Abstracts

Oral Presentations

Ab initio Spin-Orbitronics – From Spin-Orbit Interaction to Skyrmionics in real Materials

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Spin-orbitronics is an exciting direction of spintronics that exploits the spin-orbit coupling (SOC) in nonmagnetic and magnetic materials to create new types of topologically protected states [1], new novel types of topological objects as the topological orbital moment [2] or novel types of magnetic objects as the magnetic skyrmions [3], bobbers [4] or quanco balls [5], and to generate, detect or exploit spin-polarized currents or to exert spin-orbit torques [6]. The SOC is a small quantity and many of the interesting properties arise at interfaces or in bulk materials lacking inversion asymmetry and develop their full potential in the case of non-collinear magnets often together with beyond-Heisenberg interactions [7, 8]. The smallness of SOC, combined with the low symmetry, non-collinear magnetism, complex response and excitation properties, and the question of electron correlation defines an overall complexity that poses a challenge to first-principles theory. We will focus on 2 topics: (i) Topological insulators (TI): Within the all-electron FLAPW formalism [9], we show DFT, one-shot *GW*, and QS*GW* calculations for several, well-known TIs and present a comparison to photoemission spectroscopy. (ii) Skyrmions: Taking a multiscale approach combining micromagnetism, atomistic spindynamics [9] with DFT methods as implemented in the FLEUR and juKKR codes [9], we are explaining interactions and looking for materials hosting skyrmions ready of technological applications.

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Polarons from First Principles

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Polarons are among the most well-known quasiparticles in solid state physics, and are key to understanding fundamental concepts such as the electron mass enhancement in semiconductors and the formation of Cooper pairs in superconductors. Polarons have been investigated ever since the concept was formulated by Landau almost a century ago, and significant progress has been made in numerical solutions of model polaron Hamiltonians. On the other hand, first-principles calculations of polarons remain very challenging, as they require very large supercells, are sensitive to the DFT exchange and correlation functional, and suffer from the DFT delocalization problem.

In this talk I will describe a new approach to the polaron problem that aims at overcoming these limitations [1,2]. Our strategy is inspired by the related problem of calculating exciton binding energies and wavefucntions using the Bethe-Salpeter equation. Excitons can be delocalized over many crystal unit cells, but the Bethe-Salpeter equation only requires information about DFT Kohn-Sham wavefunctions and Coulomb matrix elements within a single crystal unit cell. In the same spirit, in our approach we reformulate the calculation of polarons via DFT as the solution of a coupled non-linear system of equations. The ingredients of these equations are computed in the primitive unit cell using from density-functional perturbation theory. By solving these 'polaron equations' we obtain the wavefunction, the lattice deformation, the formation energy, and the spectral decomposition of the self-trapped state into the underlying Bloch states and phonon modes. In order to demonstrate this methodology I will discuss a few case studies including large and small polarons in real materials, and show that even in the simplest crystals the structure of polarons is considerably richer than previously thought.

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Spins meet electric dipoles: modelling, discoveries and perspectives

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The discovery of novel properties, effects or phenomena in modern materials science is often driven by the quest for the coexistence and/or coupling of several functional properties into a single system. Within this framework, I will focus on the microscopic mechanisms leading to the interplay between spin and dipolar degrees of freedom, along with their theoretical modelling. I will first address the coupling of long-range magnetic and electric dipolar orders in multiferroics, in particular in electronic ferroelectrics, where the spin/charge/orbital order induces an electric polarization. Second, I will discuss (non-magnetic) ferroelectric semiconductors, where the spin-orbit interaction leads to a tight link between Rashba spin-splitting in the electronic structure, spin-texture and electric polarization. The common denominator is to achieve the electric-field control of magnetism and, therefore, the long-sought integration of spintronics with ferroelectricity.

Ab initio calculations of the lattice thermal conductivity: the discovery of new materials and multi-scale modeling

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Within the last few years it has been possible to compute the lattice thermal conductivity of bulk materials using ab initio methods. The interactions between the phonons are obtained from density functional theory and this information is incorporated into the Boltzmann to obtain the thermal conductivity. The good accuracy obtained from those calculations allows trying to use them to find new materials and perform multiscale modelling.

Here we present several strategies that we used performing such a search. The first method we used is datamining. We screened the entire Material Project library to find materials with ultra low thermal conductivity. The second method is based on polymorphism and was used to study Zn-Sb compounds. Finally we conclude showing how ab initio calculations can be combined with Monte Carlo simulations to describe thermal conduction at the micron scale and therefore achieving multiscale modeling of heat conduction.

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Machine learning enabled structure optimization

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When using Density Functional Theory (DFT) in global structure optimization, databases of structure-energy relations may be collected. Such databases may be used to direct and speed up the structure search. In this talk, I describe how machine learning methods such as clustering and regression may assist Monte Carlo [1] and evolutionary search methods [2-5] in finding the optimal structure of molecules and inorganic surface reconstructions. One key element of using machine learning methods in conjunction with DFT is the possibility of decomposing the total DFT energy into atomic contributions. Owing to the nature of the Kohn-Sham equations (the one-electron Schrödinger Equation) such a decomposition is ordinarily not supported. Nonetheless, our experience shows that the atomic energies are highly meaningful and lead to a speed up of the structure determination when used appropriately. As a final topic, I will demonstrate a search strategy in which the computer collects knowledge of atomistic interactions while searching, which eventually enables it to search in a rational way [6]. This approach combines image recognition and reinforcement learning strategies and follow the recent advancements by Google Deep Mind in their development of the Alpha Go Zero method for playing the board game Go.

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Spin and Orbital Rashba-Edelstein Effect in Noncentrosymmetric Antiferromagnets

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The efficient electrical manipulation of magnetic order is currently desirable for achieving fast devices in the field of spintronic. By exploiting spin-orbit coupling, the spin Hall effect has been explored mostly to generate a spin polarized current in magnetic nanostructures which are grown on heavy metals or semiconductors. An electrical generation of a nonequilibrium spin polarization via the Rashba-Edelstein effect in noncentrosymmetric magnetic materials is an alternative concept in the growing research field of spin-orbitronics. These effects provide current-induced staggered spin-orbit torques that can act efficiently on the magnetization of a magnetic layer on a nanometer scale in order to alter its magnetization direction. Both effects are concentrated on exploring the induced polarization of the spin part while orbital counterpart has been disregarded completely. Here, within first-principles electronic structure calculations based on density functional theory, we present a generalized theory on the Rashba-Edelstein effect, which not only generates the spin polarization but also creates the orbital polarization. Especially, the latter one is found to be far from being negligible and thereby could play a crucial role in the magnetization dynamics. We investigate computationally the full magnetic polarization induced by an applied electric field in the noncentrosymmetric antiferromagnets e.g. CuMnAs and Mn_2Au.

Topological surface states of thin films modulated with impurities

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The surface state plays an important role in condensed matter physics, and is dominant in many phenomenon, such as quantum Hall effect [1]. However, the two surfaces of a thin film may influence each other and cause the opening of energy band gaps due to quantum tunneling. In order to close the gap, we consider the absorption of and substitution by impurities [2,3], as well as contact with a heterogeneous atomic layer [4]. The Sb and SnTe thin films have been chosen for investigation by density functional calculations. The results show that the two surfaces reduce their interaction and can be identified with the original surface and interface states. The nonmagnetic impurities close the energy gap and facilitate the formation of Dirac cones characteristic of time-reversal symmetry. However, atoms with magnetic moments do just the opposite. In the case of a thin Sb film contacting with a single layer of various heterogeneous atoms, further studies are taken on the bonding and the electronic structures of the two parts of the system. The result indicates that the bonding strengthens on the condition that the contacting single layer has defects or impurities.

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Fast prediction of topological materials from first-principle calculations

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Topological electronic materials host novel linear responses in the bulk and anomalous gapless states at the boundary, and are for scientific and applied reasons under intensive research in physics and materials sciences. The detection for such materials has so far been hindered by the involved calculation of topological invariants, which requires both experience with materials and expertise with advanced theoretical tools. Here we introduce an effective, efficient and fully automated algorithm diagnosing the nontrivial band topology in a large fraction of nonmagnetic materials. It is based on recently developed exhaustive mappings between the symmetry representations of occupied bands and the topological invariants (or topological nodes). We sweep through a total of 39519 materials available in a crystal database, and find that as many as 8056 of them are topologically nontrivial. All results are available and searchable at http://materiae.iphy.ac.cn/ with an interactive user interface.

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Physics of twisted bilayer graphenes

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When 2D materials having different periodicities are overlaid with each other, the interference pattern of the lattice mismatch often leads to unusual electronic properties. For instance, the twisted bilayer graphene (rotationally stacked graphene bilayer) exhibits dramatic phenomena such as the flat band formation and emergent superconductivity. In this talk, we introduce our recent theoretical works on incommensurate twisted bilayer systems and their interesting physical properties. We will discuss the twisted bilayer graphene, where we introduce a theoretical framework to reduce its complex electronic structure into an effective nanoscale tight-binding model [1]. We then discuss the lattice relaxation and the phonon vibrations in the twisted bilayer graphene. [2] We show that the original linear dispersion of graphene's acoustic phonon is broken down into mini phonon bands separated by gaps, where the low-energy phonon modes are regarded as effective vibrations of the nanoscale moiré supercrystal.

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Density Functional Approach to Hydrogen on Electrode

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The Pt(111)-water interface has been intensively studied as benchmarking electrochemical system, while the understanding is still incomplete. The electrode surface is covered by hydrogen atoms when negatively biased, but the spectroscopic signal is missing from the adsorbed H except for few studies. This contrasts to many density functional (DFT) simulations, which unambiguously predict the fcc sites as the most stable site. To see the reason for this mystry, we have done an accurate DFT calculation at the level of the random phase approximation (RPA) and included the nuclear quantum effect using the path integral molecular dynamics method (PIMD) [1].

The RPA and PIMD calculations provide a novel picture on this system. The top site is as stable as the fcc site at full monolayer coverage condition, which is achieved at potentials lower than the equilibrium one for $2H^+(aq)+2e^-\rightarrow H_2(g)$, while the fcc and hcp sites are significantly lowered in energy at ~ 0.7 monolayer coverage, which is achieved at the equilibrium potential, because of the emerging quantum delocalization effect. The quantum effect plays a role in screening the vibrational signal and causes the striking coverage effect. H on Pt(111) has been conventionally recognized as a "hard sphere" based on the calculations done at monolayer coverage condition, but H shuld be recognized as a "quantum hard sphere" at lower coverages.

There still remains unexplained properties about H/Pt(111) that is likely to be due to incomplete DFT accuracy. We propse possible strategies for achieving higher accuracy; developing accurate exchange-correlation potential using machine learning, which will be detailed in the poster presentation [2], and utilizing empirical DFT+U(R) approach.

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A Many-Body Approach for Calculating Absolute Values of Core-Level Binding Energies

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One of the beauties of density functional theory is that it allows us to get the many-body total energy and charge density of a system by just solving "single-particle" Kohn-Sham Hamiltonian. Having the many-body total energies, it is possible to calculate quasiparticle energies, which are related to the differences between the ground-state energy and the excited-state ones, for example, the core-level binding energy. Experimentally, the core-level binding energy is widely used to fingerprint the constituents of a studied system, thank to the high-resolution X-ray photoelectron spectroscopy (XPS) measurement. In this talk, I will discuss how to calculate core-level binding energies in the absolute scale from first principles via the introduction of a projection operator and the help of an exact Coulomb cutoff technique [1]. The proposed method can be applied for studying molecules, metals, and insulators, and the results are in good agreement with experiments in general.

Three case studies will be given to illustrate the usefulness of the calculations of absolute core-level binding energies. First, I will focus on the case of silicene on $ZrB_2(0001)$, where two theoretical single-layer structures are the candidates for the silicene fabricated by experiments. The many-body core-level binding-energy calculations can clearly show that only one of the two structures is in agreement with experiments while the experimentally fabricated silicene cannot be identified via the Kohn-Sham eigenvalues [2]. Then, I will discuss a peculiar honeycomb bonding in borophene formed on Ag(111). This peculiar bonding can be confirmed by the investigation of core-level binding energies. Both theoretical and experimental data support the existence of this hidden honeycomb bonding [3]. Finally, I will discuss a large binding-energy shift measured by the XPS experiments for the samples of Pt atoms on graphene. The theoretical study supports the achievement of fabricating uniform dispersion of single Pt atoms on graphene [4].

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New materials and new states under extreme conditions

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ABSTRACT

High pressure is an important method to modify the free energy surface of materials and overcome the barriers for synthesizing new functional materials. On the other hand, crystal structure search based on ab initio calculations has been successfully used to prediction new materials. In this talk, I will introduce some of our recent work on theoretical prediction on high pressure phase transitions, functional materials with interesting properties, and new states of matter under extreme conditions, such as superionic states.

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Atomic energy mapping of neural network potential

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Recently, machine-learning (ML) approaches to developing interatomic potentials are attracting considerable attention because it is poised to overcome the major shortcoming inherent to the classical potential and first-principles method, i.e., difficulty in potential development and huge computational cost, respectively. Favored ML models are the neural network and Gaussian process. In particular, the high-dimensional neural network potential (NNP) suggested by Behler and Parrinello is attracting wide interests with applications demonstrated over various materials encompassing metals, insulators, semiconductors, and molecular clusters. In this presentation, we first discuss on the fundamental aspect of ML potentials that enables the transferability of the potential.[1] We show that the transferable atomic energy can be defined within the density functional theory, which means that the core of machine-learning potentials is to deduce a reference atomic-energy function from the given set of total energies. By utilizing invariant points in the feature space at which the atomic energy has a fixed reference value, we examine the atomic energy mapping of neural network potentials. Examples on Si consistently supports that NNPs are capable of learning correct atomic energies. However, we also find that the neural network potential is vulnerable to 'ad hoc' mapping in which the total energy appears to be trained accurately while the atomic energy mapping is incorrect in spite of its capability. We show that the energy mapping can be improved by choosing the training set carefully and monitoring the atomic energy at the invariant points during the training procedure. The energy mapping in multi-component systems is also discussed. We also introduce our in-house code for training and executing NNP called SIMPLE-NN (SNU Interatomic Machine-learning PotentiaL packagE-version Neural Network) [2] and discuss its unique feature such as GDF weighting [3]. We present application examples on the oxidation of Si-SiO$_2$ interface, phase change behavior of GeTe and Sb$_2$Te$_3$, silicidation process of Ni.

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Dynamical magnetoelectric coupling and optical effect in Ni$_3$TeO$_6$:
from *ab-initio* simiulation of optical *d-d* excitation spectra

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Nonreciprocal directional dichroism is an unusual light-matter interaction that gives rise to diode-like behavior in low symmetry materials [1,2]. The chiral varieties of nonreciprocal directional dichroism are particularly scarce due to the requirements for strong spin-orbit coupling, broken time reversal symmetry, and a chiral axis. We bring together magneto-optical spectroscopy and first principles calculations to reveal high energy, broad band nonreciprocal directional dichroism in Ni$_3$TeO$_6$ with special focus on behavior in the metamagnetic phase above 52 T [3]. Using first-principles-based methods it is shown how the Ni^{2+} *d*-to-*d* on-site excitations [4] develop magnetoelectric character via relativistic spin-orbit coupling, which present a microscopic model that unlocks the door to theory-driven discovery of chiral magnets with nonreciprocal directional dichroism.

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Design of spintronics and magnetic materials by Korringa-Kohn-Rostoker Green's function method

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First-principles electronic structure calculations are useful tools for materials development for next generation electronic devices. In general electronic structure calculation methods, the central problem is to solve the one-electron Kohn-Sham equation. The Korringa-Kohn-Rostoker (KKR) Green's function method avoids the calculation of the Kohn-Sham eigenvalues and orbitals, and directly determines the one-electron Green's function which contains the all the information about the ground state. There are several advantages in the KKR Green's function method. One of them is that order-N calculation scheme is realized by the screened transformation, where N is the number of atoms in a unit cell. The screened KKR Green's function method enables us to calculate complex large-scale systems, such as substitutional, configurational, and spin disordered systems. Additionally, combining with the linear response theory, one can efficiently calculate physical quantities (e.g., magnetic interaction, spin susceptibility and transport properties).

In this talk, we demonstrate the materials design of magnetic phase change materials[1] and quaternary magnetic high entropy alloys[2,3], showing the strongly substitutional and configurational disorders. For both the systems, we construct very large supercells to take the randomness into consideration. Such the large-scale electronic structure calculations are performed using the program KKRnano,[4] where the full potential screened KKR Green's function method is optimized by a massively parallel linear scaling (order-N) all electron algorithm. If time permits, we also talk about automatic high-throughput screening for quaternary magnetic high entropy alloys by the AkaiKKR code.[5]

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2 Dimensional Magnetic Semiconductors Based on Transition-Metal Dichalcogenides

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2D magnetism has become a very hot topic in recent years owing to significant progresses in fulfilling 2D magnetism in single layer transition metal dichalchogenides (TMD) and monolayer CrI3. Meanwhile, 2D magnetism is of high potential in future spintronics because of the capability of controlling spin polarization, spin current, and so on. In this talk, we present a newtype 2-dimensional (2D) magnetic semiconductor based on transition-metal dichalcogenides VX2 (X = S, Se and Te) via first-principles calculations. The obtained indirect band gaps of monolayer VS2, VSe2, and VTe2 given from the generalized gradient approximation (GGA) are respectively 0.05, 0.22, and 0.20 eV, all with integer magnetic moments of 1.0 μB. The GGA plus on-site Coulomb interaction U (GGA + U) enhances the exchange splittings and raises the energy gap up to 0.38~0.65 eV. By adopting the GW approximation, we obtain converged G0W0 gaps of 1.3, 1.2, and 0.7 eV for VS2, VSe2, and VTe2 monolayers, respectively. They agree very well with our calculated HSE gaps of 1.1, 1.2, and 0.6 eV, respectively. The gap sizes as well as the metal-insulator transitions are tunable by applying the in-plane strain and/or changing the number of stacking layers. The Monte Carlo simulations illustrate very high Curie-temperatures of 292, 472, and 553 K for VS2, VSe2, and VTe2 monolayers, respectively.

Hidden Corner States in Quantum Spin Hall Flakes

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We uncover the emergence of the hidden zero-dimensional corner states at a series of step edges of Bismuth thin films, which are famous for supporting the quantum spin Hall effect. Our approach is in stark contrast to the majority of the previous approaches which rely solely on the intrinsic topology of a single layer material, and here we utilize the relative difference of topology between the different film widths which match that of the two-dimensional higher-order topology. To critically examine the emergence of the corner modes, we combine the first-principle calculation, tight-binding model calculation, and analytic field theory methods. By invoking the dimensional reduction arguments, we further expose the underlying reason why such two-dimensional higher-order topology appears in Bismuth thin films and a general recipe for finding the higher-order topological insulators in similar materials.

An efficient generalized Gruneisen method for a first-principles determination of thermal expansion coefficients for all crystal types

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Phonon anharmonicity is responsible for several important thermal properties such as thermal expansion coefficient and thermal conductivity. Here we present an efficient generalized method [1-5] based on the Gruneisen approach, to calculate the thermal expansion coefficients (TECs) of any crystal system. This method avoids the extensive free energy search in the lattice-parameter space required by the quasiharmonic approximation that has an unfavorable scaling where N^p phonon calculations are usually performed, p is the number of cell parameters (e.g., $p=1$ for a cubic cell and $p = 4$ for a low-symmetry monoclinic cell). $N \sim 5$ is typically the number of lattice parameter values scanned in each cell dimension. Here we show that it is possible to perform a number of phonon calculations that is proportional to p (typically $2p$). The concept of thermal expansion tensor of low-symmetry crystals such as a monoclinic cell is presented and found to be different from the commonly known lattice-parameter TECs reported for high-symmetry cells such as a cubic cell. The relationship between two TEC concepts are presented which allows a convenient conversion between theory and experiment results. Our theory is applied to a recently reported monoclinic NiS3 crystal phase where we have calculated the TEC tensor of a monoclinic system [1] for the first time using density functional theory and phonon calculations. A negative TEC tensor component is found and attributed to elastic properties through the elastic compliances and not the population of the negative Gruneisen parameters. The success of our work demonstrated for the monoclinic cell implies all seven crystal types in nature could now be treated within the same formalism.

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Time Dependent Nonadiabatic Molecular Dynamics Investigations on the Excited Carrier Dynamics in Condensed Matter Systems

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The ultrafast dynamics of photo-excited charge carriers in condensed matter systems plays an important role in optoelectronics and solar energy conversion. Yet it is challenging to understand such multi-dimensional dynamics at the atomic scale. Combining the real-time time-dependent Kohn Sham equation (TDKS) with fewest surface hopping scheme, we use time-dependent ab initio nonadiabatic molecular dynamics (NAMD) to simulate the excited carrier dynamics in different condensed matter systems. The time-dependent dynamics of excited carriers are studied in energy, real and momentum spaces. In addition, the coupling of the excited carriers with phonons, defects and molecular adsorptions are investigated. The state of art NAMD studies provide unique insights to understand the ultrafast dynamics of the excited carriers in different condensed matter systems at the atomic scale.

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Real-time ab-initio simulations for crystalline solids driven by strong laser pulse

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The recent development of coherent light sources allows us to drive nonperturbative electron dynamics in crystalline solids [1,2]. An interesting phenomenon is accompanied by a light electric field that has field strength is comparable to V/nm scale. This electric field shakes systems with a quite nonperturbative manner because the field strength is close to typical scale for valence electron, eV divided by nm. This strong-field science has the potential to open a new type of nonequilibrium condensed matter physics.

Compared to linear optical science, many degrees of freedom takes part in strong-field science. A first-principles theoretical simulation could be beneficial to have what happens with a microscopic description. Density-functional theory (DFT) and time-dependent density-functional theory (TDDFT) are foundations for one of the most feasible theoretical framework to describe the nonperturbative electron dynamics. We have developed theoretical frameworks by solving time-dependent quantum systems in real-time based on DFT [3,4] and TDDFT [5]. The former framework is based on a quantum system whose Hamiltonian is derived from DFT matrix elements called time-dependent density-matrix (TD-DM), while the latter employs a direct solution of TDDFT.

We will talk about TD-DM and its application to high-order harmonic generation, photon energy upconversion by nonlinear interaction, from crystalline solids compared with experimental counterparts performed by our collaborators [3,4]. Recently, we have struggled to perform the exchange term of solids for the time-dependent problem [6] to capture excitonic physics within the mean-field level. Our achievement is still limited to the one-dimensional model. We would like to show the current progress of applications to three-dimensional systems and discuss with the audience.

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Apical Ion Dynamics-Modulated In-Plane Transport Properties of Cuprates

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Since the discovery of high-temperature superconductivity in hole-doped La2CuO4, the mechanism of cuprate superconductors has been one of the most important problems in condensed matter physics. Strong electronic correlations in CuO2 planes has been considered to play some essential role. However, other phenomena like abnormal phonons or lattice effects constantly add the complexity to the understanding of high-Tc superconductivity. In this talk, we show, using ab initio simulations, a new trend that the bonding strength between the apical cation (e.g. La, Hg, Bi Tl) and apical anion (O, Cl) is positively correlated with experimental $T_{c,max}$ across the hole-doped cuprates. The "apical structure unit" formed by the apical anion and the apical cation, the in-plane Cu and its nearest oxygen neighbors is a fundamental building block that can couple dynamically to control the superconductive properties. We present the underlying fundamental phenomena of coupled apical charge flux and the phonon/lattice dynamics of apical oxygen when the apical oxygen oscillates vertically. The effect not only dynamically modulates the site energy of the hole at a given Cu site to control the in-plane charge transfer energy, but also can modulate the in-plane hole hopping integral simultaneously in a dynamic way by the cooperative apical charge fluxes. We believe our understanding here can shed light on the understanding of the complicated phenomena in cuprates, especially how the transport properties are controlled by the coupled electronic and ionic dynamic oscillations.

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Search for superconducting ternary hydrides by materials informatics based on evolutionary algorithms

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In 2015, high-temperature superconductivity was discovered in sulfur hydride under high pressure and the superconducting critical temperature (T_c) reaches 203 K at pressure of 155 GPa[1]. In 2018-2019, higher T_c superconductivity was observed in lanthanum hydride (250 K at 170 GPa[2] and 260 K at 190 GPa[3]) and yttrium hydride (224 K at 166 GPa[4]). Very recently, the remarkably high-T_c superconductivity, 473 K at 250 GPa, was predicted in metastable ternary Li_2MgH_{16} by first-principles calculations[5]. These results suggest that other hydrides, especially ternary hydrides, also have a potential to show similar high-T_c superconductiviry under high-pressure conditions. However, there exist 13572 combinations for ternary hydrides formed by all elements with the atomic numbers from 2 to 118, and the number is much further increased by taking stoichiometry and crystal structure into account. Therefore, the integration approach of meterials science and data science, in other words, the materials informatics (MI) is of great help to discover potential candidates for the high-T_c superconductivity.

In this study, we present an MI method to search for superconducting hydrides, based on evolutinary algorithms. This method consists of five stages: (i) collection of physical and chemical property data for hydrides under high pressure, (ii) development of a superconductivity predictor from the data by a genetic programming technique, (iii) prediction of potential candidates for high-T_c superconductivity, (iv) search for stable crystal structures of the candidates by a genetic algorithm technique, and (v) validation of the superconductivity by first-principles calculations. By repeatedly performing the process, the database and the predictor are further improved, which leads to an efficient search for superconducting hydrides. We applied this MI method to hypothetical ternary hydrides and predicted $KScH_{12}$ with a modulated hydrogen cage showing T_c of 122 K at 300 GPa and $GaAsH_6$ showing 98 K at 180 GPa[6].

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Effect of surface doping on electron correlation in FeSe

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The single-unit-cell-thick electron-doped FeSe layer has been attracting enormous attention for high-temperature superconductivity and strong electron correlation that is enhanced with the doping. Since the doping makes Fe 3d orbitals away from half filling, the enhancement is quite anomalous. Here we investigate electronic structure of potassium-dosed FeSe layers using the density functional theory combined with the dynamical mean-field theory. We show that K dosing induces the charge transfer from K atoms to the topmost FeSe layer, and subsequently, ionized K atoms generate a strong local electric field. Role of this charge-transfer induced electric field is discussed with emphasis on its impacts on the electronic structure and electron correlation among Fe 3d orbitals. By controlling the concentration of K atoms, we systematically investigate the evolution of the electronic structures of FeSe monolayer and bilayer. Notably, K dosing reduces bandwidths of the Fe 3d bands near the Fermi level and significantly enhances electron correlation. We also discuss the structural changes of FeSe layers due to K dosing. Our results illustrate that charge transfer from external agents to FeSe layers can have substantial effects other than electron doping and account for their enhanced electron correlation. This work was supported by NRF of Korea (Grant No. 2011-0018306) and KISTI supercomputing center (Project No. KSC-2017-C3-0079).

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Effective Non-Radiative Recombination-Center Defects in Multinary and Low-Symmetry Semiconductors

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The defect-induced non-radiative recombination is a serious limit to the minority carrier lifetime and the photovoltaic performance of the light-absorber semiconductors in solar cells. The accurate determination of the carrier-capture cross sections of defects and the corresponding carrier recombination rate is challenging for both the experimental measurement and theoretical calculation. Experimentally, there is no efficient and direct method for measuring the carrier-capture cross sections or non-radiative recombination rate induced by different point defects. Usually the carrier lifetime was measured, but it is difficult to identify which defect dominates the non-radiative recombination process, especially for the multinary and low-symmetry which may have various point defects in the lattice. Through systematical defect search, we identified a series of possible recombination-center defects in three novel photovoltaic semiconductors, the quasi-one-dimensional Sb_2Se_3, quaternary Cu_2ZnSnS_4 and the organic-inorganic hybrid halide perovskite $CH_3NH_3PbI_3$. By combining the electron-phonon coupling effect and the static-coupling formalism, we calculated the carrier-capture cross sections of the possible non-radiative recombination-center defects in these semiconductors. These values are currently unavailable but critical for understanding the limiting factors of the minority carrier lifetime and simulating the photovoltaic devices. A simple empirical criterion was proposed for the quick identification of effective non-radiative recombination-center defects based on the calculated results, *i.e.*, the deep-level defects may have large carrier-capture cross sections if they are surrounded by strong bonds and undergo considerable structural relaxations after capturing a carrier.[1-3]

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DFT Sampling Studies on Interface Ionics at Disordered Heterogeneous Solid-Solid Interfaces

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Interface (and surface) processes in energy and environmental devices such as batteries and catalysts largely govern their performance and stability, where both electronics and ionics play crucial roles. To understand such interfacial processes, however, typical static geometry optimization or even molecular dynamics (MD) based on DFT framework does not work well often due to the inherent heterogeneous and multiscale characters. On the other hand, parameter-based methods such as classical MD are still insufficient from a viewpoint of predictability because their parameters are usually fitted to bulk properties, not interface. In this situation, combination of DFT calculation with another type of sampling such as microkinetics and structure prediction technique is an interesting direction.

In this talk, I will introduce DFT sampling approaches recently implemented in our group for disorderd heterogeneous solid-solid interfaces and our researches on the inonics at oxide cathode / sulfide electrolyte interfaces in all-solid-state batteries [1-4]. The sampling breakthrough was achieved by the CALYPSO technique [5,6], which allows us to explore a variety of interfacial disordered structures. The approach is compatible to massive parallel calculations with high performance computing resources like "Fugaku". The sampled interfacial structures enable to examine the chemical potential and potential energy surface of mobile ions around the solid-solid interfaces, which gives a perspective about the interfacial space-charge and electric double layers as well as their relations with the interface ionics.

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Abstracts

Poster Presentations

Electronic structure of rare-earth nitrides calculated by quasi-particle self-consistent *GW* method

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Rare-earth (RE) related compounds show broad range of functionality and their importance for our present life is taking on an added significance. Probably, the oldest application of RE is optical one and it has long history as a fluorescent material. Recently, LED was fabricated based on Eu-doped GaN and light emission due to carrier injection was realized. RE based light emitting materials are distinguished for their narrow line width, small temperature dependence and insensitivity to the environment. These characteristic properties mostly come from localized nature of 4f-states. Therefore, for accurate prediction and design of RE related functional material reasonable description of electronic structure of 4f-states is indispensable.

However, electronic structure calculation is not trivial for RE systems, because the local density approximation (LDA) or the generalized gradient approximation (GGA), they are frequently used for the band structure calculations, do not work reasonably for 4f-systems. Particularly for open 4f-shells, partially occupied 4f-bands appear around the Fermi level mostly due to the self-interaction within 4f-orbitals.

In this paper, we present systematic electronic structure calculations of RE nitrides (RE = Sc, Y, La ~ Lu) by using quasi-particle self-consistent *GW* (QSGW) method. This method was implemented to first-principles calculation package ecalj, and confirmed to give reasonable description of the electronic structure of several materials including f-electron systems [1, 2].

Fig. 1 shows calculated band structure of SmN. As shown in the figure, the system shows semi-metallic electronic structure. Calculated moments (S=2.50, L=4.96, J=2.46) are consistent to Hund's rule prediction. Present systematic calculations show calculated magnetic moments are reasonably described by the Hund's rule for most of the rare-earth nitrides with few exceptions.
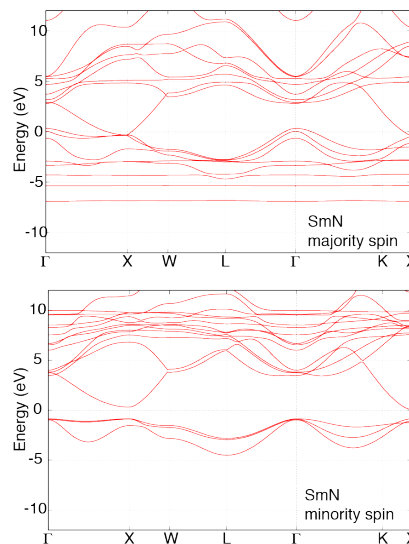

Fig. 1. Calculated band structure of SmN.

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Strain-induced electronic property modulation of monolayer tellurium: a first-principles study

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We employed density functional theory calculations to systematically investigate the strain-modulated structural, electronic and optical properties of monolayer tellurium. Our findings show that at zero strain γ-phase monolayer tellurium is found to be more energetically favorable than either the α-phase or β-phase, which were fabricated through molecular-beam epitaxy. All studied phases are found to exhibit semiconductor properties, of which α- and γ-phases exhibit indirect band gaps whereas β phase has a direct band gap. Furthermore, we found that certain strain can drive the structural transitions between α-phase and β-phase. The resulting band gap values approach zero at a large strain regime for all studied systems and the effective mass of electron and hole can be effectively altered by strain as well. These findings extend the knowledge on two-dimensional tellurium and provide potential applications in electronic devices.

Strain induced topological insulator phase in $CsPbBr_xI_{3-x}$ (x=0, 1, 2, and 3) perovskite: an *ab initio* study

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Topological insulators (TIs) with a nontrivial band structure have attracted much attention in the past decade[1]. $CsPbCl_3$, $CsPbBr_3$, and $CsPbI_3$ perovskites are known as promising materials for solar cells[2]. Recently, $CsPbI_3$ is theoretically predicted to be a TI under high pressure.[3-5] In this study, we have calculated the electronic band structures of $CsPbBr_xI_{3-x}$ (x=0, 1, 2, and 3) perovskite by using the density functional theory. The equilibrium lattice constant of $CsPbBr_xI_{3-x}$ is obtained from the minimum of the total energy as a function of lattice constant. We found that in its equilibrium state, the $CsPbBr_xI_{3-x}$ is a normal insulator with a bandgap of 0.6711 eV, 0.4849 eV, 0.3868 eV, and 0.3250 eV for $CsPbBr_3$, $CsPbBr_2I$, $CsPbBrI_2$ and $CsPbI_3$, respectively. The bandgap of $CsPbBr_xI_{3-x}$ decreases monotonically to zero when applying hydrostatic stress. The topological insulator phase then arose at about 6 GPa of pressure. The band inversion occurs with and without spin-orbit coupling. A nontrivial Z_2 topological number is obtained. Furthermore, the surface conduction band is calculated and confirmed by using a 1x1x10 supercell. If the center Pb atom is moved and the structure inversion symmetric is broken, the $CsPbBr_xI_{3-x}$ becomes polarized and the electric polarization P_{ele} is calculated. Typically, the electric polarization increases as the Pb ion moves away from its origin. However, P_{ele} of $CsPbBr_2I$ becomes negative as the Pb atom moves. We also found that at high pressure the topological phase also exists on the polar surface.

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Noncollinear Spin Torque Effect in Magnetic Heterojunctions: Combined First-Principles Calculation and TB-NEGF Method

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Our theoretical researches focus on searching for novel nm-scale ferromagnetic/barrier/ferromagnetic (FM/B/FM) magnetic tunnel junctions (MTJs) with superior spin transport properties by choosing oxides [1] and organic molecules [2] as the central barriers. The relative orientation of two FM electrodes can be controlled by a spin-polarized current via the noncollinear spin torque effect. Recently, we have successfully employed the single-band tight-binding (SBTB) model to predict the noncollinear spin torque effect in FM/I/FM and FM/I/SF/I/FM [1] magnetic tunnel junctions, where I and SF represent insulting and spin-filter barriers, respectively. However, for real complex heterojunctions, the injected spin-polarized electrons from FM electrode can be strongly influenced by the complicated interfacial spin-polarized charge transfer, which is ignored in our previous SBTB model.

In this study, our newly developed "JunPy" [3] package successfully combined the self-consistent Hamiltonian by using the first-principles calculation, including multi-band dispersion relation and complicated interfacial coupling, with our newly derived TB model and non-equilibrium Green's function (NEGF) method to investigate the noncollinear magnetotransport properties in nm-scale magnetic heterojunctions. This program is first testified for the spin-polarized currents and the noncollinear spin torque effect in conventional Fe/MgO/Fe MTJ. We further employed it to predict the giant field-like spin torque (FLST) effect in the amine-ended single-molecule magnetic junction [5], which may open a new avenue for multifunctional manipulation in next-generation organic FLST-MRAMs with lower power consumption. We believe that this newly developed calculation process not only can efficiently resolve current self-consistent difficulties in first-principles calculation for non-collinear case, but also may inspire future experimental explorations in novel magnetic heterojunctions for future spintronics applications.

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Metallic and Magnetic Edge States of HfSe$_2$ Nanoribbons

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Recently, transition metal dichalcogenides (TMDs) have become a focus of intense research due to their diversity of characteristics and interesting dimensionality effects. Among TMDs, high carrier mobility renders HfSe$_2$ a promising choice for FET and phototransistor applications [1]. In this study, the first-principles calculation are carried out to investigate size and edge effects on the structural, electronic, and magnetic properties of 1T-HfSe$_2$ from bulk to nanoribbons. The hybrid functional HSE06 is considered to meet the experimental band gap ($\sim$ 1.1 eV for monolayer HfSe$_2$ [2]). From 3D-bulk to 2D-film, the nearly invariant energy gap reveals the weak inter-layer van der Waals interaction of 1T-HfSe$_2$ [3, 4]. When we further downsize to 1D nanoribbons with zigzag and armchair edges, our first-principles calculations suggest that only Hf-terminated zigzag edge exhibits magnetic and metallic properties [4]. These surface-induced unpaired d electrons of edge Hf ions preserve spin-polarized surface states, which can be eliminated by absorbing adatoms, such as H atoms. In addition, the width dependence of spin polarization as well as the spin-polarized energy level variation are also discussed and may support potential applications in spintronics and nanodevices. This work is supported by the Ministry of Science and Technology (MOST 108-2628-M-008-004-MY3), the National Center for Theoretical Sciences (NCTS), and the National Center for High-performance Computing (NCHC).

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Poster-6

Substrate-mediated umklapp scattering at the incommensurate interface of a monatomic alloy layer

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Ultrathin Pb and Ge films deposited on Ag(111) surfaces have been investigated and compared. We found that at 1/3 ML, both films formed surface alloys, Ag_2Pb and Ag_2Ge, with $\sqrt{3} \times \sqrt{3}R30°$ and $\frac{19}{20}\sqrt{3} \times \frac{19}{20}\sqrt{3}R30°$ structures on Ag(111) but the surface electronic structures exhibit a most evident difference at the Ag(111) surface zone boundary $\bar{M}_{Ag(111)}$, where the single band and the splitting ones were observed, respectively. Up to 1 ML, Ag_2Ge subsequently develops into germanene with a striped phase and then a quasifreestanding phase, as previously reported [Lin et al., Phys. Rev. Mater. 2, 024003 (2018)], while Ag_2Pb evolves to a dense Pb(111) phase that also reveals splitting bands at $\bar{M}_{Ag(111)}$. We discover that the larger (smaller) atomic size of a Pb (Ge) atom with respect to an Ag atom causes the commensurate (incommensurate) interfaces and further demonstrate that the splitting bands of Ag_2Ge surface alloy and 1-ML Pb film originated from the commonly incommensurate interface with Ag(111), which mediates umklapp scattering by inducing the mirror image of the pristine Ag_2Ge and Pb(111) bands relative to $\bar{M}_{Ag(111)}$ [1].

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Electronic structure of a monoatomic Cu_2Si layer on a Si(111) substrate

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Fermi surfaces and band dispersion curves of a Cu_2Si layer on Si(111), quasi-"5×5" were mapped by angleresolved photoemission spectroscopy using synchrotron radiation. Two metallic bands were observed within the Si bulk band gap, which are likely assigned to the electronic bands of the Cu_2Si layer. Additional bands were found in the Si bulk band gap that originate from the interactions between the substrate state and the Cu_2Si states that have the pz character. The present research aims at investigating the changes in the electronic structure of an atomic layer when prepared on a substrate..

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Atomically precise bottom-up synthesis of
π-extended [5]triangulene

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The zigzag-edged triangular graphene molecules (ZTGMs) have been predicted to host ferromagnetically coupled edge states with the net spin scaling with the molecular size, which affords large spin tunability crucial for next-generation molecular spintronics. However, the scalable synthesis of large ZTGMs and the direct observation of their edge states have been long-standing challenges because of the molecules' high chemical instability. Here, we report the bottom-up synthesis of π-extended [5]triangulene with atomic precision via surface-assisted cyclo-dehydrogenation of a rationally designed molecular precursor on metallic surfaces. Atomic force microscopy measurements unambiguously resolve its ZTGM-like skeleton consisting of 15 fused benzene rings, while scanning tunneling spectroscopy measurements reveal edge-localized electronic states. Bolstered by density functional theory calculations, our results show that [5]triangulenes synthesized on Au(111) retain the open-shell π-conjugated character with magnetic ground states.

Sparse sampling approach to efficient ab initio calculations at finite temperature

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Efficient ab initio calculations of correlated materials at finite temperature require compact representations of the Green's functions both in imaginary time and Matsubara frequency. In this talk, we present a general procedure which generates sparse sampling points in time and frequency from compact orthogonal basis representations [1], such as Chebyshev polynomials [2] and intermediate representation (IR) basis functions [3]. These sampling points accurately resolve the information contained in the Green's function, and efficient transforms between different representations are formulated with minimal loss of information. As a demonstration, we apply the sparse sampling scheme to diagrammatic *GW* and GF2 calculations of a hydrogen chain, of noble gas atoms and of a silicon crystal.

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Structure Perturbation in Black Phosphorus Driven by Atom Intercalation

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Understanding the electronic properties of defects in black phosphorus (BP) is of great importance for both fundamental research and technological applications. Peanut-shaped defects observed in black phosphor (BP) have been attributed to structure vacancies or charged substitutional impurities in previous studies.[1-2] However, we observe a peanut-shaped defects caused by atom intercalation and demonstrate that atom intercalation in the van der Waals gap can be thermodynamically stable and causes relative shear of adjacent Bp layers, leading to significant electronic perturbation over 10 nm. We study platinum atom intercalation by both experiments and ab-initio calculation and confirm our picture from comparing partial density of states, structure distortion, and electronic band gap . In addition, we predict some atoms have also stable intercalation, for instance, Au, used as a catalyst for Bp growth and some atoms are not stable intercalant.[3]

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Spinless Charged Particle Tunneling Effect under Uniform Magnetic Field

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We analytically solve the Schrodinger equation with an external magnetic field to derive the tunneling probability in NM/I/NM junction, where NM and I represent nonmagnetic metal and insulating barrier, respectively. We first divide junction into three isolated parts, solve them individually, and then obtain the total wavefunctions of junction by matching the boundary conditions at two NM/I interfaces.

In this study, electron transport is along x-axis, a parabolic potential energy is considered in y-axis, a homogeneous magnetic field is applied along z-axis, and the Landau Gauge is chosen. Due to the existence of external magnetic field, the non-Hermitian Hamiltonian with imaginary eigenvalue of the momentum operator in transport direction is employed for the I-barrier. This can be treated as an unbroken parity-time symmetric operator with real eigenvalue and normalizable wave function ψ [1]. Since the probability of non-Hermitian Hamiltonian can be defined by $\psi(y, t) \cdot \psi^*(-y, t)$, the discontinuity of the wave function $\frac{\partial \psi^*}{\partial x}$ at the NM/I boundary renders us to calculate the transmission probability of wave functions ψ^* and ψ separately. Finally, we'll discuss about the origin of this discontinuity. This work is supported by the Ministry of Science and Technology (MOST 106-2633-M-008-002- and 107-2633-M-008-004-) and the National Center for Theoretical Sciences, Republic of China.

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First-principles study on electric field control of magnetism in bilayer VI$_3$

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Very recently, control of magnetic properties by mean of external electric field has been attracted as a key challenge in materials science [1,2]. One of the most emerging topics is the electric-field effect on magnetism in 2D magnetic materials, which is intensively considered as an ideal candidate for next-generation device fabrication [3]. A recent experiment has demonstrated electric-field control of magnetism in bilayer CrI$_3$, which reveals modification of an interlayer exchange coupling enables robust switching of magnetization [4]. This has been theoretically confirmed by the *ab initio* calculations showing that indeed the magnetic stability of bilayer CrI$_3$ can be tuned by external electric field [5]. In this work, we study electric-field control of magnetism in bilayer VI$_3$, new discovered ferromagnetic van der Walls material, based on density functional theory calculations. Two possible structures with different VI$_3$ layer stacking, i.e., rhombohedral (R-3, low-temperature phase) and monoclinic (C2/m, high-temperature phase) structures, have been investigated. All the calculations were performed using the VASP code [6] based on the projector-augmented wave (PAW) method with the generalized gradient approximation (GGA) potential. The Hubbard U correction was employed within the rotationally invariant GGA+U approach [7] to take account of the localized 3d orbital states at transition-metal atoms. We predict that the magnetic stability of bilayer VI$_3$ systems can be tuned by external electric field and the effect is comparable with the case of bilayer CrI$_3$. To understand the physics behind, the geometry, stability, and electric and magnetic properties of bilayer VI$_3$ will be discussed by presenting the projected density of state and band structure.

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Origin of Dimeric and Trimeric Formations of NO on Cu(111) from Van der Waals Density Functional Calculations

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NO adsorptions on metal surfaces have been studied intensively due to its importance in various catalytic processes. It is recognized that NO adsorbs molecularly on noble metal (111) surfaces, i.e., Cu(111), Ag(111), Au(111), at rather low temperature followed by the formations of dimeric $(NO)_2$ and N_2O with further gas exposure. Recently, by using scanning tunneling microscopy. Shiotari *et. al* observed unexpected NO adsorption in an trimer configuration on Cu(111) [1]. Their finding is then verified by electron energy loss spectroscopy which proposed that NO molecules in NO trimer are inclined from the surface normal owing to intermolecular couplings between NO molecules [2]. To provide orgin of the peculiar formation of NO trimer on Cu(111), we study the adsorptions of small NO clusters on Cu(111) by mean of density functional theory. We employed van der Waal Density Functional (vdW-DF) method as implemented[3] in the STATE code [4]. We found that the NO monomer preferably adsorbs in an upright configuration at the fcc-hollow site, whereas all NO molecules of the NO dimer and trimer are in inclined configurations at the neighboring fcc-hollow sites due to short-range attractions between molecules. The NO trimer is consistently the most stable regardless of lateral periodicities, which is consistent with the experiment [1,2]. Vibrational analysis also reveals that the N-O stretching mode shifts upward from 182 meV of NO monomer to 193 meV of NO trimer, and the latter mode agrees much better with the EELS data (~190 meV). The origin of the preferable formation of the NO trimer on Cu(111) is attributed to a subtle interplay between intermolecular and molecule – substrate interactions.

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First-principles Study of Diamond Strain engineering

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Diamond has been proved that can be deformed by advanced technolodge[1]. This discovery expands the potential of diamond application. Therefore, we introduce the density function theory to evaluate the electronic properties of diamond under strain. We elongate diamond along <100>, <110> and <111> directions, and investigate the variation of band structures. We found that diamond shows unique properties under specific direction of strain, such as bandgap reduction, indirect-to-direct bandgap transition and flat band appearance. With these impressive properties, diamond can be considered as promising material in applications of electronic and optomechanical devices.

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First-Principles Study of Piezoelectricity in Nanostructured Wurtzite Materials

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Piezoelectric properties of a variety of nanostructures such as one- or two-dimensional materials including nanowires and nanofilms have been attracted great interests as a potential material in applications for future piezo-electronics devices. Among piezoelectric materials, wurtzite-type materials such as ZnO have been actively investigated as a practically useful material for sensors, actuators, and energy harvesting devices because of their superior semiconducting and piezoelectric properties. It has been computationally demonstrated that a ZnO nanowire with a diameter less than 1 nm shows strong enhancements of longitudinal piezoelectric constants relative to the bulk value. However, microscopic origins of the piezoelectricity enhancements in nanowires are not fully understood yet. Here we study electronic, structural, and piezoelectric properties of ZnO nanowires and nanofilms by the first-principles calculations based on the density functional theory with the generalized gradient approximation. We use the model structures of ZnO nanowires ([0001]-oriented wires with several diameters in a few nm ranges) and ZnO nanofilms (polar [0001] Zn- and [000-1] O-terminated slabs with several thicknesses of a few atomic layers). The model structures are fully optimized by the first-principles, and electric polarizations and piezoelectric constants of the models are calculated by the so-called Berry phase method, as previously studied for bulk materials [1,2]. As a result, the ZnO nanowire calculated with diameters 3 to 16 angstrom shows the insulating electronic structure. The effective piezoelectric constants of the nanowires increase with decreasing diameter size as previously reported, and the model with the smallest diameter shows the highest effective piezoelectric constant enhanced by 151% relative to the bulk value. Both clamped-ion and internal-strain term in the piezoelectric constant contributes to the enhancement, and the latter is rather dominant showing that large atomic displacements in response to external strains. In the ZnO nanofilms, we find that two types of atomic structures (the wurtzite-type and the flat-layered structures) compete with respect to the film thickness as well as the in-plane strains even for a fixed thickness. Though the wurtzite-type and the flat-layered structures show metallic and insulating surface electronic states, respectively, this result provides a possibility to control piezoelectric behavior of ZnO nanofilms around the structural phase boundaries by tuning film thicknesses and in-plane strains.

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The evidence of new unconventional antiferromagnetic accompanying with insulators and semi-conductors in double-perovskite $BiPbCrMnO_6$ and $BiPbCrTcO_6$

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Based on the density-functional theory (DFT), the generalized gradient approximation (GGA), and GGA incorporated with Coulomb correlation interaction U (GGA+U), double-perovskites $BiPbCrMnO_6$ and $BiPbCrTcO_6$ are investigated for new unconventional antiferromagnetic insulators (UAF-Is) and semi-conductors (UAF-SC) by the first-principles spin-polarized calculations with electronic structure relaxation. The two compounds are UAF-Is in one spin channel and UAF-SC in the other spin in GGA, while they are totally UAF-Is in GGA+U. There are stable states for the induced UAF-Is because their UAF configuration has an about 250~500 meV lower than the ferromagnetic (FM) configuration. This discovery of property in double perovskites $BiPbCrMnO_6$ and $BiPbCrTcO_6$ contributes the useful candidates for the field of spin-based electronics, or spintronics, information utilized by ferromagnets or antiferromagnets.

Key words: First-principles, double-perovskite, unconventional antiferromagnetic insulators, unconventional antiferromagnetic semi-conductors
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Topology analysis for anomalous Hall effect in magnetic octupole Mn$_3$$A$N

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Anomalous Hall (AH) effect has been known as an important macroscopic phenomenon in materials science because of its application to spintronics such as electronic probes, switches, and memory devices [1]. The discovery of large AH effect in non-collinear antiferromagnets with no net magnetization has led an increasing amount of attention in studying the topological features of electronic band structure for the AH effect because of the insensitivity against an applied magnetic field and no stray fields interfering with the neighboring cells as well as faster spin dynamics than ferromagnets. In this work, we investigate in detail the topology features of electronic structures which produce the large AH effect in antiferromagnetic (AFM) states for a series of anti-perovskite manganese nitrides Mn$_3$$A$N ($A$ = Ni, Cu, Ga, Ge, In, Sn, Ir) by first-principles density-functional-theory calculations.

The modern formalism of the AH effect has shown that the AH conductivity is obtained as Berry curvatures integrated over the Brillouin zone. Band crossing points called Weyl points have been considered to have an essential role for large AH effect in many studies since it divergently increases the Berry curvature [2-4]. Our work revealed that small Berry curvatures which are widely spread throughout the Fermi surface in the Brillouin zone, dominantly contributes after the k-space integration to the AH conductivity. While the locally divergent Berry curvatures around Weyl points have a rather small contribution to the AH conductivity [5]. It opens a viewpoint for a relation between topology and macroscopic phenomena in noncollinear AFM. Our study might also motivate and guide further various exciting researches in associating with topology and AFM spintronics applications.

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First-principles study on the origin of structural stability of Mg-*M*-Y (*M* = Ni, Cu, Co, and Zn) alloys with long-period stacking ordered structure

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Weight reduction is one of the most important tasks for improving fuel economy and controlling CO_2 emissions of aircraft and automobiles. Therefore, it is compelling to develop light and strong materials. As a solution to this problem, the design of magnesium (Mg) based alloys has attracted much attention recently, since Mg is the lightest substance element for structural metals. Among them, a dilute Mg-based alloy with a nominal composition of $Mg_{97}Zn_1Y_2$ exhibit a remarkable high tensile yield strength and ductility. The strength of 600MPa in $Mg_{97}Zn_1Y_2$ is approximately three times larger than that of a commercial Mg based alloy called AZ31B [1]. This strength is expected to be coupled with a unique atomistic structure where a concentration of solute atoms (Zn, Y) appears on the (0001) plane in a few layers of the hexagonal close-packed (hcp) Mg matrix, and Shockley's partial dislocation (stacking fault) occurs in the solute concentrated layers [2]. The stacking sequence is relatively long along the c-axis of hcp lattice. Therefore, such structure is referred to as long-period stacking ordered (LPSO) structure. Furthermore, recent structural analysis using transmission electron microscopy (TEM) revealed that $L1_2$ type clusters of solute elements (M_6Y_8) are embedded in the Mg matrix near the stacking fault [3-5].

In this study, to clarify the microscopic origin of the phase stabilities of LPSO structures, we calculate heats of formation and electronic structure of Mg –*M*–Y alloy (*M* = Co, Ni, Cu, Zn) with different compositions and stacking sequence using first-principle calculations. In this presentation, we show how the geometry of $L1_2$ type solute clusters affects electronic structure near the Fermi level that determine the structural stabilities of LPSO phases. Furthermore, we discuss the possibility of vacancy formation in Mg-Zn-Y alloy using convex hull on the phase diagram.

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Direct and inverse magnetocaloric effects of FeRh alloy

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The magnetocaloric effect is promising for future refrigeration technology with high efficiency and environment-friendly. Temperature and entropy of magnetic material are changed with applying and removing the magnetic field. The direct magnetocaloric effect has negative isothermal magnetic entropy change, while the entropy change in the inverse magnetocaloric effect is positive. Magnetic material with a first-order phase transition has attracted much attention due to a giant isothermal magnetic entropy change and significant adiabatic temperature change. Among the magnetic materials with a first-order magnetic phase transition, FeRh alloys have peculier properties. FeRh alloys show successive antiferromagnetic (AFM) to ferromagnetic (FM) and FM to paramagnetic (PM) transitions which can be applicable for heat-assisted magnetic recording, antiferromagnetic spintronics device, and magnetocaloric effect [1,2]. In this work, we investigate the electronic structure, magnetism and magnetocaloric effect of FeRh by combining first-principles calculations and Monte Carlo simulations. The results of the magnetization curve at zero external magnetic field and isothermal entropy change (0-2 Tesla) are in good agreement with experimental works [3,4]. The isothermal entropy change (0-5 Tesla) of the inverse magnetocaloric effect (AFM-FM transition) and direct magnetocaloric effect (FM-PM transition) of FeRh alloys are reasonably predicted.

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Electronic and Topological Properties in Layered Ternary Transition Metal Chalcogenides (ABX$_4$, A and B =Ti, Zr, or Hf; X=S, Se, or Te)

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Abstract: Topological insulators (TIs) are very promising materials for new technological applications ranging from spintronics to quantum computation. Here, based on the first-principles calculations, we predict a possible new family of two-dimensional (2D) TIs in 9 ternary transition metal chalcogenides (TTMCs) ABX$_4$, where A and B = Zr, Hf or Ti and X = S, Se or Te. A total of five compounds (HfTiTe$_4$, ZrTiTe$_4$, HfTiSe$_4$, ZrTiSe$_4$ and ZrHfTe$_4$) are identified to be nontrivial under GGA functional; whereas in hybrid functional (HSE06) case, only three (ZrTiTe$_4$, ZrHfTe$_4$, and HfTiTe$_4$) remains nontrivial. The structural stabilities are confirmed by phonon spectrum analysis. The diversity of TTMCs will open up a wide possibility for tuning the bandgap and will provide a variety of new opportunities for 2D and topological material research.

First-Principles Investigations on Photoabsorption Spectra of Oxyluciferin Anions in Aqueous Solution

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The oxyluciferin anion, a well-known light emitter in firefly bioluminescence, is an important molecule for understanding the mechanism of the firefly's light emitting process. From first-principles, the environmental conditions under which the oxyluciferin anions in the excited state are generated and emit the light, are too complicated to simulate. Thus, the conventional theoretical studies have considered the environmental effects in modeled solutions or proteins and simulated only the oxyluciferin anion using quantum mechanics (QM). However, the validity of these models and the origins of detected errors have not been properly discussed to date because realistic simulations are too computationally expensive.

In this study, first-principles Born-Oppenheimer molecular dynamics (BOMD) simulations were applied to three possible oxyluciferin anions (phenolate-keto, phenolate-enol, and phenol-enolate) in a cubic box with 64 water molecules. Three long BOMD runs up to 1.4 ns were completed, corresponding to 2.9 million MD steps[1,2]. Unlike conventional simulations that use water molecules modeled by molecular mechanics (MM) or polarizable continuum mode (PCM), BOMD considers the explicit water molecules and suggests that the phenolate-enol form is the most stable isomer with the phenolate-keto and phenol-enolate isomers being less stable by 0.05 and 0.25 eV, respectively. For the three isomers, 1000 molecular structures were randomly chosen from the trajectory and the photoabsorption spectra were simulated using time-density density functional theory. Three different treatments of the water molecules (QM, MM, or PCM) and two functionals (CAM-B3LYP or B3LYP) were used. The full QM simulations with the CAM-B3LYP functional, in which the oxyluciferin anion and 64 water molecules were equally treated as QM, showed the best agreement with the available experiments. The validity of QM/MM simulations will also be discussed.

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First-principles calculation of superconductivity in hole-doped perovskite oxyhydride

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A series of oxyhdyrides $ATiO_2H$ are recently predicted as ferroelectric materials. The electronic band structures of these materials show 2D-like property and sizable contribution of the hydrogen $1s$ orbitals to the states near the Fermi level. As such interesting physical properties are expected with the hole doping. We studied the phonon property and superconductivity of these materials under the hole doping condition. To describe the Kohn-anomaly and its effect on the superconductivity accurately, we applied the Wannier interpolation scheme combined with the optimized-tetrahedron method to both the electron-phonon coupling matrix and the dynamical matrix. The hydrogen vibrational modes give the main contribution to the superconductivity, and the critical temperature reaches 10 K at the vicinity of the critical doping ratio where the hydrogen-dimerization-like distortion occurs. We found that the quartic or higher-order dispersion in electronic band structure drastically enhances phonon softening, and thus the superconductivity.

Magnetism and magneto-optical effects in bulk and multilayer CrI$_3$: An *ab-initio* study

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The magneto-optical (MO) effects not only are a powerful probe of magnetism and electronic structure of magnetic solids but also have valuable applications in high-density data-storage technology. The latest discovery of ferromagnetism in atomically thin films of semiconductors $Cr_2Ge_2Te_6$ [1] and CrI_3 [2] has unleashed numerous opportunities for fundamental physics of magnetism in two dimensional (2D) limit and also for technological applications based on 2D magnetic materials. In this contribution, we present a comprehensive theoretical study of the magnetic, electronic, optical and magneto-optical (MO) properties of multilayers (monolayer (ML), bilayer (BL) and trilayer) as well as bulk CrI_3, based on the density functional theory with the generalized gradient approximation plus on-site Coulomb repulsion scheme [3]. Interestingly, all the structures except the BL, are found to be single-spin ferromagnetic semiconductors. They all have a large out-of-plane magnetic anisotropy energy (MAE) of ~0.5 meV/Cr, in contrast to the significantly thickness-dependent MAE in multilayers of $Cr_2Ge_2Te_6$ [4]. These large MAEs suppress transverse spin fluctuations and thus stabilize long-range magnetic orders at finite temperatures down to the ML limit. They also exhibit strong MO effects with their Kerr and Faraday rotation angles being comparable to that of best-known bulk MO materials. The shape and position of the main features in the optical and MO spectra are found to be nearly thickness-independent although the magnitude of Kerr rotation angles increases monotonically with the film thickness. Magnetic transition temperatures estimated based on calculated exchange coupling parameters, calculated optical conductivity spectra, MO Kerr and Faraday rotation angles agree quite well with available experimental data [3]. The calculated MAE as well as optical and MO properties are analyzed in terms of the calculated orbital-decomposed densities of states, band state symmetries and dipole selection rules. Our findings of large out-of-plane MAEs and strong MO effects in these single-spin ferromagnetic semiconducting CrI_3 ultrathin films suggest that they will find valuable applications in semiconductor MO and spintronic nanodevices.

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First-principles study on thickness dependent electronic properties of ZrX$_2$ (X = S, Se, or Te) thin films

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Probing the effects of layer thickness and stacking on transition metal dichalcogenides (TMD) offers novel insights on their electronic properties and tunability which leads to a new avenue of research and applications[1-3]. A comprehensive first-principles study on layer-dependent stabilities and electronic properties of ZrX$_2$ (X=S, Se, or Te) thin films from 1 layer to 10 layers is performed in this study. Result shows that both ZrX$_2$ bulk and thin films adopt 1T allotrope as the most stable structures. Furthermore, 1T ZrS$_2$ and ZrSe$_2$ thin films are insulators and their band gaps decreased as the number of layers is increased to 10 layers, while 1T ZrTe$_2$ thin films and bulk are semi-metallic. Lastly, we discovered existence of van Hove singularities in unstrained 2-layer and strained 3-layer 1T ZrS$_2$ thin films. These results showcase the tunable electronic properties of two-dimensional materials due to thickness dependence and strain.

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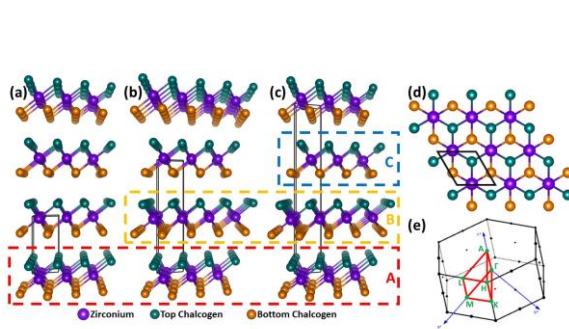[3] Villaos, R. A. B. *et al.*, *npj 2D Mater. Appl.* **3**, 1–8 (2019).

Fig. 1 Different stacking types of ZrX$_2$ with the T allotrope. **a** T-AA, **b** T-AB, and **c** T-ABC. **d** All stackings have the same Bravais unit cell and top view. **e** Hexagonal Brillouin zone with the high-symmetry points labeled.
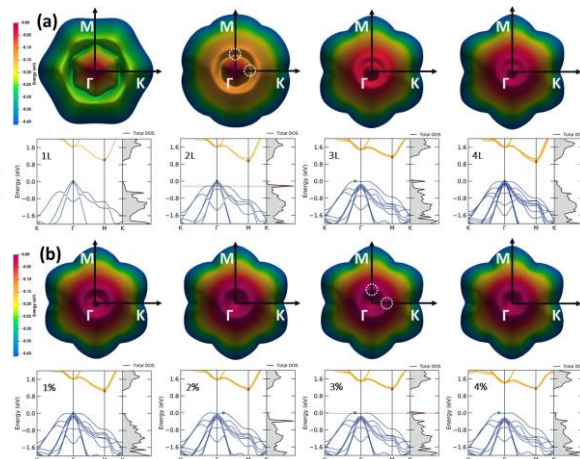

Fig. 2 Band structures, 2D contour plots of the highest occupied band, and the total DOS of **a** 1L to 4L 1T ZrS$_2$ with SOC and **b** 3L 1T ZrS$_2$ with SOC under tensile strain (1% to 4%)

The origin of high-temperature ferromagnetism in (In,Fe)Sb

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Fe-doped semiconductors have been attracting much attention as ferromagnetic semiconductors due to the possibility of high Curie temperature, fascinating functions such as low power consumption and high-speed operation, and fabrication of both *n*- and *p*-type. In fact, Fe-doped InSb not only exhibits the high Curie temperature but can also be *n*-type doping [1,2]. Our previous calculations reveal that, (In,Fe)Sb has complex magnetic properties, which are determined by the correlation between magnetic exchange coupling constants and chemical pair interactions [3]. In the isoelectronic (In,Fe)Sb case, the Fe atoms show strong antiferromagnetic interactions due to the superexchange mechanism. We have demonstrated that by modulating the chemical potentials corresponding to *n*- or *p*-type doping, the magnetic property can be changed drastically from antiferromagnetism to ferromagnetism. This transition can be well understood in terms of the Alexander-Anderson-Moriya mechanism. Moreover, ferromagnetic path containing of Fe-rich clusters occurs by the spinodal nano-decomposition and enhances the Curie temperature. However, we have obtained high Curie temperature in only *p*-type (In,Fe)Sb case. We suspect that there are the other ferromagnetic mechanisms in addition to the magnetic transition and spinodal nano-decomposition in (In,Fe)Sb. In this study, we have clarified the origin of high Curie temperature by the density functional theory calculations using the Korringa-Kohn-Rostoker Green's function method with the coherent potential approximation (KKR-CPA).

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Magnetism and Charge Densiy Wave localized on One-Dimensional MoS2 Grain Boundaries

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On the grain boundary of two-dimensional materials, localized one-dimensional (1D) electronic states can be created. By density-functional theory calculations, on the grain boundary of monolayer MoS_2, we have studied ordered phases of 1D electrons: magnetism or charge density wave. We have constructed 1D boundary structures which exhibits 1D electron bands with significantly low bandwidth. Interactions between electrons become more important due to very low bandwidth. Moreover, the system is electron doped, so that the Fermi level on the boundary levels. These low bandwidth and Fermi surface instabilities drive the system into spin or charge ordered phases. Among ordered phases, magnetism can be emerged without structural distortion, while charge density is accompanied with distortion on the boundary. Our research suggests the existence of 1D interacting electrons on transition metal dichalcogenide grain boundaries.

First-principles-based scanning Seebeck microscope simulation of epitaxial graphene on 6H-SiC

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Epitaxial graphene on 6H-SiC has been attracted attention as a promising electronic engineering candidate along with the fact that band-gap is formed due to the interaction between substrate and graphene which is a so-called buffer layer.

We recently have reported scanning Seebeck microscope (SSM) simulation of freestanding monolayer graphene in order to explain scanning thermoelectric microscope (SThM) images of monolayer graphene on 6H-SiC. The SSM simulation of freestanding monolayer graphene could reproduce the experimental thermoelectric contrast between carbon and void sites at atomic scale. More importantly, SThM for the supported graphene showed the unique oblique slope of the thermoelectric voltage between A and B sites in graphene unit cell.

Here, upgraded SSM simulations for free-standing bilayer graphene and supported monolayer graphene on 6H-SiC are numerically demonstrated. The simulations in both cases could show the opposite results for the Seebeck profile of A and B sites depending on the Fermi energy, and the oblique slope of the previous study was also able to be explained.

First-principles study on magnetism and phase stability of antiferromagnetic V_2 based Heusler alloys

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Recently, antiferromagnetic (AFM) materials have drawn a lot of attention in spintronics field because of possible device applications. Anisotropic magnetoresistance (AMR) and spin-orbit torque (SOT) can open the possibility for realizing AFM devices based on microelectronic designs. Although both SOT and AMR have been observed for a ferromagnetic (FM) Heusler alloy NiMnSb [1], AFM Heusler alloys are expected to reveal the same phenomena as the ferromagnetic one. However, almost all AFM Heusler alloys have relatively moderate Néel temperatures ($Ru_2MnGe:T_N$~300 K) compared to FM Heuser alloys with high Curie temperatures ($NiMnSb:T_C$~700 K). Quite recently V_3Al has been found to be an AFM spin-gapless semiconductor with significantly high T_N (~600 K) [2]. Therefore, V_2 based, namely V_2YZ, AFM Heusler alloys have potentials for room-temperature AFM spintronics device applications. In this work, we investigate the magnetism and phase stability of AFM V_2 based Heusler alloys by using first-principles calculations. First, we find that V_2YAl (Y=V, Nb, and Ta) have higher phase stability than V_2YSi (Y=Ti, Zr, and Hf) from an cluster-expansion analysis. Calculated T_N of V_2TaAl, V_2TiSi, and V_2HfSi are much higher than V_3Al. In the case of V_2TaAl, the high T_N originates from the larger lattice constant than that of V_3Al. The expanded lattice constant enhances the localized character of the magnetic atoms. From the distance dependence of the magnetic exchange interaction, the first-nearest and third-nearest pairs have AFM interactions, and the second-nearest interaction is ferromagnetic, totally stabilizing a G-type AFM ordering. Finally, we discuss the origin of the strong AFM coupling between the neighboring V sites by using rigid-band models.

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First-principles magnetic dipole-dipole energy calculation: Application to magnetic chain, film, and bulk systems

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We have developed a computational method to evaluate magnetic dipole-dipole energy based on density functional theory [1-2]. In general, magnetic dipole-dipole interaction between nearby magnetic moments is much weaker than other magnetic interactions; direct-, indirect-, super-, and double-exchange interactions. However, due to the long-range property of magnetic dipole-dipole interaction, it is one of the important factors for determining the magnetization direction of magnets as well as the spin-orbit interaction. Furthermore, it may also contribute to emergences of peculiar non-collinear magnetism, such as magnetic skyrmions, multiferroic polarization, etc. Up to the present, magnetic dipole-dipole interaction has been used to be estimated by the configuration of atomic magnetic moments. However, in the case where there is a large spatial anisotropy in the electron spin density, reliability of such treatment remains uncertain.

In our approach, the spin density predicted by density functional theory is employed to estimate magnetic dipole-dipole energy. Although the contribution from magnetic dipole-dipole interaction to Kohn-Sham potential might be small, it may allow us to access useful properties; the electronic structure and spin density change due to magnetic dipole-dipole interaction, the optimization of local magnetic moment direction, and more. For these reasons, we developed a self-consistent scheme with magnetic dipole-dipole interaction [1]. The magnetic dipole-dipole interaction is useful for qualitative analysis of the stability of magnetic anisotropy. Due to the long-range interaction, it is much better to adopt an appropriate periodic boundary condition (one-, two-, and three-dimension) in the computation of magnetic dipole-dipole interaction.

We calculated magnetic dipole-dipole energy in magnetic chain, film, and bulk materials. The elements of Fe, Ni, and Co are used as ferromagnetic materials. The results of two-dimensional magnetic material (CrI_3) will also be shown. We found that magnetic dipole-dipole energy depends on the distribution of spin density. For example, the quadrupole component of spin density appears in film systems, and it leads to enhancement or suppression of magnetic dipole-dipole energy. This result indicates that the previous approach may not be appropriate for precise quantitative discussion of magnetic anisotropy energy. Moreover, our method can be employed in antiferromagnetic materials [2]. In the presentation, we will explain the method and discuss its application results.

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Spin-wave dispersion of 3d ferromagnets based on quasiparticle self-consistent *GW* calculations

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There are three methods for calculating spin-wave (SW) dispersion: Lichtenstein formula (LF)[1], frozen magnon method (FMM)[2], and linear response (LR) method[3]. So far, most of the calculations have been performed based on the ground states determined by the local density approximation (LDA) or the generalized gradient approximation (GGA). In fcc Ni, however, the calculated stiffness constant D of SW dispersion is almost two times larger than the experiment. In hcp Co, the LDA also overestimates the SW energy. Thus, the LDA can not reproduce the optical magnon, which is observed by the experiment.

In this study, we calculate the transverse susceptibility $R(\boldsymbol{q},\omega)$ in the LR based on the ground states determined within the quasiparticle self-consistent *GW* (QSGW) method. We calculate SW dispersions in bcc Fe, hcp Co, fcc Ni, and B2-type FeCo. In the QSGW, D for Fe, Co, and Ni agree with experiments. For Co, Fig. 1 shows that both acoustic and optical branches agree with experiment[4]. As for FeCo, we have some discrepancies between the D in QSGW and that in the experiment. There may need further theoretical and experimental investigations on the discrepancies.
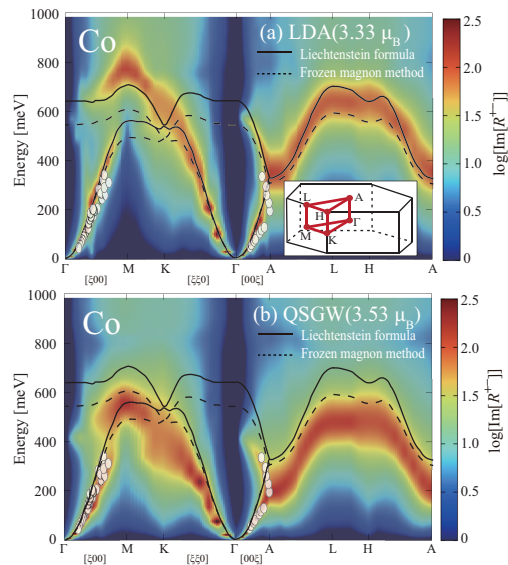

Fig.1: SW dispersions for Co in LDA (a) and in QSGW (b)[4]. The LF (bold line) and the FMM (broken line), experimental data (circle) are also shown. The inset shows the Brillouin zone for hcp Co and its symmetry lines.

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A Novel Method of Removing Defects from α-T boron

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Boron crystals have very complicated structures. They have many allotropes and all the structures have intrinsically disordered structures. Deviation from stoichiometry and the presence of partially occupied interstitial atoms are the common features of boron crystals, and it is very difficult to remove these intrinsic defects. Among allotropes of boron, the tetragonal (T) phase is a confusing phase. Theoretically, α-T boron with B composition 52 (α-T-B_{52}) is predicted [1]. Experimentally, several HPHT syntheses of α-T-B_{52} were reported. In spite of similar conditions of preparations, all the reported structures are more or less different. A theoretical study by Uemura *et al* has identified the characteristic features of pure α-T-B_{52} [2]. Their study demonstrates that α-T-B_{52} certainly exists at high pressures and high temperatures. The crystal has interstitial atoms, which randomly occupy c sites. This disorder is thought to be unavoidable to α-T boron. Their result well describes experimental observations. Recently, yet another one has been discovered by Ekimov *et al* [3]. They obtained α-T boron by thermal decomposition of decaborane by HPHT method. The obtained crystal is actually hydrogenated variant of α-T boron: α-T-$B_{52}H_m$ with the estimated H content $m=7.7$. After recovering samples from the high pressure, on annealing at $T_{a2}=700°C$, hydrogen was completely released, and, to our surprise, the α-T boron was transformed to a new phase of orthorhombic lattice. We term this as δ orthorhombic phase, δ-O-B_{52}. The structure of δ-O-B_{52} is very similar to that of α-T-B_{52}, but occupation of c sites is not random. This is a sharp contrast to our understanding of boron crystals: disorder is an intrinsic property of boron.

A number of interesting questions are aroused.
1) Why did only Ekimov's method create δ-O-B_{52} during a long history of research?
2) Why did hydrogen release show two-stepwise transition?
3) Why did hydrogen release associate a phase transition to δ-O-B_{52}.
4) Why were randomly distributed interstitial atoms in order on hydrogen release?

A quick answer to these questions is that δ-O-B_{52} is an ordered state of α-T-B_{52}, and is the low-temperature variant of the latter type. Our calculation shows that the transition temperature is 1260K. However, the extremely low mobility of atoms hinders the transition to the low-temperature phase. Here, hydrogen takes a role. There is another phase transition from hydrogenated boron to pure boron at $T=900K$. Hydrogen diffusion assists migration of interstitial boron atoms. The removing intrinsic defects is a consequence of an ingenious cooperative phase transition. This mechanism can be utilized for other materials, which are difficult to dope.

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O₂-release and Structure Evolution of Layered Li$_{1.2}$$M$$_{0.4}$$M'$$_{0.4}O_2$ (M, M' = Cr, Mn, Ti) Cathodes for Li-Ion Batteries: First-Principles Calculations

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Large-scale energy storage systems are vital to spread future electric vehicles and renewable energy power plants, and Li-ion batteries have been extensively developed due to their noteworthy battery characteristics. Therefore, alternative cathode materials have been explored because the layered LiCoO$_2$ cathode, which is a commercially successful and most polupar cathode oxide, includes rare- and highly toxicic-metal Co.

Recently, cation-disordered rock-salt Li$M$$_{0.5}Ti_{0.5}O_2$ (M = transition metals) cathodes, in which cations are romdomly disstributed in the MO$_6$ octahedral geometry, have been studied, and O-redox reaction has been reported theoretically [1,2]. However, experimental discharge capcities have not achived to theoretical ones so far, and O$_2$-release has been confirmed during charge and discharge reaction processes even though O atoms are expected to be hardly released from the MO$_6$ framework. The theoretical study has shown that such a tendency of easier O$_2$-release is strongly correlated with hybridization between M-d and O-p states, and LiCr$_{0.5}$Ti$_{0.5}$O$_2$ may be the best candidate among this series for suppressing the O-redox reaction and O$_2$-release.

In this study, we have theoretically investigated layered Li$_{1.2}$$M$$_{0.4}$$M'$$_{0.4}O_2$ (M, M' = Cr, Mn, Ti) to improve rechargeable capacities. Cr-oxide cathode are expected to show less covalency between Cr-d and O-p states, and layered structures show high Li percolation. First-principles calculations are performed with supercell models, and geometric and electronic structure changes associated with Li-insertion and desorption are investigated to claryfy microscopic reaction mechanisms in the cathodes. The computations are carried out with Quantum ESPRESSO [3] within PAW method under GGA+U. Optimized structures show that Cr atoms migrate from original octahedral sites to tetrahedral sites during the reaction processes because of no-ligand electronic structure of Cr^{6+}, and the Cr^{3+}/Cr^{6+} redox reaction predominately contributes to the charge compensations in the cathodes. The O-redox reaction and O$_2$-release can be remarkably suppressed in the Cr systems compaed to the Mn system.

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A First Principle Study on Magneto-Optical effects and Magnetism in $Y_3Fe_5O_{12}$ and $Bi_3Fe_5O_{12}$

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The magneto-optical (MO) effects not only are a powerful probe of magnetism and electronic structure of magnetic solids but also have valuable applications in high-density data-storage technology. Yttrium iron garnet ($Y_3Fe_5O_{12}$) (YIG) and bismuth iron garnet ($Bi_3Fe_5O_{12}$) (BIG) are two widely used magnetic semiconductors with strong magneto-optical effects and have also attracted the attention of physics society for fundamental physics studies. In particular, YIG has been routinely used as a spin current injector. Perhaps because of their complex structures with 80 atoms per unit cell, to date no *ab-initio* theoretical calculation has been carried out to studty the magneto-optical (MO) effects in these systems. In this contribution, we present a thorough theoretical investigation on magnetism, electronic, optical and MO properties of YIG and BIG, based on the density functional theory with the generalized gradient approximation plus onsite Coulomb repulsion scheme. We find that both semiconductors exhibit large MO effects with their Kerr and Faraday rotation angles being comparable to that of best-known bulk MO materials, suggesting that they will find valuable applications in semiconductor MO nanodevices. The calculated optical conductivity spectra, MO Kerr and Faraday rotation angles agree quite well with available experimental data. We analyse the MO effects in both systems through calculating the contribution to the optical conductivity at Γpoint in terms of the band state symmetries and dipole selection rules.

First-principles study of two-dimensional higher-order topology in monolayer graphdiyne

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Based on first-principles calculations and tight-binding model analysis, we propose monolayer graphdiyne as a candidate material for a two-dimensional higher-order topological insulator protected by inversion symmetry. We show that, despite the absence of chiral symmetry, the higher-order topology of monolayer graphdiyne is manifested in the filling anomaly and charge accumulation at two corners. Although its low energy band structure can be properly described by the effective tight-bind Hamiltonian constructed by using only the p_z orbital of each atom, the corresponding bulk band topology is trivial. The nontrivial bulk topological invariant can be correctly captured only when the contribution from the core levels derived from $p_{x,y}$ and s orbitals are included, which is further confirmed by the Wilson loop and the nested Wilson loop calculations. We also show that higher order topology of a monolayer graphdyine is the fundamental origin of the nontrivial band topology of the corresponding three dimensional material, ABC-stacked graphdiyne, which hosts monopole nodal lines and hinge states.

Understanding Ion Migration in Metal Halide Perovskites

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Metal halide perovskites have received extensive interest due to their exceptional photovoltaic performance. In addition to their semiconductivity, it was reported that halide ions in the halide perovskite are highly mobile, [1] and this gives negative effect on the stability of perovskite solar cells.[2] On the other hand, using high ion conductivity, new applications of these halide perovskite is possible, including memristors which can imitate biological synapses.[3] Herein, control the rate of ionic conductivity is becomes a key point. In most commonly studied halide perovskites, $CH_3NH_3PbI_3$, ion migration would be mediated by vacancies.[4]

During device operation, various physical stimuli such as light, heat, pressure and applied bias can affect movement of ions. However, their impact on each are still poorly understood. Using first-principles density functional theory (DFT) calculations, we simulate the ion migration mediated by vacancies changing conditions in electrical, chemical and mechanical ways. We quantify the activation energies of ions migration and how these device-relevant conditions influence the path and the energy barrier of ion migration.

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Curvature-driven atomic localization and dipole alignment of quantum emitters in h-BN

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Hexagonal boron nitride (h-BN) has recently emerged as a promising materials platform for developing various solid-state quantum technologies owing to its attractive electronic, thermal, and optical properties [1]. In particular, a number of color centers in h-BN have been found to be stable and bright single-photon-emitters (SPEs) operating at room temperature, which are crucial elements for quantum information processing and quantum communication [2]. In this talk, we combine first-principles theory and experiment to investigate the intimate relation between curvature in h-BN and atomic localization of SPEs. We use density functional theory to calculate the energetically stable configuration of various defect models of the SPEs in a buckled h-BN plane with different curvatures. We show that the vacancy-derived point defects in h-BN prefer to form in the highest-curvature area of the buckle and we find that the high curvature induces a dimer reconstruction for the atoms surrounding vacancy. Our result provides not only a microscopic understanding on the recent experimental observations of the SPEs being located in h-BN buckles [3], but also strongly suggests that the atomic origin of the SPEs may be vacancy-derived. We also discuss several key features of the SPEs formed on h-BN buckle such as dipole orientation, which would be helpful to design future experiments of the SPEs in h-BN.

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A Strategy for Finding Better Oxygen-ion Conductors beyond Database

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Recently, combinations of first principles calculations and machine learning (ML) techniques have been widely used for new material designs due to their efficiency. The prediction model for a demanded property constructed by the ML is applied to the materials in the database, then good candidates are suggested according to their predicted properties by the screening. This strategy is powerful to rediscover an unknown functional property of the previous materials that have been already registered in the database such as Inorganic Crystal Structure Database (ICSD) [1]. However, there is a shortcoming that this strategy is restricted in searching already existing materials.

"New" materials can exist beyond the database. In this study, we suggest two strategies for expanding the candidate (derivate) materials beyond the database. As the demanded target functional materials, the oxygen-ion conductors with high oxygen-ion conductivity (σ_O) were chosen. For constructing derivate materials from a good candidate suggested in the database, chemical elements or crystal structures can be changed. For changing the constituting chemical elements, a total ion exchange was used with isovalent elements [2,3]. For changing the crystal structures, a combination of evolutionary algorithm and ML was used [4]. Then, for new candidates in the expanded database, we performed various first-principles validations for the σ_O. In the result, new materials with σ_O higher than the material in the original database were proposed.

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First-principles Study of Superconducting Al-Zn-Mg Quasi-crystals: Comparison between 1/1 and 2/1 Approximants

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Recently, the bulk superconductivity at a transition temperature $T_c \sim 0.05$K has been discovered by Kamiya *et al*. in the Al-Zn-Mg quasicrystal and its approximant [1] and has attracted much attention for its symmetry and pairing mechanism. As for the theoretical study, the $Al_{30}Mg_{40}Zn_{30}$ 1/1 approximant was previously studied on the basis of the first-principles calculation and the origin of the pseudo gap near the Fermi level was discussed [2]. However, the electronic state with the composition ratio corresponding to the superconducting compound [1] was not discussed there. Then, we perform the first-principles calculations using OpenMX for the Al-Zn-Mg 1/1 and 2/1 approximants with various composition ratios including the superconducting one and find that there are significant differences in the energy bands (see Figs. 1 (a) and (b)) and the density of states (not shown) near the Fermi level. On the basis of the obtained electronic states, we discuss the features of physical quantities and superconductivity.
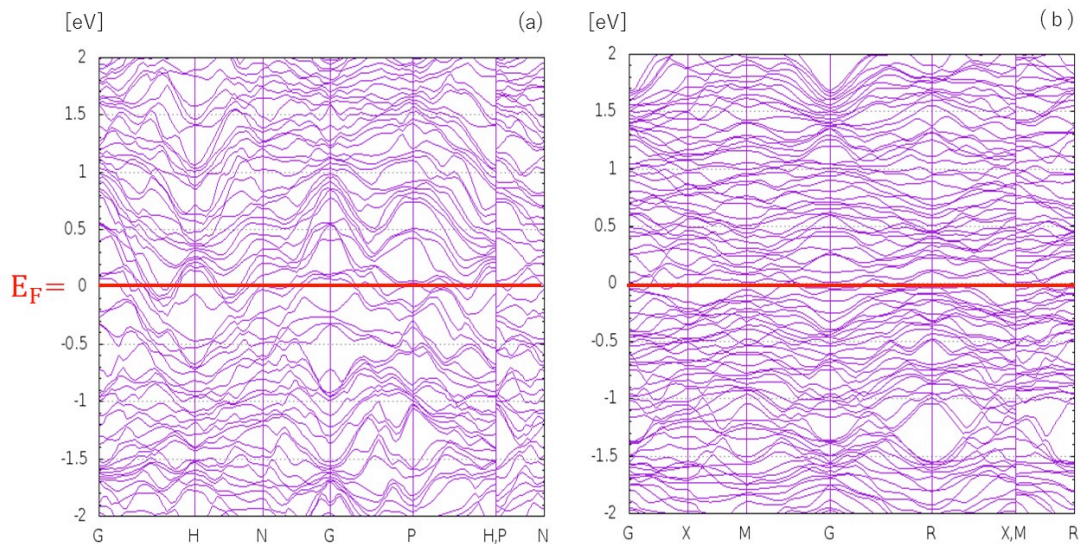
Fig. 1. Energy band dispersions of Al-Zn-Mg 1/1 (a) and 2/1 approximants (b) with the composition ratio corresponding to the superconducting compounds.

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Accessing the Accuracy of Density Functional Theory through Structure and Dynamics of the Water–Air Interface

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Density functional theory-based molecular dynamics simulations are increasingly being used for simulating aqueous interfaces. Nonetheless, the choice of the appropriate density functional, critically affecting the outcome of the simulation, has remained arbitrary. Here, we assess the performance of various exchange–correlation (XC) functionals, based on the metrics relevant to sum-frequency generation spectroscopy. The structure and dynamics of water at the water–air interface are governed by heterogeneous intermolecular interactions, thereby providing a critical benchmark for XC functionals. We find that the XC functionals constrained by exact functional conditions (revPBE and revPBE0) with the dispersion correction show excellent performance. The poor performance of the empirically optimized density functional (M06-L) indicates the importance of satisfying the exact functional condition. Understanding the performance of different XC functionals can aid in resolving the controversial interpretation of the interfacial water structure and direct the design of novel, improved XC functionals better suited to describing the heterogeneous interactions in condensed phases.

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Control of Magnetic Interactions in Eu-doped GaN

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Toward information technology, we invetsigate magnetic semiconductors with electronic structures suitable for controlling magnetism by gate-voltage and/or charge injection from a theoretical viewpoint. Such electronic structures might be provided with magnetic states obtained by doping impurities into the semiconductors and internal nanostructures induced by nano-spinodal decomposition, which is thermodynamic phase separation at the nanoscale.

Magnetic semiconductors that possessing high-spin and low-spin states as stable electronic states may be favorable to switching between magnetic stable structures by an external field and/or charge injection. This provides a possibility for use as an element of a nonvolatile random access memory or a high-speed arithmetic element. In addition, by utilizing the nanostructure in the crystal, not only the enhancement of the magnetization characteristics but also the possibility as a quantum computer or a reservoir computing element expected as a non-Neumann computer can be considered.

Eu-doped GaN, which is a gallium nitride (GaN) crystal doped with the rare earth element europium (Eu) is known as an active layer material of red light emitting diodes, but also exhibits behavior as room temperature ferromagnetism. We simulated the electronic properties of the virtual sample built in the computer and the magnetic properties of the nanostructure using a DFT calculation package, Akai-KKR. [1,2] The simulation results suggest that this system changes the dominant magnetic interaction depending on the concentration of the impurity element, and has multiple stable magnetic structures. It is also shown that the magnetic properties of paramagnetic, superparamagnetic, and ferromagnetism differ depending on the nanostructure in the crystal. For compositions near the boundaries of the different magnetic orders, the magnetic transition is also reproduced by charge variations. In this presentation, magnetic semiconductor crystals that can be controlled by gate voltage and/or charge injection will be considered.

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Investigation of Interface Structures of Au(111)/Li$_3$PO$_4$ using High-Dimensional Neural Network Potential

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Recently, the construction of interatomic potentials using first-principles calculation data and machine-learning technique has been widely tried because of higher reliability and low computational costs. Our group previously constructed the interatomic potentials of amorphous-Li$_3$PO$_4$ using high-dimensional neural network (HDNN) [1], and showed that the calculated Li-ion conductivites agree well with experimental data [2]. In the present study, we have tried to extend the application range of the HDNN potential (HDNNP) to metal/solid electrolyte interface systems. The understanding of the interface structures and Li-ion distribution near the Au/Li$_3$PO$_4$ interface is of significance for the development of all-solid-state Li-ion batteries and novel memory devices [3].

The construction of NNP for Au/Li$_3$PO$_4$ is challenging because this is a four-element system which involves a large number of input structures and parameters. We used density functional theory (DFT) calculations to construct the dataset, which includes bulk and surface structures of Au and Li$_3$PO$_4$, and interface structures of Au(111)/Li$_3$PO$_4$. We optimized the NNP using both total energies and atomic forces obtained by DFT calculations. The root-mean-square-errors of the predicted total energies and atomic forces were 18.9 meV/atom and 0.301 eV/Å, respectively. Using the constructed NNP, we then performed structure optimization of the Au(111)/Li$_3$PO$_4$ interface system containing 9,840 atoms, where the lattice mismatch of Au and Li$_3$PO$_4$ is as small as ca. 1.6%. In the meeting, we will discuss the calculated interface structures and comparison with the DFT results.

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Bulk Rashba Effect in Ferroelectric Transtion-Metal Oxides

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Perovskite transition-metal oxides ABO_3 show versatile physical properties such as magnetism, superconductivity, ferroelectricity, and ionic conduction. In multiferroic systems, one can combine more than two ferroic order parameters, for example, ferroelectricity and ferromagnetism. When non-magnetic materials have non-centrosymmetric crystal structure and strong spin-orbit coupling, the spin degeneracy of bandstructure is removed and Rashba or Dresselhaus effect can emerge. The induced spin polarizaton in the momentum space has appealing potential for spin-FET applications. When the Rashba spin splitting extends over two valleys located at the Brillouin-zone corners and results in two non-degenerate spin states, it is called 'spin-valley coupling'. By using a first-principles approach, we study 'bulk spin-valley coupling' in newly designed perovskite oxide multilayers. Aiming at a sizable spin splitting, low-spin *5d* transition metal ions are embedded in ferroelectric $BiAlO_3$ host, showing a polar $LiNbO_3$-type structure. The ferroelectric structural distortion indeed induces spin-valley coupled properties [1]. By tuning the atomic number of the B site element in the perovskite-structure, we also show that the polar distortion is also responsible for a topological transition from a quantum spin-Hall insulating phase to a trivial band insulator [2].

Here we are taking a step further by considering a multifunctional oxide which shows both ferroelectric and magnetic spontaneous orders as a playground for bulk Rashba effect. In systems which are both ferroelectric and ferromagnetic, the on-site exchange interaction largely splits the bands so that up- and down-spin bands become unpaired. On the other hand, in ferroelectric and antiferromagnetic systems, there is a spin-flipping symmetry operation (such as a mirror symmetry operation) which behaves as time-reversal symmetry and makes the bands cross at the Γ point. In this case, magnetic bulk Rashba effect can occur. In this presentation, the electronic structure, the spin texture, and its correlation with spin canting in magnetic transition-metal oxides will be discussed.

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Topological crystalline insulator: from symmetry indicators to material discovery

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Topological crystalline insulators (TCIs) are insulating electronic phases of matter with nontrivial topology originating from crystalline symmetries. Recent theoretical advances have proposed new TCI states protected by rotational symmetries. Distinct from topological insulators, rotational symmetry-protected TCIs are expected to show unique topologically protected boundary modes: First, the surface normal to the rotational axis features "unpinned" Dirac surface states whose Dirac points are located at generic k points. Second, due to the "higher-order" bulk boundary correspondence, a 3D TCI also supports 1D helical edge states. Despite the unique topological electronic properties, to date, rotational symmetry-protected TCIs remain elusive in real materials. Building upon recent theoretical works, we demonstrate a feasible method to identify new TCI states based on first-principles calculations. We systematically unveil the topological properties of the TCI states in Ca_2As. On both top and side surfaces, we observe topological surface states protected independently by rotational and mirror symmetries [1]. In addition, we identify the van der Waals material α-Bi_4Br_4 and the pristine bulk bismuth are TCI purely protected by rotation symmetry [2,3], even through these two materials have been long thought to be topologically trivial. Our work reveals rich and exotic TCI physics. Such a method can be broadly applied in searching for new TCIs.

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The Oxidative Coupling of Methane Catalyzed by MgO; A First-Principle Based Microkinetics and Ab-initio Molecular Dynamics Study

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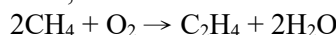
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1. Introduction

The oxidative coupling of methane (OCM) has long been studied to provide an efficient way to convert natural gas into high-value chemicals. For the full understanding of the reaction mechanism of OCM, we carried out density functional theory (DFT) calculations combined with microkinetics and chemical reactor modeling, because this approach enables the activity and selectivity prediction from atomic or molecular level simulations.

2. Theoretical method

We investigated the OCM reaction,

$$2CH_4 + O_2 \rightarrow C_2H_4 + 2H_2O$$

catalyzed by Li-doped MgO, found by Lunsford et al.[1] The present reaction model includes 109 gas-phase and 54 surface elementary reactions. Using DFT, reaction energy (ΔE) was calculated and activation barrier (E_a) were evaluated from the calculated ΔE values using Polanyi rule. For the DFT calculation, VASP 5.4 program package was used. We used spin polarized DFT method using RPBE functional. From ΔE and E_a values, we evaluated the reaction rate constants and constructed reaction rate equations. Composition of inlet and outlet gas was calculated using the transient continously stirred tank model.

3. Results and Discussions

In Figure 1, the conversion of CH_4 and the composition of C-containing compouns were plotted at reaction temperatures from 700 to 1000 ^{o}C. With the increase of temperature, the CH_4 conversion becomes higher but the composition of C_2 compounds i.e. C_2H_4 or C_2H_6 decreases because combustion compounds CO and CO_2 increases. This agrees with experimental tendency.[1,2] Our study has sucessfully shown that the DFT combined with microkinetics and reactor simulation correctly reproduces the activity and selectivity tendency of the OCM catalyst, which is important property to enable the computational catalyst screening.
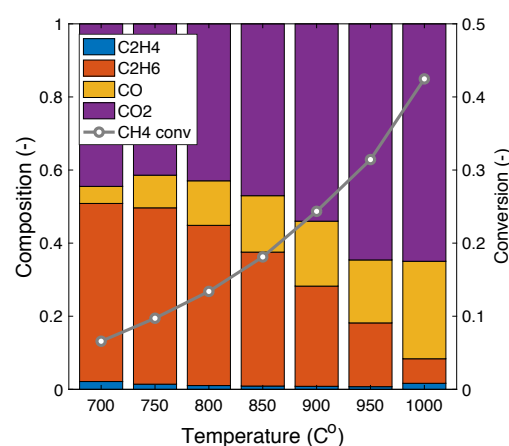

Figure 1. The CH_4 conversion (right axis) and the composton of carbon containing compounds (left axis) were plotted at reaction temperature of 700-1000 ^{o}C.

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First-principles theory of Cr-vacancy in BaZrO₃ as a solid-state qubit candidate

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Recently, remarkable advances have been made in the development of solid-state quantum bits (qubits), which are the basic hardware units of quantum information processing [1]. One of the leading solid-state qubit platforms is the nitrogen-vacancy (NV) center in diamond [2,3]. Furthermore, a significant interest has been emerging in the literature to develop such defect-based qubits in diverse wide-gap semiconductors for broadening the scope of the solid-state quantum information. In this study, we explore a Cr-O vacancy complex in BaZrO₃ as a potential solid-state qubit candidate. We use first-principles density functional theory to examine the stability and the electronic and spin properties of the Cr-O vacancy pair in BaZrO₃. To investigate the stability of the defect in various charge states, we calculate the defect formation energy of Cr-vacancy in BaZrO₃. In addition, we use HSE06 hybrid functionals [4] to accurately calculate the defect level diagram and the zero-phonon line of the Cr-O vacancy. In the poster, we also discuss the recent progress and challenges in computational design of new defect qubits in complex wide-gap materials.

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Atomistic Study of Electronic Structure and Electron-Phonon Coupling in Twisted Graphene Layers

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Twisted graphene layers have become new experimental platform for exploring correlated electron physics with various tunable degrees of freedom as demonstrated by the recent observations of superconductivity and correlated insulating states in magic-angle twisted bilayer graphene (MA-TBG) and twisted double bilayer graphene (TDBG). As a starting point to understand the rich physics emerging in these moiré superlattices, it is crucial to determine accurate atomic and electronic structures as well as their interplays. In this work, we investigate electronic structure and electron-phonon coupling in twisted graphene layers starting from an atomistic description of the system. Electronic structure, phonon spectrum, and electron-phonon coupling strength λ are obtained before and after atomic-position relaxation both in- and out-of-plane. We find that the lattice relaxation strongly affects the low-energy electronic structure, and λ is very large for the low-energy flat bands owing to the enhanced density of states. Furthermore, we show that the layer-number degree of freedom offers additional tunability to the low-energy electronic structure through the sensitivity to the external electric fields.

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First-Principles Calculation of DNA Energy Band Responsible for Superconductivity

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In 2001, Kasumov and coworkers observed proximity-induced superconductivity in a double-stranded λ-DNA below 1K [1], where the resistance shows the power low temperature dependence as expected in the Tomonaga-Luttinger liquid in the on-dimensional electron systems. However, most of the experimental results except for the Kasumov's report show that the DNA is an insulter with a finite band gap and/or with the Anderson localization due to randomness [2]. Theoretically, Artacho and coworkers studied the energy band of DNA on the basis of the first-principles calculation using SIESTA and found that an infinite poly-guanine - poly-cytosine DNA molecule (poly-(GC)) is the insulator with the band gap Δ=2.0eV [3]. In addition to the periodic poly-(GC), they also discussed the ragged and the swapped structures which show relatively small band gaps as compared with the poly-(GC). The other periodic structures, however, were not discussed there. The purpose of this paper is to investigate the energy band of DNA with various types of periodic structures on the basis of the first-principles calculation and to explore the periodic structures of DNA responsible for the metallic and the superconducting behaviors.

We carry out the first-principles calculation using OpenMX for the periodic structures of DNA, where the unit cell includes ten base-pair and 650 atoms. The geometry is initially set to be an α-helix structure and then relaxed by means of ab initio linear-scaling DFT. The obtained energy gap of the poly-(GC) is Δ=3.15eV which is considerably larger than the previous result [3] due to the recent improvement of the first-principles calculation. Remarkably, a tiny band gap Δ=0.003eV is observed in the periodic poly(GC-CG) structure where the base-pairs of guanine-cytosine and cytosine-guanine stacked alternatingly. In addition, the obtained band dispersions of the both LUMO and HOMO bands are double-well types which are responsible for the superconducting behavior based on the Tomonaga-Luttinger liquid [4]. The energy bands for the other periodic structures are also examined.

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Oxidation of Diamond (100) Surface Studied Using Density Functional Theory

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The extreme properties of diamond such as high breakdown field, high charge carrier mobilitiy, high thermal conductivity, and room temperature UV exciton emission and optical defect centers make it suitable for the next generation of power electronics and quantum technologies [1]. A major obstacle in developing diamond devices is the difficulty of growing single crystal wafers of sufficient size [2]. Molecular level understanding of the fundamental diamond surface reactions may lead to insights that will improve the current diamond processing. In this work, the oxidation of diamond (100) surface is studied using van der Waals-corrected density functional theory calculations. For single O_2 adsorption on reconstructed surface, dissociative adsorption on the bridge site is energetically favorable compared to the molecular adsorption on the top site, with adsorption energies of −4.32 eV and −2.71 eV, respectively. With a small energy barrier of 0.21 eV, O_2 on the top site can dissociate and lead to surface dereconstruction, enhancing the adsorption energy to −5.09 eV. Intersystem crossing from triplet O_2 to singlet O_2 occurs as a result of initial metastable adsorption and molecular vibrations at finite temperature. The monolayer adsorption energies per O atom on dereconstructed surface for bridge and top site are −5.33 eV and −4.49 eV, respectively. However, vibrational mode analysis shows that monolayer adsorption on the top site is unstable. Desorption of CO is initiated by breaking C-O bond on an ether group, followed by severing of two C-C bonds with subsurface atoms and a C-O bond from the adjacent ether group. The surface defect will cause the O atom to migrate to the top of site of adjacent C atom forming ketone. Desorption from the top site requires lesser activation energy, while further desorption from the bridge site will lead to reconstruction of the second layer. The desorption of CO from two oxygen adsorption sites agrees with the existence of multiple desorption peaks reported in experiments [3,4].

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Hydrogen Induced Caesium Desorption from Caesium-Decorated Tungsten(110) Surface

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We studied the influence of H adsorption on the stability of Cs on W(110) by performing density functional theory (DFT) based calculations. We also examined the adsorption of H and Cs on clean W(110) for reference. We found that the average adsorption energy of Cs decreases when H is present on the surface. Also, H adsorption is weaker in the co-adsorbed case, as compared to its adsorption on the clean W surface. We analyzed the electronic properties of the system to understand the obtained trend in the adsorption energies. With co-adsorbed H and Cs, the charge density difference profiles reveal that W atoms interact both with H and Cs. This leads to a weaker interaction between H and W, and between Cs and W. Higher coverage of H further weakens the adsorption of Cs on the surface. These results suggest that the presence of H could induce Cs desorption from the surface [1]. It is therefore important to reconsider the design of Caesiated metal surfaces [2], which are usually employed as electrodes in the production of negative hydrogen ions [3].

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Thermoelectric Properties of Nanoribbons of Phosphorene, Arsenene, Antimonene, and Bismuthene from Non-Equilibrium Green's Function Calculations

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Two-dimensional (2D) atomic layer materials have been attracting attention as flexible thermoelectric conversion elements that are suitable for waste heat recovery [1]. Recent theoretical studies based on the first-principles calculations [2-4] reported that the 2D pnictogens, i.e., phosphorene, arsenene, antimonene, and bismuthene, can be good thermoelectric materials that exhibit high power factor and figure of merit (ZT values) by controlling their chemical potentials appropriately.

In this study, we explore the possibility to further improve the thermoelectric performance of these 2D pnictogens by reducing their dimensions, i.e., making their nanoribbon structures. It was theoretically predicted by Saito and Dresselhaus et al. that the power factor of the low-dimensional materials can be enhanced by reducing their confinement length [5]. In fact, Liu et al. reported that the thermoelectric properties of phosphorene will be improved by making their nanoribbon structures with the appropriate width and cutting direction on the basis of first-principles calculations [6].

Here we calculated the power factor of H-passivated puckered armchair nanoribbons of phosphorene, arsenene, antimonene, and bismuthene using the non-equilibrium Green's function method. We fixed the ribbon length and studied the dependence of power factor on the ribbon width. It was found that Seebeck coeffcents of these pnictogen nanoribbons increase with reducing their width, while the conductances decrease. As a result, the power factors exhibit complex dependence on the ribbon width. The power factor properties also depend on the type of doping (n- or p-type). It was found that the nanoribbons of arsenene, antimonene, and bismuthene exhibits higher power factor than the phosphorene nanoribbon. The physical origin of these properties are discussed in the presentation.

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Spin-Dependent O_2 Binding to Hemoglobin

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Abstract

Oxygen adsorption on a metal active site is mediated by spin transitions. Specifically, the spin magnetic moment of the system affects the activation barrier for the reaction to proceed. Thus, consideration of spin states in adsorption is of significance. An important metal-oxygen reaction is the adsorption of O_2 to hemoglobin. In this case, we investigate the spin transition of O_2-Fe complex as it proceeds from deoxyhemoglobin to oxyhemoglobin state. We show that its reaction involves a transition from triplet to quintet to singlet spin state [1].

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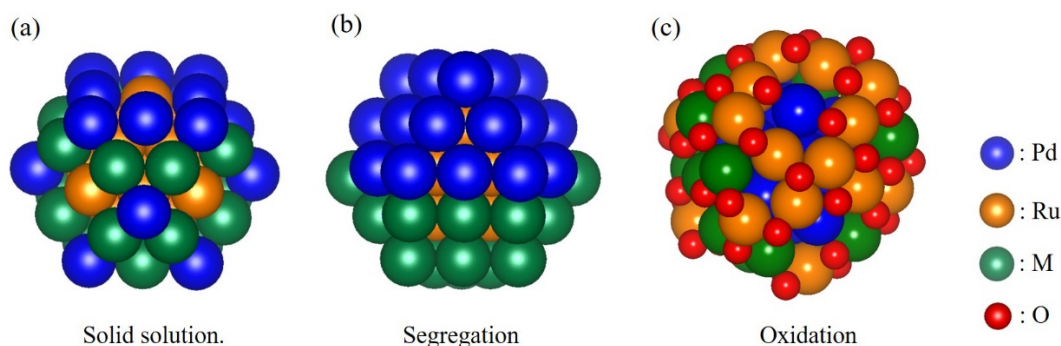# Structural investigation on ternary PdRuM (M=Pt, Rh or Ir) nanoparticles using first-principles calculations

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PdRu NPs have been studied as a catalysis for NO_x conversion with hcp and fcc coexisting phase. [1,2] However, the bimetallic NPs shows instability in higher working temperature (T>1100 K) due to the evaporation of Ru atoms. Therefore, Pt, Rh or Ir atoms are added into PdRu NPs for reinforcement the stability. For understanding the atomic arrangement of these ternary NPs (PdRuPt, PdRuRh and PdRuIr), we have utilized three different morphologies (hcp, tOh and Ih) for modeling. By using Monte Carlo sampling, we have found the Ru atoms form a core in the ternary NPs. Pd-M solid solution alloy (Fig. 1(a)) are as shell for all hcp, tOh-PdRuPt and tOh-PdRuRh NPs. For the rest of NPs (such as tOh-PdRuIr and all Ih NPs), Pd and M atoms segregate from each other forming two semi-spheres cover the Ru core (Fig. 1(b)). While in the oxygen-rich environment, the Ru atoms prefer to oxidize comparing to other elements. Therefore, Ru atoms penetrate outward to the surface (shown in Fig. 1(c)) for respective NPs. The result demonstrates the shell structures of oxidized NPs are almost in solid solution excerpting Ih-PdRuRh NPs. The free energy calculation indicates that the ternary pure hcp NPs are thermodynamically unstable under oxygen-rich condition. In summary, this work demonstrates the elements in ternary NPs can migrate bilaterally under different conditions.

(a) Solid solution. (b) Segregation (c) Oxidation

: Pd : Ru : M : O

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Force Field Parameterization using Genetic Algorithm for Lithium-ion Battery Applications

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Commercially viable and environmental friendly electrochemical storage systems are playing crucial roles in solving social problems. Among them, further improvement of lithium-ion batteries (LIB) are regarded as promising and realistic solution.The performance of LIB depends on varuous factors, but ion transport properties are one of the most important items.[1] However, the mechanism of lithium ion transport in electrolytes remains elusive so far. Many force fields such as AMOEBA and APPLE&P have the potential to be applied to large scale simulation for LIB applications, but the problem is in lack of reliable parameter sets. Force field parameterization is a challenging and time-consuming process. In order to elucidate the mechanism of lithium transport, we have developed a genetic algorithm based force field for the lithium-ion battery applications.

As a demonstration of our approach, performance of the genetic algorithm based force field parameterization reproducing the behavior of first-principles MD simulation with rev-vdW-DF2 density functional is assessed through the applications to pure EC, EC with one Li^+, and EC doped with multiple Li^+ systems. For condensed phase systems, the MD simulation with optimized parameters successfully generated reasonable density and diffusion coefficient for pure EC system, and the solvation structure of lithium ions in EC solvent were well reproduced. Basing on the premise of accurate simulations, the lithium ion transport mechanisms under different concentrations are discussed further. The lithium diffusion becomes slower with increase of concentration. As shown in Figure 1, the reason might come from that in the high concentrated systems, the lack of disengaged EC molecules triggers the decrease of attachment/detachment events, which further caused the lithium diffusion to slow down. These statistical evaluations above demonstrate that the genetic algorithm based force field parameterization is robust enough to get reliable parameter sets for various systems from pure system to high concentrated condensed systems. We have also almost finished the parametrization for anions, and comparison of the Li^+ ions dynamics with and without counter anions is ongoing.
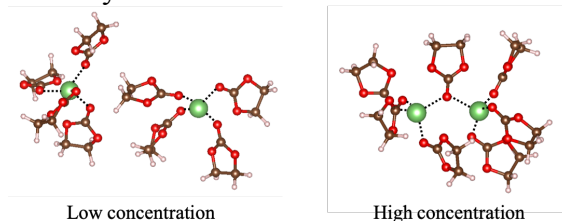

Low concentration   High concentration

Figure 1. Solvated structures of lithium ions in different concentration systems

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First-principles study on structures of crystalline Nd-Fe alloys as candidates for grain-boundary phases in Nd-Fe-B sintered magnets

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Nd-Fe-B sintered magnets are used in a wide range of industry products due to their good magnetic properties. However, there are significant drawbacks of the low coercivity of the magnets at high temperatures. The key to improving the coercivity lies not only in the physical property of the main phase but also in the microstructure of subphases in Nd-Fe-B sintered magnets[1]. The grain-boundary (GB) phases have impact on achieving the high coercivity since the demagnetized GB phases suppresses the magnetic coupling between the main phases. Elucidation of the detailed structures in GB phases is essential to understand the relationship between the coercivity and the microstructure. Therefore, we investigated candidate structures for GB phases.

We determined the most stable fcc Nd-Fe alloy structure by comparing the formation energies using first-principles calculations[2]. Figure 1 shows the formation energies E_{form} defined as follows:

$$E_{\text{form}} = E_{\text{Nd}_x\text{Fe}_{1-x}} - \{x\mu_{\text{dhcp-Nd}} + (1-x)\mu_{\text{bcc-Fe}}\}, \quad (1)$$

where $E_{\text{Nd}_x\text{Fe}_{1-x}}$ represents the total energies of Nd-Fe alloys. $\mu_{\text{dhcp-Nd}}$ and $\mu_{\text{bcc-Fe}}$ are chemical potentials corresponding to the total energy of double hcp Nd and bcc Fe per atom, respectively.

Fig. 1. The bars represent the formation energies E_{form} defined by Eq. (1) per unit cell.

Fig. 2. (a-j) The optimized crystal structures of fluorite $\text{Nd}_x\text{Fe}_{1-x}$ alloys from $x=0$ .67 to $x=1.00$.

From the comparison of formation energies, the most stable crystalline structure is determined as fluorite Nd-Fe alloys. These structures can keep the initial structure in a wide range of composition after optimizing structures as depicted in Fig. 2. Therefore, we conclude that the best candidate structures for the GB phase are fluorite Nd-Fe alloys.

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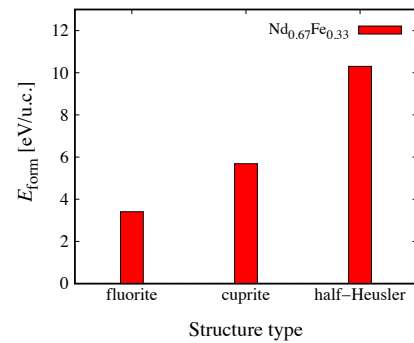[2] T. Ozaki, Phys. Rev. B **67**, 155108 (2003).

Development of First-Principles Crystal Structure Search Method with High Precision and High Efficiency and Its Implementation

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The crystal structure search method using the first-principles calculation becomes difficult to search effectively because the calculation time of the first-principles calculation becomes enormous with increasing the number of atoms. Moreover, none of the methods currently used can completely guarantee that the structure obtained is the most stable structure at the given element and composition ratio, and the correctness can only be confirmed by experimental data. One may consider impossible to cover the search space to be considered because of its large number. In this presentation, we show that it is possible to narrow down the candidate structure by checking all possibilities from 230 space groups. When examining interatomic distances taking into account physical quantities such as atomic radii, many crystal structures are physically impossible and the significant number of the crystal structures can be eliminated. Further refinement can be achieved by taking into account physical and chemical factors. By examining their enthalpies, the most stable structure with the given element and composition ratio is determined. Since the crystal structure is narrowed down, including the degree of freedom of internal coordinates, the time required for structure optimization in first-principles calculations can be greatly reduced. The most important thing is that the most stable structure of the given element and its composition ratio is definitely included in the refined structure.

Intrinsic Doping Limit and Defect-Assisted Luminescence in Cs_4PbBr_6

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Halide perovskite families have been widely studied for photovoltaics [1-2] due to their unique opto-electronic properties, but recently, they are being studied for light-emitting applications [3]. Beyond regular perovskites based on a corner-sharing octahedral network, Cs_4PbBr_6, which is called as zero-dimensional perovskite, is getting attention because of its highly-efficient green light luminescence [4]. The reported band gap of Cs_4PbBr_6 of 3.9 eV is in the ultraviolet (UV) region, thus green light emission in this material cannot be due to its intrinsic band gap. The origin of the green light emission has not clarified and two hypotheses have been suggested to explain this phenomenon. In this work, we investigate the unusual defect chemistry and physics of Cs_4PbBr_6 using first-principles theory and self-consistent Fermi level analysis. Our results show (i) strong charge compensating behavior of Cs_4PbBr_6 due to the dominant donor and acceptor defects and (ii) defect-assisted light absorption and emission from Br_3 tribromide defect whose concentration is controllable.

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Machine Learning Kohn-Sham Density Functionals from Molecules

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Kohn-Sham density functional theory (DFT) [1] is the basis for standard simulation methods of electronic structures of materials with well-balanced computational cost and accuracy. Accuracy of the simulation depends critically on the accuracy of its approximated exchange-correlation functional. The typical strategy to construct the functionals is to refer to exact asymptotic dependences. However, the interpolation of the functionals for intermediate regime must rely on heuristics such as the empirical determination of the function form and parameter fitting with more accurate calculations or experiments, etc. We propose a simply alternative strategy involving minimum human heuristics based on the machine-learning (ML) scheme.

We use an extremely flexible neural-network (NN) form for the functional structure. In our recent study, we have demonstrated that the accurate functional can be constructed with the NN for a model system [2]. Here, we demonstrate a NN-based constructions of functionals for real materials within several levels of semi-local approximation, including local spin density approximation (LSDA), generalized gradient approximation (GGA), and meta-GGA (Fig. 1) [3]. The NNs in those functionals are optimized (trained) to reproduce training data composed of densities and energies of a few molecules obtained by accurate wave-function theories such as the coupled cluster method. The trained NN-based functionals show comparable accuracies to existing functionals, even for hundreds of molecules which are not included in the training database. We also demonstrate the constuction of a non-local functional, which is difficult with the conventional semi-empirical scheme. Those results show that our ML scheme is effective for advancing the development of DFT, opening a novel path of data-driven functional construction.

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Design of isostructural metal-insulator transition in VO$_2$

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In correlated materials, electron and lattice distortion is coupled, therefore metal-insulator transition (MIT) of correlated materials accompanies the structural phase transition (SPT). We focused on the near room temperature phase transition process, i.e. insulating monoclinic to metallic rutile phase transition of the vanadium dioxide (VO$_2$), a representative correlated materials. By the first principles calculation, we designed the rutile/monoclinic heterogeneous interface that electron-electron interaction of monoclinic VO$_2$ region is successfully reduced without crystal structure changing.[1] This interface design can stabilize unstable metallic monoclinic phase. The MIT without SPT of VO$_2$ is also confirmed by combination with experimental approaches like thin-film growth, structural and electrical characterizations. Our results clarify the long-time controversy about origin of phase transition in VO$_2$ and will be applied by the high speed functional devices which are based on phase transition.

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Temperature Dependent Scattering Mechanism of ZrS$_2$ from First-principles Calculations

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Multi-valley structure is one of the favorable band structures for enhancing thermoelectric efficiency, which is realized in several good thermoelectric materials such as PbTe and Bi$_2$Te$_3$. N-type TiS$_2$, whose crystal structure is shown in Fig. 1, also has electronic structure with multi-valley character, and its power factor is relatively high: ~40 µW/cmK^2 at room temperature [1]. In previous experimental studies, the electrical resistivity of TiS$_2$ has been found to exhibit strong temperature dependence of ~T^2. One of the previous studies indicated that inter-valley scattering among conduction band valleys may be related to the peculiar temperature dependence of the electrical resistivity [2].

Since understanding the inter-valley scattering effect on the transport properties may provide a clue toward designing materials with higher thermoelectric efficiency, in the present study, we perform electronic and phonon band calculations based on DFT, and calculate the electronic transport properties by considering the electron-phonon scattering effect using the EPW code [3-6]. As a target material in this study, we consider not TiS$_2$, but instead its analogous compound ZrS$_2$. The calculated temperature dependence of the electrical resistivity is shown in Fig. 2. We will discuss the role of the inter-valley scattering played in the electronic transport properties of ZrS$_2$.

Fig. 1. Crystal structure of TiS$_2$

Fig. 2. Calculated electrical resistivity

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Negatively charged carbon dimer defect as a potential spin qubit candidate in hexagonal boron nitride: an *ab-initio* study

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Hexagonal boron nitride (h-BN) has been recently found to host a variety of quantum defects that are potentially useful for developing solid-state quantum technologies [1]. Interestingly, two recent experiments [2,3] reported observation of optically addressable spin defects in h-BN, paving the way to develop coherent spin qubits in h-BN. These defects feature non-zero spin ground state (S=1) in the C_{3v} point group symmetry, whose principal axis is aligned with the h-BN out-of-plane direction. The defect origin is, however, largely unknown. In this talk, we propose negatively charged carbon dimer(C_2) defect as a new potential C_{3v} S=1 qubit candidate. We use first-principles density functional theory to investigate the structural stability and the spin properties of the defect model. We calculate the defect formation energy of the C dimer defect and other related C-derived defects in h-BN to examine their relative stability. We also consider their spin and electronic structures and we find that the electronic structure of the defect is similar to that of the diamond NV center [4], consisting of one a_1 state and two degenerated e_x, e_y defect states localized in band-gap. To assist potential future experiments, we calculate the zero-phonon line, the zero-field splitting tensor, and the hyperfine tensor of the defect model at various DFT levels of theory including the HSE hybrid functionals [5]. Finally, we discuss several important issues of spin defects in h-BN, which would be relevant for the development of h-BN-based quantum optical devices.

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Band gap engineered ternary semiconductor $Pb_xCd_{1-x}S$: first-principles study

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Ternary Semiconductor $Pb_xCd_{1-x}S$ has attracted intense interest, because the open circuit voltage and power conversion efficiencies can be tunned by modifing the Pb doping concentrations. These advantages of $Pb_xCd_{1-x}S$ have the opportunity to be used to improve the efficiency of solar cells. To understand the nature of the evolution of the band structure as function of Pb doping, we perform first-principles calculations on the bulk band structure of $Pb_xCd_{1-x}S$ based on GGA method with HSE band gap correction. Our GGA+HSE reveals an insulating ground state with 2.3 eV direct energy band gap (E_g) at Γ point, consistent quite well with recent experimental measurements. The finite doping effect is calculated by a linear interpolation of tight-binding model matrix elements of CdS and PbS. Our calculation shows that the energy band gap decrease rapidly and the binding energy increase simultaneously as increasing Pb doping concentration. In addition, we find the lattice effect is not the major effect that modify the energy band gap. The significant the band structure evolution of $Pb_xCd_{1-x}S$ may resulted from the different type of orbital bonding between Cd-S and Pb-S. Our study reveals the origin of the evolution of the band gap in $Pb_xCd_{1-x}S$, paving a way for searching high efficiency solar cell.

First-principles study of Raman spactra of Yb silicates under pressure

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Components of next-generation gas turbines using lightweight SiC-based ceramics need environmental barrier coatings (EBCs) to protect from high-temperature water vapor because of the vaporization of the Si-based ceramics under such environments. Yb silicates Yb_2SiO_5 and $Yb_2Si_2O_7$ are known to as promising EBC materials. In sucn coating systems, residual stresses develop during thermal cycling due to mismatch between the thermal expansion coefficients of the silicates and the underlying ceramics, resulting in critical fatigue of the coating structure [1]. Raman microscopy can be used for measuring stress distributions in coating materials to diagnose the coatings after operation cycles, but its suitability for analyzing residual stress of Yb silicates has been unknown.

In this study, we calculated Raman spectra of Yb_2SiO_5, and $Yb_2Si_2O_7$ under hydrostatic pressure based on the density functional theory and we compared to the spectra of $Yb_2Si_2O_7$ under uniaxial compressive stress in experiments using polycrystalline samples [2]. When no external pressures applied, good agreement between calculated and experimental spectra is obtained. The differences in the spectra between the silicates demonstrate the utility of using Raman microscopy to detect compositional changes in Yb-silicate coatings. From the calculations, lattice vibrations associated with a Raman peak are identified as exemplified by the characteristic mode of Si_2O_7 units in $Yb_2Si_2O_7$. The calculated changes in Raman spectra as a function of pressure are as large as those for yttria-stabilized zirconia, suggesting that Raman microscopy is suitable for monitoring residual stresses in both Yb silicates. This is experimentally confirmed for the large intensity peaks at around 950 cm^{-1} of $Yb_2Si_2O_7$. We also analyzed the vibrational modes using Invarse Participation Rations, indicating that vibrations at frequencies bellow and above 200 cm^{-1} are localized on Yb atoms and Si-O units, respectively.

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Anharmonicity and two-channel phonon transport in thermoelectric tetrahedrite

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Thermoelectric chalcogenides with tetrahedral framework structures, $Cu_{12-x}Tr_xSb_4S_{13}$, have been attracting increasing attention as efficient thermoelectric materials composed of abundant and environmentally friendly elements [1]. One of the key factors that realize the high thermoelectric figure-of-merit ZT of tetrahedrites is their low thermal conductivities, $\kappa \sim 0.5$ W/mK, which have often been attributed to the strong phonon-phonon scattering induced by the "planar rattling" motion [1] of Cu(2) atoms located on triangular planes formed by sulfur atoms. However, the actual role of the planar rattling motion and thermal transport physics in tetrahedrites have not been clarified by *ab initio* theory due to the technical limitations of the conventional approach.

In this study, we investigate phonon anharmonicity and thermal transport physics of $Cu_{12}Sb_4S_{13}$ by using the self-consistent phonon theory [2]. We show that low-energy phonons of $Cu_{12}Sb_4S_{13}$ are strongly temperature-dependent, leading to an unusual temperature dependence of κ. Also, phonons are strongly damped, and their mean-free-path can be as short as the phonon wavelength, which is known as the Ioffe-Regel limit. We demonstrate that, for $Cu_{12}Sb_4S_{13}$, the Peierls-Boltzmann theory underpredicts κ because it only considers the band transport of phonons and omits the hopping contribution. After including the hopping (band off-diagonal) contribution based on Ref. [3], the calculated κ values agree nicely with the experimental result.

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AMP²: A Package for Automated *Ab-initio* Calculation for Crystalline Materials

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With high accuracy, *ab-initio* calculation based on the density functional theory (DFT) become essential components of material science in understanding material properties. However, DFT calculations require a deep knowledge of the theoretical background for reliable data, which can be an obstacle for less experienced users. Developing a fully automated package for DFT calculations makes it easier to use DFT calculations, which would greatly increase the use of DFT calculations. Recently, several automation packages have been developed to build material databases such as AiiDa, atomate, pymatgen and aflow. However, these utilities still assume that users have knowledge of theoretical details.

Here, we introduce 'automated *ab initio* modeling of materials property package' (AMP²) for fully automating DFT calculations with minimal user interference. Starting from crystalline structure, AMP² calculates various crystalline properties of the material such as band gap, band structure, density of states, effective mass and dielectric constant using the preset algorithms that incorporate well-established know-hows. In particular, AMP² identifies the ground-state antiferromagnetic ordering by applying the Ising model, and overcomes the band gap underestimation in semi-local functionals with the help of hybrid functional method. In conclusion, we expect that AMP² would help to understand mateial properties and predict new high performance materials.

Correlation effects seen in one-particle spectra by Green's function coupled-cluster singles and doubles method

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The coupled-cluster singles and doubles (CCSD) is a useful many-body approach in quantum chemistry, not based on the density functional theory (DFT). Recently there exists renewed interest for CCSD and the Green's functions (GFs)[1] thanks to the progress of computational techniques and resources. The GFs are important for understanding the electronic properties since the correlation effects are directly reflected in the spectral features.[2]

We implemented the scheme for obtaining the one-particle GFs applicable not only to isolated systems[3] but also to periodic ones.[4] In this presentation, we will explain the scheme for periodic systems in detail and show the results for C, LiH, and Be chains. Furthermore, we will propose two interpolation schemes[5] for the CCSD GFs primarily for ruducing the computational cost. Specifically, they use Wannier orbitals[6] for circumventing huge cost for a large number of sampled k points. One of the schemes is the direct interpolation, which obtains the GF straightforwardly by using Fourier transformation. The other is the self-energy-mediated interpolation, which obtains the GF via the Dyson equation. We apply the schemes to a LiH chain and trans-polyacetylene and examine their validity in detail. It is demonstrated that the direct-interpolated GFs suffer from numerical artifacts stemming from slow convergence of CCSD GFs in real space, while the self-energy-mediated interpolation provides more physically appropriate GFs due to the localized nature of CCSD self-energies.

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Construction of Green's functions on a quantum computer: quasiparticle spectra of molecules

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Quantum chemistry calculations on quantum computers[1] for obtaining the ground-state energies have been realized by using photonic[2], superconducting[3, 4] and trapped-ion[5] quantum computers. The electronic Hamiltonian for an interacting system is mapped to that for the qubits comprising a quantum computer via some transformation, which leads to the Hamiltonian made up of Pauli tensors.

In order for the quantum computation for electronic structure to come to practical use, quantum algorithms not only for the total energy but also for other various physical quantities have to be developed. One of such quantities is the one-particle Green's functions (GFs), where the quasiparticle spectra[6] are encoded and hence are important for material science. We propose a scheme for the construction of GF of an interacting electronic system via statistical sampling on a quantum computer.[7] Although the non-unitarity of creation and annihilation operators for the electronic spin orbitals prevents us from preparing specific states selectively, probabilistic state preparation is demonstrated to be possible for the qubits. We provide quantum circuits equipped with at most two ancillary qubits for obtaining all the components of GF.

We perform simulations of such construction of GFs for LiH and H_2O molecules based on the unitary coupled-cluster (UCC) method to demonstrate the validity of our scheme by comparing the quasiparticle and satellite spectra exact within UCC and those from full configuration interaction calculations. We also examine the accuracy of sampling method by exploiting the Galitskii-Migdal formula, which gives the total energy only from the GF.

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Why is the electronic density of states peaked in H_3S?

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Hydrogen sulfide becomes superconductor with Tc=200K under pressure of more than 100GPa [1]. The emergent phase H_3S is thought to be responsible for this superconductivity, as its previously predicted crystal structure shows good agreement with the X-ray diffraction measurement[2,3].

The first-principle calculations shows that this crystal structure hosts sharp peak in the electronic density of states (DOS) at the vicinity of the Fermi level. Theoretical studies on the superconductivity of this system focuses on the role of this peak, though direct experimental evidence of its existence has yet been available. For example, the high Tc is explained according to the BCS theory for the conventional phonon-mediated pairing by assuming the DOS peak; the pairing strength is proportional to the DOS at the Fermi level and therefore the peak yields the strong pairing[4]. On the other hand, there has been an argument that the sharpness of the peak implies competition of the electronic and phononic energy scales and breakdown of the BCS picture[5].

In this study, we examine the microscopic mechanism of the formation of the DOS peak to see if it is more than a consequence of the accidentally fine-tuned crystal structure and robust against subtle approximations[6]. From the first-principle electronic structure calculations, we find that the DOS peak is attributed to extended loops of saddle points in the Brillouin zone. Being the one-dimensional structure not passing the special points, it is difficult to recognize with the standard band structure analysis along linear k-point paths. The saddle loop generally emerge by turning isolated saddle points into minimum by any perturbation. We clarify the specific perturbation serving this effect in H_3S, establishing the robustness of the mechanism behind the DOS peak.

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AtomREM: Non-empirical seeker of the minimum energy escape paths on many-dimensional potential landscapes without coarse graining

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The generation of rare trajectories escaping from the minima of the potential surface with minimum energy barriers is a pivotal task in various simulations; chemical reaction, structural deformation, etc.

Recently, we developed a non-empirical scheme to search for the minimum-energy escape paths from the known minima to unknown saddle points nearby [1]. Our method is based on the Smoluchowski equation for distribution $p(x)$, which represents the distribution of variable x obeying the Langevin equation with potential $U(x)$. We derive the Master equation for a biased distribution $q(x, t) = \exp[(1-\delta)U(x)/T]p(x, t)$ with parameter δ. We have found that $q(x,t)$ climbs up the potential surface through the "valley" on the potential surface, if the initial distribution $q(x, t=0)=\delta(x-x_0)$ is properly set at the entrance of the valley. A stochastic walker algorithm that reproduces $q(x, t)$ is constructed to move the walkers up the surface through the potential valleys. Later, we designed a systematic algorithm to generate the initial atomic positions as entrances to the potential valleys, with which we can execute non-empirical search of the escape paths free from a priori collective variables or artificial force field [2]. Currently the code is available online as AtomREM (Atomistic Rare Event Manager [3,4]). Further development for accurate potential functions comparable to the first-principle calculations with the neural network is also under way. In the presentation, we demonstrate the applications of AtomREM to the Lennard-Jones clusters, hydrocarbon chains and some solid systems.

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Enectrode potential at the metal electrode/electrolyte solution interface studied by DFT with reference interaction site method

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To solve the energy resource issue, electrochemical devices, which utilize the electrochemical reaction, have been continuously developed. The electrode potential plays an important role to understand the electrochemical reaction at an interface between an electrode and electrolyte solution. However, theoretically, the precise description for the realistic electrode potential is problematic, because the theory under the grand canonical condition is required.

For solving the issue of grand canonical condition and understanding the electrochemical reaction, the density functional theory (DFT) combined with the effective screening medium technique and reference site interaction model, terminated as the ESM-RISM, is applied to the electrochemical interface [1, 2]. The electronic structure of electrode and the structure of electrolyte solution is respectively treated by the DFT and Laue-represented RISM. In the ESM-RISM framework, both the electronic and solution systems can be treated under the grand-canonical condition. All calculations have been carried out using plane-wave basis set and pseudopotential code QUANTUM ESPRESSO [3].

Figure 1 shows the results of laterally averaged electrostatic potentials at charged Al(111)/NaCl aq. interface, where the total charge of the electronic system is +0.02e. The electrostatic potential near the Al (111) surface show the oscilations. This is caused by the charge of solution. The electric field induced by surface charge is completely screened by the solution. As a result of this, the flat potential level of whole electrostatic potential is appeared as shown in Fig. 1. We will discuss the details of electrode potential at the interface between electrode and electrolyte solution, in the presentation.
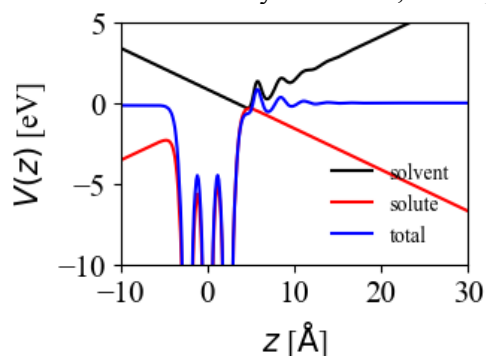

Fig.1 Laterally averaged electrostatic potential at the Al(111)/NaCl aq. interface.

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Formic Acid Adsorption and Decomposition on the Cu(111) Surface : Monomeric and Polymeric Structures

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We performed theoretical study on adsorption and decomposition mechanism of formic acid (HCOOH) on the close packed Cu(111) surface. We found that by using the van der Waals (vdW) inclusive density functionals (optB86b-vdW and rev-vdW-DF2), the activation energy for desorption (E_{des}) is significantly improved over GGA-PBE.[1] Moreover, the vdW inclusive density functionals give better agreement with experimental results. The activation energies of decomposition (E_{dec}) calculated by using vdW inclusive density functionals are lower compared with desorption energies, seemingly in contradiction with experimental findings at room temperature, in which no decomposition of HCOOH on Cu(111) is observed when the surface is exposed to the gas phase HCOOH. We performed the reaction rate analysis based on the first-principles calculations for desorption and decomposition processes to clarify this contradiction. Figure 1. Shows that the desorption of monomeric HCOOH is faster than that of its decomposition rate at room temperature because of a much larger pre-exponential factor. Thus, no decomposition of monomeric HCOOH should take place at room temperature.

Fig. 1. Calculated reaction rates per surface adsorbate for the desorption and decomposition of monomeric HCOOH. Red dashed line with circles and blue solid line with squares represent the decomposition and desorption rates, respectively.

At low temperature, HCOOH on Cu(111) surface was reported to form polymeric HCOOH structures.[2] We found that the E_{des} for polymeric HCOOH structure is 0.21 eV more stable rather than that monomeric HCOOH. We also calculated the IRAS spectra for polymeric HCOOH structures. Polymeric HCOOH structures show higher peak assigned for π(O-H) and π(C-H) and small double peaks assigned for ν(C=O) vibrational mode as observe in the experimental studies.

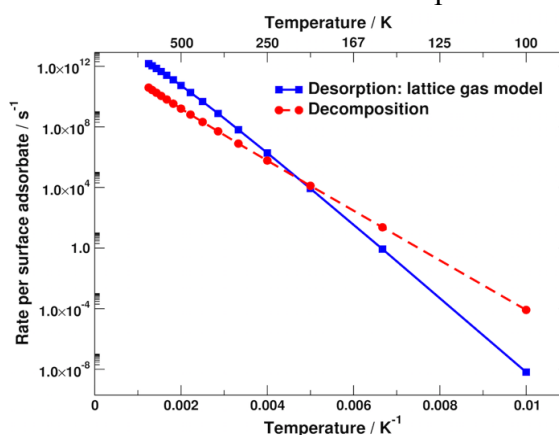[1] S. E. M. Putra, F. Muttaqien, Y. Hamamoto, K. Inagaki, I. Hamada, Y. Morikawa, *J. Chem. Phys.* **150**, 154707 (2019).
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Antiferromagnetic topological insulator EuSn$_2$P$_2$

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In the past decade, there are a lot of materials have been demonstrated as 3D topological insulators in experiments. Besides nonmagnetic topological insulators, the related study of antiferromagnetic topological insulator (AFM-TI) is still lacking. Very recently, Gui et al. [1] synthesized an entirely new quantum material, EuSn$_2$P$_2$, that never been proposed previously. Neutron scattering displays an antiferromagnetic phase below 30K. Based on density functional theory, we find a band inversion near the Γ point, which is a classical feature of topological insulators. The Z2 invariant calculation presents a nontrivial topological phase through the Wilson loop. In addition, we observe Dirac topological surface states protected by the $S = \Theta T_{1/2}$ symmetry on the (010) plane. Our calculations demonstrate that EuSn$_2$P$_2$ is a solid candidate of antiferromagnetic topological insulator, providing a new platform to study the novel phenomena in magnetic topological system.

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Machine learning analysis on tunnel magnetoresistance of Fe/disordered-MgAl$_2$O$_4$/Fe(001) magnetic tunnel junction

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  MgO-based MTJs have succeeded to provide large tunnel magnetoresistance (TMR) ratios at room temperature[1]. This can be attributed to the coherent tunneling through Δ_1 evanescent state in single-crystalline MgO and the half-metallic behavior on the Δ_1 state in Fe and CoFe [2]. However, epitaxial growth of MgO crystalline is difficult because of lattice mismatch with ferromagnetic electrode, which is about 4% for bcc-Fe.The normal-spinel type MgAl$_2$O$_4$(MAO) showed a great advantage in the epitaxial growth compared to the MgO because of a small lattice mismatch less 1% with bcc-type ferromagnetic metal at (001) face[3]. Recently, it is found that the cation-site disordered MAO provides higher TMR ratio compared with the ordered MAO due to the suppression of the band folding effect[4]. In order to enhance the TMR ratio of MAO-based MTJs, it will be important to identify local structures of cation-site disordered MAO providing high TMR ratios. However, at present stage, experiments cannot determine detail local structures of cation-site disordered MAO.

  In this work, we investigate and discuss the TMR effect of Fe/disordered-MAO/Fe(001) magnetic tunnel junctions to clarify optimal structures of disordered-MAO giving larger TMR ratios by using the Bayesian optimization and the least absolute shrinkage and selection operator (LASSO) technique combined with the first-principles calculations. The largest TMR ratio is successfully obtained from the Bayesian optimization with 1728 cases. Furthermore, we find that the in-plane distance between two Al atoms plays an important role to characterize the TMR ratio. Since the Al-Al distance of disordered MAO significantly affects to the imaginary part of complex band structures, the majority-spin conductance of Δ_1 state in Fe/disordered-MAO/Fe MTJs increases with increasing the in-plane Al-Al distance. This leads to enhancement of the TMR ratio. Furthermore, TMR ratio tends to increase when the stoichiometry of Al, Mg, and Vacancy in the [001] plane is 2:1:1. These results indicate that control of Al atomic positions is important to maximize TMR ratios in MTJs with disordered MAO.

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Electronic structure characterization of NiCo$_2$O$_4$: QSGW *ab-initio* calculation

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Nickel cobaltite (NiCo$_2$O$_4$) is one of the promising materials to be used in applications of catalysis and spintronics. In catalysis, NiCo$_2$O$_4$ have a role as heterogenous catalyst with a basic reaction of oxygen reduction reaction (ORR) constructing many complex reactions, such as energy generation (fuel cell), weathering of materials, and most of biological process [1]. One of the main interests of this reaction is focused on its role in the oxygen electrode used inelectrical-power related systems, i.e. metal-air batteries and fuel cells. From this point of view, being an abundantly available transitional metal oxides, NiCo$_2$O$_4$ undisputedly has many advantages, compared toalready available alternatives. However the mechanism of ORR on metal surfaces is still remains unclear. We have theoretically investigated electronic and magnetic properties of bulk NiCo$_2$O$_4$ to understand its basic features. NiCo$_2$O$_4$ indicates a half-metallic property according to low field magneto-resistance (LFMR) measurements [2]. One can easily tune its electrical and magnetic properties by varying the crystal growth temperature [3].

In our theoretical approach, quasi-particle self-consistent GW (QSGW) calculations [4] were performed in proper and inverse spinels of NiCo$_2$O$_4$. The calculational results were also compared with those of generalized gradient approximation (GGA). The QSGW enables to calculate metallic electronic structures. Such advantage may be useful for a wide range of materials. We obtained the results of one-shot GW and QSGW as well as those of GGA.

In the inverse spinel of NiCo$_2$O$_4$, the electronic states near the Fermi level consist mainly of the octahedral site 3d orbitals on Co and Ni in the majority spin state, and of the tetrahedral site 3d orbitals at Co in the minority spin state. Half-metallicity was found on all results and the metallicity is associated with the 3d orbitals on tetrahedral Co(Co$_{Td}$) in the minority spin state. On the other hand, a quasi-insulating nature was observed on octahedral Co (Co$_{Oct}$) and octahedral Ni (Ni$_{Oct}$) in the majority spin state. Overall, the QSGW calculation yielded more localized orbitals, compared with that of GGA. The largest contribution to the total magnetization (2.0μ_B/f.u.) is provided by Co$_{Td}$ (2.3μ_B) and Ni$_{Oct}$ (-1.1μ_B) with an anti-parallel configuration. The spin moment on Co$_{Oct}$ may be in a low spin state (less than 0.1μ_B). We will discuss our results by comparing with available experimental values. In addition, we compare the results of NiCo$_2$O$_4$with those from Co$_3$O$_4$ and Fe$_3$O$_4$ to get more insight on above properties.

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First-principles study of Rashba effect
in quantum well states of Ag/Au(111)

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An asymmetric potential together with the strong spin-orbit coupling splits the energy level of the surface- and boundary- electronic states according to their spins. This splitting which is called the Rashba effect is applicable to the spin-current generation/detection in the spintronics devices. Therefore, the materials which exhibit the Rashba effect are explored widely. Recently, part of coauthors (RN, KK, and TK) newly observed the Ag-thickness dependent energy splitting in the spin-decomposed angle resolved photo-emission spectroscopy measurement of quantum well states (QWS) in the multi-layered Ag/Au(111) system. This splitting also depends on the quantum number (number of nodes) of QWS confined in the Ag region. In this study, we performed the first-principle study based on density functional theory to reveal how such a Ag-thickness and QWS dependent Rashba effect appears in this boundary system. We reproduced quantitatively the energy splitting of QWS (Fig. 1 left) and performed further analysis by focusing on the variation of the envelope of the QWS along the depth direction. From this analysis, we found that these dependences can be explained by the linear combination of the intensities of the QWS at the Ag and Au atom locating on the boundary (Fig. 1 right). In this presentation, we explain this mechanism with the first-principles calculation and the minimum tight-binding model as well as the detail of the calculation.
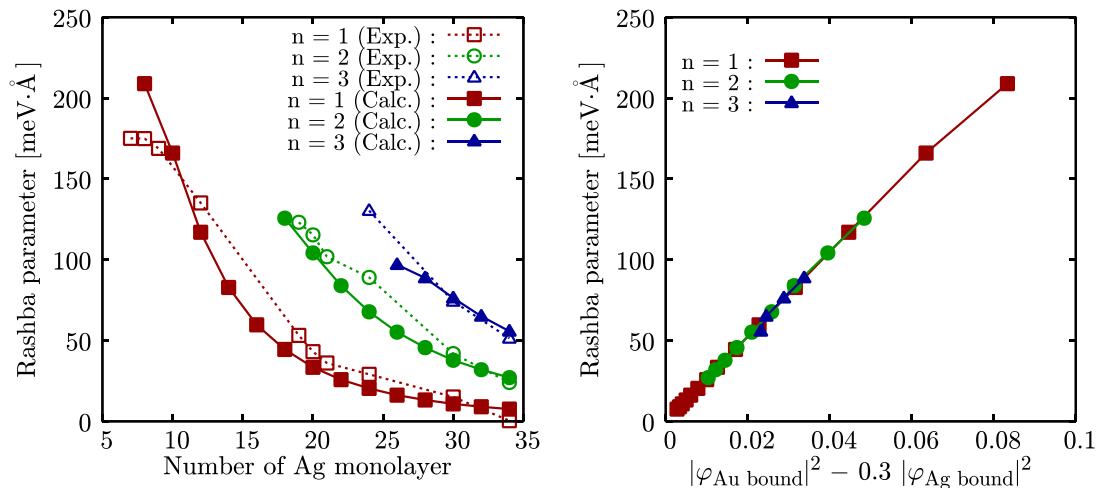

Figure 1: (Color online) (Left) The calculate and experimental Rashba splitting parameter at each QWS and Ag-monolayer thickness. (Right) The correlation between the Rashba splitting parameter and the linear combination of the intensities of the QWS at the Au and Ag atoms on the boundary.

Reaction of CO_2 and C on flat and step surfaces of Co metal

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The increase in the emission of carbon dioxide, CO_2 from human activities are the root of global warming and climate changes. A promising route for decreasing the greenhouse effect is to convert CO_2 into value-added chemicals. Among the methods of recycling CO_2, reduction of CO_2 via chemical pathway in a reducing agent such as methane (CH_4) is an efficient way of storing and recycling renewable energy. In this reaction, Co metal was identified as a good catalyst to catalyze the dissociation of both CH_4 and CO_2 [1]. In order to prevent the deposition of carbon which comes from excessive CH_4 dissociation, the reaction of CO_2 with C on the surface of Co plays the most important role. Experimentally, Co catalysts were generally exhibited as Co cluster dispersed on top of support materials [2]. Thus, Co cluster generally consist of several flat, step, and kink surfaces [3]. Fundamentally, the surface geometric structure affects the adsorption, diffusion, dissociation, formation energy barrier of CO_2, C, and their subsequent products (O and CO).

In this study, we perform *ab initio* simulations in order to model the reaction between CO_2 and C on Co metal surfaces. Our study includes the absorption, dissociation reaction (Eq. 1) of CO_2 and its subsequent reaction (Eq. 2) with adsorbed C on different flat and step surfaces, namely (111), (110), (100), (221), and (211).

$$CO_2 + 2^* \rightarrow CO^* + O^* \hspace{3cm} \text{(Equation 1)}$$
$$O^* + C^* \rightarrow CO^* \hspace{3.5cm} \text{(Equation 2)}$$

Where, CO^*, O^*, C^* indicates adsorbed CO, O and C and
2^* indicates two free adsorption sites

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Strongly Fluctuating Atomic Volumes, Charges, and Stresses in BCC Multicomponent Alloys

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Disordered multicomponent alloys, especially those called high-entropy alloys, have been attracting considerable attention from both experiment and theory [1] because of their extraordinary properties such as high temperature strength. Local lattice distortions are thought to be closely related to mechanical strength. In the present study, we investigate them in a series of body-centered cubic (BCC) multicomponent alloys, including refractory high-entropy alloys (HEAs), in terms of atomic volumes, atomic charges, and atomic stresses defined by the Bader charge analysis based on first-principles calculations [2]. The QMAS code [3] is used in calculating these atomic properties.

Analyzing the obtained results, we find surprisingly large distributions of the atomic properties for each element in each alloy, indicating a large impact of the varying local chemical environments. We show that these local-environment effects can be well understood and captured already by the first and the second nearest neighbor shells. Based on this insight, we employ linear regression models up to the second nearest neighbor shell and successfully predict the atomic properties.

Furthermore, we find that the elementwise-averaged values of the atomic properties correlate linearly with the averaged valence-electron concentration (VEC) of the considered alloys. The averaged VEC values can be evaluated without time-cosuming calculations and, therefore, are expected to be utilized as an efficient descriptor for materials screening

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First-principles Study of the Interaction between Boron Nitride and H-terminated (111) Diamond Surface

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For the better understanding of recent experimental results [1] on field effect transistors (FETs), we performed density functional theory (DFT) calculations on the hexagonal boron nitride (BN) with the hydrogen-terminated (111) diamond surface. In our work, atomic structures and electronic properties of the nanostructures of BN on the hydrogen-terminated (111) diamond surface will be addressed using first-principles DFT based simulation code CONQUEST. The small lattice mismatch of about 0.4% between BN and the H-terminated (111) diamond surface is predicted in good agreement with previous experiments[2,3]. The binding energies of the various stacking patterns, the density of states, and the charge density distribution will be discussed. It turns out that weak van der Waals interactions dominate for BN on these H-terminated (111) diamond surface. While both BN and H-diamond have the wide band gap, BN absorbed on H-diamond narrows the band gap. Our results may provide a good understanding of the interfacial properties of BN/H-diamond and pave the way to further design FETs having high mobility and high carrier density in nanoelectronics.

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First-principles analysis on superconductivity under strain in Li-intercalated bilayer MoS_2

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MoS_2, a member of transition metal dichalcogenides, has been investigated widely due to its high potential in various applications. Previous first-principles calculations revealed that the intercalation of Li [1], Na [2], and Ca [3] could turn bilayer MoS_2 from semiconductor into superconductor. Furthermore, the transition temperature T_c in the Na-intercalated bilayer MoS_2 is predicted to increase from 2.9 K to 10 K under the compressive strain of 7%, while it is suppressed under tensile strains [2]. Systematic investigation on the Li-intercalation case, however, has not been done yet. Here we report the results of our first-principles calculations within the density functional theory on the superconductivity in the Li-intercalated bilayer MoS_2 [4], which demonstrates that both of compressive and tensile strains could enhance the superconductivity.

Figure 1 shows the strain dependence of T_c calculated using the McMillan-Allen-Dynes formula with setting the Coulomb repulsion parameter to 0.1 and the density of states at the Fermi level $N(E_F)$. As seen in this figure, both of compressive and tensile strains increase T_c. The increase of T_c under tensile strain is ascribed to the increase in $N(E_F)$ and the electron-phonon coupling (EPC). On the other hand, $N(E_F)$ hardly changes under compressive strain. We found that the T_c increase in this case originates from drastic softening of phonon at a specific q-point in the Brillouin zone. The wave vector of the softning mode matches with the k-vector which connects the electron pockets on the Fermi surface. The enhancement of EPC and resuting phonon softning induced by this condition both play positive roles in incresing T_c [4].

Figure 1 : The change in transition temperature (T_c), EPC constant (λ) and density of states at Fermi level ($N(E_F)$) under strain.

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First-Principles Calculations of Point Defects and Proton Incorporation in Orthorhombic Perovskite LaScO$_3$

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LaScO$_3$ is a perovskite-type oxide that exhibits proton conductivity at intermediate temperatures (300-600°C) when acceptor-doped with Sr [1]. At low temperatures it is orthorhombic (*Pnma*) as a result of octahedral tilting away from the aristotype cubic structure. This lower symmetry compared to cubic perovskite BaZrO$_3$ is expected to influence the proton migration mechanism in this material.

We analyzed the formation energies of isolated point defects and defect clusters in Sr- and Ba-doped LaScO$_3$ using first-principles calculations to estimate their thermal equilibrium concentrations. Calculations also focused on proton incorporation and binding energies. Total energy calculations were performed using VASP code [2]. As an example, the temperature dependence of defect concentrations in 20 mol% Sr doped LaScO$_3$ under Sc$_2$O$_3$ rich conditions with $p_{(O_2)} = 0.1$ atm and $p_{(H_2O)} = 0.02$ atm is shown in Fig. 1. Using thermodynamic data of O$_2$ and H$_2$O together with the zero-K defect formation energies from first-principles calculations, the hydration reaction in LaScO$_3$ at low temperature and in a humidified atmosphere was reproduced.
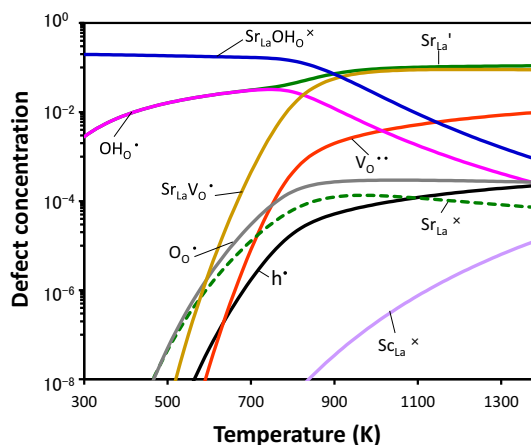
Fig. 1. Temperature dependence of various defect concentrations in LaScO$_3$.

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IrO$_3$ Desorbability of Iridium Alloys at High Temperatures from Thermodynamical Perspective

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The best material for spark plugs of an internal combustion engine is always considered as iridium. As the one of the precious metals, iridium has a remarkable mechanical strength and chemical inertness, in particular at high temperatures. The most significant problem to use iridium at high temperatures is a desorption of iridium oxides. This well-known issue was broadly studied, so it is revealed that IrO$_3$ is the dominant gaseous oxide of iridium at high temperatures[1]. However, theoretical studies for iridium desorption are yet to be performed, thus many things for the iridium desorption such as a surface index on which the reaction occurs as well as reaction paths are remain as unrevealed.

In this study, we investigate about the iridium surfaces and desorption of IrO$_3$ by first-principles calculations based on density functional theory. We used the OpenMX code based on the norm-conserving pseudopotentials and pseudo-atomic localized basis functions with GGA-PBE exchange-correlation functional. By those methods, we compared desorption free energies and oxygen adsorption free energy density. As the results, Ir(111) surface is concluded as the surface on which IrO$_3$ desorption occurs. This result is supported by our nudged elastic band calculations which show that there is no extra energy barrier for iridium desorption in addition to the desorption free energy.

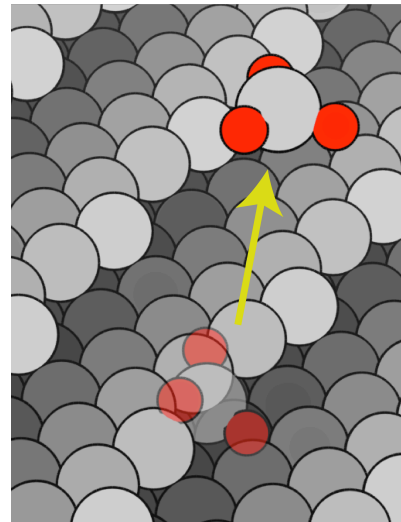Fig 1. Iridium desorption as IrO$_3$

We took a step further, we tried to design iridium alloys to prevent the problem of iridium desorption. We evaluated some transition-metal elements by substituting an iridium atom at the surfaces. By this approach, we can predict the performance of materials in terms of the desorbability, in general.

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Calculated Large Seebeck Coefficients in Fe-doped Si-Ge Alloys

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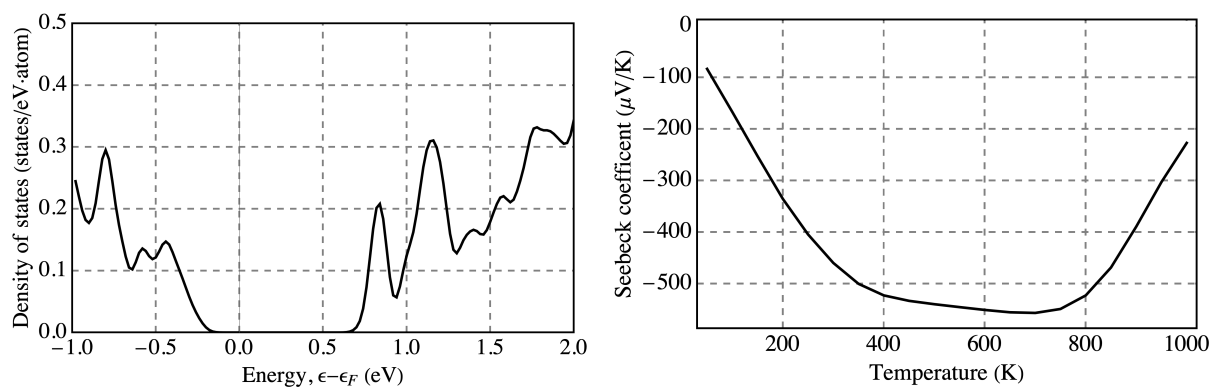[4]*Research Institute of Electrical Communication, Tohoku University, Sendai, 980-8577, Japan*
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Si-Ge alloys are known as one of the cheapest nontoxic thermoelectric materials utilized at high temperatures. The dimensionless figure of merit, ZT, of Si-Ge alloys, however, is quite small ($ZT < 1$ for both p- and n-types) because of their relatively large phonon conductivity [1]. The small ZT values in Si-Ge alloys have been somewhat improved with the use of a nano-structuring approach, where phonon conductivity is reduced by making a grain size small. To further increase the ZT values of the Si-Ge alloys, there are some attempts to modify their electronic band structure by doping Fe atoms, and it has been reported that a high ZT value, $ZT > 1.88$ (at $T = 873$ K), as well as a large Seebeck coefficient, $|S| > 517\,\mu\text{V/K}$ (at $T = 673$ K), were produced in the nanostructured $Si_{0.55}Ge_{0.35}P_{0.10}Fe_{0.01}$ sample [2]. It is believed that the large Seebeck coefficient would originate from a strong peak at the edge of the conduction band in the electronic density of states generated by the Fe-doping (a so-called impurity state), and this large Seebeck coefficient increased the ZT value through the relation, $ZT \propto S^2$. An occurerence of the impurity state by doping Fe atoms into a Si bulk sample has been confirmed using an electronic band structure calculation [2], whereas it has not been confirmed yet in the Fe-doped Si-Ge system either from experimental or theoretical approach. In this work, therefore, the impurity state in the Fe-doped Si-Ge alloys is calculated using an electronic band structure calculation, and the reported large Seebeck coefficient is reproduced with the use of the linear response theory assuming that the group velocity and relaxation time are not sensitive to the energy.

The biggest difficulty in calculating the electronic structure in the Fe-doped Si-Ge system is that Si-Ge alloys have a disordered configuration in the whole composition range. To mimic a disordered configuration, a special quasi-random structure (SQS) is used in a supercell containing 64 atoms. The calculated electronic density of states and Seebeck coefficient in the $Si_{50.0}Ge_{48.4}Fe_{1.6}$ alloy are shown below, where a hybrid functional (HSE06) is employed in the VASP code. The impurity state is successfully produced at the bottom of the conduction band, and the value of the Seebeck coefficient is in good agreement with the reported experiments, $|S| > 517\,\mu\text{V/K}$ (at $T = 673$ K) [2].

Figures. Calculated density of states (left) and Seebeck coefficient (right) in the $Si_{50.0}Ge_{48.4}Fe_{1.6}$ alloy.

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Nuclear Quantum Effect for Hydrogen Adsorption on Pt(111)

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It is generally believed that H/Pt(111) is predominantly adsorbed on the fcc site, as supported by DFT-GGA calculations. Experiments, however, observe the signal from the atop site [1]. Recently, we have revisited this problem with random phase approximation, and predicted the coexistence of fcc and atop sites at room temperature [2]. However, anharmonicity in the nuclear quantum effect is neglected in the previous work. Here, we apply the *ab initio* path integral molecular dynamics simulations [3] to H adsorption on Pt(111) near the full coverage. The coexistence of fcc and atop site is observed at the full coverage, while hollow site domains at the coverage of 2/3, in agreement with experimental observation. The transfer between fcc and hcp site would stabilize the hollow site. We distinguish the harmonic and anharmonic terms in the nuclear quantum effect, and find that the anharmonic effect is nontrivial at the room temperature. We emphasize the importance of nuclear quantum effects, where the distribution is different from that in classical case. Our results provide a new insight to consistently interpret electrochemical and spectroscopic data.

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Band Alignment and Reconstruction of ABO$_3$ (001) Surfaces

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The ionization potentials (IPs) and electron affinities (EAs) are key parameters in a wide range of technological applications including electornic devices, photocatalysis, and catalysis. The IPs and EAs are surface-dependent properties affected by the surface dipole contribution that depends on the atomic structure, stoichiometry, and adsorption. Among the perovksite oxides, much research has focused on the SrTiO$_3$ (001) surface, which has diverse reconstructed surfaces such as p(2×1), p(2×2), p(6×2), c(4×2), c(6×2), c(4×4), ($\sqrt{5} \times \sqrt{5}$)R26.6°, and ($\sqrt{13} \times \sqrt{13}$)$R$33.7°. However, a systematic study of the effects of surface reconstruction on band alignment is still lacking for perovskite oxides.

Here, we have systematically investigated the effects of surface reconstruction on the band alignment of ABO$_3$ perovksites (A = Ca, Sr, Ba, Pb; B = Ti, Zr, Hf, Ge, Sn). For cubic and distorted structures, we have considered stoichiometric slab models to determine the IPs and EAs for reconstructed surfaces. Low-surface-energy structures for diverse lateral periodicities were generated using an *ab initio* evolutionary crystal structure search method, as implemented in the AMDAEUS code [1]. We compare the surface energies of these reconstructed stoichiometric surfaces and cleaved nostoichiometric surfaces as a function of the chemical potential. As a compositional trend of perovskite oxides, we found that the IPs have an almost linear trend against the Goldschmidt tolerance factor. These results allow understanding of the band alignment of perovskite oxides with the effects of surface termination and reconstruction.

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Density Functional Theory-based Investigation of Nickel Selenide as an Electrocatalyst for Oxygen Reduction and Water Oxidation Reactions

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To reduce the cost of fuel cells, earth-abundant transition metal compounds are being developed to replace precious metals in oxygen reduction reaction (ORR) catalysts and bifunctional oxygen reduction/evolution catalysts. Recent work highlights nickel selenide (NiSe), a p-type semiconductor exhibiting high O_2 and H_2 evolution activity in alkaline media [1,2]. However, ORR catalysis on NiSe was observed to terminate at peroxide formation, albeit without a clearly defined mechanism explaining this observation [3]. Modelling its surface and reaction intermediate-surface interactions may provide insight into the ORR mechanism and other catalytic properties of NiSe.

The present study uses density functional theory (DFT) to examine the adsorption of H_2O and OH molecules on pristine NiSe(101), as well as NiSe with a surface vacancy. We found that DFT using the generalized gradient approximation (GGA) predicts NiSe as metallic and fails to reproduce the antiferromagnetic and semiconducting nature of NiSe observed experimentally [1]. Hence, to more accurately describe its electronic structure and its effect on the intermediate adsorption and reaction mechanism, DFT with the on-site Coulomb interaction (DFT+U), hybrid functional, and quasiparticle self-consistent GW method were employed. We obtained a small absorption energy of H2O (-0.093 eV/mol) along with a large adsorption distance (2.3 Å) on a pristine metallic NiSe(101) surface with PBE-GGA. This is in contrast to the chemisorption of OH with an adsorption energy and distance of -2.26 eV and 1.86 Å, respectively. We also found that H_2O and OH adsorption is enhanced by a Ni vacancy on the NiSe(101) surface. The facile formation of these oxygenous intermediates with the NiSe surface may be significant in describing ORR/OER catalysis behavior for nickel selenides. [4]

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First-principle calculation of superconducting transition temperatures of elemental transition metals including the effect of spin fluctuations

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It is widely known that spin fluctuations can be the origin of electron pairing in unconventional superconductors. On the other hand, spin fluctuations also play an important role in conventional phonon-mediated superconductors. In nearly ferromagnetic metals, spin fluctuations induced by strong exchange interaction can lead to a significant suppression of singlet pairing and thus *s*-wave superconductivity. To investigate how spin fluctuations influence conventional superconductivity, Eliashberg theory has been extended to include the effect of spin fluctuations [1] and applied to some systems, e.g. V and Nb [2]. However, this treatment is rather empirical and fully non-empirical calculation of such effect has scarcely been performed so far.

In this study, we explored the interplay of spin fluctuations and superconductivity from first-principles by using density functional theory for superconductors (SCDFT) [3] including the effect of spin-fluctuations [4]. We applied this method to the elemental transition metals V, Nb and calculated the effect of spin fluctuations on T_c. We found that spin fluctuations reduce T_c in both cases and the amount of its reduction is related with the magnitude of localization of the valence orbitals.

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Self-consistent first-principles method for extended Hubbard interactions

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We present an efficient self-consistent computational approach that extends the density functional theory plus on-site interaction (DFT+U) method further to include inter-site Hubbard Coulomb interaction (V). The extended DFT+U +V method is suitable to calculate electronic structures of periodic systems with various interactions with disparate ranges. Typical computational methods for DFT+U and DFT+U+V require additional set of computations to obtain U and (or) V. In this presentation, we suggest a scheme to compute the Hubbard parameters self-consistently with efficient algorithms..

Ground-State Structure Search for Pnictide Antiperovskites

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Antiperovskites (APs) with an M_3AB formula (M = transition metals, alkaline metals, and alkaline-earth metals, $A = p$-block elements, $B = $ C, N, and O) have recently drawn attention because of their intriguing physical properties. Most of the reported APs have undistorted structures [1], while some nitride and oxide APs have been reported to possess distorted perovskite structures. Fewer studies about the distortions of these materials, especially pnictide APs, have been performed. Some of the pnictide APs M_3PnN have been experimentally reported as narrow gap semiconductors which have a potential to be applied to photovoltaic cells and light emitting diodes.

In this study, we performed first-principles calculations using VASP [2] and the PBEsol functional [3] to investigate M_3PnN with seven representative crystal structures. We also performed *ab initio* evolutionary crystal-structure searches using AMADEUS [4] and lattice-dynamics calculations using PHONOPY [5] to explore other stable structures.

We extracted the seven representative crystal structures of ABX_3 compounds: cubic, orthorhombic, hexagonal, rhombohedral perovskites, Sn_2S_3-, $YMnO_3$-type, and ilmenite structures from Materials Project [6]. We found that the orthorhombic perovskite *Pbnm* phases are the most stable among the seven structures in the phosphide and arsenide APs except Ba_3AsN. On the other hand, in antimonide and bismuthide APs except Ba_3SbN and Ba_3BiN, the cubic perovskite $Pm\bar{3}m$ phases are the most stable. Additionally, in the three Ba-containing compounds, i.e., Ba_3AsN, Ba_3SbN, Ba_3BiN, the hexagonal perovskite $P6_3/mmc$ phases are the most stable.

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Single Platinum Atom Supported at Graphene Edges as a Promising Catalyst for CO Oxidation Reaction

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The single-atom catalysis has attracted increasing interest as one of the most promising ways to reduce the amount of precious metals used and to enhance the catalytic activity. Platinum atoms deposited on graphene are reported to show enhanced catalytic activity for some chemical reactions,[1-3] e.g. methanol oxidation in direct methanol fuel cells. Nonetheless, the precise atomic structure, the key to understand the origin of the improved catalytic activity, is yet to be clarified. Here, we present a computational study to investigate the stability of platinum adsorbed on graphene with special emphasis on the edges of graphene, taking graphene nanoribbon as a simple model for graphene edge. By means of DFT-based thermodynamics, it is found that a single Pt atom is preferentially adsorbed at the edge rather than on the basal plane,[4] the good news regarding the search of dense single atoms dispersion on a supported material. The calculated core level shift (CLS) for the stable structures are in reasonable agreement with the experiment,[5] corroborating our findings. Large positive CLSs indicate the strong interaction between single Pt atoms and graphene. Furthermore, the CO oxidation reactivity of single Pt atoms bonded to edges of graphene nanoribbons (Pt@GNRs) is predicted based on the adsorption energies from DFT calculations combined with a kinetic model. Compared with the Pt(111) surface, single Pt atoms supported by graphene nanoribbon show a better activity in CO oxidation reaction, characterized by the weakening of CO and the strengthening of OH adsorptions on Pt@GNRs. It is presumably the origin for the improved CO tolerance in the anode electrode in direct methanol fuel cell. This study can be a basis for further investigation of the development of single-atom catalysts based on platinum and graphene related materials.

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High Carrier Mobility of MoS₂ Nanoribbons Under Simultaneous Strain and Electric Field

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The one-dimensional (1D) hexagonal nanostructures of TMDC materials, such as nanotubes, nanowires, nanorods and nanoribbons have been intensively investigated in recent decades due to their diverse physical properties. With high on/off ratio, low dimensional MoS₂ is gaining more importance as promising channel layer materials for miniaturized field effective transistors (FETs). As the present concern of low dimensional MoS₂ is its low mobility at room temperature, we focus to improve the nanoribbon MoS₂'s mobility in this study. Semiconducting armchair nanoribbon can sustain quite high strain and sufficiently large external electric field to remain in its 2H-phase with nonzero bandgap. we simultaneously applied tensile strain along the ribbon direction and applied external transverse electric field along the width direction, within the nonzero bandgap region and observed that electric field with strain enhances the carrier mobility (electron & hole) as much as eight times compare to its 2D-monolayer phase. We studied the electronic and geometric structure of MoS₂-ANR with first principle calculations and calculated carrier mobility using deformation potential theory. With increased carrier mobility and high on/off ratio, MoS₂ nanoribbons could be a suitable candidate for the channel layer materials of FET.

Keywords: Carrier Mobility, MoS₂

Classical Density Functional Theory Calculations of Distribution Function of Ne Liquid

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Electrochemical energy conversion and storage have become a requisite technology in the last few decades. To consider electrochemical reactions at the interface, one needs to refer to the electrode potential as it is relevant to charge transfer (CT) reactions. This was made possible recently by using DFT calculations combined with the ESM technique [1] and the reference interaction site model (RISM) [2], called as ESM-RISM calculation [3] because of the grand canonical properties. In the ESM-RISM calculation, the number of electrons and that of the ions in the solution are determined according to the Fermi level in the metal electrode and the chemical potential in the bulk solution region, respectively, thereby enabling to relate the Fermi level with the electrochemical reaction energy as required to define the electrode potential [4].

However, RISM has a quantitative problem as has been demonstrated using classical force field models, such as the model solution characterized by hydrogen bonds typical of liquid water. The distribution functions obtained by RISM equations do not reproduce the second peak of O-O distribution of the liquid water. We consider that this disagreement is mainly caused by the many-body correlation only approximately described using the closure.

In this study, we focus on the classical density functional theory (CDFT), which is an alternative (and formally regorous) formulation for describing classical many-body system, and aim an accurate description of the solution including hydrogen bonds. In this poster presentation, we discuss the simplest approximation for CDFT, i.e. homogeneous reference fluid approximation (HRFA) [5], and compare the distribution function of Ne liquid, i.e. typical Lennard-Jones fluid, from CDFT using HRFA and that from RISM (more precisely the Ornstein-Zernike equation) using hyper-netted-chain (HNC) and Kovalenko-Hirata (KH) closures.

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Development of program code for the first-principles photoemission-spectrum calculation

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We are developing a program code to calculate a valence-band photoemission spectrum of crystal by using first-principles methods.

As the first stage of the development, we employ a simple method in which the photoemission differential cross section of crystal, $P^{\hbar\omega}(\epsilon,\Theta)$, is estimated by the sum of the product of the atomic photoionization differential cross section (APhICS), $P^{\hbar\omega}_{\nu nl}(\Theta)$, and the partical density of states (PDOS), $D^{as1}_{\nu nl}(\epsilon)$:

For the photon energy, $\hbar\omega$,
$\hbar\omega = -\epsilon + W + K$
$-\epsilon$: Binding energy
W: work function
K: kinetic energy

Polarization direction

Θ

k
Electron direction

$$P^{\hbar\omega}(\epsilon,\Theta) \equiv \frac{d^2\sigma}{d\Omega d\epsilon} = \frac{d^2\sigma}{d\Omega dK} = \sum_{\nu nl} P^{\hbar\omega}_{\nu nl}(\Theta)D^{as1}_{\nu nl}(\epsilon) \ .$$

Here , we assume that the incident photon is linearly polarized, and the photoelectron is emitted in the direction of the angle, Θ, from the photon polarization direction.

Our poster will emphasize that:
(1) We have newly developed a program code for APhICS, which works at any photon energy of $\hbar\omega \lesssim 10^4 \text{eV}$.
(2) PDOS is evaluated in 'atomic sphere' instead of muffin-tin sphere.
(3) The calculated spectrum is compared with the experiment for $CuAlO_2$.

Impurity Diffusion via Nonadiabatic Charge State Change

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An impurity in material can diffuse through hopping via charge state change by carrier capture, known as, e.g., Bourgoin-Corbett process [1]. This process could be, especially, important for hydrogen (H) impurity because H atom can have large nonadiabatic coupling due to its light mass. Regardless of its possible importance, this process has not been discussed thoroughly due to experimental difficulty to detect this kind of diffusion and computational difficulty to treat non-adiabatic process.

In this study, we investigate the diffusion of H atom in Si via charge state change from first principles. The rate for charge state change was calculated with the Fermi's golden rule by considering nonradiative carrier capture via multiphonon process [2]. Within this formalism, anharmonic effect of phonon can be taken into account by imposing 1D approximation for ionic degrees of freedom and solving the Schrödinger equation for the 1D potential energy surface [3]. Treating anharmonic effect is crucial to study H diffusion because there exists long distance between stable sites for each charge state.

Figure 1 shows the calculated potential energy surface (PES) and the calculated rate for $H^- \rightarrow H^0 + e^-$ transition. The calculations were performed with DFT/HSE06 functional. The PES shows large anharmonicity due to large difference in H atom position between the two stable sites. Compared to the experimental rate [4], the calculated rate is underestimated by 1/100 and also shows 0.2 eV difference in its activation energy. With the calculated rate, we discuss the H atom diffusion in Si via charge state change.

Figure 1. Left: calculated potential energy surface for the transition between H^- and H^0. Right: calculated rate for the transition $H^- \rightarrow H^0 + e^-$.

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Thickness and bilayer stacking dependence on the electronic properties of HfX_2 (X = S, Se, or Te)

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Two-dimensional transition metal chalcogenides (TMDs) have become well-known because of their versatile and tunable physical properties for potential applications. However, studies on the electronic properties of layered Hafnium dichalcogenides (HfX_2, X = S, Se, or Te) is limited. In this study, we explored the thickness and bilayer stacking dependence on the electronic properties of HfX_2 thin films from 1 to 5 layers using first-principles calculations. Incorporating van der Waals (vdW) interaction, our bulk calculations reveal that the most stable phase is 1T compared to 2H and 3R phases. Results show that the bulk and layered structures of $HfTe_2$ are semi-metallic while those of HfS_2 and $HfSe_2$ are insulating. All structures exhibit decreasing band gap with increasing thickness. We also investigated six stacking orders in bilayer 1T-HfX_2. Varying stacking orders can tune the system band gaps with and without SOC by as much as 8, 18 and 38 meV for HfS_2, $HfSe_2$, and $HfTe_2$, respectively. Finally, by examination of the partial density of states, we find the bands above the fermi level are mostly characterized by d orbitals of Hf and the p orbitals of the chalcogen in the bands below the fermi level. This work manifests the tunable electronic properties of 1T-HfX_2, thus we propose these materials as potential candidates for device applications.

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Structure Search and Property Analysis of Interfaces between Cathode and Solid Electrolyte in All-Solid-State Battery via DFT-CALYPSO Method

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All-Solid-state batteries (ASSBs) is of great interest because of its high safety and high energy density in comparison to traditional liquid electrolytes based batteries. However, there is a crucial issue about the interface resistance between the cathode and electrolyte in many ASSBs, which hinders the performance of batteries. To understand the microscopic mechanism of the interface resistance, we systemically investigate the interface configurations between $LiCoO_2$ cathode and Li_3PS_4 sulfide solid electrolyte using CALYPSO structure prediction approach[1]. Several efficient techniques have been applied in our method, including an automatic toolkit for searching matched superlattice, a structure generation method including the lattice displacement, and PSO algorithm.

For the obtained energetically favorable structures, we calculated the electronic states and Li vacancy formation energies. We found that the interfacial layer is formed accompanied by the cation mixing ($Co \leftrightarrow P$ exchange) and anion mixing ($O \leftrightarrow S$ exchange). We then revealed that there are interfacial Li sites where preferential Li depletion can occur upon charging, even in very disordered interfaces. Besides, the present study demonstrated how to explore the structures of solid-solid interfaces at the atomic scale.

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Parameter in a Descriptor for Efficient Crystal Structure Search Using the Bayesian Optimization

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The crystal structure search tries to find the global minimum of a potential energy by repeating generation of candidate crystal structures and relaxation of them. The crystal structure relaxation is usually a computationally expensive part in the crystal structure search. Therefore, reduction in the number of structure relaxations is important for an efficient search. Many methods have been developed with such motivation, and recently, a method using the Bayesian optimization (BO) has been developed [1]. BO is a machine learning technique for solving the minimization problem of a black-box function. The crystal structure is expressed by a numerical vector called descriptor, which often has predetermined parameters. The parameters in the descriptor would affect the efficiency of the crystal structure search because they change an input of BO.

We investigate dependency of the efficiency of the crystal structure search on parameters in a descriptor. Figure shows the number of structures required until the most stable structure is found. The result suggests that there exists an appropriate parameter value for an efficient search and the efficiency is worse than that of the random search in some cases. We also introduce an information measure based on a distribution of descriptors and confirm that the maximum of the measure corresponds to the parameter value presenting the most efficient search.
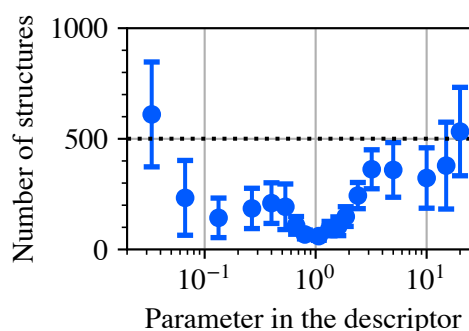

Fig. Number of structures required until the most stable structure is found among 1000 structures of Si_{16}. The horizontal dotted line is the mean of the random search.

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Theoretical Investigation on Vibrationally Enhanced Hydrogenation of CO_2 on Cu(111)

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Adsorption and reaction of CO_2 on solid surfaces are attracting enormous interest because of their importance in industrial, energy and environmental management. To clarify reaction mechanisms and to identify important factors governing the reactivity of CO_2 on solid surfaces are very important to develop more efficient catalysts or catalytic processes for utilization of CO_2. To this end, we investigated CO_2 adsorption, dissociation, and hydrogenation processes on Cu surfaces using van der Waals density functionals as implemented in our home made STATE (Simulation tool for Atom TEchnology) program code[1-5]. We theoretically proposed that the excitation of the O-C-O vibrational bending mode of CO_2 enhances the hydrogenation process of CO_2 to formate on Cu(111) [1] and very recently, this has been experimentally verified [3]. We discuss the detail of the reaction process clarified by first-principles simulations and compared with experimental results.

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Crystal structure prediction of Li(CB$_9$H$_{10}$): an example of XRD-data-assimulated molecular dynamic simulation

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All-solid-state batteriy is one of the most promising candidates to resolve the intrinsic drawbacks of current lithium-ion batteries, such as electrolyte leakage, flammability, and limited energy density. Recently, a class of complex hydrides, represented as Li(CB$_9$H$_{10}$) and Na(CB$_9$H$_{10}$), is paid considerable attention, because of its high ionic conductivity and stability against metal anode [1]. Conductivity measurement and structure analysis suggest that the disorder of (CB$_9$H$_{10}$)$^-$ anions plays a critical role of this high ionic conductivity, whereas the exact orientation of these anions has not been clarified yet. Under such circumstance, recently, our group develop an efficient approch to predict the crystal structure by combining molecular dynamic (MD) simulation and data-assimulation utilizing XRD pattern [2]. Here, we report the result of crystal structure prediction of Li(CB$_9$H$_{10}$) based on the XRD-assited MD simulation.

As a test calculation, the configurations of (B$_{10}$H$_{10}$)$^{-2}$ anions in the reported crystal strucutre of Li$_2$(B$_{10}$H$_{10}$) [3] has been predicted from the random configuration of (B$_{10}$H$_{10}$)$^{-2}$ anions with Reax-FF potential and XRD pattern of Li$_2$(B$_{10}$H$_{10}$). these XRD-assisted molecular dynamic simualtions could predict the configurations of B$_{10}$H$_{10}$ in Li$_2$(B$_{10}$H$_{10}$) crystal 8 times out of 50 simulations, suggesting that this XRD-assisted MD simulation potentially could predict the crystal structure of Li(CB$_9$H$_{10}$). 4 out of 50 MD simulations for (CB$_9$H$_{10}$)$^-$ anions with XRD pattern of Li(CB$_9$H$_{10}$) show the hcp-like trigonal structure as shown in Figure 1. (Note that the positions of Li$^+$ ions are determined by subsequent XRD-assisted *ab initio* MD simulations.) Although more than 60% of simulations show the almost same center of gravity potisions of CB$_9$H$_{10}$$^-$ anions as that of figure 1, the orientation of anions is different and the obtained XRD pattern less agrees with the experiment. In addition, it is confirmed that the potential energies of those simualtions are almost comparable suggesting the effectiveness of data-assimulation to find the crystal structure where the orientation of cluster is important.

Figure1. Li(CB$_9$H$_{10}$) structure prediceted from XRD-assisted MD simulation

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Band Structure of Group IV-VI 2D Materials

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We carry out first principle calculations on group IV-VI 2D materials and clarify band structures based on theoretical analysis. We study systems consisting of group IV (A = C, Si, Ge) and group VI elements (B = O, S, Se, Te) and find that they form buckled puckered geometries (Fig.1). We find that the highest valence bands have anion p characters and the lowest conduction bands have cation p characters. We analyze the band structures based on the group theory: The band structure of CSe is shown in Fig.2, where we use Mulliken symbols to represent irreducible representations except for the S-Y line since all the bands are doubly degenerated and the characters do not correspond to those of the Mulliken symbols. We find that the bands are also doubly degenerated on the X-S line, where the two bands belonging to different representaions have the same energy. We conclude that all the bands are doubly degenerated at the boundaries (the X-S-Y line) of the first Brillouin zone and the degeneracies are due to the symmetry ($Pmn2_1$) of the materials. Previous study clarify that this degeneracy at the zone boundary also occurs in the group V 2D materials [1].

Fig.1. Atomic structure. Fig.2. Band structure of CSe.

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First principles calculations of magnetocrystalline anisotropy of doped antiferromagnetic transition metal oxide

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Interest in antiferromagnetic materials (AFMs) is growing rapidly in technological applications, especially in the terahertz region of optoelectronics [1]. In analyzing the antiferromagnetic resonance at such high frequency, one of the important factors is magnetocrystalline anisotropy (MCA), and therefore, adjustment of the MCA may be a viable way to improve the efficiency of the AFM optoelectronic device. Despite its importance, MCA in AFMs is not well understood, as opposed to MCA, in a ferromagnetic material. Here, we performed a first principles calculation of the MCA energy of doped NiO using the full-potential linearized augmented plane wave method [2] based on generalized gradient approximation and explained the dopant dependence on MCA. The electron correlation effect at the Ni site was treated by using the DFT+U scheme using the effective on-site Coulomb interaction parameter Ueff [3]. The MCA energy was obtained by the total energy difference from spin-orbit coupling, using the force theorem [2] of magnetization along the [001], [110], and [111] directions. In the non-doped NiO, i.e., the pure NiO, although the magnetic easy axis is known from experiments to be along the [001] direction, it is difficult to determine the easy axis within the present computation accuracy. However, by doping $3d$ elements and non-magnetic elements, the MCA is found to change depending on the elements, compared to that of the non-doped NiO. Details including the mechanism of the doping effect on MCA will be discussed.

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First-principles Study of Anomalous Nernst Effect in The 2D Ferromagnetic Half-metal 1T-FeX$_2$ (X = Br, Cl)

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The discovery of magnetism in two-dimensional material [1] is open a way for realizing thermoelectric based anomalous Nernst effect (ANE). In two-dimensional system, thermal conductivity is decreased and electrical conductivity [2,3] is increased which can generate high power factor and figure of merit of thermoelectric. In this study, we performed first-principles calculations to estimate anomalous Nernst coefficient (ANC) of 2D ferromagnetic half-metal 1T-FeX_2 (X = Br, Cl) monolayer with OpenMX code [4]. We obtained large ANC around the Fermi level. The origin of large ANC originates from the asymmetric Fermi energy dependence of anomalous Hall coefficient (AHC) which can be explained by band filling and band-decomposed Berry curvature around the Fermi level. The asymmetry of AHC also tend to has the same trend with Skyrmion crystal which posesses giant ANC[5,6].

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First principals study for charge conductivity change due to the spin orbit coupling effect in metallic bilayer film system

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Understanding the transport phenomena in magnetic materials has been a crucial ingredient for designing novel materials with desired properties for spintronic applications. In bilayer systems of magnetic material (MM) and heavy metal (HM), the electrical resistance of the heavy metal layer can be tuned by varying the angle between the applied current and the magnetization direction of the magnetic material layer [1]. An effect commonly known as the spin Hall magnetoresistance (SMR) effect [2]. This attractive magnetic proximity effect (MPE) has been widely investigated in experiment. On the contrary, there are only few reports on the theoretical side. In the present work, we use the theoretical approach to study the SMR by using the first-principles calculation, full-potential linearized augmented plane wave method (FLAPW) [3] in order to design materials with high efficiency of SMR. We employed Pt|Co bilayer film system as our calculation model. In reference to previous work [4], the single layer Co is stacked on top of Pt substrates at the fcc site. Calculation of electric conductivity and spin Hall conductivity was based on linear response theory and Kubo formula [5]. We considered the electric conductivity and spin Hall conductivity while varying the magnetization directions in the system from x to z, where the z direction is perpendicular to the film plane. As a result, the resistance of the system increased by almost 2% when the magnetization direction is perpendicular to the plane compared to the in-plane magnetization case. We will present discussions with the result of charge conductivuties dispersion on the fermi surface and conduct analysis based on remarkable changes on the fermi surface.

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Crystal Orientation Dependence of Lead Vacancy Stability and Defect State on PbTiO$_3$ Grain Boundary

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Lead-based ferroelectric materials, such as PbTiO$_3$, has been recently expected to be applied in the aerospace and automotive fields, which involve severe environmental conditions. For example, automotive device standard requires the qualification of a 1000 h lifetime at 85 to 150℃. These materials typically contain vacancies, which are caused during sintering or deposition processing, and contribute to resistance degradation and leakage current. Leakage current results in the degradation of piezoelectric properties. Lead-based ferroelectric materials typically contain lead vacancies at grain boundaries because lead evaporation from grain surfaces can easily occur due to high partial pressures of lead during processes. The relation between the grain boundary conditions and lead vacancies is not clear, although it is a key to determine the performance of piezoelectric devices.

Here, we calculate the formation energies of lead vacancies (E_f^{Pb}) and the electronic structures of the PbTiO$_3$ grain boundaries using first-principles calculations based on density functional theory. PbTiO$_3$, which is a member of the PZT family, is a simple and technologically important ferroelectric material. We focus on the crystal orientation dependence of the formation energies and electronic states of lead vacancies at the grain boundaries.

Our results show that E_f^{Pb} of grain boundaries are lower than that of bulk. This indicates that lead vacancies can easily occur at grain boundaries than in bulk. The variation of E_f^{Pb} is larger at the grain boundaries with the misalignment polarization axes than that with the matched polarization axes. Especially, the grain boundary between orthogonal two grains has the negative E_f^{Pb}. This indicates that the polycrystal PbTiO$_3$ which have the orthogonal polarization axes contains a lead vacancy at the grain boundary stably.

In addition, the energy gap between the defect level in the band gap and the valence band maximum(VBM) is lower at the orthogonal grain boundary. The state in the band gap is the localized state at a part of the grain boundary, while the state of the VBM is delocalized at the grain boundary. When a hole is excited from the defect level, it can migrate through the grain boundary. It is concluded the low crystal orientation PbTiO$_3$ can easily contain lead vacancies that contribute to leakage currents at the grain boundaries.

Ab initio study of contact behaviors at 2D/3D GaX(X = S, Se, Te)/Si(111) heterojunctions

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For utilization of two-dimensional (2D) materials in nanoelectronics devices, it is necessary to investigate mixed-dimensional van der Waals (vdW) heterostructures combining those diverse 2D materials with conventional 3D ones. In this study, we have investigated atomic and electronic structures of heterostructures that consist of 2D GaX (X = S, Se, Te) and three-dimensional (3D) Si using density functional theory calculations. Especially, we focus on different contact behaviors at heterojunctions between GaX and Si depending on two types of the Si(111) surface termination. It is found that GaX/clean-Si(111) (c-Si(111)) and GaX/H-covered-Si(111) (H-Si(111)) heterostructures show semiconductor/metal and semiconductor/semiconductor contact behaviors, respectively. GaS and GaSe form n-type contact, and GaTe forms p-type contact with c-Si(111). In these cases, Fermi level pinning occurs at GaX/c-Si(111) interfaces with metal induced gap states. In the case of GaX/H-Si(111), we identify that the interfaces form p-n heterojunction, especially with type II band alignment for GaS and GaSe. From the understanding of contact behaviors between GaX and Si(111), we suggest the possibility of their applications in nano- and opto-electronic devices.

Magneto-optical conductivity in 5d transition metal by first principles calculations

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There is growing interest in spin-polarized electrons which play a central role in spintronics, in particular for optoelectronics device application using circularly polarized light. At an earlier stage in this field, an attention was paid to a spin-polarized electron emission from semiconductor heterostructures, [1] e.g., by using a strained GaAs thin-layer [2] at earlier stage in this field. However, more efficient photo spin current conversion is required for practical applications. For such purpose, designing novel materials with enhanced optical properties is inevitable, and first principles calculations are known to be an important tool. In the present work, the optical conductivity to spin currents for 5d transition metal (i.e. Lu, Hf, Ta, W, Re, Os, Ir, Pt, Au), were investigated. Self-consistent calculations were carried out on the basis of the generalized gradient approximation [3] by using full-potential linearized augmented plane wave method. [4] The conductivity parameters as function of photo energy were evaluated on the basis of Kubo-formula [5] in the linear response theory by using the calculated eigenvectors. For 5d transition metal, we find that the intensity of the optical conductivity significantly increases due to the large spin-orbit-coupling. For example, the intensity of the optical conductivity to spin currents in Pt ranging from 1 eV to 4 eV of photo energy is larger by one order of magnitude compared to that of GaAs. More details including the mechanism will be presented.

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Spin-Polarized Cation Vacancies in Compound Semiconductors: First-principles Study

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Spontaneous magnetism in wide-gap compound semiconductors has attracted scientific interests over the recent years. This d^0 magnetism is induced by cation vacancies which cause the spin polarization [1,2,3,4]. In this paper, we study spin-polarized isolated cation vacancies in wide-gap compound semiconductors by using first-principles calculations. Oxides (ZnO and BeO) and nitrides (BN, AlN and GaN) are studied. We find that the vacancies have the C_{3v} symmetry and are spin-polarized. Four near anions are displaced outwardly and therefore the bond angles of the anions become larger than the sp^3 bond angle. Due to this relaxation, the spin density is localized and thus the spin-polarized state is stabilized (Fig. 1). We clarify that two majority spin electrons occupy the E level in the case of II-VI semiconductors and the magnetic moment is 2 μB. On the other hand, three majority spin electrons occupy the E and A_1 levels in the case of III-V semiconductors and thus the magnetic moment is 3 μB. The Jahn-Teller distortion which lowers the symmetry from C_{3v} to C_s is found not to lead to the most stable geometry. This small Jahn-Teller effect is due to the outward relaxation of the four anions. We therefore conclude that the spin polarized C_{3v} geometry is the most stable.

Fig. 1. Spin density of Be vacancy in BeO.

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Band alignment in graphene/WSe$_2$ heterostructures before and after oxygen plasma treatment

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Recently, two-dimensional (2D) materials have been actively studied in various fields. It is known that graphene, one of the 2D materials consisting of carbon atoms, has excellent electron mobility and permeability. Despite these excellent properties of graphene, one drawback in use of graphene in the electronic devices is that electron mobility of graphene is considerably reduced when the band gap is generated. To overcome such drawback, diverse 2D semiconducting materials have been investigated including transition metal dichalcogenides (TMDs). Here, using density functional theory (DFT) calculations we have studied atomic and electronic structures of heterostructures between graphene and TMDs such as graphene/WSe$_2$ and graphene/WO$_2$/WSe$_2$ Especially, we focus on the band alignment in those heterostructures by noting that WSe$_2$ in graphene/WSe$_2$ can change to WO$_2$ by oxygen plasma.

Implementation of computing anomalous Hall conductivity for high throughput screening of magnetic thermoelectric materials

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The thermoelectric conversion based on the anomalous Nernst effect (ANE) has attracted attention because the ANE realizes the high-density integration more easily compared to that based on the Seebeck effect [1]. The ANE is the phenomenon that the electric power is created along the direction perpendicular to both the temperature gradient and the magnetization in magnetic materials. This effect is induced by the anomalous Hall conductivity (AHC), and if the AHC changes drastically as a function of the Fermi level, we expect the large ANE [2]. In our previous first-principles study, we predicted that an electron doped EuO skyrmion crystal shows the large ANE induced by the finite Chern number at the Fermi level [3]. For discovery and design of new materials which have the large ANE via high throughput screening, we need an efficient computational method to investigate the Fermi level dependence of the AHC.

In this study, we implemented the code of computing the AHC applicable to metallic systems in OpenMX package [4] without constructing Wannier functions. By improving Fukui-Hatsugai-Suzuki mehod [5] which calculates the Chern number in the insulating system, we have successfully obtained the Fermi level dependence of AHC comparable to those calculated by Wannier90 package [6]. In this poster session, we introduce how to implement this scheme and calculated results of the AHC in two-dimensional ferromagnetic materials [7] and bcc Fe [8].

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H_2 Nuclear Spin Transition on Metals: Physisorption Vs. Molecular Chemisorption

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It has been known that physisorbed hydrogen undergoes ortho-para (o-p) conversion in the order of minutes on non-magnetic metal surfaces such as Ag(111)[1]. This has been described by the virtual surface-molecule electron transfer and the hyperfine Fermi contact interaction within second order perturbation theory. Recent experiments have shown that on Cu(510)[2] and Pd(210)[3] surfaces, H_2 is in the so-called molecularly chemisorbed state and behaves as a 2-dimensional quantum rotor, while the o-p conversion rate is 1 order magnitude faster than on flat surfaces.

In this work, we investigate the o-p conversion of hydrogen in these two distinct adsorption states on metal surfaces by means of first-principles calculations based on the density functional theory (DFT). We revisit the o-p conversion of physisorbed H_2 and subsequently compare with the molecularly chemisorbed case. Physisorption and chemisorption states are modelled prototypically by H_2/Pd(111) and H_2/Pd(210) systems. The spin transition probability is calculated by perturbation theory using the parameters derived from DFT calculations[4], the details of which will be presented at the workshop.

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Large-scale DFT study on Pd@Ag core-shell nanoparticles

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Large-scale density functional theory (DFT) can play an important role to clarify the active sites of catalytic reactions in metallic nanoparticle systems by investigating the site- and size- dependence of atomic and electronic structures and reaction properties. Our large-scale DFT code CONQUEST [1] can treat large systems containing more than several thousands of atoms by using local orbitals and a linear-scaling (O(N)) method. CONQUEST achieves O(N) by using the locality of density matrices with the density matrix minimization method. Local orbitals, which are called support functions, are used to express the density matrices.

The computational cost scales cubically to the number of the support functions, both in the O(N) and the conventional diagonalization calculations. Therefore, to reduce the number of support functions without losing accuracy, we have introduced multi-site support functions (MSSF) [2]. MSSFs are the linear combinations of pseudo-atomic orbitals from a target atom and its neighbor atoms in a cutoff region,

$$\phi_{I\alpha}(r) = \sum_{K}^{neighbours} \sum_{\mu \in K} C_{I\alpha,K\mu} \chi_{K\mu}(r)$$

MSSFs correspond to local molecular orbitals so that the number of required support functions can be the minimal. While the O(N) method is not guaranteed to be applicable for metallic systems, the multi-site method can be applied for metallic systems stably.

In this study, we have investigated the selective semihydrogenation of alkynes with core-Pd/shell-Ag (Pd@Ag) nanoparticles by using the multi-site method. Pd@Ag accelerates the hydrogenation from alkyne to alkene but prevent the overhydrogenation from alkene to alkane [3]. To clarify the catalytic mechanism of Pd@Ag, we investigated the atomic and electronic structures of Pd, Ag and Pd@Ag nanoparticles. It is found that the surfaces of Ag nanoparticles are charged negatively, while that of the Pd nanoparticle is partially charged negatively. We will also report the electronic structure dependence of Pd@Ag on the Ag-shell thickness and the effect from the Pd/Ag interface to the particle surface.

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Thickness dependent electronic structures of Pd dichalcogenides: a first principles study

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Among the family of transition metal dichalcogenides (TMDs) [1], Pd-based TMDs have been one of the less explored materials. In this study, using first-principles calculations, we investigate the electronic properties of PdX_2 (X = S, Se, and Te) with respect to film thickness. With regards to the structural stability, the bulk and thin film (1 to 5 layers) structures of PdS_2 and $PdSe_2$ exhibit pyrite, while $PdTe_2$ exhibit 1T as their most stable configurations. For the electronic properties, the most stable bulk configurations demonstrate semi-metallic features, while their corresponding monolayer structures are insulating with band gaps of 1.14, 1.43, 0.12 eV for pyrite PdS_2 and $PdSe_2$, and 1T $PdTe_2$, respectively. For the band properties, we observe that all these materials manifest decreasing/closing of indirect band gap with increasing thickness. Moreover, all the stable monolayer band structures exhibit flat bands and diverging density of states near the Fermi level, indicating the presence of van Hove singularity [2]. Our results show the sensitivity and tunability of the electronic properties of PdX_2 for various potential applications.

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First-Principles Study of the Surface Alloys
M/Ag (111)-$\sqrt{3}\times\sqrt{3}$ R 30°

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Surface alloys are synthesized by inserting atoms into the surface. This topic has attracted a lot of research attention lately because of some of its applications, such as spintronics [1], where there is a giant spin splitting in Bi/Ag (111) surface alloys. Many types of surface alloys have been studied: for example, Ge / Ag (111) [2], Pb / Ag (111) [3], Bi / Ag (111) [4, 5], Sn / Ag (111) [6] . One structure that has been observed in experiments with surfaces is -$\sqrt{3}\times\sqrt{3}$ R 30°. Structural studies such as surfaces alloys can provide important information for forming metal-semiconductors interface and metal interfaces. The metal-semiconductor interface is very important to industry, for example, the Si / Ag interface is a promising candidate for solar cells [7].

In this study we systematically explore the structural stability of M / Ag (111) surface alloys with first-principles calculation using the OpenMX code [8], where M is group III (B, Al, Ga, In, Tl), IV (C, Si, Ge, Sn, Pb) and V atom (N, P, As, Sb, Bi). We focus on the corrugation parameter d which is determined by the height of the M atom from the Ag atom in the plane of the top-most atom. We introduce concept of atomic radii by corrugation in surface alloys. The formation energy for each M atom is also calculated to determine the stability of the atoms on the alloy surface. We will discuss the dependence of M atoms on corrugation parameters.

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Diffusion of Carbon in α-Fe in the Presence of Vacancy

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Diffusion of carbon in iron is one of important phenomena and strongly linked to processes of production of steels such as formation of iron carbide, phase transformations, and so on. In this work, the diffusion and clustering of interstitial carbon atoms in α-iron are studied taking into account their interaction with vacancy (V) by using the molecular statics and kinetic Monte Carlo (kMC) methods. Firstly, the stability of carbon atoms in a 8×8×8 bcc super-cell was examined via total energy optimization. Secondly, the diffusion mechanisms and jumping rates for carbon interstitials were analyzed and calculated. Finally, by using the calculated jumping rates as input parameters for an atomistic kMC program, diffusion coefficients of carbon depending on temperature were estimated to study the influence of vacancy on the diffusion and clustering of carbon in bulk iron. We found that in perfect α-iron lattice, carbon atoms tend to form stable C-C pairs where two atoms in the pair are slightly shifted off the octahedral sites and separated by a distance of 1.537Å. While the presence of vacancies leads to the formation of larger vacancy-carbon clusters with the most stable structure being VC_2. The results calculated by new Fe-C Tersoff potential [1] show that a single vacancy can accumulate up to 3 or 4 carbon atoms, being consistent with those from first-principles calculations. In addition, due to the presence of vacancies, the diffusion paths of carbon are strongly modified. The kMC simulations show that the diffusion coefficient is decreased as the vacancy content increases.

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Hydrogen on Reconstructed Gold Surface

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Hydrogen is the lightest element and this causes interesting quantum effects such as quantum confinement, tunneling, and nonadiabatic effect. On metal surfaces, hydrogen tends to delocalize at low temperature and tunnel diffusion dominates over thermally activated diffusion. According to a recent first-principles calculation of hydrogen diffusion in a crystalline palladium, the quantum behavior appears more apparently as the lattice constant is reduced[1]. If this occurs similarly on a surface, catalytic reactions may be controled by changing the lattice constant. In this context, we focus on the Au(111) surface. The surface is known to reconstruct resulting in an increasing of the density of surface atoms, [2] which can make quantum effect stronger. With this in mind, we calculated the reconstruction and the adsorption of hydrogen on gold surface. From the calculation, it is revealed that the density of the surface atoms increases by 4% after the reconstruction. Also, hydrogen tends to adsorp on the hollow site rather than the top site, while on platinum and palladium surfaces hydrogen is known to adsorb on both sites. On the basis of this perculiar property of Au(111), in the poster, we will discuss possible quantum effects.

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Electronic stress tensor density based on the quantum field theory in surface material systems

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Development of the observation technology such as Atomic Force Microscopy (AFM) has made a paradigm shift from researches about bulk feature in solids to investigation of surface science. In particular, in order to understand the interaction between a material surface and a tip of AFM, it is important not only to evaluate evaluation of the energy of the whole system and band structures but also to investigate the local physical properties and the nature of chemical bonding which are caused by change of a part of atomic structures and local electronic structures.

The purpose of this study is to investigate the physical and chemical nature around the interaction region betweeen a solid crystal and a molecular cluster from the picture of "action through medium" based on the local physical quantities such as electronic kinetic energy density and electronic stress tensor density[1], which are defined in the framework of relativistic quantum field theory. Specifically, we performed quantum physical property analysis[2] based on the local physical quantities by using wave packets which are obtained by the electronic structure calculations based on DFT[3]. We show some pictures of electronic stress tensor density in simple atoms, molecules and crystals, and discuss the nature of the electronic stress tensor density in the interaction region between a material surface and a tip of AFM.

Fig. Distribution of maximum eigenvalues and eigenvectors of the electronic stress tensor density and zero surface of the electronic kinetic energy density in the region between Si tip and $\sqrt{7} \times \sqrt{7}$ Si(111) surface.

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Development and Validation of a Neural Network Potential for Identifying Stable Phases in the Y-Ti-O System

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Yttrium tantalates are candidate materials for use as coatings of gas turbine engines to protect them from chemical and thermal corrosion [1], and phase diagrams of this system are important for understanding their thermochemical stabilities and compatibilities. However, it is not easy to generate phase diagrams from first principles by exhaustively performing density functional theory (DFT) calculations because of the high computational cost. Recently, there has been remarkable progress in the development of artificial neural networks (ANNs) and these can be used to generate accurate but computationally unexpensive potentials for modeling crystal structures. With the exception of rutile TiO_2, the crystal structures of oxides in Y-Ti-O system can be regarded as consisting of a double-sublattice of cation and anion sites. In this study, we develop an ANN potential to search for ground states in the Y-Ti-O system and validate it by comparing with results of DFT calculations.

First, we generated all unique configurations in cells with up to 12 sublattice sites for each, while enforcing charge neutrality conditions, using the Alloy Theoretic Automated Toolkit (ATAT) [2]. Next, we performed ionic relaxation of these structures using DFT as implemented in VASP [3,4]. We then performed data cleansing on the relaxed structures based on displacements. As the final step, we trained the ANN potential using 90% and 10% of these structures as training data and testing data, repectively, using The Atomic Energy NETwork (ænet) code [5].

The convex hull of Y-Ti-O energies calculated using the ANN potential was found to coincide with stable phases from experiment and DFT calculations. Our analysis shows that for composition Y:Ti=1, anion vacancies form more easily than cation vacancies. The results confirmed that our ANN potential can reproduce phase stabilities in the Y-Ti-O system with sufficient accuracy.

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Intrinsic and anisotropic Rashba spin splitting in Janus monolayer transition metal dichalcogenides MXY (M = Pd, Te; X/Y = S, Se, Te)

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Recently, studies on Janus monolayer transition metal dichalcogenides (TMDs) [1,2] have demonstrated the feasibility of synthesizing 2D materials which possess both in-plane inversion and out-of-plane symmetry. Here, we performed a systematic first-principles and phonon calculations to study the spin-orbit-induced anisotropic Rashba splitting and structural stability in Janus monolayer TMDs with chemical formula *MXY* (*M* = Pd, Pt and *X,Y* = S, Se, Te). Phonon dispersion calculations showed that PdSSe, PtSSe, PtSTe, and PtSeTe are dynamically stable. With regards to the spin-orbit-induced anisotropic Rashba splitting, we observed Rashba splittings both in the M-Γ and M-K directions for all the stable *MXY* structures. For M-Γ, the Rashba parameters (α_Γ^R) are equal to 0.068, 0.407, 0.418, and 0.058 eV•Å for PdSSe, PtSSe, PtSTe, and PtSeTe, respectively. Meanwhile, for M-K, the Rashba parameters (α_K^R) are equal to 0.203, 0.372, 0.455, and 0.276 eV•Å for PdSSe, PtSSe, PtSTe, and PtSeTe, respectively. Moreover, we found that the Rashba splitting can be significantly enhanced through compressive strain. Our findings indeed show that Pt and Pd Janus TMDs demonstrate the potential for spintronics applications

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Effect of External Electric Field on Methane Conversion on IrO$_2$(110) Surface: A Density Functional Theory Study

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Catalytic conversion of methane to value-added chemicals is a promising application for gas versatility. [1-3] In this work, we have investigated the methane oxidation over oxygen-rich IrO$_2$ (110) surface by DFT calculations, as IrO$_2$ is reported to be an effective catalyst for activating the C-H bond of methane. Compared to the methane reaction on the surface of stoichiometric IrO$_2$ (110), the reaction barrier for each step of forming formaldehyde on the oxygen-rich IrO$_2$ (110) is small. The calculations show that formaldehyde formation is the most favorable route in methane oxidation, but this process is limited by the high desorption energy of formaldehyde. To modify the reactivity of IrO$_2$ (110), we conducted a study of the influence of an external electric field on the methane conversion reaction. The calculations show that the effects of external electric field on methane dehydrogenation and C-O coupling reactions is not so apparent. However, it is found that the desorption energy of the adsorbates can be regulated by applying an external electric field. Our study indicates that the use of an external electric field is crucial in regulating the catalytic reaction, and especially the application of a positive electric field promotes the oxidation of methane to formaldehyde over oxygen-rich IrO$_2$ (110) surface.

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Anisotropic Mechanical and Thermal Properties of Two Dimesional Borophene

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The study of two dimensional (2D) materials, such as graphene, hexagonal boron nitride, and borophene is one of the most exciting and prolific areas in material science in recent years. The recent synthesis of an atomically thin boron sheet (i.e., borophene) on an Ag(111) substrate with a buckled triangular structure has aroused increasing interest because of its exceptional anisotropic properties. Since then, intensive theoretical studies have been performed for various thermally stable borophene phase.

The mechanical and thermal properties of asymmetric washboard borophene were studied using first principles calculations. We first calculated the elastic constants derived from the second derivative of total energy with respect to the in-plane strain at equilibrium are. Then, its mechanical properties, in-plane Young's modulus Y, and Poisson's ratio P were calculated. Furthermore, based on the second and third order interatomic force constants obtained from the super-cell method, the lattice thermal conductivity along x and y directions at room temperature were calculated. Upon obtaining thermal conductivity results from ab-initio lattice dynamics calculations, we perform molecular dynamics (MD) simulations, which is popular tool to calculate lattice thermal transport properties. Using classical nonequilibrium molecular dynamics (NEMD) simulations and lattice dynamics (LD) calculations, we also demonstrated the length-dependent lattice thermal conductivities of borophene from the ballistic to the diffusive thermal transport regime. The obtained results show that the mechanical properties and lattice thermal conductivity of borophene exhibit anisotropic feature. These findings provide important guidelines for the practical applications of borophene in nanodevice applications.

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Evaluation of solid-solution strengthening of alloys

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Evaluation of mechanical properties of materials by first-principles calculations is still in its infancy in spite of technological imporatance. Among the mechanisms for strengthning of metallic structural materials, solid-solution strengthening is effective at high temperature, for which the increase in the yield stress $\Delta\sigma$ can be evaluated using conventional theory of dislocation. Materials of practical importance used at high temperature include iridium and Ti-Al. Focusing on iridium-based alloys, it has been reported that hafnium and zirconium show large solid-solution strengthening in the high temperature region around 1950 ℃ [1]. However, there have been few studies on further exploration for the second element effective for strengthening of iridium.

In this study, various kinds of additive element for iridium are examined computationally with different concentrations by first-principles calculations based on density functional theory with norm-conserving pseudopotentials and pseudo-atomic localized basis functions as implemented in the OpenMX code [2]. The GGA-PBE exchange-correlation functional is adopted [3]. In solid-solution strengthening, the dislocation motion is inhibited by a strain field on the atomic scale formed by the difference in the sizes between a solute atom and a solvent atom. Thus, the misfit strain for each concentration is obtained using the change in the interatomic distance due to alloying. The figure of merit for the solid-solution strengthening, the increase in the yield stress $\Delta\sigma$, which is shown in Fig. 1, agrees well with the experimental tendency [1].
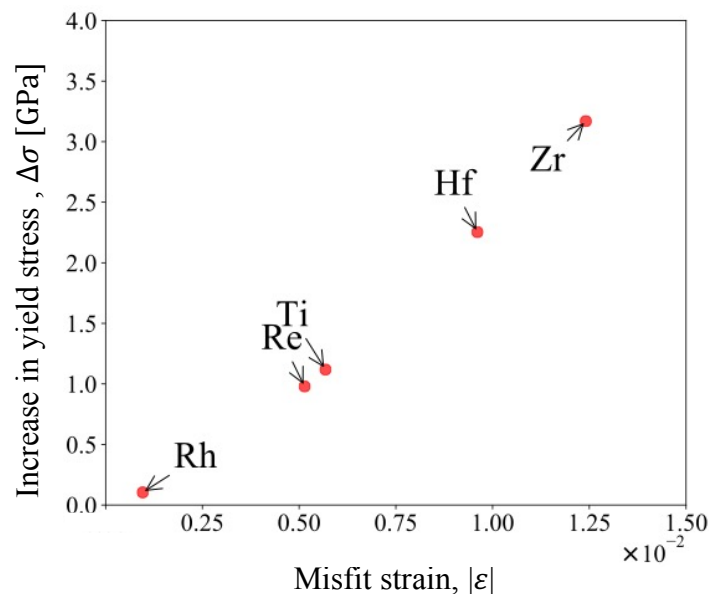

Fig. 1. Solid-solution strengthening of Ir with 0.93at% of X (X = Rh, Re, Ti, Hf, and Zr)

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Jx: An open-source software for calculating magnetic interactions based on magnetic force theory
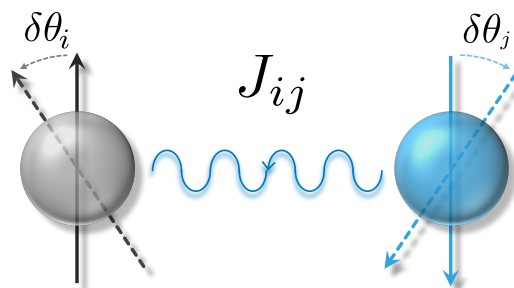
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Magnetic interactions in solids are often described in terms of effective spin Hamiltonians, $H = -S_i J_{ij} S_j$, and the determination of J is the key step to study magnetism and related phenomena. One way to calculate the J from first-principles based on linear response theory is often called magnetic force theorem (MFT) [1,2]. Here, we present our newly-developed open-source software, named by *Jx* [3,4], to perform MFT calculations. *Jx* is a user-friendly and efficient tool to calculate magnetic interaction in solids and molecules. Without supercell calculation, it computes both short- and long-range interactions. It is also possible to calculate an orbital-resolved matrix form of magnetic couplings.

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A Study about the Possibility of Reducing Coercive Field in Pristine HfO$_2$ from the Ab-initio Calculation

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The discovery of ferroelectricity in HfO$_2$ has led to many attempts to utilize HfO$_2$ in memory devices. However, the large coercive field (E_c) of HfO$_2$ is still one of the challenging issues for HfO$_2$-based devices. In recent years, to solve this problem, several studies for reducing E_c by using dopants have been reported, and among them, Si doped HfO$_2$ have dramatically reduced E_c. Despite these studies, doping processes require high processing temperature, which near 580K [1] and it makes doped HfO$_2$ difficult to use in industrial area. Therefore, here, we studied how to reduce E_c of HfO$_2$ without doping process with intuition from the crystal structure of Si-doped HfO$_2$. From Ab-initio calculations, we figured out the structural property of Si doped HfO$_2$ and found that the unique bonding character of Si dopant, which favors sp3 bonding, chemically induces internal strain on the lattice. We calculate how the ferroelectricity of HfO$_2$ responds to strain, and how strain affects the energy barrier of polarization switching. From the calculation results, we find that the polar symmetry is almost preserved and the energy barrier is reduced isotropically within the appropriate strain. Finally, we discuss how the strain affects on the energy barrier through structural analysis of the intermediate state in the polarization switching path and further study the origin of these phenomenon through strain-phonon coupling analysis.

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First-principles Fully Relativistic Calculations of Insulators Under Finite Electric Fields by Using the LCAO Method

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In density functional calculations, when one consider applying electric fields to bulk systems under periodic boundary conditions, it is possible to perform self-consistent field calculations of electronic states under electric fields by introducing effective potential based on the modern theory of electric polarization using the Berry phase method [1-3]. It is useful in investigating physical properties of materials under electric fields and obtaining physical quantities such as Born effective charges and dielectric constants. There are various applications of the effective potential to vacancies in oxides [4], ferroelectric transitions in molecular crystals [5], and magnetoelectric effects [6].

We have implemented the effective potential of finite electric fields to the OpenMX code [7, 8], which utilizes linear conbination of pseudo atomic orbitals as the basis set. Our implemented code is applicable to various systems from fundamental materials to materials where fully-relativistic effects are important. By using the code, Our calculated values of the Born effective charge, electronic dielectric constant and static dielectric constant for a Mg atom in MgO bulk are 1.96, 2.97 and 9.57, respectively. We will explain how we implemented it and discuss various applications of it.

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Cu$_4$ Complex in Silicon and Impurity Reaction between Cu$_4$ and Hydrogen

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In semiconductor manufacturing, defects of copper impurities in silicon often have deleterious effects. On the other hand, some of these defects are useful. One such defect is one that emits photoluminescence at 1.014 eV. Since this photoluminescence emission is sharp, this defect can be a promising candidate for an optical device. High-resolution optical spectroscopic experiments using copper radioisotopes revealed that this defect consists of four copper atoms [1]. The Cu$_4$ complex is stable, but it is observed that Cu$_4$ is easily decomposed when hydrogen is added. This is undesirable for applications, and it is necessary to clarify the defect reaction between Cu$_4$ and hydrogen for practical use.

In this study, first-principles calculations are used to investigate the reaction process between Cu$_4$ and hydrogen in silicon. We also reviewed the most stable structure of Cu$_4$ defect in silicon. Previously, the Cu$_4$ defect structure was proposed to be Cu$_{(s)}$Cu$_{3(i)}$ with one copper atom at the substitutional site and three copper atoms at the interstitial site. However, in this study, the structure of Cu$_4$ is found to be most stable with the Cu$_{4(i)}V_{Si}$ structure. Cu$_{4(i)}V_{Si}$ defect are composed of vacancy and four copper atoms in the interstitial positions surrounding it. In the reaction between Cu$_4$ and hydrogen, the final product is found to be Cu$_{(s)}$H$_3$. In addition, by taking into account Cu$_{(H)}$H defects, we reproduced the intermediate products Cu$_{(s)}$H and Cu$_{(s)}$H$_2$ observed in the experiment [2].

From the above, we are able to clarify the structure of Cu$_4$ in silicon and the complex reaction between Cu$_4$ and hydrogen. This research provides an important contribution to the control of defects in semiconductor processes.

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First-principles calculation on defects in LiFeAs

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In recent years, iron-based superconductors have attracted attention as novel superconductors. Using scanning tunneling microscope, Grothe et al. found that various types of defects exist in the cleaved LiFeAs, and that the differential conductance differs significantly among defect types [1]. The effect of defects on the electronic states remains to be investigated for understanding the mechanism of superconductivity. Therefore, in the present study, we aim at elucidating the stable defect types and their magnetic properties using first-principles calculations based on the density functional theory.

Our results reveal the followings. First, there are two types of antiferromagnetic phases (hereafter referred to as the stripe AFM phase and weak AFM phase). They have different values of magnetic moments of the Fe atoms each other.

Defect species (phase)	Defect formation Energy (eV)
Fe (weak AFM)	-0.71
Fe (stripe AFM)	-0.56
Li (weak AFM)	0.02
Li (stripe AFM)	0.01
As (weak AFM)	2.33
As (stripe AFM)	1.65

Table 1: Calculated defect formation energies.

Second, as seen from the calculated defect formation energies (the minimum value in the allowable range of the chemical potential of the defect atomic species) in Table 1, the Fe atom defect model is the most stable both in the stripe and weak AFM phases.

Third, the stable magnetic state differs depending on the type of defect. The weak AFM structure is the more stable for the Fe defect, while the strip AFM structure is the more stable for the As defect. In the case of the Li defect, the difference in the defect formation energy between the two magnetic phases is slight.

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Interlayer Exciton in WS$_2$

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The few layer transition metal dichalcogenides (TMDCs) are two dimensional materials that have an intrinsic gap of the order of ≈ 2 eV. The reduced screening in two dimensions implies a rich excitonic physics and, as a consequence, many potential applications in the field of opto-electronics.

Continuously decreasing band gap with the application of an increasing electric field, and eventually a metallic behavior has been observed in bilayers of group VI TMDCs MoS$_2$, MoSe$_2$, MoTe$_2$ and WS$_2$ both experimentally [1] and theoretically [2-3]. We employed the GW+BSE method for studying the gate induced band gap tunability of bilayer MoS$_2$ for the first time and found excellent agreement with experiment. We reported an anomalous interlayer exciton in MoS$_2$ bilayer with a binding energy that is independent of the applied electric field unlike the intralayer exciton [4]. We speculate and expect this unique feature to be generic for 2D TMDCs. To confirm this, we have explored BL WS$_2$ under the influence of electric field using GW+BSE method.

For zero field, there are two excitonic peaks, A and B and when the electric field is applied it splits these two main peaks into A-2, A-1 and B-2, B-1. A closer look at the BSE results in low energy region shows the emergence of new excitonic peaks II-2 and II-1. We find that the position of peaks A-2 and A-1 stays almost the same as a function of the applied field whereas position of peaks II-2 and II-1 changes strongly upon gating. We found that the trends in binding energy are the opposite of the peak position trends: the binding energy of exciton II is weakly dependent on the field, while that of A is strongly dependent on the field as observed for MoS$_2$ BL. Exciton II, therefore is the anomalous exciton we had found in MoS$_2$ BL and proposed for WS$_2$ BL also. In WS$_2$ BL the critical field strength is ~ 4 V/nm and below this value distinct II and A excitons cannot be resolved.

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First-principles study of van der Waals magnetic materials: CrOX (X = Cl, Br) bulk to monolayer

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To estimate the magnetic ground state of CrOX (X = Cl, Br) monolayers, we performed detailed first-principles study for both bulks and the monolayers. We performed charge density functional theory plus U calculations for CrOX. While the previous calculations were based on spin density functionals [1, 2], our result and analysis show that the use of spin-unpolarized charge-only density is more reasonable in terms of the magnetic phase diagram, electronic structure, and magnetic property of both bulks and monolayers. Unlike several previous studies [3, 4], our results show that all CrOX monolayers are antiferromagnetic, which is same as the bulk. Furthermore, we performed magnetic force linear response calculation [5] to understand orbital-resolved magnetic exchange couplings J matrix. The Cr t_{2g}-t_{2g} component mainly contributes to the antiferromagnetic state for both the bulks and the monolayers.

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Logarithmic Violation of Mott Relation for Anomalous Nernst Effect

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The anomalous Nernst effect (ANE), which is a thermoelectric effect induced by the anomalous Hall effect has attracted attention as a probe to observe magnetic Weyl fermions due to the experimental observation of $-T\log T$ behavior which deviate from the Mott relation.[1] Two-band model analysis predicted that $-T\log T$ behavior originates from peculiar electronic states of type-II Weyl semimetal; however in real materials, when considering the multi-band case, it is unclear whether this $-T\log T$ behavior is reproduced or not.

In this study, we investigate the origin of logarithmic violation of Mott relation for ANE. We have performed first-principles fully-relativistic density functional calculations[2] and computed anomalous Hall and Nernst coefficients[3] for typical Weyl semimetals; Co_2MnGa[1] and $Co_3Sn_2Sn_2$[4]. We clarified that $-T\log T$ behavior in ANE originates from nodal lines as well as Weyl node. Moreover, we found that $-T\log T$ behavior can be detected a singularity of density of states composed by nodal line. In the presentation, we will present a detailed discussion for the origin of logarithmic violation of Mott relation based on Berry curvatures associated with peculiar topological electronic structures.

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First-principles study of ferromagnetic metal Fe$_5$GeTe$_2$

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We predicted electronic and magnetic properties of Fe$_5$GeTe$_2$ using density functional theory calculation. Band structures and density of states for both the monolayer and bulk are similar and demonstrate Fe d orbital mainly contribute to their metallic and ferromagnetic properties. Band splitting occurs for spin-up and spin-down states, presenting exchange splitting in Stoner's theory of ferromagnetism. The monolayer Fe$_5$GeTe$_2$ reveal itself as another Stoner-type 2D magnet, satisfying Stoner's criterion. Furthermore, we observed that the ferromagnetic ordering is robust and even significantly enhanced by applying biaxial strain. Our findings suggest Fe$_5$GeTe$_2$ could be a promising 2D magnetic material in the application of flexible spin devices..

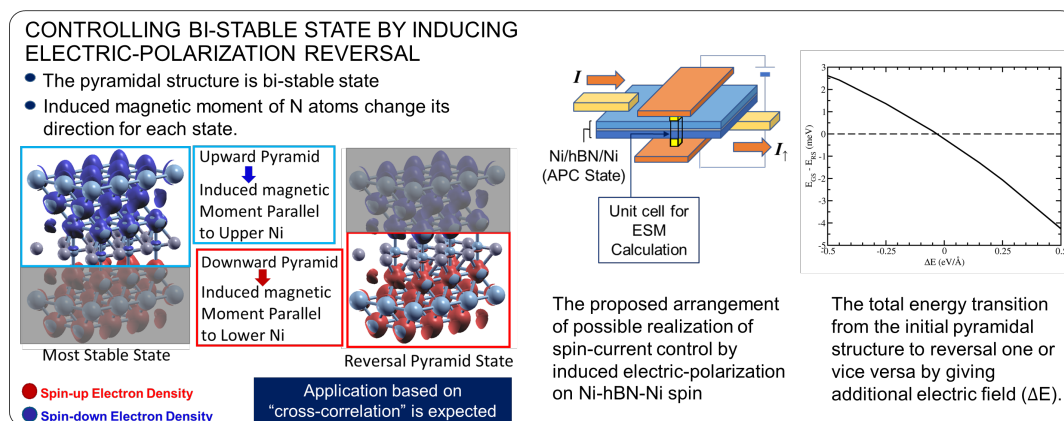[1] M. Joe, U. Yang, and C. Lee, Nano Materials Science, in press, (2019).

Optimizing the Spin-Current Control by Induced Electric-Polarization Reversal in hBN-based Magnetic Tunnel Junction

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Our recent study on hexagonal boron nitride (hBN) sandwiched by Ni(111) slabs reveals that the most stable stacking arrangement among 36 possible stacking arrangements shows the cross-correlation functionality coming from the bi-stable state of a rugged hBN plane and the controllable BN polarization [1]. The bi-stable state of the rugged hBN plane appears when the magnetic alignment of the upper and lower Ni(111) slabs is anti-parallel. The rugged hBN plane forms a unique pyramidal structure having two folding state, upward and downward pyramidal structure, which is interchangeable by controlling the BN polarization. The charge transfer from Ni(111) slabs to N atoms of hBN, which comes from pd-hybridization between Ni atoms with N atoms, leads to induced magnetic moment on N atoms. The induced magnetic moment direction of N atoms depends on the pyramidal structure of rugged BN. The transmission probability calculations show a spin-filtering effect where the spin-polarized current is controlled by the electric field when a field-induced reversal of the polarization is realized. The efficiency of spin-filtering effect depends on induced magnetic moment of N atoms. A consideration of other ferromagnetic slabs leads to different mechanism of charge transfer resulting different characteristics of induced magnetic moment on hBN layer.

CONTROLLING BI-STABLE STATE BY INDUCING ELECTRIC-POLARIZATION REVERSAL
- The pyramidal structure is bi-stable state
- Induced magnetic moment of N atoms change its direction for each state.

Upward Pyramid / Induced magnetic Moment Parallel to Upper Ni

Downward Pyramid / Induced magnetic Moment Parallel to Lower Ni

Most Stable State — Reversal Pyramid State

Spin-up Electron Density
Spin-down Electron Density

Application based on "cross-correlation" is expected

Ni/hBN/Ni (APC State)

Unit cell for ESM Calculation

The proposed arrangement of possible realization of spin-current control by induced electric-polarization on Ni-hBN-Ni spin

The total energy transition from the initial pyramidal structure to reversal one or vice versa by giving additional electric field (ΔE).

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Role of the molecular configuration on the electronic structure of organic crystals: A combined DFT and *GW* study

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Organic molecular materials have attracted considerable attention as candidates for next-generation flexible electronics materials. However, there still remains open questions on electronic properties such as mechanisms of the carrier transport.

The intermolecular van der Waals (vdW) forces are responsible for the cohesion in organic molecular crystals. To discuss the structure and stability of the organic crystal precisely, it is crucially important to describe the intermolecular and intramolecular interactions at the same accuracy with reliable first-principles theoretical methods.

In this study, we show, by using oligoacene crystals as examples, that the recent vdW inclusive method[1] is able to predict accurate geometries and energetics of the organic crystals. Furthermore, we demonstrate that by using the theoretical crystal structure, it is also possible to calculate the accurate electronic structure based on the methodology such as the many-body perturbation theory within the *GW* approximation[2].

The calculated lattice constants of the most stable polymorph were found to be in good agreement with the diffraction measurements at temperature far lower than room temperature, with the maximum relative deviation being within 1.0 %.

To demonstrate the impact of the crystal geometry upon the electronic structure, the theoretical density of states for three crystal geometries of tetracene were investigated, i.e., one with its lattice constants measured at room temperature or higher (Structure I), second one with the lattice constants measured at 175 K (Structure II), and third one whose crystal geometry was theoretically optimized (Structure III). It is found that as the cell volume decreases from Structure I to III, the band widths increase due to the increasing overlap between the neighboring molecular orbitals (MO's). The fundamental gap of Structure III becomes smaller by as large as 0.2 eV than that of Structure I. The gap narrowing originates from both the increasing overlap between the neighboring MO's and the screening effect upon the injected charge enhanced by the denser molecular packing.

Based on the Wannier function localized at each of the molecular sites, the impact of the molecular configuration upon the intermolecular transfer integrals could be discussed. The detail of the transfer integrals will be presented.

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Spin-wave dispersion in magnetic compounds: $RFe_{11}Ti$ (R=Y, Nd, Sm) and $Nd_2Fe_{14}B$

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Spin-wave dispersion offers information of collective magnetic excitations that is essential in the analysis of magnetic properties at finite temperatures. In the context of the development of hard magnets, exchange stiffness is important for understanding the macroscopic behavior, and one can derive it from the curvature of the lowest branch around the Γ point.

We have recently developed an efficient method to calculate spin-wave dispersion[1] by using Liechtenstein's formula[2] that constructs a Heisenberg model from a self-consistent field obtained by first-principles calculation. Our method can obtain the spin-wave dispersion for a system with multi-sublattices within a practical time consumption.[1] In the previous paper, we reported our results for $Sm(Fe, Co)_{12}$, which is a promising magnetic compound that has a potential of surpassing the current champion magnet $Nd_2Fe_{14}B$.[1] However, we did not perform a calculation for a spin-wave dispersion of $Nd_2Fe_{14}B$ itself, partly because it needs more computational resources than $Sm(Fe, Co)_{12}$ due to its largeness of the primitive unit cell.

In this paper, we report our results of calculated spin-wave dispersions with our refined program, which is now capable of calculation for $Nd_2Fe_{14}B$. We show that the spin-wave dispersion of $Nd_2Fe_{14}B$ has an almost isotropic curvature in the lowest branch around the Γ point, which agrees with the experimental observation by Mayer et al.[3,4] This isotropy is in contrast to the anisotropy found in $Sm(Fe, Co)_{12}$ in our previous paper.[1]

We also show the existence of anisotropy in the lowest branch of the calculated spin-wave dispersion for $RFe_{11}Ti$ (R=Y, Nd, Sm), which have the same crystallographic structure to $Sm(Fe, Co)_{12}$. The $RFe_{11}Ti$ compounds are known to be much stabler as a bulk material than $Sm(Fe, Co)_{12}$. Therefore, we consider it is more feasible to verify the anisotropy experimentally.

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Vibrational properties of small ring structure molecules

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We present the electronic and vibrational properties of small ring structured molecules, based on first-principles calculations. Among the isoelectronic structures, benzene shows more stability than borazine (~0.36 eV) and carborazine (~0.46 eV). The bond length (X-H, X=B, C, N) in ring structures is shorter than their aliphatic counterpart, revealing higher in stretching frequency. Exploring other small ring structure molecules, we reveal X-H stretching frequency increases as one moves down the periodic table. For skeletal ring structure, we also deduce a relationship between their intrinsic radial breathing part and stretching part.

A finite electric-field approach to evaluate the vertex correction for the screened Coulomb interaction in the quasiparticle self-consistent GW method

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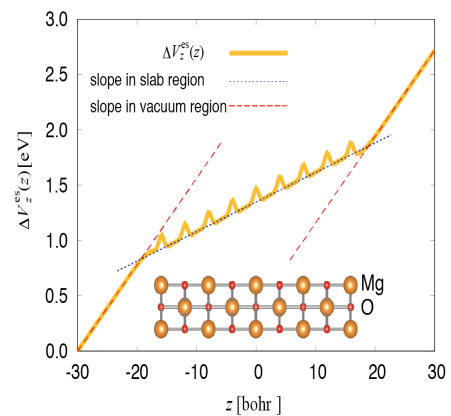
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Quasi-particle Self-consistent GW (QSGW) is a reliable first-principles-method which can take the effect of electron excitation into account. However, QSGW has several problems. For example, it systematically overestimates the size of band gaps. QSGW80, which is a hybrid method combining QSGW and LDA/GGA in the ratio of 80:20, overcomes such over-estimations [1]. The improvement of QSGW80 can be interpreted that the hybrid methods work as an addition of vertex corrections.

In this study [2], we have investigated vertex corrections. By a self-consistent calculation applying electric fields on slab models, we have calculated optical dielectric constants for LiF, KF, NaCl, MgO, and CaO. Here the constants are determined via a ratio of slopes of voltages in vacuum region and medium region as shown in the right Figure. In this method, higher-order terms are taken into account in the self-consistent manner. These terms can be interpreted to work as vertex corrections. In QSGW, we have applied this approach to the compounds, resulting in good

agreements with experimental measurements. Comparing the results of the slab calculations with the conventional random phase approximations for bulk systems, we can find such vertex corrections give significant differences. This means that QSGW with vertex corrections can explain electronic structure of such ionic materials very well.

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Piezoresistivity of InAsP Nanowires : Role of Crystal Phases and Phosphorus Atoms in Strain-Induced Channel Conductances

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Strong piezoresistivity of InAsP nanowires is rationalized with atomistic simulations coupled to Density Functional Theory. With a focal interest in the case of the As(75%)-P(25%) alloy, the role of crystal phases and phosphorus atoms in strain-driven carrier conductance is discussed with a direct comparison to nanowires of a single crystal phase and a binary (InAs) alloy. Our analysis of electronic structures presents solid evidences that the strong electron conductance and its sensitivity to external tensile stress are due to the phosphorous atoms in a Wurtzite phase, and the effect of a Zincblende phase is not remarkable. With several solid connections to recent experimental studies, this work can serve as a sound framework for understanding of the unique piezoresistive characteristics of InAsP nanowires.

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Origin of sub-band gap optical excitation in low-temperature GaAs-based THz emitter by DFT+SO method

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Terahertz (THz) wave, which lies between the infrared and microwave regions of the electromagnetic spectrum, corresponds to photon energies within the energy scale of excitation of many materials, making it attractive for low-energy spectroscopy, biomedical imaging, and security applications. However, this newly explored electromagnetic region has very limited low-cost, broadband and powerful THz sources. So far, 4.5 THz has been demonstrated in photoconductive antenna (PCA) with low-temperature grown GaAs (LT-GaAs) pumped at 1560nm [1]. Experimentally, this below band-gap excitation is considered stable and is proposed to be due to the presence of mid-gap states [1, 2]. In this theoretical work, we confirmed the presence of mid-gap state in GaAs with point defect (As-antisite) at ~1.00% concentration through bandstructure calculations using DFT+SO. SO is included because of the broken-inversion symmetry of GaAs. Also, we calculated the absorption spectra within the independent particle approximation and compared with experiment. At wide frequency window, spectral shape and intensity of the calculated and experimental absorption spectra are consistent. Using the above results, we derive the two-step photon absorption mechanism for the sub-band gap optical excitation in LT-GaAs-based THz emitter. For design purposes, we also derived the structure of the point defect that gives rise to the mid-gap state [3].

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Diagnosing and observing topological degeneracies in AI class systems

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Topological states have been widely studied in Fermionic systems for many years [1-6], wherever, they are sporadically explored in Bosonic systems [7-8], even both Fermions and Bosons are fundamental particles in condensed matter physics. Actually, Bosons can offer a better platform to get spinless band structures where nodal lines can be realized with only parity-time reversal (PT) symmetry, such as phonons. By combining first-principles calculations and meV-resolution inelastic x-ray scattering, we demonstrate the first realization of PT symmetry protected helical nodal lines of phonons in a real material [9]. The combination of theoretical analysis and experimental measurements demonstrate the PT-protected helical nodal lines for the first time in phononic materials. This nodal line is robust because of the spinless nature of phonons, which will open a new route to explore exotic topological states in crystalline materials.

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Six-nearest-neighbor parameterized Slater-Koster tight-binding model for bismuth

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We report a six-neighbor Slater-Koster tight-binding model[1], with spin-orbit coupling included, to treat the electronic and topological properties of Bi crystals. We contrast two distinct parameter sets showing seemingly same electronic energy bands with different Z2 index of bulk Bi. The parameter set which shows trivial Z2 index (v=[0;0,0,0]) for Bulk Bi and nontrivial Z2 index (v=1) for few layer Bi reproduces semi metallic properties of bulk Bi and its topologically trivial surface states, while semiconducting nature of few layer Bi films and their topologically non-trivial edge states. This highlights that the topological index or parameter needs to be treated carefully for proper parameterization of first-principles energy bands of topological materials.

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