

# Design of spintronic semiconductors via first-principles statistical-mechanics

Alex Zunger

*National Renewable Energy Laboratory, USA*

*E-mail: alex\_zunger@nrel.gov*

This talk describes a novel way by which one can scan a huge number of structural configurations of magnetic ions (e.g, Mn) in a host semiconductor (e.g, GaAs), identifying the structure that has the highest  $T_c$ . This is done in 4 steps: (1) Select a chemical configuration. This can include any superlattice (MnAs)<sub>n</sub>/(GaAs)<sub>m</sub>, or any Mn-clustering, or any random arrangement. The calculation is done within a supercell. (2) Compute the magnetic spin-spin interactions for this chemical configuration via Linear Response Theory (LRT). This involves calculating the band structure from first principles, as well as doing Greens-function Linear Response. (3) Using the spin-spin interactions for this chemical configuration, compute the critical temperature using Heisenberg Monte-Carlo method. (4) Now that we have  $T_c$  for 20-40 different chemical configurations of Mn in GaAs, we need to search which one has the highest  $T_c$ . But we can not limit our search to these configurations, for the maximum  $T_c$  configuration can be different. Yet, we can not compute directly the  $T_c$  for all  $2^N$  chemical configurations created by positioning either Mn or Ga on each of the N lattice sites in the supercell (N~ 64). Thus, we use the 20-40 computed  $T_c$  to construct a "Cluster Expansion" which gives  $T_c$  for any lattice configuration. This expansion is tested by its ability to predict  $T_c$  values for structures outside the "input set" of 20-40 structures, then comparing the prediction to actual LDA-LRT values. Using this Cluster Expansion we search for the configuration that has the highest  $T_c$  by scanning  $2^N$  possibilities. This method allow you (a) to find high  $T_c$  structures in a given semiconductor/metal system, and (b) to understand the origin of this  $T_c$  by analysing the connection between the configuration ( Step 1 ), its Spin-Spin interaction ( Step 2 ), and  $T_c$  ( step 3 ). This shed light on the mechanism of Ferromagnetism.

Early model -theories have suggested that Mn in the III-V semiconductors such as GaAs and GaN will introduce a host-like-hole state ( acceptor ) that will interact via RKKY-type coupling with the Mn local moment to produce the observed ferromagnetism (FM). This widely publicized model theory predicted that FM will be enhanced with the reduction of spin-orbit splitting and the interatomic distances, thus prompting a search for host materials with such properties. I will show how modern first-principles electronic structure theory can be used to establish if the basic properties of Mn and other 3d impurities in III-V semiconductors are indeed similar to what was widely assumed in the phenomenological model theories. Surprisingly, such calculations (via LDA, GGA, and LDA+U) reveal that the hole induced by Mn is not host-like, and that the ensuing FM is not RKKY-like, but has a characteristic dependence on the lattice-orientation of the Mn-Mn interactions in the crystal which is unexpected by RKKY. I will describe the chemical trends as the host is altered (GaN/GaP/GaAs/GaSb and Chalcopyrite semiconductors such as CuInSe<sub>2</sub> ) ; as the impurity is altered along the 3d series, and as the Fermi level is altered ( via doping ). The effects of clustering of impurities, as well as the site-preference ( substitutional, interstitial ) will be revealed. These calculations suggest a rather general picture of bonding and ferromagnetism in 3d-doped semiconductors. They open the way to "Inverse Design", by searching the atomic configuration of Mn in the lattice having the highest FM  $T_c$ .

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