Impurity Clustering and Ferromagnetic Interactions that are not Carrier Induced in Dilute Magnetic Semiconductors: The Case of Cu_2O:Co

Hannes Raebiger, Stephan Lany, and Alex Zunger

N . . . R . . E . L . , G . . , C . 80401, USA

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Current models for ferromagnetism in diluted magnetic semiconductors, such as ''*p-d* exchange'' or ''double-exchange'', rely on the presence of partially filled gap states. We point out a new mechanism, not requiring partially filled states, in which ferromagnetic coupling arises from the occupation of previously unoccupied levels when two transition metal impurities form a close pair. We find from first-principles calculations that this mechanism explains strong ferromagnetic coupling between Co impurities in $_2$, and at the same time gives rise to Co clustering.

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Nonmagnetic semiconductors (GaAs, ZnTe) can be made ferromagnetic by doping via 1% of substitutional magnetic ions (e.g., V, Cr, and Mn), leading to the interesting situation where magnetism is controlled via the density of free carriers (electrons or holes) in the sample [1–3]. Despite early attempts at RKKY-descriptions involving the picture of a delocalized hole [4,5] or a polaron [6,7] leading to magnetic coupling, the picture that now emerges is simpler [8–11]: The substitutional impurity [denoted TM in Fig. 1(a)

tor augmented-wave method as implemented in the VASP code [21]. Plane waves are included up to the cut-off energy of 400 eV. Doped and pure $_2$ are modeled in supercells consisting of 96 atoms using lattice constant for pure $_2$ of 4.313 Å calculated within GGA-PBE, and allowing all atomic positions to relax. For total energies a k-point mesh of 4 4 4 including the -point is used,

2 Bohr magnetons per Co atom, but as none of the levels are partially filled, $_2$ $'$ is not expected to be ferromagnetic within the conventional carrier-mediated mechanism of Fig. 1(a).

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