

# Impurity Clustering and Ferromagnetic Interactions that are not Carrier Induced in Dilute Magnetic Semiconductors: The Case of $\text{Cu}_2\text{O}:\text{Co}$

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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  $\text{Cu}_2\text{O}$ , 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  $\text{Fe}_2$  are modeled in supercells consisting of 96 atoms using lattice constant for pure  $\text{Fe}_2$  of 4.313 Å calculated within GGA-PBE, and allowing all atomic positions to relax. For total energies a  $k$ -point mesh of  $4 \times 4 \times 4$  including the  $\Gamma$ -point is used,

2 Bohr magnetons per Co atom, but as none of the levels are partially filled,  $\chi_{23}$  is not expected to be ferromagnetic within the conventional carrier-mediated mechanism of Fig. 1(a).

S  $a$   $a$  Co-Co

