Cu-based spin-1/2 spin-gap tetramer and trimer system: new generation materials for quantum computation

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Search for newer spin-1/2 low-dimensional magnetic system with a singlet ground state explores the prospects of its utilization as a newer generation device material suitable for quantum computation. Such geometrically frustrated complex systems materialize ideal two-level system with a wide range of excited state properties. We investigate the magnetostructural details of magnetic exchange properties for two such series: one tetramer Libethenite series and another trimer series with the help of first principles density functional appropriate multistep approaches enabling the construction of an effective minimal model Heisenberg spin-Hamiltonian for the series.

Exchange interactions in spin-tetramer mineral Libethenite series A_2PO_4OH [A = Cu, Co] appear to be more exciting than described by a simple square spin tetramer model in the earlier literature.[1] Evolution of localized band-structure for the series of system 1) Co_2PO_4OH, 2) $CoCuPO_4OH$ (Cu replacing Co_2), 3) $CuCoPO_4OH$ (Cu replacing Co_1) and 4) Cu_2PO_4OH reveals that, gradual bandwidth reduction and modification of ligand-field splitting due to substitution-induced change in bond-length and bond-angle along the series, occurs in such a way that it evolves from a three dimensional long-range ordered antiferromagnet (CO_2PO_4OH) to a low-dimensional geometrically frustrated spin-1/2 d^9 system (Cu_2PO_4OH) with a singlet ground state. Experimental investigation predicts a spin-gap ~ 141 K in Cu_2PO_4OH and long-range antiferromagntic order in Co_2PO_4OH below 70 K. [1,2]

Doping induced spin-manipulation with magnetic (Ni) and non-magnetic (Mg) dopants constitutes the experimental attempts to obtain a singlet ground state system from the linear chain Heisenberg antiferromagnetic Cu-based (d^9) spin-1/2 trimer compound $Ca_3Cu_2X(PO_4)_4$ [X = Ni, Mg] with doublet ground state. The current study is a first-principles based investigation of the effects of such doping on the spin-exchange mechanism and electronic structure of the parent compound. Site-selective doping with zero-spin dopants like Mg is proved to be more efficient than an integral spin dopant Ni in obtaining a spin-gap system with singlet ground state, as also observed in the experimental studies. Doping induced dimerized state is found to be the lowest in ground-state energy. Calculated spin exchange couplings along various possible pathways are observed to attain good agreement with earlier experimental results with suitable optimization of Coulomb repulsion (U) and exchange (J) parameters. [3]

First principles Density functional multistep approaches determine a generalized minimal model of spin-exchange interactions for these systems. The structural parameters after ionic relaxation are used for obtaining the N-th order Muffin-Tin Orbital (NMTO) downfolded band-structure and to identify the significant hopping paths. LSDA+ U Planewave calculations with PAW potentials are performed to calculate the exchange integrals along these paths and to construct a minimal Heisenberg model, the solution of which under first-order perturbation theory predicts the magnitude of the spin-gap close to the experimental value. Effects like hydrostatic pressure and size-confinement are also thoroughly investigated.

References:

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[2] Debjani Karmakar and J. V. Yakhmi, J. Phys.: Condens. Matter **24**, 436003 (2012); Solid State Phenomena **194**, 284 (2013).

[3] Debjani Karmakar and Alexander Yaresko (submitted).