Systematic analysis of ab initio low-energy effective Hamiltonians for $Pd(dmit)_2$ molecular conductors

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Quantum spin liquids, which do not show any symmetry breaking even at zero temperature, have attracted much interest since their elementary excitations in the quantum spin liquids are expected to show exotic elementary excitations such as Majorana particles. Because the exotic elementary excitations may be useful for next-generation devices such as quantum computers, a huge amount of works on searching and identifying the quantum spin liquids in solids has been done in a decade.

Among several candidates of the quantum spin liquids, the molecular solids β'-$X[Pd(dmit)_2]_2$ (X represents a cation) offers an ideal platform for realizing the quantum spin liquid induced by the geometrical frustration in the magnetic interactions because the geometrical frustration can be systematically controlled by changing cations X. In experiments, it has been proposed that the Neel temperatures are systematically controlled by changing cations X and the quantum spin liquid realizes in X =$EtMe_3Sb$ [1, 2].

In this project, to identify the microscopic origin of the quantum spin liquid found in X =$EtMe_3Sb$, we have performed systematic and comprehensive *ab initio* derivations of low-energy effective Hamiltonians for available 9 compounds of β'-$X[Pd(dmit)_2]_2$ (X = Me_4Y, $EtMe_3Y$, Et_2Me_2Y and , Y = As, Sb, and P) [3]. In the derivation the low-energy effective Hamiltonians, we first obtain the global band structure for dmit-salts using Quantum ESPRESSO [4]. Then, using the open-source software package RESPACK [5, 6], we evaluate the transfer integrals and two-body interactions such as the Coulomb interactions. As a result, we have found that the anisotropy of the transfer integrals and correlations effects systematically change by changing cations.

Moreover, we have analyzed the low-energy effective Hamiltonians using the exact diagonalization method [7, 8]. From the numerical exact analyzes of the low-energy effective Hamiltonians, we have shown the significant reduction of the antiferromagnetic ordered moment occurs around X =$EtMe_3Sb$. This reduction is consistent with the experimentally observed quantum spin liquid behavior in X=$EtMe_3Sb$. We have also shown that the reduction is induced by both the geometrical frustration in the hopping integrals and off-site Coulomb interactions. This result indicates that accurate evaluation of the microscopic parameters in the Hamiltonians is essential for reproducing the quantum spin liquid behavior in the dmit-salts.

In addition to the *ab initio* study for the dmit-salts, we have also analyzed the long-range spin transport in the topological Dirac semimetals [9] using the real-time evolution of the quantum systems. Furthermore, using $\mathcal{H}\Phi$ [7, 8], we have analyzed the magnetization process of the antiferromagnetic Heisenberg model on the kagome lattice [10]. We have also developed an open-source library for the shifted Krylov subspace method (Kω) [11] and an *Ab initio* tool for derivation of effective

low-energy model for solids (RESPACK) [5, 6].

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