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Low-energy Excitations in Quantum Spin Liquid Derived from Molecular Mott Insulator

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A molecular Mott insulator β' -EtMe₃Sb[Pd(dmit)₂]₂ (dmit = 1,3-Dithiol-2-thione-4,5-dithiolate) is a quantum spin liquid (QSL) candidate. In the crystal with the space group $C2/c$, Pd(dmit)₂ anion radicals are strongly dimerized to form a dimer with spin 1/2. The dimers are arranged in an approximately isosceles-triangular lattice, which leads to a frustrated $S = 1/2$ Heisenberg spin system.

The system shows no magnetic order down to a very low temperature (~ 19 mK) that corresponds to $J/12,000$, where J (~ 250 K) is the nearest-neighbor spin interaction energy. The HOMO-LUMO mixing in the dimer unit induces fragmentation of $S=1/2$ electron spin with strong quantum fluctuation.

Low-energy excitations in the QSL state are open to debate even now. Heat capacity and magnetization indicate gapless fermion-like excitations, while ¹³C-NMR indicates an existence of a nodal gap. ESR and μ SR probed the spinons, revealing their gapless character and an unexpectedly large degree of in-plane anisotropy in the spin dynamics. This anisotropic spin dynamics indicates quasi-one-dimensional diffusive motion in the direction of the weakest magnetic coupling in the triangular lattice.

In 2010, it was reported that thermal conductivity is characterized by its large value and gapless behavior (a finite temperature-linear term). In 2019, however, two other research groups reported opposite data (much smaller value and a vanishingly small temperature-linear term) and the discrepancy in the thermal conductivity measurement data emerges as a serious problem concerning the ground state of QSL. An origin of the discrepancy will be discussed.

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