Quantum spin liquid state of Pd(dmit)$_2$ salts

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Quantum spin liquid proposed by P. W. Anderson in 1973 [1] exhibits the absence of magnetic or valence bond solid order among entangled quantum spins even at zero temperature. Although this third fundamental state for magnetism is a long-sought state of matter that has attracted much theoretical attention, the ground state and low-energy excitations of the S=1/2 antiferromagnetic triangular lattice are still far from full understanding and furthermore there are few candidates of real materials [2]. We found that an anion radical salt $\beta'$-EtMe$_3$Sb[Pd(dmit)$_2]$$_2$ (dmit = 1,3-dithiol-2-thione-4,5-dithiolate) with a quasi triangular lattice of [Pd(dmit)$_2]$$_2$' dimers is a promising candidate for the quantum spin liquid [3]. Measurements of various properties including magnetic susceptibility, $\mu$SR, magnetic torque, $^{13}$C-NMR, specific heat, electrical/thermal transport, vibrational spectra, dielectric constant, and low-temperature crystal structure, have been performed. First-principles DFT calculations followed by tight-binding fitting were also carried out. The spin liquid in the $\beta'$-type Pd(dmit)$_2$ salts exists as a "phase" and is situated between an antiferromagnetic phase and a charge ordering phase. Ground states are classified by the anisotropy of the triangular lattice that can be finely tuned by mixed cations with a minimum of disorder effect on the magnetic layer.