Molecular Quantum Spin Liquid and Related Electronic States

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Solid state properties are determined by behavior of electrons. Electron with properties of either particle or wave obeys the laws of quantum mechanics. However, it remains impossible to predict properties of solids completely. Assemblies of interacting electrons with charge and spin frequently exhibit unexpected behavior. The phrase “More is Different” by P. W. Anderson indicates such a situation. In 1973, Anderson proposed an idea of “quantum spin liquid” that exhibits liquid-like properties among the spins even at zero temperature. Although this is a long-sought state of matter that has attracted much theoretical attention, there are few candidates of the real materials. Recently, we have found a molecular system that can be a candidate of quantum spin liquid. I will demonstrate the quantum spin liquid state and related electronic states that are generated by strong correlation and frustration of π-electrons in molecular crystals.

Anion radical salts of Pd(dmit)$_2$ (dmit=1,3-dithiol-2-thione-4,5-dithiolate) with monovalent cations Et$\text{Me}_x$,$\text{Z}^+$ (Et = C$_2$H$_{5-}$, Me = CH$_3-$, Z= P, As, Sb; $x = 0–2$) belong to a two-dimensional strongly correlated electron system with a quasi-triangular lattice. At ambient pressure, they are Mott insulators where one spin ($S=1/2$) is localized on each dimer unit [Pd(dmit)$_2$]$^2$. Due to a quasi-triangular arrangement of the dimer units, the spin frustration operates in this system. The degree of frustration can be tuned by the choice of the counter cation. Among them, EtMe$_3$Sb [Pd(dmit)$_2$]$_2$ is a promising candidate of the quantum spin liquid, where there is no spin ordering/freezing down to 19 mK. Choice of the cation provides fine tuning of the interdimer interactions and leads to various magnetic states including an antiferromagnetic ordered state (Me$_4$Sb salt), a charge ordered state (Et$_2$Me$_2$Sb salt), and a valence bond solid state (EtMe$_3$P salt).

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