Novel phase transition in $Et_2Me_2Sb[Pd(dmit)_2]_2$ at 70 K: a possible mechanism based on strong dimerization of two-level molecules

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A series of anion radical salts of $[Pd(dmit)_2]$ with tetrahedral cations such as Me_4P^+ are paramagnetic Mott insulators under ambient pressure. They exhibit strong frustration effect due to the triangular lattice like arrangement of the spin-1/2 units, $[Pd(dmit)_2]_2^-$. $Et_2Me_2Sb[Pd(dmit)_2]_2$ also behaves as such a frustrated magnet above about 100 K.

Recently, we have found that sufficiently pure samples of the Et_2Me_2Sb salt undergo a novel first-order phase transition at about 70 K. The spin susceptibility exhibits a sharp drop to zero, accompanied by a steep rise of resistivity. In the low temperature phase, two types of $[Pd(dmit)_2]_2$ with different geometries are found by X-ray analysis (A. Nakao *et al.*, this conference). These observations indicate the charges on $[Pd(dmit)_2]_2$ units are separated as, $2[Pd(dmit)_2]_2^- \longrightarrow [Pd(dmit)_2]_2 + [Pd(dmit)_2]_2^{2-}$, in the low temperature phase.

We will discuss the possible mechanism of this phase transition, emphasizing the role of strong dimerization in $[Pd(dmit)_2]_2$. We will point out that the observed charge separation is favored by the two-level electronic structure of $[Pd(dmit)_2]_2$, which can stabilize the doubly occupied state of the dimer by two-bond resonance. This mechanism is not expected for the conventional charge-ordered systems such as the ET salts with quarter-filling, in which the intermolecular Coulomb repulsion is believed to be responsible for charge separation.