Spin-1/2 Heisenberg Antiferromagnets on Anisotropic Triangular Lattice, [Pd(dmit)$_2$] Salts – How Do They Release Frustration?

Masafumi Tamura, Reizo Kato
Condensed Molecular Materials Lab., RIKEN, JST-CREST, Wako, Saitama 351-0198, Japan;
e–mail: tam@riken.jp, reizo@riken.jp

The outstanding character of a metal complex [Pd(dmit)$_2$] is its strong dimerization in a solid state. In a series of the 1:2 anion radical salts of [Pd(dmit)$_2$] with a tetrahedral cation such as Me_4P^+, the unpaired electron occupies the antibinding level of HOMO's of each dimer, $[Pd(dmit)_2]_2^-$. They are Mott insulators under ambient pressure. The dimers are arranged so as to provide a triangular-lattice-like layer with spatial anisotropy. The paramagnetic state of these salts is characterized by strong frustration arising from the nearly triangular arrangement, as shown by the temperature dependence of susceptibility (χ) explained only by the spin-1/2 Heisenberg triangular antiferromagnet model in the high temperature range. As temperature is lowered, χ forms a round peak, indicating the growth of antiferromagnetic correlation. Except the Et_2Me_2Sb salt, it is followed by steap decrease until the Néel temperature T_N is approached. From this behavior, we can learn that the system crossovers from a frustrated paramagnet to a state with growing antiferromagnetic correlation. The large entropy and energy cost due to frustration are thus released by the spatial anisotropy.

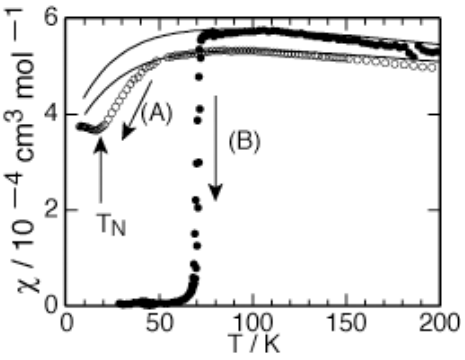

Fig. 1. Temperature dependence of χ showing the growth of antiferromagnetic correlation in $Et_2Me_2P[Pd(dmit)_2]_2$ (A) and charge separation in $Et_2Me_2P[Pd(dmit)_2]_2$ (B). Solid curves are the theoritical values for the triangular antiferromagnet.

In the case of the Et_2Me_2Sb salt, the frustration is released in another way. This salt undergoes charge separation, $2[Pd(dmit)_2]_2^- \longrightarrow [Pd(dmit)_2]_2^{2-} + [Pd(dmit)_2]_2^0$, at 70 K. This transition is driven by HOMO-LUMO interplay such that the electrons are paired on $[Pd(dmit)_2]_2^{2-}$ with the formation of HOMO-LUMO double-bond in the neutral dimer $[Pd(dmit)_2]_2^0$.