Anisotropic Triangular Lattice Antiferromagnets, [Pd(dmit)$_2$] Salts – Frustration and Dimensional Crossover in Spin-1/2 Heisenberg Systems

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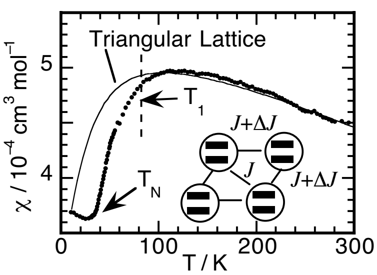

Fig. 1 Susceptibility of the Me_4As salt showing two crossovers is plotted versus T.

The [Pd(dmit)$_2$] salts with tetrahedral cations such as Me_4P^+ behave as Mott insulators under ambient pressure, in which each [Pd(dmit)$_2$]$_2$ dimeric unit carries an S = 1/2 spin. The temperature dependence of magnetic susceptibilities of the [Pd(dmit)$_2$] salts show a peak profile characteristic of a frustrated spin-1/2 Heisenberg antiferromagnet on a two-dimensional (2D) triangular lattice [1]. Unlike other triangular antiferromagnets, *e.g.*, κ-ET_2X, the [Pd(dmit)$_2$] salts appear to provide a transitional series deviating from the regular triangular lattice, with suppression of frustration due to the in-plain spatial anisotropy depending on the cation. For example, the Et_2Me_2Sb salt shows no magnetic order down to 4 K, while the susceptibility of the Me_4As salt varies as shown in Fig. 1, suggesting two crossovers near T_1~ 80 K and T_N ~ 40 K [1,2]. This suggests a significant role of the anisotropy (ΔJ in Fig. 1) to determine the low temperature magnetic properties. The crossover near T_1 is the release of the frustration to afford exponential growth of correlation with $1/T$ due to the presence of the square-lattice-like anisotropy, ΔJ. This explains the decrease of susceptibility below T_1, followed by the other crossover to a 3D system, which enables the system to undergo the long-range order at T_N, much higher than $J_\perp/k_B$, where $J_\perp$ is the interlayer coupling. From the analysis based on this picture, the anisotropy and the interlayer couplings can be estimated. We also comment on the role of the frustration in stabilizing the metallic state under pressure [1].

References

[1] M. Tamura and R. Kato: *J. Phys. Condens. Matter* **14** (2002) L729, and R. Kato *et al.*: this symposium.
[2] T. Nakamura *et al.*: *J. Mater. Chem.* **11** (2001) 2159, and S. Ohira *et al.*: this symposium.