PHASE TRANSITIONS AND CROSSOVERS IN QUANTUM TRIANGULAR ANTIFERROMAGNETS WITH SPATIAL ANISOTROPY, [Pd(dmit)$_2$] SALTS.

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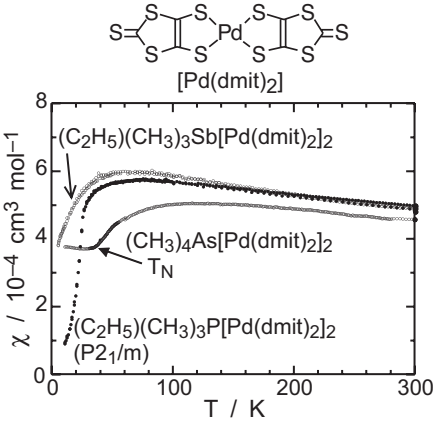
We report a trend observed in the temperature dependence of magnetic susceptibilities (χ) of a series of molecule-based triangular antiferromagnets, [Pd(dmit)$_2$] salts, $(C_2H_5)_x(CH_3)_{4-x}Z[Pd(dmit)_2]_2$ (x = 0, 1, 2; Z = P, As, Sb). In the salts, the spin-1/2 units $[Pd(dmit)_2]_2^-$ form spatially anisotropic triangular lattice layers.



The finite temperature behavior of χ of these salts is basically explained in terms of a geometrically frustrated spin-1/2 Heisenberg triangular antiferromagnet [1]. A crossover to unfrustrated states appears at low temperatures [2], which allows the spins to order antiferromagnetically. The Néel temperature, as well as the crossover temperature, increases from 18 K (the $(C_2H_5)_2(CH_3)_2P$ and $(CH_3)_3Sb$ salts) to 40 K (the $(CH_3)_4P$ and $(CH_3)_4As$ salts) with the increasing anisotropy (the deviation from the regular triangular lattice) as regulated by the cation size and geometry. If the anisotropy is sufficiently small, the frustrated paramagnetic state is retained down to low temperature, as observed in the $(C_2H_5)(CH_3)_3Sb$ salt.

A spin-Peierls-like phase transition to a non-magnetic state is found at 25 K in the $(C_2H_5)(CH_3)_3P$ salt ($P2_1/m$ phase), for the first time in a two-dimensional spin system close to the triangular lattice. Such a spin-dimer ground state has been expected to appear, when the long-range order is broken by the frustration. The spin-dimer state, evidently accompanied by the lattice symmetry lowering, is preferred in this salt, because it comprises parallel molecular stacking unlike the others.

[1] M. Tamura, R. Kato: J. Phys.: Condens. Matter **14** (2002) L729.
[2] M. Tamura, R. Kato: J. Phys. IV France **114** (2004) 383; Polyhedron, **24**, (2005) 2817; S. Ohira et al. Phys. Rev. B **70** (2004) 220404.