Antiferromagnetic Molecular Metal (Me-3,5-DIP)[Ni(dmit)2]2 (Me-3,5-DIP = N-methyl-3,5-diiodopyridinium; dmit = 1,3-dithiole-2-thione-4,5-dithiolate)

Yosuke Kosaka, a,b,c Hiroshi M. Yamamoto, b,c Akiko Nakao, d Masafumi Tamura, b,c Reizo Kato a,b,c,*

a Department of Chemistry, Faculty of Science, Saitama University, Saitama-shi, Saitama 338-8570, Japan
b RIKEN, Wako-shi, Saitama 351-0198, Japan
c JST-CREST, Wako-shi, Saitama 351-0198, Japan
d High-Energy Accelerator Research Organization, Tsukuba-shi, Ibaraki 305-0801, Japan
E-mail: reizo@riken.jp

We have succeeded in the preparation of a new antiferromagnetic molecular metal (Me-3,5-DIP)[Ni(dmit)2]2. The crystal consists of two crystallographically independent layers, which are alternatively arranged along the c-axis (Layers I and II). In Layer I, Ni(dmit)2 anions form strongly dimerized columns. In Layer II, on the other hand, the anion arrangement is similar to so-called the ‘spanning-overlap’ packing as seen in α-Et2Me2N[Ni(dmit)2]2,1 but slightly different in the anion orientation. These uncommon structural features are considered to be furnished by the short cation-anion interactions (I···S). Tight-binding band calculations suggest that this material has two contrastive characters: Mott-insulating state (Layer I) and two-dimensional metallic conduction in the ab plane (Layer II).

Temperature dependence of electrical resistivity depends strongly on current directions. For the a-axis directions, the resistivity decreases monotonically with lowering temperature. The b-axis resistivity is also metallic but it is accompanied by a broad maximum at around 72 K. In contrast, for the interlayer direction (//c), the resistivity at room temperature is about two orders of magnitude larger than those for the other directions and increases with lowering temperature down to 100 K. Magnetic susceptibility χ shows Currie-Weiss-like temperature dependence in a range of 20-300 K. The χ behavior would be explained by the superposition of the contribution from the localized spins in Layer I and the Pauli paramagnetism of conduction electrons in Layer II. In fact, the χ-T curve can be fitted by the Boner-Fisher model2 (The Hamiltonian is defined as $H = -2J \sum \langle \vec{S}_i \cdot \vec{S}_j \rangle ; J/k_B = -16.8$ K) with an additional constant term down to 10 K. At 10 K, χ has an anomaly and becomes anisotropic below this temperature. This is the evidence for the antiferromagnetic transition. In this material, both conduction and magnetism are based only on π electrons of Ni(dmit)2.

Reference