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)

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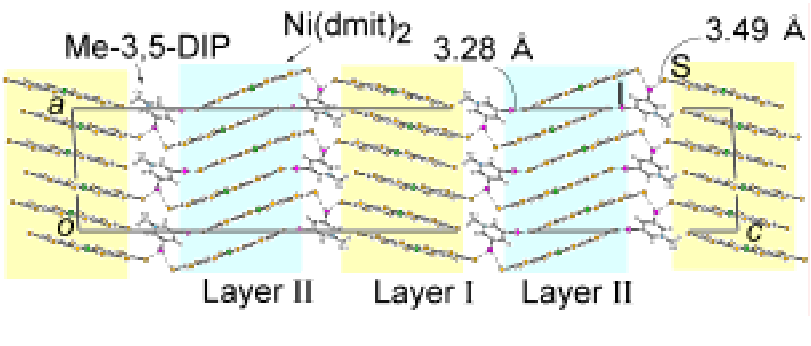
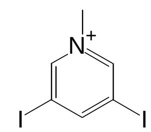
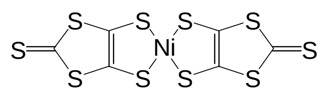
We have succeeded in the preparation of a new antiferromagnetic molecular metal (Me-3,5-DIP)[Ni(dmit)$_2$]$_2$. The crystal consists of two crystallographically independent layers, which are alternatively arranged along the *c*-axis (Layers I and II). In Layer I, Ni(dmit)$_2$ anions form strongly dimerized columns. In Layer II, on the other hand, the anion arrangement is similar to so-called the 'spanning-overlap' packing as seen in α-Et$_2$Me$_2$N[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 model[2] (The Hamiltonian is defined as $H = -2J\sum_i(\hat{\mathbf{S}}^i \cdot \hat{\mathbf{S}}^{i+1})$; $J/k_B = -16.8$ 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)$_2$.





(Me-3,5-DIP)$^+$



Ni(dmit)$_2$

Reference

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