A New Antiferromagnetic Molecular Conductor With Modified λ-type Structure, λ'-$(BETS)_2FeBr_4$

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It has been reported that the molecular conductors based on BETS (bis(ethylenedithio)-tetraselenafulvalene) and magnetic anions FeX_4^- (X = Cl, Br) gave novel systems showing attractive electromagnetic properties such as magnetic field induced superconductivity [1,2]. Here, we report the crystal structure, electrical resistivity, magnetic susceptibility of new BETS conductor, λ'-$(BETS)_2FeBr_4$.

λ'-$(BETS)_2FeBr_4$ is isostructural to λ'-$(BETS)_2GaBr_4$ [3] and has a modified λ-type structure. The planar BETS molecules are stacked along the *b* axis, and form two-dimensional conduction layers parallel to (010). The $FeBr_4^-$ anions are located in the spaces between the BETS layers. Although the intermolecular overlap integrals between HOMOs along the *b* direction are much larger than those along the *a* and *c* directions, the tight-binding band calculation gave two-dimensional Fermi surfaces.

Resistivities of λ'-$(BETS)_2FeBr_4$ were measured by conventional four probe method. The room-temperature conductivity was fairly high ($\sigma_{r.t.}$ = 30 S·cm^{-1}), but the resistivities were almost temperature independent at 300-50 K. Then the resistivities increased rapidly below 50 K, indicating a metal-insulator (MI) transition. The resistivity at 4 K was about 300 times larger than the room-temperature value.

The magnetic susceptibilities of λ'-$(BETS)_2FeBr_4$ were measured by SQUID magnetometer. A Curie-Weiss behavior was observed in the high temperature region (χ_{rt} = 1.40 ×10^{-2} emu mol^{-1}, C = 4.24 emu K mol^{-1}, θ = -2.8 K) indicating the existence of a localized S = 5/2 Fe^{3+} spin on each structure unit. However, a sudden decrease in susceptibilities was observed below *ca*. 3 K, suggesting an antiferromagnetic (AF) transition. In contrast to λ-$(BETS)_2FeCl_4$ [4], where π electron system undergoes a coupled AF and MI transition, the Fe^{3+} spins in λ'-$(BETS)_2FeBr_4$ are 3-D ordered below 3 K.

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