A New Antiferromagnetic Molecular Conductor With Modified λ-type Structure, λ'-(BETS)$_2$FeBr$_4$

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It has been reported that the molecular conductors based on BETS (bis(ethylenedithio)-tetrasedlenafulvalene) 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)$_2$FeBr$_4$.

λ'-(BETS)$_2$FeBr$_4$ is isostructural to λ'-(BETS)$_2$GaBr$_4$ [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)$_2$FeBr$_4$ 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)$_2$FeBr$_4$ were measured by SQUID magnetometer. A Curie-Weiss behavior was observed in the high temperature region ($\chi_0 = 1.40 \times 10^{-2}$ emu mol$^{-1}$, $C = 4.24$ emu K mol$^{-1}$, $\theta = -2.8$ 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)$_2$FeCl$_4$ [4], where $\pi$ electron system undergoes a coupled AF and MI transition, the Fe$^{3+}$ spins in λ'-(BETS)$_2$FeBr$_4$ are 3-D ordered below 3 K.