Fe-doping Effect on π-d interaction in λ-$BETS_2Fe_XGa_{1-X}Cl_4$ system

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A quasi-two-dimensional organic conductor λ$BETS_2FeCl_4$ exhibits a phase transition from a paramagnetic metal (PM) to an antiferromagnetic insulator (AFI) at 8.3 K. On the other hand, an isostructural nonmagnetic conductor λ$BETS_2GaCl_4$ shows a superconducting transition at 5.5 K [1]. Previously, the PM-AFI transition in λ$BETS_2FeCl_4$ was considered to be induced by the antiferromagnetic transition of the 3d spins via the π-d interaction. Recently, however, a measurement of specific heat for λ$BETS_2FeCl_4$ clarifies that the π and 3d spins do not cooperatively form the antiferromagnetic ordering as a ground state of λ$BETS_2FeCl_4$, but the 3d spins remain the paramagnetic states, while the π electrons are localized and forms the antiferromagnetic ordering [2]. To clarify the mechanism of the PM-AFI transition, we investigate Fe-doping effect on the π-d interaction in λ-$BETS_2Fe_xGa_{1-x}Cl_4$ system, where the ground states transfer from the superconducting states to the antiferromagnetic insulating ones at $x > 0.3$. We measured the specific heat of the salts with $x = 0.4, 0.6, 0.7, 1.0$ down to 0.2 K (Fig.1), and observed the six-level Schottky-typ hump derived from the paramagnetic 3d spins under the π-d interaction in the all salts. Therefore, we succeeded to quantitatively estimate the π-d interactions in the ground state in each salt from the Zeeman splitting of the paramagnetic 3d spins.

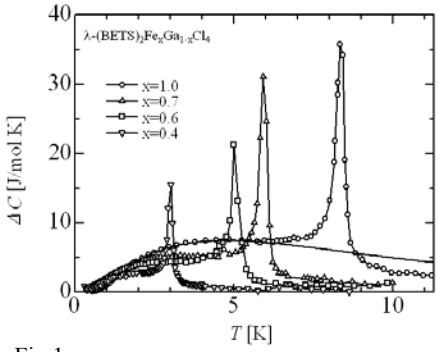

Fig.1
Excess specific heat ΔC of λ$BETS_2Fe_xGa_{1-x}Cl_4$ ($x = 0.4, 0.6, 0.7, 1.0$) obtained by subtracting the lattice and electric specific heats estimated for λ$BETS_2GaCl_4$. The Solid curve show the calculated specific heat based on the paramagnetic Fe 3d spins under the πd interaction.

[1] H. Kobayashi et al., J. Am. Chem. Soc. 118 (1996) 368.
[2] H. Akiba et al., J. Phys. Soc. Jpn. 78 (2009) 033601.