Fe-doping Effect on \( \pi \)-d interaction in \( \lambda \)-BETS\(_2\)Fe\(_x\)Ga\(_{1-x}\)Cl\(_4\) system

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A quasi-two-dimensional organic conductor \( \lambda \)BETS\(_2\)FeCl\(_4\) 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 \( \lambda \)BETS\(_2\)GaCl\(_4\) shows a superconducting transition at 5.5 K [1]. Previously, the PM-AFI transition in \( \lambda \)BETS\(_2\)FeCl\(_4\) was considered to be induced by the antiferromagnetic transition of the 3d spins via the \( \pi \)-d interaction. Recently, however, a measurement of specific heat for \( \lambda \)BETS\(_2\)FeCl\(_4\) clarifies that the \( \pi \) and 3d spins do not cooperatively form the antiferromagnetic ordering as a ground state of \( \lambda \)BETS\(_2\)FeCl\(_4\), but the 3d spins remain the paramagnetic states, while the \( \pi \) 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 \( \pi \)-d interaction in \( \lambda \)-BETS\(_2\)Fe\(_x\)Ga\(_{1-x}\)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 \( \pi \)-d interaction in the all salts. Therefore, we succeeded to quantitatively estimate the \( \pi \)-d interactions in the ground state in each salt from the Zeeman splitting of the paramagnetic 3d spins.