Temperature Dependence of Spin Susceptibility in α–(BEDT-TSF)$_2$I$_3$: Comparison with that in the Zero-gap State of α–(BEDT-TTF)$_2$I$_3$ under pressure

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A zero-gap state (ZGS) with Dirac cone-type energy dispersion for massless fermion has been found theoretically for an organic conductor, α-(BEDT-TTF)$_2$I$_3$ under pressure [1]. The band structure derived from structural data under uniaxial compression could explain systematically the successive change of ground states in this material. Accumulated numbers of experimental evidences have confirmed the existence of ZGS [2,3] and its unique features have been attracting wide interest of researchers. We found experimentally that the existence of charge ordering (CO) in this salt under pressure and suggested that the ZGS was accompanied by CO which has a different pattern from that in the insulating state at ambient pressure. We also observed that spin susceptibility of this salt under pressure tends to vanish linearly in temperature, which is characteristic to the ZGS.

The title material is an isostructural compound based on BEDT-TSF molecule and the electronic structure is believed to correspond to that of the BEDT-TTF salt under pressure. This salts behaves metallic at temperatures above 50 K. We have carried out ^{13}C-NMR at ambient pressure on a selectively ^{13}C-labeled single crystal sample and determined local susceptibility at each of different molecular sites. We found there exist the same pattern of CO as in the BEDT-TTF salt under pressure. Spin susceptibility was also found to decrease with decreasing temperature. However, the temperature dependence of spin susceptibility was not linear but almost exponential. The spin-lattice relaxation rate was also found to decrease exponentially. It may suggest a gap-opening at high temperatures instead of the ZGS stabilization. However, a real gap-opening is not likely since the resistivity is metallic in this temperature region. We discuss the difference in the temperature dependence of spin susceptibility between these two systems in connection with the electronic properties of ZGS.

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