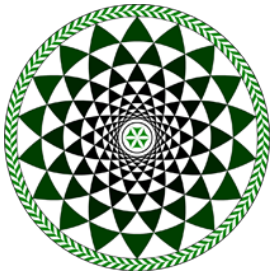

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Ferromagnetism in two-dimensional Mott insulating compounds with strong electron correlations

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The Mott insulating state of organic charge transfer complexes with layered structures usually has strong antiferromagnetic interactions in two dimensional characters. They induce various novel electronic phases such as unconventional superconductivity, spin liquids, and anomalous conductivities in π-electrons. It is well understood that not only spins but also charge and lattice degrees of freedom are coupled to each other and therefore organic π-electrons systems give interesting crossover or quantum mechanical proximity at low temperature region. As a further variation of molecule-based Mott-insulating state, we focused on the multi-layers dimer-Mott system, in which the counter ions have dipole-moments and aligned in one direction to form net-polarization to the organic donor/acceptor layers. Depending on the direction and magnitude of the net polarization, different molecular packing and physical properties appears in alternative layers. We report systematic experimental results of low-temperature heat capacity measurements as well as the magnetization, and transport measurements for the multi-layer dimer-Mott compounds of X[Ni(dmit)$_2$]$_2$ (X = ethyl-4-bromothiazolium (Et-4BrT), ethyl-2-iodo-5-bromopyridinium (Et-2I-5BrP), and ethyl-2-iodo-5-bromopyridinium (Et-2,5-DBrP)), where dmit is 1,3-dithiole-2-thione-4,5-dithiolate. The order of the magnitude of net polarization was evaluated as Et-4BrT>Et-2I-5BrP>Et-2,5-DBrP in them.

We succeeded to detected distinct peak structures in the temperature dependences of heat capacity which are in association with long-range ordering of π-electron spins around 1 K. The large transition entropy of 20˜40% of Rln2 in these compounds is suggesting that long-range ordering occurs in bulk feature. From the heat capacity measurements under magnetic fields, we observed an upward shift of the peak structure in (Et-4BrT)[Ni(dmit)$_2$]$_2$, which indicates that the ferromagnetic behavior occurs as a bulk feature. (Et-2I-5BrP) [Ni(dmit)$_2$]$_2$ and (Et-2,5-DBrP) [Ni(dmit)$_2$]$_2$ showed small magnetic field dependence which indicates paramagnetic like and anti-ferromagnetic behavior, respectively. The variation of the magnetic states can be understood as a kind of doping between layers. Furthermore, we detected that the application of pressure up to 1.0 GPa for (Et-4BrT) [Ni(dmit)$_2$]$_2$, the ground state changes to metallic state where the electronic heat capacity coefficient γ is nearly 2-3 times rather than usual metallic compound. The ferromagnetic fluctuations in the metallic states are expected in these multi-layer compounds. Systematic variation of the magnetic and conductive states will be discussed.

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