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Low temperature magnetic properties of bi-layer Mott insulators

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In this work, we measured heat capacities of bi-layer Mott insulators X[Ni(dmit)₂]₂ (X = Et-4BrT, Et-2I-5BrP, Et-2,5-DBrP) under magnetic fields in order to clarify the low temperature magnetic properties. X[Ni(dmit)₂]₂ forms low dimensional electronic structure based on the segregated stacking of Ni(dmit)₂ molecules and their counter cations. In these compounds, Ni(dmit)₂ molecules form dimerized structure. Since X is monovalent cation, each dimer has one electron. When the band width W is not so large compared to on site Coulomb repulsion U, this system behaves as a Mott insulator which is known to antiferromagnetic system with S = 1/2. In Mott insulating compounds (Et-4BrT)[Ni(dmit)₂]₂, (Et-2I-5BrP)[Ni(dmit)₂]₂ and (Et-2,5-DBrP) [Ni(dmit)₂]₂, two types of Mott insulating layers are realized due to asymmetric structure and stacking of cations. Due to the existence of alternative layer, these compounds are called as bi-layer Mott insulators. In these compounds, any long range ordering was not detected down to 2 K and the competition of ferromagnetic and anti-ferromagnetic behavior was observed in magnetic susceptibility. We performed heat capacity measurements and detected a peak structure around 1 K with large transition entropy (20~40% of Rln2) in all compounds. We consider that the existence of peak structure and large transition entropy is a common feature of bi-layer Mott insulators. We also observed sensitive magnetic field dependence which indicates bulk ferromagnetic behavior in (Et-4BrT)[Ni(dmit)₂]₂. In contrast to (Et-4BrT)[Ni(dmit)₂]₂, (Et-2I-5BrP)[Ni(dmit)₂]₂ and (Et-2,5-DBrP)[N(dmit)₂]₂ showed small magnetic field dependence which indicates paramagnetic like and anti-ferromagnetic behavior, respectively. In terms of the asymmetry of cation structure, the magnetic behavior changes from anti-ferromagnetic to ferromagnetic behavior with an increase in the asymmetry of cation structure. We speculate that the magnetic state and the effect based on asymmetric cations are strongly coupled in bi-layer Mott insulators.

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