LOW TEMPERATURE HEAT CAPACITIES OF BI-LAYER MOTT INSULATOR X[Ni(dmit)$_2$]$_2$ (X=Et-4BrT, Et-2I-5BrP)

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A part of organic charge transfer compounds forms low dimensional electron systems based on segregated stacking of organic molecules and their counter ion. A series of anion radical salts X[Ni(dmit)$_2$]$_2$ is known to such kind of low dimensional system. 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)$_2$]$_2$[1] and (Et-2I-5BrP)[Ni(dmit)$_2$]$_2$[2], two different types of Ni(dmit)$_2$ layer are realized due to asymmetric structure and stacking of cations(Fig.1). Therefore, two types 2D magnetic system are realized. Due to the existence of alternative Ni(dmit)$_2$ layer, these compounds are called as “bi-layer”. In these compounds, any long range ordering is not observed down to 2K. The low temperature magnetic properties of these antiferromagnetic spin systems were not clarified. In this work, we have performed heat capacity measurements down to 0.7 K of two bi-layer compounds in order to clarify low temperature magnetic properties.

The heat capacity data of (Et-4BrT)[Ni(dmit)$_2$]$_2$ is displayed in Fig. 2 by $C_p T^{-1}$ vs $T$ plot. We have observed a sharp peak structure around 1 K, which is indicative of formation of long range ordering. Interestingly, the transition entropy is estimated as about 40% of $R \ln 2$ despite that the intra-layer interaction was estimated as 10-10$^2$ K. Moreover, as illustrated in the inset of Fig. 2, the sharp peak was affected by magnetic fields like ferromagnetic system. The similar transition behavior was also observed in (Et-2I-5BrP)[Ni(dmit)$_2$]$_2$. The transition temperature and transition entropy were estimated as about 0.5-0.6 K and about 50% of $R \ln 2$, respectively. However the field dependence of peak structure is not so sensitive. We suggest that the transition behavior with the sharp peak structure and large transition entropy is a common feature of bi-layer Mott systems. However, the detailed difference such as the field dependence cannot be explained by the difference of intra-layer interactions. The important role of inter-layer interactions is speculated.