UNIQUE MAGNETO-CONDUCTING PROPERTIES OF NOVEL BI-LAYER MOTT INSULATOR BASED ON Ni(DMIT)$_2$ ANION RADICALS

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We have developed Bi-layer molecular conductors based on (alkyl-dihalopyridinium)[Ni(dmit)$_2$]$_2$ (dmit = 1,3-dithiole-2-thione-4,5-dithiolate), in which halogen bonding (halogen···S association) between cations and anions leads to two different kinds of Ni(dmit)$_2$ anion layers in a crystal. Such a bi-layer system can exhibit novel physical properties which cannot be obtained in conventional mono-layer molecular conductors. Indeed, the bi-layer system (Me-3,5-DIP)[Ni(dmit)$_2$]$_2$ exhibits both metallic conduction and localized spin paramagnetism attributed to one kind of molecule [1]. On the other hand, two isostructural derivatives (Et-2,5-DBrP)[Ni(dmit)$_2$]$_2$ and (Et-2I-5BrP)[Ni(dmit)$_2$]$_2$ are constructed from two kinds of Mott insulating layers. Their low-temperature magnetic properties, however, are quite different from each other.

We now focus on alkyl-monohalothiazolium cation as a candidate to develop new bi-layer Ni(dmit)$_2$ anion radical salts. Since the monohalothiazolium cation is less bulky than the dihalopyridinium cation, denser packing of Ni(dmit)$_2$ anion radicals leading to better conducting property as well as stronger spin-spin exchange interaction is expected.

Detailed magnetic investigations for a new bi-layer Mott insulator (Et-4BrT)[Ni(dmit)$_2$]$_2$ disclosed coexistence of antiferromagnetic layer and ferromagnetic layer below 30 K. Furthermore, large negative magnetoresistance (-75 % at 7 T) was observed at 4 K under 1 GPa.