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Pulsed and Multi-frequency ESR Investigation for Low-dimensional Molecular-based Conductors

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We carried out pulsed and multi-frequency ESR investigation for low-dimensional molecular-based conductors to clarify the low-temperature electronic states and carrier dynamics. In this paper, we introduce our recent progress of investigations for TMTTF and BEDT-TTTF low-dimensional organic conductors and related materials.

1) The organic conductors, $(TMTTF)_2X$, are well-known quasi-one-dimensional conductors possessing various ground states, such as spin-Peierls, antiferromagnetic and superconductivity states realized by applied pressure or variation of counter anions, X. [1] However, the recent progress in the investigation of charge ordering (CO) phenomena has cast doubts on the validity of the simple Mott-Hubbard insulator scenario. [2] Hence, we performed pulsed ESR measurements to clarify the low-temperature electronic state. We present possible charge redistribution transitions observed in $(TMTTF)_2X$.

2) Multi-frequency ESR measurements using X-(9.5GHz), Q-(34GHz), and W-(95GHz)bands microwaves, magnetic measurement using SQUID and energy band calculation were carried out for ζ-(BEDT-TTF)$_2$PF$_6$(THF) and γ-(BEDT-TTF)$_2$PF$_6$. The temperature dependence of the spin susceptibility of the two salts shows that of typical paramagnetic insulators with low-dimensional antiferromagnetic interaction. The macroscopic magnetic behaviors are apparently similar to each other, and the absolute values of the intra-chain antiferromagnetic interaction, $J_{\mathrm{intra}}/k_{\mathrm{B}}$, of the two salts are also close to each other. However, their ground states and microscopic behaviors indicate obvious difference. ζ-(BEDT-TTF)$_2$PF$_6$(THF) undergoes an antiferromagnetic transition at around 5 K, while γ-(BEDT-TTF)$_2$PF$_6$ shows no magnetic long-range ordering down to 4 K. The ESR linewidth, ΔH_{pp}, of ζ-(BEDT-TTF)$_2$PF$_6$(THF) is almost temperature independent in the paramagnetic region. On the other hand, the ΔH_{pp} of γ-(BEDT-TTF)$_2$PF$_6$ gradually decreases as the temperature decreases. The low temperature electronic states of these salts are discussed in the aspect of the magnetic dimensionality.

[1] for example: T. Ishiguro, K. Yamaji and G. Saito, *Organic Superconductors, 2nd* (Springer-Verlag, Berlin/Heidelberg, 1998).
[2] T. Nakamura, K. Furukawa and T. Hara: to appear in J. Phys. Soc. Jpn. **76** (2007), and references therein.