IV-B Magnetic Resonance Studies for Molecular-Based Conductors

Molecular based conductors are one of the extensively studied materials. The development of the understanding of the electronic phases of these materials enables us systematic investigations of low-dimensional highly correlated electrons systems.

Magnetic resonance investigations are powerful microscopic investigations; they are advantageous for studying the fundamental electronic properties and for understanding the detailed electronic structures of molecular based compounds. Competition of the electronic phases in molecular based conductors has attracted much attention. Investigation of such electronic phases in molecular based conductors is important to understand the unsolved fundamental problems in the field of solid state physics.

In this project, we performed the ESR, and NMR measurements for molecular based conductors to clarify the low temperature electronic states.

IV-B-1 Low-Temperature Electronic Phases of EDT-TTF Based Molecular Conductors

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[*Mol. Cryst. Liq. Cryst.* **380**, 233–237 (2002)]

Magnetic investigation was carried out for low temperatures electronic phases in $(EDT\text{-}TTF)_2AuBr_2$, which undergoes an SDW transition at 16 K. In the SDW phase, we observed an abrupt change of ^{1}H-NMR absorption line around 6 K where the ^{1}H-NMR spin-lattice relaxation rate shows an anomalous second-peak. The electronic phase is discussed by microscopic point of view.

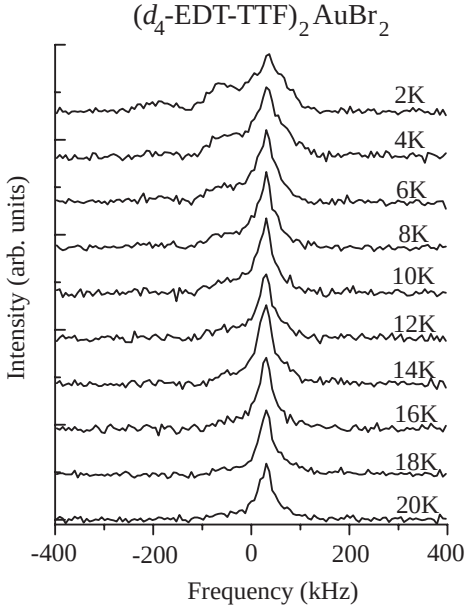

Figure 1. Temperature dependence of the ^{1}H-NMR absorption lines of the $(d_4\text{-}EDT\text{-}TTF)_2AuBr_2$ single crystal.

IV-B-2 Microscopic Investigation of a New Two-Component Organic Conductor with Itinerant and Localized Spins: $(CHTM\text{-}TTP)_2TCNQ$

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[*J. Phys. Soc. Jpn.* **71**, 2208–2215 (2002)]

Low-temperature electronic phases in a new two-component organic conductor, a segregated-stack charge-transfer salt called $(CHTM\text{-}TTP)_2TCNQ$, are investigated. The ESR g tensor analyses indicate that there exist itinerant CHTM-TTP spins and localized TCNQ spins at R.T. The temperature dependence of the physical parameters reveals that this salt undergoes two drastic, successive phase transitions at low temperatures. The effective moment of the localized TCNQ spins decreases at the 245 K transition and completely disappears at the transition around 195 K. These curious physical properties are explained by the drastic changes in the electronic states of the two different types of spins. The spin susceptibility was decomposed into the contribution of each of the two spin species by using ESR, ^{1}H-NMR, and static susceptibility analyses. We present a microscopic investigation of the two-spin system with itinerant and localized moments.

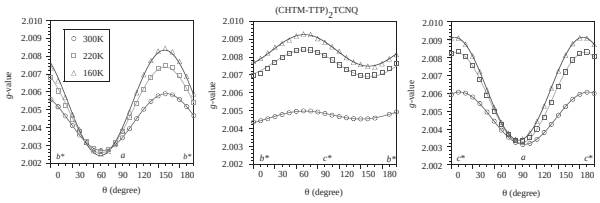

Figure 1. Angular dependence of the g values of $(CHTM\text{-}TTP)_2TCNQ$ at 300 K (circle), 220 K (square) and 150 K (triangle), applying the static field static field within the ab^*, b^*c^*, and c^*a planes, respectively. The solid curves are the least square fits to the measured data using the standard anisotropic g tensor equation.

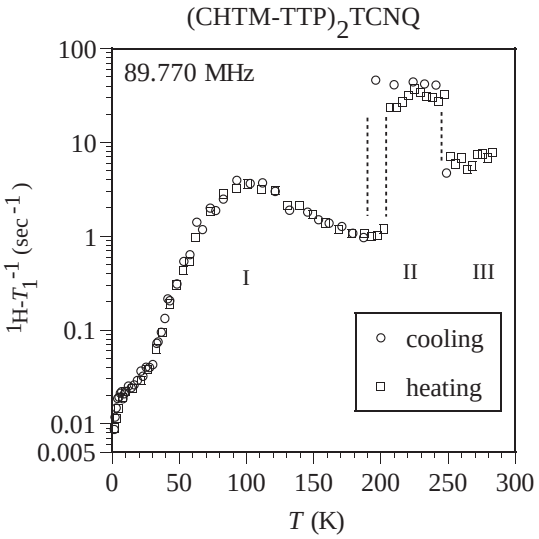

Figure 2. Temperature dependence of the ^{1}H-NMR spin-lattice relaxation rate, T_1^{-1}, of (CHTM-TTP)$_2$TCNQ, operated at 89.770 MHz, using a polycrystalline sample.

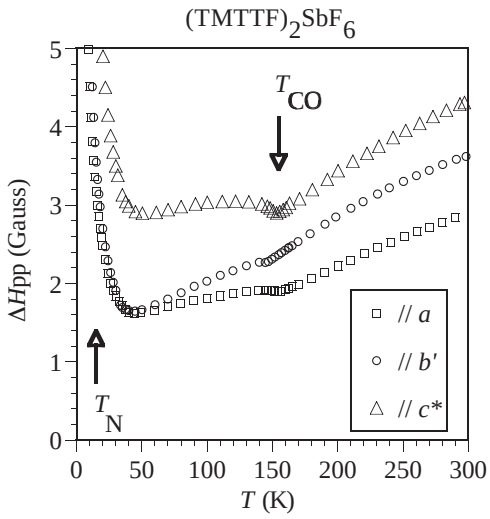

Figure 1. Temperature dependence of the peak-to-peak ESR linewidth, ΔHpp of (TMTTF)$_2$SbF$_6$ for a single crystal. The arrows indicate the charge-ordering and antiferromagnetic transitions.

IV-B-3 *g*-Tensor Analyses of β'-Type Pd(dmit)$_2$ Metal Complexes

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[*Mol. Cryst. Liq. Cryst.* **379**, 53–58 (2002)]

ESR measurements and *g*-tensor analyses were performed for metal complexes, β'-type Pd(dmit)$_2$. The ESR *g*-values of β'-type Pd(dmit)$_2$ are found to be beyond one radical description which is a good approximation for conventional molecular based conductors. In this paper we focus on the anomalous behavior of the ESR *g*-values in β'-type Pd(dmit)$_2$. We discuss the possible explanation of the electronic structure of β'-type Pd(dmit)$_2$ metal complexes from microscopic points of view.

IV-B-4 Possible Charge Ordering Patterns of the Paramagnetic Insulating States in (TMTTF)$_2$*X*

NAKAMURA, Toshikazu

[*J. Phys. Soc. Jpn.* **72**, 213–216 (2003)]

ESR investigations were performed for a series of quasi-one-dimensional organic conductors, (TMTTF)$_2$*X*. The ESR linewidth shows abrupt jumps or humps in the paramagnetic insulating region. The (TMTTF)$_2$*X* salts are roughly divided into three groups according to the anisotropy of the ESR linewidth at low temperatures. In this paper, we discuss the possible charge-ordered patterns of three typical salts (*X* = ReO_4, SbF_6 and Br) from the microscopic point of view.

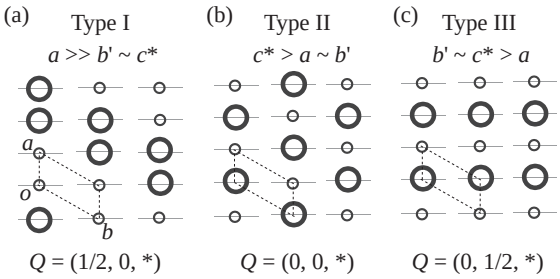

Figure 2. Schematic of possible charge-ordering patterns in the conducting *ab*' planes of TMTTF compounds: (a) Type I (ReO_4, ClO_4), (b) Type II (SbF_6, AsF_6) and (c) Type III (Br).

IV-B-5 NMR Investigation of (TMTTF)$_2$*X*: Charge Configurations and Spin Dynamics

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[*Synth. Met.* **133-134**, 67–68 (2003)]

We performed ^{13}C NMR study for a quasi-one-dimensional organic conductor (TMTTF)$_2$Br. Although the resistivity has its minimum value around 100 K, the NMR spectra showed no significant change down to 30 K. Instead, we found slight broadening of the spectra above the magnetic phase transition temperature (T_N ~ 15 K). Similar anomaly is also observed for the uniform susceptibility (χ_s).

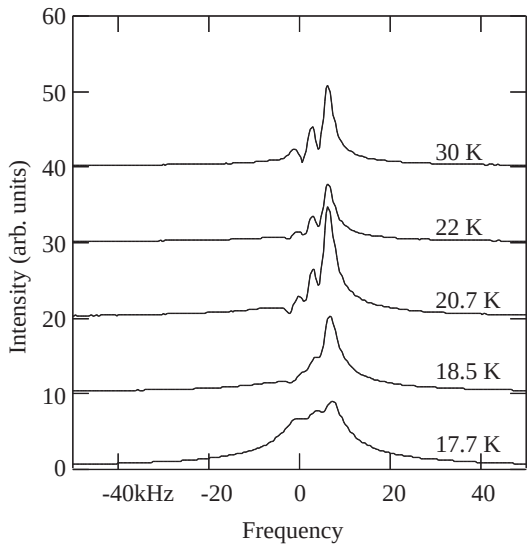

Figure 1. Temperature dependence of the ^{13}C NMR absorption line below 30 K.