IV-I Charge and Spin Dynamics of Organic Conductors

The spin and charge dynamics in organic conductors play important role in the emergence of the exotic properties in organic conductors, for example, superconductivity, magnetic ordering and charge ordering. It is important to reveal not only magnetic properties, but also the total picture of organic conductors. As known well, ^{13}C-NMR is a one of the most powerful tool in the point of the magnetism. Since nuclear magnetic moment, *I*, is 1/2, ^{13}C-NMR is not sensitive to the charge properties. On the other hand, optical studies, which are sensitive to the charge properties, are complementary to NMR study. In order to study both magnetic and charge properties, we performed ^{13}C-NMR and optical works.

IV-I-1 Electron Delocalization on κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ under Pressure

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κ-(BEDT-TTF)$_2$X is a system, whose bandwidth is comparable to the effective onsite coulomb repulsion. The phase diagram of this system has been considered to represent the competition between anti-ferromagnetic insulating behavior and superconductivity.

The κ-(BEDT-TTF)$_2$X system have been well explained using a parameter, *U*/*W* as well as the high T_c cuprates. Although κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ exhibited the superconductivity under pressure, this salt behaves as an insulator without magnetic ordering under ambient pressure. Similar to the case of other salts, AF fluctuations intensified with a decrease in temperature. In addition to that, line broadenings, which suggested inhomogeneous electron localization, was observed.

We have proposed that the electron localization effect is significant in a relatively narrow bandwidth of the salt and the scenario in which, pressure application increases the bandwidth, suppresses the localization and produces the superconductivity.

In order to determine the relationship between the insulator phase and the superconductivity and confirm the proposed scenario, we measured the ^{13}C-NMR of κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ under pressures. We observed a decrease in the spin susceptibility with an increase in pressure and the suppression of the line broadening above critical pressure.

The temperature dependences of $(T_1T)^{-1}$ under pressures exhibited the same behavior as that of κ-(BEDT-TTF)$_2$X, where X = Cu[N(CN)$_2$]Br and Cu(NCS)$_2$.

These results suggested that the electronic structure of κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ under pressures is same as that of other κ-(BEDT-TTF)$_2$X salts. Moreover, they supported our scenario of the emergence of the superconductivity.

IV-I-2 Coherent-Incoherent Crossover Behavior of Electron on κ-(BEDT-TTF)$_2$Ag(CN)$_2$·H$_2$O

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κ-(BEDT-TTF)$_2$X system showed the variety electronic states, for example, superconductivity (SC), anti-ferromagnetic ordering (AF), *etc.* Experimentally, the most impressive aspects are an adjustment between the superconducting and the magnetic ordering state and the peak of $(T_1T)^{-1}$ at T^* in NMR study as observed in high T_c cuprates. These result suggests the mechanism of superconductivity intermediated by magnetic fluctuations as same as high Tc cuprates. ^{13}C-NMR is a one of the most powerful tool in the point of magnetic dynamics. On the other hand, optical studies, which are sensitive to the charge dynamics, are complementary to NMR study which detects the magnetism. We performed single crystal ^{13}C-NMR and optical works. Many researchers paid the attention to mainly T_c = 10 K class samples near the AF-SC boundary. However κ-(BEDT-TTF)$_2$Ag(CN)$_2$·H$_2$O, which is far from the boundary, should be paid the attention. Is there the peak structure in $(T_1T)^{-1}$ in this sample and the relationship between T^* and SC?

We measured ^{13}C-NMR and reflection spectrum in κ-(BEDT-TTF)$_2$Ag(CN)$_2$·H$_2$O. We could not observed the increase of the AF magnetic fluctuation as in κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl. Results of ^{13}C-NMR suggested the emergence of superconductivity did not require the increase of the AF magnetic fluctuation but the crossover to the Fermi liquid regime. Optical studies suggested the crossover corresponds to the development of the coherency of conduction electron.

IV-I-3 Charge Ordering State on (BEDT-TTF)$_3$Cl$_2$·2H$_2$O

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The organic superconductors, as represented by κ-(BEDT-TTF)$_2$X, are attractive compound, because of the relationship between anti-ferromagnetic fluctuation and superconductivity well as High-T_c cuprates. Since these κ-type salts are well known to have the nature of the strong dimerization, they can be considered as a half-filled electron system. Many experiments on κ-

$(BEDT\text{-}TTF)_2X$ have been well explained by the universal phase diagram using a parameter, U/W, where U is the effective on-site coulomb repulsive energy and W is the bandwidth.

On the other hand, if the dimerization is weak and the U is small, the system cannot be regarded as half-filled system. In addition to the U, the effective off-site Coulomb repulsive energy V makes the system form Charge Ordering (CO) state.

In some of organic conductors, such as α-type, θ-type salts, their insulator phase was said not to be antiferromagnetic ordering state. Since the dimerization in these salts is not so strong, the insulator phase of these salts is likely to form the CO state. Applying pressure, some of these salts show the superconductivity. Therefore we are also interested in the relationship between charge fluctuation and superconductivity. Quasi-two-dimensional (Q2D) organic conductor $(BEDT\text{-}TTF)_3Cl_2\cdot 2H_2O$ is metallic at 300 K and it undergoes metal-insulator transition (MIT) at $T \sim 150$ K from magnetic susceptibility and electric conductivity measurements. The MIT has been believed to be connected with charge density wave (CDW) formation. However, the formation of the charge ordering (CO) in the insulator phase was also expected. Using ^{13}C-NMR measurement, we observed the split of the NMR spectrum which corresponded to the charge rich and poor sites below the MIT temperature and could conclude the insulator state is the CO state.