IV-E 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, charge ordering. For these purposes, it is important to reveal not only magnetic properties, 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-E-1 Charge Ordering State on Organic Conductors

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Quasi-two-dimensional (Q2D) organic conductor (BEDT-TTF)$_3$Cl$_2$·2H$_2$O 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 in the CO state.

From electric conductivity measurement under pressures for this compound, it is found that the metal–insulator transition is suppressed by applying pressure and this compound shows superconductivity above 1.6 GPa. In order to inspect how CO state formed under ambient pressure is changed, we measured T_1 under pressures. Regrettably, we could get no information from NMR spectrum because of the broadening spectrum due to magnetism of the pressure cell. Below 1.4 GPa, we observed $(T_1T)^{-1}$ = constant at high temperature and decreased at low temperature, just same as ambient pressure. This indicates that the formation of the gap Δ also exists under pressures. As we have done above, we estimated the gap Δ at each pressure by means of thermal activation model fitting. We could confirm the existence of the gap Δ until 1.4 GPa. Both the gap Δ and T_g decreased with increasing pressure until 1.4 GPa. By applying pressure, transfer integral *t* in conductivity chain increases and enhance itinerant-electron system. As the results, the amplitude of the charge dispropotionation becomes small and the gap Δ is decreasing. In addition, as compared to metal–insulator transition temperature T_{MI} estimated from electric conductivity measurement, T_g behaves in a way similar to T_{MI}, just as ambient pressure. We predict that a strong correlation between two temperatures; T_g and T_{MI}, exists.With decreasing temperature, $(T_1T)^{-1}$ shows almost constant like a metallic behavior. This indicates that the system changes an itinerant-electron system and the localization of the carrier by CO vanishes. It is important for the elucidation of the mechanism of superconductivity to consider whether the system is a simple metallic state or an exotic metallic state in which itinerant-electron system and charge disproportionation coexist.

IV-E-2 ^{13}C-NMR Study of Single Crystal of β'-(BEDT-TTF)(TCNQ) under Pressure

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Organic conductor β'-(BEDT-TTF)(TCNQ) is the material in which BEDT-TTF molecules form two-dimensional structure and TCNQ molecules form one-dimensional stacked structure, and both molecules are in the form of dimer. Properties of this salt shows metal insulator transition (M–I transition) at 330 K at ambient pressure, and transition temperatures decrease by applying pressure. It is known that BEDT-TTF molecule and TCNQ molecule show different magnetic properties so that an antiferromagnetic ordering occurs in the former at 20 K and the latter occurs at 3 K. In addition, properties of this salt under pressure are interested very much from the point of view that the layer structure of BEDT-TTF have the same type of structure of β'-(BEDT-TTF)$_2$ICl$_2$ which has the highest T_C under pressure among the organic conductors. So we performed ^{13}C-NMR measurements using single crystal of (BEDT-TTF)(TCNQ) where one site of the central double-bonded carbon in BEDT-TTF molecule is labeled by ^{13}C. BEDT-TTF forms a dimer in a layer. It is expected that two peaks are observed in the high temperature. Actually, two peaks were seen in the observed spectrum in the high temperature side. We can estimate a local susceptibility only for a BEDT-TTF site by using NMR at selected site. One peak was observed in the vicinity of almost knight shift 0 at antiferromagnetic transition temperature (20 K) and understood that line width increased as lowering temperature. We measured in a range of ±1 MHz at 10 K, but the other peaks were not observed. It suggests that antiferromagnetic transition of this material is not commensurate but incommensurate. We are also going to report about the result of under pressure.