博士論文（要約）

Transport and NMR studies of an organic crystal α-(BEDT-TTF)$_2$I$_3$ in the critical region of charge order-to-Dirac fermion transition

（電荷秩序-ディラックフェルミオン転移の臨界領域にある有機結晶 α-(BEDT-TTF)$_2$I$_3$ の電荷輸送および NMR 研究）

劉　東

In this thesis, we studied the critical region of charge order-to-Dirac fermion transition in an organic crystal α-(BEDT-TTF)$_2$I$_3$ (α-I$_3$) under pressure. Through electrical resistance and ^{13}C NMR measurements, the electronic states was investigated in both charge and spin degrees of freedom. The charge ordered (CO) state is stabilized by intersite short-range (SR) Coulomb interactions while, in the Dirac fermion (DF) state with conical dispersions, unscreened long-range (LR) Coulomb dominate the electronic properties. In α-I$_3$, the two distinct electron-electron (el-el) interactions, SR and LR, are both vital and provide an unprecedented research arena for the CO-to-DF critical region, which has been explored in the present thesis for the first time.

In the CO state realized at $T < T_{CO}$ and $P < P_C \approx 11$ kbar, where T_{CO} means the CO transition temperature and P_C does the CO-DF phase boundary, the CO transition accompanied by spin singlet formation is found to be gradually suppressed by pressure until discontinuously vanishing at P_C. The spin excitation gap (Δ_{NMR}) is pressure-insensitive and drops discontinuously to zero at P_C in contrast to the pressure dependence of T_{CO}, pointing to the spin excitations, very likely spin-triplet excitations, low-lying below quasi-particle excitations. However, Δ_{NMR} ($\approx$ 44 meV) is far smaller than the singlet-triplet excitation gap of around 130 meV estimated on the assumption that the spin-rich horizontal strip forms a one-dimensional spin-1/2 alternate Heisenberg chain. The discrepancy may be related to the CO nature of the system, where spins are not completely confined on the spin-rich stripes but also distributed on the charge-poor sites to some extent due to the incomplete charge disproportionation (CD). Assuming that the spin gap is dominated by $J \propto t^2/U$, an increase in t with pressure is expected to increase the spin gap; on the other hand, however, CD is gradually suppressed by pressure. These two effects are considered to compensate the pressure dependence of spin gap, causing the observed pressure-insensitivity. T_{CO} and Δ_{NMR} characterize the bulk CO state. However, the charge excitation derived by electrical resistivity decreases prominently under pressure and even vanishes in a pressure range below P_C, which is most reasonably explained in terms of the emergence of the edge states suggested theoretically.

The static spin susceptibility at high temperatures above the energy scale of the van Hove singularities (~ 130 K) is suppressed with pressure increase near ambient pressure but its pressure variation is moderate above P_C. Among the three site-specific spin susceptibilities (χ_s^j; j = A, B, or C), the decrease of χ_s^B under elevated pressure is the most prominent, while that of χ_s^C is the least. The contrasting behavior between χ_s^B and χ_s^C may indicate a progressive cone tilting by pressure until the stabilization of the DF state above P_C.

For the DF state at low temperatures, a critical regime in the vicinity of P_C was found where charge transport and dynamic spin susceptibility ($1/T_1T$, where T_1 is the nuclear spin-lattice relaxation time) show quasi-critical behaviors toward P_C, being associated with the development of SR interactions, as evidenced by the emergence of CO. On the other hand, the site-specific static spin susceptibilities show no remarkable pressure-dependence at low temperatures. This indicates that the LR part of the el-el interactions, which renormalizes the electron velocity upward and reshapes the cone, is pressure-insensitive. This suggests that the mechanism of the mass acquisition of the massless Dirac fermions at P_C is cause by the SR interactions. In addition, the ferrimagnetic fluctuations on site-B, evidenced by negative spin susceptibility, are found to persist in a wide pressure range from P_C up to 23 kbar at least.

Specifically, the T-square temperature dependence of $1/T_1T$ previously observed at P = 23 kbar changes to the T-cubic dependence at $P_C \approx 11$ kbar. A conceivable scenario is that the spin-singlet fluctuations is strengthened near DF-CO transition due to the development of the SR el-el interactions.

A low-temperature resistivity upturn appears robustly irrespective of pressure and is enhanced while approaching the transition pressure of CO. The contrast between the experimental observation and the prediction within the framework of the renormalization of unscreened LR Coulomb interactions show that the insulating behavior can originate from SR ones.

As seen above, the present thesis finds various manifestation of long-range and short-range Coulomb interactions in the electronic and magnetic properties of massless Dirac fermions and thus contributes to the foundation of the research field of interacting Dirac fermions.