論文の内容の要旨

論文題目：A Study of Electronic States in the Doped Triangular-Lattice Organic Conductor κ-$(ET)_4Hg_{2.89}Br_8$ under Pressure

（ドープされた三角格子有機伝導体 κ-$(ET)_4Hg_{2.89}Br_8$ の圧力下電子状態の研究）

氏名　大池広志

1. Introduction

This thesis is on electronic states in the doped triangular-lattice organic conductor κ-$(ET)_4Hg_{2.89}Br_8$ (κ-HgBr) under pressure. Our study has been motivated by the distinctive features of κ-HgBr in clarifying the nature of doped Mott insulator, which is one of the central issues in the physics of strongly correlated electron systems.

When U/W takes large value, electrons in half-filled band (n = 0.5) localize to form Mott insulating state, where U, W and n are on-site Coulomb repulsion, bandwidth and band filling, respectively. While most of Mott insulators undergo antiferromagnetic order in low temperature, in case of triangular lattice, the ground state is non-trivial because of spin frustration. Recently, Mott insulating states with no magnetic order, called spin liquid, have been found in organic Mott insulator with triangular lattice κ-$(ET)_2Cu_2(CN)_3$ [1]. What occurs when carrier is doped to spin liquid is an intriguing issue, because doped spin liquid state is the possible origin of high-T_c superconductivity as proposed by P. W. Anderson [2]. However, it has been difficult to make an experimental access to this issue because mother materials of doped Mott insulators are not spin liquid in most cases.

The title compound κ-HgBr (Fig .1) is an exceptional case of the possible doped Mott insulator whose mother material is spin liquid, where ET denotes bis(ethylenedithio)-tetrathiafulvalene [3]. κ-HgBr has conducting ET layers sandwiched by insulating anion layers composed of Hg and Br ions. The in-plane structure of the ET layer is characterized by a checker board-like

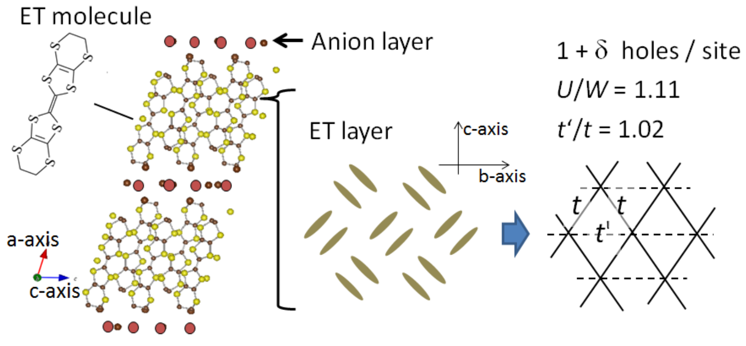

Fig. 1 Crystal structure of κ-$(ET)_4Hg_{3-\delta}Br_8$ (δ = 0.11).

arrangement of strongly dimerized ET molecules, which is called κ-type. A band-structure calculation shows that *U*/*W* of κ-HgBr is larger than that of Mott insulator κ-$(ET)_2Cu_2(CN)_3$, and *t'*/*t*, which is an indicator of anisotropy of triangular lattice as depicted in Fig.1, is close to unity (nearly isotropic triangular lattice), where *t* and *t'* are nearest-neighbor and next-nearest-neighbor transfer integrals, respectively. Assuming that valences of Hg and Br ions are +2 and -1, respectively, the valence of an ET dimer turns out to be +1.11due to the non-stoichiometry; namely, *n* of κ-HgBr is 0.445. Therefore, κ-HgBr is reasonably viewed as a 11-% hole-doped Mott insulator with triangular lattice.

Organic conductors are advantageous in that the highly compressible nature allows a wide range of variation in correlation strength. Actually, pressure experiments for organics with half-filled band (n = 0.5) demonstrated the first-order phase transition from a Mott insulator to a less correlated metal, and revealed its criticality [4]. In order to clarify the nature of doped Mott insulator with triangular lattice and seek for novel electronic state in doped region ($n \neq 0.5$), we performed transport measurements on κ-HgBr under pressure.

This thesis is divided into two parts. Part 1 is on *U*/*W* control under hydrostatic pressure. In this part, we suggested quantum phase transition from doped Mott insulator to a less correlated metal. Part 2 is on control of both *U*/*W* and *t'*/*t* with new pressurizing technique. We found a novel electronic state which is not observed under hydrostatic pressure.

2. Experimental

Part 1: *U*/*W* control in doped region

In order to probe bulk property of superconducting (SC) transition and normal state in-plane resistivity $\rho_{//}$, we performed contactless conductivity measurement under pressure, which utilizes the technique of AC susceptibility measurement in MHz-frequency range [5]. When SC regions appear in a sample, diamagnetic response depends on the volume fraction of SC regions, so we can discuss the bulk property of SC transition. In addition, this method can detect normal state properties because eddy current due to electromagnetic induction causes diamagnetic response and the characteristic length of flux penetration is the skin depth, δ (so called skin effect), which is the function of resistivity. We applied AC magnetic field perpendicular to the sample so that δ gives the value of $\rho_{//}$. The pressure medium was Daphne 7474 oil.

Part 2: *U*/*W* and *t*'/*t* control in doped region

In order to control both *U*/*W* and *t*'/*t*, we used Demnum oil s-20 as pressure medium, which is in a solid state above 1.1GPa. While hydrostatic pressure is applied below 1.1GPa, we can apply uniaxial pressure above 1.1GPa and vary *t*'/*t*. When we lower the solidification pressure by cooling, the hydrostatic component in pressure decreases. Then, the variation of *t*'/*t* starts at lower pressure. Thus, we can control both U/W and t'/t. With this pressurizing technique, we performed four-probe resistivity measurement.

In both parts, we use single crystals of κ-HgBr, which are obtained by the standard electrochemical method. The pressure cell is a hybrid system consisting of a NiCrAl inner shell and a BeCu outer one.

3. Results and Discussions

Part 1: *U/W* control in doped region

We observed SC demagnetization. SC transition temperature T_c shows dome-shaped pressure dependence although non-SC fractions exist in a sample in low-pressure side of SC dome. In the normal state, crossover temperature from Fermi liquid to non-Fermi liquid decreases by lowering pressure pointing to the T_c dome. The obtained pressure-temperature phase diagram (Fig. 2 left panel) suggests that quantum phase transition from doped Mott insulator to a less correlated metal occurs around 0.5GPa. We proposed that this quantum phase transition continuously connects to metal-insulator transition at n = 0.5 as described in the schematic band filling-U/W phase diagram (Fig. 2 right panel).

Part 2: *U/W* and *t'/t* control in doped region

We succeeded to control both U/W and t'/t with new pressurizing technique. We found peak structure in temperature dependence of $\rho_{//}$ in low-t'/t region. This peak structure grows up systematically by lowering t'/t at fixed U/W (Fig. 3). We considered that this peak structure came from phase separation of insulating and metallic phases in a sample. In half-filled band system, lowering t'/t changes metallic state into Mott insulating state because antiferromagnetic

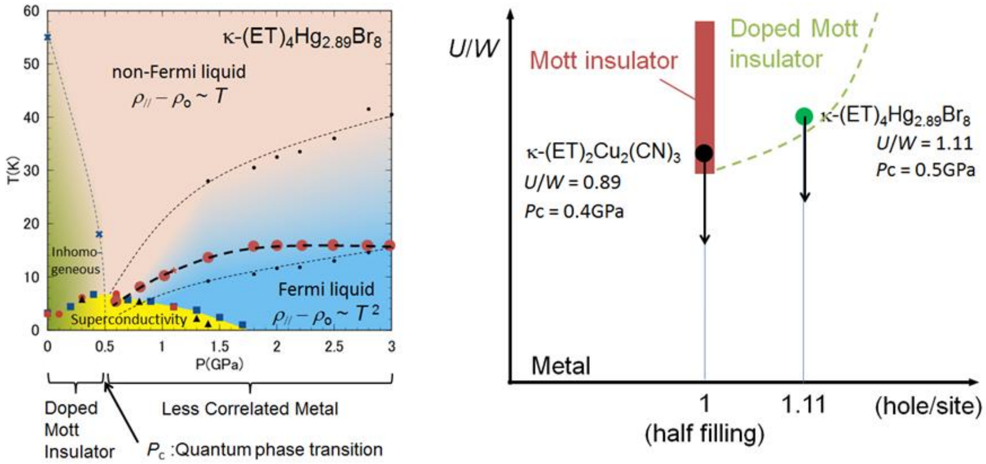

Fig. 2 Pressure-temperature phase diagram of κ-$(ET)_4Hg_{2.89}Br_8$ and schematic band filling-U/W phase diagram.

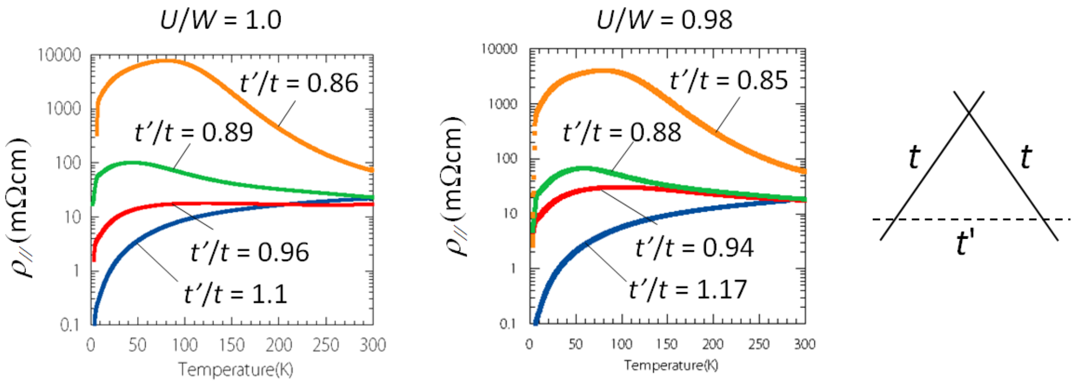

Fig. 3 Temperature dependences of $\rho_{//}$ under variation of t'/t at fixed U/W. The values of U/W and t'/t are obtained by extend Huckel and tight binding methods under some assumptions about crystal structure.

order assists localization of carriers. If this tendency to be insulating state applies to doped case, we can understand the insulating temperature dependence of $\rho_{//}$ in high temperature range as formation of Mott insulating region in a sample. Because whole sample cannot be in a Mott insulating phase when $n \neq 0.5$, metallic regions should also exist in a sample. We ascribed the metallic temperature dependence of $\rho_{//}$ in low temperature range to the existence of metallic regions. This drastic dependence of $\rho_{//}$ on t'/t can be a novel property of doped Mott insulator with triangular lattice.

4. Conclusion

This thesis presents a transport study on the doped triangular-lattice organic conductor κ-HgBr under pressure. In the experiments under hydrostatic pressure, we suggested that quantum phase transition from doped Mott insulator to a less correlated metal occurs in κ-HgBr. In the experiments under pressure with solid-state oil, we succeeded to control both U/W and t'/t and found a peak structure in temperature dependence of $\rho_{//}$ in low-t'/t region. These results indicate that this study clarified the nature of doped Mott insulator with triangular lattice and the novel electronic state in doped region.

5. Reference

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