Anisotropic Pressure and Career Doping Effect for Molecular Conductor $EtMe_3P[Pd(dmit)_2]_2$

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Molecular conductor $EtMe_3P[Pd(dmit)_2]_2$ (dmit = 1,3-dithiole-2-thione-4,5-dithiolate) consists of alternate stack of cation layer of $EtMe_3P^+$ and anion dimer layer of $[Pd(dmit)_2]_2^-$. A spin correlation between dimers in the anion dimer layer has a geometry close to a regular triangular lattice. In addition, the system has a half-filled band structure at ambient pressure, and electrons are localized on each dimer site one by one due to the strong on-site Coulomb repulsion, resulting in the Mott insulator. From previous study for this salt, it is known that, at ambient pressure, the transition to the valence-bond order (VBO) phase with lattice modulation occurs at 25 K and leads to the nonmagnetic state with a spin gap [1]. Furthermore, the VBO transition is suppressed by the weak hydrostatic pressure and superconducting phase appears above 0.4 GPa [2, 3].

In order to investigate the electronic state of the superconducting phase adjacent to the Mott insulating phase using device fabrication, we have mainly investigated κ-$(ET)_2X$ (ET = Bis(ethylenedithio)tetrathiafulvalene, X : monovalent anion). Using a thin crystal laminated on a flexible substrate, we have attempted control of the bandwidth by the bending of the substrate and doping of carriers by the field effect [4,5]. In the present research, we newly investigated $EtMe_3P[Pd(dmit)_2]_2$, and studied universal physics of the Mott insulating phase and superconducting phase, and compared with other molecular conductors having a triangular lattice. We aim to elucidate the relationship between the VBO phase, which appears only in $EtMe_3P[Pd(dmit)_2]_2$, and the magnetic frustration and the band structure.

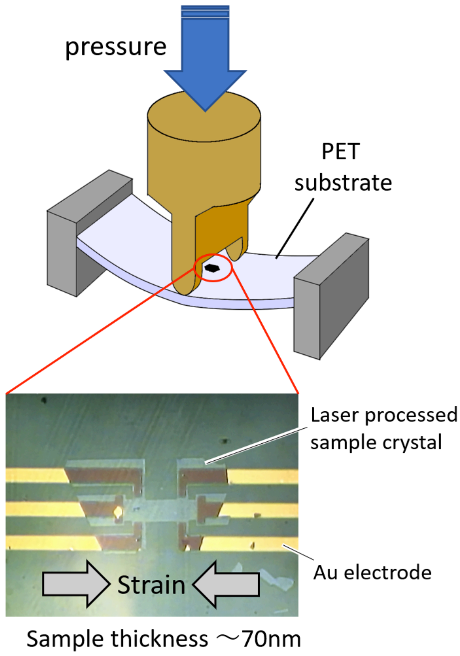

Fig. 1: Schematic figure of the setup for one-dimensional pressure measurement and the thin crystal of $EtMe_3P[Pd(dmit)_2]_2$ laminated on the PET substrate.

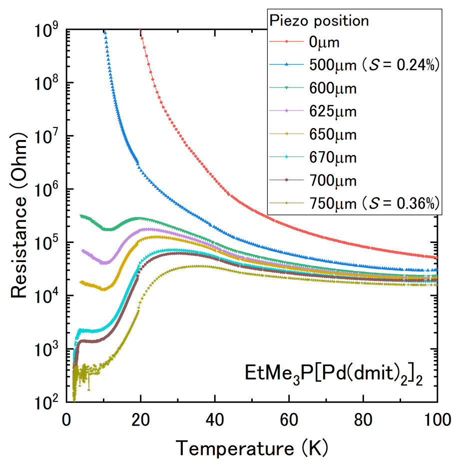

Fig.2: Temperature dependence of the electrical resistance of $EtMe_3P[Pd(dmit)_2]_2$ for various uniaxial pressure.

Figure 1 shows the setup used in this study to apply the uniaxial pressure. A single crystal of $EtMe_3P[Pd(dmit)_2]_2$ with a thickness of the several tens of nanometers is laminated on the soft PET (polyethylene terephthalate) substrate and the center of the substrate is pushed by the piezo actuator. The sample is bent according to the bending of the substrate and compressed in the direction of the long axis of the substrate. The PET substrate has larger coefficient of thermal expansion than that of $EtMe_3P[Pd(dmit)_2]_2$ and when it cooled, a weak two-dimensional positive pressure is applied to the sample.

Figure 2 shows temperature dependence of the electrical resistance for $EtMe_3P[Pd(dmit)_2]_2$ thin crystal with uniaxial pressure. The units "μm" and "*S*" in the legend represent the elongation of the piezo actuator and the magnitude of the distortion, relatively. When the substrate is not bent, the system remained insulating in the whole temperature range. When the uniaxial pressure was applied to the sample, the insulation behavior was suppressed, and in the high-pressure region, a superconducting phase appeared with a transition temperature of 4 K. This behavior is roughly same as that of the bulk sample under the hydrostatic pressure [2, 3]. In the intermediate pressure region, reentrant behavior was observed. In the bulk sample under the hydrostatic pressure, the VBO transition accompanied by the hysteresis is observed [3]. However, the sharp resistance jump or clear hysteresis was not observed at the present case. Magnetic frustration due to the triangular lattice structure is considered to play an important role for the VBO transition and it is possible that the differences in the transport behavior are caused by the difference between hydrostatic and uniaxial pressure.

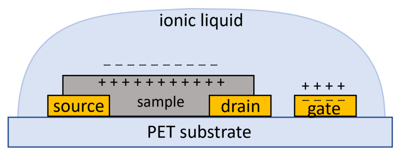

Fig. 3: Schematic picture of organic FET with ionic liquid.

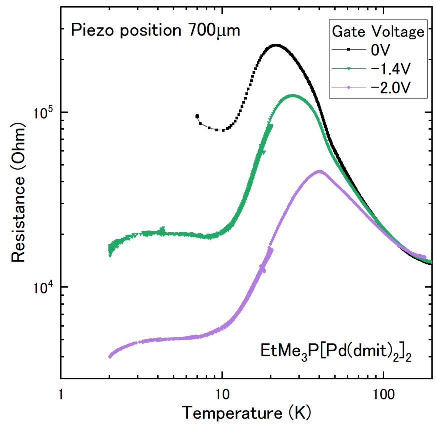

Fig. 4: Electrical resistance of FET processed $EtMe_3P[Pd(dmit)_2]_2$ under the uniaxial pressure for various gate voltage.

Finally, in order to investigate the effect of the carrier doping near the boundary between the superconducting and VBO phases, we fabricated a field effect transistor structure using an electric double layer structure and attempted the hole-doping. Figure 3 shows a schematic picture of an electric double layer transistor. When the gate voltage is applied, carriers are induced on the sample surface. Figure 4 shows temperature dependent electric resistance of hole-doped $EtMe_3P[Pd(dmit)_2]_2$ for various gate voltage when the expansion of piezo actuator fixed at 700 μm. Although the measurement is insufficient, we can say that the conductivity rose with increasing hole density and the small drop of the resistance accompanying the superconducting transition in the high-doped region was observed. In the research of the next year, we will verify the gate voltage dependence in more detail, and conduct the Hall resistance measurement to discuss the relation between the carrier concentration and the electronic state.

Reference

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