Investigation of Superconducting Phase Diagram of an Organic Mott Insulator Using Simultaneous Control of Electric Field and Strain

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In materials with a half-filled band, strong on-site Coulomb repulsion localizes the conduction electron at each site. This type of insulator is called Mott insulator. The phase transition between the Mott insulating and metallic states (the Mott transition) has attracted much attention for these decades because unconventional superconductivity emerges in the proximity of the Mott transition in some cuprates and molecular conductors (organic conductors).

There are two routes for the Mott transition. One is to weaken the electron correlation effectively by increasing the electron transfer energy. This is experimentally achieved through pressure control. The other is to reduce the commensurability between the electron density and lattice potential. Carrier doping enables this operation. If we are able to tune the lattice spacing and doping concentration in a single Mott insulator, we can thoroughly investigate the relationship between the Mott transition and the unconventional superconductivity. Nevertheless, such an experiment had not been done yet because of lacking suitable samples. Inorganic Mott insulators can be precisely doped with chemical substitution but are mostly not compressible enough for the Mott transition. On the other hand, organic Mott insulators can be easily compressed, but have been difficult to dope.

In RIKEN Condensed Molecular Materials Lab, we have been developing the field-effect doping to molecular conductors. Recently, we achieved doping into an organic Mott insulator κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl (hereafter κ-Cl) up to approximately 20%, using the principle of electric-double-layer transistor (EDLT) [2]. In this study, we attempt exploring the superconducting phase diagram in the proximity of the Mott insulating state by combining the EDLT doping and pressure control. Without using a pressure cell, the effective pressure can be varied by bending the substrate [3].

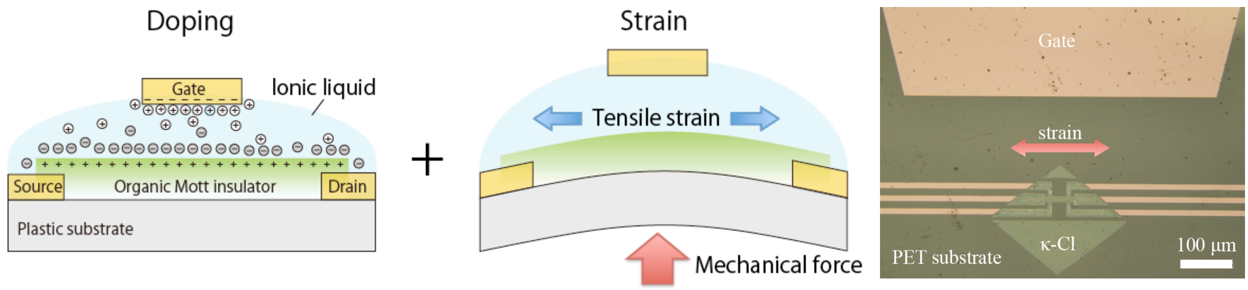

Fig. 1. Schematic side view and optical top view of the organic Mott EDLT based on κ-Cl. A gate voltage varies doping and a strain modifies the effective pressure in the same sample.

Figure 2 shows the temperature dependence of the resistivity in our organic Mott EDLT based on κ-Cl [4]. Without gating (Fig. 2a), the sample changes from a superconductor to an insulator with varying the tensile strain. Non-monotonic temperature dependences are observed in the intermediate state, indicating that the insulating state is antiferromagnetically ordered at low temperature. This reentrant behavior is consistent with the pressure dependence of the resistivity in the bulk κ-Cl crystals [5].

Namely, the effective pressure for κ-Cl was successfully tuned by bending of the substrate enough for the pressure-control phase transition. Next, at a fixed strain, we observed electric-field-induced superconductivity in the insulating state of the same sample (Fig. 2b). This also indicates that the EDLT doping is enough for the doping-control phase transition. Thus, by precisely varying the strain and gate voltage, we can investigate the outlines of the superconducting and insulating phases.

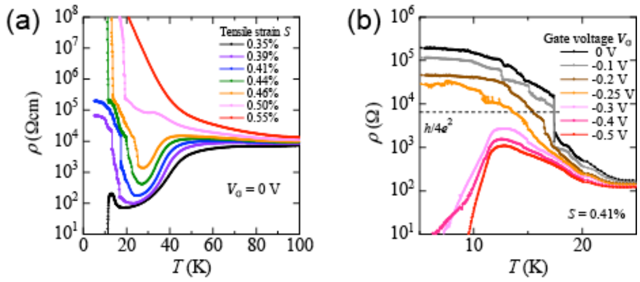

Fig. 2. Temperature dependences of the resistivity (a) at various tensile strains with no gate voltage and (b) at various gate voltages at a fixed tensile strain.

Figure 3 shows color plots of the resistivity against gate voltage and tensile strain, which indicate the outlines of those phases. The superconducting phase appears highly doping-asymmetric and surrounding the Mott insulating phase. On the hole-doped side, the resistivity monotonically decreases against gate voltage and the superconducting state appears in a region where the hole doping level is above ca. 10%. On the other hand, the superconducting state emerges more drastically on the electron-doped side and it disappears by further doping.

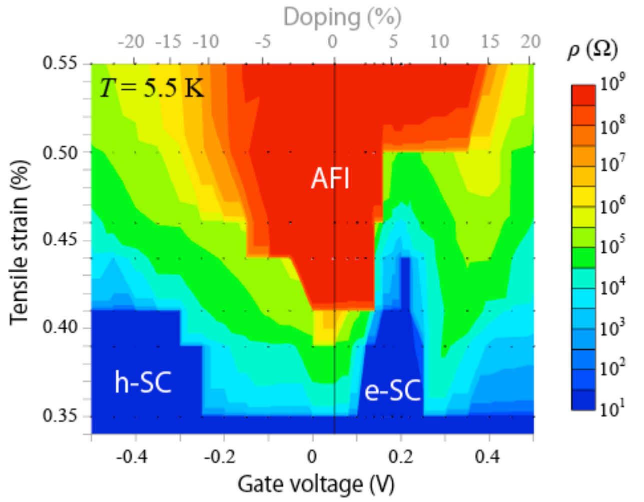

Fig. 3. Gate voltage and tensile strain dependence of the resistivity at 5.5 K. The dark blue regions indicate the superconducting phases. AFI, h-SC, e-SC denote the antiferromagnetic insulating, hole-doped superconducting, and electron-doped superconducting phases, respectively.

The above doping asymmetry (except for the disappearance of the superconducting state under high electron doping) has been qualitatively reproduced by our variational cluster approximation (VCA) calculations for the Hubbard model on an anisotropic triangular lattice. Seemingly it originates from the difference of the density of states between the bottom of the upper Hubbard band and the top of the lower Hubbard band. For future works, we will investigate the origin of the disappearance of the electron-doped superconductivity and apply the method for quantum spin liquid candidates (paramagnetic Mott insulators) to gain insights into the relationship between the superconductivity and magnetism.

References

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