Light-induced superconductivity in an organic strongly-correlated material using a photoactive electric double layer

Electric-double-layer (EDL) transistors has become one of the state-of-the-art methods in condensed matter science that realizes high carrier density at low gate voltage, which enables researchers to obtain gigantic response including field-induced superconductivity. However, this method has one drawback, where modulation of the carrier density is limited only to the high-temperature regime because of freezing of the ionic motions at low temperature.

The current research expands this idea beyond the limit of thermal ionic motion by means of photo-induced EDL formation using a spiropyran monolayer. Spiropyran is a photochromic molecule that can photoisomerize reversibly between the nonionic spiropyran (SP) form and the zwitter ionic merocyanine (MC) form upon UV or visible light irradiation. Therefore, an EDL-like large electric field across the film can be induced or be eliminated by UV light or visible light irradiation, respectively. The advantage of the photochromic EDL system over the conventional one is that it retains the modulation capability even at low temperatures, because these photochromic reactions proceed with photon-energy dissipation under non-equilibrium conditions. Hence, the interface hole-carrier density of an organic strongly-correlated material has become able to be largely tuned even at low temperature (2 K~). Finally, we have succeeded in continuous observation of the reversible light-induced phase transition between Mott insulating phase and superconducting phase.

The photo-active superconducting devices have been fabricated by laminating a thin-single crystal of κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br (κ-Br) (BEDT-TTF = bis(ethylenedithio)tetrathiafulvalene) on top of a Al_2O_3/Nb:$SrTiO_3$ substrate covered with a self-assembled monolayer of spiropyran-derivatives (SP-SAM). Conducting properties of the devices were monitored by four-probe resistivity and magnetic measurements. Because of the weak tensile strain-effect from the substrate, the initial resistances for the devices showed Mott insulator phase in the resistance measurements. After UV irradiation, however, superconducting transition was clearly observed, and remained even after the irradiation was stopped. The resistance recovered to nearly the initial value by visible light irradiation, showing a reversible switching capability. This photo-induced superconducting transition was also confirmed by magnetic susceptibility measurements.

It has been known that, when organic molecules with sizeable dipole moments are organized as a self-assembled monolayer, a cooperative effect of charge transfer occurs between the monolayer and the substrate. The electric field produced by such a monolayer dipole can exceed the field that can be applied across the gate dielectric in a standard FET configuration. We can estimate the difference in the electric fields between the SP and MC states as 4.3 MV/cm, which is much larger than the electric fields that can be produced by applying a gate voltage using the bottom-gate structure. Furthermore, the field-induced carriers and the photo-induced carriers were working as the same type of carriers that are indistinguishable from one another in the κ-Br crystals. This means that SP-SAM EDL can expand the limit of electrostatic carrier doping beyond the density that can be accumulated only by means of a normal FET configuration.