Photo-Induced Phase Transition due to Cooperative Coupling of Phonon and Intra- and Inter-Molecule Electronic Transitions

The photo-induced phase transitions, in which material properties such as color, conductivity, magnetism, etc. suddenly change under light illumination, occur as a result of interactions among light, electrons, and lattice vibrations. Because a sudden phase transition can be controlled by light illumination, the photo-induced phase transitions are attracting attention also from the point of view of applications such as ultrafast switching devices. Photo-induced melting of a charge order is a typical example of such phenomena and has been observed in various materials. In a charge ordered state, the number of electrons localized in a molecule becomes different between neighboring molecules, causing the formation of a symmetry-broken superlattice as shown in Fig. 1.

Recently the research group in the Physics Department of Chuo University performed an extensive theoretical study for the purpose of understanding the initial processes of photo-induced melting in quasi-two-dimensional organic conductor $Et_2Me_2Sb[Pd(dmit)_2]_2$. In this study, a model including the internal structure of molecules was introduced and a new mechanism due to cooperative couplings of lattice vibrations and electron transitions within a molecule and between neighboring molecules was proposed for its photo-induced melting of the charge order.

In this material, strong electron-phonon interaction causes a significant structure difference between a molecule with high electron density and that with low density in the charge-ordered state. In fact, the molecular size is enlarged for high electron density, while it is reduced for low electron density, and the electron transition within a molecule due to light absorption is strongly coupled with the vibration corresponding to this size change. After light absorption due to this transition in a molecule with lower electron concentration, the resulting change in the electronic states causes molecular vibrations and the molecule becomes bigger. Then, the electron density starts to move from a molecule with a bigger size to that with a smaller size. When the size of these two molecules becomes the same, electron transfer between the molecules is resonantly induced, causing sudden melting of the charge order, i.e., photo-induced melting of charge order. This mechanism of the melting is illustrated in Fig. 2.

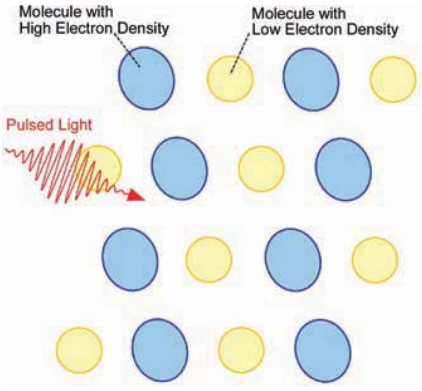

Fig. 1: Molecular crystal with charge order.

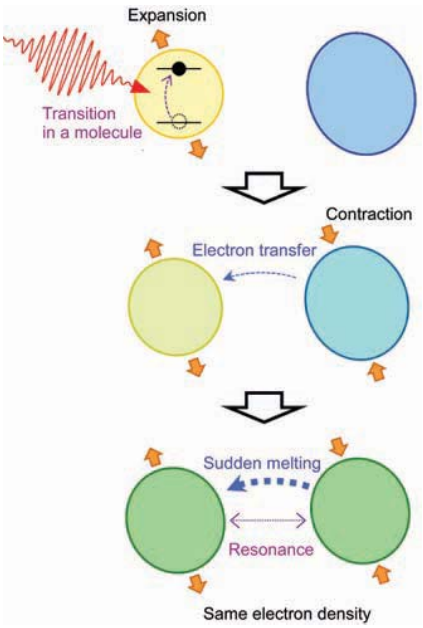

Fig. 2: Schematic illustration of the process of photo-induced melting of charge order.

Reference

K. Nishioka and K. Yonemitsu, J. Phys. Soc. Jpn. 82 (2013) 094716.