

Dynamics of cooperative lattice-charge (spin) coupled phenomena under far equilibrium condition induced by fs laser light irradiation

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In the dynamical process of the photo-induced phase transition, it has been expected that new exotic phase appears under nonequilibrium condition, so called as a false ground state which can never be achieved in the thermally equilibrium ground state. In this talk, we introduce that such new phase can be really achieved in the molecular based correlated system as a result of transformation of charge order pattern due to competition between electron-electron and electron-lattice interactions.

Here, we mainly discuss about the gigantic photo response observed for the quasi-one-dimensional, quarter-filled (EDO-TTF)₂PF₆ (EDO-TTF = ethylenedioxy-tetrathiafulvalene) crystals. Compared to other quasi-one-dimensional, quarter-filled systems, (EDO-TTF)₂PF₆ is known to have unique features: the phase transition temperature from metal to insulator is anomaly high ($T_c = 280$ K), the charge order in the low temperature phase is (0110), which represents the order of $D^0D^+D^+D^0$ ($D = \text{EDO-TTF}$), and the EDO-TTF ion is largely deformed in the low temperature phase. In addition, ultrafast gigantic reflectivity change within 200 fs has been recently observed by photo-excitation in the low temperature phase and this reflectivity change was assigned to the photoinduced insulator-to-metal phase transition. We show that photoinduced state is never in agreement with any one in thermal equilibrium utilizing ultrafast spectroscopic method and attributed to creation of intrinsic photoinduced state (false ground state) comparing with the time-dependent numerical calculation. Also we found that the coherent reflectivity oscillation becomes incoherent at mid-infrared probe photon energies and showed that the calculation reproduces this phenomenon

These result clearly shows a key role of dynamical structural science for achieving artificial control of the nature of matters utilizing false ground state under non-equilibrium condition.