

Short- and long-range charge correlations and their photoinduced dynamics in charge-ordered organic ferroelectrics (TMTTF)₂X

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Short-range electronic correlations have been intensively investigated for their contribution to exotic condensed matter phenomena such as high-temperature superconductivity, colossal magnetoresistance, etc [1]. Particularly intriguing is their nontrivial dynamics seen during photoinduced phase transitions [2], illustrating a promising pathway to unprecedented nonequilibrium states which might be thermodynamically inaccessible. We report herein ultrafast photoresponses of strongly-correlated charges in quarter-filled organic conductors (TMTTF)₂X (TMTTF: tetramethyl-tetrathiafulvalene; X: anion) [3], which exhibit prototypical electronic ferroelectricity [4].

By employing terahertz (THz) emission and absorption spectroscopy, we were able to disentangle long-range charge order (CO), forming ferroelectric domains with a hundreds-micrometer scale [Fig. a], and short-range charge correlations that activate the absorption at ~ 65 cm⁻¹ well above CO transition temperature T_{CO} [Fig. b] [5]. For X = PF₆ and AsF₆, photoexcitation triggers ultrafast (< 0.1 picosecond) enhancement of short-range charge correlations in contrast to melting of long-range CO [Fig. c], the latter of which is also seen in other electronic ferroelectrics [6].

For tetrahedral anion cases (X = ReO₄ and BF₄), domain walls separating antiparallel polarization domains were found to be controllable by anion order, and to host, upon photoexcitation, real-time oscillation of THz emission amplitude with a period of a few picoseconds. The key factor for these ultrafast dynamics is the photoexcitation in the vicinity of phase boundaries or charge fluctuations therein.

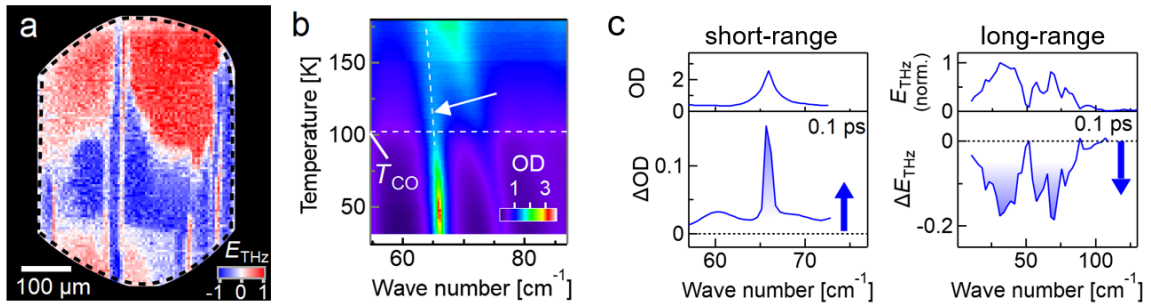


Figure. Results for X = AsF₆. (a) Domains of antiparallel ferroelectric polarization, or long-range CO, observed via THz emission microscopy. (b) THz absorption peak of intermolecular vibration mode activated by short-range charge correlations. (c) Photoinduced (50 K, 0.89 eV, 0.3 mJ/cm²) enhancement of the absorption peak (left) and decrease of THz emission amplitude (right).

References

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