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Domain Structures and Photoinduced Dynamics of Electronic Ferroelectrics Investigated by Terahertz-Emission Microscopy

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Abstract

Electronic ferroelectricity, which is peculiarly driven by charge ordering (CO) or strong Coulomb repulsion among charges,[1] has become an important topic in contemporary condensed matter physics, toward gigantic optical responses such as ultrafast photoinduced insulator-to-metal transition or large optical nonlinearity.[2,3] We report herein recent results of terahertz (THz)-wave-emission microscopy to reveal domain structures of prototypical electronic ferroelectrics: strongly-correlated quasi-one-dimensional organic conductors (TMTTF)₂X (TMTTF: tetramethyl-tetrathiafulvalene, X = SbF₆, AsF₆, PF₆, ReO₄, BF₄).[4,5]

Fig.: (a) Schematics of the THz emission microscopy. (b) THz emission images of (TMTTF)₂AsF₆, measured before and after photoexcitation (0.89 eV, 7 mJ/cm², 30 min). E_{THz} represents THz electric field (a.u.) at an extremum of the time-domain waveform. (c) Images of (TMTTF)₂ReO₄ at $T = 165$ K ($T_{\text{AO}} < T < T_{\text{CO}}$), measured during cooling or heating procedures.

Upon irradiating femtosecond pulses to the crystals, the THz emission occurs via 2nd-order optical nonlinearity activated by ferroelectricity or concomitant non-centrosymmetry. Therein, as shown in Fig. (a), the resultant THz waveform inverts upon inversion of ferroelectric polarization hence antiparallel ferroelectric domains are distinguishable [6].

The observed characteristic domain structures with size of ~ 100 μm (resolution: 5 μm), and their changes upon photoexcitation [Fig. (b)] or temperature cycles [Fig. (c)] will be discussed.

Keywords: strongly correlated electrons, charge order, electronic ferroelectricity, ferroelectric domains, terahertz, photoinduced phase transition, ultrafast spectroscopy

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