Terahertz-driven activation of ferroelectric mode in a molecular material studied by pump-probe spectroscopy

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Controlling ferroelectric materials with light is a major challenge. Among different materials, the mixed-stack TTF-CA molecular system is of particular interest because its ferroelectricity originates from coupled inter-molecular charge transfer and symmetry breaking, which are both simultaneously modified at the first-order phase transition (T_c=81 K). Indeed, asides from structural modifications, such as dimerization and ferroelastic ordering [1], the intermolecular charge transfer changes molecular ionicity from neutral (0.3 e) to ionic (0.6 e). Due to this coupling, probing a charge-transfer sensitive band of TTF-CA can be used to track vibrational motions.
We present here a Terahertz pump-Optical probe experiment performed within the neutral and paraelectric phase of TTF-CA. The use of THz pulses with a bandwidth spanning from 0 to 2 THz enables the excitation of low frequency polar phonon modes [2] involved in the ferroelastic ordering toward the low temperature phase. Following this excitation, we use the transient change of reflectivity probed on a CT sensitive band [3,4] for tracking the THz-driven activation of such polar lattice vibrations. We will present the temporal evolution of the response with electric field's amplitude and on lowering the temperature. On approaching the phase transition, the photoresponse is clearly enhanced. Since the amplitude of the THz electric field is two orders of magnitude higher than the coercive field observed in the ferroelectric hysteresis loop of TTF-CA, THz-driven activation of polar modes is promising for inducing ferroelectric order and controlling ferroelectricity on ultrafast timescales.

References

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