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Ultrafast dynamics of a charge density wave in non-equilibrium in 1T-TiSe2

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Spontaneously broken symmetries in solids often result from many-body interactions that fall outside the scope of conventional solid-state models. As a consequence, some of the most fascinating phase transitions such as stripe formation, unconventional superconductivity or colossal magnetoresistance are still not fully understood. Time-resolved multi-terahertz spectroscopy is an ideal optical technique to resolve the microscopic interactions in complex materials and to trace their dynamics on an ultrafast time scale. We apply this optical technique to investigate the non-equilibrium dynamics of a charge density wave (CDW), an intriguing phase characterized by a static modulation of the electronic charge distribution accompanied by a periodic lattice distortion (PLD). In the transition metal dichalcogenide 1T-TiSe2, the CDW occurs at temperatures below 200 K and has been studied with several experimental techniques such as x-ray diffraction and photoemission experiments. These experiments have highlighted two potential driving mechanisms of the phase transition: excitonic correlations and a Jahn-Teller lattice distortion. Despite intense efforts, both in experiments and theory, no consensus could be reached on which one of these interaction mechanisms is the actual driving force of the CDW and which one is simply a consequence of it. Here we perturb the CDW in a 80 nm thin film of single crystalline TiSe2 and monitor the subsequent ultrafast dynamics of the two constituent orders via PLD induced phonons resonances and characteristic terahertz fingerprints of excitonic order [1]. Our experiment reveals a new transient phase in which the PLD persists in a coherently oscillating state while the excitonic order is entirely quenched. These results prove that the CDW transition in 1T-TiSe2 is not solely driven by excitonic correlations, but also by a Jahn-Teller distortion in a cooperative process. Our low-energy spectroscopic technique of ultrafast dynamics associated to a complex long-range order provides a new benchmark for exploring this fascinating class of quantum phenomena in strongly correlated materials.

[1] M. Porer et al. Non-thermal separation of electronic and structural orders in a persisting charge density wave. Nature Materials 13, 857 (2014)

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