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Transient hexatic phase observed by ultrafast high-coherence nanobeam diffraction

T. Domröse [1], T.H. Danz [1], S. Yalunin [1], C. Ropers [1]

[1]Max Planck Institute for Multidisciplinary Sciences - Göttingen (Germany)

Abstract

Ultrafast transmission electron microscopy (UTEM) is a powerful technique that enables the investigation of non-equilibrium structural dynamics by means of diffraction in a laser pump/electron probe approach [1]. The sensitivity of such experiments to structural modifications induced by the optical excitation is often limited by the coherence of the electron source. In order to achieve sufficient reciprocal-space resolution, diffractive probing typically averages over nanoscale heterogeneity by forming probes with a diameter of several 100µm.

In the Göttingen UTEM, ultrashort electron pulses are generated by linear photoemission from a tip-shaped field-emitter, yielding a particularly high coherence (Fig. 1a). This enables ultrafast diffraction experiments with a collimated nanometre-sized electron beam and sub-picosecond temporal resolution (Fig. 1b) [2]. In this work, we harness these capabilities to investigate the formation kinetics of an incommensurate charge-density wave (CDW) phase following optical excitation of a layered material [3]. The high-coherence electron source in conjunction with the small probing volume allows us to track the establishment of three-dimensional long-range order encoded in the temporal evolution of the CDW reciprocal lattice rods.

At early times and within the material layers, the CDW is characterised by a strongly anisotropic broadening of the associated diffraction spots, suggesting a transient state with intact bond-orientational order, but reduced translational symmetry (Fig. 1d). This anisotropy decays after around 10 ps, followed by ongoing spot sharpening on longer timescales.

Along the out-of-plane directions, a reconstruction of the CDW rocking curve from a time-dependent tilt series reveals an initial increase of scattered CDW intensities that is broadly spread across the entire out-of-plane wave vector component (Fig. 1c). Only afterwards, a well-defined diffraction peak indicative of the equilibrium CDW stacking sequence emerges.

This behaviour implies an initial loss of interlayer correlations and the formation of a quasi-two-dimensional intermediate at early times. We identify this intermediate as a transient hexatic phase characterised by a high density of unbound topological defects. Induced by the optical excitation, their presence translates into the observed characteristic broadening of diffraction spots along the azimuthal direction (green arrow in Fig. 1d) [4]. From here, the ongoing pairwise recombination of dislocations and the transition into a three-dimensional system lead to the establishment of long-range order associated with the thermalised crystalline IC phase found at late times.

In the future, we expect our experimental approach to provide further insights into phase formation kinetics in other correlated materials and to elucidate the impact of spatial heterogeneity on ultrafast structural dynamics.

References

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Ultrafast CDW dynamics probed by UTEM