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Measuring Collective CDW Excitations throughout momentum space using XUV ARPES

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We use time- and angle-resolved photoemission spectroscopy with sub-30-fs XUV pulses to map the time- and momentum-dependent electronic structure of photoexcited 1T–TaS2. This compound is a two-dimensional Mott insulator with charge-density wave (CDW) ordering. The ultrafast time-resolution and wide angular acceptance of the experiment allow us to clock the melting dynamics of the CDW state and decompose its charge and lattice order throughout momentum space. Upon charge transfer photo-excitation, or photo-doping, not only the Mott gap at the Fermi level but also the CDW gaps at finite binding energy melt on sub-vibrational timescales: Charge order, evidenced by splitting between occupied sub-bands at the Brillouin zone boundary melt. Surprisingly, this occurs well before the lattice responds. This indicates that the CDW gaps close in response to a redistribution of charge, rather than a repositioning of atom. Subsequently, spectral intensity migrates from shadow bands in the first zone back out to higher momentum, revealing the unfolding of the Brillouin-zone that accompanies incipient lattice relaxation along the coordinate of the Raman active amplitude mode following photo-doping. At longer timescales, charge order and lattice distortions lock again, displaying a modulation of spectral weight at the frequency of the amplitude mode.
In blue bronze (K0.3MoO3), a prototypical 1D-CDW system, we investigate the collective response of a melting CDW wave. We find the excitation of both amplitude and phason response are excited coherently, making it possible to reconstruct the coherent dynamics of a complex order parameter and to connect it to the complex band structure.
These experiments show the power of time- and angle resolved photoemission experiments performed with XUV radiation, a significant technical improvement compared to the more common use of 6-7 eV radiation.

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