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Femtosecond time-resolved photoemission of layered charge-density-wave compounds

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Charge-density waves (CDWs) are broken-symmetry states of low-dimensional materials that are brought about by strong electron-phonon interaction. Yet, surprisingly, a clear microscopic understanding beyond this statement has not really evolved for this classical paradigm of condensed matter physics. In quasi-two-dimensional systems, for example, the common approaches based on ARPES band structure results—looking for nested sections of the Fermi surface or for a peak in the electronic susceptibility—have almost no predictive power. Apparently, a more successful explanation has to take into account the delicate balance between several factors including not only electronic and phononic structure, but also electron-electron (electron-hole) and electron-phonon interactions. Here, we will explore whether femtosecond time-resolved XPS and ARPES using pulsed extreme ultraviolet radiation generated by a free-electron laser (FLASH) and a table-top high-harmonic-generation source can provide novel insights into the relative roles that the various factors play in CDW formation. We will focus on three conspicuous CDWs in prominent members of the family of layered transition-metal dichalcogenides: the (√13×√13) CDW in the Mott insulator 1T-TaS2, the c(2√3×4)rect. CDW in the Peierls insulator RbxTaS2, and the (2×2×2) CDW in the possible excitonic insulator 1T-TiSe2. Our particular goal will be to reveal the relative importance of electronic (excitonic) or phononic contributions to each CDW transition by relating measured vaporization and relaxation times of CDW-induced spectral features to typical elementary time scales in layered compounds.

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