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Shooting the electronic structure movie: Femtosecond time-resolved photoemission of layered charge-density-wave systems

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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 universal 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, more successful explanations have to take into account the delicate balance between several factors including not only electronic and phononic structure, but also electron-electron and electron-phonon interactions. Here, we will show that femtosecond time-resolved XPS and ARPES using pulsed extreme ultraviolet radiation generated by the free-electron laser FLASH [1] as well as by a table-top high-harmonic-generation source [2] 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 $(\sqrt{13} \times \sqrt{13})$ CDW in the Mott insulator $1T$-TaS_2, the $c(2\sqrt{3} \times 4)rect.$ CDW in the Peierls insulator Rb_xTaS_2, and the $(2 \times 2 \times 2)$ CDW in the possible excitonic insulator $1T$-$TiSe_2$. The specific program is to reveal the relative importance of electronic and phononic contributions to the various CDW transitions by relating measured melting and relaxation times of CDW-induced spectral features to typical elementary time scales in layered materials.

[1] S. Hellmann *et al.*, Phys. Rev. Lett. **105**, 187401 (2010).
[2] T. Rohwer *et al.*, Nature **471**, 490 (2011).