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Ultrafast Dynamics in $(\text{TaSe}_4)_2\text{I}$ Triggered by Optical and X-Ray Excitation

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Dimensionality plays a key role for the emergence of ordered phases such as charge-density-waves (CDW), which can couple to, and modulate, the topological properties of matter.

In this work, we study the out-of-equilibrium dynamics of the paradigmatic quasi-one-dimensional material $(\text{TaSe}_4)_2\text{I}$, that exhibits a transition into an incommensurate CDW phase when cooled just below room temperature, namely at $T_{\text{CDW}} = 263\text{K}$.

We make use of both optical laser and free-electron laser (FEL) based time-resolved spectroscopies in order to study the effect of a selective excitation on the normal-state and on the CDW phases, by probing the near-infrared/visible optical properties both along and perpendicularly to the direction of the CDW, where the system is metallic and insulating, respectively.

Excitation of the core-levels by ultrashort X-ray FEL pulses at 47 eV and 119 eV induces reflectivity transients resembling those recorded when only exciting the valence band of the compound - by near-infrared pulses at 1.55 eV - in the case of the insulating sub-system. Conversely, the metallic sub-system displays a relaxation dynamics which depends on the energy of photo-excitation.

Moreover, excitation of the CDW amplitude mode is recorded only for excitation at low-photon-energy. This fact suggests that the coupling of light to ordered states of matter can predominantly be achieved when directly injecting delocalized carriers in the valence band, rather than localized excitations in the core levels.

On a complementary side, table-top experiments allow us to prove the quasi-unidirectional nature of the CDW phase in $(\text{TaSe}_4)_2\text{I}$, whose fingerprints are detected along its c -axis only.

Our results provide new insights on the symmetry of the ordered phase of $(\text{TaSe}_4)_2\text{I}$ perturbed by a selective excitation, and suggest a novel approach based on complementary table-top and FEL spectroscopies for the study of complex materials.

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