

Comment on “Photoinduced Dynamics and Momentum Distribution of Chiral Charge Density Waves in 1T-TiSe₂”

A recent time-resolved x-ray diffraction (trXRD) study reported that under femtosecond laser excitation, the charge-density-wave (CDW) peak of 1T-TiSe₂ shows a notable difference (20%) in its intensity reduction between left and right circularly polarized (LCP and RCP) pumps [1]. This observation was attributed to the helicity-dependent coupling between circularly polarized light and chiral CDW domains. Here we raise technical concerns about the pump-probe geometry used by the authors and the resulting polarization state of the pump laser, which appear unsuitable for detecting the circular dichroism they claim to have observed.

In Ref. [1], the circularly polarized 800-nm laser was incident at a grazing angle of $\sim 10^\circ$ [Fig. 1(a)]. The pump beam hence became elliptically polarized as it transmitted into the sample at a 19° refraction angle [2]. This elliptically polarized beam, as represented by the triangles in the Poincaré sphere in Fig. 1(c), can be decomposed into circular ($\mathbf{E}_{cp}$) and linear ($\mathbf{E}_{lp}$) components with an amplitude ratio of $E_{cp}/E_{lp} \approx 2:1$, indicating a substantial $\mathbf{E}_{lp}$ contribution to the CDW dynamics. However, under purely linearly polarized excitation, the transient CDW suppression shows a polarization angular dependence that markedly deviates from expectations based on the transmission amplitudes of p - and s -polarized light [1]. This unexplained discrepancy raises questions about the interpretation of the CDW dynamics driven by elliptical pump light.

To isolate the effect of the circularly polarized pump field, we conducted ultrafast electron diffraction (UED) measurements [3] with collinear laser pump and electron probe beams under a much reduced angle of incidence [Fig. 1(a)]. The polarization state of the pump was carefully characterized at the sample position [see Poincaré spheres in Figs. 1(c) and 1(d)] and found to be much closer to pure LCP/RCP. Our sample was prepared using a similar method reported previously [4,5]. A typical static diffraction pattern at 90 K is shown in Fig. 1(b), featuring the CDW peaks at $(\pm \frac{3}{2}, \pm \frac{1}{2}, \mp \frac{1}{2})$. Following 800 nm excitation with nominally LCP and RCP pulses, the CDW peak intensity decreases within 1 ps and subsequently recovers over the next few picoseconds [Fig. 1(c)]. We also carried out experiments at a $10.6 \mu\text{m}$ pump wavelength, which is known to optically train the chiral CDW domains [6].

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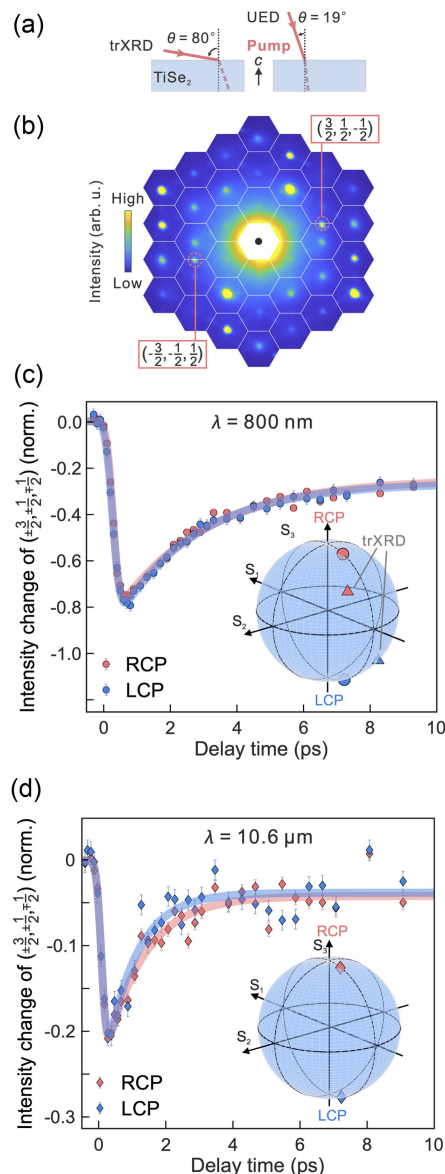


FIG. 1. (a) Pump laser propagation directions in the trXRD [1] and UED experiments. (b) Static electron diffraction pattern in the CDW phase at 90 K. (c),(d) Evolution of the averaged intensity of CDW peaks in (a) following photoexcitation by 55-fs (FWHM), 1-mJ/cm², 800-nm pulses (c) and 150-fs, 2.4-mJ/cm², 10.6- μm pulses (d). Curves are fits to Eq. (1) in Ref. [1]. The UED was operating with 3.2 MeV electron kinetic energy at a 360 Hz repetition rate. Insets of (c),(d): Poincaré sphere representations of the pump polarizations inside the sample. Triangles indicate the polarizations used by trXRD in Ref. [1].

For both wavelengths, the overall CDW peak dynamics are very similar to those reported in Ref. [1], but there is no discernible difference in the intensity reduction between the RCP and LCP pumps [Figs. 1(c) and 1(d)].

To summarize, the grazing-incidence geometry and the unexpected CDW response to linearly polarized excitation in Ref. [1] complicate a reliable assessment of helicity-dependent effects when the pump substantially deviates

from circular polarization. In our measurements, where the pump is much closer to being circularly polarized, no circular dichroism was detected in the CDW dynamics.

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Data availability—The data that support the findings of this Comment are freely available at [7].

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