Ultrafast x-ray scattering from collective modes of the charge density wave in (TaSe$_4$)$_2$I

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We report ultrafast x-ray scattering experiments of the quasi-1D charge density wave (CDW) material (TaSe$_4$)$_2$I following photoexcitation with femtosecond infrared laser pulses, carried out at the LCLS and SACLA hard x-ray free-electron laser facilities. Time-resolved diffraction signals were obtained at the first- and second-order CDW sidebands surrounding the (2 2 4) and (4 2 0) crystal Bragg peaks. Two modes with sub-THz frequencies are observed at the CDW sidebands, one of which we identify as the amplitude mode derived primarily from the transverse acoustic component of the CDW at a frequency 0.11 THz based on the polarization dependence of the atomic displacements relative to the static CDW distortion. The dynamics of this acoustic amplitude mode are described well by a model of a displacive excitation. We explain the displacive excitation of an acoustic amplitude mode as excited indirectly through its coupling to the optical phonon component associated with the tetramerization of the tantalum chains. We observe linearly-dispersive features at GHz frequencies in the vicinity of the CDW sidebands that do not coincide with the acoustic velocities of the high symmetry phase.

Charge density wave (CDW) materials are low dimensional systems that exhibit spontaneously broken symmetries associated with instabilities in the Fermi surface. These systems are characterized by a modulation of the valence electron density and a corresponding lattice distortion due to the electron-lattice interaction. The fundamental collective modes of the CDW ground state correspond to excitations of the amplitude and phase of the condensed electron density and concomitant lattice modulation. While frequency-domain inelastic scattering techniques with neutrons or x-rays can generally access collective excitations, they probe the equilibrium spectrum and cannot disentangle couplings among these modes or with other degrees of freedom. Time-domain scattering techniques can provide a direct probe of these couplings by perturbing and observing different degrees of freedom. Here we use an x-ray free electron laser (XFEL) to study the collective modes of the CDW in (TaSe$_4$)$_2$I (TSI) after ultrafast near-infrared excitation. Exploiting the extreme high-frequency and wavevector resolution enabled by the time-domain approach, we find that the amplitude mode of this CDW has a strong acoustical character, a manifestation of the complex nature of the order parameter. Furthermore, we find that near-IR excitation produces coherent low-frequency modes that disperse linearly around the CDW wavevector, consistent with phase excitations of the CDW. However, these also modulate the average lattice Bragg peaks and may correspond to lattice strain, but their propagation speeds do not match the known speeds of sound for acoustic modes of the crystal.

(TaSe$_4$)$_2$I (TSI) is a quasi-one-dimensional Weyl semimetal that exhibits a CDW instability at a temperature of $T_c = 263$ K. As a Weyl-CDW material it has attracted much recent interest due to its potential
for realizing a so-called dynamical axion insulator state \[12\], a correlated topological phase where the phase mode of the CDW becomes an analogue of the proposed axion field in high energy physics \[13, 14\]. The high-symmetry tetragonal structure, with space group I422, is chiral and is comprised of parallel screw-like TaSe\(_2\) chains separated by rows of iodine atoms as shown in Figs. 1(a,b).

On account of its chain-like structure TSI has a very high electronic anisotropy \[15\], with a relatively high conductivity along the chain resulting from a band formed by the Ta 5\(d_2\) orbitals \[16\]. Although the Ta-Ta distances along a chain are all equivalent there are actually two crystallographically and chemically distinct Ta sites due to slightly varying iodine environments, which have different formal oxidation states of +4 and +5. This yields a filling of the Ta 5\(d_2\) band up to the Z-point, which corresponds to a quarter-filling of this band in the approximately one-dimensional Brillouin zone of a single tantalum chain along the c-axis \[16\]. The strongly 1D and partially-filled Ta 5\(d_2\) band suggests that the material may exhibit an instability to a tetramerization of the Ta atoms along the chain axis, and \textit{ab initio} density functional theory indeed predicts that the I422 phase is unstable to such a distortion \[17\]. However, the dominant component of the CDW distortion below \(T_c\) is not a homogeneous Ta tetramerization but rather a frozen transverse acoustic (TA) phonon at a finite wavevector \(q_{\text{CDW}}\) \[18\]. A component of the lattice distortion that corresponds to the Ta-tetramerization pattern modulated at \(q_{\text{CDW}}\) is also present, albeit with an amplitude of less than \(1/6\) that of the TA component. Given its much smaller amplitude relative to the dominant TA distortion, this secondary lattice distortion can only be detected unambiguously using resonant x-ray diffraction techniques \[19\]. Our results presented here show that the amplitude mode, which strongly modulates the CDW Bragg peaks, has a strong transverse acoustic character. This shows that the CDW instability involves multiple distortions rather than the naively expected optical tetramerization mode.

The TSI crystals were grown by chemical vapor transport of the elements in a thermal gradient of 600 °C to 500 °C, as described in Refs. \[20, 21\]. The experiments were performed at the XPP instrument of the Linac Coherent Light Source (LCLS) at the SLAC National Accelerator Laboratory and at the BL3 hutch of the Spring-8 Angstrom Compact free electron LAser (SACLA) at the RIKEN SPring-8 Center. A schematic of the experimental configuration is shown in Fig. 1(c). In the experiments at LCLS (SACLA) a 2 \(\mu\m (800 \mn)\) infrared laser pulse with a duration of 30 fs (45 fs) was used to excite the TSI sample with a (1 1 0) surface normal at an incidence angle of \(\gamma = 25°\) (45°). Following excitation the sample was probed with a time-delayed \(\sim 30\) fs x-ray pulse, and the delay-dependent single-shot diffraction patterns were recorded on a 2D array detector positioned 600 mm (620 mm) away from the sample. The x-ray photon energy and bandwidth were 9.55 keV and 0.5 eV respectively, and the timing jitter between the pump and probe beams was corrected on a single-shot basis using a spectroencoding diagnostic tool to achieve a time resolution of \(< 80\) fs \[22, 23\]. The pump laser pulses were p-polarized with respect to the sample surface and focused to a spot on the sample with area of 0.15 \(\times\) 1.2 mm\(^2\) (0.15 \(\times\) 0.2 mm\(^2\)). The x rays were incident at grazing angles between 0.5°-1° with respect to the sample surface in order to match

![FIG. 1. (a,b) Conventional unit cell of TSI in the high-temperature (I422) phase. The lattice constants are \(a, b = 9.5317\) Å and \(c = 12.761\) Å. (c) Schematic of the experimental setup. (d) Integrated diffraction image of a scan of the sample azimuth \(\phi\) around the (2 2 4) Bragg peak in the CDW phase. Several first- and second-order sidebands are visible. (e) Reciprocal space mapping of the scan represented in (d), showing the first-order sidebands as the corners of a tetragonal box as well as two second-order sidebands. The colorbar indicates the value of \(\phi\) in degrees relative to the (2 2 4) Bragg condition. The discrete spots passing diagonally through the Bragg peak are the intersections of the Bragg rod with the Ewald sphere as \(\phi\) is varied.](image-url)
the x-ray and optical penetration depths, and the x-ray
spot sizes on the sample were 0.1 × 1.2 mm² (0.03 × 3.4
mm²). The sample temperature was maintained at ∼150
K using a nitrogen gas cryostream cooling system. We
observe virtually no pump wavelength dependence of the
response.

Fig. 1(d) shows the sum of the detector images ob-
tained by scanning the sample azimuthal angle φ (i.e., the
angle parametrizing rotations about the sample normal)
early (2 2 4) crystal Bragg reflection at a temperature
of ∼150 K. Several sidebands corresponding to the CDW
lattice distortion are visible around the central crystal
Bragg peak. Fig. 1(e) shows a mapping of this data into
reciprocal lattice indices (h k l). The eight first-order
sidebands closest to the crystal Bragg peak are at posi-
tions (2±α 2±α 4±β), where α ≈ 0.055 reciprocal lattice
units (r.l.u.) and β ≈ 0.112 r.l.u. Also visible are some
of the second-order sidebands at (2 ±2α 2 ±2α 4 ±2β).
For simplicity of notation, we will refer to sidebands in-
dices by their sign, e.g. (−−+) represents (−α −α −β),
whereas 2× (−++−) is (−2α −2α 2−β). The correspond-
ing labels for each of the sidebands observed are provided
in Fig. 1(d). Two labels are provided in the cases where
two different sidebands fall on the same detector posi-
tion at different values of φ. According to the structural
determination [18] the CDW phase is monoclinic and fea-
tures a single wavevector with 4 possible orientational do-
 mains, which has been confirmed experimentally by the
observation of single-domain x-ray diffraction [19]. Thus
in Fig. 1(e) the sidebands that lie on a common line
passing through the central (2 2 4) crystal Bragg peak
belong to the same domain, and sidebands related by
90 deg rotations about the l-axis correspond to different
domains.

Figs. 2(a-c) show the normalized diffracted signals
I(t)/I_{eq} integrated over regions of interest (ROIs) around
the (+ + +), (+ −−), and 2× (+ −−) sidebands as a
function of pump-probe delay, where I(t) is the recorded
x-ray intensity and I_{eq} is the equilibrium intensity. As
is evident from the amplitude of the Fourier transforms
shown in Figs. 2(d-f), there are three prominent oscilla-
tory components at 0.11, 0.23, and 0.46 THz. The 0.11
THz mode appears at all sidebands with approximately
equal normalized amplitude, while the 0.23 and 0.46 THz
modes vary significantly with sideband index. For exam-
ple, the (+ + +) sideband in Fig. 2(d) contains all three
components, while the (+ −−) sideband in Fig. 2(e)
only shows the 0.11 THz mode. Based on the appearance
of the 0.11 THz mode in all sidebands with equal rela-
tive magnitude we assign this mode to be the amplitude
mode of the CDW. Furthermore, this mode modulates
both the first- and second-order sidebands with the same
0.11 THz frequency, clearly indicating that it is related
to the amplitude mode of the system rather than to an
unrelated acoustic phonon which would double its fre-
quency between q_{CDW} and 2q_{CDW}. We also note that,
based on measured elastic constants, 0.11 THz is close to
the transverse acoustic (TA) phonon frequency at q_{CDW}
for the high-temperature phase [24]. We note that this
acoustic mode dominates the static CDW distortion [18].

We note that similar frequencies as those in Fig. 2 have
been reported in optical reflectivity measurements [25].
These have been tentatively associated with the CDW
phase because they disappear at T > T_c. However, op-
tical measurements are only sensitive to long-wavelength
excitations and have limited sensitivity to the order pa-
rameter, and thus they cannot easily distinguish a col-
lective mode of the CDW from a zone-folded spectator
phonon mode at q_{CDW}. On the other hand, in our x-ray
measurement we are able to determine the polarizations
of the observed modes at q_{CDW} relative to the the CDW
distortion. Furthermore, the observation of the 0.11 THz
mode at 2q_{CDW} allows us to identify this mode as being
an amplitude mode of the CDW. The other two acoustic
modes at q_{CDW} as determined from calculations based on
experimental values of the I422-phase elastic constants
[24] are a quasi-transverse acoustic (QTA) mode at a
frequency of 0.18 THz and a quasi-longitudinal acoustic
(QLA) mode at a frequency of 0.34 THz. The calculated
polarization of either of these modes is consistent with
the observed variation of the 0.23 THz mode amplitude
among the different CDW sidebands. We thus assign
the 0.23 THz mode to a renormalized acoustic degree of
freedom that is derived from either the high-temperature
QTA or the QLA acoustic modes. Given that the 0.46
THz mode appears only when the 0.23 THz mode is
prominent, it is likely that this component of the signal
arises due to a nonlinear dependence of the structure
factor on the amplitude of this mode. This is supported
by the fact that no component with a frequency similar to
0.46 THz was observed in optical transient reflectivity
measurements [27].

The main distortion associated with the CDW seems
to have acoustic character, however x-ray diffraction re-
finement reported a small optical component [18]. Fur-
thermore, ultrafast optical experiments and Raman scat-
tering report a prominent mode at ∼2.7 THz, which dis-
appears at T > T_c and is presumably the collective mode
corresponding to this optical component of the CDW.
We note that the structure factor of (2 2 4) is not sen-
sitive to this type of distortion [26] and thus we do not
expect this mode in the data in Fig. 3. The optical com-
ponent of the CDW, associated with tantalum motion,
is extremely small but its signature can be enhanced by
resonant diffraction on the Ta edge [19]. Furthermore,
density functional theory (DFT) calculations [17] predict
an instability of an optical mode of 1422 with symme-
try B_1 and/or B_2 that would produce a tetramerization of
the one-dimensional Ta chain similar to the distor-
tion associated with this optical mode. All these facts
taken together suggest that the CDW transition should be
thought of as two combined distortions, one optical and
one acoustical [28].

We model the signals at the (2 2 4) sidebands as a
displacive excitation of the harmonic oscillator [27] as-
associated with the acoustic component of the CDW. We
define the amplitude of the CDW relative to the equilibrium distortion as \((1 + \xi)\), with \(\xi\) the mode displacement, which oscillates around a displaced equilibrium position at \(t > 0\). The structure factor of the CDW is \(\propto (1 + \xi)^2\) [28] (details provided in the Appendix). Fig. 3 shows the normalized signals observed at the \((- + -)\) sideband for a range of excitation fluences. Dashed black lines are the corresponding fits based on the displacive harmonic oscillator [27]. The agreement shown in Fig. 3 suggests that the acoustic mode is excited through a sudden displacement of its equilibrium position (the cosine phase in the oscillation is a signature of this effect [27]). However, this mode is not itself expected to couple directly to the valence electrons, and is inconsistent with a displacive excitation by the photoexcited charge. However, as mentioned above, the optical-like component of the CDW lattice distortion is derived from a linear combination of the lowest-frequency \(B_1\) and \(B_2\) modes of the \(I422\) phase, related to the Ta-tetramerization distortion, which have been predicted to be unstable [17]. These modes couple strongly to the charge and through anharmonic coupling can induce a displacive mechanism on the acoustic mode. In fact, a phenomenological Ginzburg-Landau model of coupled optical and acoustic modes was proposed to explain several features of the CDW in TSI [24]. In this model, two soft optical modes of \(B_1\) and \(B_2\) symmetries with quartic bare potentials are coupled to the lattice strain. By including coupling terms that involve the gradients of the phonon amplitudes they find a CDW lattice modulation at finite wavevector with mixed acoustic and optical character, qualitatively matching the observed distortion [18]. This mode of optical character associated with the Ta-tetramerization likely corresponds to the prominent 2.7 THz mode observed in ultrafast reflectivity and Raman scattering [25, 29], however, its contribution to the CDW distortion is an order of magnitude smaller than the acoustic one, and was not visible in our experiment. We note that a quartic Ginzburg-Landau potential is usually associated with a second order phase transition showing critical behavior and mode softening, but in the case of multiple coupled degrees of freedom it may not exhibit a frequency softening [30]. Thus, a model of this order, as proposed in [26] is still appropriate even in the absence of a soft mode.

Whereas coherent amplitude-mode dynamics yield homogeneous modulations of the CDW sideband, the fine-scale momentum- and time-dependent features in the scattering around the sidebands can contain informa-
FIG. 4. (a) Normalized time-domain data obtained from the line-cut of pixels shown in the inset of (c). Dashed lines are the LP decompositions to the data. (b) the same data with oscillatory components at < 2 and > 20 GHz subtracted. Dashed lines are the portions of the LP decompositions shown in (a) that have frequencies within the range of 2-20 GHz. (c) Dispersion plot of the low-frequency components shown in (b). The damping rates are given by the span of the bars on each point.

ζ (in r.l.u.) parametrizes the line cut in the inset of (c), which is given by $q(ζ) = q_0 + ζ \hat{n}$, where $q_0 = (3.945, 2.055, -0.112)$ r.l.u. and the unit vector $\hat{n} = (-0.916, -0.226, 0.331)$.

In conclusion, we present an ultrafast x-ray study of the low energy lattice modes of (TaSe$_4$)$_2$I in the CDW phase. The high frequency resolution enabled by the time-domain approach allows our measurement to identify a 0.11 THz mode as an acoustic component of the amplitude mode of the CDW. This mode does not soften with excitation fluence even at excitations reaching the high-symmetry phase. This is suggestive of a coupling between this mode and the Ta-tetramerization distortion, which is known to couple to the charge. Finally, low frequency linearly dispersive modes observed around the CDW behave as phase excitations, but they also modulate the Bragg peak and we cannot rule them out as acoustic modes of the crystal. The measured speed of sound for these modes does not match the known values for the I422 phase.

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Appendix: Modeling the excitation of the amplitude mode

The time-dependent potential used to model the excitation of the 0.11 THz amplitude mode shown in Fig. 3 is given by

\[ V(\xi, t) = \frac{1}{2} a[\xi - \xi_0(t)]^2 \]

where \( \xi_0(t) \) is the potential minimum and \( \Theta(t) \) is the Heaviside step function. For \( t < 0 \) the potential minimum is at \( \xi_0 = 0 \), and at \( t = 0 \) it instantaneously shifts by a magnitude given by \( \eta \) and gradually relaxes back with a timescale given by \( \tau \). The equation of motion for \( \xi \) is

\[ \frac{1}{c} \ddot{\xi} = -\xi + \xi_0(t) - \gamma \dot{\xi} \]

where \( \gamma \) is the damping rate, \( c = a/m \), and \( m \) is the mass.

We now motivate the use of a displacive excitation as a model for our experiment. As mentioned in the main text the CDW modulation contains two components, a transverse acoustic distortion and Ta-tetramerization derived from optical phonons. The optical component of the distortion is found to be involved in a predicted structural instability by DFT calculations [17]. We thus begin by considering a simple coupled-mode Ginzburg-Landau potential in which a quartic mode \( \xi_1 \) is bilinearly coupled to a harmonic mode \( \xi_2 \),

\[ V(\xi_1, \xi_2, t) = \frac{1}{2} a_1 r(t) \xi_1^2 + \frac{1}{4} b_1 \xi_1^4 + \frac{1}{2} a_2 \xi_2^2 + g \xi_1 \xi_2, \quad r(t) \equiv \eta \Theta(t) e^{-t/\tau} - 1 \tag{A.3} \]

where we include the effect of the pump through the time-dependent term \( r(t) \). The equilibrium values for this potential are given by

\[ \xi_1^{(0)} = \pm \sqrt{\frac{a_2}{ba_1}}, \quad \xi_2^{(0)} = \frac{g}{a_2} \xi_1^{(0)} \tag{A.4} \]

Solving Eq. A.3 for the equations of motion in the \( \xi_i \), normalizing by the equilibrium values given in Eq. A.4 and taking the weak-coupling limit where \( g \ll a_1, a_2 \) for simplicity, we obtain

\[ \frac{1}{c_1} \ddot{\xi}_1 = -r(t) \dot{\xi}_1 - \dot{\xi}_2^2 - \gamma_1 \dot{\xi}_1 \]

\[ \frac{1}{c_2} \ddot{\xi}_2 = -\xi_2, \dot{\xi}_1 - \gamma_2 \dot{\xi}_2 \tag{A.5} \]

where \( \xi_i \equiv \xi_i/\xi_i^{(0)} \) and \( c_i \equiv a_i/m_i \) where \( m_i \). The mode \( \xi_1 \) directly experiences the effect of the pump through the term \( r(t) \), and the subsequent motion of \( \xi_1 \) then drives the mode \( \xi_2 \). Note the similarity between the equation of motion for \( \xi_2 \) in Eq. A.5 and the displacive equation of motion given in Eq. A.2.

In our experiment the high-frequency optical component of the CDW corresponds to \( \xi_1 \), while the harmonic low-frequency acoustic component corresponds to \( \xi_2 \). Thus \( c_1 \gg c_2 \) and the oscillatory dynamics in \( \xi_1 \) following excitation are too fast for \( \xi_2 \) to register. Instead \( \xi_2 \) will respond to the slower timescale associated with the recovery of the potential, such that the effective driving of the mode \( \xi_2 \) will resemble the expression for \( \xi_0(t) \) in Eq. A.1.

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