Hot collective modes govern the ultrafast quench of charge-density waves

G. Storeck, J. G. Horstmann, T. Diekmann, S. Vogelgesang, G. von Witte, S. V. Yalunin, K. Rossnagel, C. Ropers

14th Physical Institute, Solids and Nanostructures, University of Göttingen, Göttingen 37077, Germany
2Institute of Experimental and Applied Physics, Kiel University, 24098 Kiel, Germany
3Ruprecht Haensel Laboratory, Deutsches Elektronen-Synchrotron DESY, 22607 Hamburg, Germany
4International Center for Advanced Studies of Energy Conversion (ICASEC), University of Göttingen, Göttingen 37077, Germany

Abstract

Spontaneous symmetry-breaking and the excitations of symmetry-broken states are two interdependent aspects of great relevance in condensed matter. This work shows how transient populations of collective modes affect the structural order parameter of a symmetry-broken charge-density wave system. We study the non-equilibrium structural dynamics of the incommensurate and nearly-commensurate charge-density wave (CDW) phases in 1T-TaS$_2$. Employing ultrafast low-energy electron diffraction (ULEED) with 1 ps temporal resolution, we disentangle the ultrafast quench of the CDW-coupled periodic lattice distortion (PLD) from the excitation of fluctuation modes. We identify an unexpectedly long-lived, non-thermal suppression of the structural order parameter, governed by hot populations of CDW-coupled lattice modes. The observations are attributed to an amplitude renormalization induced by low-energy phasons protected by the electronic gap, with topological defects arising only beyond an excitation threshold.

I. INTRODUCTION

The spontaneous breaking of a continuous symmetry is a fundamental concept of physics with broad relevance in such diverse areas as particle physics [1], cosmology [2,3], and condensed matter physics [4,5]. An essential consequence of this symmetry breaking is the emergence of new amplitude and phase excitations of the fields considered, with examples in the Higgs mechanism [6] and massless Nambu-Goldstone bosons [7,8], respectively. Moreover, the degenerate ground state allows for nontrivial topological states, as in the case of magnetic vortices [4].

Electron-lattice interaction is an important source of symmetry breaking in solids, most prominently in superconductivity and the formation of charge-density wave (CDW) phases [9–12]. Specifically, CDWs constitute a periodic modulation of the charge density by electron-hole pairing [12], coupled to a periodic lattice distortion (PLD) [13–15] and an electronic gap [16–19]. The emergence, correlations and fluctuations of symmetry-broken CDW states can be revealed in the time domain by ultrafast measurement techniques. In this way, quenches of the electronic gap coupled to coherent amplitude oscillations [20–28], light-induced PLD dynamics [29–32] and phase transitions have been probed [20,33,34].
particular, ultrafast structural probes trace changes of structural symmetry [35,36] and long-range ordering following a phase transformation [37,38].

However, while the initial quench and coherent amplitude dynamics of CDW systems following short-pulsed excitation are rather well-characterized, the subsequent path to thermal equilibrium, including the roles of different collective modes in re-establishing a thermal CDW state, are far less understood. This is exemplified in the case of phasons, phase fluctuations of the order parameter [22]. Despite early theoretical and experimental work [13,14,39,40], a unifying picture of phason dynamics has not been reached, and recent assignments of their contribution in diffraction experiments range from largely negligible [41] to dominant [42]. These questions call for a sensitive experimental probe that distinguishes a time-dependent quench of the lattice distortion from the transient excitation of fluctuation modes.

Here, we demonstrate that the transient population of collective CDW modes has a lasting impact on the structural order parameter. We experimentally trace the non-equilibrium structural dynamics of charge-density wave phases at the surface of 1T-TaS$_2$. Employing ultrafast low-energy electron diffraction, a recently developed surface-sensitive structural probe [37,43–45], we isolate the dynamics of an optically-induced amplitude quench from a multi-stage excitation of phonons. Following a rapid partial recovery, we observe a surprisingly long-lived non-thermal amplitude suppression that equilibrates only after approximately 60 ps. Energy transfer to acoustic phonons is required to re-establish a thermal value of the PLD amplitude, which suggests a persistent amplitude renormalization by long-lived, low-energy CDW-coupled lattice modes, namely phasons. At high excitation fluence, a spot broadening associated with topological defects is observed, suggesting a mechanism based on threshold phase fluctuations.

II. MATERIAL AND EXPERIMENTAL APPROACH

In this work, we study one of the most prominent CDW systems, 1T-TaS$_2$, which is part of the class of transition metal dichalcogenides. The atomic structure of this material consists of weakly interacting S-Ta-S trilayers [46–48], in which the tantalum atoms are octahedrally coordinated between the sulfur atoms (Fig. 1a). This compound has attracted much attention for its various ordered CDW equilibrium states [12,46,47,49,50], excitations [21,24–26,29,51] (Fig. 1c), and correlation effects [52–54], serving as a model system to study, for example, Peierls- versus Mott-type metal-insulator transitions [23,55], pressure-induced superconductivity in coexistence with CDWs [56], transitions to metastable “hidden” CDW states [34,57], the emergence of complex orbital textures [58], or quantum spin liquid behavior [59].

The material exhibits multiple temperature-dependent phases (Fig. 1c) with characteristic lattice deformations coupled to electronic structure changes [46,55,60]. Starting from a metallic phase with an undistorted trigonal structure (Fig. 1a) above 543 K, the system undergoes a sequence of CDW transitions, forming a commensurate (C) (Mott-insulating) state below 187 K. At intermediate temperatures, two incommensurate phases are found, namely the so-called “nearly-commensurate” (NC) phase (187-353 K), exhibiting commensurate patches separated by discommensurations [47,61–63] and a homogeneous, fully incommensurate (IC) structure (Fig. 1b) between 353 K and 543 K. Ultrafast transitions
between and manipulation of these phases, as well as their collective modes (Fig. 1c) have been observed in various diffraction and spectroscopy studies [20,21,23–26,29,31,35,37,51,64–69].

In our experiments, we employ pulses of electrons at low energies, typically in the range of 40-150 eV, to probe the structural evolution of the NC and IC states in backscattering diffraction. Ultrafast LEED [37,43–45] allows us to trace the changes of the diffraction pattern in the time domain, following intense fs-laser illumination (red pulse in Fig. 1e). In this optical-pump/electron-probe scheme, excitation and relaxation processes are sampled by varying the time delay $t$ between the optical pump pulse (red) and the photoemission pulse (blue) generating the electron probe (green). Minimizing electron pulse broadening by short propagation lengths, a miniaturized electron gun (Fig. 1g) [44] allows for a temporal resolution of 1 ps (Fig. 1f). Further experimental details are provided in Appendix A.

**III. RESULTS AND DISCUSSION**

To facilitate the discussion, we focus the presentation on the response of the IC phase, which has not been studied by ultrafast diffraction, and provide a comprehensive data set of related observations for the NC phase in Appendix B. In the IC phase, the PLD leads to characteristic arrangements of satellite peaks [40,70] around the main lattice diffraction spots, seen in the ULEED pattern displayed in Fig. 2c. As the IC state wave vectors are collinear to the lattice vectors, the satellites are located on the lines connecting the main reflexes, and due to the harmonic (and weak) structural modulation [61,62], only first-order satellites are observed [40].
We study the excitation and relaxation of the IC and NC phases, without driving the system across a phase transition [31,35,37,38,61,71]. The dynamics of this incommensurate Peierls system can be discussed based on a simplified picture of three coupled subsystems, namely, the electronic system exhibiting a gapped band structure (Fig. 2a, top), the collective amplitude and phase excitations around the symmetry-broken CDW ground state (center) [12], and the ordinary lattice modes far from the CDW wavevector in reciprocal space, i.e., regular phonons (bottom).

Fig. 2. Dynamics and excitations in CDW systems influencing diffraction. (a) Electron and lattice subsystems (right) governing CDW dynamics. Gapped band structure (top, left), symmetry broken CDW state with phase and amplitude excitations (middle), and non-CDW phonons (bottom). (b) Changes in average amplitude and all lattice excitations (CDW and non-CDW) lead to a redistribution of intensity in the electron diffraction pattern. (c) Diffraction pattern of the IC phase of \(1T\)-TaS\(_2\) showing main lattice reflexes and first-order PLD-induced satellites (integration time: 90 s, electron energy: 100 eV). (d) Time-dependent measurement of reflexes (blue and red circles in (c)) and diffuse background (fluence \(F = 2.5\) mJ/cm\(^2\)). The main lattice signal is averaged over (10) and (-1 1) spots (blue), the satellite signal over several reflexes. Curves are normalized to the signal at negative times.

It is widely established that electronic excitation by an ultrashort laser pulse induces a carrier population above the band gap, which results in a quench of the CDW/PLD amplitude that recovers upon carrier cooling by electron-phonon scattering [24,29,51]. The corresponding sequence of energy redistribution and relaxation processes involving the three subsystems causes characteristic changes to the diffraction pattern. In a simplified approximation ignoring dynamical diffraction, the intensities of the satellite spots \(I_{\text{sat}}\), the main peaks \(I_{\text{main}}\), and the diffuse background \(I_B\) are expected to scale as [40,70,72]:

\[
I_{\text{sat}} \sim \eta^2 \exp (-2W), \quad (1)
\]

\[
I_{\text{main}} \sim (1 - c \cdot Q^2 \eta^2) \cdot \exp (-2W), \quad (2)
\]

\[
I_B \sim (1 - \exp (-2W)), \quad (3)
\]

Here, \(\eta\) is the mean PLD amplitude, \(Q\) is the CDW wavevector, and \(\exp (-2W)\) is a Debye-Waller suppression from populated phonons [72,73]. As we will see, different main reflexes are sensitive to the PLD to a varying degree, which requires the introduction of the factor \(c\). Consistent with the expressions above, a light-induced amplitude quench leads to a
redistribution of intensity from the satellites to the main reflexes [29,30,41], and inelastic scattering by generated phonons transfers intensity from the reflexes to a diffuse background (Fig. 2b) [73–75]. Dislocation-type topological defects may broaden the superlattice peaks and also reduce the PLD in the dislocation core [37,67].

In our ULEED experiments, such changes are directly observed in the exemplary data displayed in Fig. 2d: Main lattice peaks (blue) exhibit a transient intensity increase after the pump pulse, before experiencing an initially rapid and then slowed suppression to a minimum at \( t = 60 \text{ ps} \). The satellite peaks (red), on the other hand, are first suppressed, before they approach a similar trend as the main peaks beyond approximately 10 ps. Both the satellite and main peak intensities are significantly reduced by phonon populations [73]. These are evident in the diffuse background (gray), which mirrors the suppression of the reflexes, with a step-like increase in the first ps and a slower rise to a maximum at the delay of 60 ps. The initial step can be interpreted as the excitation of a broad population of optical and acoustic phonons on the timescale of electron-phonon energy relaxation (<1 ps) [26], while the slower timescale corresponds to phonon-phonon equilibration [76] and the population of low-energy acoustic modes. LEED intensities are rather sensitive to the large amplitudes of low-frequency modes, particularly those with out-of-plane polarization. Specifically, phonon modes with out-of-plane displacements \( \vec{u}_{ph} \) have a more pronounced Debye-Waller factor \( W \sim \langle (\hat{s} \cdot \vec{u}_{ph})^2 \rangle \) [72,73] due to the backscattering geometry with a primarily out-of-plane scattering vector of the electron \( \hat{s} \). In addition, these modes exhibit comparatively slow phase velocities, as is typical for layered van-der-Waals materials [77]. Thus, the prominent main lattice suppression evolving over tens of picoseconds primarily stems from the increasing population of low-frequency acoustic modes modulating the layer distance.

**FIG. 3.** Amplitude dynamics of PLD obtained from main lattice reflexes. (a) Time-dependent intensity of visible main lattice reflexes and integrated background intensity, for a fluence of \( F=3.8 \text{ mJ/cm}^2 \). Two classes of inequivalent spot groups are found, featuring a strong (dark blue) and weak (light blue) sensitivity to the amplitude quench. (b) Sketch of IC diffraction pattern, and parts of the difference diffraction image (\( I_{t=1\text{ps}} - I_{t<0} \)) around the (10) and (01) main reflexes (insets). (c) Extracted PLD amplitude quench and relaxation (see also Appendix C) for three fluences, showing a rapid and a slower relaxation component. (Time constants from a biexponential fit (black line) to the highest fluence data: 2.2 ps and 66 ps).

These strong Debye-Waller type peak suppressions complicate an analysis of the temporal evolution of the amplitude quench. In order to disentangle the dynamics of the structural order parameter \( \eta \) from the phonon population, we exploit the different sensitivities of inequivalent
main lattice reflections to the PLD. Whereas all five visible main peaks show a suppression opposite to the increase in diffuse background (Fig. 3a), the transient amplitude signal is only present in the (1,0) and (-1,1) peaks, and not in the (01), (-10), and (1,-1) peaks (see also difference maps in Fig. 3b) [78]. This experimental observation does not directly follow from simple symmetry considerations, but it is also not unexpected, due to the lack of a mirror symmetry of both the unit cell and the resulting backscattering diffraction pattern. The different sensitivities of the peaks to the PLD are a robust feature which does not depend on the specific details of the lattice distortion. In particular, we found the same trend in experiments on the NC phase (see Appendix B). Moreover, the sensitivity to the PLD is strongly energy-dependent, and we have found that the dependency of the main peak intensities on $\eta$ is generally much weaker at an energy of 80 eV (see additional data in Appendix D).

We now derive a phonon-corrected amplitude signal by a scaled ratio of the intensities of the two groups of peaks (see Appendix C for details), displayed in Fig. 3c for three pump fluences. In each case, the amplitude exhibits a rapid initial quench (within our temporal resolution), and a recovery with an exponential time constant of about 2 ps. The re-establishment of the amplitude is, however, incomplete, slowing down considerably beyond 4 ps, and lasting well into the range of tens to hundreds of picoseconds.

The question now arises whether this long-lived partial suppression is merely a result of the system being thermalized at a higher temperature and with reduced equilibrium amplitude [29], or, alternatively, if there are non-equilibrium dynamics inhibiting the recovery of the order parameter. As shown in the following, we have evidence for a sustained non-thermal suppression of the order parameter. In Fig. 4, we consider in more detail the path to thermal equilibrium. We first plot the logarithm of the satellite and main peak intensities (normalized to the signal at $t < 0$), divided by the fluence (Fig. 4a). For all three fluences, the traces of the main lattice peaks collapse to a single curve (blue), highlighting the importance of the Debye-Waller suppression for these peaks. In contrast, the satellite peaks show a strong, non-exponential fluence dependency in the suppression and recovery which warrants further analysis.

An instructive depiction is obtained by plotting the main and satellite intensities against each other, resulting in cyclic trajectories in a two-dimensional plane (Fig. 4b), traced out over time in a clockwise fashion. At long delays (beyond 100 ps), the curves for all fluences follow a universal path (dashed line) representing a thermalized system at elevated temperatures, cooling down. Different trajectories reach the same combination of intensities at different times. For instance, the high-fluence trajectory exhibits the same combination of intensity suppressions at 1500 ps as the intermediate fluence at a somewhat earlier time of 290 ps (black circle in Fig. 4b). On this line, the satellite peak suppression is composed of a Debye-Waller factor as well as a reduction of the equilibrium amplitude, and the further progression (i.e. cooling) is only governed by thermal diffusion to the bulk.
FIG. 4. Path to equilibrium. (a) Logarithm of normalized main lattice and satellite peak intensities, divided by fluence, versus time delay. While the main peak intensities (blue) collapse to a single curve due to the exponential (in fluence) Debye-Waller-type suppression, the satellite intensities (red) show a strong fluence-dependent behavior for early times, before converging for long time delays. (b) Curves from (a) (not normalized to fluence) plotted against each other, leading to cyclic trajectories in a 2D plane with varying size. Note that all curves reach a common equilibrium line after approximately 60 ps. The gray color scale highlights certain time intervals (dark gray: 0-1 ps, intermed. gray: 1-60 ps, light gray: 60-1500 ps). The same combination of intensity suppressions is found for different fluences at different times (black circle corresponds to 1500 ps/290 ps at high/intermediate fluence). (c) Ratio of time-integrated frames exhibit prominent pedestals around diffraction peaks, pointing to an enhanced acoustic phonon population on the equilibrium line. Late frames (dark magenta in inset, $t = 790 \ldots 1500$ ps) divided by early frames (light magenta in inset, $t = 4.5 \ldots 10$ ps).
All points displaced from the dashed line represent deviations from a thermal state, with the distance being a very sensitive measure of the structural non-equilibrium. For example, within the first picosecond after the excitation (dark segments of the curves), the rapid quench of the order parameter causes a reduction of satellite intensity and a moderate enhancement of the main lattice signal, with a fluence-dependent maximum displacement from thermal equilibrium (corresponding curves for the main peaks insensitive to the amplitude are found in Appendix E). The recovery to the thermal state now proceeds through various stages and in a fluence-dependent manner. After about 4 ps (see marks), the fast component of the amplitude recovery is completed (cf. Fig. 3c, compare also Ref. [79]). However, the system remains far from the equilibrium state, i.e., exhibits a lower-than-thermal satellite intensity. Interestingly, for all curves, a surprisingly long time of approximately 60 ps is required to reach the thermal state. This depiction directly shows that the persistent amplitude suppression discussed in Fig. 3c is in fact not of a thermal nature, and that we have a pronounced deviation from equilibrium between the degrees of freedom affecting the diffraction intensities.

To identify the origin of this long-lived amplitude suppression, we first note that the time at which the system reaches a thermal amplitude nearly coincides with the strongest suppression of the main lattice peaks. As this time also corresponds to the maximum intensity of the diffuse background (cf. Figs. 2d, 3a), the full equilibration of lattice fluctuations appears to be critical in controlling the structural order parameter. In particular, this lattice equilibration induces a significant increase of diffuse background intensity around main lattice peaks (difference image in Fig. 4c), directly pointing to the excitation of low-energy acoustic modes near the center of the Brillouin zone.

Taken together, these observations indicate the sequence of relaxation processes illustrated in Fig. 5c, which can be related to the intensity curves (Fig. 5a) and the cyclic trajectories introduced above (simplified sketch in Fig. 5b). Within the first picosecond, optical excitation of the electronic system leads to a CDW amplitude quench and a strong deformation of the potential energy landscape, which triggers cooperative motion of the lattice towards its unmodulated state, including the excitation of coherent amplitude modes (stage 1) [26,51]. Facilitated by the generation of high-energy lattice modes, the electron system cools down within few ps (stage 2), and as a result, the amplitude partially recovers. The remaining PLD suppression at the end of this stage strongly indicates a substantial population of CDW lattice excitations, i.e. amplitudons, phasons, and possible topological defects.
Generally, cooling of the carrier temperature below the energy scale of the electronic gap will affect inelastic scattering near and between gap regions. Assuming, for simplicity, a constant density of states, the scattering rate $\gamma_\varepsilon$ of electrons creating lattice excitations at energy $\varepsilon$ by transitions in the electronic gap regions are given by the integration over the density of occupied initial times unoccupied final states,

$$\gamma_\varepsilon \sim \int_{\Delta/2}^{\infty} dE \left[ f(E, T_e) \cdot (1 - f(E - \varepsilon, T_e)) \right],$$

where $f(E, T_e)$ is the electronic Fermi-Dirac distribution as a function of energy $E$ and temperature $T_e$, and $\Delta$ is the gap width. For low-energy excitations with $\varepsilon \ll \Delta$, the rates are strongly suppressed with temperature as $\gamma_\varepsilon \sim \left[ e^{\Delta/(2k_B T_e)} + 1 \right]^{-1}$, effectively decoupling electronic and CDW-associated lattice degrees of freedom. Generating phasons requires momenta around the CDW wavevector $Q$, and therefore, the gap contributes to enhanced phason lifetimes. As a result, upon further cooling of $T_e$ by regular lattice modes, the CDW modes may even exceed the electronic system in effective temperature, facilitating the persistent amplitude suppression during stage (3). Full lattice thermalization and the excitation of zone-center acoustic modes is then only achieved after 60 ps, from which point on the equilibrated system cools down (stage 4).
We now discuss the possible roles of different CDW excitations in the long-lived amplitude suppression, namely amplitudons, phasons, and topological defects. Figure 6 illustrates these mechanisms based on the familiar free-energy surface of a Mexican hat potential. Spatiotemporal variations of the amplitude and phase affect the observable value of $\eta$. Specifically, amplitudons represent amplitude oscillations $\Delta A$ around an equilibrium amplitude $A_0$, leading to an observed average value of $\langle A_0 + \Delta A \rangle$. By an anharmonicity of the potential [69] (top in Fig. 6a), these oscillations become asymmetric, and a high population of amplitudons can in principle reduce the value of $\eta$. Phase oscillations, on the other hand, proposed to be generated by, e.g., parametric decay of amplitudons [22], also reduce the time-averaged order parameter, resulting in a “renormalized” amplitude by a factor of $\langle \cos(\varphi) \rangle$, where the expectation value is over the phase deviations $\varphi$ from the unperturbed CDW [39]. Together, both contributions lead to $\eta = \langle A_0 + \Delta A \rangle \langle \cos \varphi \rangle$. Finally, topological defects should also be considered as a possible cause for the long-lived order parameter suppression, as they have been observed as a consequence of phase transitions, e.g. in $1T$-TaS$_2$ [37] or LaTe$_3$ [67,80].

From our observations, the long lifetime of the suppression and the relaxation concurring with the thermalization of acoustic phonons points to lower-energy excitations instead of anharmonic amplitudons, and suggests that phason-induced renormalization is the dominant...
factor. In this picture, the observed suppressions of $\eta$ at $t = 4$ ps (subsequent to the initial recovery) correspond to standard deviations of the phase fluctuation of $\sqrt{\langle \varphi^2 \rangle} = 0.35$, $0.53$ and $0.74$ for the fluences of $1.3$, $2.5$ and $3.8$ mJ/cm$^2$. At higher excitation densities, an increase of phase distortions presents a possible mechanism to create defects. This process is expected to show a fluence-dependent threshold behavior. In our measurements, only at high fluence, we observe significant spot broadening of the satellites (Fig. 6c), determined by the change of the reflex width. The fluence-dependent spot broadening points to a strongly nonlinear behavior in the density of topological defects, as expected for critical phase fluctuations and ruling out a defect density depending linearly on fluence (see Appendix F). As the long-lived amplitude suppression is present even for the lowest fluences, we conclude that topological defects are only relevant at the highest fluence, while the long-lived suppression is predominantly phason-induced.

III. CONCLUSIONS

The impact of fluctuations on symmetry breaking transitions has long been considered, for example in the Peierls instability [81–84]. Providing a time-domain view of the structural relaxation pathways, the present measurements highlight that non-equilibrium populations of collective modes directly affect the transient expectation value of the order parameter. Relevant further questions pertain to the phason generation mechanism, as well as that of topological defects, and their coupling to regular lattice modes. Also the link between fluctuation modes and the creation and relaxation of metastable states [34,57], as well as the role of partial and full commensurability in the different CDW states warrant further investigation.

Considering methodical aspects, this work represents the first comprehensive study employing ultrafast low-energy electron diffraction with a temporal resolution of 1 ps. Future investigation using ULEED will enable a quantitative analysis of the three-dimensional structural evolution based on time- and energy-dependent diffraction. Moreover, the method is applicable to a wide variety of other surface systems and low-dimensional structures, harnessing its strengths of high momentum resolution, efficient scattering and enhanced sensitivity to lattice fluctuations.

ACKNOWLEDGMENTS

This work was funded by the European Research Council (ERC Starting Grant `ULEED', ID: 639119) and the Deutsche Forschungsgemeinschaft (SFB-1073, project A05). We gratefully acknowledge insightful discussions with H. Schwoerer and B. Siwick. Furthermore, we thank K. Hanff for help with sample preparation.
Here, we briefly describe our experimental ULEED apparatus (Fig. 7). The time-resolved measurements are conducted in an ultra-high vacuum chamber (base pressure \( p = 5 \cdot 10^{-11} \text{ mbar} \)) into which samples are transferred via a load-lock system and cleaved in-situ. Inside the chamber, the electron source (microgun [44]) and a microchannel plate detector are mounted. A cooled CMOS camera records the detected electron diffraction patterns from outside the UHV chamber.

A femtosecond laser system (Amplifier, NOPA and OPA) generates three femtosecond laser beams of different center wavelength. An ultraviolet beam (center wavelength of 400 nm) is focused on a nanometric tungsten needle that is embedded inside the microgun [44] generating ultrashort electron pulses via two-photon photoemission. An electrostatic lens assembly controls the collimation of the electron beam having an energy range of 40-100 eV. With a gun front diameter of 80 µm and a working distance of around 150 µm, we achieve a temporal resolution of 1 ps and an electron beam diameter of approximately 10 µm at the sample. An infrared beam (1030 nm center wavelength) optically excites the sample at specific times controlled by an automated linear delay stage. In order to heat the 1T-TaS\(_2\) sample and stabilize it in the IC phase slightly above \( T \approx 353 \text{ K} \), a third beam (800 nm) is aligned collinearly with the infrared beam. This pulse is delayed by about 3 ns with respect to the electron and 1030 nm pulses (i.e., it arrives 10 µs before the next pulses) and thus leads to an average increase in sample temperature.

**APPENDIX B: DATA FOR NEARLY-COMMENSURATE (NC) PHASE**

Figure 8 displays the complete analysis discussed above applied to the nearly-commensurate phase. Similar features are found in the pump-probe curves for the main and satellite diffraction peaks as well as the background (Fig. 8c), the long-lived amplitude suppression (Fig. 8d-e) and the relaxation cycles (Fig. 8f). In particular, the two-stage amplitude relaxation process (first stage up to \( \sim 4 \) ps, second stage up to \( \sim 60 \) ps) is very pronounced at all fluences.
FIG. 8. Measurements in the NC phase for an electron energy of 100 eV. (a) Top view of nearly-commensurate (NC) CDW phase illustrating charge density (green), distorted lattice (black dots, displacements exaggerated) and superstructure unit cell (orange). (b) Diffraction pattern of the NC phase of 1T-TaS$_2$ showing main lattice reflexes and several orders of PLD-induced satellites (integration time: 90 s). (c) Time-dependent measurement of reflexes (blue and red circles in (b)) and diffuse background (for three fluences). The main lattice signal is averaged over the (10) and (-1 1) spots (blue), the satellite signal over several reflexes. Curves are normalized to signal at negative times. (d) Time-dependent intensity of visible main lattice reflexes and integrated background intensity, for a fluence of $F = 2.0 \text{ mJ/cm}^2$. Two classes of inequivalent spot groups are found, featuring a strong (dark blue) and weak (light blue) sensitivity to the amplitude quench. (e) Extracted PLD amplitude quench and relaxation for three fluences, showing a rapid and a slower relaxation component. (Time constants from a biexponential fit (black line) to the highest fluence data: 1.3 ps and 88.5 ps). (f) Main lattice peak intensities vs. satellite peak intensities, leading to cyclic trajectories in a 2D plane with varying size. Following a two-stage relaxation, all curves reach a common equilibrium line after approximately 60 ps. The gray color scale highlights certain time intervals (dark gray: 0-1 ps, intermed. gray: 1-60 ps, light gray: 60-1500 ps).

APPENDIX C: DATA ANALYSIS

In order to obtain time curves (Fig. 2d) from the stacks of diffraction patterns, we process the data in a sequence of operations. First, a binary circular mask is laid on top of each individual reflex (Fig. 2c, blue and red circle, diameter $\Delta k_{\text{main}} = 0.6 \text{ Å}^{-1}$ and $\Delta k_{\text{sat}} = 0.36 \text{ Å}^{-1}$ respectively) for each time delay. Second, we fit 2D Cauchy distributions (background: slope and constant offset) to the satellite reflexes and 2D pseudo Voigt profiles (background: constant offset) to the main lattice reflexes, to determine a background profile and subtract it from each spot.
\[ C(x, y) = \frac{A}{(2\pi \sigma_1 \sigma_2)} \cdot (1 + (x/\sigma_1)^2 + (y/\sigma_2)^2)^{-3/2} + \text{plane}(x, y) + \text{const.} \]  
\[ PV(x, y) = \frac{A}{(2\pi \sigma_1 \sigma_2)} \cdot (1 + (x/\sigma_1)^2 + (y/\sigma_2)^2)^{-3/2} + B \cdot \text{Gaussian}(x, y, \sigma_3) + \text{const.} \]  

Here, the x and y axes correspond to the azimuthal and radial directions (with respect to the main peak) for a given spot. Third, from the background-corrected segment, the average and the maximum intensity (average over brightest 4% within a mask) are determined for each reflex within the mask. The remaining intensity outside the circular masks forms the integrated background. For improved signal-to-noise ratio, several spot curves are averaged. Furthermore, from the 2D fit functions, we obtain the azimuthal (\( \sigma_1 \)) and radial (\( \sigma_2 \)) widths for each reflex (Fig. 6c).

Next, we describe the separation of the amplitude-quench-related intensity changes from Debye-Waller type peak suppression (Fig. 3a-c). To this end, we need to correct the intensities for the different effective Debye-Waller coefficients of different reflexes. For each fluence, light blue (less sensitive to amplitude) and dark blue curves (more sensitive to amplitude) (in Fig. 3a) are averaged (\( I_{\text{non-amp}} \) and \( I_{\text{amp}} \)). For the lowest fluence (\( F_1 = 1.3 \text{ mJ/cm}^2 \)), for which a negligible amplitude change is expected at very long delays, we logarithmically scale \( I_{\text{non-amp}, F_1} \) such that it coincides with \( I_{\text{amp}, F_1} \) for delay times beyond 1 ns, determining a constant \( C = 0.81 \):

\[ I_{\text{ratio}, F} = \frac{I_{\text{amp}, F}}{I^{F/F_1 \cdot C}_{\text{non-amp}, F_1}} \]  

Last, in order to relate the intensity to a relative change in PLD amplitude \( \eta/\eta_{t<0} \), we assume that the intensity \( I_{\text{ratio}, F} \) and satellite intensity \( I_{\text{sat}, F} \) scale quadratically as given in Eq. (1), with the maximum suppression given by the lowest value of the (Debye-Waller-corrected) satellite intensity.

The fit function in Fig. 3c is based on a step-like decrease followed by two exponential relaxations

\[ S(t) = 1 - \theta(t - t_0) \cdot (-A_1 + A_2 \cdot (1 - e^{-(t-t_0)/\tau_1}) + A_3 \cdot (1 - e^{-(t-t_0)/\tau_2}) \)  

where \( \theta \) is the Heaviside-function, \( t_0 \) time zero, \( A_1, A_2 \) and \( A_3 \) are the amplitudes and \( \tau_1 \) and \( \tau_2 \) time constants. The complete fit function is the convolution of \( S(t) \) with a Gaussian (FWHM of 1 ps) corresponding to the temporal resolution in our experiment.

**APPENDIX D: DATA AT 80eV ELECTRON ENERGY**

Figure 9 shows additional data recorded in the IC phase at 80 eV energy. The main lattice peaks show a much weaker dependency on the PLD amplitude (Fig. 9b-d).
FIG. 9. Measurements in the IC phase for an electron energy of 80 eV. (a) Diffraction pattern of the IC phase of 1T-TaS$_2$ showing main lattice reflexes and first-order PLD-induced satellites (integration time: 90 s). (b) Time-dependent measurement of reflexes (blue and red circles in (a)) and diffuse background (for three fluences). The main lattice signal is averaged over the (10) and (-1 1) spots (blue), the satellite signal over several reflexes. Curves are normalized to signal at negative times. (c) Time-dependent intensity of visible main lattice reflexes and integrated background intensity, for a fluence of $F = 3.8$ mJ/cm$^2$. Two classes of inequivalent spot groups are found, but none shows a strong dependence on the amplitude quench. (d) Main lattice intensity vs. satellite peak intensity, leading to cyclic trajectories in a 2D plane with varying size. Note that all curves reach a common equilibrium line after approximately 60 ps. The gray color scale highlights certain time intervals (dark gray: 0-1 ps, intermedi. gray: 1-60 ps, light gray: 60-1500 ps).

APPENDIX E: RELAXATION CYCLES FOR MAIN PEAKS (-1 1), (0 1) and (1 -1)

Figure 10 shows the relaxation cycles in the IC phase as in Fig. 4b, using the intensities of the main lattice peaks (-1 1), (0 1) and (1 -1) without sensitivity to the PLD amplitude.
FIG. 10. Main lattice peaks without amplitude feature vs. satellite peak intensities, leading to cyclic trajectories in a 2D plane with varying size. All curves reach a common equilibrium line after approximately 60 ps. The gray color scale highlights certain time intervals (dark gray: 0-1 ps, intermed. gray: 1-60 ps, light gray: 60-1500 ps).

APPENDIX F: IMPACT OF TOPOLOGICAL DEFECTS ON PEAK WIDTH

Here, we argue that our data rules out a linear scaling of topological defect density with fluence and is only consistent with a nonlinear or threshold behavior. Assuming a linear relation of the defect density with the fluence \( n \sim F \) and a correlation length \( L \sim 1/\sqrt{n} \) [37], the defect-induced broadening should scale as \( \sigma_{\text{top}} \sim 1/L \sim \sqrt{n} \sim \sqrt{F} \). A doubling of the normalized peak widths \( \sigma_{\text{tot}} = \sqrt{\sigma_0^2 + \sigma_{\text{top}}^2} \) with respect to the instrument resolution \( \sigma_0 \) for the highest fluence would then imply considerably higher broadening values for lower fluences (\( \sigma_{\text{tot,2.5}} = 1.7 \) and \( \sigma_{\text{tot,1.3}} = 1.4 \)) than observed in our measurement (Fig. 6c). Experimentally, we find maximum broadening values at \( t \approx 1 \) ps of \( \sigma_{\text{tot,3.8}} \approx 2 \), \( \sigma_{\text{tot,2.5}} \approx 1.3 \) and \( \sigma_{\text{tot,1.3}} \approx 1 \) for the highest, intermediate and lowest fluence, respectively (see Fig. 6c).

Thus, we infer that the density of topological defects does not scale linearly with fluence.
References

[1] S. Weinberg, *The Quantum Theory of Fields* (Cambridge University Press, 1995).
[2] T. W. B. Kibble, *Topology of Cosmic Domains and Strings*, J. Phys. A: Math. Gen. 9, 1387 (1976).
[3] W. H. Zurek, *Cosmological Experiments in Superfluid Helium?*, Nature 317, 505 (1985).
[4] A. Auerbach, *Interacting Electrons and Quantum Magnetism* (Springer Science & Business Media, 2012).
[5] A. Altland and B. D. Simons, *Condensed Matter Field Theory* (Cambridge University Press, 2010).
[6] P. W. Higgs, *Broken Symmetries and the Masses of Gauge Bosons*, Phys. Rev. Lett. 13, 508 (1964).
[7] Y. Nambu, *Quasi-Particles and Gauge Invariance in the Theory of Superconductivity*, Phys. Rev. 117, 648 (1960).
[8] J. Goldstone, *Field Theories with « Superconductor » Solutions*, Nuovo Cim 19, 154 (1961).
[9] H. Fröhlich, *On the Theory of Superconductivity: The One-Dimensional Case*, Proceedings of the Royal Society of London. Series A. Mathematical and Physical Sciences 223, 296 (1954).
[10] R. E. Peierls, *Quantum Theory of Solids* (Clarendon Press, 1955).
[11] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, *Theory of Superconductivity*, Phys. Rev. 108, 1175 (1957).
[12] G. Grüner, *Density Waves in Solids* (Addison-Wesley Pub. Co., Advanced Book Program, Reading, Mass, 1994).
[13] L. D. Chapman and R. Colella, *Experimental Evidence from X-Ray Diffraction for Phase Excitations in Solids*, Physical Review Letters 52, 652 (1984).
[14] W. Minor, L. D. Chapman, S. N. Ehrlich, and R. Colella, *Phason Velocities in TaS 2 by X-Ray Diffuse Scattering*, Physical Review B 39, 1360 (1989).
[15] J. P. Pouget, B. Hennion, C. Escribe-Filippini, and M. Sato, *Neutron-Scattering Investigations of the Kohn Anomaly and of the Phase and Amplitude Charge-Density-Wave Excitations of the Blue Bronze K 0.3 MoO 3*, Phys. Rev. B 43, 8421 (1991).
[16] G.-H. Gweon, J. D. Denlinger, J. W. Allen, E. DiMasi, M. C. Aronson, B. Foran, and S. Lee, *Direct Observation of Complete Fermi Surface, Imperfect Nesting, and Gap Anisotropy in the High-Temperature Incommensurate Charge-Density-Wave Compound SmTe 3*, Phys. Rev. Lett. 81, 886 (1998).
[17] S. Uchida and S. Sugai, *Infrared and Raman Studies on Commensurate CDW States in Transition Metal Dichalcogenides*, Physica B+C 105, 393 (1981).
[18] T. Hirata and F. S. Ohuchi, *Temperature Dependence of the Raman Spectra of 1T-TaS2*, Solid State Communications 117, 361 (2001).
[19] J. R. Duffey, R. D. Kirby, and R. V. Coleman, *Raman Scattering from 1T-TaS2*, Solid State Communications 20, 617 (1976).
[20] S. Hellmann, M. Beye, C. Sohrt, T. Rohwer, F. Sorgenfrei, H. Redlin, M. Kallâne, M. Marczynski-Bühlow, F. Hennies, M. Bauer, et al., *Ultrafast Melting of a Charge-Density Wave in the Mott Insulator 1 T − TaS 2*, Physical Review Letters 105, (2010).
[21] L. Perfetti, P. A. Loukakos, M. Lisowski, U. Bovensiepen, M. Wolf, H. Berger, S. Biermann, and A. Georges, *Femtosecond Dynamics of Electronic States in the Mott Insulator 1 T−TaS2 by Time Resolved Photoelectron Spectroscopy*, New J. Phys. 10, 053019 (2008).
[22] H. Y. Liu, I. Gierz, J. C. Petersen, S. Kaiser, A. Simoncig, A. L. Cavalleri, C. Cacho, I. C. E. Turcu, E. Springate, F. Frassetto, et al., *Possible Observation of Parametrically Amplified Coherent Phasons in K 0.3 MoO 3 Using Time-Resolved Extreme-Ultraviolet Angle-Resolved Photoemission Spectroscopy*, Physical Review B 88, (2013).
[23] J. C. Petersen, S. Kaiser, N. Dean, A. Simoncig, H. Y. Liu, A. L. Cavalleri, C. Cacho, I. C. E. Turcu, E. Springate, F. Frassetto, et al., *Clocking the Melting Transition of Charge
and Lattice Order in 1T-TaS2 with Ultrafast Extreme-Ultraviolet Angle-Resolved Photoemission Spectroscopy, Phys. Rev. Lett. 107, 177402 (2011).

[24] L. Perfetti, P. A. Loukakos, M. Lisowski, U. Bovensiepen, H. Berger, S. Biermann, P. S. Cornaglia, A. Georges, and M. Wolf, Time Evolution of the Electronic Structure of 1 T – TaS 2 through the Insulator-Metal Transition, Physical Review Letters 97, (2006).

[25] C. Sohrt, A. Stange, M. Bauer, and K. Rossnagel, How Fast Can a Peierls–Mott Insulator Be Melted?, Faraday Discuss. 171, 243 (2014).

[26] J. Demsar, L. Forró, H. Berger, and D. Mihailovic, Femtosecond Snapshots of Gap-Forming Charge-Density-Wave Correlations in Quasi-Two-Dimensional Dichalcogenides 1 T − TaS 2 and 2 H − TaSe 2, Physical Review B 66, (2002).

[27] R. Yusupov, T. Mertelj, V. V. Kabanov, S. Brazovskii, P. Kusar, J.-H. Chu, I. R. Fisher, and D. Mihailovic, Coherent Dynamics of Macroscopic Electronic Order through a Symmetry Breaking Transition, Nature Physics 6, 681 (2010).

[28] A. Tomeljak, H. Schäfer, D. Städter, M. Beyer, K. Biljakovic, and J. Demsar, Dynamics of Photoinduced Charge-Density-Wave to Metal Phase Transition in K 0.3 MoO 3, Physical Review Letters 102, (2009).

[29] M. Eichberger, H. Schäfer, M. Krumova, M. Beyer, J. Demsar, H. Berger, G. Moriena, G. Sciaini, and R. J. D. Miller, Snapshots of Cooperative Atomic Motions in the Optical Suppression of Charge Density Waves, Nature 468, 799 (2010).

[30] N. Erasmus, M. Eichberger, K. Haupt, I. Boshoff, G. Kassier, H. Berger, J. Demsar, and H. Schwoerer, Ultrafast Dynamics of Charge Density Waves in 4 H b − TaSe 2 Probed by Femtosecond Electron Diffraction, Phys. Rev. Lett. 109, 167402 (2012).

[31] C. Laulhé, L. Cario, B. Corraze, E. Janod, T. Huber, G. Lantz, S. Boullaat, A. Ferrer, S. O. Mariager, J. A. Johnson, et al., X-Ray Study of Femtosecond Structural Dynamics in the 2D Charge Density Wave Compound 1T-TaS2, Physica B: Condensed Matter 460, 100 (2015).

[32] T.-R. T. Han, Z. Tao, S. D. Mahanti, K. Chang, C.-Y. Ruan, C. D. Malliakas, and M. G. Kanatzidis, Structural Dynamics of Two-Dimensional Charge-Density Waves in CeTe 3 Investigated by Ultrafast Electron Crystallography, Physical Review B 86, (2012).

[33] C. W. Nicholson, A. Lücke, W. G. Schmidt, M. Puppin, L. Rettig, R. Ernstorfer, and M. Wolf, Beyond the Molecular Movie: Dynamics of Bands and Bonds during a Photoinduced Phase Transition, Science 362, 821 (2018).

[34] X. Shi, W. You, Y. Zhang, Z. Tao, P. M. Oppeneer, X. Wu, R. Thomale, K. Rossnagel, M. Bauer, H. Kapteyn, et al., Ultrafast Electron Calorimetry Uncovers a New Long-Lived Metastable State in 1 T -TaSe 2 Mediated by Mode-Selective Electron-Phonon Coupling, Sci. Adv. 5, eaav4449 (2019).

[35] K. Haupt, M. Eichberger, N. Erasmus, A. Rohwer, J. Demsar, K. Rossnagel, and H. Schwoerer, Ultrafast Metamorphosis of a Complex Charge-Density Wave, Physical Review Letters 116, (2016).

[36] T.-R. T. Han, F. Zhou, C. D. Malliakas, P. M. Duxbury, S. D. Mahanti, M. G. Kanatzidis, and C.-Y. Ruan, Exploration of Metastability and Hidden Phases in Correlated Electron Crystals Visualized by Femtosecond Optical Doping and Electron Crystallography, Science Advances 1, e1400173 (2015).

[37] S. Vogelgesang, G. Storeck, J. G. Horstmann, T. Diekmann, M. Sivis, S. Schramm, K. Rossnagel, Schäfer, and C. Ropers, Phase Ordering of Charge Density Waves Traced by Ultrafast Low-Energy Electron Diffraction, Nature Physics 14, 184 (2018).

[38] G. Lantz, C. Laulhé, S. Ravy, M. Kubli, M. Savoini, K. Tasca, E. Abreu, V. Esposito, M. Porer, A. Ciavardini, et al., Domain-Size Effects on the Dynamics of a Charge Density Wave in 1 T − TaS 2, Physical Review B 96, (2017).

[39] J. D. Axe, Debye-Waller Factors for Incommensurate Structures, Physical Review B 21, 4181 (1980).

[40] A. W. Overhauser, Observability of Charge-Density Waves by Neutron Diffraction, Physical Review B 3, 3173 (1971).
[41] J. Li, J. Li, K. Sun, L. Wu, H. Huang, R. Li, J. Yang, X. Wang, H. Luo, R. J. Cava, et al., Ultrafast Decoupling of Atomic Sublattices in a Charge-Density-Wave Material, ArXiv:1903.09911 (2019).
[42] W. S. Lee, Y. D. Chuang, R. G. Moore, Y. Zhu, L. Pattthey, M. Trigo, D. H. Lu, P. S. Kirchmann, O. Krupin, M. Yi, et al., Phase Fluctuations and the Absence of Topological Defects in a Photo-Excited Charge-Ordered Nickelate, Nature Communications 3, 838 (2012).
[43] M. Gulde, S. Schweda, G. Storeck, M. Maiti, H. K. Yu, A. M. Wodtke, S. Schafer, and C. Ropers, Ultrafast Low-Energy Electron Diffraction in Transmission Resolves Polymer/Graphene Superstructure Dynamics, Science 345, 200 (2014).
[44] G. Storeck, S. Vogelgesang, M. Sivis, S. Schäfer, and C. Ropers, Nanotip-Based Photoelectron Microgun for Ultrafast LEED, Structural Dynamics 4, 044024 (2017).
[45] J. G. Horstmann, B. Wit, G. Storeck, and C. Ropers, Coherent Control of a Structural Phase Transition in a Solid-State Surface System, ArXiv:1906.11155 (2019).
[46] C. B. Scruby, P. M. Williams, and G. S. Parry, The Role of Charge Density Waves in Structural Transformations of 1T TaS$_2$, Philosophical Magazine 31, 255 (1975).
[47] A. Spijkerman, J. L. de Boer, A. Meetsma, G. A. Wiegers, and S. van Smaalen, X-Ray Crystal-Structure Refinement of the Nearly Commensurate Phase of 1T-TaS$_2$ in (3+2)-Dimensional Superspace, Phys. Rev. B 56, 13757 (1997).
[48] F. Jellinek, The System Tantalum-Sulfur, Journal of the Less Common Metals 4, 9 (1962).
[49] I. Vaskivskyi, J. Gospodaric, S. Brazovskii, D. Svetin, P. Sutar, E. Goreshnik, I. A. Mihailovic, T. Mertelj, and D. Mihailovic, Controlling the Metal-to-Insulator Relaxation of the Metastable Hidden Quantum State in 1T-TaS$_2$, Science Advances 1, e1500168 (2015).
[50] P. Bak, Commensurate Phases, Incommensurate Phases and the Devil’s Staircase, Reports on Progress in Physics 45, 587 (1982).
[51] S. Hellmann, T. Rohwer, M. Kalläne, K. Hanff, C. Sohrt, A. Carr, M. M. Murnane, H. C. Kapteyn, L. Kipp, et al., Time-Domain Classification of Charge-Density-Wave Insulators, Nat Commun 3, 1069 (2012).
[52] P. Fazekas and E. Tosatti, Electrical, Structural and Magnetic Properties of Pure and Doped 1T-TaS$_2$, Philosophical Magazine B 39, 229 (1979).
[53] P. Darancet, A. J. Millis, and C. A. Marianetti, Three-Dimensional Metallic and Two-Dimensional Insulating Behavior in Octahedral Tantalum Dichalcogenides, Phys. Rev. B 90, 045134 (2014).
[54] K. T. Law and P. A. Lee, 1T-TaS$_2$ as a Quantum Spin Liquid, Proc Natl Acad Sci USA 114, 6996 (2017).
[55] K. Rossnagel, On the Origin of Charge-Density Waves in Select Layered Transition-Metal Dichalcogenides, Journal of Physics: Condensed Matter 23, 213001 (2011).
[56] B. Sipos, A. F. Kusmartseva, A. Akrap, H. Berger, L. Forró, and E. Tutiš, From Mott State to Superconductivity in 1T-TaS$_2$, Nature Mater 7, 960 (2008).
[57] L. Stojevska, I. Vaskivskyi, T. Mertelj, P. Kusar, D. Svetin, S. Brazovskii, and D. Mihailovic, Ultrafast Switching to a Stable Hidden Quantum State in an Electronic Crystal, Science 344, 177 (2014).
[58] T. Ritschel, J. Trinckauf, K. Köpennik, B. Büchner, M. v. Zimmermann, H. Berger, Y. I. Joe, P. Abbamonte, and J. Geck, Orbital Textures and Charge Density Waves in Transition Metal Dichalcogenides, Nature Physics 11, 328 (2015).
[59] M. Klanjšek, A. Zorko, R. Žitko, J. Mravlje, Z. Jagličić, P. K. Biswas, P. Prelovšek, D. Mihailovic, and D. Arčon, A High-Temperature Quantum Spin Liquid with Polaron Spins, Nature Phys 13, 1130 (2017).
[60] B. Dardel, M. Grioni, D. Malterre, P. Weibel, Y. Baer, and F. Lévy, Temperature-Dependent Pseudogap and Electron Localization in 1T-TaS$_2$, Phys. Rev. B 45, 1462 (1992).
[61] K. Nakanishi and H. Shiba, *Domain-like Incommensurate Charge-Density-Wave States and the First-Order Incommensurate-Commensurate Transitions in Layered Tantalum Dichalcogenides. I. 1T-Polytype*, J. Phys. Soc. Jpn. 43, 1839 (1977).

[62] K. Nakanishi, H. Takatera, Y. Yamada, and H. Shiba, *The Nearly Commensurate Phase and Effect of Harmonics on the Successive Phase Transition in 1T-TaS2*, J. Phys. Soc. Jpn. 43, 1509 (1977).

[63] T. Ishiguro and H. Sato, *High-Resolution Electron Microscopy of Discommensuration in the Nearly Commensurate Phase on Warming of 1T-TaS2*, Phys. Rev. B 52, 759 (1995).

[64] C. Laulhé, T. Huber, G. Lantz, A. Ferrer, S. O. Mariager, S. Grübel, J. Rittmann, J. A. Johnson, V. Esposito, A. Lübcke, et al., *Ultrafast Formation of a Charge Density Wave State in 1 T − TaS2 : Observation at Nanometer Scales Using Time-Resolved X-Ray Diffraction*, Physical Review Letters 118, (2017).

[65] L. Le Guyader, T. Chase, A. H. Reid, R. K. Li, D. Svetin, X. Shen, T. Vecchione, X. J. Wang, D. Mihailovic, and H. A. Dürr, *Stacking Order Dynamics in the Quasi-Two-Dimensional Dichalcogenide 1 T−TaS2 : Probed with MeV Ultrafast Electron Diffraction*, Structural Dynamics 4, 044020 (2017).

[66] I. Avigo, F. Queisser, P. Zhou, M. Liggies, K. Rossnagel, R. Schützhold, and U. Bovensiepen, *Doublon Bottleneck in the Ultrafast Relaxation Dynamics of Hot Electrons in 1T-TaS2*, ArXiv:1907.11677 (2019).

[67] A. Zong, A. Kogar, Y.-Q. Bie, T. Rohwer, C. Lee, E. Baldini, E. Ergeçen, M. B. Yilmaz, B. Freelon, E. J. Sie, et al., *Evidence for Topological Defects in a Photoinduced Phase Transition*, Nature Physics 15, 27 (2018).

[68] A. Mann, E. Baldini, A. Odeh, A. Magrez, H. Berger, and F. Carbone, *Probing the Coupling between a Doublon Excitation and the Charge-Density Wave in TaS2 by Ultrafast Optical Spectroscopy*, Phys. Rev. B 94, 115122 (2016).

[69] P. Kusar, T. Mertelj, V. V. Kabanov, J.-H. Chu, I. R. Fisher, H. Berger, L. Forró, and D. Mihailovic, *Anharmonic Order-Parameter Oscillations and Lattice Coupling in Strongly Driven 1 T − TaS2 and TbTe 3 Charge-Density-Wave Compounds: A Multiple-Pulse Femtosecond Laser Spectroscopy Study*, Phys. Rev. B 83, 035104 (2011).

[70] G. F. Giuliani and A. W. Overhauser, *Structure Factor of a Charge-Density Wave*, Phys. Rev. B 23, 3737 (1981).

[71] W. L. McMillan, *Landau Theory of Charge-Density Waves in Transition-Metal Dichalcogenides*, Phys. Rev. B 12, 1187 (1975).

[72] A. Ichimiya and P. I. Cohen, *Reflection High-Energy Electron Diffraction* (Cambridge University Press, Cambridge, U.K.; New York, 2004).

[73] M. A. V. Hove, W. H. Weinberg, and C.-M. Chan, *Low-Energy Electron Diffraction: Experiment, Theory and Surface Structure Determination* (Springer-Verlag, 1986).

[74] M. J. Stern, L. P. René de Cotret, M. R. Otto, R. P. Chatelain, J.-P. Boisvert, M. Sutton, and B. J. Siwick, *Mapping Momentum-Dependent Electron-Phonon Coupling and Nonequilibrium Phonon Dynamics with Ultrafast Electron Diffuse Scattering*, Phys. Rev. B 97, 165416 (2018).

[75] L. Waldecker, R. Bertoni, H. Hübener, T. Brumme, T. Vasileiadis, D. Zahn, A. Rubio, and R. Ernstorfer, *Momentum-Resolved View of Electron-Phonon Coupling in Multilayer WSe2*, Physical Review Letters 119, (2017).

[76] X. Gu and R. Yang, *Phonon Transport in Single-Layer Transition Metal Dichalcogenides: A First-Principles Study*, Appl. Phys. Lett. 105, 131903 (2014).

[77] M. Mohr, J. Maultzsch, E. Dobardzić, S. Reich, I. Milošević, M. Damnjanović, A. Bosak, M. Krisch, and C. Thomsen, *Phonon Dispersion of Graphite by Inelastic X-Ray Scattering*, Phys. Rev. B 76, 035439 (2007).

Note that the different behavior of these inequivalent peaks is unrelated to a potential decoupling of atomic sublattices, as reported in Ref. [41] based on different groups of peaks. In fact, the diffraction signal is inherently mostly affected by the sulfur atoms due to the much larger atomic scattering factors at 100 eV energy.
[79] M. Eichberger, Probing Structural Dynamics in Dichalcogenides with Ultrashort Electron Pulses, Universität Konstanz, 2014.
[80] A. Kogar, A. Zong, P. E. Dolgirev, X. Shen, J. Straquadine, Y.-Q. Bie, X. Wang, T. Rohwer, I.-C. Tung, Y. Yang, et al., Light-Induced Charge Density Wave in LaTe$_3$, ArXiv:1904.07472 (2019).
[81] P. A. Lee, T. M. Rice, and P. W. Anderson, Fluctuation Effects at a Peierls Transition, Phys. Rev. Lett. 31, 462 (1973).
[82] R. H. McKenzie and J. W. Wilkins, Effect of Lattice Zero-Point Motion on Electronic Properties of the Peierls-Fröhlich State, Phys. Rev. Lett. 69, 1085 (1992).
[83] L. Degiorgi, G. Grüner, K. Kim, R. H. McKenzie, and P. Wachter, Optical Probing of Thermal Lattice Fluctuations in Charge-Density-Wave Condensates, Phys. Rev. B 49, 14754 (1994).
[84] H. Monien, Exact Results for the Crossover from Gaussian to Non-Gaussian Order Parameter Fluctuations in Quasi-One-Dimensional Electronic Systems, Phys. Rev. Lett. 87, 126402 (2001).