

Concurrent probing of electron-lattice dephasing induced by photoexcitation in 1T-TaSeTe using ultrafast electron diffraction

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It has been technically challenging to concurrently probe the electrons and the lattices in materials during nonequilibrium processes, allowing their correlations to be determined. Here, in a single set of ultrafast electron diffraction patterns taken on the charge-density-wave (CDW) material 1T-TaSeTe, we discover a temporal shift in the diffraction intensity measurements as a function of scattering angle. With the help of dynamic models and theoretical calculations, we show that the ultrafast electrons probe both the valence-electron and lattice dynamic processes, resulting in the temporal shift measurements. Our results demonstrate unambiguously that the CDW is not merely a result of the periodic lattice deformation ever present in 1T-TaSeTe but has significant electronic origin. This method demonstrates an approach for studying many quantum effects that arise from electron-lattice dephasing in molecules and crystals for next-generation devices.

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Observation of the incoherent movements of electrons and lattice, i.e., electron-lattice dephasing in excited states, is of fundamental importance to understand charge-lattice interactions [1–5]. Recently, the rapid development of ultrafast methods offers the opportunity to trace dynamics in structures for characterizing the charge-lattice interactions induced by photoexcitations [1–3,6–12]. Many ultrafast studies have focused on charge-density-wave (CDW) materials [1,7–12] that are of great interest due to their intimate relation to a variety of captivating electronic properties including metal-insulator transition and superconductivity [4,5,13–15]. The symmetry-broken states of CDW are depicted by real-space charge-density redistributions, which often result in a periodic lattice distortion (PLD) at equilibrium [16], or vice versa. It is this “electron dichotomy” reflecting distinct electronic and lattice contributions to the CDW that gives rise to much of the ongoing debates about the nature and origin of CDWs in various systems. One of the most compelling challenges in studying CDW materials is to probe the dynamics of charge states and the lattice distortions concurrently because only the coevolution can yield the correct understanding of the driving mechanisms [1,6,7]. Limited by technical difficulties, however, the electron dynamics and lattice evolution in a material during nonequilibrium processes are commonly investigated by discrete methods; for instance, angle-resolved photoemission spectroscopy for electron dynamics [17,18]

and x-ray diffraction for lattice dynamics [19]. Correlating the observations of the different experimental methods requires careful synchronization; however, it is often an insurmountable problem due to the distinct nature of the probes and the different experimental and material conditions employed.

Electron diffraction techniques have been developed for nearly a century, with intrinsic advantages compared to other scattering tools [20] due to the high electron-scattering cross sections. Unlike for x rays, which interact with all the electrons surrounding an atom, and are therefore sensitive to atom positions, electrons interact with the electrostatic potential of an atom—its positively charged nucleus screened by its negatively charged electron cloud [21–23]. Thus, the scattering amplitude of an atom for incident electrons at small scattering angles is determined mainly by an atom’s valence charge, rather than by the total density of electrons [see Fig. 1(a)]. In other words, the electron-scattering atomic form factor at small scattering angles is strongly influenced by the valence electron distribution of atoms and charge transfer in a crystal. This is particularly advantageous for crystals with large unit cells that have reflections at small scattering angles [21]. In contrast, in a complementary fashion, electrons scattered to high scattering angles are extremely sensitive to subtle changes of atom positions or atomic motion.

Inspired by this feature of the scattering mechanism, we analyze the intensity of superlattice reflections (SLRs) that correspond to the CDW superstructure in the single-crystalline layered transition metal dichalcogenide 1T-TaSeTe using MeV ultrafast electron diffraction (UED) [24,25], taking

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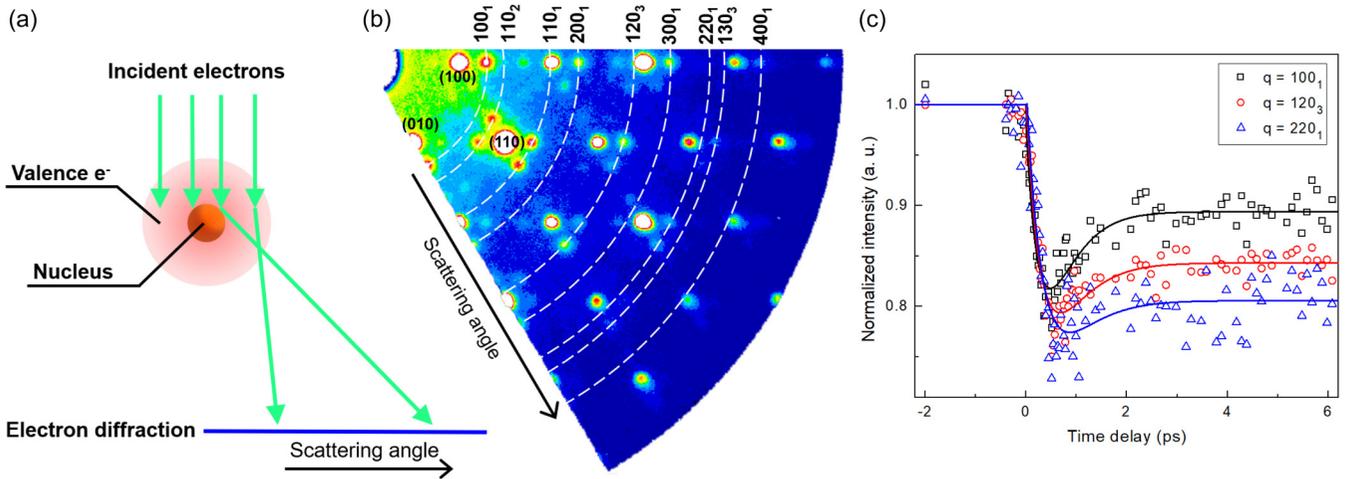


FIG. 1. Scattering-angle-dependent dynamics measured via UED. (a) A schematic representation of electron scattering, illustrating that scattering from valence electrons is predominant at small scattering angles, while it is from nuclei at high scattering angles. (b) A part of an experimental UED pattern showing multiple Bragg reflections and SLRs. Nine sets of SLRs were selected for the measurements with their scattering angles marked by the dashed lines. (c) Intensity variation (normalized by the averaged intensity before pumping) vs time delay measured from representative SLRs. The solid lines are fittings of the experimental data to a two-exponential model.

advantage of its sensitivity to the valence charge and atomic displacements, which dominate at different scattering angles. The 1T-TaSeTe single crystals were grown by chemical vapor transport with iodine as a transport agent [26]. In fact, the PLD in our samples exists at all the measurable temperatures until they melt. This raises a serious question about whether the CDW in this material is truly of some electronic origin or merely a result of the ever-present lattice deformation. Understanding this extreme case is instrumental to resolving the aforementioned ongoing debate about the nature and origin of CDWs in various systems. The UED experiments were performed on the 3.5-MeV-UED setup at the Stanford Linear Accelerator Laboratory excited by a laser pulse with a photon energy of 1.55 eV (center wavelength of 800 nm) and a fluence of 3.5 mJ/cm^2 [27]. This classic layered 1T dichalcogenide has a random Te-Se distribution on the dichalcogenide lattice site [26]; the Te/Se mixture is required to stabilize the 1T structure. The measurement temperature is 26 K to minimize the thermal atomic motion and diffuse scattering background. A portion of the UED pattern (at $t = -2$ ps; before pumping) is shown in Fig. 1(b), with nine sets of the SLRs and their positions in reciprocal space as a function of scattering angles marked by the dashed line [28]. The subscripts 1, 2, and 3 correspond to indexes $(hkl100)$, $(hkl010)$, and $(hkl0\bar{1}0)$, respectively, owing to the triple- q states in the CDW [26,27]. Normalized intensities (normalized to the averaged intensity before $t = 0$) as a function of time delay from three selected SLRs are shown in Fig. 1(c). Through quantitative intensity analysis we determine the atomic positions during the lattice evolution upon photoexcitation [27]. The SLR intensities as a function of time are simulated and shown in Supplemental Material Fig. S4 [28]. The simulation is consistent with the experiment, revealing that there is a rapid drop of the normalized intensities, which reach a minimum at a few hundred femtoseconds, then recover on a slower timescale. We use a two-exponential function to fit the SLR intensities in Fig. 1(c) which well describes the experimental observations.

Compared with the reconstructed lattice evolution using the same set of UED data [27], we notice that at the time when the SLR intensities reach the minimum (the cusp), both the Ta and Se/Te atoms depart furthest from their PLD state with distorted positions (i.e., before pumping). The temporal characteristics in the lattice evolution, particularly the time of the cusp t_c , should be equally reflected by all the SLRs suggested by the diffraction simulations of all the SLRs that t_c is the same given the same set of atomic displacements through the dynamics (see simulated intensity variations in Figs. S4 and S5 using both kinematic and dynamic electron-scattering simulations) [28]. However, unexpectedly we observe an intriguing difference in values of t_c measured for the SLRs at different scattering angles. To better visualize the shift of t_c , we renormalized the intensity plots [Fig. 2(a)]. The intensity renormalization affects the time constants in the two-exponential curve fitting but it does not affect the value of t_c . In Fig. 2(a), the curve fits of the intensity variations of all nine SLRs show a shift of t_c toward higher values as the scattering vector length s increases (also see inset). The measured t_c vs s behavior is plotted in Fig. 2(b) (black dots), indicating that the value of t_c changes from ~ 0.48 ps for the (100_1) reflection to ~ 0.88 ps for the (400_1) reflection.

Since the evolution of atomic displacements in the lattice do not induce the measured shift in t_c , we move further to examine the lattice vibration effect on the SLRs, which are sensitive to scattering angle, on the measured t_c values. The normalized SLRs intensity at time t' can be expressed by $\frac{I(t')}{I(t_0)} = \frac{I(u')}{I(u_0)} e^{-2B(t')s^2}$, where t_0 is time zero, u' the lattice distortion at t' , and u_0 the lattice distortion at t_0 ; $B(t')$ is the effective Debye-Waller (D-W) factor at t' assuming isotropic and identical for all the atoms; and $s = \sin\theta/\lambda$ (θ being the half of scattering angle and λ is the electron wavelength) as the scattering vector [20,22]. It is widely accepted during the warmup that $B(t')$ can be expressed by $B(t') = a(1 - e^{-t'/\tau_{DW}})$ assuming the lattice has negligible vibration at time zero,

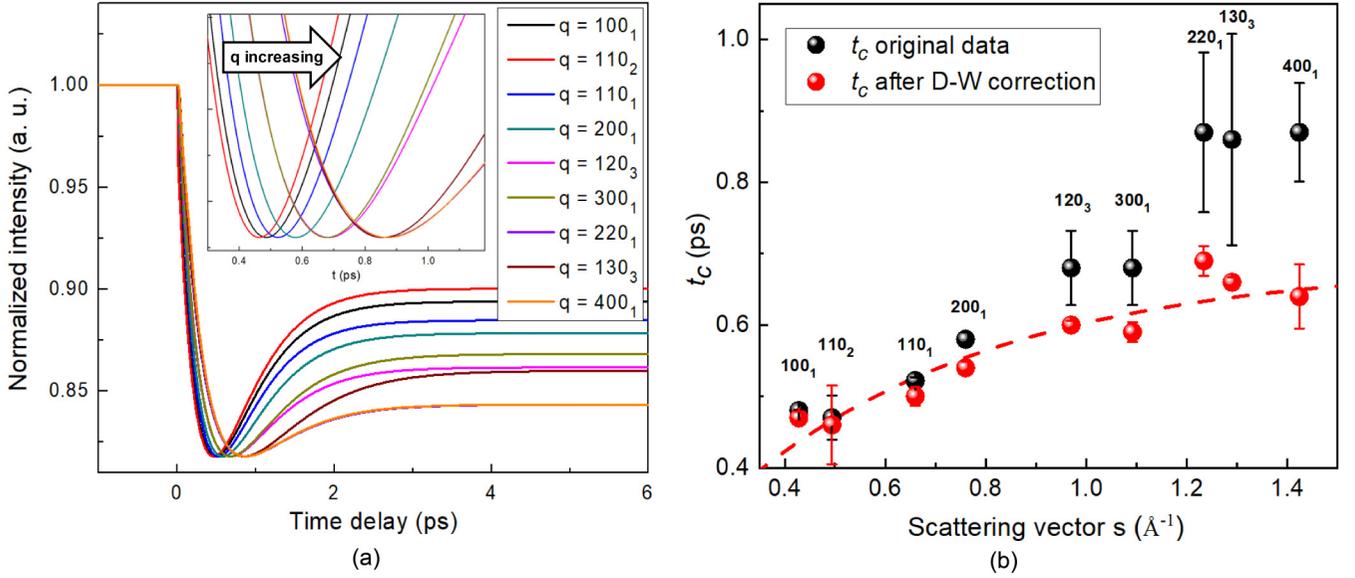


FIG. 2. Measurement of the time at the cusp point t_c of the SLRs during the lattice relaxation process as a function of the scattering vector length s . (a) Two-exponential fitting curves for nine sets of SLRs, showing a shift of t_c toward increased time delay with the increase of scattering vector s . (b) A plot of t_c vs the scattering vector length s of the SLRs. The black dots are measured directly from the fitting of the raw diffraction data. The red dots are the measurements after the removal of the influence of the D-W factors on the values of t_c [28]. The red dashed line is a guide to the eye for the red dots. The error bars mean the deviation of the t_c values determined in two distinct fitting methods, as shown in Figs. S2, S2-1, and S2-2 [28].

where τ_{DW} is the time constant for the change in the lattice vibration and a describes the value of $B(t')$ where it reaches saturation [29–31]. By applying the D-W term to the intensities of the SLRs, we find that the t_c in the intensity variation can be shifted to higher values, depending on the parameters a and τ_{DW} in the $B(t')$ expression. According to the intensity expression, we measured $\frac{I(t')}{I(t_0)}$ and calculated the $\frac{I(t')}{I(t_0)}$ for all nine SLRs at $t' = 6$ ps when the system reaches a quasistable condition. Then the parameter a is determined to be $\sim 0.134(\text{\AA}^2)$ by the slope of the plot of $\ln[\frac{I(t')}{I(t_0)} / \frac{I(t')}{I(t_0)}]$ vs s^2 , as shown in Fig. S6 [28]. While it is hard to accurately determine τ_{DW} , we find that the D-W term has a maximum effect on the t_c value (i.e., t_c exhibits the largest shift) when $\tau_{DW} \sim 0.9$ ps [28]. Given all the above considerations in measurements, we remove the lattice vibration effect on the intensity variations measured from the nine SLRs by dividing the raw data by $e^{-2B(t')s^2}$, in which $B(t') = 0.134(1 - e^{-t'/0.9})$, and fit the processed data again. The corrected values of t_c are plotted as the red dots in Fig. 2(b), with the correction as scattering-angle dependent. Note that the correction for the D-W term is the upper limit of the effect in this case for the reason stated above. Therefore, the lattice behavior, including the atomic displacements and the lattice vibrations, cannot be responsible for the t_c shift that is unambiguously observed in the experiment.

To explore the origin of the t_c shift, we employ theoretical models to explain the dynamic behavior of the system. The electrons are excited abruptly by the pumping photons, and the excitation and relaxation process takes place within a few femtoseconds [6,32,33]. Assuming that an electronic order parameter η in the material relaxes in an exponential decay

from the excited states, then

$$\eta(t) = \eta_f + (\eta_i - \eta_f)e^{-t/\tau_\eta}, \quad (1)$$

where τ_η is the time constant for the electron relaxation, and η_i and η_f are the initial and a semifinal stage (when the measurements reach metastable values after $t \sim 3$ ps [27]) of the electronic order, respectively. On the other hand, the lattice order parameter Q can be written in a dynamic equation

$$\frac{dQ}{dt} = \frac{Q - Q_f(\eta)}{\tau_Q}, \quad (2)$$

where lattice order Q_f in the final states (after 3 ps) is a function of η instead of a simple time-independent constant and τ_Q describes how fast the lattice follows the change in the electronic order. Taking linear coupling between the lattice and electrons for simplicity, i.e., $Q_f(\eta) = \eta$, Eq. (2) is $\frac{dQ}{dt} = \frac{Q - [\eta_f + (\eta_i - \eta_f)e^{-t/\tau_\eta}]}{\tau_Q}$, which has an analytical solution as follows.

$$Q(t) = e^{-t/\tau_Q} + \eta_f(1 - e^{-t/\tau_Q}) + \frac{(\eta_i - \eta_f)\tau_\eta}{\tau_Q - \tau_\eta}(e^{-t/\tau_Q} - e^{-t/\tau_\eta}). \quad (3)$$

Equations (1) and (3) are plotted as the black and red (both bold) curves in Fig. 3 to represent the dynamics of electron and lattice, respectively, by setting $\eta_i = 0.6$, $\eta_f = 0.9$, $\tau_\eta = 0.4$, and $\tau_Q = 0.3$. Equation (3) provides a phenomenological interpretation of the fitting functions that the two time constants (τ_η and τ_Q) are associated with the electron relaxation speed (even when we measure the response of the lattice) and the speed that the lattice follows the change in electrons. The amplitudes of the two-exponential fittings, which have been

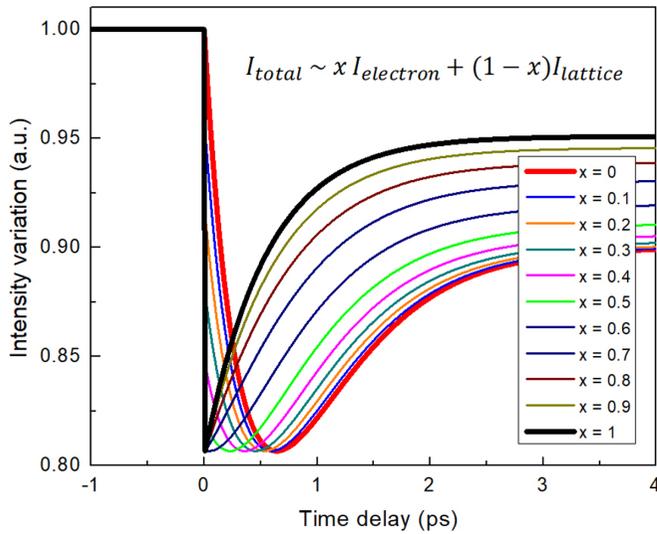


FIG. 3. Intensity variation as a function of time derived from dynamic models. The bold black curve reflects the electron while the bold red curve reflects the lattice dynamics considering the electron relaxation rate and the electron-lattice coupling time constant. Colored curves are the linear combinations of the black and the red curves, indicating a shift of t_c as a function of x , or the weight of the electron dynamics, in the total measurement.

often employed in UED data analysis [8,12,27,34], can now be explicitly expressed by Eq. (3). Indeed, to be comparable to the experimental results, τ_η and τ_Q need to have similar values (~ 0.3 ps in this case). This indicates that the relaxation time of the electronic order is not independent of its environment, but strongly coupled to the lattice dynamics. Such an implication is consistent with polaron-type behavior (i.e., the electron and lattice dynamics are intertwined) suggested by previous ultrafast observations in a doped manganite [34]. Most interestingly, the theoretical plots clearly show that a mixture of the lattice order with the electronic order (see a linear combination of the black and red bold curves in Fig. 3) can explain the shift of the cusp of the curve in ultrafast-time regime. The more weight of electronic order in the mixed intensity, the faster the curve reaches its cusp. Imagining an electron beam that probes mainly the lattice dynamics with a certain portion of its diffraction intensity as being due to the electron dynamics, the intensity variation would be identical to the curves depicted in Fig. 3. In comparison to the experimental findings in Fig. 2(b) and based on the scattering principles illustrated in Fig. 1(a), we interpret the shift of measured t_c in Fig. 2(b) as arising from the coevolution of the lattice and the electron dynamics, which are both reflected in the diffraction intensity variations. Even a few percent of electron contribution to the total intensity yields a shift of t_c for the data at small scattering vector length s , while at higher scattering angles (larger s), the value of t_c is predominantly dictated by the lattice dynamics because the electron contribution is nearly zero.

The reflection of the electron dynamics in the UED measurements as a function of scattering vector are substantiated by density-functional-theory (DFT) calculations TaSe₂ in the 1T structure. The normal state, with a high-symmetry electron/lattice arrangement (no CDW), and the CDW state,

with symmetry breaking, were both calculated; their charge-density distributions in real space are illustrated in Fig. 4(a). Structure factors (and intensities I_{total}) using total charge for the nine SLRs were further derived from the calculated structures and charge-density mapping for both x-ray and electron diffraction. In addition, structure factors (and intensities I_{valence}) using valence electrons only, that are identified to be 5d and 6s electrons for Ta atoms and 4p electrons for Se atoms (note that other orbital electrons can also be involved in the photoexcitation), were calculated as well using the DFT results. The ratios of I_{valence} to I_{total} are plotted in Fig. 4(b) as a function of s for both x rays and electrons. It clearly shows that both techniques manifest a scattering-angle-dependent intensity variation and the weight of valence electrons in the total intensity is much higher in electron diffraction than that in x-ray diffraction, particularly at small scattering angles. Thus the (valence) electron dynamics can well be reflected in the temporal characteristics measured from UED. Note that the DFT calculations in Fig. 4 are from (3 × 3)-type CDW structures based on the experimental observations. Moreover, similar DFT calculations and the derived intensities for electron and x-ray diffractions from the Star-of-David-type (with a $\sqrt{13} \times \sqrt{13}$ unit cell) CDW, which is the low-temperature state of 1T-TaSe₂, can be found in Fig. S7 with discussions [28]. Both CDW patterns have in common that in each cluster the six nearest-neighbor Ta atoms of the central Ta atom move toward the center. With the help of theoretical modeling and calculations, the results show that the lattice dynamics are driven by the change in the electronic structure in this material, which addresses the question of whether the origin of the superstructure originates in chemical order or electronic instability (i.e., a CDW) [4,5,26,35].

Separating scattering contributions of valence electrons from a lattice of atomic nuclei and inner-shell electrons is of great importance but not a trivial task with diffraction. Because incident electrons interact with electrostatic potentials of the sample, i.e., atomic nuclei screened by the electron clouds, electron diffraction has a better capability, compared to x-ray techniques, to distinguish the contribution of valence electrons from the total scattering intensity (Ref. [21], and quantitatively manifested in this work). Our findings demonstrate an experimental approach to concurrently probe both lattice and electron dynamics at an ultrafast timescale because the lattice and electrons move incoherently with distinct dynamics, manifesting that 1T-TaSe₂ is a bona fide CDW material in which the CDW is not merely a result of the ever-present PLD and suggesting that CDW in systems that otherwise exhibit a PLD phase transition are even more likely to have significant electronic origin.

In a broader scope, in many correlated electron systems the low-energy electrons near the Fermi level tend to self-partition into fast (itinerant) and slow (more localized) ones via various mechanisms. Well-known examples include the cuprates [38] and the iridates [39] in which the partition occurs in the momentum space as node and antinode regions, the iron-based superconductors in which the partition takes place via the orbital-selective Mott transition [40,41], and the orbital-selective Peierls transitions in CuIr₂S₄ spinel [42] and NaTiSi₂O₆ pyroxene [43]. Again, it is this “electron dichotomy” that gives rise to much of the ongoing debates about

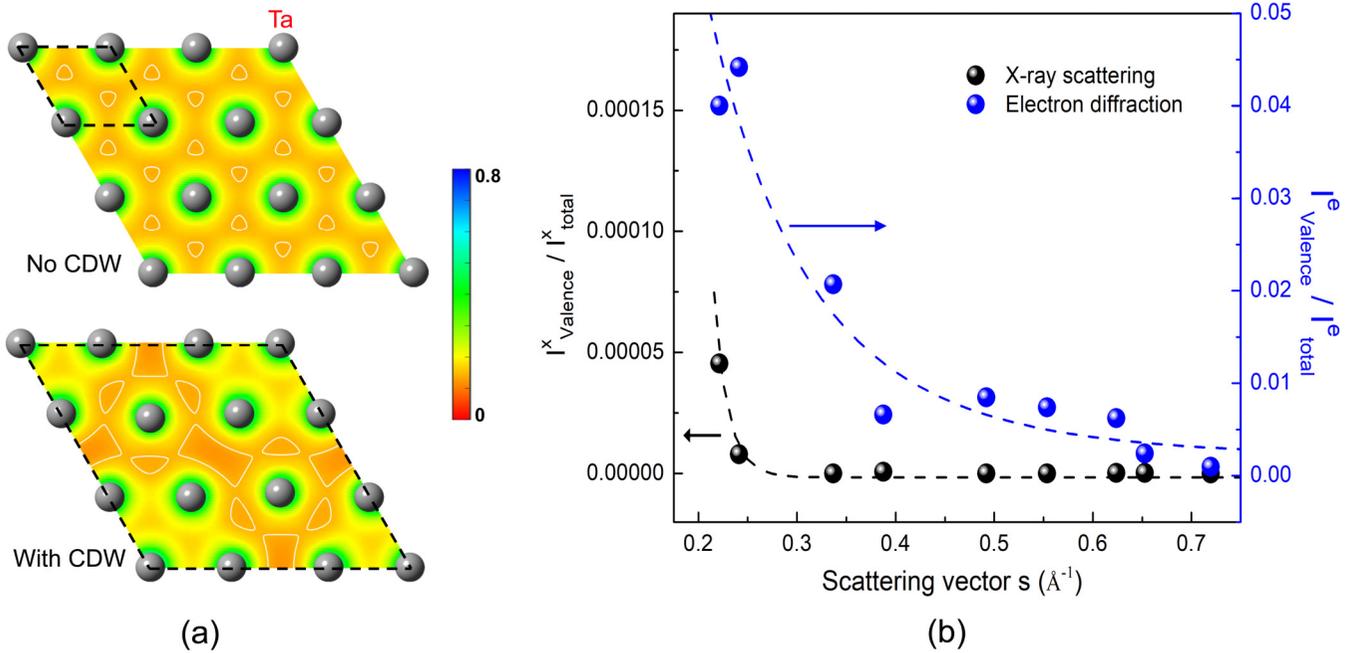


FIG. 4. Calculated (planar) charge-density distributions and associated x-ray/electron-scattering intensities. (a) Charge-density mapping of the Ta plane (Ta atoms are shown) in $1T$ -TaSe₂ based on DFT calculations for the state with no CDW (upper; unit cell marked by the black dashed lines) and the CDW state (lower; unit cell is 3×3 times larger). The color scale shows the number of electrons in unit-cell volume. (b) Structure factors of the CDW state were calculated, corresponding to the nine measured SLRs, using valence charges ($5d$ and $6s$ electrons for Ta atoms and $4p$ electrons for Se atoms) and total charges of Ta and Se atoms based on the electronic structures in (a). The ratios of the scattering intensity from valence electrons to the total intensity are obtained and plotted for x-ray (black dots) and electron diffraction (blue dots). Note that the scales are different for x-ray and electron scattering. The dashed lines are guides to the eye.

the nature and origin of the various unusual phenomena in those systems. As the electrons move fast or slow through the lattice, their couplings to the lattice are substantially different. Hence, the present technique is anticipated to provide insights into many quantum materials by temporally separating and ultimately quantifying electron-lattice coupling on its fundamental timescales.

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