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ABSTRACT

We investigate the optically driven phase transition between two charge-density wave (CDW) states at the surface of tantalum disulfide (1*T*-TaS_2). Specifically, we employ a recently improved ultrafast low-energy electron diffraction setup to study the transition from the nearly commensurate to the incommensurate (IC) CDW state. The experimental setup allows us to follow transient changes in the diffraction pattern with high momentum resolution and 1-ps electron pulse duration. In particular, we trace the diffraction intensities and spot profiles of the crystal lattice, including main and CDW superstructure peaks, as well as the diffuse background. Harnessing the enhanced data quality of the instrumental upgrade, we follow the laser-induced transient disorder in the system and perform a spot-profile analysis that yields a substantial IC-peak broadening for very short time scales followed by a prolonged spot narrowing.

Charge-density waves (CDW) emerge in materials with considerable anisotropy and electron-lattice interaction.[1] As a result, CDW materials undergo a symmetry-breaking phase transition into a new ground state that is characterized by a modulation of the charge density, a periodic distortion of the crystal lattice (PLD), and the formation of an electronic gap. Many studies investigated the interdependence of electronic and lattice degrees of freedom and the characteristic collective modes[2–5] in the time domain using ultrafast techniques. For example, these works focused on the initial gap quench followed by coherent amplitude oscillations,[6–11] the dynamics of the periodic lattice distortion via diffractive techniques,[12–16] and the emergence as well as the transition between different CDW states.[17–23] An important aspect is the amount of disorder in the system introduced by CDW fluctuations and dislocation-type topological defects after photo-excitation.[16,24–27] Sensitive structural probes are ideal tools to capture this disorder as they provide access to transient changes of long-range order.

In this work, we study the ultrafast structural dynamics at the surface of 1*T*-TaS_2 using ultrafast low-energy electron diffraction (ULEED). Specifically, we revisit the phase transition between the NC and IC CDW states, which we investigated previously,[22,23] and provide data from a recently upgraded experimental setup. We employ ULEED to trace the optically induced phase transition and the subsequent relaxation dynamics. High momentum resolution at the 1-ps timescale combined with large scattering efficiency yields clear time-dependent traces of peak intensities and spot profiles. This allows us to track the energy relaxation pathways in the presence of transient structural disorder in the system.

The ULEED technique is part of a wider family of stroboscopic pump-probe schemes which provide access to laser-induced non-equilibrium dynamics on surfaces[16,22,28,29] or thin films.[30] In our setup, the *in situ* cleaved sample is optically excited by a fs-laser pulse (wavelength 1030 nm center wavelength, 200 fs duration, 100 kHz repetition rate, 100 μm pump spot diameter) and probed by a low-energy electron pulse (100 eV electron energy, 1 ps duration, 10 μm electron beam size) at a controlled time delay Δt [Fig. 1(b)]. By sweeping the pump-probe delay, we record a series of diffraction patterns mapping the entire dynamical process (see the supplementary material for a movie file). The time-dependent redistribution of electron diffraction intensity gives a detailed insight into the excitation and relaxation pathways of the structure. In order to achieve ultrafast temporal

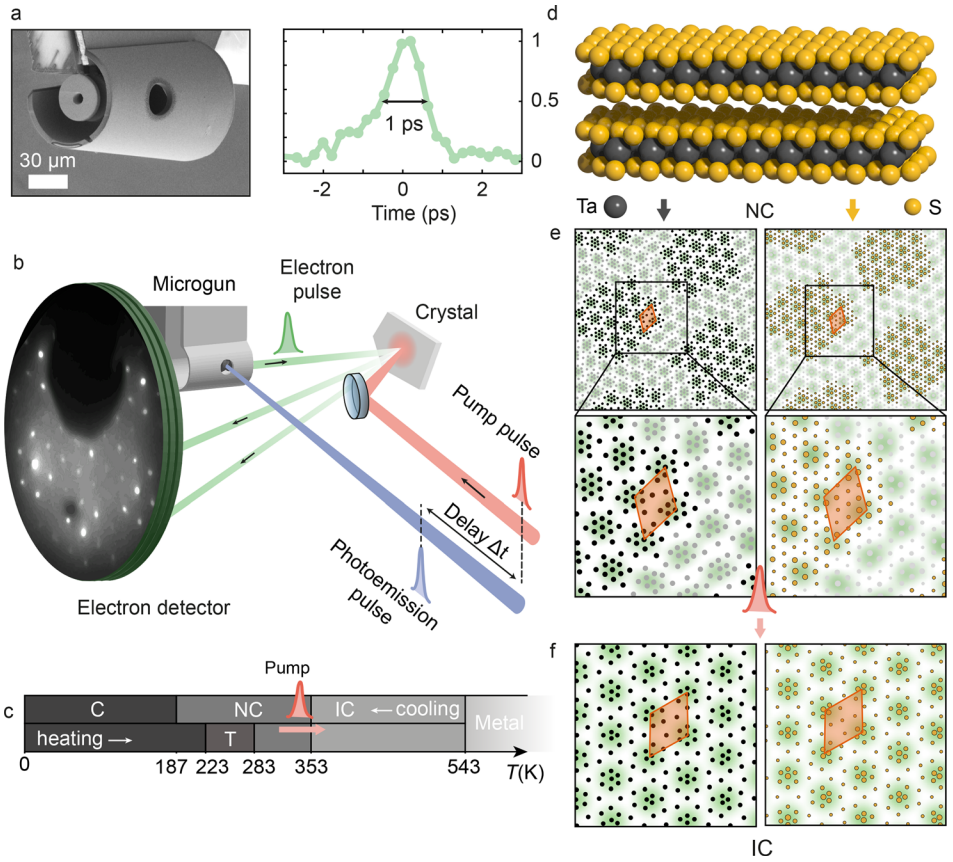

FIG. 1. Experimental setup and materials system. (a) Micrograph of electron microgun (left). The graph (right) shows the derivative of the fastest intensity change [blue curve in Fig. 2(d)] yielding an upper limit for the temporal resolution. (b) Sketch of ULEED setup illustrating the pump-probe approach. (c) 1*T*-TaS_2 exhibits several temperature-dependent CDW phases. (d) Layered tantalum disulfide (1*T*-TaS_2) (e) and (f) Illustrations of the in-plane and out-of-plane displacements in the NC and IC phase. The columns correspond to the displacement of tantalum (left column) and sulfur (right column) atoms, respectively. For sulfur, the size of circles indicates the displacement magnitude or direction. The orange rhomb specifies the CDW unit cell, while green areas illustrate electron charge accumulations. Values of NC displacements are obtained from Ref. 32, while, for the IC phase, we assume purely sinusoidal modulation. Displacements are shown exaggerated fivefold for illustration.

resolution at such electron energies, the development of a miniaturized photoelectron source, the microgun[16,31] [Fig. 1(a), left], was an essential element, providing 1 ps electron pulses at a 150 m working distance in the sample plane [Fig. 1(a), right].

Tantalum disulfide is a van der Waals material that consists of weakly interacting S-Ta-S stacks [Fig. 1(d)]. In the 1T polytype, the sulfur atoms are octahedrally coordinated around the sulfur atoms[33] [Fig. 1(d)]. The material properties of 1*T*-TaS_2 are known to be multifaceted, featuring charge order,[34,35] electron correlation effects,[36–38] orbital texture,[39,40] and different types of stacking order.[41–43] Multiple temperature-dependent charge-density wave states[34] [Fig. 1(c)] emerge at a temperature of below 543 K, each introducing a characteristic superstructure in the system. The orientation and size of the corresponding CDW unit cell change with temperature and lead to CDW phases of different symmetry, namely the incommensurate (IC), nearly commensurate (NC), trigonal and commensurate phase (C) [Fig. 1(c)].[44] In this work, we focus on the NC and IC phases whose unit cells are rotated by approximately 12° with respect to each other.[21,22] Each CDW phase features a characteristic PLD which is probed via electron diffraction. Moreover, it is important to note that the PLD affects both the displacements of tantalum and sulfur atoms, however, to a different extent concerning the respective directions and magnitudes[32,43] [Fig. 1(c)]. Commonly the prominent in-plane displacements of tantalum atoms are depicted, leading to clusters of 13 atoms, called "Star of David" formations (NC and C phase).[33] For the sulfur atoms, however, the situation is reversed, since the out-of-plane displacements are significantly larger than the in-plane displacements[32,43] [Figs. 1(e) and 1(f)]. This is important because, at the applied electron energy of 100 eV, ULEED primarily probes the lighter sulfur atoms and is also more sensitive to out-of-plane changes due to diffraction in backscattering.[45]

In the following, we show the experimental results of the ultrafast structural NC-IC phase transition at the surface of 1*T*-TaS_2 employing ULEED. The recorded diffraction patterns in both phases exhibit bright main lattice peaks that are each surrounded by six first-order CDW superlattice peaks, labeled by $Q_{NC/IC}$ [Figs. 2(a) and 2(b)]. In the NC phase, also higher-order spots are visible pointing to a stronger and more anharmonic structural modulation.[46] By pumping the room-temperature phase using a fs-laser pulse (2.7 mJ/cm^2 fluence), NC peaks are strongly suppressed within a few picoseconds, while IC spots appear on the line connecting main lattice peaks corresponding to the unit cell rotation of approximately 12° [Figs. 2(a) and 2(b)]. Figure 2(c) shows in more detail the time traces for spot intensities and the diffuse background. The fast drop of NC spot intensity (blue line) is paralleled by a transient increase in the main lattice peak (black line), and a rapid increase in diffuse background (gray line) and IC spot intensity (red line). The increase in the main lattice peak intensity indicates the transient loss of CDW order driving the system toward the high-symmetry metallic phase.[12,16,27] For longer time delays, the main lattice intensity (black) mirrors the diffuse background curve

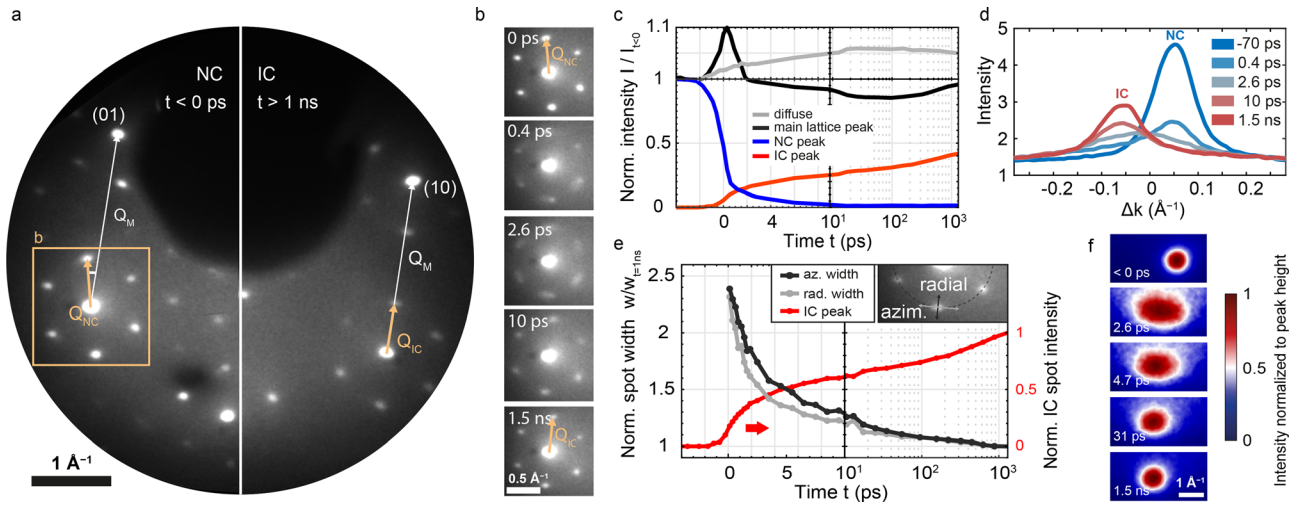

FIG. 2. Experimental results. (a) Measured diffraction pattern for NC (negative delay) and IC CDW phase (large positive delay). Five bright main lattice spots and PLD-induced superstructure spots are shown. The dark area corresponds to the electron gun shadow on the detector. The reciprocal vectors $Q_{NC/IC}$ (orange arrows) and Q_M (white arrows) indicate the structural change which is realized as a rotation of the superlattice by about 12°. (b) Selected time frames of inset in (a). (c) Normalized time-dependent peak intensities and diffuse background signal are shown (see methods in supplementary material). (d) Line profiles through NC and IC diffraction peak for different time delays. (e) Normalized azimuthal (black) and radial (gray) spot widths vs time. IC spot peak intensity (red) shown for comparison. (f) Snapshots of NC and IC peaks showing substantial broadening after excitation followed by peak narrowing.

(gray) which gives insight into the relaxation path of structural modes. Similar observations were made for the laser-induced structural dynamics starting from the IC state and in the absence of a phase transition.[16] The main lattice and the diffuse background show a trend reversal after approximately 100 ps indicating Debye–Waller-type suppression and subsequent cooling of the system. In contrast, the IC spot intensity (red) exhibits a continuous increase over the entire delay range of up to 1.5 ns. With the given repetition rate of 100 kHz, the system returns to its initial state within 10 s, the time between two pump cycles. Optically pumping the sample at this repetition rate results in a minor cumulative increase in the sample's base temperature by a few Kelvin. Figures 2(d) and 2(f) show the time-dependent spot profiles and cutouts for several time delays. IC spot analysis (see methods in the supplementary material) yields azimuthal (black) and radial (gray) widths [Fig. 2(e)]. Normalized to long delay times, the spot widths show a significant increase for early times, pointing to a substantial disorder in the system, followed by a steady narrowing over longer time scales. While this agrees with our previous observations,[22] we additionally find slightly different relaxations for the correlations in radial and azimuthal direction. This potentially points to an intermediate transient hexatic phase in the material which is defined by a specific translational and orientational order of the 2D CDW lattice. In impurity-doped $Nb_xTa_{1-x}S_2$, a hexatic phase emerges and leads to a significant azimuthal broadening in reciprocal space.[47,48]

In order to describe the dynamics in the incommensurate phase, we proposed a model[22] based on the kinetics of dislocation-like CDW defects.[25] After photoexcitation, a high density of phase singularities of the CDW order parameter leads to strong disorder and substantially broadened diffraction peaks. Incommensurate long-range order emerges over time as pairs of CDW defects annihilate. From pulse-to-pulse, the generation and annihilation process of CDW defects take different realizations precluding an exact microscopic reproducibility. The ensemble dynamics and transient changes of crystalline symmetry, however, are consistent and reversible. In the pump-probe configuration, over many pulses, we form the incoherent sum over many realizations, measuring a pattern with regular diffraction peaks and well-defined widths.

In conclusion, we applied our improved ULEED setup to the surface of 1*T*-TaS_2 in order to study the ultrafast structural dynamics of the NC-IC CDW phase transition. With high momentum resolution at the 1 ps time regime, diffraction spot analysis yields substantial broadening of IC peaks indicating strong disorder in the system after the phase transition. Referring to our previous study,[22] we attribute this to the kinetics of dislocation-like CDW defects. More generally, ULEED technique facilitates ultrafast structural probing of monolayers and superstructures and, due to the backscattering geometry, is particularly sensitive to out-of-plane displacements. In the future, the approach will allow for the investigation of a wide range of low-dimensional non-equilibrium phenomena, including topological phase transitions,[49] the formation of light-induced metastable phases,[50] and coherently controlled surface transformations.[29]

See the supplementary material for a methods section and a movie of the measured diffraction images showing the structural phase transition between the NC and IC CDW state and emerging broadening of CDW peaks.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

REFERENCES

[1]G. Grüner, *Density Waves in Solids*, Frontiers in Physics (Addison-Wesley Pub. Co., Advanced Book Program, Reading, MA, 1994), Vol. 89.

[2]A. W. Overhauser, "Observability of charge-density waves by neutron diffraction," Phys. Rev. B **3**, 3173 (1971).

[3]P. Lee, T. Rice, and P. Anderson, "Conductivity from charge or spin density waves," Solid State Commun. **14**, 703–709 (1974).

[4]W. L. McMillan, "Collective modes of a charge-density wave near the lock-in transition," Phys. Rev. B **16**, 4655–4658 (1977).

[5]G. F. Giuliani and A. W. Overhauser, "Structure factor of a charge-density wave," Phys. Rev. B **23**, 3737–3743 (1981).

[6]J. Demsar, K. Biljaković, and D. Mihailovic, "Single Particle and collective excitations in the one-dimensional charge density wave solid $K_{0.3}MoO_3$ probed in real time by femtosecond spectroscopy," Phys. Rev. Lett. **83**, 800–803 (1999).

[7]S. Hellmann, M. Beye, C. Sohrt, T. Rohwer, F. Sorgenfrei, H. Redlin, M. Kalläne, M. Marczynski-Bühlow, F. Hennies, M. Bauer, A. Föhlisch, L. Kipp, W. Wurth, and K. Rossnagel, "Ultrafast melting of a charge-density wave in the Mott insulator 1*T*-TaS_2," Phys. Rev. Lett. **105**, 187401 (2010).

[8]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*-TaS_2 by time resolved photoelectron spectroscopy," New J. Phys. **10**, 053019 (2008).

[9]H. Y. Liu, I. Gierz, J. C. Petersen, S. Kaiser, A. Simoncig, A. L. Cavalieri, C. Cacho, I. C. E. Turcu, E. Springate, F. Frassetto, L. Poletto, S. S. Dhesi, Z.-A. Xu, T. Cuk, R. Merlin, and A. Cavalleri, "Possible observation of parametrically amplified coherent phasons in $K_{0.3}MoO_3$ using time-resolved extreme-ultraviolet angle-resolved photoemission spectroscopy," Phys. Rev. B **88**, 045104 (2013).

[10]J. C. Petersen, S. Kaiser, N. Dean, A. Simoncig, H. Y. Liu, A. L. Cavalieri, C. Cacho, I. C. E. Turcu, E. Springate, F. Frassetto, L. Poletto, S. S. Dhesi, H. Berger, and A. Cavalleri, "Clocking the melting transition of charge and lattice order in 1*T*-TaS_2 with ultrafast extreme-ultraviolet angle-resolved photoemission spectroscopy," Phys. Rev. Lett. **107**, 177402 (2011).

[11]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," Phys. Rev. Lett. **97**, 067402 (2006).

[12]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**, 799–802 (2010).

[13]N. Erasmus, M. Eichberger, K. Haupt, I. Boshoff, G. Kassier, R. Birmurske, H. Berger, J. Demsar, and H. Schwoerer, "Ultrafast dynamics of charge density waves in 4H^b-$TaSe_2$ probed by femtosecond electron diffraction," Phys. Rev. Lett. **109**, 167402 (2012).

[14]C. Laulhé, L. Cario, B. Corraze, E. Janod, T. Huber, G. Lantz, S. Boulfaat, A. Ferrer, S. Mariager, J. Johnson, S. Grübel, A. Lübcke, G. Ingold, P. Beaud, S. Johnson, and S. Ravy, "X-ray study of femtosecond structural dynamics in the 2D charge density wave compound 1*T*-TaS_2," Physica B **460**, 100–104 (2015).

[15]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," Phys. Rev. B **86**, 075145 (2012).

[16]G. Storeck, J. G. Horstmann, T. Diekmann, S. Vogelgesang, G. von Witte, S. V. Yalunin, K. Rossnagel, and C. Ropers, "Structural dynamics of incommensurate charge-density waves tracked by ultrafast low-energy electron diffraction," Struct. Dyn. **7**, 034304 (2020).

[17]C. Laulhé, T. Huber, G. Lantz, A. Ferrer, S. Mariager, S. Grübel, J. Rittmann, J. Johnson, V. Esposito, A. Lübcke, L. Huber, M. Kubli, M. Savoini, V. Jacques, L. Cario, B. Corraze, E. Janod, G. Ingold, P. Beaud, S. Johnson, and S. Ravy, "Ultrafast formation of a charge density wave state in 1*T*-TaS_2: Observation at nanometer scales using time-resolved x-ray diffraction," Phys. Rev. Lett. **118**, 247401 (2017).

[18]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**, 821–825 (2018).

[19]X. Shi, W. You, Y. Zhang, Z. Tao, P. M. Oppeneer, X. Wu, R. Thomale, K. Rossnagel, M. Bauer, H. Kapteyn, and M. Murnane, "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).

[20]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," Sci. Adv. **1**, e1400173 (2015).

[21]K. Haupt, M. Eichberger, N. Erasmus, A. Rohwer, J. Demsar, K. Rossnagel, and H. Schwoerer, "Ultrafast metamorphosis of a complex charge-density wave," Phys. Rev. Lett. **116**, 016402 (2016).

[22]S. Vogelgesang, G. Storeck, J. G. Horstmann, T. Diekmann, M. Sivis, S. Schramm, K. Rossnagel, S. Schäfer, and C. Ropers, "Phase ordering of charge density waves traced by ultrafast low-energy electron diffraction," Nat. Phys. **14**, 184–190 (2018).

[23]T. Danz, T. Domröse, and C. Ropers, "Ultrafast nanoimaging of the order parameter in a structural phase transition," Science **371**, 371–374 (2021).

[24]W. L. McMillan, "Landau theory of charge-density waves in transition-metal dichalcogenides," Phys. Rev. B **12**, 1187–1196 (1975).

[25]W. L. McMillan, "Theory of discommensurations and the commensurate-incommensurate charge-density-wave phase transition," Phys. Rev. B **14**, 1496–1502 (1976).

[26]W. Lee, Y. Chuang, R. Moore, Y. Zhu, L. Patthey, M. Trigo, D. Lu, P. Kirchmann, O. Krupin, M. Yi, M. Langner, N. Huse, J. Robinson, Y. Chen, S. Zhou, G. Coslovich, B. Huber, D. Reis, R. Kaindl, R. Schoenlein, D. Doering, P. Denes, W. Schlotter, J. Turner, S. Johnson, M. Först, T. Sasagawa, Y. Kung, A. Sorini, A. Kemper, B. Moritz, T. Devereaux, D.-H. Lee, Z. Shen, and Z. Hussain, "Phase fluctuations and the absence of topological defects in a photo-excited charge-ordered nickelate," Nat. Commun. **3**, 838 (2012).

[27]A. Zong, A. Kogar, Y.-Q. Bie, T. Rohwer, C. Lee, E. Baldini, E. Ergeçen, M. B. Yilmaz, B. Freelon, E. J. Sie, H. Zhou, J. Straquadine, P. Walmsley, P. E. Dolgirev, A. V. Rozhkov, I. R. Fisher, P. Jarillo-Herrero, B. V. Fine, and N. Gedik, "Evidence for topological defects in a photoinduced phase transition," Nat. Phys. **15**, 27–31 (2019).

[28]B. Wit, O. Bunjes, M. Wenderoth, and C. Ropers, "Structure and nonequilibrium heat-transfer of a physisorbed molecular layer on graphene," Adv. Mater. Interfaces **7**, 2000473 (2020).

[29]J. G. Horstmann, H. Böckmann, B. Wit, F. urtz, G. Storeck, and C. Ropers, "Coherent control of a surface structural phase transition," Nature **583**, 232–236 (2020).

[30]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**, 200–204 (2014).

[31]G. Storeck, S. Vogelgesang, M. Sivis, S. Schäfer, and C. Ropers, "Nanotip-based photoelectron microgun for ultrafast LEED," Struct. Dyn. **4**, 044024 (2017).

[32]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 1*T*-TaS_2 in $(3+2)$-dimensional superspace," Phys. Rev. B **56**, 13757–13767 (1997).

[33]K. Rossnagel, "On the origin of charge-density waves in select layered transition-metal dichalcogenides," J. Phys.: Condens. Matter **23**, 213001 (2011).

[34]C. B. Scruby, P. M. Williams, and G. S. Parry, "The role of charge density waves in structural transformations of 1*T*-TaS_2," Philos. Mag. **31**, 255–274 (1975).

[35]J. Wilson, F. D. Salvo, and S. Mahajan, "Charge-density waves and superlattices in the metallic layered transition metal dichalcogenides," Adv. Phys. **24**, 117–201 (1975).

[36]P. Fazekas and E. Tosatti, "Electrical, structural and magnetic properties of pure and doped 1*T*-TaS_2," Philos. Mag. B **39**, 229–244 (1979).

[37]P. Fazekas and E. Tosatti, "Charge carrier localization in pure and doped 1*T*-TaS2," Physica B **99**, 183–187 (1980).

[38]B. Sipos, A. F. Kusmartseva, A. Akrap, H. Berger, L. Forró, and E. Tutiš, "From Mott state to superconductivity in 1*T*-TaS_2," Nat. Mater. **7**, 960–965 (2008).

[39]T. Ritschel, J. Trinckauf, K. Koepernik, 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," Nat. Phys. **11**, 328–331 (2015).

[40]Y. Chen, W. Ruan, M. Wu, S. Tang, H. Ryu, H.-Z. Tsai, R. Lee, S. Kahn, F. Liou, C. Jia, O. R. Albertini, H. Xiong, T. Jia, Z. Liu, J. A. Sobota, A. Y. Liu, J. E. Moore, Z.-X. Shen, S. G. Louie, S.-K. Mo, and M. F. Crommie, "Strong correlations and orbital texture in single-layer 1*T*-$TaSe_2$," Nat. Phys. **16**, 218–224 (2020).
[41]K. Nakanishi and H. Shiba, "Theory of three-dimensional orderings of charge-density waves in 1*T*-TaX_2 (X: S, Se)," J. Phys. Soc. Jpn. **53**, 1103–1113 (1984).
[42]T. Ritschel, H. Berger, and J. Geck, "Stacking-driven gap formation in layered 1*T*-TaS_2," Phys. Rev. B **98**, 195134 (2018).
[43]G. von Witte, T. Kißlinger, J. G. Horstmann, K. Rossnagel, M. A. Schneider, C. Ropers, and L. Hammer, "Surface structure and stacking of the commensurate (13x13)R13.9 charge density wave phase of 1*T*-TaS_2 (0001)," Phys. Rev. B **100**, 155407 (2019).
[44]R. E. Thomson, B. Burk, A. Zettl, and J. Clarke, "Scanning tunneling microscopy of the charge-density-wave structure in 1*T*-TaS_2," Phys. Rev. B **49**, 16899 (1994).
[45]M. A. Van Hove, W. H. Weinberg, and C.-M. Chan, *Low-Energy Electron Diffraction: Experiment, Theory and Surface Structure Determination, Softcover Reprint of the Hardcover*, Springer Series in Surface Sciences (Springer, Berlin, 1986).
[46]K. Nakanishi and H. Shiba, "Domain-like incommensurate charge-density-wave states and the first-order incommensurate-commensurate transitions in layered tantalum dichalcogenides. I. 1*T*-polytype," J. Phys. Soc. Jpn. **43**, 1839–1847 (1977).
[47]H. Dai and C. M. Lieber, "Solid-hexatic-liquid phases in two-dimensional charge-density waves," Phys. Rev. Lett. **69**, 1576–1579 (1992).
[48]I. R. Peterson and V. M. Kaganer, "Diffraction line profile of a two-dimensional hexatic," Phys. Rev. Lett. **73**, 102–105 (1994).
[49]J. M. Kosterlitz, "Ordering, metastability and phase transitions in two-dimensional systems," J. Phys. C **6**, 1181–1203 (1973).
[50]L. Stojchevska, 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**, 177–180 (2014).