

Ultrafast electronic structure engineering in 1T-TaS₂: Role of doping and amplitude mode dynamics

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In strongly correlated transition metal dichalcogenides, an intricate interplay of polaronic distortions, stacking arrangement, and electronic correlations determines the nature of the insulating state. Transient changes in pristine and Mo-doped 1T-TaS₂ are measured by femtosecond time-resolved photoelectron spectroscopy and modeled using nonequilibrium dynamical mean-field theory complemented by density functional theory. Inspired by vibrational spectroscopy we analyze real-time oscillatory spectral changes induced by the charge density wave amplitude mode through modulations of electronic interactions. We distinguish effects induced by electron doping and structural disorder which can modify the layer stacking and conclude that doped crystals might feature an enhanced fraction of monolayers. Our work establishes substitutional doping as a pathway to engineer the dynamic electronic structure in correlated transition metal dichalcogenides.

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I. INTRODUCTION

Many-body effects in strongly correlated materials give rise to density waves, superconductivity, Mott insulating ground states, and interesting nonequilibrium phenomena [1–5]. A much-studied group V transition metal dichalcogenide is 1T-TaS₂, which is an undistorted metal at temperatures T above 540 K. As T is lowered, a charge density wave (CDW) is formed with increasing long-range order. The CDW consists of individual Star-of-David shaped entities formed out of 13 Ta atoms each [2,6,7]. At $T < 350$ K these stars form clusters and a nearly commensurate CDW (NC-CDW). Below 200 K a long-range ordered, commensurate CDW develops into a $(\sqrt{13} \times \sqrt{13})R13.9^\circ$ superstructure of the Ta atoms. In recent years, the early proposal of a Mott insulating ground state for 1T-TaS₂ by Fazekas and Tosatti [6] has been challenged. The stacking of individual layers with identical atomic structure was shown to be essential to understand the insulating character [8–10]. Surface and bulk contributions were considered [11–13], bilayer formation and Mott localization were disentangled, and it was shown that

Mott insulating monolayers make up only about 20% of the bulk [5,14]. Substitutional [15,16] and hole doping [17] have been investigated and coexisting metallic and insulating domains were reported. On this basis, 1T-TaS₂ has become an interesting material platform to study intertwined electronic and vibrational excitations [7,18–24]. Despite considerable efforts [12,25–27], the electronic structure changes associated with the CDW amplitude mode [7,12,22,28] are, so far, not fully understood.

In this work, we model the transient electronic structure change of the amplitude mode in 1T-TaS₂ in real time and distinguish the response of mono- and bilayers in the pristine and the electron-doped compound. Time-resolved photoemission spectra compare well with the developed theory. Our results establish a coherent real-time observable of the dynamic electronic structure in strongly correlated two-dimensional materials that provides additional insights into the stacking of mono- and bilayers.

II. SAMPLE PREPARATION AND CHARACTERIZATION

Single crystals of 1T-Ta_(1-x)Mo_xS₂ with chosen dopant concentrations $x = 0.00, 0.02, 0.04, 0.06$ were grown using iodine vapor transport. High-purity tantalum and molybdenum wires, as well as sulfur powder in a near-stoichiometric ratio (with a 10% excess of sulfur, or 1 mg/cm³), were placed at one end of evacuated quartz ampoules with an inner diameter of 12 mm and a length of 200 mm. Then, 5 mg/cm³

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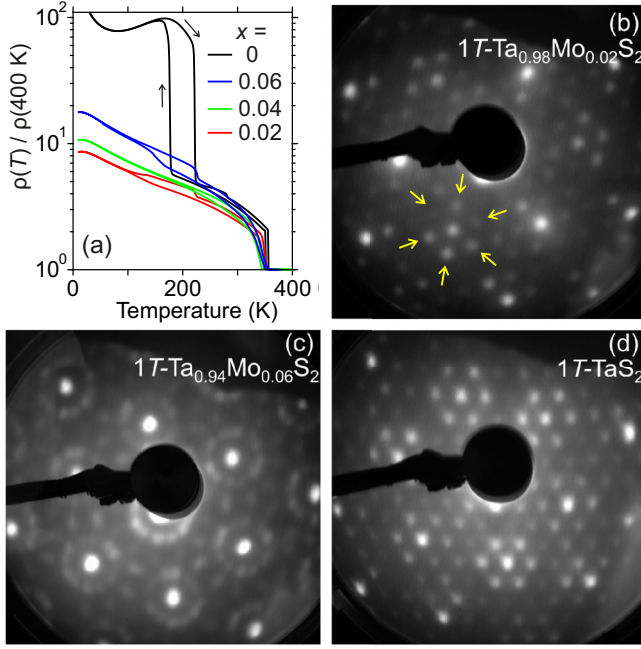


FIG. 1. (a) Temperature-dependent electrical resistivity ρ of $1T\text{-Ta}_{(1-x)}\text{Mo}_x\text{S}_2$ normalized to the value at 400 K at the indicated doping level x . (b)–(d) LEED images for $x = 0.02$ (b), 0.06 (c), and the parent compound $x = 0$ (d), recorded at $T = 30$ K with an electron kinetic energy of 123 eV at crystal surfaces obtained by cleaving at room temperature. For $x = 0.04$, see Supplemental Material [29].

of iodine granules were added. The ampoules were sealed and subjected to a 100°C temperature gradient in two growth phases. The hotter end, containing the starting materials, was maintained at 970°C . Crystals formed near the colder end over 120 h in the first phase and 648 h in the second. After both growth phases, the ampoules were rapidly quenched in cold water from the growth temperature to preserve the metastable $1T$ polymorph. To characterize the different CDW phases in the doped crystals, we performed electrical resistivity and low-energy electron diffraction (LEED) measurements. Figure 1(a) shows the temperature-dependent electric resistivity measured for single crystals of $1T\text{-Ta}_{(1-x)}\text{Mo}_x\text{S}_2$. The transition to the NCCDW phase prevails for all x , while the long-range order below $T = 200$ K is strongly suppressed upon doping. The satellite features observed in LEED [see Figs. 1(b)–1(d)] indicate that locally the Star-of-David superstructure remains, although at reduced intensity. Thus, Mo doping reduces the fraction containing the superstructure. For $x = 0.06$ we identify more complex satellite features indicating two domains rotated with respect to each other by $30^\circ \pm 4^\circ$. From these results we conclude that electric conductivity channels persist in the presence of Mo doping at all temperatures. We attribute this double-domain formation to the existence of two equivalent domains rotated clockwise and anticlockwise by $\pm 13.9^\circ$ from the unperturbed crystallographic axes.

In order to study the homogeneity of the doped crystals, spatially and angle-resolved photoelectron spectroscopy was conducted using the extreme ultraviolet beamline AU-SGM4

at the ASTRID2 synchrotron. For details of the instrumentation, see Ref. [30]. In Fig. 2, we present spatially resolved angle-resolved photoemission spectroscopy (ARPES) measurements over a large area of a 6% Mo-doped $1T\text{-TaS}_2$ sample using a step size of $25\ \mu\text{m}$. The raw variations in the Ta $4f$ core-level intensity across the sample measured with photon energy 80 eV are shown in Fig. 2(a). Normalized core-level spectra extracted from three distinct regions indicated by colored areas in Fig. 2(a) are plotted in the top of Fig. 2(b), demonstrating no significant variations across the sample surface. To further confirm the uniformity of the sample, we investigated variations in the valence band electronic structure at different positions marked by colored symbols in Fig. 2(a), using a photon energy of 50 eV. High statistics measurements of the valence band spectra at normal emission, represented by energy distribution curves (EDCs) at the Γ point using a $0.06\ \text{\AA}^{-1}$ integration range [see bottom panel of Fig. 2(b)], indicate that observed spectral differences arise primarily due to slight angular offsets between measurement positions. Additionally, the homogeneity was evaluated by fitting the Ta $4f$ core-level spectra at each spatial point of the map using four peaks convoluted with a Gaussian function and a linear background. The resulting spatial variation of the binding energy for the third peak component is indicated by the gray arrow in Fig. 2(b) and is plotted in Fig. 2(c), revealing only minor variations in the region near the surface of the cleaved crystal.

In Fig. 3 we present the equilibrium ARPES data of a crystal with $x = 0.06$ measured below the CDW transition temperature at the SGM4 end station of Astrid II in Aarhus, Denmark [30]. For this measurement, the sample was cleaved *in situ* at room temperature and a pressure of 2×10^{-8} mbar before being transferred into the main chamber. The pressure during the experiment was 2×10^{-10} mbar.

The dispersions along two high-symmetry directions, shown in Fig. 3, were extracted from the same high-resolution angle scan. The EDCs through the Γ point are plotted next to the corresponding spectra to highlight the band features at a binding energy of approximately $E - E_F = -0.5$ eV. Crucially, no significant deviations are observed when comparing the data to that of a pristine $1T\text{-TaS}_2$ sample. Such data were reported previously (see, e.g., Refs. [22,31]). Based on these results, we found homogeneous crystal surfaces without dopant agglomeration and the overall electronic band structure does not vary with doping compared to the pristine crystal. Spectral response functions are, however, expected to reveal the effect of doping.

III. RESULTS AND DISCUSSION

A. Experiment

In the femtosecond time-resolved ARPES experiments, performed in Duisburg, femtosecond laser pulses with a photon energy of $h\nu = 1.51$ eV and 50 fs pulse duration were used for pumping, combined with probing at 6.04 eV at delay time t [32]. The incident pump fluence was $0.2\ \text{mJ cm}^{-2}$. The crystals were cleaved at $T = 300$ K *in situ* at $p = 2 \times 10^{-10}$ mbar. All measurements were performed at 30 K using p -polarized light pulses. The kinetic energy of the

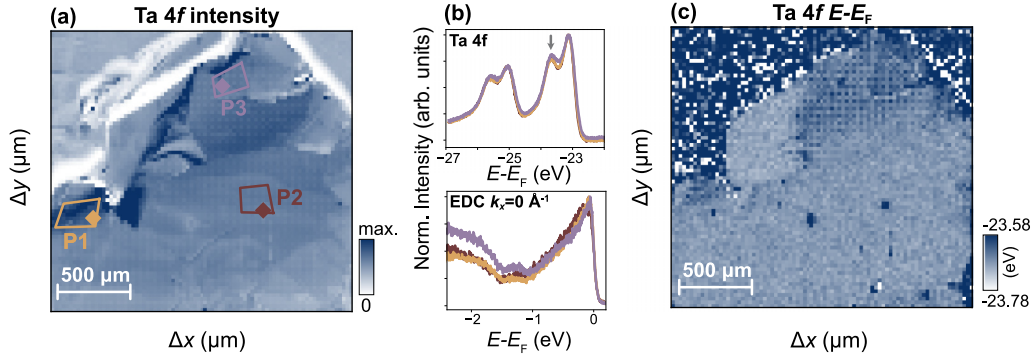


FIG. 2. (a) Spatially resolved ARPES measurements of $1T\text{-Ta}_{0.94}\text{Mo}_{0.06}\text{S}_2$ at room temperature scanning over the Ta 4*f* core levels using a photon energy of $h\nu = 80 \text{ eV}$ and linear horizontal-polarized light. (b) Top: Extracted core levels from the map shown in (a), integrated over small regions around three different probing spots. Bottom: Extracted EDCs at the Gamma point ($k_x = 0 \text{ \AA}^{-1}$) from fixed scans measured at the same three positions, indicated by the colored markers in (a) using a photon energy of $h\nu = 50 \text{ eV}$. (c) Variations in the Ta 4*f* binding energy of the third peak component indicated by the arrow in (b), showing only small variations in the binding energy within the topmost part of the sample.

photoelectrons emitted within $\pm 3^\circ$ collection angle along the surface normal direction was measured by electron time-of-flight spectroscopy [33]. The Fermi level E_F was determined by the high-energy spectral cutoff without pumping at 300 K [7].

Time-dependent photoelectron emission spectra were taken for $1T\text{-Ta}_{(1-x)}\text{Mo}_x\text{S}_2$ with $x = 0.00, 0.02, 0.04$, and 0.06 . The results, which represent the central experimental finding of this work, are shown in Fig. 4. Panel (a) shows the intensity for $x = 0.02$ as a function of energy $E - E_F$ and t . The spectrum at $t < 0$ before excitation is 0.6 eV wide. At $t = 0$ a slight intensity reduction below E_F and a clear increase above E_F represents the strong nonequilibrium response to the optical excitation. At delays $0.4 < t < 0.5 \text{ ps}$, oscillations in the vicinity of E_F are observed. We find a single frequency of $\Omega = 2.40 \pm 0.05 \text{ THz}$ and dephasing or damping during 5 ps. This frequency represents the breathing of the Star-of-David and the CDW amplitude mode [7,18,34].

We observe transient spectral changes with characteristic differences for the parent and the electron-doped compound

and focus on the amplitude mode response at $t > 0.4 \text{ ps}$. The early nonequilibrium dynamics has been studied before [7,22,24]. For $x = 0$ the oscillatory changes extend over 0.5 eV with rich details. For $x = 0.02$ the oscillatory changes are confined to 100 meV around E_F for $t > 0.4 \text{ ps}$. The transient spectrum of the $x = 0.06$ sample, shown in Fig. 4(d), closely resembles that of the $x = 0.02$ sample. The characteristic features are (1) a spectral width of 0.6 eV observed before excitation ($t < 0$), (2) a similar electronic response to optical excitation close to $t = 0$, and (3) the frequency of the CDW-amplitude mode. However, the oscillatory changes are in this case strongly confined close to E_F . Comparing ΔI_{PES} of the samples $x = 0.00$, $x = 0.02$, and $x = 0.06$ establishes that doping by Mo confines the oscillatory changes to a narrow range around E_F .

B. Theory

We combine density functional theory (DFT) calculations with nonequilibrium dynamical mean-field theory (DMFT)

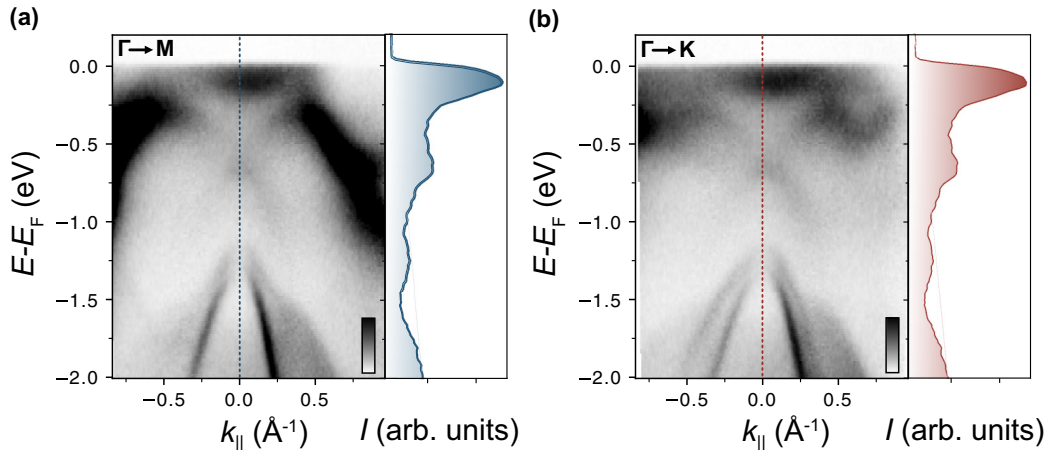


FIG. 3. Equilibrium ARPES measured for $1T\text{-Ta}_{0.94}\text{Mo}_{0.06}\text{S}_2$ at 45 K using $h\nu = 50 \text{ eV}$ along (a) the $\Gamma\text{-}M$ and (b) the $\Gamma\text{-}K$ high-symmetry directions. The corresponding EDCs extracted at the Γ point ($k_{||} = 0 \text{ \AA}^{-1}$, indicated by dashed lines) over an integration range of $0.05 \text{ \AA}^{-1}$ are plotted next to the data.

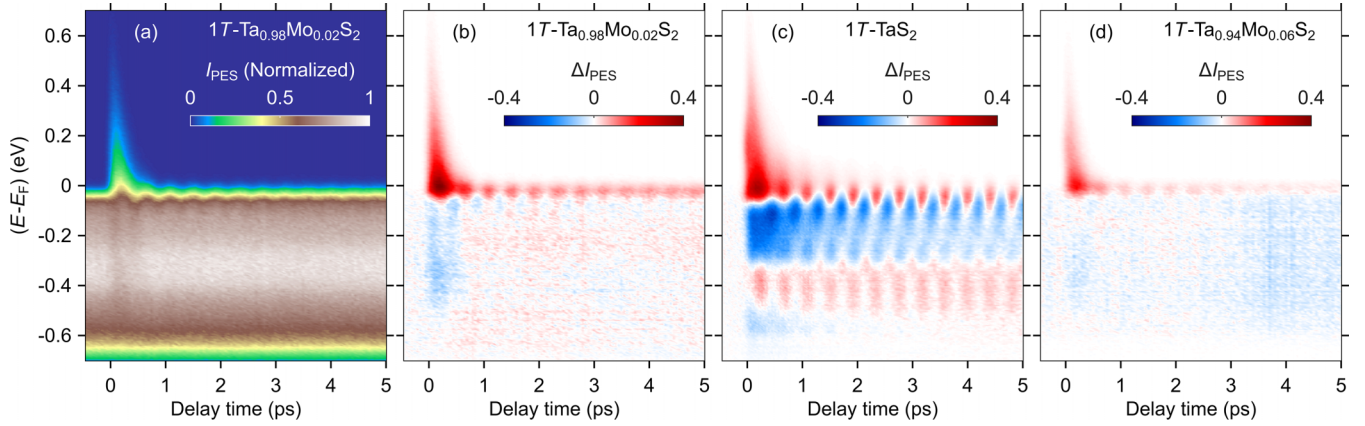


FIG. 4. Time-dependent photoelectron intensity I_{PES} as a function of energy $E - E_F$ and delay time t . (a) for $T = 30$ K and $x = 0.02$ after normalization to the intensity maximum using a false color map. (b)–(d) pump-induced change for $x = 0.02, 0$, and 0.06 , respectively. Panel (b) shows the differential spectrum after subtracting the static spectrum ($t < 0$) from the dataset shown in panel (a). Positive and negative changes are indicated in red and blue, respectively, in (b)–(d). For $x = 0.06$ the results are similar to $x = 0.02$. For $x = 0$, full time-resolved spectra were previously published in Refs. [7,12,28].

simulations to describe how the amplitude mode changes the electronic structure in real time.

1. DFT calculations

A relevant question is the distinction between the amplitude mode response of monolayers and bilayers of Star-of-David clusters [see illustration in Fig. 5(b)].

DFT calculations were performed with QUANTUM ESPRESSO [35] using the Perdew-Burke-Ernzerhof approximation for the exchange-correlation potential [36] and norm-conserving pseudopotentials [37]. Plane waves were included until an energy cutoff of 1360 eV and a

Fermi-Dirac-type smearing of 68 meV was imposed. We used a lattice constant of 3.43 Å and a $6 \times 6 \times 1$ k-point grid for the $\sqrt{13} \times \sqrt{13}$ supercell. A vacuum of approximately 12 Å was included above the monolayer/bilayer. For calculations of the doped system, we considered a doping of 0.02/0.04/0.06 e^- per TaS₂ unit cell, which corresponds to 0.26/0.52/0.78 e^- per $\sqrt{13} \times \sqrt{13}$ supercell.

We compare modulations of the electronic structure for a monolayer and a bilayer of 1T-TaS₂ [see Figs. 6(d)–6(f) and 6(g)–6(i)], respectively. For monolayers (bilayers), the emergence of one (two) flat bands near E_F is found upon approaching the relaxed CDW structure characterized by a distortion $\alpha = 1$. Here, α represents the scaled CDW distortion with the relaxed CDW structure corresponding to $\alpha = 1$ and the high-temperature structure without CDW to $\alpha = 0$. In the case of the bilayer, the two flat bands near E_F are split by a bonding-antibonding splitting due to interlayer hopping, which dominates over the in-plane kinetic energy contributions of the flat band. Upon reducing the CDW displacement amplitude by 40%, corresponding to $\alpha = 0.6$, the flat bands become more dispersive in both cases and most strikingly, the bonding-antibonding splitting in the bilayer case is reduced.

As visualized in Fig. 5(a) for different α , the interlayer hopping in a bilayer is one order of magnitude larger than in-plane kinetic energy contributions within a monolayer. This finding motivates the modulation of the interlayer hopping terms in the subsequent many-body treatment to mimic the dynamic effect of the Ta ion core displacement on the electronic structure.

2. DMFT calculations

To model the transient electronic structure in the DMFT calculations, we use a single-orbital multi-layer description of 1T-TaS₂ in the CDW phase [38,39] supplemented with a local Hubbard repulsion to incorporate electronic correlations. To describe the stacking order, the neighboring layers are connected via two hopping amplitudes: $h^A = 0.2$ eV for the intrabilayer hybridization and $h^L = 0.045$ eV for the hopping between two van der Waals bonded layers [38,39] [see

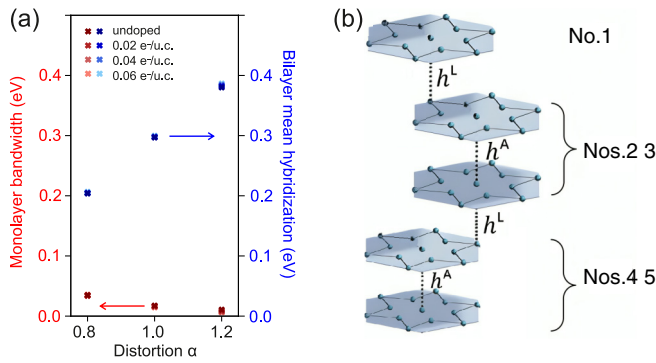


FIG. 5. (a) Representative energy scales for mono- and bilayers of 1T-TaS₂ calculated by DFT for the indicated doping levels. Left axis: Bandwidth for the monolayer single band as a function of the distortion α to simulate the influence of the breathing mode on the intralayer physics. Right axis: Calculated mean hybridization gap of the bilayer band structure for identical α representing the effect of the breathing mode on the interlayer physics. The interlayer energy scale is one order of magnitude larger than the intralayer one. (b) Stacking of the different planes are indicated by adjacent Star-of-David clusters. A monolayer (No. 1) is located on top with two bilayers (Nos. 2 3 and Nos. 4 5), which represent the bulk. Two matrix elements describe hopping along the c axis: h^A for hopping within a bilayer and h^L for hopping two adjacent clusters are considered.

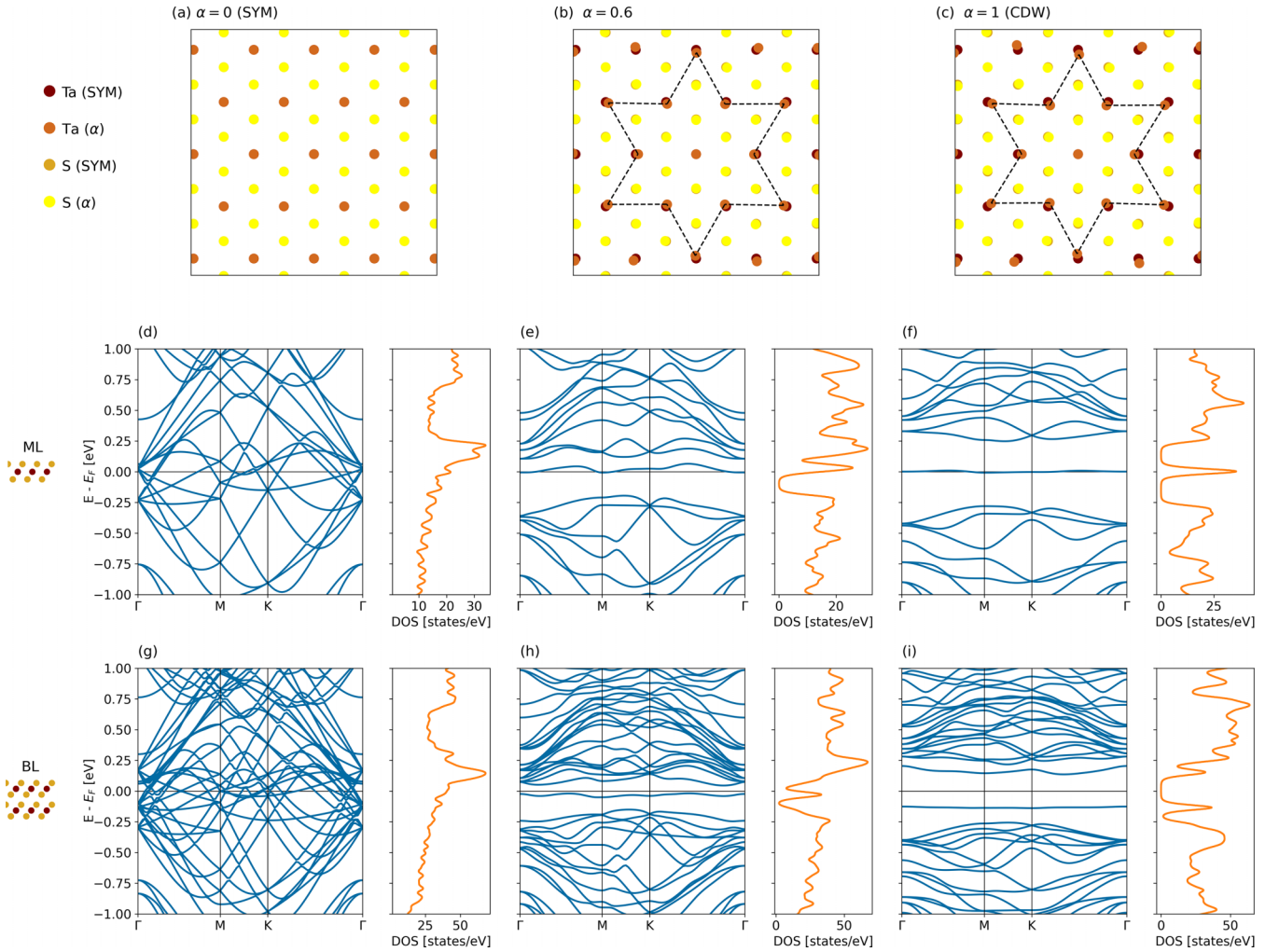


FIG. 6. Results of DFT calculations for monolayer and bilayer 1T-TaS₂. (a)–(c) Zoom into the $\sqrt{13} \times \sqrt{13}$ supercell with a lattice constant of 3.43 Å for different percentages of displacement towards the distortion in the Star-of-David, from (a) the symmetric phase $\alpha = 0$ over (b) $\alpha = 0.6$ to (c) the Star-of-David CDW $\alpha = 1$. As a guide to the eye, the star shape of the CDW is marked via dashed lines. (d)–(f) Noninteracting band structure (blue) and density of states (DOS; orange) of the 1T-TaS₂ monolayer obtained via DFT for the different percentages of distortion α from (a)–(c). The emergence of a single band around the Fermi level can be seen. (g)–(i) Noninteracting band structure (blue) and DOS (orange) of the 1T-TaS₂ bilayer obtained via DFT for the different percentages of distortion α from (a)–(c).

Fig. 5(b)]. The kinetic part of the Hamiltonian matrix is

$$\mathcal{H}_{ab}(\mathbf{R}) = \mathcal{H}_{\text{SOD}}(\mathbf{R})\delta_{ab} - h_{ab}^A\delta_{\mathbf{R},\mathbf{0}} - h_{ab}^L(\mathbf{R}), \quad (1)$$

where $\{a, b\}$ is a layer index, $\mathcal{H}_{\text{SOD}}$ is the Hamiltonian of the Star-of-David, and $\mathbf{R}$ is the unit-cell vector. To study the time evolution after a photoexcitation event, we use the nonequilibrium generalization of DMFT [40] in combination with a noncrossing approximation [41,42] impurity solver. A simplified Bethe-lattice type self-consistency allows one to analyze the essential correlation and hybridization effects by constructing an approximate real-space hybridization function [43–46].

To capture the bilayer hybridization h^A , we employ a two-site cluster DMFT [47]. The two sites of a cluster represent the two planes of a bilayer. The local Hamiltonian for this cluster DMFT description of the bilayer has the form

$$\begin{aligned} \mathcal{H}_i(t) = & -\mu(n_{i1} + n_{i2}) + U(n_{i1\uparrow}n_{i1\downarrow} + n_{i2\uparrow}n_{i2\downarrow}) \\ & - \sum_{\sigma} [h_{12}^i(t)c_{i1\sigma}^{\dagger}c_{i2\sigma} + \text{H.c.}], \end{aligned} \quad (2)$$

where U is the local Hubbard interaction and μ is the chemical potential, which are adjusted to realize the desired doping.

Upon laser excitation, the hopping matrices become time dependent and complex $h_{mn}(t) = h_{mn}(0)e^{i\phi_{mn}(t)}$. The modeling of the time dependence and the self-consistency of DMFT is described in the Supplemental Material [29]. In essence, the Peierls phase $\phi_{mn}(t) = -\int_0^t dt' \mathbf{E}(t') \cdot \mathbf{r}_{mn}$ is determined by the time integral of the vector product of the electric laser field $\mathbf{E}(t)$ and the displacement $\mathbf{r}_{mn}$ between the centers of orbitals m and n [40]. The chosen laser field is resonant with the local Hubbard repulsion U . Its duration is 10 fs. This differs from the experimental situation that employs a 1.5 eV pump photon energy for excitations of sulfur-derived states [31]. Consequently, the induced nonequilibrium state might vary regarding the electronic excitations during the early dynamics between experiment and theory. However, the amplitude mode dynamics is, in both approaches, triggered by the sudden excitation shorter than the duration of half the

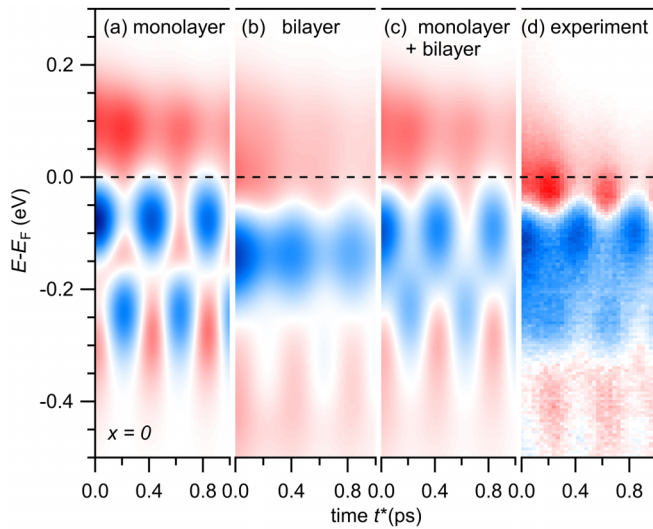


FIG. 7. False color representation of the calculated (a),(b) and measured (d) transient spectral changes for the monolayer at the surface (No. 1) (a) and the first bilayer (Nos. 2 3) (b) for the pristine crystal ($x = 0$). Panel (c) depicts the sum of the monolayer and the bilayer contribution. The time interval $t^* > 0$ focuses on the oscillatory behavior upon relaxation of the electronic nonequilibrium. Experimental results are replotted from Fig. 4 with $t^* = 0$ after one amplitude mode period has elapsed, i.e., at delay time of 450 fs. Theoretical results are plotted with $t^* = 0$ after a delay time of 160 fs. The color map of Figs. 4(b) and 4(c) is used. The horizontal dashed line indicates the Fermi energy.

amplitude mode period of 208 fs. In the following discussion we will, therefore, focus on the oscillatory spectral response.

C. Discussion

In Figs. 7 and 8, which present the main combined theoretical and experimental results of this work, we plot the oscillatory response in time intervals t^* that start once the hot-electron signature has relaxed and the oscillatory response dominates, for the pristine ($x = 0$) and the doped ($x = 0.02$) case, respectively. Panels (a) show the calculated time-dependent changes of the spectral function for the monolayer No. 1 at the surface. Panels (b) show the result for the first bilayer Nos. 2 3, while panels (c) depict the average of the mono- and bilayer responses. For the pristine compound, this average reproduces the experimental finding reasonably well [see Fig. 7(d)], and in particular better than the individual mono- or bilayer calculations.

The results for $x = 0.02$, Fig. 8, demonstrate that upon doping the amplitude mode persists in both experiment and theory. However, the behavior differs significantly from the pristine case. The calculated response for the monolayer is most pronounced near E_F , where it is very weak for the bilayer, while at lower $E - E_F \approx -0.3$ eV the monolayer and bilayer response is similar. The experiment, on the other hand, detects an oscillating response exclusively near E_F .

Considering that photoemission spectroscopy is surface sensitive, a monolayer and bilayer surface termination could be distinguished in experiments. Since for $x = 0$ a mixture of mono- and bilayer response reproduces the experiment better

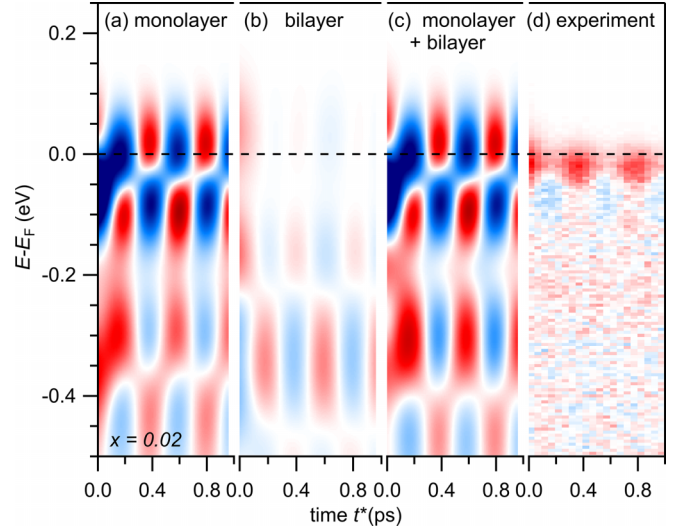


FIG. 8. False color representation of the calculated (a),(b) and measured (d) transient spectral changes for the monolayer at the surface (No. 1) (a) and the first bilayer (Nos. 2 3) (b) for the substituted crystal ($x = 0.02$). Panel (c) depicts the sum of the monolayer and the bilayer contribution. Experimental results are replotted from Fig. 4 with $t^* = 0$ after a delay time of 690 fs. Theoretical results are plotted with $t^* = 0$ after a delay time of 200 fs. The color map of Figs. 4(b) and 4(c) is used. The horizontal dashed line indicates the Fermi energy.

than pure mono- or bilayers [cf. Figs. 7(c) and 7(d)], we conclude that the cleaved crystal surface probed in time-resolved angle-resolved photoelectron spectroscopy (trARPES) contains both mono- and bilayer contributions.

Adding valence electrons by Mo doping allows one to fill parent holes in the pristine crystal [2,31]. The observed spectral change induced by the amplitude mode in the doped system is similar to results obtained in the nearly commensurate phase of the pristine crystal reported in Ref. [7]. Near $T = 350$ K the electrical resistivity of doped crystals exhibits the abrupt change associated with the onset of the NCCDW phase for pristine crystals [see Fig. 1(a)]. This finding indicates—in agreement with our LEED results (Fig. 1)—that Star-of-David clusters persist upon doping, while long-range order in the commensurate CDW is suppressed. It is, therefore, plausible that at $T = 30$ K Mo-doped crystals behave similar to pristine crystals in their NCCDW phase.

In Fig. 8, our calculations find an oscillatory response over a wide energy range, from E_F to $E - E_F = -0.5$ eV, whereas the experimental response is largely constrained to E_F . This difference between experiment and theory for the doped crystal might be associated with disorder. In the LEED images (Fig. 1 and Supplemental Material [29]) we recognize increasing disorder with x . Due to technical limitations the calculations do not take disorder into account. Disorder has likely two effects. Following the electric transport measurements, we conclude that doped crystals conduct currents similar to pristine crystals in the NCCDW phase in channels at boundaries of ordered Star-of-David regions [2]. These conducting channels carry mobile charge carriers that are likely to screen the vibrational response in the NCCDW

phase. Given the similarity of the doped and the NCCDW phase, such screening of the oscillatory response is very likely to occur also in the doped crystals, which explains the absent oscillatory response at lower energy in the experiment [48]. We find the following support for this scenario in our data. The electrical resistance at $T = 10$ K increases with x [see Fig. 1(a)], while the oscillatory response recedes [cf. Figs. 4(b) and 4(d)] for $x = 0.02$ and $x = 0.06$, respectively. This dependence implies that in the macroscopic resistivity, disorder-induced scattering reduces the electric conductivity for larger x , while locally the screening of the oscillatory response increases due to a larger mobile electron density.

A recent x-ray scattering study [5] reported a ratio of bilayers to monolayers in the bulk of pristine 1T-TaS₂ of 2:1, i.e., 20% of the layers are monolayers. Our results for cleaved surfaces and $x = 0$ are consistent with a significant bilayer contribution, as discussed above. For the doped compound, the calculated monolayer spectral function shows a better agreement with the experimental result than the bilayer spectral function, since only the monolayer reproduces the oscillatory response observed experimentally at E_F . Summing up the bi- and the monolayer contributions [see Fig. 8(c)], essentially reproduces the monolayer response near E_F , due to the weak bilayer contribution. A possible interpretation of this observation is that the contribution of monolayers at surfaces of the doped system is higher than in the pristine case (while the discrepancies between theory and experiment at higher binding energies are a consequence of the above-mentioned inhomogeneity). The implication of this scenario is that doping does not only inhibit commensurate CDW order but also hinders the formation of bilayers.

Indeed, dopant-induced disorder is likely to prevent bilayer formation. As depicted in Fig. 5(b) for layers Nos. 2 3 and Nos. 4 5, the bilayer formation requires an identical stacking of the Star-of-David clusters formed in adjacent layers. Doping by Mo substitutes Ta with a statistical distribution and will reduce the probability of bilayer formation since it is unlikely that the substitution follows a stacking pattern. Since the stacking order influences the physics in profound ways, this finding is significant. It suggests that, in addition to the appearance of metallic channels between polaronically distorted domains, chemical doping triggers a restructuring of the layered crystal and fundamentally changes the nature of electronic correlations.

Having drawn this conclusion we note that the match of the oscillatory spectral response between experiment and theory is not perfect. Besides the importance of disorder addressed above, we observe in the experiment for $x = 0$, at $E - E_F = -0.4$ eV an additional oscillatory signature [see Figs. 4(c) and 7(d)]. This energy agrees well with the CDW energy gap at the Brillouin zone boundary at the M point detected in static ARPES [21,22]. In trARPES the CDW gap oscillates at the amplitude mode frequency, in agreement with the $\Omega = 2.4$ THz identified in the present work using $h\nu = 6$ eV. At this

low photon energy, ARPES does not reach the M point. Moreover, we detect the photoelectron in normal emission at Γ . Following Petersen *et al.* [21], we assign the oscillation at -0.4 eV to the amplitudon whose excitation coherently modifies the scattering probability leading to backfolding from M to Γ . Due to computing limitations, our modeling does not take the long-range ordered crystal lattice into account and lacks this backscattered feature.

IV. CONCLUSIONS

We presented microscopic insights into dynamic electronic structure changes in response to the amplitude mode of the charge density wave in pristine and Mo-doped 1T-TaS₂. Comparing the results of time-resolved photoelectron spectroscopy with real-time DMFT simulations combined with parameter modulations inspired by density functional calculations, we analyzed periodic variations in the electronic structure. This approach allowed us to discuss the effects of disorder and stacking in the material. While for the parent compound the experiment-theory comparison suggests a coexistence of mono- and bilayers near the surface, our findings for the doped systems are consistent with an increased monolayer fraction induced by Mo doping. This restructuring leads to enhanced electronic correlations and a strongly modified oscillatory response near E_F . At the same time, enhanced screening in the metallic state results in strong damping at higher binding energies.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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