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Pudelko, W., Liu, H., Petocchi, F. et al (2024). Probing enhanced superconductivity in van der Waals polytypes of VxTaS2. PHYSICAL REVIEW MATERIALS, 8(10). http://dx.doi.org/10.1103/PhysRevMaterials.8.104802

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Editors' Suggestion

Probing enhanced superconductivity in van der Waals polytypes of V_xTaS₂

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(Received 25 January 2024; revised 15 July 2024; accepted 24 September 2024; published 25 October 2024)

Layered transition metal dichalcogenides (TMDs) stabilize in multiple structural forms with profoundly distinct and exotic electronic phases. Interfacing different layer types is a promising route to manipulate TMDs' properties, not only as a means to engineer quantum devices but also as a route to explore fundamental physics in complex matter. Here we use angle-resolved photoemission (ARPES) to investigate a strong layering-dependent enhancement of superconductivity in TaS₂, in which the superconducting transition temperature, T_c , of its 2*H* structural phase is nearly tripled when insulating 1*T* layers are inserted into the system. The study is facilitated by a vanadium-intercalation approach to synthesizing various TaS₂ polytypes, which improves the quality of the ARPES data while leaving key aspects of the electronic structure and properties intact. The spectra show the clear opening of the charge density wave gap in the pure 2*H* phase and its suppression when 1*T* layers are introduced to the system. Moreover, in the mixed-layer 4*Hb* system, we observe a strongly momentum-anisotropic increase in electron-phonon coupling near the Fermi level relative to the 2*H* phase. Both phenomena help to account for the increased T_c in mixed 2*H*/1*T* layer structures.

DOI: 10.1103/PhysRevMaterials.8.104802

I. INTRODUCTION

Layered transition metal dichalcogenides (TMDs) are quasi-two-dimensional materials with rich phase diagrams involving numerous exotic electronic phases. Tantalum disulfide is a widely studied TMD, whose properties are emblematic of many compounds in this family. It typically stabilizes in one of two structural phases, 2H or 1T. While chemically equivalent, these phases exhibit markedly different electronic properties. The former is a good metal, which enters a charge density wave (CDW) phase at 78 K and becomes a superconductor below 0.8 K [1]. The latter is insulating and undergoes multiple CDW transitions: incommensurate at 543 K to nearly commensurate at 352 K to fully commensurate (CCDW) below 183 K [2]. In the CCDW phase, the reconstructed unit cell forms a Star-of-David (SoD) cluster, consisting of 13 Ta atoms [3], which has been proposed to host a quantum spin liquid [4,5]. Despite the two-dimensional character of 1T-TaS₂, many theoretical and experimental studies have shown non-negligible out-of-plane dispersion [6] or stacking dependence [7,8], which can further influence its electronic properties and change the gap character from band to Mott insulator [3,9,10].

Due to the delicate balance of interactions, the properties of many TMDs are highly sensitive to minute changes in virtually any material or external parameter, allowing one to, e.g., tune metal-insulator transitions [11], manipulate magnetic interactions [12], or induce or enhance superconductivity [13–18]. In addition to these diverse tunable properties, the weak couplings between layers make TMDs an especially attractive platform for engineering exotic electronic phenomena at their interfaces and manufacturing novel quantum devices [19–23].

One testament to the promise of interfacing various TaS_2 structures is the 4*Hb* phase, a polytype consisting of 1*T* and 1*H*/1*H'* elements (half-layers of the 2*H* structure) stacked alternately along the *c* axis [24,25]. This structure has recently attracted attention, due to evidence that it hosts a chiral superconducting state [25–27]. At the same time, the superconducting *T_c* of the 4*Hb* phase (2.2 K) is nearly triple that of 2*H*-TaS₂. These observations suggest that investigations of

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FIG. 1. (a) Crystal structures of 2*H*- and 1*T*-TaS₂. (b) Illustrations of how the 4*Hb*, 6*R*, and disordered phases are composed of the 2*H* (1*H*/1*H'*) and 1*T* layer elements. (c) Powder XRD measurements of V_x TaS₂ indicating changes in the structure as a function of *x*. (d) Normalized resistivity ρ/ρ_{4K} of V_x TaS₂ for various *x* values. (e) T_c as a function of *x*.

mixed 2H/1T-TaS₂ layer systems can yield profound insights into the interplay of superconductivity with topology, dimensionality, many-body interactions, and competing orders.

This work investigates the strong enhancement of T_c in 4*Hb* and other mixed-layer phases of TaS₂. We use angleresolved photoemission spectroscopy (ARPES) to probe the electronic structure of various polytypes of TaS₂. Our study utilizes vanadium intercalation to obtain the different TaS₂ structural configurations. We find that V_x TaS₂ samples grant clearer ARPES spectra than conventionally grown TaS₂ samples of the same polytypes, while possessing quantitatively similar band structures [2,28] and the same key electronic behaviors, including—most importantly—the same elevated T_c in the 4*Hb* phase relative to the 2*H* phase.

The ARPES measurements focus on the 2H, 4Hb, and 1T phases in order to identify factors at play in the strong enhancement of superconductivity in 4Hb and other mixed 2H/1T-TaS₂ systems. Our results show that when 1T layers are inserted into the 2H structure, as in the 4Hb phase, the CDW within the 1H(') layers is suppressed while electron-phonon (*e*-ph) interactions are strongly enhanced. Surprisingly, this enhancement in electron-phonon coupling (EPC) is highly anisotropic in momentum space and only manifests in certain regions of the band structure.

II. RESULTS

Figure 1 presents an overview of the $V_x TaS_2$ system. The 2*H* and 1*T* layer components and their arrangements in various polytype structures are illustrated in Fig. 1(a). Data from powder x-ray diffraction (XRD) performed on samples with various x values are shown in Fig. 1(b), with labels indicating phases interpreted using standard structural data. Below x = 0.03, $V_x TaS_2$ stabilizes in the 2*H* phase. Samples synthesized in the range of about $0.03 \le x \le 0.05$ have the 4*Hb* structure. In a narrow intercalation window near x = 0.07, the structure is assigned to the 6*R* phase. At slightly higher *x* levels, the arrangement of 1H(') and 1T layers appears to be disordered. Finally, at x = 0.15 the compound is in the pure 1T phase. Differences in the in-plane lattice constants of V-intercalated and conventional TaS₂ samples are well below 1%. Larger interlayer spacings (up to 2%) may be notable, though, particularly as they might promote cleaner sample cleavage and thereby account for the improvements in the quality of ARPES data [29]. The XRD analysis is consistent with core-level spectroscopy results, which identify distinct chemical environments of the Ta and S atoms in the 1H(') and 1T layers [29].

As shown in Fig. 1(d), a superconducting transition occurs in all samples that contain 1*H* structural components. Intercalating vanadium into the 2*H* (x = 0) system, and thereby inserting 1*T* layers into the structure, increases the T_c . The T_c reaches a maximum of 2.2 K in the 4*Hb* structure (x = 0.05), which matches the T_c in other 4*Hb*-TaS₂ samples [25]. Further details about the structure and transport properties of the V_x TaS₂ samples are provided in the Supplemental Material (SM) [29].

Figures 2(a)-2(c) show ARPES momentum maps in the surface $k_x \cdot k_y$ plane, evaluated 100 meV below E_F for the x = 0, 0.05, and 0.15 samples. The electronic structures of the x = 0 and 0.15 samples are consistent with results reported for the 2*H* and 1*T* structures, respectively [2,3,30,31]. The 2*H* phase [Fig. 2(a)] is a metallic system consisting of two so-called "barrel"-shaped Fermi surface (FS) sheets centered around the Γ and *K* points, plus a "dogbone" sheet located at the *M* point [30,32], labeled as 2*H*-B and 2*H*-D, respectively. The 1*T* phase [Fig. 2(b)] is composed of a single gapped band that, when viewed at the energy of the gap, forms a "flower



FIG. 2. (a)–(c) ARPES *k*-space maps evaluated at $E - E_F = -100$ meV obtained from 2*H* (x = 0), 1*T* (x = 0.15), and 4*Hb* (x = 0.05) samples, respectively. (d) DMFT calculation of a 2*H* + 1*T* model system, evaluated at $E - E_F = -100$ meV. The overlaid contours indicate contributions of the 2*H* and 1*T* components, which are evident in panel (c).

petal" in the k_x - k_y plane centered around M (1*T*-FP). In the x = 0.05 sample [Fig. 2(c)], spectral features originating from both the 2*H* and the 1*T* phases are visible, consistent with data reported on 4*Hb*-TaS₂ [25]. The contributions of the 2*H* and 1*T* components to the band structure of the mixed-layer phases are highlighted by density mean-field theory (DMFT) calculations, which consider a 1*T* layer inserted into the 2*H* structure. Unlike density functional theory, the DMFT method accounts for a Hubbard interaction energy, *U*, within the 1*T* layers. The model results, shown in Fig. 2(d), are in reasonable agreement with the ARPES data from the 4*Hb* (x = 0.05) phase when assuming U = 0.4 eV, similar to the 1*T* system [9].

Figure 3(a) shows the evolution of the band dispersions in $V_x TaS_2$ with increasing x. The top left panel displays the 2H (x = 0) phase, consisting of 2H-B and 2H-D bands. The 1T (x = 0.15) phase, consisting of the 1*T*-FP band, is shown in the bottom right panel. In the mixed 4Hb (x = 0.05) and 1H + 1T (x = 0.1) systems, all three bands are visible. While the most salient features of the 2H- and 1T-derived bands are preserved in these phases, there are notable differences among the spectra, which could be points of future investigation. These include a large downward shift in the energy of the 2*H* valence bands around Γ , and faint replicas of the 1*T*-FP band in the 4Hb phase. In addition, compared to the pure 2H(x = 0) phase, the 4*Hb* (x = 0.05) and disordered 1*H* + 1*T* (x = 0.1) structures show a slight upward shift of the 2*H*-B and -D conduction bands. The corresponding changes in Fermi momenta appear consistent with a recent observation



FIG. 3. (a) ARPES measurements of electron dispersions in 2H, 4Hb, mixed 1H + 1T, and 1T samples with the noted x values. Barrel, dogbone, and flower-petal bands are labeled as 2H-B, 2H-D, and 1T-FP, respectively. Rectangles in the spectra of the 2H and 4Hb samples highlight regions that are analyzed in detail in Fig. 4. (b) ARPES k_x - k_z maps of 4Hb and 1T samples, evaluated 100 meV below E_F . (c) Bulk and surface Brillouin zones. For simplicity, the "prime" notation of the surface Brillouin zone labels is dropped when considering in-plane results.

of charge transfer between the 1T and 1H(') layers within 4Hb-TaS₂ [28].

We also note that, although we observe signatures of SoD reconstruction in 1T-V_xTaS₂ (x = 0.15), its ARPES spectrum does not exhibit a shallow, gapped spectral feature at Γ , which is seen in conventionally grown 1T samples and widely ascribed to a Mott gap [6,7,33]. The lack of this feature is attributable to a difference in the vertical stacking alignment of layers [3,7,9]. As the "Mott gap" feature does not carry over into the 4*Hb* system where we investigate the enhancement of T_c [25,28], its absence in 1T-V_xTaS₂ does not affect the current study.

Varying the photon energy in ARPES allows the electronic structure to be probed as a function of the out-of-plane momentum, k_z . Figure 3(b) compares momentum maps from the x = 0.05 (4Hb, left) and 0.15 (1T, right) samples in the k_x - k_z plane, evaluated 100 meV below E_F . Here the 1T-FP band in the x = 0.15 sample exhibits weak but observable momentum dependence along k_z , similar to previous reports [10,34–39]. By contrast, the same 1T-derived bands in the 4Hb (x = 0.05) phase show little to no variation with respect to k_z , signaling enhanced two-dimensionality. For orientation, the bulk and surface Brillouin zones are sketched in Fig. 3(c).



FIG. 4. (a) FSs of 2H (x = 0, left panel) and 4Hb (x = 0.05, right panel) samples measured at 18 K. Gapped portions of the 2H FS (arrows) are signatures of CDW order. (b) Normalized resistivity measurements as a function of x. A cusp in the resistivity due to the CDW transition at $T \sim 76$ K in the 2H phase (x = 0 and 0.01) is suppressed in the 4Hb phase and other mixed polytypes ($x \ge 0.05$). (c) EPC constant λ of the 2H (x = 0) and 4Hb (x = 0.05) samples along the Γ -K, Γ -M, and M-K directions. (d) [i] Fitted band dispersion (white squares), as well as the renormalized Fermi velocity (dashed black line) and extracted bare band velocity (solid black line) along the Γ -K direction for the x = 0 (right panel) and 0.5 samples (left panel). [ii] ([iii]) Real (imaginary) part of the self-energy, Σ' (Σ''), along the Γ -K direction for x = 0 and 0.05. Two methods of computing $\Sigma'(\omega)$ are shown: Kramers-Kronig transformation of $\Sigma''(\omega)$ (KK) and subtraction from the bare band dispersion (disp). Panels (e) and (f) show the same analysis as in panel (d), applied along the Γ -M and M-K directions, respectively.

The FS presented in Fig. 4(a) (left panel) exhibits welldefined gapped regions along the dogbone sheets-clear signatures of a CDW reconstruction. The locations of the gapped portions in the FS sheet (indicated with the white arrows) match those of the sister compound 2H-TaSe₂, where the $q_{\text{CDW}} = 2/3 |\Gamma - M|$ ordering wave vector leads to a commensurate 3×3 reconstruction of the unit cell [40–42]. Analysis of the Lindhard susceptibility of 2H-TaS₂, included in the SM [29], further establishes the similarities between its CDW state and that of 2H-TaSe₂. Moving from the 2H(x = 0) system to the 4Hb structure (x = 0.05), the CDW instability is suppressed, as seen in the right panel of Fig. 4(a). A comparison of the energy distribution curves taken at the k points where the CDW gap opens is included in the SM [29]. The suppression of the CDW phase transition in samples containing a mixture of 2H and 1T layers ($0.03 \le x \le 0.10$) is confirmed by resistivity measurements, as shown in Fig. 4(b).

In addition, the ARPES data show signatures of electron interactions in the 2*H* band structure in the form of a dispersion "kink" anomaly roughly 30 meV below E_F . The kink can be seen in ARPES dispersion cuts along various momentum directions. It is highlighted by the boxes in Fig. 3(a) and examined in detail in Figs. 4(d)-4(f)[i]. This energetically sharp feature, which has also been noted in previous work [30], ostensibly signals that the electrons couple to a particular boson mode at the same energy. We ascribe the kink to *e*-ph interactions; calculations of the 2*H*-TaS₂ phonon band structure find an optical phonon branch with a flat dispersion at the same ~30-meV scale, where there is substantial EPC over an extended region centered around the Γ -*M* wave vector [43].

In Figs. 4(d)–4(f), we analyze the dispersion kink to assess changes in the EPC parameter, λ , between the 2*H* (*x* = 0) and 4*Hb* (x = 0.05) samples. We evaluate λ from analysis of the complex electronic self-energy, $\Sigma(\omega) = \Sigma'(\omega) +$ $i\Sigma''(\omega)$, where $\omega = E - E_F$. We follow the general approach of Refs. [44,45], which is based on Lorentzian fitting of the ARPES momentum distribution curves (MDCs) at fixed energies. For each sample, $\Sigma(\omega)$ is analyzed along the Γ -K, Γ -*M*, and *M*-*K* directions. $\Sigma'(\omega)$ and $\Sigma''(\omega)$ are related to the spectra as the difference between the renormalized and noninteracting "bare" dispersions and from the widths of the MDCs at the corresponding energies, respectively. The methodology involves extracting $\Sigma'(\omega)$ by two independent methods— Kramers-Kronig (KK) transformation of $\Sigma''(\omega)$ and analysis of the dispersion (disp)-which self-consistently determine the noninteracting Fermi velocity v_F^0 and thus the absolute scaling of $\Sigma(\omega)$. Details of the calculations and results are described in the SM [29].

The extracted λ values are presented in Fig. 4(c). For the 2*H* phase (x = 0), we find λ ranges from 0.236 along the Γ -*M* direction to 0.494 along the Γ -*K* direction. This variation in λ is not surprising, as the EPC is known to be *k* dependent in 2*H*-TaS₂ [30,46]. It is remarkable, however, that the *enhancement* in EPC in going from the 2*H* phase to the 4*Hb* phase (x = 0.05) is strongly momentum dependent. Namely, λ is virtually unchanged at the measured points along the Γ -*K* and *M*-*K* directions, while a large enhancement in the interactions is found along the Γ -*M* direction, where $\lambda = 0.440$ (an increase of about 85%).

III. DISCUSSION

The results here offer spectroscopic insights into key factors that lead to a strong enhancement of T_c in the mixed-layer

polytypes. The suppression of CDW order restores states to the FS that are especially likely to couple strongly to phonons, allowing them to form Cooper pairs in the competing superconducting phase. Meanwhile the enhancement in EPC at the \sim 30-meV scale, while strongly momentum anisotropic, is nonetheless substantial in terms of its likely impact on T_c . As an illustration, by applying the McMillan equation [47] and averaging the λ values extracted along the three momentum directions in Figs. 4(d)-4(f), the measured T_c values of 0.8 K and 2.2 K in the 2*H* and 4*Hb* systems, respectively, can be rationalized under reasonable assumptions, even before other factors are taken into account [29].

It follows from this that Mott-like interactions originating from the 1T planes should not play a leading role in elevating the T_c in 4Hb-TaS₂. This aligns with the negligible k_z dispersion of the 1*T*-FP band within the 4*Hb* system [Fig. 3(b)], as well as the similar U values inferred from DMFT modeling of 1T layers in pure and 4Hb-like systems, which both suggest that Mott interactions remain largely confined within the 1Tplanes. The ARPES measurements furthermore suggest that changes in carrier density, either due to inter- or intralayer charge transfer or doping by the intercalants, probably have little *direct* impact on T_c . Observed changes in the FS volume of the 2H bands are too small to account for the large T_c enhancement via the density of states alone and, if anything, would tend to reduce the states at E_F and hence the T_c [28]. Still, small charge transfer and/or doping effects could indirectly influence superconductivity in profound ways, e.g., by altering the *e*-ph interactions.

The strongly momentum-dependent increase in λ highlights the complexity of the interactions in these systems and the need for further study. It is tempting to draw inferences from the fact that we observe a dramatic EPC enhancement at a point in the dispersion along the same momentum direction (Γ -M) as the 2H CCDW ordering wave vector. It is possible the the same phonon interactions at play in the CDW order of the 2H phase are strengthened and bolster superconductivity in the 4Hb system, similar to recent findings in charge-ordered cuprates [33]. Drawing such conclusions, however, will require further studies to the identify the phonon wave vectors where EPC is strongly enhanced.

The observations here may have broader relevance outside of the TaS_2 bulk polytypes. Suppression of CDW order and enhancement of EPC were also observed in other intercalated TMDs [48] and atomically thin layers of 2*H*-TaS₂ [18,19]. The similar behaviors in these systems may be general consequences of isolating individual 1H(') layers, which may reduce metallic screening and alter the phonon dispersions and FS nesting conditions.

IV. CONCLUSIONS

In conclusion, using ARPES, we have probed the electronic structures of 2H-, 1T-, and 4Hb-TaS₂ in order to investigate the strong enhancement of superconductivity in mixed 2H/1T layer polytypes relative to the pure 2H structure. Our work utilized vanadium intercalation as a route to synthesize high-quality TaS₂ polytypes. The ARPES measurements demonstrate the clear presence of the CDW gap in pure 2H-TaS₂, which by a comparison to isovalent 2H-TaSe₂, might be associated with a commensurate 3×3 reconstruction. ARPES and transport data further show that the CDW is suppressed in mixed 2H/1T layer forms of V_xTaS₂ (0.03 \leq $x \leq 0.1$). The spectra exhibit a kink dispersion anomaly in the 2H-derived bands as a signature of energetically sharp electron excitations—presumably e-ph interactions—roughly 30 meV below E_F . In the 4Hb structure (x = 0.05), the EPC associated with the kink is strongly enhanced in a highly momentum-dependent manner. Both the suppression of the CDW and the momentum-anisotropic increase of EPC are likely to be key factors in explaining the large enhancement of T_c when 2H-TaS₂ layers or half-layers are electronically isolated—either as polytypes incorporating 1T layers (4Hb, 6R, and other phases), or in monolayer or true heterostructure form.

ACKNOWLEDGMENTS

W.R.P., N.C.P., J.K., K.v.A., and J.C. acknowledge support from the Swiss National Science Foundation through Projects No. 200021_185037 and No. 200021_188564. Q.W. acknowledges support by the Research Grants Council of Hong Kong (ECS No. 24306223). We acknowledge MAX IV Laboratory for time on Beamline Bloch under Proposal No. 20230375. Research conducted at MAX IV, a Swedish national user facility, is supported by the Swedish Research Council under Contract No. 2018-07152, the Swedish Governmental Agency for Innovation Systems under Contract No. 2018-04969, and Formas under Contract No. 2019-0249. The authors thank A. Kanigel for helpful discussions.

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