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Enhanced superconductivity upon weakening of charge density wave transport in 2H-TaS$_2$ in the two-dimensional limit

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1. INTRODUCTION

Transition-metal dichalcogenides (TMDs) 2H-MX$_2$ (where $M = Nb$, Ta and $X = S$, Se) have attracted considerable attention as novel two-dimensional (2D) crystalline superconductors [1]. In these materials, superconductivity (SC) occurs in an environment of preexisting charge-density wave (CDW) order [2,3], making them an ideal platform to study many-body ground states and competing phases in the 2D limit. In bulk crystals, the reported critical temperature of the CDW transition decreases from 120 K in 2H-TaSe$_2$ down to 30 K in 2H-NbSe$_2$. Superconductivity weakens in approximately reverse order, with $T_c$ increasing from around 0.2 K in 2H-TaSe$_2$ to 7.2 K in 2H-NbSe$_2$. The relationship between CDW and superconductivity in such systems is still under debate [4,5]. It is generally believed that their mutual interaction is competitive, but evidence to the contrary, indicating a cooperative interaction, has also been reported in angle-resolved photoemission spectroscopy (ARPES) studies [3].

In TMDs, superconductivity and CDW instability can be investigated by adjusting the interlayer interactions through pressure [6,7] or molecule intercalation [8,9]. Recently, mechanical exfoliation has emerged as a robust method for producing ultraclean, highly crystalline samples with atomic thickness [10]. This offers a useful way to assess the effect of dimensionality and interlayer interactions on both superconductivity and CDW. A material whose behavior as a function of layer thickness has been recently studied is NbSe$_2$ [11–14], in which the superconducting state is progressively weakened in samples thinner than five layers (5L), with $T_c$ lowered from 7.2 K in bulk crystals to 3 K in the monolayer. The thickness dependence of CDW order is still under debate, with different results from Raman and scanning tunneling microscopy/spectroscopy (STM/STS) studies [15,16].

Bulk 2H-TaS$_2$, another member of the 2H-MX$_2$ family, exhibits a CDW transition at 70 K and a SC transition at 0.8 K [2,17–19]. Compared to NbSe$_2$, TaS$_2$ manifests a stronger signature of the CDW transition in transport in the form of a sharp decrease of resistivity [9], and thus serves as a desirable platform to study the thickness dependence of the CDW instability. STM/STS measurements on monolayer TaS$_2$ epitaxially grown on Au(111) substrates show suppression of the CDW instability [20]. Regarding superconductivity, an enhanced $T_c$ down to a thickness of 3.5 nm (5L) was recently reported, utilizing TaS$_2$ flakes directly exfoliated on a Si/SiO$_2$ substrate [19]. Considering the fact that an appreciable decrease of $T_c$ in NbSe$_2$ only occurs in samples thinner than five layers, it would be interesting to explore the trend of $T_c$ in TaS$_2$ toward the exact 2D limit. Prior work found that samples thinner than 5L become insulating, indicative of its particular susceptibility to degradation in ambient atmosphere. Therefore, exfoliation and encapsulation in an inert atmosphere become crucial in order to obtain high-quality samples.

This paper is arranged as follows: in Sec. II, we report electronic transport studies of the properties of mono- and few-layer 2H-TaS$_2$ at different temperatures and magnetic fields. We show that superconductivity persists in TaS$_2$ down to the monolayer limit, with a pronounced increase in $T_c$ from...
0.8 K in bulk crystals to 3.4 K in the monolayer. At higher temperatures, two electronic transport signatures of the CDW transition are found to vanish in ultrathin samples: (1) a kink in the temperature dependence of the resistivity, and (2) a change of sign in the Hall coefficient versus temperature. In Sec. III, we report calculations on the electronic and vibrational properties to explore the competition between SC and CDW in this material. We found that suppression of the CDW order leads to a substantial increase in the density of states at the Fermi level, which ultimately enhances \( T_c \). We also revisit the impact of quantum fluctuations of the CDW order on the enhancement of \( T_c \). Our study provides insights into the importance of reduced dimensions on many-body ground states and their interactions.

II. EXPERIMENTAL RESULTS

A. Device fabrication

We exfoliate and fabricate samples with a transfer setup built inside a glovebox filled with argon gas, and encapsulate the TaS\(_2\) flake between two sheets of hexagonal boron nitride (hBN). We build our devices utilizing a polymer pick-up technique \cite{24} as illustrated in Fig. 1(a), taking advantage of the van der Waals adhesion between 2D layers. Using this technique, hBN and TaS\(_2\) flakes are picked up in sequence during the device fabrication process, which makes it possible to produce high-quality TaS\(_2\) samples as thin as monolayer in our experiment.

B. Enhanced superconductivity in the monolayer limit

As seen in Fig. 1(b), when the temperature is sufficiently low, a clear superconducting transition is observed for one-, two-, three-, five-, seven-layer, and bulk samples. By fitting the resistance to the Aslamazov-Larkin expression \cite{25}, we are able to determine the mean-field superconducting transition temperature \( T_c \). When the sample thickness is decreased from bulk to monolayer, the corresponding \( T_c \) monotonically increases from 0.9 to 3.4 K \cite{26}. The trend observed in our experiment is strictly opposite that of a previous finding on NbSe\(_2\) \cite{12}, where \( T_c \) decreases monotonically with thickness reduction, despite the fact that the two materials are isostructural and isochemical. In NbSe\(_2\), the decrease in \( T_c \) is attributed to a weaker interlayer Cooper pairing as the layer number \( N \) is reduced. In our case, however, it is surprising to see that even with a reduced interlayer Cooper pairing, the \( T_c \) is dramatically enhanced when the thickness is reduced. To verify that the thickness dependence of \( T_c \) is independent of extrinsic factors (such as level of disorder, substrate, source of crystal, sample quality), we measure an additional set of bilayer and trilayer samples and plot our results alongside previously reported \( T_c \) values \cite{19} in Fig. 1(c). Independent of different sample preparation procedures and substrates, the trend of \( T_c \) versus thickness is consistent between the two sets of experimental results, indicative of an intrinsic origin underlying the enhancement of \( T_c \).

To further characterize the superconducting properties of thin TaS\(_2\), we measure the superconducting transition under both out-of-plane and in-plane magnetic fields. Figure 1(d) shows the \( H_c^2 \) as a function of temperature. Close to \( T_c \), the dependence of \( H_c^2 \) is fitted well by the phenomenological 2D Ginzburg-Landau (GL) model, which yields \( H_c^2 = \sqrt{2} \phi_0/\xi(0) \sqrt{(\xi(0)/\lambda(0))} \). The purple background indicates the Pauli limit regime. (e) Normalized critical current as a function of \( T/T_c \). Dashed and dotted lines denote the models proposed by Bardeen \cite{22} and Ambegaokar and Baratoff \cite{23}, respectively.

![FIG. 1.](image)

(a) Schematic of device fabrication and crystal structure of 2H-TaS\(_2\). (b) Resistance normalized by the normal state (\( R/R_N \)) as a function of temperature for one-, two-, three-, five-, seven-layer, and bulk (\( d = 40 \) nm) samples near the SC transition. The superconducting \( T_c \) is 3.4, 3.0, 2.5, 2.05, 1.6, and 0.9 K, respectively, determined by fitting the transition curve to the Aslamazov-Larkin formula (black solid lines). (c) \( T_c \) reported in this work (circles) and in a prior study (crosses) \cite{19}. The dashed line guides the eye to the general trend. (d) Out-of-plane critical field \( H_c^2 \) for two-, three-, and five-layer samples. The dashed lines are linear fits to \( H_c^2 = \phi_0/\xi(0)^2 (1 - T/T_c) \), where \( \phi_0 \) and \( \xi(0) \) denote the flux quantum and in-plane GL coherence length at zero temperature, respectively. Inset: In-plane critical field \( H_c^1 \) normalized by Pauli limit (\( H_p \approx 1.86 T_c \)) for bilayer and bulk samples. The dashed line for the bilayer is a fit to the Tinkham formula [21] for 2D samples \( H_c^1 = \sqrt{2} \phi_0/\xi(0)^2 \sqrt{(\xi(0)/\lambda(0))} \). The purple background indicates the Pauli limit regime. (e) Normalized critical current as a function of \( T/T_c \). Dashed and dotted lines denote the models proposed by Bardeen [22] and Ambegaokar and Baratoff [23], respectively.
R observations verify that thin TaS$_2$ behaves as a 2D superconductor, which both show a peak in $R_N$ down. Data for the bulk crystal is from Ref. [8].

The periodicity of the CDW is more than six times the Pauli paramagnetic limit $H_p$, obeying a square root rather than a linear temperature dependence [inset Fig. 1(d)]. This dramatic enhancement of in-plane $H_{c2}$, often referred to as Ising superconductivity, has been observed in other 2D crystalline superconductors [12,13,29,30]. The above observations verify that thin TaS$_2$ behaves as a 2D superconductor. We also show that the superconducting transition exhibits the Berezinskii-Kosterlitz-Thouless (BKT) transition as expected in 2D in Appendix B. Additionally, the critical current density increases by orders of magnitude as the devices become thinner (bulk $J_c \approx 700$ A/cm$^2$, trilayer $J_c \approx 7 \times 10^5$ A/cm$^2$, bilayer $J_c \approx 1.2 \times 10^6$ A/cm$^2$). The trend of critical current density versus temperature for representative thicknesses is shown in Fig. 1(e), with a more detailed discussion given in Appendix C.

C. Transport properties and the CDW transition

In addition to the SC transition, bulk TaS$_2$ is known to exhibit CDW order below 70 K. The pattern of atomic displacements in the CDW state is illustrated in Fig. 2(a). In our experiment, we characterize the temperature-dependent resistance of two-, three-, five-, seven-layer, and bulk samples, and plot the normalized resistance versus temperature on a linear scale in Fig. 2(b). All samples manifest a linear decrease of resistance at high temperatures, consistent with phonon limited resistivity in a normal metal [31]. Below 70 K, the seven-layer and bulk devices undergo a CDW phase transition, producing a kink in resistance, which is manifested as a peak in $dR/dT$. In the inset of Fig. 2(b), the temperature derivative of the resistivity is shown, and a peak in $dR/dT$ develops in the seven-layer and bulk samples close to the transition. In two-, three-, and five-layer samples, however, such a peak in $dR/dT$ is absent.

To further investigate the temperature dependence of the sample resistance, we plot the subtracted resistance $R - R_N(4 K)$ on a log-log scale in Fig. 2(c). For bulk, the linear temperature dependence is disrupted by a sudden switch to $T^2$ near $T_{CDW}$, a well-known consequence of electron-phonon scattering at temperatures lower than the Debye temperature $\Theta_D$. In contrast, a gradual transition to $R \sim T^2$ is observed in two- and three-layer samples. It is important to note that these powers are only observed over the range between $T \approx 55$ K and SC $T_c$, and thus extend over a bit less than a decade.

In bulk TaS$_2$, the Hall coefficient undergoes a broad transition between 70 and 20 K including a change in sign at 56 K [8], which is another indicator of the electronic structure change induced by the CDW transition. It has been shown that a two-carrier model with light holes and heavy electrons is necessary to explain the opposite signs for the Seebeck and Hall coefficients measured above the CDW transition temperature, as opposed to a single-carrier model describing the low-temperature behavior [8]. In Fig. 2(d), we plot the Hall coefficient ($R_H$) of three representative thicknesses below 100 K. In the five-layer sample, a significant deviation from the bulk behavior is already apparent: the overall magnitude of $R_H$ and temperature where it switches sign are both diminished. However, the most striking fact is the weak temperature dependence and absence of a sign change in the two-layer sample. This implies that the same electronic structure modification caused by the CDW transition that affects the Hall coefficient is also absent in the 2D limit.

III. DISCUSSION

As superconductors are thinned down to the 2D limit, their critical temperature $T_c$ typically decreases [32]. This suppression of SC is normally attributable to either disorder-induced localization of Cooper pairs, weaker Coulomb screening in 2D, or reduction of the superfluid stiffness which leads to a reduction of the BKT transition temperature. Recent studies reveal more novel origins of suppressed superconductivity in atomically thin NbSe$_2$, including a weaker interlayer Cooper pairing [12] and suppression of the Cooper pair density at the superconductor-vacuum interface [14]. Beyond these mechanisms, here we consider a key factor affecting $T_c$ that was not quantitatively studied in atomically thin layered $2H$-MX$_2$: the interaction with CDW order. Anticorrelated trends between SC and CDW transition have been observed in bulk $2H$-TaS$_2$ crystals under pressure [6,28] and in single-crystal alloys [33–35]. Here we hypothesize that the enhancement of $T_c$ as sample thickness decreases all the way to the 2D limit is associated with a suppression of the electronic structure reconstruction induced by the CDW order.
figuring on the inner pocket around K along Ε-Κ and Κ-M. In
addition, the saddle point located along Ε-Κ, is shifted to en-
ergies above the Fermi level. The reconstruction of electronic
structures induced by the CDW order is also seen in the Fermi
energy contour plots shown in Figs. 3(d) and 3(e). After
the CDW transition, the pockets around K and Ε are either partially
or fully gapped. This reconfiguration of electronic structures
may account for the abnormal change of sign seen in the Hall
measurement. However, a quantitative analysis is not possible
without access to both the local curvature of the Fermi surface
and the anisotropy in the scattering time at different η points.
Given that the k-dependent CDW-gap and SC-gap formation
in 2H-MX₂ have been intensively studied by ARPES [3,43],
our DFT calculations can serve as a check of these results.

We also computed the phonon dispersion for the bulk and
monolayer (see Appendix F). We find that in both cases,
an acoustic mode that involves in-plane motion of Ta atoms
softens and becomes unstable as the electronic temperature is
lowered. In both cases the instability occurs at approximately
the same wave vector that corresponds to the CDW ordering
Q̄ CDW ≈ 2/3ΓM [44]. This is in good agreement with the
3 × 3 periodicity of the CDW order observed in 2H-MX₂.

To investigate the change of Tc that may be induced by a
suppression of the CDW order, we calculate Tc, as a function of
CDW amplitude based on electronic and phononic structure
calculations. We recall that in McMillan’s theory, the critical
temperature is expressed as [45]

\[ Tc = \frac{\Theta_D}{1.45} \exp \left[ \frac{-1.04(1 + \lambda)}{\lambda - \mu^*(1 + 0.62\lambda)} \right] \]  

where \( \mu^* \) is the Coulomb pseudopotential of Morel and And-
erson, and \( \lambda \) is the electron-phonon coupling constant. We find
that if the CDW distortion is completely suppressed, the cor-
responding change in DOS(\( E_F \)) can result in an enhancement of
Tc up to 3.75 K (see Appendix G). The result is summarized in
Fig. 3(f). This provides a good approximate estimate on the
impact of the suppression of the CDW instability on the
superconducting Tc. Interestingly, this estimate agrees reason-
ably well with our experiment, indicating the importance of
including electronic structure modifications induced by the
CDW as a contributing factor. We note that there are several
other factors that can impact Tc, such as substrate effects, the
presence of a van Hove singularity near the Fermi level, and
enhanced electron-phonon coupling due to reduced screening
[19] in 2D. These factors have been discussed in great detail
in the literature and we do not reiterate them here.

The above analysis investigates primarily the direct com-
petition between CDW and superconductivity over DOS. We
further note that a continuous suppression of the CDW phase
as a function of thickness would imply a softening of the
CDW amplitude mode. If the presence of quantum fluctuations
caused by proximity to a CDW transition is considered, the
analysis of its impact on SC can go beyond a simple “competing
order” scenario [46]. The competition between fermionic
incoherence and the strong electron-mediated pairing near a
quantum critical point can yield a pairing instability at tempera-
ture Tc which increases and saturates as the order correlation
length \( \xi \to \infty \). This reveals potentially rich relationships
between CDW and SC that cannot be simply interpreted in the

FIG. 3. (a) Band structure for monolayer 2H-TaS₂. The gray lines
show the unfolded band structure compared with the original band
structure in the normal phase (red lines). (b) Density of states for
monolayer/bilayer/bulk in the normal phase. Monolayer in the CDW
phase is plotted as a gray shade for reference. (c) Density of states for
monolayer/bilayer/bulk in the CDW phase. Monolayer in the normal
phase is plotted as a gray shade for reference. (d) Energy contour at
the Fermi energy in the CDW phase for monolayer TaS₂. (e) Energy
contour at the Fermi energy in the normal phase for monolayer TaS₂.
(f) Left axis: density of states at the Fermi level DOS(\( E_F \)) as a function of
CDW amplitude for the monolayer. Right axis: \( Tc \) from McMillan’s
formalism [45] using the calculated DOS(\( E_F \)).
BCS framework as the material thickness is reduced toward the 2D limit.

After careful analysis of possible correlations between SC and CDW in TaS$_2$ by DFT calculations, we turn to a discussion on the transport data shown in Sec. IIIC. We will argue that the transport data is also consistent with the scenario of suppression of the CDW transition as sample thickness decreases. Let us start from the disappearance of the kink in the resistivity curve in Fig. 2(a). While the exact relation between the CDW ordering and the kink is not fully understood, its disappearance is definitely consistent with suppression of the CDW transition temperature or a complete disappearance of the ordering phase. The evolution of this kink was also studied in bulk samples of TaS$_2$ with intercalation, showing similar disappearance of the kink when the CDW order is suppressed [8]. The change in the power of the temperature-dependent resistivity from $T^3$ to $T^2$ can also be explained within the same framework. However, in this case it is only consistent if the suppression of the CDW order is continuous, such that the gap of the amplitude mode of the CDW becomes very small in ultrathin samples. As we show in the Supplemental Material [47], when the gap of the amplitude mode is smaller than thermal excitations, scattering of electrons off these fluctuations leads to a strong $T^2$ contribution to the resistivity [48]. We also note that using a Kadawaki-Woods scaling [49] one finds that the $T^2$ resistivity from electron-electron scattering is expected to be much smaller than the one we measured. Finally, the absence of a change in the sign of the Hall coefficient indicates that the change in electronic structure occurring in the bulk is either absent or different in the 2D limit. Thus, it could be that the CDW order disappears, or a different type of CDW order is stabilized in the ultrathin limit. In summary, we find that both the superconducting and transport data are consistent with the hypothesis that the CDW order is continuously suppressed when reducing the sample thickness toward the 2D limit. It is, however, important to point out that none of our probes directly measure such an effect and at this point it can only be viewed as a hypothesis consistent with multiple different measurements.

IV. CONCLUSION

We observe enhanced superconductivity in atomically thin 2$H$-TaS$_2$ accompanied with suppression of the CDW-induced transport signatures. Our electronic band structure calculations show that suppression of the CDW phase leads to a substantial increase in DOS($E_F$), which acts to boost the superconducting $T_c$. Our result reveals that competition of the two ordered phases in determining the DOS can provide a reasonable explanation of the enhanced superconducting $T_c$ in this material down to the monolayer limit. Future layer-dependent studies that directly probe the CDW order, for example, through STM/STS, Raman, and ultrafast spectroscopy measurements, will be essential to understanding the origin of the CDW formation and its stability against dimensional reduction. Future studies on the microscopic origin of the mechanisms (electronic, magnetic, electron-phonon, etc.) will be of paramount importance to a comprehensive understanding of the mutual interactions between the CDW and SC.

Note added. Recently, we became aware of a related work [50].

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APPENDIX A: SAMPLE PREPARATION

The sample fabrication process takes place in a glovebox filled with argon gas. TaS$_2$ crystals (from HQgraphene) are exfoliated onto Si/SiO$_2$ wafers inside the glovebox. hBN flakes that support bottom contacts are exfoliated onto Si/SiO$_2$ wafers. Wafers with hBN flakes are then annealed in Ar-H$_2$ atmosphere at 350°C for 3–5 hours. Selected hBN flakes, with thickness ranging from 20 to 30 nm, are patterned with e-beam lithography using MMA/ZEP followed by XeF$_2$ gas etch for 20s × 5 cycles. Then Cr/Pd 2/25 nm is evaporated by thermal or e-beam deposition. In this way, bottom contacts that are embedded in hBN are fabricated. Using a slide consisting of a poly(bisphenol A carbonate) film held on top of a piece of polydimethylsiloxane (PDMS) elastomer, hBN and TaS$_2$ flakes can be picked up in sequence and transferred onto the bottom contacts. Such encapsulation allows for a van der Waals seal between two hBN flakes and therefore prohibits exposure to air during the sample transportation outside the glovebox.

APPENDIX B: BKT TRANSITIONS

In 2D superconductors, when $d < \xi(0)$ and the lateral size is smaller than the in-plane magnetic penetration length ($\lambda_{ab} = \lambda^2/d$, estimated to be ~30 µm in thin-layer 2H-TaS$_2$, assuming $\lambda = 210$ nm and $d = 1.4$ nm [27]), the SC transition is expected to be of the BKT type [51]. In BKT theory, when the temperature drops below the Bardeen-Cooper-Schrieffer (BCS) mean-field transition temperature $T_c$, a loss of global phase coherence and dissipation due to a finite flux resistance
occur with thermally excited vortices. In this regime, a finite supercurrent can flow below the BKT transition temperature $T_{\text{BKT}}$, as a consequence of formation of bound vortex-antivortex pairs. Experimentally, $T_{\text{BKT}}$ can be determined by analyzing the $I-V$ characteristic curves at various temperatures, expected to manifest a universal relation $V \propto I^3$ when vortex-antivortex pairs dissociate. $I-V$ analysis for a bilayer ($T_c = 3$ K) and another trilayer ($T_c = 2.78$ K) sample is shown in Fig. 4, where voltage obeys nonlinear current dependence $V \propto I^3$ during the BKT transition. By plotting the voltage-current data in a log-log scale and fitting the slope to $\alpha$, we can get $\alpha$ as a function of temperature (shown in inset of Fig. 4). In this way, we estimate $T_{\text{BKT}}$ as when $\alpha$ crosses the value of 3. $T_{\text{BKT}}$ is found to be 2.8 and 2.6 K for bilayer and trilayer, respectively. They are close to and slightly lower than the BCS mean field $T_c$ measured with $R(T)$ curves, which is also consistent with the BKT theory. Here we note that there might be inhomogeneity in this trilayer sample, which shows two sections of upturning voltages, possibly due to vortex pinning by a selected disordered region.

**APPENDIX C: CRITICAL CURRENT**

In Fig. 1(e), it is remarkable that all the critical current data collapse into a single trace. By analysis of this curve, we observe that it clearly deviates from the GL theory [21], which predicts that $J_c$ is proportional to $(1 - T/T_c)^{1/2}$. In addition, we compare our result with Bardeen’s phenomenological expression [22] $J_c(T)/J_c(0) = [1 - (T/T_c)^{\alpha}]^{1/2}$ [dotted line in Fig. 1(e)], which is an approximation of numerical calculations based on density-limit microscopic theory that extends the GL theory to low temperatures. There is still a discrepancy between our result and Bardeen’s model. Ambegaokar and Baratoff [23] then proposed that if the system comprises of superconducting grains connected with an insulating region, the maximum dc Josephson current is expressed as

\[ I_{0}(T) = \frac{\pi \Delta(T)}{2eR_N} \tan \left[ \frac{\Delta(T)}{2k_B T} \right], \]

where $\Delta(T)$ is the temperature-dependent BCS gap parameter and $R_N$ is the normal-state tunneling resistance of a junction. Among the aforementioned models, the Ambegaokar-Baratoff model fits best with our experimental data.

**APPENDIX D: ELECTRONIC BAND STRUCTURE FOR PRISTINE CRYSTALS**

$2H$-TaS$_2$ and the TMD family exhibit a trigonal prismatic structure, in which the Ta atoms in all the layers are aligned vertically but the S sublattice is rotated by 60° with respect to that of the neighboring layer. In the simplest monolayer structure with spin-orbit coupling, the relevant group of bands near the Fermi level are 22 bands that are mostly composed of the $d$ orbitals of Ta atoms and the $p$ orbitals of S atoms. Thus, we include all these relevant $p-d$ orbital hybrids in deriving the Wannier tight-binding Hamiltonians. Due to the broken inversion symmetry, spin splitting in the bands from spin-orbit coupling is present at generic $k$ points. To give the essential physics picture and elucidate the features in the electronic structure, here we provide more electronic structure details for the pristine monolayer, bilayer, and bulk crystals. In Fig. 5, we summarize the band plots along $\Gamma$-$K$-$M$-$\Gamma$ density of states and the energy contour at the Fermi energy. We find that the Fermi contour and the DOS at the Fermi level in the monolayer are not very different from the bulk case. Therefore band structure modification due to reduction of dimensionality.
FIG. 6. CDW-ordered monolayer electronic band structure, the folding effects from the supercell geometry, and the inverse unfolding process. (a) BZ (red hexagon) and sBZ (blue hexagons) in the repeated zone. (b) Energy contour of the CDW-ordered monolayer crystal in the repeated zone scheme (contrast with the ones for the pristine crystal without CDW order in Fig. 5). (c) Folded CDW band structure (magenta) along Γ-K-M-Γ as outlined in (a). For comparison, bands from the pristine crystal without CDW order in the 1×1 unit cell are also plotted (cyan). (d) Folded supercell bands (magenta) compared with the unfolded and weighted effective bands. The unfolded effective bands differ from the pristine electronic band structure by additional features such as gap opening from CDW perturbations [as in Fig. 3(a)]. The Fermi level is indicated by the white horizontal line. When the CDW energy contour in (b) is decorated with the unfolding weight as the band structure, the unfolded CDW energy contour is obtained in Fig. 3(d).

itself cannot result in the enhancement of $T_c$. More details and numerical implementations can be found in the Supplemental Material [47].

APPENDIX E: ELECTRONIC BAND STRUCTURE UNFOLDING

The CDW phase leads to a deformed crystal lattice as shown in Fig. 2(a) with 3×3 periodicity, whose band structure can be plotted in a supercell Brillouin zone. It can be viewed as the “folded” bands from the original bands of the pristine crystal. We describe here the details for the reverse “unfolding” procedure [41,52,53], which aims to present the CDW effects as perturbation on the pristine band structure.

When the primitive unit cell in real space is enlarged to the supercell, the corresponding BZ in the reciprocal space is shrunk to the sBZ. In Fig. 6(a), we compare in reciprocal space the original BZ (red hexagon) and the sBZ (blue hexagons) in the repeated zone scheme. We use $\tilde{a}_i$ to denote the primitive vectors of the original 1×1 unit cell, and $\tilde{a}_i^{sc} = 3\tilde{a}_i$ for the 3×3 supercell primitive vectors. The corresponding reciprocal lattice vectors are $\tilde{G}_i$ and $\tilde{G}_i^{sc} = \tilde{G}_i/3$. The area of one original BZ contains nine sBZs. Suppose the crystal momentum $\tilde{k}^{sc}$ lies within sBZ; any crystal momentum $\tilde{k} = \tilde{k}^{sc} + p_1\tilde{G}_1^{sc} + p_2\tilde{G}_2^{sc}$ with integers $p_i$ in original BZ would be classified as the same $\tilde{k}^{sc}$ in the sBZ (repeated zone). This can be seen from the Bloch theorem derived from the translational symmetry under the supercell translation vectors $\tilde{T}^{sc} = m_1\tilde{a}_1^{sc} + m_2\tilde{a}_2^{sc}$ with integers $m_i$.

$$e^{i\tilde{k}^{sc}\tilde{T}^{sc}} = e^{i(k^{sc} + p_1G_1^{sc} + p_2G_2^{sc})(m_1\tilde{a}_1^{sc} + m_2\tilde{a}_2^{sc})}$$  \hspace{1cm} (E1)$$

$$= e^{i\tilde{k}^{sc}(m_1\tilde{a}_1^{sc} + m_2\tilde{a}_2^{sc})}$$  \hspace{1cm} (E2)$$

$$= e^{i\tilde{k}^{sc}\tilde{T}^{sc}}$$  \hspace{1cm} (E3)$$

for distinct $p_i$ integers. Various crystal momentum states in the original 1×1 unit cell description are thus mapped onto the “folded” $\tilde{k}^{sc}$ crystal momenta where the state is labeled in the sBZ. The perturbation from CDW deformation introduces coupling and mixing between these momentum states. Because the states and bands are folded in the sBZ, for each $\tilde{k}^{sc}$ there are nine times the number of bands in the sBZ than the original BZ description. In the repeated sBZ scheme, the energy contour at Fermi energy for the monolayer with CDW is shown in Fig. 6(b). The topography of the energy contour from the band folding is substantially more complicated than that of the pristine crystal without CDW, shown in Fig. 5. The band structure in Fig. 6(c) along the momentum line as in Fig. 6(a) also shows the supercell bands that are drastically different from those of the pristine crystal.

As shown in Fig. 5, the band folding can be traced back to the associated phase factor and the crystal momentum of the Bloch theorem. Within the supercell description, the crystal momentum $\tilde{k}^{sc}$ is defined under the supercell translation $\tilde{T}^{sc}$. As a result, the translational properties under the smaller original primitive vectors are not resolved with the $\tilde{k}^{sc}$ supercell crystal momentum labeling. Any state with $\tilde{k} = \tilde{k}^{sc} + p_1\tilde{G}_1^{sc} + p_2\tilde{G}_2^{sc}$ (original crystal momentum defined with translation $\tilde{T}$ spanned by primitive vector $\tilde{a}_i$) is compatible with the supercell crystal momentum $\tilde{k}^{sc}$, with the same reasoning as in Eq. (E1). However, in many cases such as CDW order, there is weak translational symmetry breaking, and the translational symmetry for the 1×1 unit cell can be treated as the approximate symmetry of the crystal. That is, we can decompose a supercell Bloch state or an energy eigenstate at $\tilde{k}^{sc}$ into components with distinct translational properties under the original $\tilde{T}$ and label them with $\tilde{k} = \tilde{k}^{sc} + q_1\tilde{G}_1^{sc} + q_2\tilde{G}_2^{sc}$:

$$\psi_k^{sc} = \sum_{\tilde{k} \in \text{sBZ}} \psi_{\tilde{k}}^{sc},$$  \hspace{1cm} (E4)$$

which has nine components in the 3×3 supercell. In the perturbative regime, we can start with the pristine crystal without CDW order and the translational symmetry under $\tilde{T}$ is recovered. This translation symmetry eliminates the mixings between distinct $\tilde{k}$ components classified under the same supercell $\tilde{k}^{sc}$ CDW order breaks the $\tilde{T}$ translation symmetry and induces mixing between $\tilde{k}$ components. To compare and contrast with the original pristine crystal band structure in the repeated zone scheme, the $\psi_{\tilde{k} \in \text{sBZ}}$ component is singled out and the unfolding weight for the state is defined as the norm of this specific component $|\psi_{\tilde{k}}^{sc}|^2$. When the supercell band structure is
plotted with the unfolding weight for each state as in Fig. 6(d), the band structure with weights is greatly simplified and they are defined as the unfolded band structure. These unfolded and weighted bands are then compared with the pristine bands without CDW in Fig. 3(a). Clearly, these unfolded CDW bands introduce additional effective electronic features to the pristine band structure such as the splittings and the gap opening from CDW order. The same unfolding procedure can also be applied to the Fermi energy contour change due to CDW as in Fig. 3(d).

APPENDIX F: PHONON DISPERSION

For TaS$_2$ crystals, the phonon properties are calculated within density functional perturbation theory, using the VASP code with the PHONOPY package [55]. A $6 \times 6 \times 2 (8 \times 8 \times 1)$ supercell geometry is employed for the bulk (monolayer) crystal, with the calculated phonon spectrum and the density of states shown in Fig. 7. We find that the average phonon frequency slightly decreases for the monolayer case, with squared phonon frequency ($\omega^2$) changing from 58.12 THz$^2$ in bulk to 56.59 THz$^2$ in monolayer.

APPENDIX G: McMillan’s Formalism

In McMillan’s theory [45], the critical temperature is expressed by Eq. (1), where $\mu^*$ is the Coulomb pseudopotential of Morel and Anderson given by

$$\mu^* = \frac{\text{DOS}(E_F)(V_c)}{1 + \text{DOS}(E_F)(V_c) \ln \left( \frac{E_F - \mu^*}{\Delta} \right)},$$  \hspace{1cm} (G1)$$

and $\lambda$ is the electron-phonon coupling constant

$$\lambda = \frac{\text{DOS}(E_F)(I^2)}{M \langle \omega^2 \rangle},$$  \hspace{1cm} (G2)$$

with DOS($E_F$) the density of states, $V_c$ the matrix element of the screened Coulomb interaction, $E_B$ the electronic bandwidth, $\omega$ ($\omega_0$) the (maximum) phonon frequency, $I$ the electron-phonon matrix elements, $M$ the atom mass, and $\langle \cdots \rangle$ the average over the Fermi surface. Assuming $\mu^* = 0.15$ as suggested by McMillan, one can evaluate the inverted form of the above equation and obtain $\lambda = 0.482$ for TaS$_2$ by assuming $\Theta_D = 250$ K [56] and $T_c = 0.8$ K [19], indicating that TaS$_2$ lies in the intermediate coupling regime.

We then investigate the impact of progressive weakening of the CDW by varying the magnitude of atomic distortion. A scaling factor, from 1 to 0, is used to define the fraction by which the magnitude of the atomic displacement is reduced with respect to the stable distorted configuration. The corresponding DOS as a function of atom displacement amplitude is calculated by DFT. Using the bulk as a starting point, we take into account the change in DOS($E_F$) and a small phonon energy shift ($\omega^2$) calculated for the monolayer, and predict $T_c$ within the McMillan formalism.

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