



This is the accepted manuscript made available via CHORUS. The article has been published as:

Charge density waves in the 2.5-dimensional quantum heterostructure

F. Z. Yang, T. T. Zhang, F. Y. Meng, H. C. Lei, C. Nelson, Y. Q. Cai, E. Vescovo, A. H. Said, P. M. Lozano, G. Fabbri, and H. Miao

Phys. Rev. B **111**, L041101 — Published 2 January 2025

DOI: [10.1103/PhysRevB.111.L041101](https://doi.org/10.1103/PhysRevB.111.L041101)

Charge Density Waves in the 2.5-Dimensional Quantum Heterostructure

F. Z. Yang,¹ T. T. Zhang,² F. Y. Meng,^{3,4} H. C. Lei,^{3,4} C. Nelson,⁵ Y. Q. Cai,⁵
E. Vescovo,⁵ A. H. Said,⁶ P. Mercado Lozano,⁶ G. Fabbri,⁶ and H. Miao^{1,*}

¹*Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA*

²*Institute of Theoretical Physics, Chinese Academy of Sciences, Beijing 100190, China*

³*Department of Physics and Beijing Key Laboratory of Opto-electronic Functional Materials & Micro-nano Devices, Renmin University of China, Beijing 100872, China*

⁴*Key Laboratory of Quantum State Construction and Manipulation (Ministry of Education), Renmin University of China, Beijing 100872, China*

⁵*National Synchrotron Light Source II, Brookhaven National Laboratory, Upton, New York 11973, USA*

⁶*Advanced Photon Source, Argonne National Laboratory, Lemont, Illinois 60439, USA*

(Dated: December 5, 2024)

Charge density wave (CDW) and their interplay with correlated and topological quantum states are forefront of condensed matter research. The $4H_b$ -TaS₂ is a CDW ordered quantum heterostructure that is formed by alternative stacking of Mott insulating 1T-TaS₂ and Ising superconducting 1H-TaS₂. While the $\sqrt{13} \times \sqrt{13}$ and 3×3 CDWs have been respectively observed in the bulk 1T-TaS₂ and 2H-TaS₂, the CDWs and their pivotal role for unconventional superconductivity in the $4H_b$ -TaS₂ remain unsolved. In this letter, we reveal the 2-dimensional (2D) $\sqrt{13} \times \sqrt{13}$ chiral CDW in the 1T-layers and intra-unit cell coupled 2D 2×2 CDW in the 1H and 1H' layers of $4H_b$ -TaS₂. Our results establish $4H_b$ -TaS₂ a novel 2.5D quantum heterostructure, where 2D quantum states emerge from 3D crystalline structure.

Electrons confined in 2-dimensional (2D) space can give rise to unprecedented quantum phases, including integer and fractional quantum Hall liquids [1, 2] and topological superconductivity [3, 4]. While 2D electronic states can be created by making 2D lattice structures such as thin films [5], interfaces [6, 7], and exfoliated thin flakes [8], its realization in 3D crystalline materials remains an unresolved challenge. Recently, the 3D stacking of heterostructured transition metal dichalcogenides is emerging as a promising platform hosting unusual 2D electronic orders [9–17]. A prototypical example is the $4H_b$ -TaS₂, which is formed by periodic stacking of 1H and 1T-TaS₂ layers and features cascade of symmetry-breaking orders [9–12, 18]. Figure 1a shows the crystal structure and 3×3 charge density wave (CDW) of the monolayer 1H-TaS₂. Due to the absence of inversion symmetry, $\mathcal{P}$, the Ising-type spin-orbital coupling (SOC) lifts the spin degeneracy near the K and K' point in the momentum space (Fig. 1b) [19, 20] and gives rise to Ising superconductivity below $T_{SC} \sim 3$ K [21–25]. Figure 1c shows the structure of $\mathcal{P}$ -preserving 1T-TaS₂ that hosts $\sqrt{13} \times \sqrt{13}$ CDW and star-of-David (SoD) lattice distortions. The CDW induced flatband is pushed away from the Fermi level by developing a Mott gap, which may support a quantum spin liquid ground state (Fig. 1d) [26, 27]. By adding the stacking degree of freedom to combine the CDW ordered 1H and 1T-TaS₂ layers, the $4H_b$ -TaS₂ offers a unique opportunity to realize 2D topological quantum phases in 3D crystal structures [9, 10, 18, 28–30]. Here, using hard x-ray diffraction (XRD), we show the emergence of 2D CDW in the 1T-layers that is sandwiched between intra-unit cell coupled 2D 2×2 CDWs

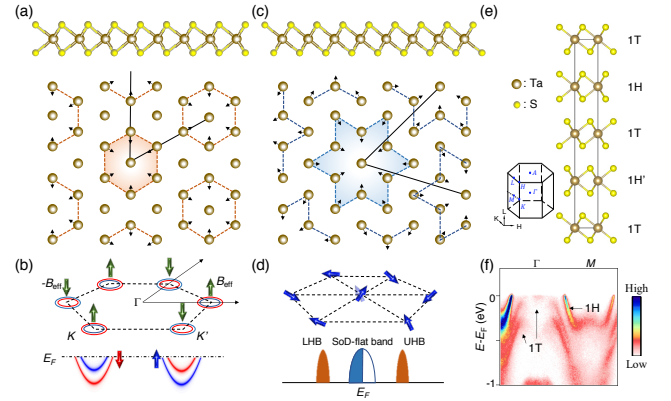


Figure 1. (a) Side view of monolayer 1H-TaS₂ that breaks $\mathcal{P}$. Top view of Ta triangular lattice that displays 3×3 CDW [32, 33]. (b) The electronic structure of 1H-TaS₂. The Ising-type SOC induces effective local magnetic field, B_{eff} , in the momentum space that lifts the spin degeneracy. (c) Side view of monolayer 1T-TaS₂ that preserves $\mathcal{P}$. (d) The SoD CDW induced band folding results in single-occupied flat band that eventually forms the upper Hubbard band (UHB) and lower Hubbard band (LHB). (e) Crystal structure of $4H_b$ -TaS₂. (f) ARPES determined electronic structure confirms the coexistence of 1H and 1T electronic states.

in the 1H and 1H' layers. The unconventional 2D CDW structures yield Josephson coupled bilayer superconductivity at low temperature that can host orbital driven finite momentum pairing under external magnetic field [31].

Figure 1f shows the ARPES determined electronic structure of $4H_b$ -TaS₂ along the Γ –M direction, confirming the coexistence of 1H and 1T electronic states [10, 34]. Due to the charge transfer between 1H and 1T layers, the

* miaoh@ornl.gov

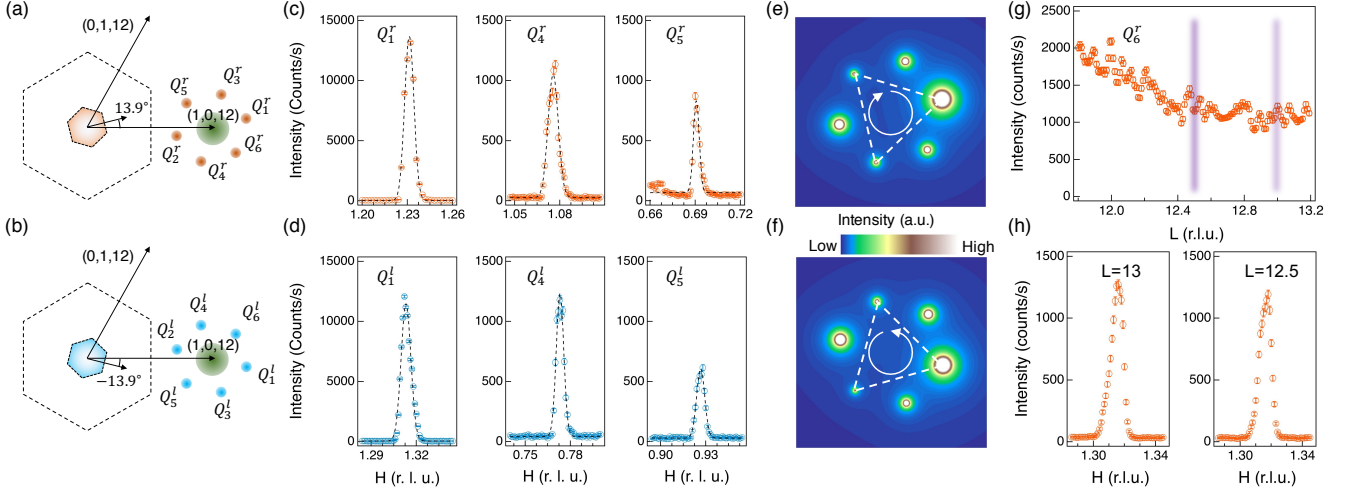


Figure 2. (a) and (b) depicts Brillouin zones of undistorted 1T-TaS₂ (dashed hexagon), right-handed CDW phase (pink hexagon), and left-handed CDW phase (blue hexagon). $Q^{r/l} = q^{r/l} + \mathbf{G}$ represent the right and left-handed CDW superlattice peak positions near the $\mathbf{G}=(0, 1, 12)$ fundamental Bragg peak. (c) and (d) shows the H-scan of CDW superlattice peaks at 300 K. Dashed lines are fittings of the CDW peak intensity using a Gaussian function. (e) and (f) summarize the right and left-handed CDW peak intensity distributions in the HK scattering plane. Both the peak positions and intensity distributions of the right and left-handedness are related by M_y mirror symmetry. The white triangles connect CDW superlattice peaks, whose intensity distribution reflects the handedness of the CDW domains. (g) and (h) show L and H scans at Q_6^r , establishing 2D CDW in the 1T layers. Error bars represent 1 standard deviation assuming Poisson counting statistics.

Mott gap in the 1T layer is collapsed leaving a flat band on the Fermi level. Figure 2 shows the $\sqrt{13} \times \sqrt{13}$ CDW superlattice peaks in the 1T layers measured at 300 K below the transition temperature at 315 K [32, 33]. As shown in Fig. 1e, the SoD lattice distortions break in-plane mirror symmetries giving rise to right-handed and left-handed domains as depicted in Fig. 2a and b. For the right-handed CDW domains, the CDW peaks are located at $q_1^r = -q_2^r = (3/13, 1/13)$, $q_3^r = -q_4^r = (-1/13, 4/13)$, and $q_5^r = -q_6^r = (-4/13, 3/13)$ in the reciprocal lattice unit (r.l.u.). For the left-handed CDW domains, the CDW peaks are located at $q_1^l = -q_2^l = (4/13, -1/13)$, $q_3^l = -q_4^l = (3/13, -4/13)$, and $q_5^l = -q_6^l = (-1/13, -3/13)$ r.l.u. Here $q^{r/l}$ are reduced momentum transfers that are related to $Q^{r/l}$ (Fig. 2a and b) by the reciprocal lattice unit, $\mathbf{G} = (0, 1, 12)$. Figures 2c and d show the CDW superlattice peaks of the right and left-handed domains. Dashed curves are fittings using a Gaussian function that yield half-width-at-half-maximum (HWHM) ~ 0.004 r.l.u. and in-plane CDW correlation length, $\xi_{//} = 1/\text{HWHM} \sim 130$ Å. Figures 2e and f depict the extracted right and left-handed CDW peak intensities. These plots are constructed based on experimentally determined peak widths and intensities [35]. Both superlattice peak positions and intensities of the left and right-handed domains are related by the mirror, M_y , symmetry, as expected for the scattering near the $(0, 1, 12)$ Bragg peak. Interestingly, we find that the handedness of the CDW domain is encoded in the diffraction intensities. As highlighted by the white dashed triangles in Fig 2e and f, the peak intensity of q_1^r , q_4^r , and q_5^r decreases clockwise (Fig. 2c), whereas the

peak intensity of q_1^l , q_4^l , and q_5^l decreases counterclockwise (Fig. 2d). These handed peak intensities arise from the CDW-induced SoD lattice distortions and can be reproduced by calculating relative x-ray scattering amplitude [35]. Most surprisingly, we find that despite the intermediate CDW correlation length in the HK plane, the CDW is decoupled along the crystalline c-axis as revealed by the L -scan shown in Fig. 2g and H-scans at $L=13$ and 12.5 shown in Fig. 2h. These observations are in stark contrast with bulk 1T-TaS₂ that forms 3D CDW [32, 36], and hence establish a rare 2D CDW in 3D structure.

We now focus on CDWs in the 1H layers. Figure 3a shows the XRD intensity along the $(H, 0, 12)$ direction at 10 K. Previous scanning tunneling microscopy studies of $4H_b$ -TaS₂ show 3×3 CDW on the surface 1H-TaS₂ [28, 37], in agreement with monolayer 1H-TaS₂ and bulk 2H-TaS₂ [32, 33]. Unexpectedly, our bulk sensitive XRD shows CDW superlattice peaks at $H = \text{Integer} + 0.5$, corresponding to 2×2 CDW in the 1H layers of $4H_b$ -TaS₂. The insets of Fig. 3a show x-ray diffraction intensity near $(0.5, -0.5, 12)$ and along the $(1.5, 0, L)$ direction. The extracted in-plane and out-plane correlation lengths are $\xi_{//} \sim 170$ Å and $\xi_{\perp} \sim 13$ Å, respectively. While the 2×2 CDW in the 1H layers displays a peak at $L=12$, the short $\xi_{\perp}$ reveals an in-phase and intra-unit cell coupled 2×2 CDW between the 1H and 1H' layers. Figures 3b and c show the temperature dependence of CDW peak intensity and peak width, respectively. In agreement with resistivity measurement (Fig. 4d) [38], we find that the CDW peak intensity displays a sudden jump at $T_{1H}=20$ K. Warming (yellow) and cooling (cyan) of the sample reveal

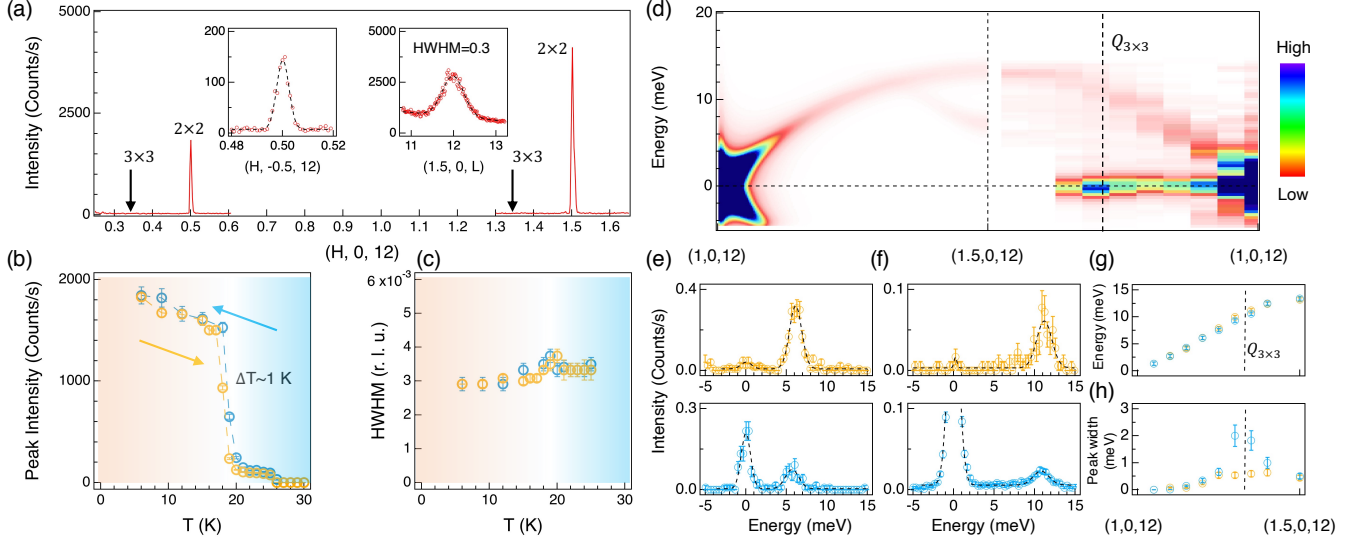


Figure 3. (a) XRD along the $(H, 0, 12)$ direction at $T = 10$ K shows 2×2 CDW. Inset shows the K -scan near the $(0.5, -0.5, 12)$ superlattice peak position and L -scan near the $(1.5, 0, 12)$ superlattice peak position. Dashed lines are Gaussian function fittings of the experimental data. The extracted in-plane and out-plane correlation lengths are 170 and 13 Å, respectively. (b) and (c) show the temperature-dependent peak intensity and HWHM, respectively. Yellow and cyan data were collected by warming and cooling respectively. Since the 2×2 CDW is broadly peaked at $L = \text{even integers}$, the intra-unit cell coupled CDW in the 1H and 1H' layers are in-phase. The 1 K temperature hysteresis supports a weak first-order transition. (d) compares DFT calculated (left) and IXS determined $S(Q, \omega)$ at 30 K. (e) and (f) show IXS spectra at $q = 0.25$ and $q = 0.33$, respectively. (g) and (h) show extracted phonon dispersion and peak width, respectively. Yellow and cyan in (e)-(h) represent $T = 300$ and 30 K, respectively. The error bars are standard deviation of the fitting parameters.

small hysteresis, $\Delta T \sim 1$ K, confirming a weak first-order phase transition [39–42]. Interestingly, despite the significantly suppressed peak intensity above $T_{1H} = 20$ K, the CDW peak remains observable until $T^* = 25$ K. As we discuss later, we attribute the persistent CDW to competing CDW instabilities in the 1H layers.

To understand the origin of 2×2 CDW in the 1H layers, we determine the low energy phonon dynamical structure factor, $S^{DFT}(Q, \omega)$. Figure 3d shows the comparison of the density functional theory (DFT) calculated $S^{DFT}(Q, \omega)$ of 1H-TaS₂ and experimental $S(Q, \omega)$ measured by meV resolution inelastic x-ray scattering (IXS) at $T = 30$ K. The inelastic phonon band corresponds to the transverse acoustic phonon. The overall good agreement between experiment and DFT calculation confirms that the experimental spectra is dominated by the lattice excitations from the 1H layers. Figure 3e-f compares IXS spectra measured at $Q = (1.25, 0, 12)$ and $(1.33, 0, 12)$. Yellow and cyan curves represent data collected at room temperature and $T = 30$ K, respectively. Black dashed lines are fittings of the experimental data. The extracted phonon dispersion and phonon peak width are respectively shown in Figs. 3g and h. Interestingly, we find that while the phonon dispersion remains temperature-independent in our experimental geometry, we observe enhanced phonon anharmonicity near the 3×3 CDW wavevector. This observation suggests competing 3×3 and 2×2 CDW instabilities in the 1H layer of $4H_b$ -TaS₂.

To confirm this scenario, we show DFT calculated

phonon dispersion of 1H-TaS₂, 0.25 electron/Ta doped 1H-TaS₂, and 0.5 electron/Ta doped 1H-TaS₂ in Figs. 4a-c. In the monolayer 1H-TaS₂, phonon dispersion displays lattice instabilities at both 3×3 and 2×2 wavevectors, where the negative phonon mode reaches global minimum at the 3×3 wavevector, consistent with the leading 3×3 CDW in monolayer 1H-TaS₂. However, we find that in the 0.25 electron/Ta doped 1H-TaS₂, 3×3 CDW is completely suppressed, leaving 2×2 CDW as the leading instability. The evolution of the phonon energy minimum suggests that the electron doping of the 1H-TaS₂ will first enhance the competition between the 3×3 and 2×2 CDWs, then favor the 2×2 CDW, and eventually kills all CDW instabilities. In $4H_b$ -TaS₂, the electron charge transfer from the 1T to 1H layers is less than 1/13 electron/Ta, supporting the nearly degenerate 3×3 and 2×2 CDWs in $4H_b$ -TaS₂.

Figure 4e depicts the side view of competing 3×3 and 2×2 CDWs in the 1H layers. The competing lattice distortions induced by 3×3 and 2×2 CDWs are highlighted in the dashed rectangles. This competition frustrates the lattice structure and yields enhanced lattice fluctuations in a way similar to frustrated spin systems. Consequently, the CDW order in the 1H layer is expected to be fragile. To confirm this expectation, we show the resistivity curve and XRD of Se doped $4H_b$ -TaS_{1.99}Se_{0.01} in Fig. 4d. Remarkably, the 1% Se doping completely melts CDW in the 1H layers, resulting in a homogeneous Fermi liquid normal state. This observation is in strong

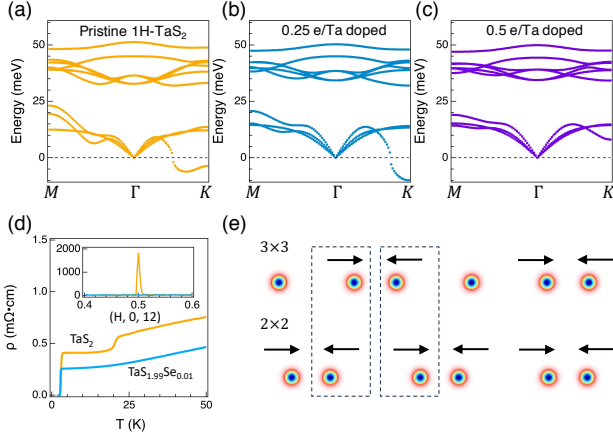


Figure 4. (a)-(c) Calculated phonon dispersions of pristine 1H-TaS₂ (yellow), 0.25 electron/Ta doped 1H-TaS₂ (blue), and 0.5 electron/Ta doped 1H-TaS₂ (purple). (d) Resistivity and x-ray diffraction of 4H_b-TaS₂ (yellow) and 4H_b-TaS_{1.99}Se_{0.01} (cyan). The CDW is completely suppressed by 1% Se substitution of S. (e) Schematics of competing 3 × 3 and 2 × 2 CDWs in the Ta atoms in 1H layers.

contrast with the $\sqrt{13} \times \sqrt{13}$ CDW in the 1T layers, which is robust against change transfer and Se doping. This lattice frustration picture also explains the weak CDW peak between 20 and 25 K, where the CDW transition temperature is sensitive to the local environment, such as strain force and defects. Similarly, the competing CDW scenario reconciles the different CDW patterns on the surface [28, 30].

Our observations of 2D handed CDWs in the 1T layers and intra-unit cell coupled 2D 2 × 2 CDW in the 1H and 1H' layers make 4H_b-TaS₂ as a novel 2.5-dimensional (2.5D) quantum heterostructure [43]. As shown in Fig. 1f, the 1T layer is a doped Mott-insulator with dilute electron carriers. The 1T layers thus provide weak screening of the inter-layer Coulomb interactions and allow intra-unit cell coupled CDWs between 1H and 1H' layers. The 1H/1H' layers, on the other hand, are good metals with large Fermi surface. Therefore, the Coulomb interactions between 1T layers are significantly suppressed, resulting in 2D CDW localized on each 1T layer. It is important to note that even for the well-known van der Waals materials, including the cuprate high T_c -superconductors [44], transition metal dichalcogenides [45–47], and AV₃Sb₅ kagome superconductors [39], the CDWs are 3D or quasi-2D. Our observations thus establish a rare realization of pure 2D electronic states in a 3D

crystal structure.

The emergence of the 2D electronic states in the 3D bulk structure has significant implications for low-temperature quantum states. The isolated $\sqrt{13} \times \sqrt{13}$ CDW in the 1T-layers yields 2D flat band that is pinned at the E_F [34, 48]. The 2D nature of the 1T layers provides the possibility to realize the Mott physics and quantum spin liquid in a 3D bulk crystal 4H_b-TaS₂. The experimental observation of flat band on the Fermi level and handed CDW provide electronic and symmetry constraints to realize unconventional superconductivity, including the $\mathcal{T}$ -breaking superconducting pairing [9, 10, 28, 29]. On the other hand, the intra-unit cell coupled CDW forms an effective 2D bilayer structure, which has been predicted to host orbital Fulde-Ferrell-Larkin-Ovchinnikov state [31, 49–51]. Furthermore, the formation of intra-unit cell 1H-1H' bilayer breaks the global mirror, M_z , symmetry of 4H_b-TaS₂ allowing the Lifshitz invariant term in the Ginzburg-Landau free energy [52]. This Lifshitz invariant can stabilize finite momentum pairing state in 3D systems under external magnetic field [18].

In summary, we revealed 2D handed CDW in the 1T-layers and intra-unit cell coupled 2D CDW in the 1H layers. Our results highlight 4H_b-TaS₂ as a 2.5D quantum heterostructure that can host emergent macroscopic quantum phases.

Acknowledgments: We thank Gang Chen, Hong Ding, Patrick A. Lee, Chao-Xing Liu, Lingyuan Kong, Wenya Liu, Qiangsheng Lu, Ziqiang Wang, Binghai Yan, Jiaqiang Yan, Noah F. Q. Yuan, Rui-Xing Zhang, and Yang Zhang for stimulating discussions. This research was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Sciences and Engineering Division (X-ray and ARPES). X-ray scattering used resources (beamline 4-ID and 30-ID) of the Advanced Photon Source, a U.S. DOE Office of Science User Facility operated for the DOE Office of Science by Argonne National Laboratory under Contract No. DE-AC02-06CH11357. ARPES and X-ray scattering measurements used resources at 21-ID-1, 4-ID and 10-ID beamlines of the National Synchrotron Light Source II, a US Department of Energy Office of Science User Facility operated for the DOE Office of Science by Brookhaven National Laboratory under contract no. DE-SC0012704. H.C.L. was supported by Beijing Natural Science Foundation (Grant No. Z200005), National Key R&D Program of China (Grants No. 2022YFA1403800, No. 2023YFA1406500), National Natural Science Foundation of China (Grants No. 12274459).

-
- [1] K. v. Klitzing, G. Dorda, and M. Pepper, New method for high-accuracy determination of the fine-structure constant based on quantized hall resistance, *Phys. Rev. Lett.* **45**, 494 (1980).
 [2] D. C. Tsui, H. L. Stormer, and A. C. Gossard, Two-

- dimensional magnetotransport in the extreme quantum limit, *Phys. Rev. Lett.* **48**, 1559 (1982).
 [3] L. Fu and C. L. Kane, Superconducting proximity effect and majorana fermions at the surface of a topological insulator, *Phys. Rev. Lett.* **100**, 096407 (2008).

- [4] X.-L. Qi and S.-C. Zhang, Topological insulators and superconductors, *Rev. Mod. Phys.* **83**, 1057 (2011).
- [5] C.-Z. Chang, J. Zhang, X. Feng, J. Shen, Z. Zhang, M. Guo, K. Li, Y. Ou, P. Wei, L.-L. Wang, *et al.*, Experimental observation of the quantum anomalous hall effect in a magnetic topological insulator, *Science* **340**, 167 (2013).
- [6] A. Ohtomo and H. Hwang, A high-mobility electron gas at the $\text{LaAlO}_3/\text{SrTiO}_3$ heterointerface, *Nature* **427**, 423 (2004).
- [7] D. Hsieh, D. Qian, L. Wray, Y. Xia, Y. S. Hor, R. J. Cava, and M. Z. Hasan, A topological Dirac insulator in a quantum spin Hall phase, *Nature* **452**, 970 (2008).
- [8] K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. Grigorieva, and A. A. Firsov, Electric Field Effect in Atomically Thin Carbon Films, *Science* **306**, 666 (2004).
- [9] E. Persky, A. V. Björliig, I. Feldman, A. Almoalem, E. Altman, E. Berg, I. Kimchi, J. Ruhman, A. Kanigel, and B. Kalisky, Magnetic memory and spontaneous vortices in a van der waals superconductor, *Nature* **607**, 692 (2022).
- [10] A. Ribak, R. M. Skiff, M. Mograbi, P. K. Rout, M. H. Fischer, J. Ruhman, K. Chashka, Y. Dagan, and A. Kanigel, Chiral superconductivity in the alternate stacking compound 4Hb-TaS_2 , *Science Advances* **6**, eaax9480 (2020).
- [11] I. Silber, S. Mathimalar, I. Mangel, A. Nayak, O. Green, N. Avraham, H. Beidenkopf, I. Feldman, A. Kanigel, A. Klein, *et al.*, Two-component nematic superconductivity in 4hb-tas_2 , *Nature Communications* **15**, 824 (2024).
- [12] A. Almoalem, I. Feldman, I. Mangel, M. Shlafman, Y. E. Yaish, M. H. Fischer, M. Moshe, J. Ruhman, and A. Kanigel, The observation of π -shifts in the little-parks effect in 4hb-tas_2 , *Nature Communications* **15**, 4623 (2024).
- [13] A. Devarakonda, A. Chen, S. Fang, D. Graf, M. Kriener, A. Akey, D. Bell, T. Suzuki, and J. Checkelsky, Evidence of striped electronic phases in a structurally modulated superlattice, *Nature* **631**, 526 (2024).
- [14] S.-B. Liu, C. Tian, Y. Fang, H. Rong, L. Cao, X. Wei, H. Cui, M. Chen, D. Chen, Y. Song, *et al.*, Nematic ising superconductivity with hidden magnetism in few-layer 6r-tas_2 , *Nature Communications* **15**, 7569 (2024).
- [15] Z. Wan, G. Qiu, H. Ren, Q. Qian, Y. Li, D. Xu, J. Zhou, J. Zhou, B. Zhou, L. Wang, *et al.*, Unconventional superconductivity in chiral molecule- tas_2 hybrid superlattices, *Nature* **632**, 69 (2024).
- [16] A. Devarakonda, T. Suzuki, S. Fang, J. Zhu, D. Graf, M. Kriener, L. Fu, E. Kaxiras, and J. Checkelsky, Signatures of bosonic landau levels in a finite-momentum superconductor, *Nature* **599**, 51 (2021).
- [17] A. Devarakonda, H. Inoue, S. Fang, C. Ozsoy-Keskinbora, T. Suzuki, M. Kriener, L. Fu, E. Kaxiras, D. C. Bell, and J. G. Checkelsky, Clean 2d superconductivity in a bulk van der waals superlattice, *Science* **370**, 231 (2020).
- [18] F. Z. Yang, H. D. Zhang, S. Mandal, F. Y. Meng, G. Fabbri, A. Said, P. M. Lozano, A. Rajapitamahuni, E. Vescovo, C. Nelson, S. Lin, Y. Park, E. M. Clements, T. Z. Ward, H. N. Lee, H. C. Lei, C. X. Liu, and H. Miao, Signature of orbital driven finite momentum pairing in a 3d ising superconductor (2024), arXiv:2407.10352 [cond-mat.supr-con].
- [19] D. Xiao, W. Yao, and Q. Niu, Valley-contrasting physics in graphene: Magnetic moment and topological transport, *Phys. Rev. Lett.* **99**, 236809 (2007).
- [20] X. Xu, W. Yao, D. Xiao, and T. F. Heinz, Spin and pseudospins in layered transition metal dichalcogenides, *Nature Physics* **10**, 343 (2014).
- [21] J. M. Lu, O. Zheliuk, I. Leermakers, N. F. Q. Yuan, U. Zeitler, K. T. Law, and J. T. Ye, Evidence for two-dimensional ising superconductivity in gated MoS_2 , *Science* **350**, 1353 (2015), doi: 10.1126/science.aab2277.
- [22] X. Xi, Z. Wang, W. Zhao, J.-H. Park, K. T. Law, H. Berger, L. Forró, J. Shan, and K. F. Mak, Ising pairing in superconducting NbSe_2 atomic layers, *Nature Physics* **12**, 139 (2016).
- [23] W. Li, J. Huang, X. Li, S. Zhao, J. Lu, Z. V. Han, and H. Wang, Recent progresses in two-dimensional ising superconductivity, *Materials Today Physics* **21**, 100504 (2021).
- [24] H. Zhang, A. Rousuli, K. Zhang, L. Luo, C. Guo, X. Cong, Z. Lin, C. Bao, H. Zhang, S. Xu, R. Feng, S. Shen, K. Zhao, W. Yao, Y. Wu, S. Ji, X. Chen, P. Tan, Q.-K. Xue, Y. Xu, W. Duan, P. Yu, and S. Zhou, Tailored ising superconductivity in intercalated bulk NbSe_2 , *Nature Physics* **18**, 1425 (2022).
- [25] S. C. de la Barrera, M. R. Sinko, D. P. Gopalan, N. Sivasada, K. L. Seyler, K. Watanabe, T. Taniguchi, A. W. Tsen, X. Xu, D. Xiao, and B. M. Hunt, Tuning ising superconductivity with layer and spin-orbit coupling in two-dimensional transition-metal dichalcogenides, *Nature Communications* **9**, 1427 (2018).
- [26] K. T. Law and P. A. Lee, 1T-TaS_2 as a quantum spin liquid, *Proceedings of the National Academy of Sciences* **114**, 6996 (2017).
- [27] W. Ruan, Y. Chen, S. Tang, J. Hwang, H.-Z. Tsai, R. L. Lee, M. Wu, H. Ryu, S. Kahn, F. Liou, C. Jia, A. Aikawa, C. Hwang, F. Wang, Y. Choi, S. G. Louie, P. A. Lee, Z.-X. Shen, S.-K. Mo, and M. F. Crommie, Evidence for quantum spin liquid behaviour in single-layer 1T-TaSe_2 from scanning tunnelling microscopy, *Nature Physics* **17**, 1154 (2021).
- [28] A. K. Nayak, A. Steinbok, Y. Roet, J. Koo, G. Margalit, I. Feldman, A. Almoalem, A. Kanigel, G. A. Fiete, B. Yan, Y. Oreg, N. Avraham, and H. Beidenkopf, Evidence of topological boundary modes with topological nodal-point superconductivity, *Nature Physics* **17**, 1413 (2021).
- [29] R. L. Luo and G. V. Chen, Is spontaneous vortex generation in superconducting 4Hb-TaS_2 from vison-vortex nucleation with z_2 topological order?, *Quantum Frontiers* **3**, 10 (2024).
- [30] C. Wen, J. Gao, Y. Xie, Q. Zhang, P. Kong, J. Wang, Y. Jiang, X. Luo, J. Li, W. Lu, Y.-P. Sun, and S. Yan, Roles of the narrow electronic band near the fermi level in 1T-tas_2 -related layered materials, *Phys. Rev. Lett.* **126**, 256402 (2021).
- [31] C.-X. Liu, Unconventional superconductivity in bilayer transition metal dichalcogenides, *Phys. Rev. Lett.* **118**, 087001 (2017).
- [32] R. Brouwer and F. Jellinek, The low-temperature superstructures of 1T-TaSe_2 and 2H-TaSe_2 , *Physica B+C* **99**, 51 (1980).
- [33] G. A. Scholz, O. Singh, R. F. Frindt, and A. E. Curzon, Charge density wave commensurability in 2H-TaS_2 and AgxTaS_2 , *Solid State Communications* **44**, 1455 (1982).
- [34] A. Almoalem, R. Gofman, Y. Nitzav, I. Mangel,

- I. Feldman, J. Koo, F. Mazzola, J. Fujii, I. Vobornik, J. S'anchez-Barriga, O. J. Clark, N. C. Plumb, M. Shi, B. Yan, and A. Kanigel, Charge transfer and spin-valley locking in 4Hb-TaS₂, *npj Quantum Materials* **9**, 36 (2024).
- [35] See Supporting Information for further details.
- [36] B. BURK, R. E. THOMSON, J. CLARKE, and A. ZETTL, Surface and bulk charge density wave structure in 1 t-tas₂, *Science* **257**, 362 (1992).
- [37] S. Shen, T. Qin, J. Gao, C. Wen, J. Wang, W. Wang, J. Li, X. Luo, W. Lu, Y. Sun, and S. Yan, Coexistence of quasi-two-dimensional superconductivity and tunable kondo lattice in a van der waals superconductor, *Chinese Physics Letters* **39**, 077401 (2022).
- [38] F. Meng, Y. Fu, S. Pan, S. Tian, S. Yan, Z. Li, S. Wang, J. Zhang, and H. Lei, Extreme orbital *ab*-plane upper critical fields far beyond the pauli limit in 4H_b-Ta(S,Se)₂ bulk crystals, *Phys. Rev. B* **109**, 134510 (2024).
- [39] H. Li, T. T. Zhang, T. Yilmaz, Y. Y. Pai, C. E. Marvinney, A. Said, Q. W. Yin, C. S. Gong, Z. J. Tu, E. Vescovo, C. S. Nelson, R. G. Moore, S. Murakami, H. C. Lei, H. N. Lee, B. J. Lawrie, and H. Miao, Observation of unconventional charge density wave without acoustic phonon anomaly in kagome superconductors AV₃Sb₅ (*A* = Rb, Cs), *Phys. Rev. X* **11**, 031050 (2021).
- [40] H. Miao, H. X. Li, W. R. Meier, A. Huon, H. N. Lee, A. Said, H. C. Lei, B. R. Ortiz, S. D. Wilson, J. X. Yin, M. Z. Hasan, Z. Wang, H. Tan, and B. Yan, Geometry of the charge density wave in the kagome metal AV₃Sb₅, *Phys. Rev. B* **104**, 195132 (2021).
- [41] H. Li, G. Fabbri, A. H. Said, J. P. Sun, Y.-X. Jiang, J.-X. Yin, Y.-Y. Pai, S. Yoon, A. R. Lupini, C. S. Nelson, Q. W. Yin, C. S. Gong, Z. J. Tu, H. C. Lei, J.-G. Cheng, M. Z. Hasan, Z. Wang, B. Yan, R. Thomale, H. N. Lee, and H. Miao, Discovery of conjoined charge density waves in the kagome superconductor CsV₃Sb₅, *Nature Communications* **13**, 6348 (2022).
- [42] H. Miao, T. T. Zhang, H. X. Li, G. Fabbri, A. H. Said, R. Tartaglia, T. Yilmaz, E. Vescovo, J.-X. Yin, S. Murakami, X. L. Feng, K. Jiang, X. L. Wu, A. F. Wang, S. Okamoto, Y. L. Wang, and H. N. Lee, Signature of spin-phonon coupling driven charge density wave in a kagome magnet, *Nature Communications* **14**, 6183 (2023).
- [43] Y. M. K. M. M. K. K. U. Hiroki Ago Susumu Okada and K. Nagashio, Science of 2.5 dimensional materials: paradigm shift of materials science toward future social innovation, *Science and Technology of Advanced Materials* **23**, 275 (2022).
- [44] R. Comin and A. Damascelli, Resonant X-Ray Scattering Studies of Charge Order in Cuprates, *Annual Review of Condensed Matter Physics* **7**, 369 (2016).
- [45] S. Tanda, T. Sambongi, T. Tani, and S. Tanaka, X-ray study of charge density wave structure in 1t-tas₂, *Journal of the Physical Society of Japan* **53**, 476 (1984).
- [46] T. Ritschel, J. Trinckauf, K. Koepernik, B. Büchner, M. v. Zimmermann, H. Berger, Y. Joe, P. Abbamonte, and J. Geck, Orbital textures and charge density waves in transition metal dichalcogenides, *Nature physics* **11**, 328 (2015).
- [47] C. Butler, M. Yoshida, T. Hanaguri, and Y. Iwasa, Mottness versus unit-cell doubling as the driver of the insulating state in 1 t-tas₂, *Nature communications* **11**, 2477 (2020).
- [48] H. F. Yang, K. Y. He, J. Koo, S. W. Shen, S. H. Zhang, G. Liu, Y. Z. Liu, C. Chen, A. J. Liang, K. Huang, M. X. Wang, J. J. Gao, X. Luo, L. X. Yang, J. P. Liu, Y. P. Sun, S. C. Yan, B. H. Yan, Y. L. Chen, X. Xi, and Z. K. Liu, Visualization of chiral electronic structure and anomalous optical response in a material with chiral charge density waves, *Phys. Rev. Lett.* **129**, 156401 (2022).
- [49] P. Wan, O. Zheliuk, N. F. Q. Yuan, X. Peng, L. Zhang, M. Liang, U. Zeitler, S. Wiedmann, N. E. Hussey, T. T. M. Palstra, and J. Ye, Orbital Fulde-Ferrell-Larkin-Ovchinnikov state in an Ising superconductor, *Nature* **619**, 46 (2023).
- [50] D. Zhao, L. Debbeler, M. Kühne, S. Fecher, N. Gross, and J. Smet, Evidence of finite-momentum pairing in a centrosymmetric bilayer, *Nature Physics* **19**, 1599 (2023).
- [51] Y.-M. Xie and K. T. Law, Orbital fulde-ferrell pairing state in moiré ising superconductors, *Phys. Rev. Lett.* **131**, 016001 (2023).
- [52] V. M. Edelstein, The ginzburg - landau equation for superconductors of polar symmetry, *Journal of Physics: Condensed Matter* **8**, 339 (1996).