Evidence for clean 2D superconductivity and field-induced finite-momentum pairing in a bulk vdW superlattice

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The recent development of two-dimensional (2D) van der Waals (vdW) materials has enabled the rapid exploration of novel low-dimensional electronic phenomena[1-7]. The family of hexagonal transition metal dichalcogenides (H-MX$_2$) has proven to be a particularly rich host of exotic quantum phases[1]. Owing to their crystal structure and strong spin-orbit coupling, experiments with monolayer and few-layer H-MX$_2$ have demonstrated optical control of valley polarization[2,3], the valley Hall effect[4], and Ising superconductivity[5-7]. However, these materials are often subject to degradation, and for exfoliated materials, reduction in quality during the fabrication process can constrain the phase space for potential ground states[8]. Here we show that high-quality H-NbS$_2$ monolayers with electronic mobilities more than three orders of magnitude larger than in bulk 2H-NbS$_2$ can be realized in a bulk single crystal superlattice formed with a commensurate block layer. We find that these materials are clean-limit 2D superconductors exhibiting a Berezinskii-Kosterlitz-Thouless (BKT) transition at $T_{BKT} = 0.82$ K and prominent 2D Shubnikov de-Haas quantum oscillations. Furthermore, we observe an enhancement of the superconducting upper critical field $\mu_0H_{c2}$ beyond the Pauli limit for field applied within a narrow angular window $\delta\theta \lesssim 2^\circ$ of the layer plane, which we show is consistent with field-induced finite momentum Cooper pairing[9,10] enhanced by local symmetry breaking[11,12]. Our results demonstrate the ability of these commensurate superlattices to support clean monolayer H-MX$_2$ beyond that possible in their bulk 2H-MX$_2$ counterparts and monolayers exfoliated therefrom. Their structure and exfoliability offer pathways to direct probing of pair density wave superconductivity[13] and, more broadly, the possibility of engineering other high quality 2D MX$_2$ layers in a new class of bulk single crystal superlattices.

The fundamental structural unit in hexagonal transition metal dichalcogenides is the H-MX$_2$ layer. As shown in Fig. 1(a), this structure, with point group symmetry $\bar{6}m2$ ($D_{3h}$), breaks inversion symmetry in the layer plane due to the trigonal prismatic coordination of X around M (the missing inversion partner is shown in dashed lines). In monolayers with heavy transition element constituents, this in-plane symmetry breaking gives rise to a large out-of-plane (Ising) spin texture equivalent to applying magnetic fields of order 100 T in scale[5-7]. An additional mirror symmetry breaking exists across the MX$_2$ plane for materials
deposited on substrates (see Fig. 1(b)) giving rise to an in-plane (Rashba) spin-orbit texture characterized by the local symmetry breaking electric field\(^{11}\). The overall spin texture (Fig. 1(c)) is determined by a mixture of the intrinsic Ising and extrinsic Rashba contributions and, in the case of multilayer \(H-MX_2\), the layer coupling\(^{11}\).

The impact of the band spin texture on superconductivity is of particular interest. For monolayers (the 2D limit) dominated by the out-of-plane texture, superconducting pairing involves electrons with Ising-like spin anisotropy that are robust to application of in-plane magnetic fields significantly beyond the Pauli limit\(^{5-7}\). More recently, systems showing an interplay of Ising and Rashba textures, as well as finite interlayer coupling in multi-layer materials (towards the three dimensional limit), have been utilized to tune the degree of Pauli limit breaking\(^{15-17}\). It has been theorized that with a significant Rashba texture, an alternative state with finite momentum Cooper pairs robust to in-plane magnetic fields may arise\(^{18,19}\). This Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) phase\(^{9,10}\) leverages combined spin, momentum, and in-plane magnetic field to produce a pairing between momentum-shifted Fermi surfaces (see Fig. 1(d)). This novel pairing state requires superconductivity in the clean limit wherein the normal state mean free path exceeds the Pippard coherence length \(\xi_0 \approx 0.18 \hbar v_F/k_B T_c\), where \(v_F\) is the Fermi velocity and \(T_c\) is the superconducting transition temperature. However, as depicted in Fig. 1(e), \(H-MX_2\) materials have thus far not achieved the required combination of two-dimensionality and clean limit superconductivity.

Here we describe an alternative method to realizing high quality 2D \(H-MX_2\) layers for \(M = Nb\) and \(X = S\) in the form of natural bulk superlattice material \(Ba_3Nb_5S_{13}\) composed of \(H-NbS_2\) layers and \(Ba_6NbS_8\) block layers. Figure 1(f) shows a cross section of the structure imaged by high angle annular dark field scanning transmission electron microscopy (HAADF-STEM) along with the model structure. As determined by electron diffraction, the unit cell (space group \(P\overline{3}1c\) with \(a = 10.8\ \text{Å}, c = 22.2\ \text{Å}\)) is composed of two inversion-related \(H-NbS_2\) layers across each of which mirror symmetry is broken by the neighboring block layers while the unit cell retains both inversion and mirror symmetries. The \(H-NbS_2\) interlayer distance \(d = 8.4\ \text{Å}\) is approximately three times that of \(2H-NbS_2\), leading to a significant reduction of the interlayer transfer integral \(t_\perp\). This amplifies the two-dimensionality of the electronic structure and enables local symmetry breaking induced spin-orbit textures on the \(H-NbS_2\) layers\(^{11,12}\). Compared to traditional misfit compounds which combine incommensurate layers in a superlattice, \(Ba_3Nb_5S_{13}\) exhibits a \(3 \times 3\) in-plane, commensurate superstructure due to
the lattice mismatch between the two layer types (see supplementary materials), which leads to additional modification of the electronic structure.

Figure 1(g) shows the dependence of electrical resistivity $\rho_{xx}(T)$ on temperature $T$ for Ba$_3$Nb$_5$S$_{13}$. The system is a metal, eventually becoming a superconductor below $T = 1$ K. This can be compared to bulk 2$H$-NbS$_2$ which is also metallic and becomes a superconductor at $T_c = 5.7$ K$^{21}$. Unlike several other related $MX_2$ systems, neither Ba$_3$Nb$_5$S$_{13}$ nor 2$H$-NbS$_2$ show signs of a density wave transition$^{20}$. The inset of Fig. 1(g) shows a detailed view of the superconducting transition, which onsets near $T = 1.6$ K and reaches zero resistance at $T = 0.85$ K. At the latter temperature, the magnetic susceptibility $4\pi \chi$ with field along the c-axis shows a Meissner signal reaching a volume fraction of 75% (Fig. 1(g) (inset, green)). The reduction in resistivity is well captured by the Halperin-Nelson model, $\rho_{xx}^F(T) = \rho_{xx}^N e^{-b/\sqrt{t}}$, where $\rho_{xx}^F$ and $\rho_{xx}^N$ are the fluctuation and normal state resistivity respectively, $t = T/T_{HN} - 1$, and $b$ is a fitting parameter on the order of one (dashed curve in Fig. 1(g))$^{22}$. The agreement with the Halperin-Nelson model evidences phase fluctuations of the superconducting order parameter above a two-dimensional BKT transition at $T_{HN} = 0.85$ K. Such behavior is rare in bulk single crystals, but has been reported in La$_{1.875}$Ba$_{0.125}$CuO$_4$ and attributed to the decoupling of superconducting CuO$_2$ planes by stripe order$^{23}$.

Magnetotransport measurements show further evidence for a 2D electronic structure. Figure 2(a) shows the magnetoresistance, $MR = (\rho_{xx}(H)/\rho_{xx}(0)) - 1$, measured to 31 T where a series of quantum oscillations are observed which respond to the component of the magnetic field perpendicular to the $ab$-plane (the tilt angle $\theta$ is measured between the c-axis and applied field). The Fast Fourier Transform (FFT) computed after subtracting a monotonically increasing background (see supplementary materials) plotted versus inverse field shows this more clearly (Fig. 2(b, c)). Here the frequency multiplied by cos($\theta$) has little variance, demonstrating the 2D nature of the Fermi surface. This is qualitatively different than in 2$H$-NbS$_2$, for which electronic structure calculations indicate strongly warped and elliptical Fermi surfaces$^{24}$. Instead, owing to the reduced coupling between the layers, the observed bands (labeled here as $\alpha$, $\beta_{(1,2)}$, and $\gamma_{(1,2)}$) can be understood by zone-folding the 2D electronic structure of a monolayer $H$-NbS$_2$, which consists of bands at the $\Gamma$, $K$, and $K'$ points in the hexagonal Brillouin Zone (Fig. 2(d)), with the $3 \times 3$ superstructure of the block layers (see Fig. 2(e)) (see supplementary materials). In particular, the approximate order of magnitude reduction in the pocket size from monolayer $H$-NbS$_2$ caused by this zone-folding
quantitatively captures the size of the observed pockets (Fig. 2(f)). An important aspect of this structure is that the large ratio of the spin-orbit coupling to $t_{\perp}$ enables local symmetry breaking to affect the bulk electronic structure. The zone-folding promotes the Rashba-textured pockets associated with the $\Gamma$ point in monolayer $H$-NbS$_2$ to be of the largest size (rather than the Ising-split pockets at $K$ and $K'$, (see supplementary material)) and has important implications for superconducting pairing.

More generally, it is noteworthy that quantum oscillations have not been reported in $2H$-NbS$_2$; there, the typical transport mobilities reported for bulk single crystals are of order 1 cm$^2$/Vs. In Ba$_3$Nb$_5$S$_{13}$ we see the onset of quantum oscillations in magnetic fields between 2 – 3 T, indicating quantum mobilities of order $10^3$ cm$^2$/Vs. Analysis of the quantum oscillations indicates that the associated transport mean free path significantly exceeds the Pippard coherence length $\xi_0 \approx 0.18 \hbar v_F/k_B T_{BKT} = 254$ nm (see supplementary materials), placing Ba$_3$Nb$_5$S$_{13}$ in the clean limit of superconductivity.

Turning to properties of the superconducting state, Fig. 3(a) shows the current voltage $I(V)$ characteristics of Ba$_3$Nb$_5$S$_{13}$ across the superconducting transition. As expected for a BKT transition$^{22}$, a linear response at $T = 0.95$ K and above crosses over to a non-linear dependence $V \propto I^\alpha$ with $\alpha \sim 3$ at $T_{BKT} = 0.82$ K, close to $T_{HN} = 0.85$ K. Analysis of the slope of $\alpha(T)$ at $T_{BKT}$, suggests the superconducting interlayer coupling is vanishingly small compared to the intralayer coupling$^{22}$ (supplementary materials). In addition to the Halperin-Nelson scaling in the fluctuation regime, this provides further evidence for 2D superconductivity in Ba$_3$Nb$_5$S$_{13}$.

Figure 3(b) shows the evolution of $\rho_{xx}(H)$ as a function of magnetic field for different $\theta$. While for $\theta = 0$ superconductivity is suppressed with relatively low fields and gives rise to quantum oscillations, for larger $\theta$ the upper critical field $\mu_0 H_{c2}$ rapidly increases (herein we define $\mu_0 H_{c2}$ to be when $\rho_{xx}$ reaches half of the normal state value). Fig. 3(c) summarizes this behavior with $\mu_0 H_{c2}(\theta)$ showing a sharp cusp for in-plane fields. Recent studies of 2D superconductors have demonstrated that a distinguishing feature of such systems from anisotropic 3D superconductors is the profile of $\mu_0 H_{c2}(\theta)$ following the 2D Tinkham form $\left( \frac{H_{c2}(\theta) \sin \theta}{H_{c2}^a} \right)^2 + \left( \frac{H_{c2}(\theta) \cos \theta}{H_{c2}^c} \right)^2 = 1$ \cite{Tinkham}, where $H_{c2}^a$ and $H_{c2}^c$ are the upper-critical fields for field applied in-plane and out-of-plane respectively. The response in Ba$_3$Nb$_5$S$_{13}$ can be fit by such a form, but notably there is an enhancement of the scale of $\mu_0 H_{c2}(\theta)$ for angles below 1.7$^\circ$. As shown in the inset of Fig. 3(c), this anomalous enhancement coincides with $\mu_0 H_{c2}(\theta)$
crossing the Pauli paramagnetic limit $\mu_0 H_p \approx 1.84 T_{BKT} = 1.51 \text{T.}$

To further examine the anomaly in $\mu_0 H_{c2}$, we measured $\rho_{xx}(H, T)$ with $\theta$ systematically tuned away from 90°. Plotted as the excess conductivity $\delta \sigma \equiv 1 - \rho_{xx}/\rho_{xx}^N$, the significant enhancement at low $T$ and high $H$ can be seen to quickly disappear as $\theta$ is moved away from 90°, while by $\theta = 86°$ there is little variation with further field tilt (Fig. 4(a)). A distinct feature at all $\theta$ is the finite $\delta \sigma$ for low $H$ extending to $T$ beyond $T_{BKT}$ associated with fluctuating superconductivity. To remove this fluctuation background, we plot the difference $\delta \sigma(\theta = 90°) - \delta \sigma(\theta = 84°)$ in Fig. 4(b). The expected 2D paramagnetic limit is shown as a green line; the transition line follows this response below $T_{BKT}$ until approximately $T/T_{BKT} \approx 0.6$, below which a significant enhancement is observed. As shown in Fig. 4(c), this behavior is confined to low temperature and to a small angular region $\delta \theta \approx 1.7°$ about the ab-plane.

Various theoretical scenarios have been discussed for Pauli breaking in 2D superconductors including spin-orbit scattering, Ising superconductivity, and FFLO states. Given the clean-limit nature of superconductivity here, spin-orbit scattering enhancements cannot account for the present observations (see supplementary material). The dominant local Rashba spin-orbit coupling in the present system reduces the importance of the local Ising coupling; moreover, the acute angular dependence of this effect recalls that of organic FFLO materials such as $\kappa-(ET)_2\text{Cu(NCS)}_2$ and $\beta''-(EH)_2\text{SF}_5\text{CH}_2\text{CF}_2\text{SO}_3$. Viewed more broadly, the clean limit, highly anisotropic superconductivity, Pauli breaking, and Fermi surface nesting without density wave order in Ba$_3$Nb$_5$S$_{13}$ satisfy all the requirements for an FFLO phase. The degree of anisotropy is large enough to exhibit 2D superconductivity and BKT behavior, not previously possible in a candidate FFLO system. Amongst the various FFLO phases, we find that a multi-gap FFLO scenario with mixed $s$-$p$ pairing best fits the data (Fig. 4(b)) where the ratio $T_{pc}^p/T_{sc}$ between the bare $p$-wave and $s$-wave transition temperatures, $T_{pc}^p$ and $T_{sc}$ respectively, is the only free parameter. This is consistent with the multi-gap superconductivity in 2$H$-NbS$_2$ and strong Rashba coupling in Ba$_3$Nb$_5$S$_{13}$ (see supplementary material).

The possibility of an FFLO phase in Ba$_3$Nb$_5$S$_{13}$ offers a significant new pathway to studying pair density wave order. Compared to misfit layered compounds and exfoliated monolayers, the high electronic quality allows for clean limit superconductivity. Compared to CeCoIn$_5$ in which an incommensurate antiferromagnetic state may be intertwined with an
FFLO phase\textsuperscript{30}, $\text{Ba}_3\text{Nb}_5\text{S}_{13}$ is non-magnetic and may offer a simpler phase space. While strong evidence for FFLO phases have been reported in organic crystals\textsuperscript{30}, the robust, inorganic nature of $\text{Ba}_3\text{Nb}_5\text{S}_{13}$ enables a broader range of experimental probes including scattering\textsuperscript{35}, tunneling\textsuperscript{36}, and fabrication of Josephson junctions\textsuperscript{37} that may allow for direct probing of modulated superconducting order.

Finally, we hypothesize that the significant enhancement of electronic mobility observed for the $H$-$\text{NbS}_2$ layers in $\text{Ba}_3\text{Nb}_5\text{S}_{13}$ may be attributed to screening by the highly polarizable block layer akin to that observed in engineered semiconductor heterostructures\textsuperscript{38}. Our DFT calculations suggest that the lowest energy cleavage occurs between the $H$-$\text{MX}_2$ and block layers. Thus, mechanically exfoliated $\text{Ba}_3\text{Nb}_5\text{S}_{13}$ may yield naturally encapsulated $H$-$\text{NbS}_2$ monolayers akin to vdW structures made by stacking $\text{MX}_2$ layers and h-BN\textsuperscript{38} (see supplementary material). Additionally, there is scope for functionalizing the spacer layer by, for example, introducing magnetic constituents. Extending the materials family of natural commensurate superlattices to other $\text{MX}_2$ materials may pave the way for stabilizing high quality materials as platforms for unconventional superconducting\textsuperscript{39}, topological\textsuperscript{40} and excitonic vdW devices\textsuperscript{41}.

**METHODS**

**Single Crystal Synthesis** Single crystals of $\text{Ba}_3\text{Nb}_5\text{S}_{13}$ were grown by the flux method. The crystal structure was analyzed by electron diffraction patterns and high-angle annular dark-field (HAADF-STEM) Scanning Transmission Electron Microscopy images.

**Transport Measurements** Electrical transport measurements were performed using standard AC lock-in techniques. The longitudinal voltages were field symmetrized to correct for contact misalignment. Measurements in magnetic fields up to 31 T were conducted at the National High Magnetic Field Laboratory.

**Magnetization Measurements** Magnetization down to 0.39 K was measured in a magnetic property measurement system (MPMS3) equipped with a $^3\text{He}$ refrigerator, Quantum Design.

**Scanning Transmission Electron Microscopy (STEM)** STEM experiments were conducted at a CEOS Cs probe corrected cold emission gun JEOL JEM-ARM200F STEM operated at 200 kV acceleration voltage. HAADF-STEM images were acquired with 75
mrad convergence semi-angle and 2D Wiener filter applied to reduce the noise. Samples were prepared by a FEI Helios Focused Ion Beam, operated at 30 kV acceleration voltage for the Gallium beam during lift-out and 2 kV during polishing. Additional polishing was performed at 0.5 kV with a Fischione NanoMill for 10 minutes on each side at a milling angle of ±10°.

**Density Functional Theory Calculations** We performed electronic structure calculations implemented in the Vienna ab initio simulation package using the projector augmented wave pseudo-potential method and exchange-correlation functional within the generalized gradient approximation parametrized by PerdewBurkeErnzerhof.

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**Author Contributions** A.D. synthesized and characterized the single crystals. A.D. and H.I. performed the electrical transport experiments. A.D. and M.Kr. performed the magnetization experiments. C.O.-K. and D.B. performed the electronic microscopy experiments. A.D. and S.F. performed theoretical calculations. All authors contributed to discussions and writing the manuscript. J.G.C. coordinated the project.

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Figure 1. Symmetry Breaking and Superconductivity in $H$-$MX_2$ and $\text{Ba}_3\text{Nb}_5\text{S}_{13}$

a In-plane inversion symmetry breaking and b out-of-plane mirror symmetry breaking in monolayer $H$-$MX_2$. c Depiction of momentum space spin-orbit texture for monolayer $H$-$MX_2$ with varying degrees of Rashba and Ising coupling. d Depiction of finite momentum Cooper pair and momentum shift $q_y$ arising from Rashba coupling and in-plane magnetic field $\mu_0H$. e Schematic phase diagram of superconducting $H$-$MX_2$ with in-plane magnetic field as a function of disorder and dimensionality. f HAADF-STEM image of $\text{Ba}_3\text{Nb}_5\text{S}_{13}$ taken along [1100] (1 nm scale bar). A simulation of the crystal structure is overlayed with one unit cell shaded in green. g Resistivity as a function of temperature $\rho_{xx}(T)$ and superconducting transition in $\text{Ba}_3\text{Nb}_5\text{S}_{13}$. The upper inset shows a magnified view of the transition in $\rho_{xx}(T)$ and magnetic susceptibility $4\pi\chi$. The former is well-fit to the Halperin-Nelson model shown in black (see text). The lower inset shows the $H$-$\text{NbS}_2$ layer and mirror symmetry breaking $\text{Ba}_6\text{NbS}_8$ block layers.

Figure 2. Quantum Oscillations and Electronic Structure of $\text{Ba}_3\text{Nb}_5\text{S}_{13}$

a Magnetoresistance as a function of perpendicular field $MR \equiv (\rho_{xx}(\mu_0H) / \rho_{xx}(0)) - 1$ at temperature $T = 0.39$ K for different field rotation angle $\theta$ (geometry defined as shown in the inset). Curves are vertically offset by 150%. b Low frequency and c full range of Fast Fourier Transform (FFT) in inverse field of quantum oscillation amplitude as a function of perpendicular frequency $F \cos(\theta)$. The FFT amplitude for the higher frequency pockets are multiplied by 25. d DFT calculation of monolayer $H$-$\text{NbS}_2$ Fermi surfaces including spin-orbit coupling. e Depiction of zone folding scheme due to $3 \times 3$ superstructure imposed by $\text{Ba}_6\text{NbS}_8$ block layer. f Zone folded monolayer $H$-$\text{NbS}_2$ electronic structure with comparison to observed FFT frequencies.

Figure 3. 2D Superconductivity and Pauli Limit Breaking in $\text{Ba}_3\text{Nb}_5\text{S}_{13}$

a Current voltage characteristics $I(V)$ from temperature $T = 0.95$ K to $T = 0.28$ K. The inset shows the evolution of the power law $V \propto I^\alpha$ with the horizontal line marking $\alpha = 3$. b Longitudinal resistivity $\rho_{xx}$ as a function of field $\mu_0H$ for different $\theta$. Curves are vertically offset by 20 $\mu\Omega$ cm for clarity (horizontal lines). Vertical ticks separate regions measured with low current (7 $\mu$A) and higher current (70 $\mu$A) to avoid Joule heating suppression of superconductivity. For $\theta = 80^\circ$ and 90$^\circ$, only low current is used. c Angular dependence of
upper critical field $\mu_0 H_{c2}$ measured at $T = 0.28$ K with fits to the 2D-Tinkham model, purple and black lines, computed using data in the range $|\theta - 90^\circ| < 1.7^\circ$ and $|\theta - 90^\circ| > 1.7^\circ$, respectively. The inset shows a detailed view near $\theta = 90^\circ$ where an enhancement of $\mu_0 H_{c2}(\theta)$ is observed across the Pauli limit $\mu_0 H_p$.

**Figure 4. Angular Dependence of Excess Conductivity and FFLO Enhancement**

**a** Excess conductivity relative to the normal state $\delta\sigma(\mu_0 H, T)$ for field angles $\theta$ near the $ab$-plane ($\theta = 90^\circ$). **b** Difference between $\delta\sigma(\mu_0 H, T)$ for $\theta = 90^\circ$ and $84^\circ$. The temperature axis is normalized to $T_{BKT}$. The 2D paramagnetic limiting boundary of $\mu_0 H_{c2}$ is shown in green and numerical calculations for the FFLO phase boundary with mixed $s$-$p$ pairing are shown in purple ($T_p^c/T_s^c$ is the ratio of the associated triplet and singlet transition temperature, see text). **c** The angular dependence of $\mu_0 H_{c2}$ at $T/T_{BKT} = 0.3$ (orange) and $\mu_0 H_{c2}$ at $T/T_{BKT} = 0.8$ (green, magnified by a factor of 3). The enhancement above the Pauli limit $\mu_0 H_p$ at low $T$ and small deflection away from $90^\circ$ are suggestive of a crossover from conventional (inset left) to finite-momentum (inset center) Cooper pairing within the angular window $\delta\theta$. The measurement geometry is shown in the inset, right.
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