Transition from three- to two-dimensional Ising superconductivity in few-layer NbSe$_2$ by proximity effect from van der Waals heterostacking

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Abstract

We report the experimental observation of Ising superconductivity in 3-dimensional NbSe$_2$ stacked with single-layer MoS$_2$. The angular dependence of the upper critical magnetic field and the temperature dependence of the upper parallel critical field confirm the appearance of two-dimensional Ising superconductivity in the 3-dimensional NbSe$_2$ with single-layer MoS$_2$ overlay. We show that the superconducting phase has strong Ising spin-orbit correlations which make the holes spin non-degenerate. Our observation of Ising superconductivity in heterostructures of few-layer NbSe$_2$ of thickness $\sim$ 15 nm with single-layer MoS$_2$ raises the interesting prospect of observing topological chiral superconductors with nontrivial Chern numbers in a momentum-space spin-split fermionic system.

I. INTRODUCTION

An important focus in condensed matter physics is on creating 2-dimensional (2D) topological chiral superconductors (SC) with non-trivial Chern numbers [1–4]. Naturally occurring topological SC being rare [5, 6], recent efforts have concentrated on inducing superconductivity in systems with strong spin-orbit coupling (SOC) [7]. The fundamental idea is that in a time-reversal-invariant non-centrosymmetric system with strong spin-valley locking, the spin degeneracy (in momentum space) of the fermions gets lifted. In such a system, when the chemical potential of the system lies in between the spin-split valence band maxima, repulsive interactions can lead to topological pairing mechanisms [8, 9].

For a single atomic layer of a system with hexagonal lattice, the measured in-plane critical magnetic field is significantly larger than the Pauli limit. The origin of this enhancement lies in the intrinsic Ising SOI (arising as a consequence of in-plane inversion symmetry breaking) in the layer. This SOC manifests itself as an effective out-of-plane Zeeman-like field, $B_{SO}$ with opposite orientations at $K$ and $K'$ valley. Thus, for a cooper pair in such a system, spin-valley locking leads to a large $B_{||}^{c}$. Ising superconductivity is thus the most promising candidate for the observation of topological effects. This phenomena have earlier been observed in ionic-gated 2D-MoS$_2$ [10, 11] and in single-layer NbSe$_2$ [12].

Single-layer of 2H-MoS$_2$ (hereafter referred to as SL-MoS$_2$), owing to its strong SOC [13], holds the promise of forming the base for such exotic SC with tailor-made properties. There have been previous reports of Ising superconductivity induced in ionic-gated 2D-
MoS$_2$ [10, 11]. But both these methods suffer from certain inherent drawbacks – structural changes in the material, rapid degradation of the sample under ionic liquid, the difficulty of fabricating a complex device structure and a superconducting transition temperature much lower than theoretical predictions. Single-layer NbSe$_2$ degrades very rapidly on exposure to the ambient making it unfeasible as the basis for complex device architectures. A practical path to overcome this hurdle is by engineering novel heterostructures of NbSe$_2$ with other materials having properties whose combination can give rise to an atmosphere stable Ising superconducting phase.

Additionally, in all cases, the SL-MoS$_2$ had been electron-doped to achieve the SC. Note that the strength of SOC (and the resulting spin-valley coupling and spin-splitting of the bands) in single-layer TMD is significant in the valence band. This implies that in order to have spinless fermions, such a system must be hole-doped [8, 9]. A controllable mechanism of inducing 2-dimensional superconductivity in transition metal dichalcogenide that can achieve relatively high transition temperatures while maintaining the long-term stability of the device is elusive.

In this letter, we report the observation of Ising superconductivity in few-layer NbSe$_2$ (∼15 nm) with an overlayer of single-layer MoS$_2$. Having both materials from the TMD family avoids many of the problems arising from lattice mismatch at the interface [14–17]. Through systematic magnetotransport measurements, we establish that system develops an Ising superconductivity that is distinct from that of pristine NbSe$_2$. Though Ising superconductivity has been observed in ionic-gated MoS$_2$ [10, 11] and in single-layer NbSe$_2$, our approach has distinct advantages over previous attempts in terms of yield, stability of the system, and is more amenable for the realization of complex device structures.

II. DEVICE FABRICATION

The devices were fabricated by transfer of SL-MoS$_2$ using the dry transfer technique on gold probes pre-patterned on a thick hBN substrate. This was followed by the transfer of a NbSe$_2$ flake (thickness ∼15 nm) such it lay partially on the SL-MoS$_2$ and partially on the Au-probes. We refer to the pristine NbSe$_2$ part of the device as ‘region-I’ and the SL-MoS$_2$/NbSe$_2$ heterostructure as ‘region-II’. The entire device was capped with a thin flake of hBN (thickness ∼30 nm) to protect the device from environmental degradation (for
Figure 1. (a) A schematic of the device structure. The SL-MoS$_2$ is transferred on top of gold contact pads etched in hBN. A multi-layer NbSe$_2$ is transferred on top such that it lies partially on the MoS$_2$ (shown in light blue) and partially on the gold probes (shown as an orange rectangle). (b) False color DIC image of a final device. (c) Temperature dependence of the four-probe resistance of the pristine NbSe$_2$ (orange line) and of the SL-MoS$_2$ (olive line) sections of the device.

details of device fabrication, see Appendix S2). This device architecture allowed us to study electrical transport simultaneously in both the pure NbSe$_2$ and in the SL-MoS$_2$/ lying on top of NbSe$_2$ (Fig. 1(a)). Fig. 1(b) is the false-color optical image of a final device showing the SL-MoS$_2$ (blue area), the NbSe$_2$ (∼15 nm) (pink area, region-I) and the overlap area (region-II).

III. RESULTS AND DISCUSSIONS

Electrical transport properties were measured in four-probe configurations using standard low-frequency lock-in detection technique with an excitation current of 1µA in a dilution refrigerator. At room temperature and under zero gate bias, the SL-MoS$_2$ capped by NbSe$_2$ (‘region-II’) had significantly lower resistance (∼700Ω) as compared to that measured for
Figure 2. Angular dependence of the $B_{c2}$ for (a) region-I and, (b) region-II. The red and black lines in both the panels are plots of 2D Tinkham model and 3D GL anisotropic mass model, respectively. The size of data points in both the plots are larger than the error in the data. The insets show the direction of $B_{c2}(\theta)$.

MoS$_2$ on SiO$_2$ or on hBN substrates (> 1MΩ) (data not shown). Fig. 1(c) shows the temperature dependence of the resistance $R$ of the two regions for $T < 10$ K – the resistances of both region-I and region-II decrease with decreasing temperature $T$ before becoming smaller than our measurement resolution.

Fig. 2(a) and Fig. 2(b) show the angle (measured with respect to the out-of-plane direction of the device) dependence of $B_{c2}$ normalized by the in-plane $B_{c2}$ at $T = 0.9T_c$ for region II and region I, respectively. The angular dependence of $B_{c2}(\theta)$ for the region II is well described by the 2D Tinkham model $\left(\frac{B_{c2}(\theta)}{\cos\theta/B_{c2}^{\perp}} + \frac{B_{c2}(\theta)\sin\theta/B_{c2}^{||}}{1}\right)^2 = 1$ [18]. On the other hand, $B_{c2}(\theta)$ in the NbSe$_2$ region follows the 3D Ginzburg-Landau model $\left(\frac{B_{c2}(\theta)\cos\theta/B_{c2}^{\perp}}{1} + \frac{B_{c2}(\theta)\sin\theta/B_{c2}^{||}}{1}\right)^2 = 1$ [19] establishing that in contrast to the 3D superconductivity of the pure NbSe$_2$ in region-I, the superconductivity in region-II is of 2D nature. This is the central point of this letter.
Having established that the SC in region-II is 2-dimensional, we estimate the Berezinskii-Kosterlitz-Thouless transition temperature, $T_{BKT}$ for region-II from both non-linear current-voltage characteristics and from the $R - T$ characteristics [20, 21]) to be 6.14 K (see Appendix S4 for details).

Fig. 3(a), and Fig.3(b) show the normalized magnetoresistance as a function of a magnetic field applied in-plane for the region-II and region-I, respectively. The in-plane critical field ($B_{c2}^\parallel$) are plotted versus the reduced temperature, $T/T_c$ in Fig. 3(c). Note that $B_{c2}^\parallel$ for the region-II is significantly larger than that of the pristine NbSe$_2$ in region-I. The data from region-II fits well with the 2D GL equation $B_{c2} = B_{c2}^0 \sqrt{1 - T/T_c}$ [19] giving a $B_{c2}^\parallel(0) \sim 36.5$ T. This is well beyond the Pauli Paramagnetic limit, $B_P (= \sim 1.86T_c) \sim 10.5$ T for the system. For the region-I the data, as expected for NbSe$_2$ flake thicker than the superconducting coherence length, fits with the 3D formula, $B_{c2} = B_{c2}^0 (1 - T/T_c)$ [19]. The fact that the SC phase in region-II has a dimension $d=2$ (in contrast to $d=3$ for NbSe$_2$ in region-I) is the central result of this letter.

Fig. 3(d) shows the plots of both $B_{c2}^\parallel$ and $B_{c2}^\perp$ (normalized by $B_P$) versus the reduced temperature for region II. Here $B_P (= \sim 1.86T_c) \sim 10.5$ T is the Pauli paramagnetic limit for the SL-MoS$_2$/NbSe$_2$ heterostructure. As can be seen, $B_{c2}^\parallel(0)$ is $\sim 3.55$ times above the Pauli limit. This is a direct indication of Ising superconductivity. The spin-splitting energy, $\Delta_{SO} \approx 2\mu_B B_{c2}^\parallel(0)^2/B_P$ [12, 22, 23] estimated from the parallel-field magnetoresistance data was $\sim 15$meV which matches quite well with the theoretical estimation of Zeeman type SOI, $\Delta_{SO} \sim 13$ meV in 2-dimensional TMD [11].

To further distinguish between transport in the two regimes, we probed the vortex dynamics of the system through measurements of DC non-linear current-voltage characteristics in the presence of an out-of-plane magnetic field, $B^\perp$. Fig. 4(a) and (c) show the $E$-$J$ characteristics for the two regions of the device measured at 2.5 K for different values of $B^\perp$ (the data for another device are presented Appendix ??). For a disordered superconductor in the flux-flow regime, the charge current density is given by $E = \rho_{ff} (J - J_p)$ [24], where $E$ is the electric field between the voltage probes, $\rho_{ff}$ is the flux flow resistivity, and $J_p$ the depinning current density. A comparison of Fig. 4(b) and (d) shows that $J_p$ for the region-II is much higher than that in region-I. The pinning force per unit length of the vortex, extracted using the equation $F_p^d = J_p(h/2e)$ [25], is plotted in Fig. 4(e) at a few representative values of $T/T_c$. The value of $F_p^d$ for region-II is an order of magnitude higher than that of region-I.
Figure 3. Plot of magnetoresistance of the (a) region-I and (b) region-II (normalized in both cases by the normal state resistance) for in-plane magnetic field. (c) Plot of $B_{c2}^\parallel$ versus $T/T_c$ for region-I (green open circles) and region-II (orange open circles) with the 2D GL fit (dashed red line) for region-II and 3D fit (dashed black line) for region-I. (d) Plot of $B_{c2}/B_P$ versus $T/T_c$ for both out of plane (red circle) and in-plane (blue circle) magnetic field direction for region-II – the dashed lines show the fits to the data.

This is expected given the significantly higher defect levels in SL-MoS$_2$ [26] as compared to that in NbSe$_2$. This establishes that in region-II of the device, the presence of MoS$_2$ has a major effect on the transport of supercurrent through the underlying NbSe$_2$.

To recapitulate our principal results, we observe superconductivity at the heterojunction of single-layer MoS$_2$ and few-layer NbSe$_2$(\sim 15 nm) which differs from that of NbSe$_2$ in several different aspects – the primary being its 2-dimensional Ising nature. We turn now to the discussion of the possible origin of this Ising SC.

A plausible mechanism is that the SL-MoS$_2$ can have cracks facilitating current to flow through it into the NbSe$_2$. This artefact was ruled out from AFM topography mappings on multiple devices, which did not show any structural damage on the SL-MoS$_2$ upon transfer
Figure 4. $E - J$ characteristics at different $B^\perp$ measured at $T = 2.5$ K for the (a) region-II, and (b) region-I. Depinning current density $J_p$ as a function of $B^\perp$ at 2.5 K for the (c) region-II, and (d) region-I. (e) Plots of the pinning force per unit length of the vortex, $F^l_p$ versus the reduced temperature $T/T_c$ region-I (orange filled circles) and region-II (olive filled circles). The lines are guided to the eye.

on the pre-patterned hBN probes (See Appendix S3). An alternate proposition is that the transport in region-II is solely through the underlying NbSe$_2$ whose superconducting properties change from that of 3D to 2D-like due to the presence of the single-layer MoS$_2$. We note that further experimental and theoretical studies are necessary to ascertain the mechanism of superconductivity in this hybrid system.

In an attempt to understand the effect of the presence of the single-layer MoS$_2$ on the superconductivity of the underlying NbSe$_2$, we carried out first-principles density functional theory calculations (see Appendix S7 for technical details). We constructed a heterojunction comprising of 6 layers of NbSe$_2$ and 1 layer of MoS$_2$, as shown in Fig. 5(a). The resulting density of states is presented in Fig. 5(b). The total density of states (olive curve) shows
that the system is metallic, with many states around the Fermi level. We further calculate
the density of states projected on the MoS$_2$ layer (orange curve). Notably, the MoS$_2$ layer
has also lost its semiconducting character when in proximity to the NbSe$_2$, as illustrated by
the finite density of states around the Fermi level on the MoS$_2$ layer. To further corroborate
our findings, we present a plot of the charge density of the heterojunction in Fig. 5(c). The
isosurfaces show substantial charge density at the various layers. Most notably, we find
that the charge densities of the SL-MoS$_2$ layer and the NbSe$_2$ layer in contact with it are
overlapping, confirming the hybridization between the two. This hybridization may result
in the change in superconducting properties of NbSe$_2$ from 3D to 2D-like, as we observed. From
the Hall data in Appendix S5 Fig. S5 the overlap of charge density is evident. We can also
notice that the adding the SL-MoS$_2$ in region-II varies the density from that of the pristine
NbSe$_2$. The implication of this is that the coherence length of region-II becomes larger than
the typical $\sim 9$ nm value of pristine NbSe$_2$ which consequently lead to the observation of
2D superconductivity in heterostructure.

An interesting observation is that the charge carriers in region-II are holes – this is
in accordance with previous observations [27]. Our observation that region-II becomes a
hole-doped 2-dimensional Ising SC is exciting because of the prospect that it can host a
topologically non-trivial superconducting phase [8, 9] and SC with finite-momentum-pairing
per the predictions by Fulde and Ferrell [28] and by Larkin and Ovchinnikov [29]. This
makes our system unique; as in all earlier reports, SC in TMD was achieved through electron
doping [30].

IV. CONCLUSION

In summary, we have observed a transition from 3-dimensional to 2-dimensional supercon-
ductivity in heterostructure of few-layer NbSe$_2$(\(\sim 15\) nm) and SL-MoS$_2$. Magnetotransport
measurements establish that the superconductivity in the underlying NbSe$_2$, albeit physically
in the 3-dimensional limit, has a strong Ising pairing. The stability of the superconducting
phase and the system being in the hole-doped region makes our device structure unique and
opens up the scope of observing topological superconductivity in van der Waals systems.
Figure 5. (a) Illustration of the heterojunction with 6 layers of NbSe₂ and 1 layer MoS₂. (b) The total density of states (olive) and density of states projected on the MoS₂ layer (orange) of the system. (c) Charge density isosurfaces for the heterojunction. (d) The average local potential along the stacking direction, z.

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S1. SUNKEN ELECTRICAL PROBES ON HBN SUBSTRATE

Figure S1. Optical image at different stages of fabrication process of a hBN sunken Au probe (a) ~ 25 nm hBN flake transferred on pre-patterned substrate. (b) Electron-beam lithography patterns for contact on the hBN flake before dry etching. (c) The hBN flake after the dry etching process (d) Final substrate after metal deposition.

One disadvantage of dealing with transition metal dichalcogenide (TMD) materials, especially NbSe$_2$, is their low stability in ambient conditions [31]. Keeping this fact in mind we went for an encapsulated device structure (as described earlier). The flakes were trans-
Figure S2. (a) Room temperature Raman spectra of MoS$_2$ (red open circles) along with the Lorentzian fits for the $E_{2g}$ peak (violet line) and $A_{1g}$ peak (pink line). The difference in Raman Shift between the two peaks is 19.50 cm$^{-1}$ indicating the MoS$_2$ to be a single-layer. False color optical image of few other measured devices in two different probe geometry (b) Linear probe (c) Hall measurement device for estimation of carrier no density discussed in Appendix S5.

ferred on pre-fabricated hBN sunken Au probes to avoid degradation due to exposure to moisture as chemicals such as e-beam resist, organic solvents etc. The ‘sunken’ probes also help avoid strain (usually present in such structures [32]) to the thin flakes of MoS$_2$ which may result in a ‘tear’ that can electrically short it with the NbSe$_2$ above it. So for this purpose we first fabricated a pre-patterned substrate where big electrical contact pads of dimensions $\sim$ 200 $\mu$m x 200 $\mu$m were initially patterned using photo-lithography technique on a $\sim$ 2 mm x 2 mm piece of a Si/SiO$_2$ substrate. The SiO$_2$ in the exposed region of the pattern were then etched by $\sim$ 45 nm using a Buffered HF solution with a subsequent step of deposition of Cr-Au of thickness 5-55 nm respectively creating big pads of an effective
height of 15 nm with respect to the substrate surface. hBN flakes were procured using micro-
mechanical exfoliation with standard scotch-tape method [33] on commercial PDMS gel 
stamp(Gel-Pak,PF-3-X4). Flakes of thickness ~ 25 nm were roughly selected using optical 
microscope contrast. Selected flakes were then transferred using a micro-manipulator and 
an optical microscope in the middle section of a pre-patterned probes on Si/SiO₂ substrate 
as shown in Fig. S1(a). After the transfer of hBN onto the substrate the thickness of the 
flake were confirmed using AFM technique and substrates having hBN flakes of thickness 
~ 25 ± 2 nm were only selected for further process. Subsequently patterns of electrical 
contacts on hBN as well as the connecting lines between hBN and the pre-deposited big 
pads were defined using e-beam lithography technique (Fig. S1(b)) with a follow up step 
of selective etching of hBN using a proportionate mixture of CHF₃ and O₂ in a Reactive 
Ion Etching system(RIE-Cl) to etch off the hBN in the exposed part of the flake along the 
contact lines as shown in Fig. S1(c). As the final step Cr-Au of thickness 5-22 nm were then 
deposited giving an almost planar contact surface on hBN (Fig. S1(d)). The advantage of 
having only 15 nm of effective height of big pads is that the connecting lines of total thick-
ness of ~ 27 nm would ‘climb’ on top of it upon deposition ensuring continuous electrical 
connection. As a final step the prepared probes were vacuum annealed at 250°C for 3 hours 
to get rid of any residue on the hBN surface before further process.

S2. DETAILS OF DEVICE FABRICATION

Single-layer flakes of MoS₂ were obtained from high-quality single crystal by mechanical 
exfoliation [33] on PDMS gel stamp. The thickness of the flakes was verified through room-
temperature Raman spectroscopy [34]. Fig. S2(a) is the Raman spectra of one such MoS₂ 
flake – shows the presence of E₂g and A₁g peaks with a peak difference of ~ 19.5 cm⁻¹ in 
Raman shift confirming the flake to be a single-layer [34]. The MoS₂ flake was transferred 
partially on the pre-patterned Au contact probes on hBN (see Appendix S1) using a dry 
transfer technique [35] with the help of a high precision electrical micromanipulator and a 
digital camera fitted with an optical microscope. AFM measurements were carried out to 
rule out any cracks in the MoS₂ layer (see Appendix S3).

NbSe₂ flakes were exfoliated in similar fashion in the inert environment of a glove-box 
having O₂ and H₂O concentrations < 0.5 ppm. The thickness of the flake was estimated to
Figure S3. (a) Optical image of monolayer of MoS$_2$ transferred on hBN sunken gold probes with false color dashed line showing the boundary of the MoS$_2$ (white) and hBN (red). The black dashed circle marks a region with cracks in the MoS$_2$ layer. (b) Zoomed in image of the area marked by circle in Fig. S3(a) showing cracks in MoS$_2$ (c) AFM image of the system in enhanced coloring. (d) Zoomed in AFM image of the section on probe showing MoS$_2$ on probe (darker shade) and bare probes (lighter shade).

To be ∼ 15 nm from optical contrast. The NbSe$_2$ flake was then transferred very precisely using the remotely controlled micromanipulator onto the probes such that half of the NbSe$_2$ flake lay on the exposed Au probes, whereas the other half got transferred on the single-layer MoS$_2$. We refer to the region having only NbSe$_2$ as ‘region-I’ while the region with MoS$_2$ on NbSe$_2$ is referred to as ‘region-II’. Finally, an hBN flake of thickness ∼ 30 nm was transferred to the device to avoid environmental degradation of the heterostructure. As a final step, the device was vacuum annealed at 200° C to improve the coupling between the MoS$_2$ and NbSe$_2$ layers [36].

S3. VERIFYING THE TOPOGRAPHY OF MOS$_2$ TRANSFERRED ON PROBES

To choose regions of defect-free single-layer MoS$_2$, AFM measurements were carried out after transfer on the Au-probes. Fig. S3 shows an example of both the optical (Fig. S3(a)) and AFM (Fig. S3(c)) images. The regions having cracks on MoS$_2$ (marked with blacked circles) as shown in Fig. S3(b) gives a contrast difference both in optical as well as AFM which makes it easier to identify them. These areas were stringently avoided in making the
Figure S4. (a) Zero magnetic field current-voltage characteristics of the region-II with varying temperature ranging from 5.0 K to 14.0 K. The red dashed line represents the range of current within which linear fit was done for each curve. (b) Plot of $\alpha$ as a function of temperature, $T$ to evaluate $T_{BKT}$ for region-II. (c) Plot of $(\text{dlnR/dT})^{-2/3}$ as a function of temperature, $T$ for the MoS$_2$ where the intersect of the black dashed line with the x axis gives $T_{BKT}$.

final device. The zoomed in image of the single-layer MoS$_2$ on the Au-probes in Fig. S3(d) also shows the continuity of flakes. The few bubbles which are present in Fig. S3(d) were eliminated by the vacuum annealing after final assembly of the entire device.
S4. ESTABLISHING BKT PHYSICS

In the main text, we have established, through angular dependence of the critical field, that in region-II the superconductivity has an effective dimensionality of two. In this section we evaluate the Berezinskii-Kosterlitz-Thouless transition temperature, $T_{BKT}$ of region-II from the temperature dependent current-voltage characteristics (Fig. S4(a)). For a 2D superconductor, electric field induced unbinding of the the vortex-antivortex pairs give rise to non-linear current-voltage characteristics of the form $V \sim I^\alpha$. At $T = T_{BKT}$, the non-linearity exponent, $\alpha$ becomes 3 as shown in Fig. S4(b) [20, 37]. This analysis yields $T_{BKT} = 6.13$ K.

One can also evaluate $T_{BKT}$ from the $R$-$T$ characteristics using the relation $R = R_0 \exp[-b_R/(T - T_{BKT})^{1/2}]$, where $b_R$ is quantity indicating the vortex-antivortex interaction strength [20, 21, 38]. The formula is valid over a very small range of above $T_{BKT}$ as within that range superconductivity is destroyed by thermal unbinding of vortex-antivortex pairs. Thus by plotting its reduced form $(dlnR/dT)^{-2/3} = (2/b_R)^{2/3}(T - T_{BKT})$ we can deduce the $T_{BKT}$ from the x-axis intercept as shown in Fig. S4(c) which comes out to be 6.14 K. The estimation of $T_{BKT}$ from both the analysis is very close to the value of 6.3 K reported for ion-gated TMD in earlier results [10].

S5. HALL MEASUREMENTS

Fig. S5(a) and Fig. S5(b) show respectively the plots of $R_{xy}$ versus $B$ measured for the region-I and region-II. The charge carrier densities calculated from these plots are shown Fig. S5(c). One can see that the estimated number densities in the region-II closely follows that of the pristine NbSe$_2$ in region-I.

S6. DEPINNING CURRENT, $J_p$ EVALUATION

To obtain the depinning current, $J_P$ we have to consider the linear section of the $E - J$ curve (Fig. S7) near the onset of resistance as following the definition, for $J > J_P$ the system enters into the flux flow regime for which the resistance i.e. $R_{ff}$ becomes independent of current [24]. For that purpose we select a section of the curve just above the $E$ value 1 V/m and perform a linear fit in that region. We then extract the value of $J_P$ from the intersect
Figure S5. Magnetic field ($B$) dependence of Hall resistance, $R_{xy}$ for (a) region-I over the temperature range 7.5 K–100 K, and, (b) region-II over the temperature range 8 K–100 K. (c) Plot of the charge carrier density, $n$ versus $T$ for both region-I (orange filled circles) and region-II (olive filled circles) showing similar carrier densities, i.e. holes.

of the best fit to the $J$ axis [39], as shown in Fig. S7.

S7. AB-IN-ITIO CALCULATION

Density functional theory computations were performed using the VASP code [40, 41]. The Perdew-Burke-Ernzerhof approximation to the exchange-correlation functional, including the van der Waals correction, was employed [42]. A plane wave cutoff of 300eV was used, and the Brillouin zone was sampled using a $9 \times 9 \times 1 \Gamma$–centered $k$–point mesh. All atoms were relaxed until the forces were less than 0.01eV/Å. A vacuum of 10 Å was used to avoid spurious interaction between periodic images.
Figure S6. $E - J$ characteristics at different perpendicular magnetic fields measured at $T = 5\, \text{K}$ for the (a) region-I, and (b) region-II. (c) Depinning current density $J_p$ as a function of the applied perpendicular magnetic field $B$ at 5 K for the region-I (orange filled circles) and region-I (green filled circles). (d) Plots of the pinning force per unit length of the vortex, $F^l_p$ as a function of reduced temperature $T/T_c$ for the region-I (orange filled circles) and region-II (green filled circles).
Figure S7. $E - J$ characteristics at $T = 5$ K and $B = 0.03$ T for (a) region-I, and (b) region-II. The dashed lines in both the plots are the linear fits - the intersect of this line with the $J$ axis gives an estimate of the depinning current density.
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