

Quantum Coherent Transport of 1D Ballistic States in Second-Order Topological Insulator Bi_4Br_4

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We investigate quantum transport in micrometer-sized single crystals of Bi_4Br_4 , a material predicted to be a second-order topological insulator. 1D topological states with long phase coherence times are revealed via the modulation of quantum interference with magnetic field and gate voltage. In particular, we demonstrate the existence of Aharonov-Bohm interference between 1D ballistic states several micrometers long, that we identify as phase-coherent hinge modes on neighboring step edges at the crystal surface. These Aharonov-Bohm oscillations are made possible by a disordered phase-coherent contact region, the existence of which is confirmed by scanning transmission electron microscopy combined with energy-dispersive x-ray spectroscopy (STEM-EDX) of FIB lamellae. Their coherent nature modulates the transmission of the 1D edge states, leading to weak antilocalization and universal conductance fluctuations with surprisingly large characteristic fields and a strongly anisotropic behavior. These complementary experimental results provide a comprehensive, coherent description of quantum transport in Bi_4Br_4 and establish the material as a second-order topological insulator with topologically protected 1D ballistic states.

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I. INTRODUCTION

An important recent achievement in condensed matter physics is the discovery of a new class of materials, topological insulators (TIs), whose bulk is insulating, while the boundaries are conducting [1–3]. In particular, the 1D edges of 2D TIs realize the quantum spin Hall state [4,5], where current is carried dissipationlessly by two counter-propagating ballistic channels with opposite spins, forming a helical edge state. This opens many possibilities, ranging from dissipationless charge and spin transport at room temperature, to new avenues for quantum computing. Among the recent additions to the TI classification, second-order topological insulators (SOTIs) [6,7] are n -dimensional crystals exhibiting $(n - 2)$ -dimensional topological states. In particular, 3D SOTIs have insulating

bulk and surfaces but topologically protected, one-dimensional helical “hinge” states. 1D helical states have been evidenced in several systems in 2D such as HgTe quantum wells [8], monolayer WTe_2 [9,10], or TaIrTe_4 [11] and in 3D systems such as crystalline bismuth [12,13]. Unfortunately, the expected topological protection has turned out to be less robust than anticipated, notably due to the existence of conduction in the bulk and surfaces, or charge puddles [14]. This complicates the fundamental study of the edge states and motivates the search for different TIs with a reduced contribution of the nontopological bulk states. Among newly discovered TIs, Bi_4Br_4 appears to be a very promising material, with a large bulk gap ($\gtrsim 200$ meV) [15–21] and strong experimental indications of a SOTI character [22–25].

This material, which in its α form has a bilayer structure of chains oriented along the crystal's b direction [Figs. 1(a) and 1(b)], has indeed been predicted to show the quantum spin Hall (QSH) state for the monolayer [15,16], and the multilayer has been classified as a SOTI [17–21,23,26–29]. The SOTI character in the multilayer case results from the coupling of helical edge states upon stacking of 2D monolayers, leading to gapped side surfaces with a gap of the order of 30 meV. On a perfect crystal, only two

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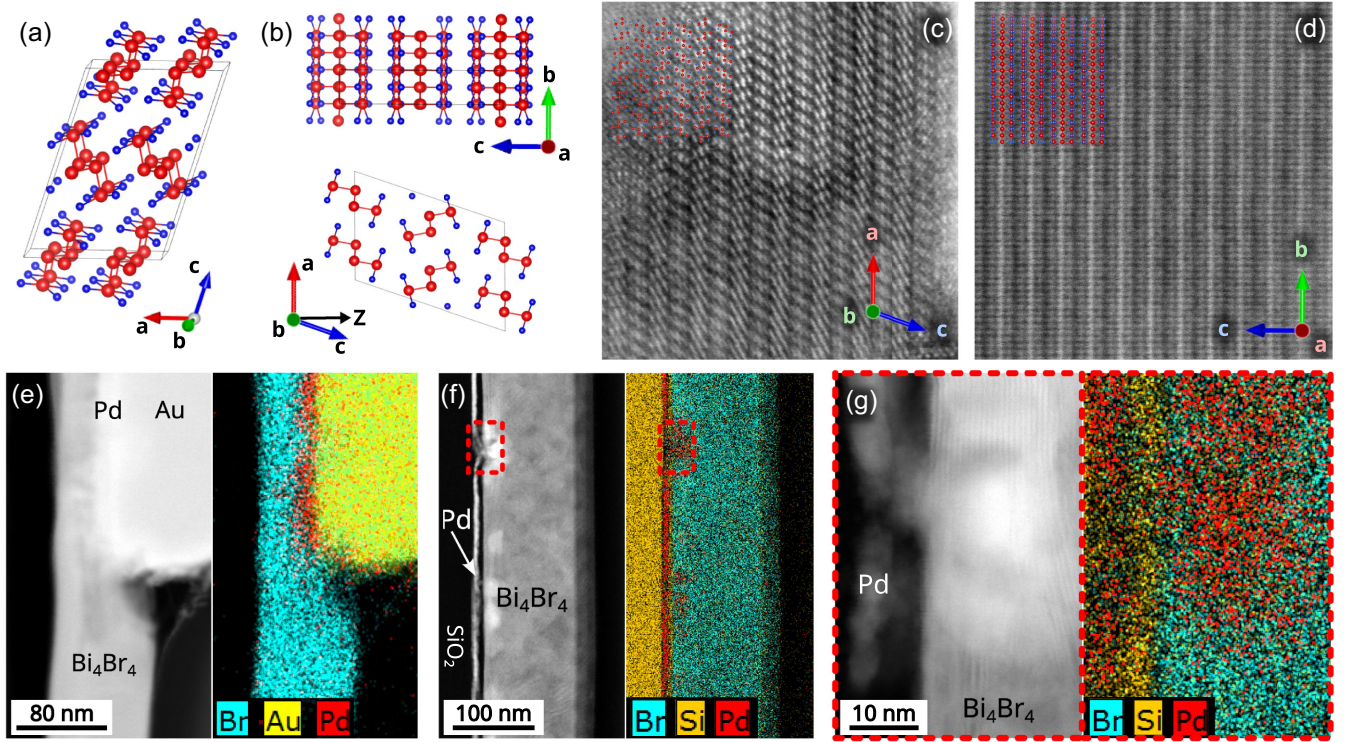


FIG. 1. Structure of Bi_4Br_4 crystals far from and in the contact region. (a) Crystalline structure of Bi_4Br_4 and crystal orientation. Bi atoms are figured in red and Br in blue. (b) Structure of Bi_4Br_4 along the a and b axis. The Z direction, perpendicular to the (a), (b) plane of the flakes, is indicated. (c) High-resolution high-angle-annular dark-field STEM images of FIB lamellae of contacted Bi_4Br_4 flakes obtained along the b direction, away from the contact area. The expected crystalline structure is superimposed on the picture. The agreement between theoretical and real atoms positions is excellent. (d) Similar image along the a direction. (e) Contact region of Au/Pd (100 nm/7 nm) top-contacted electrodes, showing crystal damage both under and away from the electrode. The EDX analysis (right image) shows a diffusion of Bi in the Pd and, to a lesser extent, in the Au layer. The spurious Si signal in the gold electrode is due to the proximity of a fluorescence band of Au with the Si $K\alpha$. (f) Contact region for 7-nm-thick Pd electrodes placed below the Bi_4Br_4 flake. Diffusion of Pd in the crystal is also visible. (g) Enlargement of the area denoted by the red dashed rectangle in (f), which evidences disordered contact regions of size around 100 nm.

helical states located at hinges remain uncoupled, which location depends on the parity of the number of layers.

The band structure of Bi_4Br_4 has been probed by angle-resolved photoemission spectroscopy (ARPES) [22–24] and micro-ARPES [25]. This latter work confirmed the existence of gapped lateral surfaces, with a gap ranging from 28 to 40 meV, and resolved their spin texture, indicating a spin polarization perpendicular to the direction of the chains. In addition, optical pump-probe microscopy at room temperature revealed a relatively long carrier lifetime for boundary states attributed to their linear dispersion relation [26]. Lastly, conducting hinge states have been probed by STM [23,27–29] up to room temperature, on mono- and few-layers-high step edges. These states are gapped by a magnetic field perpendicular to chains with a Landé factor of 53, indicating a spin quantization axis along the b axis, in stark contrast to the spin polarization of bulk [20] and lateral surface states [25].

With regard to electronic transport, experiments carried out on macroscopic samples indicate that the temperature

dependence of resistivity is close to that of a semiconductor below 50 K and often shows a local maximum around 200 K [22,23,30–34]. It has been proposed that the former results from either competition between bulk and hinge states [22,30] or the appearance of a charge density wave [32,33] or a variable-range hopping induced by Br vacancies [34], while the 200 K feature is attributed to a Lifshitz transition [23,30]. At low temperature, macroscopic samples exhibit weak localization or weak antilocalization depending on the field orientation [35,36]. High-pressure experiments found a metal-insulator transition that can even lead to a superconducting state at pressures above 3.8 GPa [33]. By probing micrometer-size crystals obtained by exfoliation, several temperature behaviors were obtained: a nearly temperature-independent resistivity for relatively long flakes [37], a gate tunable resistance attributed to competition of bulk and surface conduction [38], or a thermal activation of bulk carriers [39]. This latter reference demonstrated an Aharonov-Bohm effect associated to quantum interference of coherent hinge states around single lateral surfaces in four- and six-layer flakes.

In the present work, we study quantum transport in micrometer-sized Bi_4Br_4 flakes with thicknesses ranging from 50 to 150 nm. We find evidence of phase-coherent ballistic 1D states over several microns via the Aharonov-Bohm effect, as well as negative four-wire resistances. Interestingly, the samples also consistently display weak antilocalization (WAL) and universal conductance fluctuations (UCFs), which are staples of 2D coherent transport in the diffusive regime. These apparently conflicting observations can be reconciled by considering a small, coherent, but disordered region between the pristine crystal and the contact, which enables both interhinge coherence and anomalous 2D quantum interference. The existence of this region is confirmed by direct imaging and elemental analysis using STEM-EDX. While this contact regime prevents observation of the quantification of conductance generally expected in QSH systems, it provides extremely rich signatures of the underlying topological transport.

II. EXFOLIATION AND CONTACTING OF Bi_4Br_4 FLAKES

The Bi_4Br_4 crystals were synthesized by the chemical vapor transport method [22,40]. The samples were exfoliated in a glove box under argon atmosphere with $\text{O}_2 < 0.2$ ppm and $\text{H}_2\text{O} < 5$ ppm. The exfoliation and transfer of micrometer-size Bi_4Br_4 flakes was done using polydimethylsiloxane (PDMS) stamps. We check with STEM imaging that the crystalline quality of the exfoliated flakes is preserved before any contact is made (Sec. 1 in Supplemental Material [41]).

In order to contact the flakes, we developed two strategies. The first one is based on contacts made on top of the flake. This is done by e-beam lithography, with low-temperature processes to preserve the quality of the flakes (PMMA resist baking below 100°C), and metal deposition in an e-beam evaporator after argon ion beam etching. Different contact materials were tried, but the best results were obtained with palladium (Pd). The samples are, thus, contacted by Pd(7 nm)/Au(100 nm) electrodes; this leads to $\text{k}\Omega$ two-wire resistances at room temperature. However, this method damages the crystal under the contacts and exposes the flakes to organic solvents and ambient air during the e-beam lithography and lift-off processes. To avoid these problems, we also tried contacting the flakes by transferring them onto predefined metallic electrodes, fabricated by e-beam and optical lithography, with e-beam metal deposition. The Bi_4Br_4 flakes are then pressed onto the electrodes in Ar atmosphere. During this process, we found that heating the transfer stage to 80°C produced Ohmic contacts more reliably. The best results were obtained, as in the case of contacts on top, with palladium electrodes and also yields two-wire resistances in the $\text{k}\Omega$ range.

To understand the nature of the contact region between Pd electrodes and Bi_4Br_4 flakes, we produced FIB lamellae

of the region of the contacts, as well as slices away from the contacts and analyzed them in a STEM. The results are presented in Fig. 1. Sections taken far from the contacts show excellent crystalline quality, even after exposure to ambient air [Figs. 1(c) and 1(d)]. By contrast, for both contacting schemes, the contact regions are disordered, with Pd/Bi interdiffusion, and have a typical size of approximately 100 nm.

III. ANISOTROPIC ELECTRONIC TRANSPORT IN Bi_4Br_4 FLAKES

We have performed electronic transport measurements on more than ten samples. In the following, we focus on four samples. One, denoted $\mathcal{L}$, is a flake with six contacts [top contacts with Pd(7 nm)/Au(100 nm) electrodes], thus defining five segments, with length ranging from 1 to 5 μm . The flake has a total length of 30 μm , a width of 2.1 μm , and a thickness of around 100 nm [Fig. 4(a)]. The flake is deposited on a doped silicon substrate, which serves as a back gate, covered by 500 nm of oxide.

We also discuss three samples with a Hall bar geometry. Sample $\mathcal{H}_1$, shown in Fig. 2(a), is a flake deposited on top of Ti(5 nm)/Au(10 nm)/Pd(5 nm) contacts. Sample $\mathcal{H}_3$ has similar contacts, while sample $\mathcal{H}_2$ has Pd(7 nm)/Au(100 nm) contacts deposited from the top (Fig. 5 and Sec. 2 in Supplemental Material [41]).

All samples display four-wire resistance values in the $\text{k}\Omega$ range at room temperature that do not change significantly down to 10 K [Fig. 3(a)]. There is a sharp increase of resistance below 1 K, which we attribute to the mesoscopic nature of our sample rather than to the band structure (Sec. 7 in Supplemental Material [41]). The absence of a strong temperature dependence in our measurements contrasts with other works [22,23,30,32–34,38] and suggests that our samples are relatively clean, with a low density of doping defects, and, therefore, have a rather temperature-stable chemical potential. Moreover, the absence of a temperature-activated behavior across the whole temperature range suggests that the Fermi level lies at least 26 meV away from a bulk band edge, thus clearly in the bulk gap.

The four-wire resistances of the segments of sample $\mathcal{L}$ do not increase proportionally to their lengths, which indicates that they are dominated by a contact resistance (Sec. 3 in Supplemental Material [41]). This can be explained by two competing scenarios: either a resistive interface between the Pd/Au contacts and well-conducting bulk or surface states, or ballistic conduction, likely due to hinge states. The variations in resistance from segment to segment can, thus, be explained either by fluctuations in interface quality or by differences in the number of edge modes contacted due to insulating resist residues under the electrodes.

In order to elucidate the flow of current inside the Hall bar devices, we use a formula derived in the context of graph theory [63,64]:

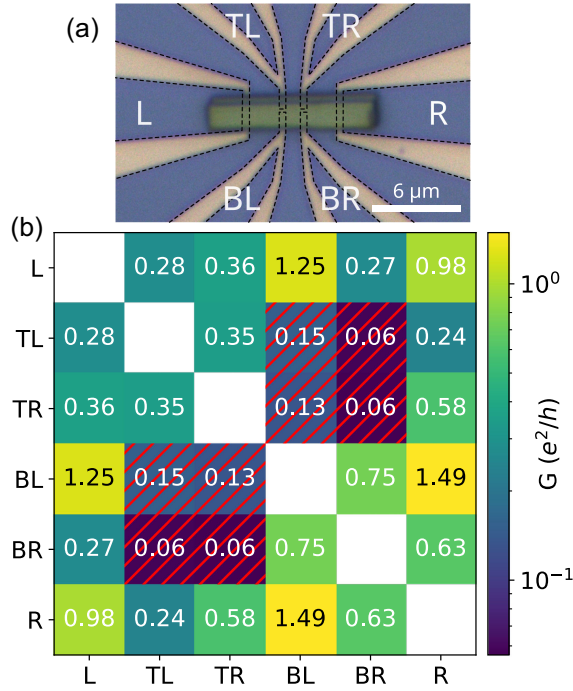


FIG. 2. Anisotropic transport in Bi₄Br₄ sample H_1 at 10 mK and under a 500 nA dc bias. (a) Optical picture of sample H_1 with labeling of the electrodes. (b) Conductance matrix of sample H_1 . The conductances are small in the transverse direction (a axis of the crystal), highlighted by red hatching, and much higher, by a factor 10, in the longitudinal direction (b axis of the crystal).

$$\mathcal{G} = -2\mathcal{R}^{-1} + 2\frac{\mathcal{R}^{-1}\mathbf{u}\mathbf{u}^T\mathcal{R}^{-1}}{\mathbf{u}\mathcal{R}^{-1}\mathbf{u}^T}, \quad (1)$$

which relates two quantities—the Laplacian matrix $\mathcal{G}$ and the resistance distance matrix $\mathcal{R}$, with $\mathbf{u}$ a vector with all coefficients equal to 1. Modeling the sample as a network of lumped resistances with nodes representing terminals, the resistance distance matrix $\mathcal{R}$ contains the two-wire resistances between each possible pair of terminals of the resistance network. The Laplacian matrix $\mathcal{G}$ has matrix elements $\mathcal{G}_{ij} = -G_{ij}$, with G_{ij} the discrete node-to-node conductances, and diagonal coefficients ensuring current conservation $\mathcal{G}_{ii} = \sum_{j \neq i} G_{ij}$. $\mathcal{G}$, thus, corresponds to the conductance matrix in the context of mesoscopic transport [65]. This result, therefore, provides a direct determination of the conductance matrix from simple resistance measurements [66].

The conductance matrix of $\mathcal{H}_1$ is given in Fig. 2(b) (see Sec. 2 in Supplemental Material [41] for samples $\mathcal{H}_2$ and $\mathcal{H}_3$). All contacts to the flake are doubled, so that the lead resistance can be removed for every two-wire configuration. We find transport to be very anisotropic, with relatively high conductances, of the order of the conductance quantum, along the b direction of the crystal. Notably, the conduction along the a direction is consistently among

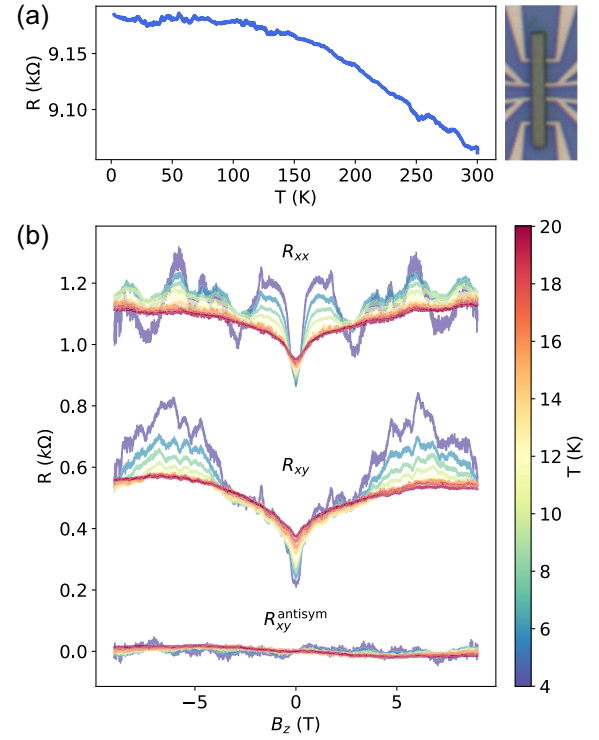


FIG. 3. Temperature dependence of electronic transport in Bi₄Br₄. (a) Temperature dependence of a representative Bi₄Br₄ sample, measured in a physical property measurement system. A Savitzky-Golay filter was applied to the data to get rid of the high-frequency noise. Right: optical picture of the sample used for temperature dependence. (b) Longitudinal (R_{xx}) and transverse (R_{xy}) magnetoresistance at temperatures between 4 (purple) and 20 K (red) for sample H_1 . R_{xy}^{antisym} corresponds to the antisymmetric part of the transverse resistance and shows no Hall effect.

the smallest in all of our devices, with conductances under $0.15 e^2/h$ for contacts 500 nm apart for $\mathcal{H}_1$. Nonzero transverse conductances may result from residual interface resistances that are not eliminated by our measurement procedure.

Finally, we have carried out magnetotransport measurements (Fig. 3) on the same sample, which revealed an essentially symmetrical four-wire transverse resistance dominated by quantum interferences at low temperature (see the next section). The antisymmetrical fraction of the contribution to the magnetoresistance is extremely small, with no clear linear dependence in magnetic field, indicating the absence of Hall effect in the sample. Accordingly, the longitudinal resistance exhibits no Shubnikov–de Haas oscillations (Sec. 8 in Supplemental Material [41]), further demonstrating the absence of transverse transport.

These results are a striking demonstration that the bulk and (001) surfaces are insulating. It suggests that conduction is either due to the lateral (100) surfaces or to 1D states along the b direction.

IV. LOW-TEMPERATURE QUANTUM TRANSPORT

At low temperature, the differential resistance of all samples exhibits a zero-bias peak, which cannot be attributed to self-heating (Sec. 7 in Supplemental Material [41]). Considering the linear regime, with a low excitation current, and for temperatures below 20 K, magnetoconductance has two main characteristics: a weak-antilocalization (WAL) peak at zero magnetic field and sizable universal conductance fluctuations (UCFs) [Fig. 4(c) for sample $\mathcal{L}$].

The low-field WAL peaks are fitted using the Hikami-Larkin-Nagaoka relation:

$$\delta G(B) = \frac{\alpha e^2}{2\pi h} \left[\Psi\left(\frac{1}{2} + \frac{\hbar}{4eBl_\phi^2}\right) - \ln\left(\frac{\hbar}{4eBl_\phi^2}\right) \right] \quad (2)$$

with Ψ the digamma function, B the magnetic field, l_ϕ the phase coherence length, and α a prefactor corresponding to the number of independent channels in parallel [67,68]. Equation (2) is valid for a 2D diffusive conductor in the limit of very strong spin-orbit coupling (spin-orbit length $l_{\text{SO}} \ll l_\phi$). The fitting is done in the magnetic field range $[-1 \text{ T}, 1 \text{ T}]$ for temperatures under 2 K and $[-3 \text{ T}, 3 \text{ T}]$ otherwise with two adjustable parameters: the prefactor α , which is kept constant for a given segment at all temperatures, and the phase coherence length l_ϕ . Formula (2)

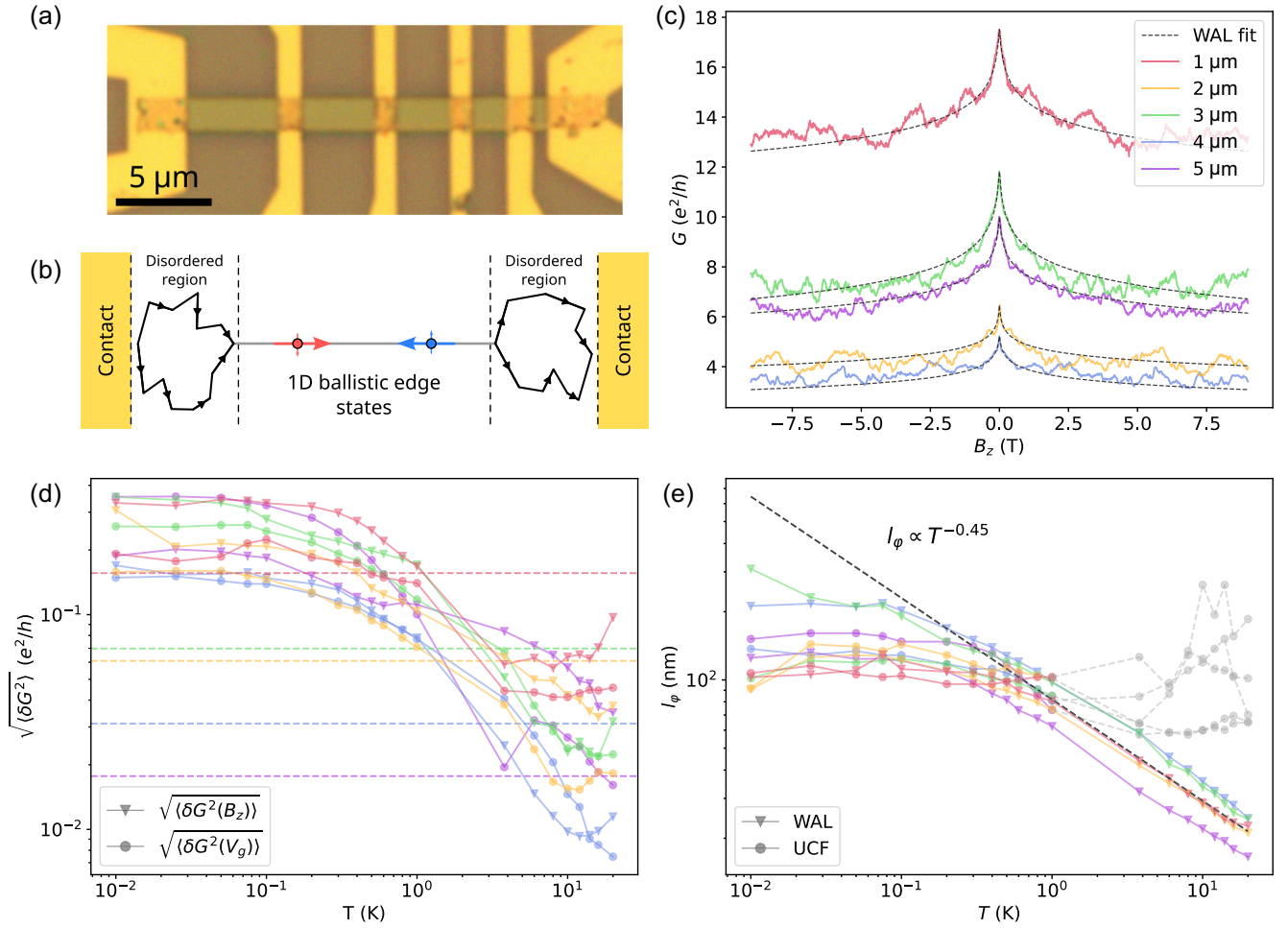


FIG. 4. Coherent quantum transport measurement in sample $\mathcal{L}$. (a) Optical image of sample $\mathcal{L}$. (b) Sketch of the different sample regions contributing to the quantum transport. (c) Magnetoconductance at 10 mK of all segments for an out-of-plane magnetic field. Correspondence between the segment and curve is indicated in the legend. The dashed line is a fit of the WAL component using formula (2). (d) Dependence of UCF amplitudes (in units of e^2/h) as a function of temperature. The amplitude is deduced from the gate dependence (round symbols) or the magnetic field dependence (triangle symbols) of the UCFs. The color code is the same as (c). The horizontal dashed lines are the expected amplitude of the UCF for a diffusive conductor of the same length and same α as the considered segment if self-averaging is assumed. (e) l_ϕ extracted from both the UCFs (round symbols) and WAL (triangle symbols), showing a power-law dependence in temperature with exponent -0.45 . l_ϕ saturates for temperatures below 100 mK. The part of the curve in gray indicates the range of temperature where UCFs cannot be reliably extracted due to experimental noise.

gives an excellent agreement over the entire field range. This seems to indicate the absence of other contributions to the magnetoconductance. However, fits using the WAL expression in the case of quasi-1D conductors yielded both a poor agreement with the experimental data and inconsistent values for l_φ (Sec. 6 in Supplemental Material [41]). We find relatively short l_φ across all segments, between 100 and 200 nm at low temperatures [Fig. 4(e)] and values for α between 2 and 8, which suggests the existence of several uncorrelated electron paths in parallel [67,69,70].

We now turn to the analysis of the UCFs, which were measured as a function of magnetic field [Fig. 4(c)] and gate voltage (Sec. 3 in Supplemental Material [41]). The UCFs exhibit a typical magnetic field scale, given by the half-width of the autocorrelation, which provides another determination of l_φ [71–73]. As shown in Fig. 4(e), these values are close to those extracted from WAL, except at temperatures above 1 K, for which the experimental noise outweighs the UCF signal. For all sections of sample $\mathcal{L}$, l_φ extracted from both WAL and UCFs follows a power-law dependence down to approximately 100 mK, below which it saturates. We find an exponent of -0.45 , consistent with the expected $-1/2$ for dephasing in 2D dominated by electron-electron interactions [67].

Despite l_φ being much shorter than the segment length, we find sizable fluctuation amplitudes with no systematic decrease with increasing segment length. We recall that UCF in diffusive conductors should decrease as the segment length extends beyond l_φ , due to self-averaging. By contrast, the absence of a systematic decrease in fluctuations amplitude with length [Fig. 4(d)] shows that the quantum interference we measure does not originate from a diffusive path within the flake.

Rather, we propose that diffuse transport occurs only in the damaged region near the contacts, roughly 100 nm in size as discussed above. Those diffusive regions are connected by 1D ballistic channels, up to 5 μm long, so that coherent transport occurs over up to 5 μm . We conjecture that it is the transmission from contacts to ballistic channel that is modulated by quantum interference in the disordered regions. This is what leads to the absence of self-averaging and to the high field scale of UCF and WAL. This scenario was already pointed out in the context of the conductance of edge states in the integer quantum Hall effect [74] and is qualitatively reproduced by our numerical simulations [Fig. 8(a) and Sec. 5 in Supplemental Material [41]].

To confirm this picture, we studied the dependence of quantum interference on magnetic field orientation. We performed magnetoconductance measurements on sample $\mathcal{L}$ using a vector magnet, which allows to apply magnetic fields in arbitrary directions up to 1 T. We present field cuts in three different planes: in-plane, longitudinal and transverse, all taken at 3.8 K to remove the contribution from conductance fluctuations, which complicates the analysis [Figs. 5(a)–5(c)]. The first two cuts clearly highlight a near

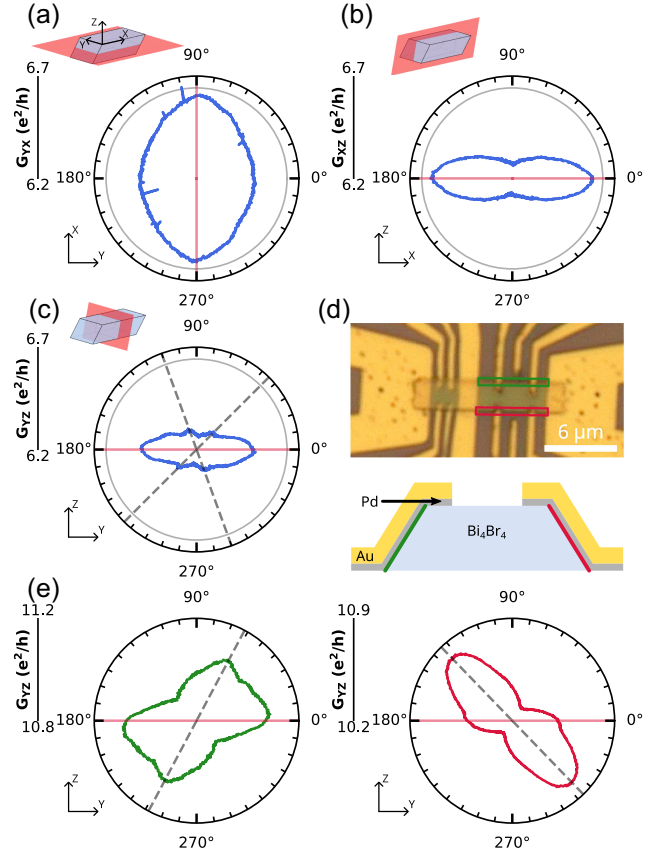


FIG. 5. (a)–(c) Magnetoconductance anisotropy at 3.8 K for the 4 μm segment of sample $\mathcal{L}$. (a) Top: schematic image of the flake and the XY plane in which the magnetic field is applied. Bottom: variation of the conductance by sweeping the magnetic field angle in the XY plane, for a magnetic field modulus of 1 T. The angle 0° corresponds to a field aligned along the Y direction. Magnetoconductance is highest in the Y direction, defining an extended lobe in the perpendicular X direction, as indicated by the red line. (b) Similar data for a magnetic field in the XZ plane, showing a maximum magnetoconductance along the Z direction. (c) Similar data for a magnetic field in the YZ plane, showing a maximum magnetoconductance along the Z direction and side lobes, indicated by gray dashed lines, attributed to the geometry of the contacts (see the text). (d) Top: optical image of the sample $\mathcal{H}_2$. Bottom: sketch of the cross section of the flake $\mathcal{H}_2$. (e) Magnetoconductance with a magnetic field of 1 T in the YZ plane for lateral contacts indicated in (d) in green (left part) or red (right part). The direction of maximum magnetoconductance is not related to the crystalline orientation but rather to the geometry of the lateral contacts.

absence of magnetoconductance along the b axis of the flake [X direction in Figs. 5(a) and 5(b)]. The longitudinal and transverse cuts reveal that the decrease of conductance with magnetic field is maximum for an out-of-plane field, leading an extended lobe in the polar plot in the perpendicular direction (indicated by the red lines in Fig. 5), demonstrating that the surface of diffusion essentially lies in the plane of the sample. This behavior, together

with the absence of self-averaging of the UCFs, rules out diffusion on the sides surfaces of Bi_4Br_4 as the source of the observed quantum interferences.

In addition to the large out-of-plane magnetoconductance observed in the transverse cut, several devices contacted from the top exhibit other lobes [gray dashed lines in Figs. 5(c), 5(f), and 5(g)] that are not related to any specific crystalline direction of Bi_4Br_4 . Rather, these axes correspond to the planes of the electrodes deposited on the lateral side of the flake. Indeed, the disordered region induced by the contacts follows the slope of the etched crystal, which defines the plane of diffusion for the WAL [Fig. 5(e)]. Moreover, for sample $\mathcal{H}_2$, with a Hall bar geometry, we find that, for contacts on the side of the flakes, only one of the side lobes is present and that it is more prominent than the out-of-plane one. This confirms that each side lobe comes from one flank of the flake. In contrast, samples contacted from below only show magnetoconductance out of plane, confirming that the origin of the side lobes lies in the contact method.

In conclusion, the magnetotransport at low temperature and its anisotropy with magnetic field orientation confirms that the regions determining the mesoscopic fluctuations are the diffusive contact regions, leading to a field-dependent transmission to 1D ballistic channels.

V. NEGATIVE FOUR-WIRE RESISTANCE

While the behavior of samples contacted from above and from below are similar on many aspects, there are a few clear differences. The first is the presence of a higher interface resistance for bottom contacts, and the second is the absence of a four-wire negative differential resistance in top-contacted samples.

Indeed, we observe negative four-wire resistance fluctuations that extend between -6 and 10 k Ω with a two-wire resistance of 20 – 35 k Ω for $\mathcal{H}_1$ (Fig. 6). This corresponds to a sizable fraction of the theoretical limit of $\pm R_{2\text{wires}}$, predicted for ballistic conduction measured with moderately invasive voltage probes [75]. Indeed, the two-wire resistance measured through the voltage probes indicated in blue in Fig. 6 is high (approximately 150 k Ω), which may indicate a weakly perturbative contact to the edge states that can favor the appearance of these negative fluctuations. Indeed, contacts from below may allow for the hinge states to be preserved to a certain extent, if they are not cut by a large Pd/Bi alloy region (Fig. 1). We emphasize that this prediction of a four-wire negative differential resistance for ballistic systems has not been reported by many studies. Experiments on single-walled carbon nanotubes have found negative fluctuations reaching the theoretical limit [76], as well as measurements on high-mobility GaAs 2DEGs, which were observed only in a specific field range [77,78]. Both of these experiments featured exceptionally clean systems with noninvasive voltage probes, which contrasts with the disorder induced at the

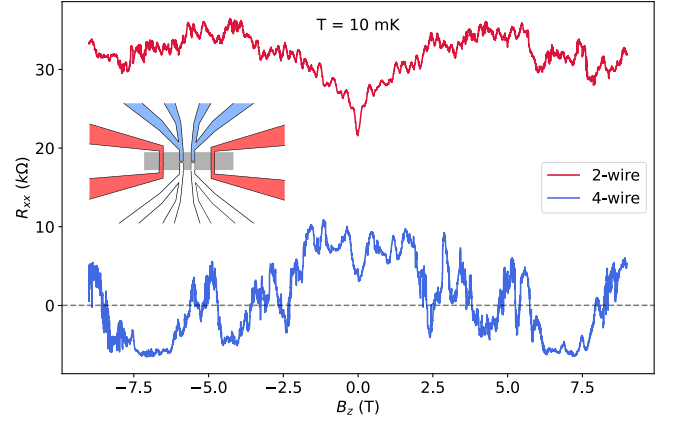


FIG. 6. Two- and four-wire longitudinal resistances of sample $\mathcal{H}_1$ as a function of magnetic field at $T = 10$ mK, measured with a 10 nA ac current. The four-wire measurement is negative in several field ranges. Inset: sample geometry with the Bi_4Br_4 flake in gray, current injection in red, and voltage probes in blue. For the two-wire measurement, the voltage probes are the red wires.

contacts with Bi_4Br_4 . Therefore, this observation of negative differential resistance in our samples could be a consequence of the expected topological protection of hinge states against elastic disorder.

VI. AHARONOV-BOHM INTERFERENCE

In addition to the aperiodic UCFs and WAL displayed in the magnetoconductance, several samples also display periodic oscillations, leading to well-defined peaks in their Fourier transform (FFT).

In particular, the longitudinal magnetoresistance of sample $\mathcal{H}_1$ shows a very clear periodicity of around 0.24 T at low temperatures leading to a FFT peak at 4 T $^{-1}$ [Figs. 7(a) and 7(c)]. Note that the periodicity of the oscillations does not depend on temperature but the amplitude does (Sec. 4 in Supplemental Material [41]). We interpret this as Aharonov-Bohm (AB) interference [79] between two 1D states of length 6 μm and separated by 2.7 nm, allowed by the partially coherent contact region, as confirmed by numerical simulations [Fig. 8(b) and Sec. 5 in Supplemental Material [41]]. One possible reason for this particularly clear signature of AB interference in this sample is the small separation between the lateral electrodes, which selects only a few channels that can carry current coherently across the flake without being interrupted by a contact [Fig. 7(b)]. The good visibility of the AB oscillations is also favored by the small lateral distance between edge states, comparable to the mean free path in the diffusive contact (Sec. 5 in Supplemental Material [41]). Besides, we find an exponential decay with temperature of the AB amplitude [inset in Fig. 7(c)]. This temperature behavior has been reported in quasi-1D ballistic rings [80], 3D topological insulators [81–83], and hinge state loops in thin MoTe_2 flakes [84].

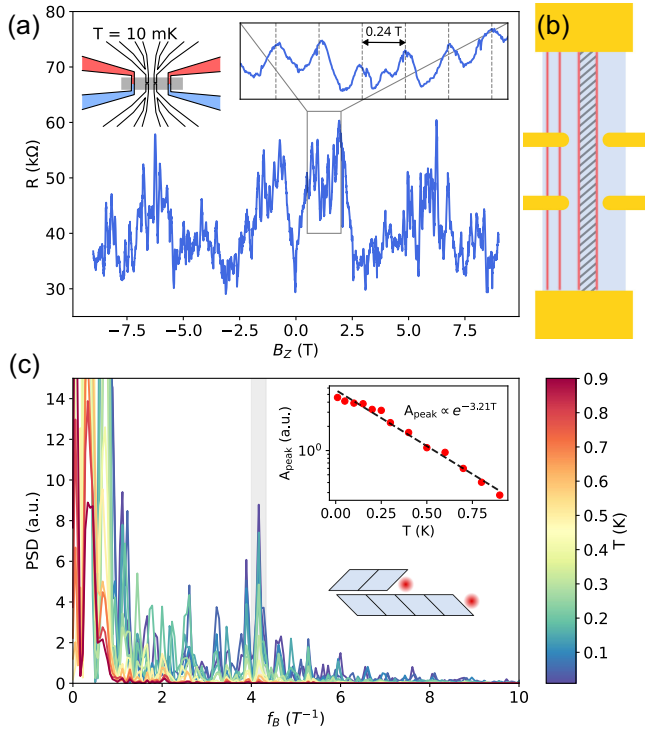


FIG. 7. Aharonov-Bohm effect of sample $\mathcal{H}_1$. (a) Magnetoconductance at low temperature exhibiting periodic conductance oscillations, with a field period of 0.24 T. Left inset: configuration of the contacts used for current injection (red) and voltage probing (blue). Right inset: enlargement of a reduced field range. (b) Schematic picture of the Bi_4Br_4 flake (blue) with its contacts (yellow). The red lines represent 1D states, some of which are interrupted by lateral contacts. The two central 1D states delimit a surface (hatched area) defining the magnetic field periodicity of the AB effect. (c) Power spectral density of the magnetoconductance of sample $\mathcal{H}_1$ at different temperatures. The gray area indicates the peak corresponding to the observed conductance oscillations, taking into account an uncertainty of $\pm 20 \times 20 \text{ nm}^2$. Top inset: temperature dependence of the FFT peak area. It is consistent with an exponential decay with temperature, of exponent -3.21 . Bottom inset: schematic image of a cross section of Bi_4Br_4 exhibiting a configuration of edge states compatible with the observed AB periodicity. The blue diamonds represent individual crystalline unit cells, and the red dots show the position of edge states.

This dependence is, however, at odds with the result of Hossain *et al.* [39] on thin flakes of Bi_4Br_4 , which instead exhibits a power-law dependence that is attributed to the extremely long, effectively temperature-independent, phase coherence length of edge states. The functional difference between the decay of the UCFs and AB oscillations with temperature in our samples suggests that their coherence is limited by distinct mechanisms. One would expect the amplitude of AB oscillations to be proportional to the probability of an electron trajectory to complete a loop without phase-breaking event $\exp(-P/l_\phi)$, with P the perimeter of the closed trajectory [79]. The exponential

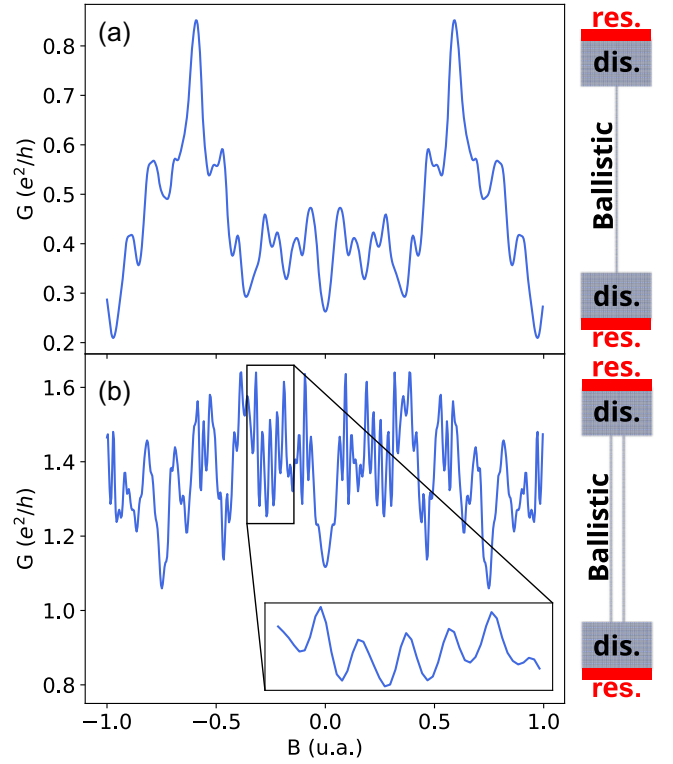


FIG. 8. Numerical simulations of the conductance of ballistic channels connected to reservoirs via a disordered coherent region realized with the KWANT software package [85]. (a) Case of a single ballistic channel showing WAL and UCFs. Right: tight binding network used to calculate the conductance (Sec. 5 in Supplemental Material [41]). The long, narrow region is the ballistic channel (noted ballistic). It is terminated by two diffusive coherent regions (noted dis). The reservoirs are represented by the red rectangles (noted res). (b) The same for two ballistic channels in parallel connected to reservoirs via a disordered coherent region. The conductance exhibits AB oscillations on top of WAL and UCFs. Right: corresponding tight binding network [41].

decay of the peak amplitude in the FFT with temperature, therefore, implies a $1/T$ dependence of the phase coherence length and taking $P = 12 \text{ } \mu\text{m}$, $l_\phi = 3.7 \text{ } \mu\text{m}$ at 1 K. The fact that we can measure such a long phase coherence length implies that phase breaking occurs predominantly within the hinge states, so that the electron trajectories contributing the most to the AB effect do not explore the diffusive regions much, thanks to the very small separation between hinge states (2.7 nm). These results emphasize the coherence of electronic transport in relatively long Bi_4Br_4 flakes.

Strikingly, for device $\mathcal{L}$, several of the FFT peaks correspond to a field scale proportional to the length of the considered segment. Hence, if the magnetic frequency is rescaled by the segment length [Fig. 9(b)], it is possible to identify some peaks in the FFT of different segments that correspond to the same rescaled magnetic frequency. We interpret this as AB interference around surfaces of fixed widths across each segment. We postulate that these oscillations originate from coherent 1D hinge states that

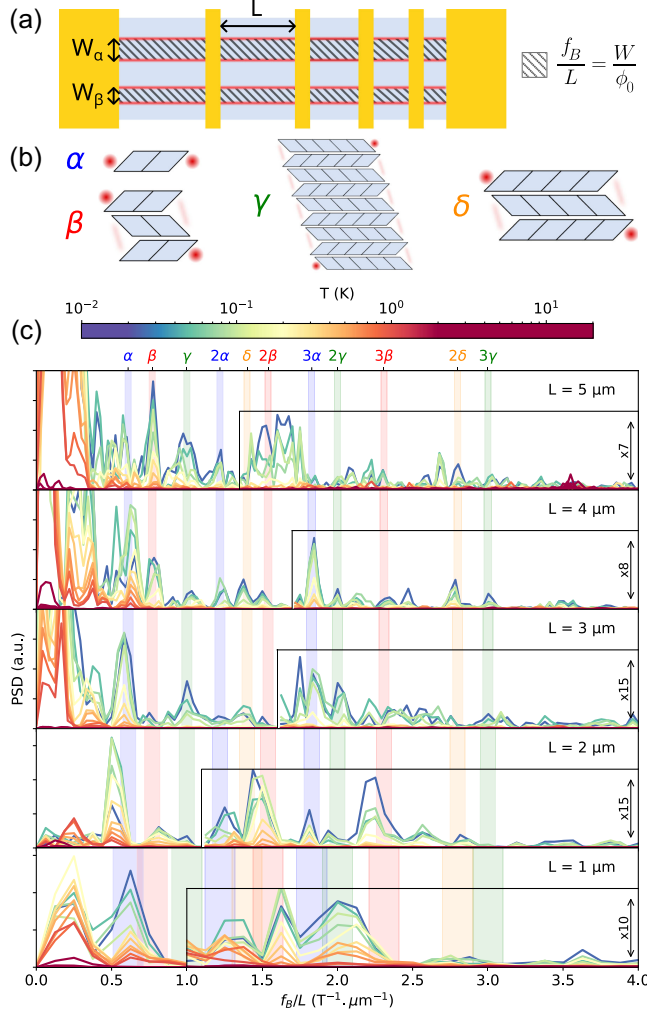


FIG. 9. AB effect in sample $\mathcal{L}$. (a) Schematic picture of the sample with the Bi₄Br₄ flake in blue and the contacts in yellow. The red lines represent 1D states, which are separated by a distance W and interrupted by the contacts. For a segment of length L , the AB magnetic field period is $\Phi_0/(WL)$, thus leading to a magnetic frequency $f_B = WL/\Phi_0$. (b) Hinge state configurations consistent with the observed AB peaks in the FFT [see (c)], where a blue diamond represents a crystalline unit cell and the red dots correspond to the hinge states. (c) Power spectral density of the magnetoconductance of the different segments of sample $\mathcal{L}$ and for temperatures between 10 mK and 10 K, indicated by the color bar. The FFT frequencies are rescaled by the length of the corresponding segment. Shaded areas correspond to an uncertainty of $\pm 20 \times 20 \text{ nm}^2$ around a few identified peaks [see (b) and Sec. 4 in Supplemental Material [41]], denoted α (blue), β (red), γ (green), and δ (orange), and their harmonics.

extend over the entire length of the flake, which were subsequently divided into several segments by the fabricated contacts. Consequently, from segment to segment, there is a constant lateral separation between the hinge states, as illustrated in Fig. 9(a). In fact, the magnetic frequencies identified in the FFT spectrum can be related to some specific configurations of hinge states that are

consistent with the crystalline structure of Bi₄Br₄ [Fig. 9(b)]. As the AB effect is visible for all segments, including the longest ones, the phase coherence length of the edge states must be of the order of 5 μm or more. However, the limited phase coherence in the contact regions induces an upper cutoff to the lateral distance between 1D channels allowing for AB interferences, such that we do not observe peaks in the Fourier spectrum corresponding to distances greater than a few tens of nanometers. Moreover, we attribute the absence of a clear magnetic field periodicity in the magnetoconductance signal to the multiplicity of frequencies present in the Fourier transform, which are due to the many 1D coherent channels of the flake that may interfere with each other. The temperature dependence of the identified peaks exhibits an exponential decay consistent with a $1/T$ dependence of phase coherence length and a value at 1 K similar to the one of sample $\mathcal{H}_1$. Moreover, the existence of AB interference in all segments of $\mathcal{L}$ up to 5 μm is consistent with long phase coherence length.

VII. CONCLUSION

We have probed the electronic transport in micrometer-size Bi₄Br₄ flakes and have demonstrated a strongly anisotropic transport. We have highlighted two distinct types of contributions to low-temperature coherent transport and argue that they reveal the existence of long coherent 1D hinge states contacted to reservoirs via a small coherent disordered region. The first type of contribution yields universal conductance fluctuations and weak antilocalization, arising from phase coherence in the region close to the contacts, which modulate the transmission through the 1D states. The second type of contribution is the periodic Aharonov-Bohm conductance oscillations due to interference between adjacent 1D coherent channels. We emphasize that both of these effects are due to the very nature of the contacts, which exhibit some phase coherence despite being rather invasive. These effects indicate that the 1D states are ballistic and coherent over several micrometers at low temperatures. The signature of the 1D ballistic states, located at step edges distributed at the surface of the Bi₄Br₄ crystal, and their interference are, thus, paradoxically revealed thanks to the coherent diffusive nature of the contact regions. It would be interesting to achieve the opposite limit of clean, crystal-preserving contacts in small samples with a reduced number of edge states, in order to access the conductance quantization regime. Also, the signature of the helical nature of these 1D states in multilayer samples remains to be demonstrated. Lastly, the use of superconducting contacts to enter the regime of proximity-induced superconductivity is of great interest, with expected perfect Andreev reflection, and would further enhance the potential of Bi₄Br₄ as a second-order topological insulator.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [86].

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