

Two-dimensional Shubnikov–de Haas oscillations in PtSe₂: A fermiological charge carrier investigation

Julian Max Salchegger^{1,*}, Rajdeep Adhikari^{1,2,†}, Bogdan Faina¹, and Alberta Bonanni^{1,‡}

¹*Institut für Halbleiter-und-Festkörperphysik, Johannes Kepler University, Altenbergerstr. 69, A-4040 Linz, Austria*

²*Linz institute of Technology, Johannes Kepler University, Altenbergerstr. 69, A-4040 Linz, Austria*



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High magnetic field and low temperature transport measurements are carried out in order to gain insight into the properties of the charge carriers of PtSe₂. In particular, the Shubnikov–de Haas oscillations arising at applied magnetic field strengths $\gtrsim 4.5$ T are found to occur exclusively in plane. An analysis of the oscillations via the Lifshitz-Kosevich formalism allows determining the charge carrier's cyclotron mass, quantum transport time, Berry phase, Fermi surface cross section, and Dingle temperature. The oscillations emerge at a layer thickness of ≈ 18 nm and decrease in frequency for thinner PtSe₂ flakes. Further, weak antilocalization (WAL) is observed despite the presence of magnetic moments from Pt vacancies, which typically inhibit such effects. An explanation is provided on how WAL and the Kondo effect can be observed within the same material.

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I. INTRODUCTION

The transition-metal dichalcogenide PtSe₂ is a van der Waals-layered semimetal with type-II Dirac cones [1,2], which has been suggested for applications in electronic [3–5] and spintronic [6] devices, and has recently been considered as a platform for the detection of the orbital Hall effect (OHE) [7,8]. Unlike the conventional Hall effect, the OHE does not require an applied magnetic field to emerge: The orbital momentum of the charge carriers couples to an applied electric field, resulting in the generation of a transversal orbital current. By injecting the orbital current into a material with high spin-orbit coupling (SOC), or, e.g., through Rashba SOC [9], the orbital current can be converted to a spin current. Harnessing orbital currents promises applications in spintronic [10] and orbitronic [11,12] devices. Additionally, the OHE may account for the discrepancy between the observed and theoretical magnitudes of the spin Hall effect in materials such as Pt [13]. PtSe₂ is predicted to host significant orbital currents [8] and offers the advantage of inherent SOC, due to the presence of heavy Pt atoms. Understanding the nature of the charge carriers present in PtSe₂, as well as its electronic structure, is a fundamental step towards the experimental realization of an orbital Hall device.

Here, high magnetic field and low temperature transport studies are carried out to investigate key aspects of the charge carriers of PtSe₂ flakes, such as the cyclotron mass, the quan-

tum scattering time, the mean free path, and the Berry phase. In a previous work [14], we demonstrated that PtSe₂ flakes, exfoliated from a bulk crystal, contain Pt vacancies, resulting in a Kondo effect. Here, a further analysis reveals that the vacancy concentration is not uniform, since the Kondo effect varies across samples. A reduced number of Pt vacancies leads to the emergence of weak antilocalization (WAL) in the four-terminal resistance measurements, heralding SOC in the samples. To investigate the above-mentioned charge-carrier properties, Shubnikov–de Haas (SdH) oscillations, which are closely linked to the orbital motion of the charge carriers and to their Fermi surface, are analyzed [15–17]. The oscillations arise in both the longitudinal and the transverse resistance when the applied magnetic field exceeds ~ 4 T.

The spectral analysis of the oscillations points at only one dominant type of charge-carrier orbit. The quantum scattering time τ and the cyclotron mass m_c are determined together with the Berry phase Φ_B . The thickness dependence of the extremal Fermi surface cross-sectional area indicates that the bulk limit of the electronic structure establishes only for thicknesses $t > 20$ nm. Using the angular dispersion of the SdH oscillations, insights into the Fermi surface shape are gained.

II. EXPERIMENT

The PtSe₂ flakes considered in this work are mechanically exfoliated from bulk crystals grown by HQ Graphene [18]. Optical microscopy (Fig. 1) shows that the angles between flake edges are multiples of 60° , as expected from the C_3 symmetry of the crystal. Atomic force microscopy and x-ray photoemission spectroscopy are performed to characterize the flatness, crystallinity, and chemical constituents of the flakes, as depicted in Figs. S3 and S4 of the Supplemental Material [19]. The samples are listed in Table I: Samples A–F are obtained from bulk crystal Batch 1, while samples X1 and X2 are from Batch 2. The samples cover a range of thicknesses from 8 to 26 nm, resulting in a semimetallic band structure

*Contact author: julian.salchegger@jku.at

†Contact author: rajdeep.adhikari@jku.at

‡Contact author: alberta.bonanni@jku.at

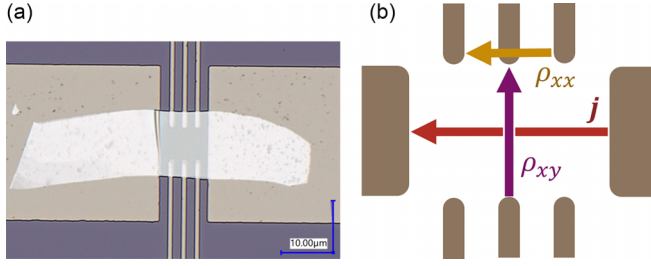


FIG. 1. (a) Optical image of sample X1. (b) Schematic of the Hall bar with the direction of the current density j . Measuring the voltage difference between two terminals that lie on a line parallel to j , results in ρ_{xx} while the voltage difference between terminals, which lies on a line normal to j , yields ρ_{xy} .

[20], while also providing variation in the extent of two-dimensional (2D) confinement of the charge carriers and in the surface-to-bulk ratio. A cover layer of hexagonal boron nitride (hBN) is included to improve the signal-to-noise ratio for the thinnest samples and to study the interface effects originating from the interaction between hBN and the Pt vacancy magnetic moments.

Prefabricated Hall bars are patterned via electron-beam lithography and 10 nm of Pt is deposited via sputtering. The flakes are then placed onto the Pt contacts. An optical image of sample X1 is shown in Fig. 1(a). In Fig. 1(b), the direction of the current density j and exemplary voltage terminal selections for the measurement of the longitudinal resistance and the Hall voltage are marked.

Au wires are bonded to the Hall bar using an In solder agent. The low T /high $\mu_0 H$ transport measurements are performed in a Janis Super Variable Temperature 7TM-SVM cryostat equipped with a 7 T superconducting magnet and a homemade rotatory sample holder (SH) with two angular degrees of freedom. A lock-in amplifier ac technique at 277 Hz is used for measuring the magnetotransport properties of the PtSe₂ flakes.

III. RESULTS AND DISCUSSION

A. Hall voltage

The Hall voltage V_H is obtained when choosing voltage probing terminals orthogonal to the source-drain direction. The slope of $V_H(H)$ is used to determine the charge-carrier density $n = \frac{\mu_0 H}{V_H} \frac{I}{te}$, with I being the source-drain current. The

TABLE I. Considered samples: thickness t , cover layer, and respective batch of bulk crystals.

Sample	t (nm)	Cover	Bulk crystal Batch
A	18		1
B	20		1
C	26		1
D	8	hBN	1
E	10	hBN	1
F	11		1
X1	17		2
X2	12		2

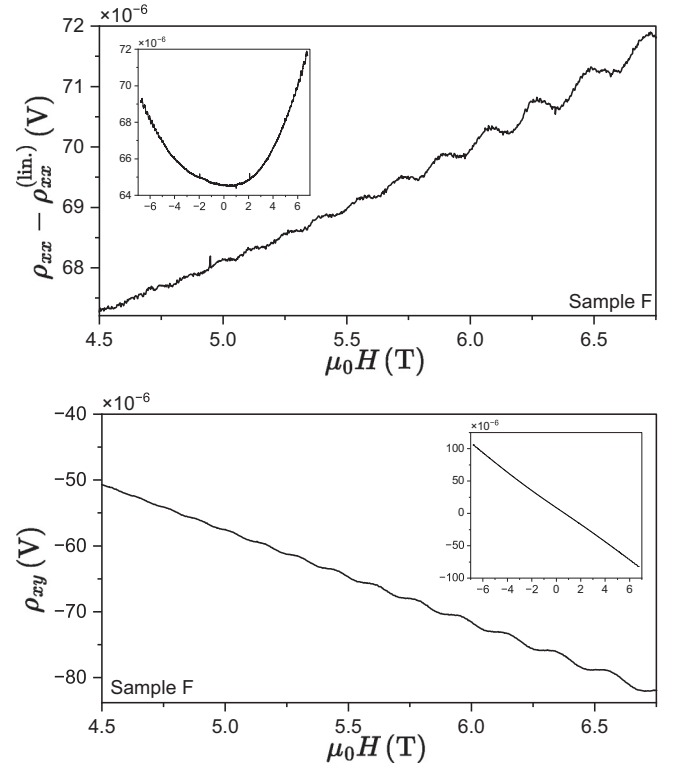


FIG. 2. Upper panel: longitudinal four-terminal resistance $\rho_{xx} - \rho_{xx}^{(\text{lin.})}$ of sample F over applied magnetic field for $\mu_0 H \geq 4.5$ T at 2 K. Inset: increased range of $|\mu_0 H| \leq 6.8$ T. Lower panel: equivalent plot for ρ_{xy} .

samples show $n(2\text{ K}) \sim (2 \times 10^{20})\text{ cm}^{-3}$. Exemplary plots of $V_H(H)$ are presented in Supplemental Material [19] Fig. S7 for samples F and X1.

B. Shubnikov-de Haas oscillations

The current density j is induced between the source-drain contacts and the four-terminal longitudinal resistivity ρ_{xx} is determined by measuring the voltage difference between two terminals that lie on a line parallel to j . The transversal resistivity ρ_{xy} (cognate with V_H) is measured between two terminals that lie on a line normal to j . The longitudinal resistance $\rho_{xx}(H)$ of sample F at 2 K is reported in the inset to the upper panel of Fig. 2, with H being the scalar value of the applied magnetic field ($H = \mathbf{H} \cdot \hat{e}_H$ and $\hat{e}_H$ is the axis along which the magnetic field is applied). Since the flake edges are, in general, not exactly parallel to j , the longitudinal resistivity contains a linear component $\rho_{xx}^{(\text{lin.})} \approx (2.62 \times 10^{-6})\text{ V/T}$, which originates from the Hall voltage. The longitudinal resistance $\rho_{xx} - \rho_{xx}^{(\text{lin.})}$ shows a dependence that is approximately quadratic in H .

For $|\mu_0 H| \gtrsim 4.5$ T, SdH oscillations emerge, as displayed in the upper panel of Fig. 2. The Hall voltage as a function of applied magnetic field captured at 2 K shows a quasilinear dependence $\rho_{xy} \propto -H$, as depicted in the inset to the lower panel of Fig. 2, until, for $|\mu_0 H| \gtrsim 4.5$ T, SdH oscillations emerge, as reported in the lower panel of Fig. 2.

The origin of the SdH oscillations is the Landau quantization of the electronic states, which modulates the density of

states (DOS) at the Fermi level when varying $\mathbf{H}$. A local maximum in $\rho_{xx}(\mathbf{H})$ corresponds to a Landau level (LL) coinciding with the Fermi level, as the increased number of available states (compared to no LL coinciding with the Fermi level) enhances the electron-electron scattering. A similar mechanism also affects ρ_{xy} , in which case the modulation of the DOS at the Fermi level alters the charge-carrier density n and thus the Hall voltage $\rho_{xy} \cong V_H \propto \frac{1}{n}$ shows a minimum when a LL coincides with the Fermi level.

Let $d \in \{xx, xy\} \equiv \{\text{longitudinal, transversal}\}$ denote the measurement direction and $\tilde{\rho}_d$ be the oscillatory part of ρ_d , such that $\rho_d = \rho_d^{(\text{bg.})} + \tilde{\rho}_d$, where $\rho_d^{(\text{bg.})}$ is a smooth background. Then, $\tilde{\rho}_d(H, T)$ follows from the Lifshitz-Kosevich (L-K) relation [21,22]:

$$\tilde{\rho}_d = AT \left(\frac{5}{2} \left(\frac{H}{2\mathfrak{F}} \right)^{1/2} + \frac{3}{2} \left(\frac{H}{2\mathfrak{F}} \right) \right) \times \frac{\exp\left(-x \frac{NM}{H}\right) \cos(\pi M)}{\sinh\left(T \frac{NM}{H}\right)} \cos\left[2\pi \left(\frac{\mathfrak{F}}{H} + \phi \right)\right], \quad (1)$$

where A is an amplitude prefactor, ϕ the phase of the oscillations, x the Dingle temperature (x is not a physical temperature, but is related to defects and impurities in the sample), $M = \frac{m_c}{m_0}$ the relative electron cyclotron mass (m_c is the electron cyclotron mass and m_0 the free electron rest mass), and $N = 2\pi^2 \frac{m_0 k_B}{e\hbar}$ a constant ($\hbar$ is the reduced Planck constant, e the elementary charge, and k_B the Boltzmann constant). Finally, $\mathfrak{F}$ is the frequency of the oscillations (periodic in $\frac{1}{H}$), which is linked to S_F , i.e., the extremal cross-sectional area of the Fermi surface normal to $\mathbf{H}$, via $\mathfrak{F} = \frac{\hbar}{2\pi e} S_F$. The adaptation of the L-K formula given in Ref. [22] is described in Appendix B. The experimental value of $\tilde{\rho}_d$ is obtained by subtracting a polynomial background from ρ_d . Equation (1) is augmented with a second-order polynomial,

$$\tilde{\rho}_d \rightarrow \tilde{\rho}_d + A^{(0)} + A^{(\text{lin.})}H + A^{(\text{quad.})}H^2, \quad (2)$$

to account for any residual background, and the fitting of $\tilde{\rho}_d$ as a function of the applied magnetic field of sample F at 2 K with Eq. (2) is reported in Fig. 3.

The maxima in $\tilde{\rho}_{xx}(H)$ occur when the scattering rate is maximal and thus correspond to (whole) integer LL numbers n_{LL} (and minima to half-integers). Therefore, in the conductivity $\sigma_{xx} = \frac{\rho_{xx}}{\rho_{xx}^2 + \rho_{yy}^2}$, minima of $\tilde{\sigma}_{xx}(H)$ correspond to integer LL numbers. The identified levels can be found in the upper panel of Fig. 4, which shows $\tilde{\sigma}_{xx}(|H|^{-1})$ for sample F at 2 K for both $H > 0$ and $H < 0$. The oscillations match in frequency and phase, which points at the underlying Landau quantization being symmetric under inversion of the applied magnetic field. The differing amplitudes are attributed to an asymmetry in the flake placement with respect to the source-drain axis, resulting in asymmetric contribution of the edge conductance channels. The index-axis cutoff of the Landau-fan diagram is the oscillation phase ϕ , which relates to the Berry phase [23] according to

$$\Phi_B = 2\pi \left(\frac{1}{2} - \phi \pm \delta \right) \mod 2\pi. \quad (3)$$

The phase shift δ is taken to be zero [17] in the case of a 2D SdH effect, which is reflected in the fact that only the

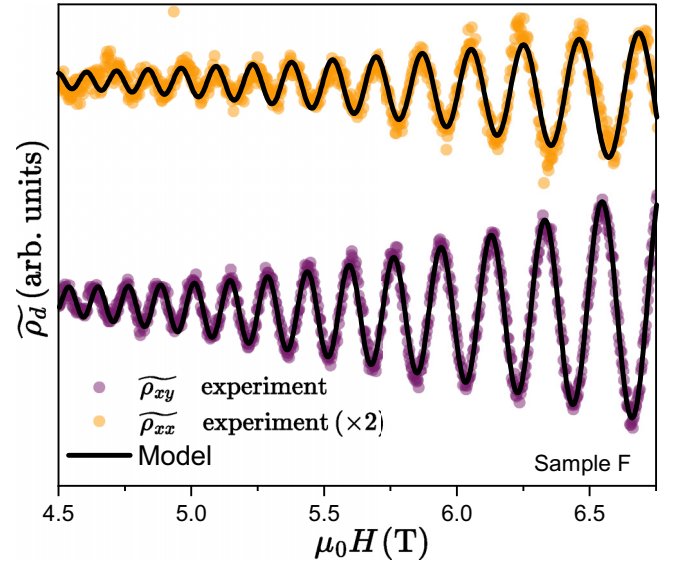


FIG. 3. $\tilde{\rho}_d$ over applied magnetic field for sample F at 2 K. Solid curves: L-K fits. The obtained values are given in Table II.

out-of-plane components of $\mathbf{H}$ contribute to the SdH oscillations. From the Landau-fan diagram, $\Phi_B = (0.82 \pm 0.09)\pi$ is determined, suggesting nontrivial charge carriers. The physical origin of this nontrivial value is unclear, since the Dirac cone is located ≈ 1.5 eV below the Fermi level, the dispersion of which forms a hole pocket at the Fermi level [24]. The transport of the investigated system is, however, dominated by electrons, as evidenced by the sign of the Hall resistance and by the value of $S_F^{(11\text{ nm})} \approx 0.02 \text{ \AA}^{-2}$ being smaller than the observed size of the hole pocket [24]. It has to be mentioned, that the extrapolation from $n_{LL} = 29$ to 0 is volatile with regard to the exact manner in which the extrema of $\tilde{\sigma}_{xx}(|H|^{-1})$ are detected and at which value of $|H|^{-1}$ the cutoff is set, since the amplitude of the oscillations damps significantly as $|H|^{-1} \rightarrow \infty$. To substantiate the value of the Berry phase, a direct L-K fit of σ_{xx}^{-1} is performed (provided in Fig. S1 of the Supplemental Material [19]), which yields $\Phi_B^{(\text{L-K})} = (1.01 \pm 0.05)\pi$.

A fast Fourier transform (FFT) of the SdH oscillations is performed to gain further insight into the charge-carrier properties. The data treatment is detailed in Sec. B of the Supplemental Material [19]. The resulting spectra are shown in the left panel of Fig. 5, where the FFT amplitude $\mathcal{A}$ is given as a function of the oscillation frequency $\mathfrak{F}$: For sample F, the position of the local maximum at $\mathfrak{F}_{\text{FFT}} \approx 195 \text{ T}$ is in agreement with the frequency $\mathfrak{F}_{\text{L-K}} = 193.6 \text{ T}$ from the L-K fit. Similarly, the frequencies obtained for sample A are $\mathfrak{F}_{\text{FFT}} \approx 250 \text{ T} \approx \mathfrak{F}_{\text{L-K}} = 254.9 \text{ T}$. The shift in the value of $\mathfrak{F}$ with increasing the sample thickness t from 11 to 18 nm, points at $t = 11 \text{ nm}$ not having a bulk band structure, because $\mathfrak{F} \propto S_F$. For $t \geq 20 \text{ nm}$ (samples B and C) no local maximum at $\mathfrak{F} > 0$ is discernible, and indeed, no SdH oscillations can be gleaned in $\tilde{\rho}_{xy}$, as evidenced in the right panel of Fig. 5. A similar effect is observed for sample E, which has a thickness comparable to that of sample F, but is covered with hBN; a peak at $\mathfrak{F} \approx 200 \text{ T}$ can be found and $\tilde{\rho}_{xy}$ does show SdH oscillations. However, the peak height of $\mathfrak{F}_{\mathcal{F}}$ and the signal-to-noise ratio

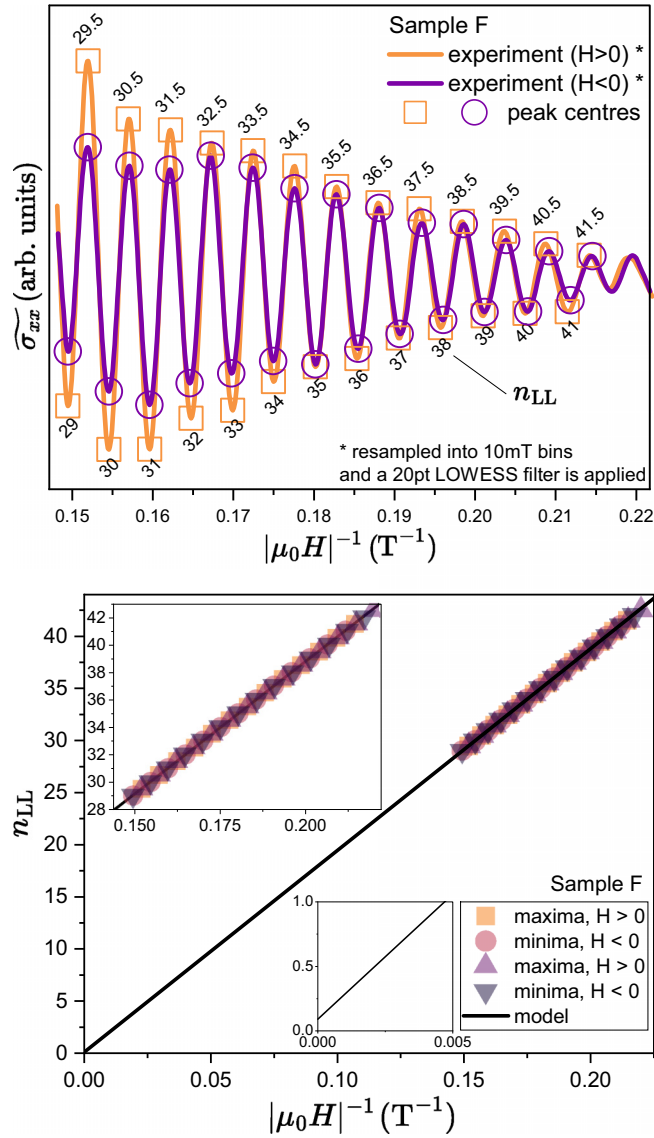


FIG. 4. Upper panel: $\tilde{\sigma}_{xx}$ of sample F at 2 K over inverse applied magnetic field magnitude and extracted peak center positions marked as squares/circles. Lower panel: Landau-fan diagram showing the LL indices over the peak positions from the upper panel and a linear fit. Insets: magnification of the identified peak positions and of the axis cutoff region. The obtained parameters are provided in Table S1 of the Supplemental Material [19].

observed in $\tilde{\rho}_{xy}$ are substantially lower than those of sample F. The hBN coating has a similar effect on sample D, with $t = 8$ nm: The maximum at $\mathfrak{F} \approx 180$ T is barely detectable, and the signal-to-noise ratio in $\tilde{\rho}_{xy}$ is reduced in comparison to sample F.

The SdH oscillations reduce in magnitude with increasing temperature due to the broadening of the Fermi edge. The oscillations $\tilde{\rho}_{xy}(H, T)$ as a function of applied magnetic field for various temperatures are given in Fig. 6 for sample F. An equivalent plot for sample X2 is reported in Fig. S9 of the Supplemental Material [19].

By fitting the amplitude of a specific LL index n_{LL} as a function of T with the temperature dependence following

Eq. (1), the reduction of the oscillation amplitude over temperature can be employed to extract the cyclotron mass m_c according to

$$\tilde{\rho}_d(H = \text{const}, T) = A_T \frac{T}{\sinh\left(T \frac{N_{PM}}{H}\right)}, \quad (4)$$

with $A_T \in \mathbb{R}_+$ representing the temperature-independent terms. In Fig. 7, the amplitudes of $\tilde{\rho}_{xy}$ corresponding to specific $n_{LL} \in [29; 38]$ are given over a temperature range $T \in [1.55; 6.00]$ K. Now, using Eq. (4), the $\tilde{\rho}_{xy}$ amplitude at each n_{LL} as a function of temperature is fitted (solid lines in Fig. 7). The resulting values of the cyclotron mass $m_c^{(n_{LL}, xy)}$ as a function of n_{LL} are plotted in the inset to Fig. 7. The errors of $m_c^{(n_{LL}, xy)}$ are not statistically independent, since they originate from the same dataset and the weighted mean value is found to be $m_c^{(xy)} = (0.32 \pm 0.05)m_e$. Similarly, the amplitude values extracted from the $\tilde{\rho}_{xx}$ data of sample F yield $m_c^{(xx)} = (0.32 \pm 0.02)m_e$ and a value of $m_c^{(xy)} = (0.39 \pm 0.05)m_e$ is obtained from the $\tilde{\rho}_{xy}$ data of sample X2. The amplitude over temperature dependence and the fitting are provided in Figs. S10 and S11, respectively, in the Supplemental Material [19].

In order to obtain the quantum transport time τ , the extrema of $\tilde{\rho}_d$ as a function of applied magnetic field, where $\frac{\partial^2 \tilde{\rho}_d}{\partial H^2} = 0$, are investigated. At these points, the L-K formula simplifies considerably, and the quantum transport time τ can be expressed as

$$\tau = \left(\frac{e}{-m_c \pi} \frac{\partial}{\partial \frac{1}{H}} \ln \mathfrak{G}_d \right)^{-1}, \quad (5)$$

with $\mathfrak{G}_d \equiv \frac{\tilde{\rho}_d}{\rho_d^{(bg)}} \sinh\left(NM \frac{T}{H}\right) \frac{1}{\sqrt{H}}$ and $\rho_d^{(bg)}$ the smooth background of ρ_d . A derivation is given in Appendix C. τ can be extracted from the slope of $\ln \mathfrak{G}_d$ over $\frac{1}{H}$, leaving only m_c as an unknown parameter. Since m_c is already determined, τ can be obtained from $\tilde{\rho}_{xx}(T = 2 \text{ K})$ and is found to be $\tau = [(0.26 \pm 0.03) \times 10^{-12}]$ s. A linear fit of $\ln \mathfrak{G}_d(\frac{1}{H})$ is provided in the upper panel of Fig. 8 (Dingle plot). The Dingle temperature x is inherently linked to τ via $x = \frac{\hbar}{2\pi k_B \tau} = (4.0 \pm 0.5) \text{ K}$. The Dingle damping is obtained by considering the bare electron mass $m_b \geq m_c$, not the cyclotron mass [22], so the results are to be understood as an upper bound of x (and lower bound of τ). A direct L-K fit of $\tilde{\rho}_{xx}(T = 2 \text{ K})$ results in $x_{xx}^{(L-K)} = (4.7 \pm 0.4) \text{ K}$, corresponding to $\tau = [(0.22 \pm 0.02) \times 10^{-12}]$ s, as depicted in the lower panel of Fig. 8. A similar value of $x_{xy}^{(L-K)} = (4.3 \pm 0.3) \text{ K}$ is found based on $\tilde{\rho}_{xy}(T = 2 \text{ K})$.

Using the rotating SH, the SdH oscillations are resolved as a function of both H and of the angle θ measured between $\mathbf{H}$ and the out-of-plane axis, as depicted in Fig. 9. The θ -dependent data for sample F at 2 K is given in Fig. 10 as $\tilde{\rho}_{xy}$ over the applied magnetic field for specific values of θ . The oscillation frequency $\mathfrak{F}$, which is periodic in $\frac{1}{H}$, experiences an apparent increment when θ is increased: $\mathfrak{F} \rightarrow \frac{\mathfrak{F}}{\cos \theta}$. This conforms to a 2D SdH model, in which only the component of $\mathbf{H}$ along the out-of-plane axis contributes to the Landau quantization. While it is possible that the 2D character of the oscillations originates from an anisotropic mobility when

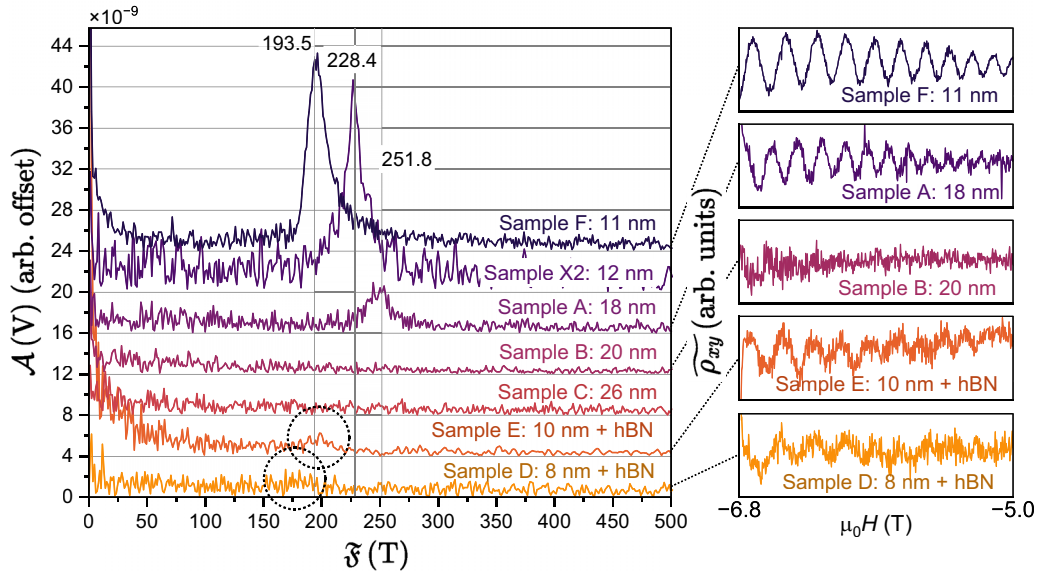


FIG. 5. Left panel: amplitude $\mathcal{A}$ resulting from the FFT spectra of $\widetilde{\rho}_{xy}$ over frequency $\mathfrak{F}$ for specific flake thicknesses. The dotted circles are a guide to the eye. Right panel: $\widetilde{\rho}_{xy}$ as a function of applied magnetic field for specific flake thicknesses.

comparing the in-plane to the out-of-plane values, geometric considerations, detailed in Sec. C of the Supplemental Material [19], show that the real-space extent of the oscillations exceeds the flake thickness. The uncorrected frequency $\mathfrak{F}^*$ is plotted in the inset to Fig. 10 and the solid line follows the theoretical:

$$\mathfrak{F}^*(\theta) = \frac{\mathfrak{F}_0}{\cos(\theta)}, \quad (6)$$

where the frequency $\mathfrak{F}_0 = \mathfrak{F}(\theta = 0)$ increases as a function of $\frac{1}{\cos(\theta)}$, while the effects from a nonspherical Fermi surface are neglected.

In order to obtain the correct values of x and $\mathfrak{F}$, the $\widetilde{\rho}_{xy}$ data is analyzed as a function of $H_{\perp} = H \cos(\theta)$. The extracted parameters x , $\mathfrak{F}$ and the extremal Fermi surface cross-sectional area $S_F = \mathfrak{F} \frac{2\pi e}{h}$ as a function of θ are given in Fig. 11. An equivalent plot for sample X2 is reported in Fig. S12 in the Supplemental Material [19].

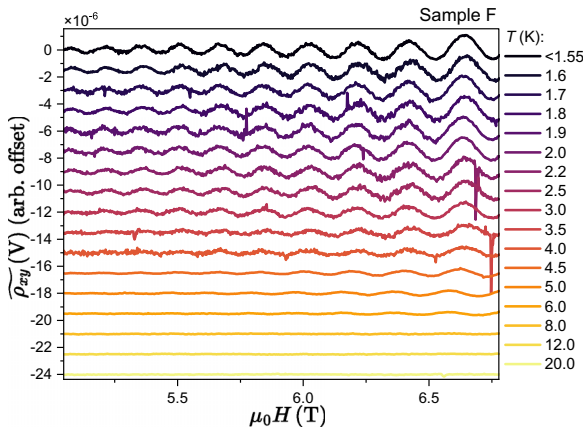


FIG. 6. Oscillations in the Hall voltage $\widetilde{\rho}_{xy}$ of sample F as a function of applied magnetic field at various temperatures.

The parameters determined previously allow for the estimation of further characteristic parameters. The carrier mobility evaluated from $\tau = \mu_q = \frac{e\tau}{m_c} = (1200 \pm 100) \frac{\text{cm}^2}{\text{Vs}}$, which is significantly higher than the Hall mobility $\mu_H = \frac{l}{w} \frac{\partial V_{xy}}{\partial (\mu_0 H)} \approx 27 \frac{\text{cm}^2}{\text{Vs}}$, with $l \approx 6 \mu\text{m}$ and $w \approx 1 \mu\text{m}$ the length and width of the flake section between the contacts over which V_{xx} and V_{xy} are measured. This is attributed to the presence of both electron and hole pockets at E_F , which diminishes the observed Hall resistance, while the underlying charge carriers exhibit (individually) a much higher mobility. Assuming a spherical Fermi surface, the Fermi wave vector k_F is estimated from the carrier density as $k_F^{(n)} = (3\pi^2 n)^{1/3} \approx 0.19 \text{ \AA}^{-1}$. Since n is derived from the Hall voltage, and assumes a single charge-carrier species, $k_F^{(n)}$ may be overesti-

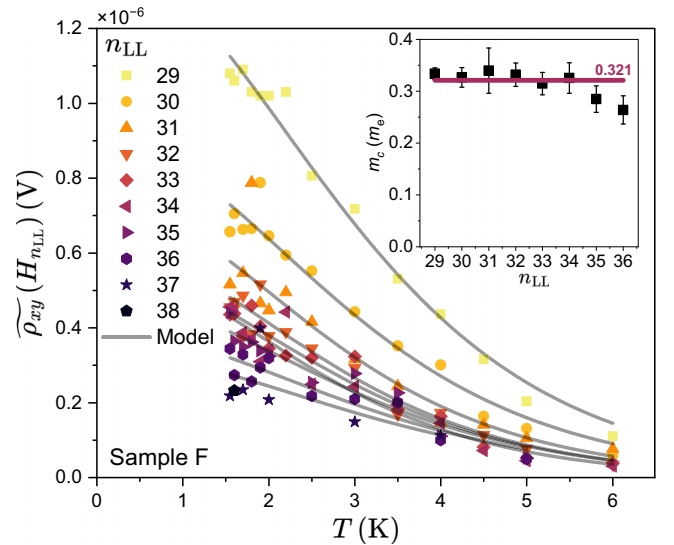


FIG. 7. Amplitudes of $\widetilde{\rho}_{xy}$ for sample F at applied magnetic fields corresponding to $n_{LL} \in [29; 38]$ over temperature and fitting with Eq. (4). Inset: m_c for each fitted n_{LL} and average m_c .

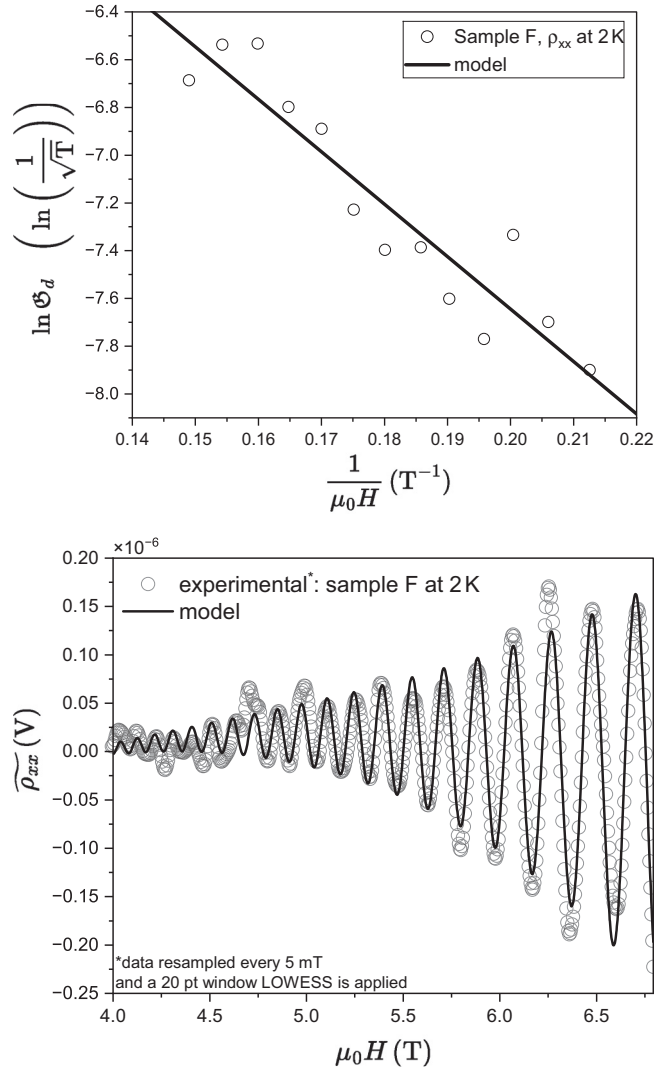


FIG. 8. Upper panel: Dingle plot of $\ln \Phi_d$ as a function of inverse magnetic field for sample F at 2 K. Solid line: linear fit. Lower panel: longitudinal resistance oscillations as a function of applied magnetic field for sample F at 2 K. Solid line: L-K fit. The obtained parameters are provided in Table II.

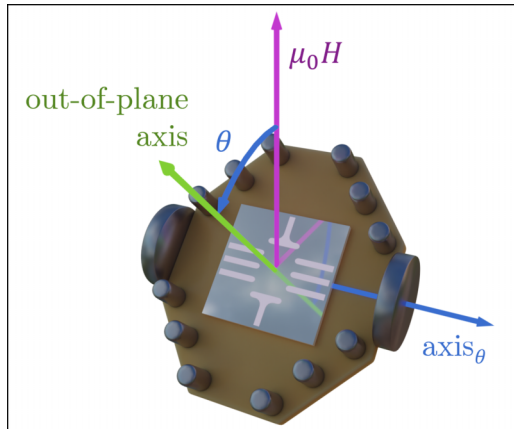


FIG. 9. Visualization of the SH after a rotation along axis_θ, the applied magnetic field encloses the angle θ with the out-of-plane axis of the sample.

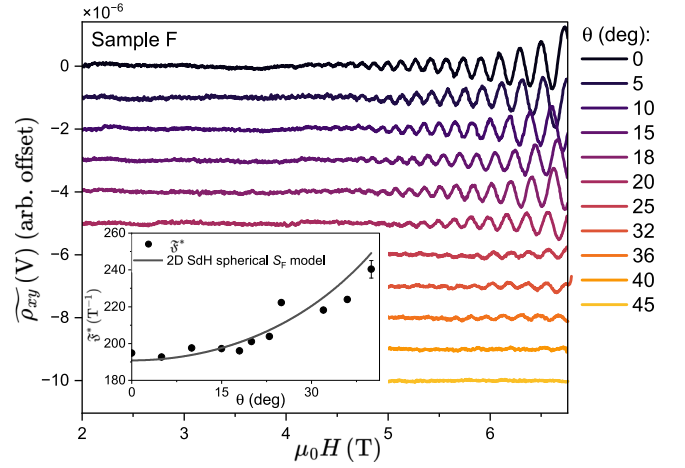


FIG. 10. Oscillations in the Hall voltage $\widetilde{\rho}_{xy}$ of sample F as a function of magnetic field, applied at various angles θ to the sample surface normal. Inset: oscillation frequency $\mathfrak{F}$ as a function of θ. Solid line: model resulting in $\mathfrak{F}(\theta = 0) = (191 \pm 1) \text{ T}^{-1}$.

Alternatively, the value found for S_F can be employed (assuming a circular Fermi surface cross section at $\theta = 0$): $k_F^{(S_F)} = (\frac{S_F}{\pi})^{1/2} \approx 0.08 \text{ \AA}^{-1}$. The mean free path length is established based on τ and m_c and yields $l_0^{(n)} = \frac{\hbar k_F^{(n)} \tau}{m_c} \approx 150 \text{ nm}$ or $l_0^{(S_F)} = \frac{\hbar k_F^{(S_F)} \tau}{m_c} \approx 80 \text{ nm}$. The parameters obtained via L-K fits of samples A and F at 2 K are summarized in Table II, and the evaluated transport parameters are tabulated in Table III.

C. Weak antilocalization

The two-terminal conductance is obtained by measuring the voltage difference directly between source and drain. Around $H = 0$, the samples show a conductance peak, reported in Fig. 12 as conductance over applied magnetic field for sample F at 2 K. Similar plots are obtained for samples X1 and X2, as given in Fig. S14 of the Supplemental Material [19]. In this two-terminal configuration, the measured conductance is affected by both the PtSe₂ film and by the (metallic) Pt contacts.

The origin of the conductance peak is assigned to the SOC of the charge carriers within the 12 nm thin Pt contacts leading to WAL: Without external magnetic field, the time-reversal

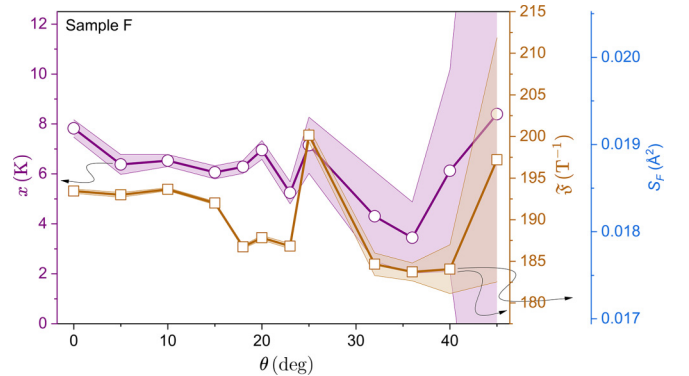


FIG. 11. Dingle temperature x (circles), oscillation frequency $\mathfrak{F}$ (squares), and extremal Fermi surface cross section S_F (also squares) of sample F as a function of θ. Shaded areas: error range.

TABLE II. Parameters obtained from L-K fits at 2 K.

Sample	Fit	x (K)	$\mathfrak{F}$ (T^{-1})	ϕ	M (m_0)
F	$\tilde{\rho}_{xx}$	3.6(0.2) ^a	192.46(0.11) ^a	0.21(0.015)	0.321 ^b
	$\tilde{\rho}_{xy}$			0.60(0.015)	
	$\tilde{\rho}_{xx}^{-1}$	4.7(0.4)	193.1(0.3)	0.19(0.04)	
	$\tilde{\sigma}_{xx}^{-1}$	3.64(0.24)	194.19(0.18)	0(0.024)	
A	$\tilde{\rho}_{xy}$	7(2.4)	254.9(1.7)	-0.5(0.2)	

^aThe values of x and $\mathfrak{F}$ are constrained to be equivalent for $\tilde{\rho}_{xx}$ and $\tilde{\rho}_{xy}$.

^bSince x and M are highly dependent, the value M is fixed to 0.321 m_0 , which is obtained from the temperature dependence of the oscillation amplitude of sample F.

symmetry (TRS) is preserved and the SOC triggers a destructive interference between the backscattering paths with their time-reversed counterparts. The applied magnetic field breaks the TRS, reducing the destructive backscattering interference, thus leading to a decrease in conductance.

In the four-terminal conductance, sample X1 shows WAL, depicted in Fig. 13 as conductance over an applied magnetic field at 1.7 K. The WAL remains resolvable until $T \approx 10$ K (Fig. S15 in the Supplemental Material [19]).

It is proposed that this four-terminal WAL originates from the SOC in the PtSe₂: The Pt vacancies in PtSe₂ contribute uncompensated magnetic moments that manifest via a Kondo effect [14]. Furthermore, it is inferred that the magnetic moments also suppress the SOC in PtSe₂. The vanishing of the Kondo effect in sample X1, correlated with the emergence of four-terminal WAL, suggests that the Pt vacancy concentration varies within the bulk crystal. The Kondo effect is discussed in Appendix A, and the resistance-over-temperature-curve (R - T) data of samples X1 and X2 are compared: The Kondo effect and the WAL are observed to be mutually exclusive, as one originates from the presence of Pt vacancies, while the other is inhibited by them.

The WAL data is understood within the (2D) Hikami-Larkin-Nagaoka (HLN) model [25,26]:

$$\Delta\sigma_{\text{WAL}}^{(2D)} \propto -\frac{\alpha e^2}{\pi h} \left[\ln\left(\frac{B_\phi}{H}\right) - \Psi\left(\frac{1}{2} + \frac{B_\phi}{H}\right) \right], \quad (7)$$

TABLE III. Parameters of the charge carriers obtained by combining the analysis of the SdH oscillations and V_H .

		Evaluated via
μ_q	$(1200 \pm 100) \frac{\text{cm}^2}{\text{Vs}}$	τ
μ_H	$27 \frac{\text{cm}^2}{\text{Vs}}$	V_{xx} and V_{xy}
n	$(2 \times 10^{20}) \text{cm}^{-3}$	Hall voltage
S_F	$0.02 \text{ \AA}^{-2}$	$\mathfrak{F}_0$ from $\mathfrak{F}(\theta)$
$k_F^{(S_F)}$	$0.08 \text{ \AA}^{-1}$	S_F
$k_F^{(n)}$	$0.19 \text{ \AA}^{-1}$	n
m_c	$(0.32 \pm 0.02)m_e$	Eq. (4)
τ	$[(0.22 \pm 0.02) \times 10^{-12}] \text{ s}$	Eq. (5)
$l_0^{(n)}$	150 nm	$k_F^{(n)}$
$l_0^{(S_F)}$	80 nm	$k_F^{(S_F)}$

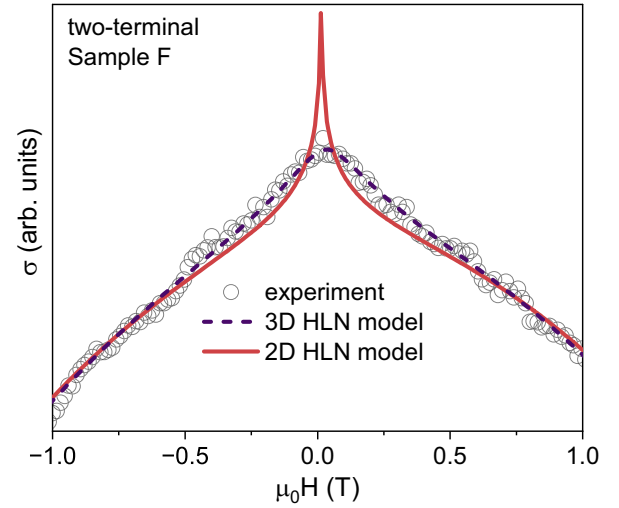


FIG. 12. Two-terminal conductance over applied magnetic field of sample F at 2 K. Dashed line 3D HLN model. Solid line 2D HLN model. The resulting parameters are given in Table S3 of the Supplemental Material [19].

where the magnetic field length is $B_\phi = \frac{h}{8\pi\hbar l_\phi}$ (l_ϕ is the phase coherence length), Ψ is the digamma function, and $\alpha \in [-\frac{3}{2}, -\frac{1}{2}]$ is a coefficient. Alternatively, a three-dimensional (3D) extension of the HLN model can be employed as in Refs. [27,28], where it is also applied to model a Dirac system:

$$\Delta\sigma_{\text{WAL}}^{(3D)} = A_{\text{WAL}} \sqrt{\frac{H}{f}} \left[2\zeta\left(\frac{1}{2}, \frac{1}{2} + \frac{f}{Hl_0^2}\right) + \zeta\left(\frac{1}{2}, \frac{1}{2} + \frac{f}{Hl_\phi^2}\right) - 3\zeta\left(\frac{1}{2}, \frac{1}{2} + 4\frac{f}{Hl_{\text{SO}}^2} + \frac{f}{Hl_\phi^2}\right) \right], \quad (8)$$

where A_{WAL} is the magnitude of the conductance peak, ζ the Hurwitz zeta function, and $f = \frac{\hbar}{4e} = \frac{\Phi_0}{8\pi}$ a constant. There are four length scales involved: (i) The mean free path l_0 ,

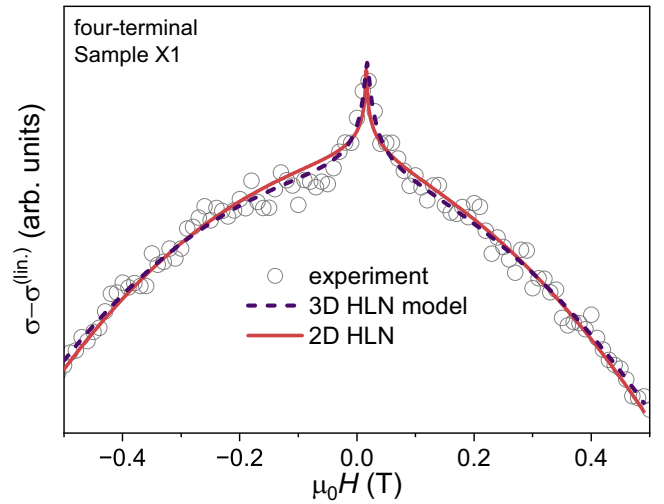


FIG. 13. Four-terminal conductance of sample X1 at 1.7 K (Hall intermix $\sigma^{(\text{lin.})}$ corrected). Comparison of the 3D HLN model (dashed line) with the 2D HLN model (solid line). The obtained parameters are provided in Table S4 of the Supplemental Material [19].

(ii) the phase coherence length l_ϕ , (iii) the spin-orbit scattering length l_{SO} , and (iv) the magnetic length $l_H = \sqrt{\frac{\hbar}{eB}}$. To fit the WAL in a region $|\mu_0 H| < 2$ T, expressions for the orbital magnetoconductance and the Hall intermix are added. The L-K term modeling the SdH oscillations is omitted, given that its magnitude is negligible at $|\mu_0 H| < 2$ T. The total conductance as a function of applied magnetic field is modeled as the WAL conductance taking place in parallel (additive) to the orbital magnetoconductance:

$$\sigma(H) = \Delta\sigma_{\text{WAL}}^{(i)} + \left(\frac{1}{\sigma_0} + A_{\text{MR}} |(H + H_{\text{lag}})|^s + kH \right)^{-1}, \quad (9)$$

with σ_0 the zero-field conductance, A_{MR} the orbital magnetoresistance (OMR) magnitude, and s the scaling exponent thereof. Since the highest considered temperature of 15 K still lies in the range in which the system is in the Fermi liquid regime (≈ 80 K), $s = 2$ is chosen. The parameter H_{lag} compensates for the lag between the nominal magnetic field and the applied magnetic field, which is about ± 16 mT when sweeping at $8 \frac{\text{mT}}{\text{s}}$, while k models the already mentioned Hall intermix. Details about the evaluation of Eq. (9) and implementation of the least-squares fit are provided in Sec. D in the Supplemental Material [19].

Both the 2D model and the 3D model are applied to the two-terminal conductivity of sample F at 2 K (Fig. 12). The 3D model captures the peak profile more precisely. The obtained length scales are compared in Table S2 of the Supplemental Material [19]. The data is comparable to the WAL exhibited by Pt thin films reported in Ref. [29].

The 2D and the 3D model are also applied to the four-terminal WAL observed in sample X1 at 1.7 K, depicted in Fig. 13 as conductance over applied magnetic field. The conductance peak shape is narrower in H than the one recorded in the two-terminal measurement. The difference between the models is not as stark as for the two-terminal conductance peak. This suggests that the four-terminal WAL is a distinct effect from the two-terminal WAL. The 3D HLN fit yields $l_0 = (350 \pm 160)$ nm and $l_\phi^{3D} = (350 \pm 120)$ nm.

IV. CONCLUSIONS

Here, key aspects of the charge carriers in PtSe₂ are extracted via the implementation of an analytic L-K framework. Under application of a magnetic field, SdH oscillations, observed in the resistance measurements, occur in plane. This provides key information about the charge carriers, including the determination of the Berry phase $\Phi_B = (1.01 \pm 0.05)\pi$, the cyclotron mass $m_c = (0.321 \pm 0.008)m_0$, in-plane mobility $\mu = (1200 \pm 100) \frac{\text{cm}^2}{\text{Vs}}$, and quantum scattering time $\tau = [(0.22 \pm 0.02) \times 10^{-12}]$ s. A spectral analysis of the oscillations yields a single species of electrons as the prevalent charge carriers, with $\mathfrak{F} = (194.19 \pm 0.18) \text{T}^{-1}$, equating to a Fermi surface cross section $S_F \approx 0.0185 \text{\AA}^{-2}$ for $t = 11$ nm. $\mathfrak{F}$ is reduced as the thickness is lowered, suggesting a thickness dependence of the band structure for $8 \text{ nm} < t < 20$ nm and potentially presenting a compelling tuning parameter for the precise electronic properties of PtSe₂ flakes and thin films. The Pt vacancies, inherently present in the grown bulk

crystals, are found to play a crucial role, as they provide uncompensated magnetic moments that establish a Kondo effect, but suppress the SOC and consequently the WAL. While the two-terminal WAL is better understood with the 3D HLN model and originates from the Pt contacts, the four-terminal conductance peak also fits the 2D HLN model, consistent with the 2D confinement observed from the SdH oscillations. It is concluded that the four-terminal WAL heralds substantial inherent SOC in PtSe₂. While it is plausible that the SOC, and following that, the Rashba effect leads to the observed nonzero Berry phase values, the measurements do not provide a conclusive picture of how the bulk Dirac cone influences the charge-carrier properties at the Fermi level. The mechanism by which the nontrivial Berry phase emerges remains to be clarified. The employed bulk crystals provide a long-range order larger than the physical measurement area and contain Pt vacancies from growth, prompting a comparative investigation to directly grown PtSe₂ structures. The adaptation of the L-K formalism is valid for a single charge-carrier species with 2D oscillations. The results motivate magnetotransport studies in highly stoichiometric PtSe₂ flakes and PtSe₂/ferromagnetic heterostructures, in order to fully harness the SOC inherent in the material and demonstrate the significance of PtSe₂ for orbitronic and orbital Hall-based prospective devices.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

APPENDIX A: EXPOSITION OF KONDO EFFECT

Although both samples X1 and X2 are exfoliated from bulk crystal Batch 2, their $R - T$ behavior is qualitatively different: The four-terminal resistance over temperature curves [field-warming (FW) and zero-field-warming (ZFW)] of sample X1 are given in Fig. 14 and the field-cooling (FC) and zero-field-cooling curves of sample X2 are provided in Fig. 15. The expression (θ, ψ) describes the orientation of the SH in relation to the applied magnetic field: ψ is the angle by which the SH is rotated with respect to axis ψ , which is normal to $\mathbf{H}$; θ is the angle of orientation about the axis θ , which is normal to axis ψ and encloses an angle $(\pi/2 + \psi)$ with $\mathbf{H}$. The angles and their respective axes are sketched in the right panel of Fig. 14. The curves for sample X1 are distinguished mainly by a temperature-independent offset, which is caused by the OMR. The OMR is strongest when the applied magnetic field is oriented along the sample surface normal, corresponding to an orientation (0,0). In the (90,0) orientation, the applied

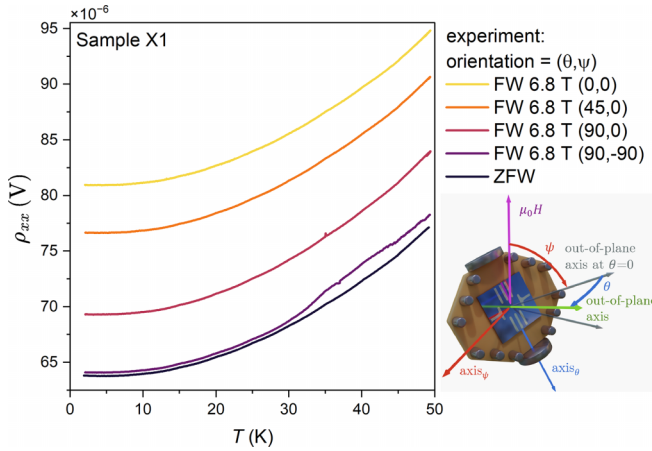


FIG. 14. Left panel: four-terminal resistance over temperature of sample X1 (bulk crystal Batch 2). Right panel: sketch of how the angles θ and ψ are related to the SH orientation.

magnetic field lies in plane and normal to the source-drain direction. The resulting OMR is reduced due to the charge carriers being confined to the sample thickness of 17 nm. Considering the structural anisotropy of PtSe₂, the out-of-plane mobility can be assumed to be lower than the in-plane value, which can also account for the reduced in-plane OMR. In the (90, -90) orientation, the applied magnetic field lies in plane and parallel to the source-drain direction. Almost no OMR is observed, since the Lorentz force is zero. The origin of the additional resistance for $T \gtrsim 35$ K still is unclear. The ZFW shows the lowest resistance because no OMR applies. As $T \rightarrow 0$, no increase in the resistance R is observed. This is in contrast to all other considered samples, which display a global minimum resistance temperature of ≈ 15 K and an increase in resistance as the temperature is lowered below $T \lesssim 15$ K. This property of samples A, B, and C is discussed in a previous work [14] and can be gleaned in Fig. 15 for

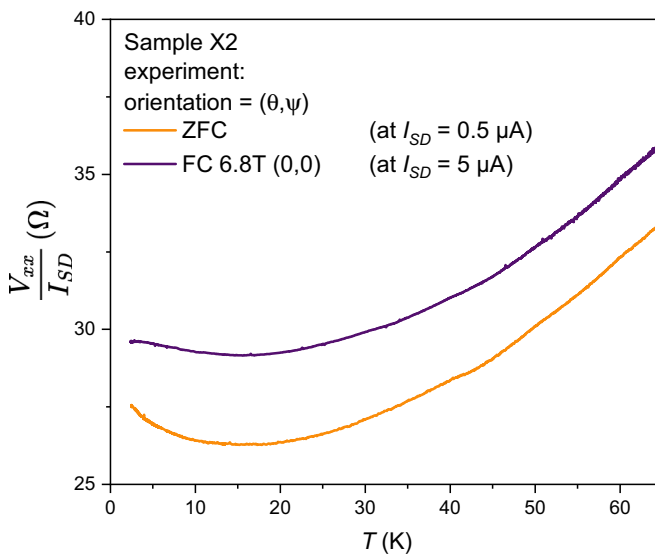


FIG. 15. Four-terminal resistance over temperature of sample X2 (bulk crystal Batch 2), normalized by the applied source-drain current.

sample X2. The anomalous increase in resistance as $T \rightarrow 0$ is mediated by Pt vacancies, which contribute an uncompensated spin when located in the top- or bottommost layers. At or below the Kondo temperature T_K , this results in a Kondo effect: The charge carriers scatter at the vacancy spin via a double antiferromagnetic spin exchange, resulting in an increase in resistance as the temperature is lowered further.

APPENDIX B: ADAPTATION OF THE LIFSHITZ-KOSEVICH FORMULA

The oscillatory part of the thermodynamic potential of the electrons due to quantum oscillations $\tilde{\Omega}$ is given by [21,22]

$$\begin{aligned} \tilde{\Omega} = & \left(\frac{e}{2\pi c \hbar} \right)^{3/2} \frac{2kTH^{3/2}V}{(S_F'')^{1/2}} \\ & \times \sum_{p=1}^{\infty} \frac{\exp[-2\pi^2 p k x / (\beta H)] \cos(\frac{1}{2} p \pi g \frac{m}{m_0})}{p^{3/2} \sinh[2\pi^2 p k T / (\beta H)]} \\ & \times \cos \left[2\pi p \left(\frac{\mathfrak{F}}{H} - \frac{1}{2} \right) \pm \frac{\pi}{4} \right] \end{aligned} \quad (B1)$$

$$\propto H^{3/2} \sum_{p=1}^{\infty} R_D R_T R_S \mathfrak{D} \quad (B2)$$

with c the speed of light, $\hbar$ the reduced Planck quantum, k the Boltzmann constant, V the sample volume, p the harmonic number, x the Dingle temperature, $\beta = \frac{e\hbar}{m}$, m the electron cyclotron mass, g the Landé g factor, m_0 the electron rest mass, and $\mathfrak{F}$ the oscillation frequency. The extremal cross-sectional area of the Fermi surface perpendicular to $\mathbf{H}$ is S_F and κ is the component of $\mathbf{k}$ parallel to $\mathbf{H}$. The parameter S_F'' describes the curvature of S_F with respect to κ :

$$S_F'' = \frac{d^2 S_F}{d\kappa^2}, \quad \kappa = \frac{\mathbf{k} \cdot \mathbf{H}}{|\mathbf{H}|}. \quad (B3)$$

When considering the conductivity oscillations $\tilde{\sigma}$ instead of $\tilde{\Omega}$, the same expressions for the Dingle damping $R_D = \exp[-2\pi^2 p k x / (\beta H)]$, the thermal smearing $R_T = T \sinh[2\pi^2 p k T / (\beta H)]^{-1}$, and the spin phase interference $R_S = \cos(\frac{1}{2} p \pi g \frac{m}{m_0})$ are applicable. The conductivity does, however, scale differently under application of a magnetic field: $\tilde{\sigma}$ is related to the variation of the density of states at the Fermi energy $\tilde{\mathcal{D}}$ [22]:

$$\frac{5}{2} \frac{\tilde{\mathcal{D}}}{\mathcal{D}_0} + \frac{3}{2} \left(\frac{\tilde{\mathcal{D}}}{\mathcal{D}_0} \right)^2, \quad (B4)$$

where $\mathcal{D}_0$ is the steady density of states at the Fermi energy (without applied magnetic field). The ratio $\frac{\tilde{\mathcal{D}}}{\mathcal{D}_0}$ relates to the applied magnetic field as

$$\frac{\tilde{\mathcal{D}}}{\mathcal{D}_0} = \left(\frac{H}{2\mathfrak{F}} \right)^{1/2}, \quad (B5)$$

$$\frac{\tilde{\sigma}}{\sigma} \propto \left(\frac{5}{2} \left(\frac{H}{\mathfrak{F}} \right)^{1/2} + \frac{3}{2} \left(\frac{H}{2\mathfrak{F}} \right) \right). \quad (B6)$$

The higher harmonic numbers $p > 1$ are omitted since they are not observed in the considered samples (a single peak

is obtained in the FFT). The expression for the phase of the oscillations in $\cos[2\pi(\frac{\mathfrak{F}}{H} - \frac{1}{2}) \pm \frac{\pi}{4}]$ is obtained for a 3D case, wherein the offset $\delta = \frac{\pi}{4}$ results from an integration over κ [22]. This offset is zero in the 2D case. The phase offset $\phi = \frac{1}{2}$ is expanded to also consider a potentially nonzero Berry curvature Φ_B [17]:

$$2\pi\left(\frac{\mathfrak{F}}{H} - \frac{1}{2}\right) \pm \frac{\pi}{4} \equiv 2\pi\left(\frac{\mathfrak{F}}{H} - \phi\right), \quad (\text{B7})$$

$$\phi = \left(\frac{1}{2} - \frac{\Phi_B}{2\pi} \pm \delta\right) \bmod 1, \quad \delta^{(2D)} = 0. \quad (\text{B8})$$

$$\iff \Phi_B = 2\pi\left(\frac{1}{2} - \phi\right) \bmod 2\pi. \quad (\text{B9})$$

Thus, for $\phi = \frac{1}{2}$ the Berry phase is zero, while for $\phi = 0$ the Berry phase is $\pm\pi$. The expression for the oscillation in the 2D case is $\cos[2\pi(\frac{\mathfrak{F}}{H} + \phi)]$. Equation (B2) is adapted to describe the oscillatory part of ρ_d by keeping the behavior $\propto R_D R_T R_S \mathfrak{D}^{(2D)}$, while also considering that the origin of $\tilde{\sigma}$ (and hence $\tilde{\rho}_d$) lies in the modulation of $\mathcal{D}$:

$$\tilde{\rho}_d = AR_D R_T R_S \mathfrak{D} \left(\frac{5}{2} \frac{\tilde{\mathcal{D}}}{\mathcal{D}_0} + \frac{3}{2} \left(\frac{\tilde{\mathcal{D}}}{\mathcal{D}_0} \right)^2 \right), \quad (\text{B10})$$

$$\begin{aligned} \tilde{\rho}_d &\stackrel{\text{L}}{=} AT \left(\frac{5}{2} \left(\frac{H}{2\mathfrak{F}} \right)^{\frac{1}{2}} + \frac{3}{2} \left(\frac{H}{2\mathfrak{F}} \right) \right) \frac{\exp(-x \frac{NM}{H}) \cos(\pi M)}{\sinh(T \frac{NM}{H})} \\ &\times \cos\left(2\pi\left(\frac{\mathfrak{F}}{H} + \phi\right)\right). \end{aligned} \quad (\text{B11})$$

To account for the four-terminal transport measurements, the amplitude A is introduced. Several constants are included into A . The parameter $M = \frac{m_c}{m_0}$ is the fermion cyclotron mass ratio. The value of $N = 2\pi^2 \frac{k m_0}{\hbar e} \approx 14.693 \frac{\text{K}}{\text{T}}$ is determined by substituting the values for the constants.

APPENDIX C: EXTRACTION OF THE QUANTUM TRANSPORT TIME FROM THE LIFSHITZ-KOSEVICH FORMULA

The quantum transport time τ is related to the Dingle temperature via $x = \frac{\hbar}{2\pi k\tau}$, and is extracted from Eq. (B11): At a local extrema $\frac{\partial^2 \tilde{\rho}_d(H)}{\partial H^2} = 0$, the phase factor $|\mathfrak{D}^{(2D)}| = 1$. A relative oscillation magnitude $\frac{\tilde{\rho}_d}{\rho_d^{(\text{bg.})}}$ is introduced to obtain a

dimensionless expression (the value of A changes accordingly). Let $\mathcal{H} = \frac{5}{2}(\frac{H}{2\mathfrak{F}})^{1/2} + \frac{3}{2}(\frac{H}{2\mathfrak{F}})$ be the magnetic field dependence of the oscillation amplitude. This yields

$$\frac{\tilde{\rho}_d}{\rho_d^{(\text{bg.})}} = AT \mathcal{H} \frac{\exp(-x \frac{NM}{H}) \cos(\pi M)}{\sinh(T \frac{NM}{H})}, \quad (\text{C1})$$

which is solved for x :

$$\begin{aligned} &\iff \frac{\tilde{\rho}_d}{\rho_d^{(\text{bg.})}} \sinh\left(T \frac{NM}{H}\right) \mathcal{H}^{-1} \frac{1}{AT \cos(\pi M)} \\ &\propto \exp\left(-x \frac{NM}{H}\right), \end{aligned} \quad (\text{C2})$$

$$\begin{aligned} &\iff \ln\left(\frac{\tilde{\rho}_d}{\rho_d^{(\text{bg.})}} \sinh\left(T \frac{NM}{H}\right) \mathcal{H}^{-1}\right) - \ln[AT \cos(\pi M)] \\ &\propto -x \frac{NM}{H}, \end{aligned} \quad (\text{C3})$$

and taking the derivative in $\frac{1}{H}$ gives

$$\frac{\partial}{\partial \frac{1}{H}} \ln\left(\frac{\tilde{\rho}_d}{\rho_d^{(\text{bg.})}} \sinh\left(T \frac{NM}{H}\right) \mathcal{H}^{-1}\right) = -xNM. \quad (\text{C4})$$

Now putting $x = \frac{\hbar}{2\pi k\tau}$ and resubstituting $N = 2\pi^2 \frac{k m_0}{\hbar e}$ the right-hand term becomes

$$xNM = \frac{\pi m_c}{e\tau}. \quad (\text{C5})$$

On the left-hand side of Eq. (C4), the expression inside the logarithm can be summarized as

$$\mathfrak{G}_d \equiv \frac{\tilde{\rho}_d}{\rho_d^{(\text{bg.})}} \sinh\left(NM \frac{T}{H}\right) \mathcal{H}^{-1}. \quad (\text{C6})$$

The expression for τ becomes

$$\tau = \left(\frac{e}{-m_c \pi} \frac{\partial}{\partial \frac{1}{H}} \ln \mathfrak{G}_d \right)^{-1}. \quad (\text{C7})$$

The second term in $\mathcal{H}$ can be neglected for $\mathfrak{F} \gg H \Rightarrow (\frac{H}{2\mathfrak{F}})^{1/2} \gg (\frac{H}{2\mathfrak{F}})$, yielding a simplified expression for $\mathfrak{G}_d$:

$$\mathfrak{G}_d \equiv \frac{\tilde{\rho}_d}{\rho_d^{(\text{bg.})}} \sinh\left(NM \frac{T}{H}\right) \frac{1}{\sqrt{H}}. \quad (\text{C8})$$

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