High quality transport in exfoliated jacutingaite crystals

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Monolayer jacutingaite (Pt$_2$HgSe$_3$) has been recently identified as a candidate quantum spin Hall system with a 0.5 eV band gap, but no transport measurements have been performed so far on this material, neither in monolayer nor in the bulk. By using a dedicated high-pressure technique, we grow crystals enabling the exfoliation of 50-100 nm thick layers and the realization of devices for controlled transport experiments. Magnetoresistance measurements indicate that jacutingaite is a semimetal, exhibiting Shubnikov-de Haas (SdH) resistance oscillations with a multi-frequency spectrum. We adapt the Lifshitz-Kosevich formula to analyze quantitatively the SdH resistance oscillations in the presence of multiple frequencies, and find that the experimental observations are overall reproduced well by band structure ab-initio calculations for bulk jacutingaite. Together with the relatively high electron mobility extracted from the experiments (∼2000 cm$^2$/Vs, comparable to what is observed in WTe$_2$ crystals of the same thickness), our results indicate that monolayer jacutingaite –once successfully exfoliated– will likely provide an excellent platform to investigate transport in 2D quantum spin Hall systems.

Two-dimensional (2D) quantum spin Hall insulators (QSHIs) constitute an important class of topological systems having a gapped insulating bulk and gap-less helical edge states that mediate transport at low energy. According to theory, these helical states are protected against elastic backscattering by time-reversal symmetry, so that edge conduction is predicted to occur in the ballistic regime over distances shorter than the inelastic scattering length. In reality, the material systems that have been identified experimentally as 2D QSHI –such as HgTe/CdTe and InAs/GaSb quantum wells– do not exhibit the expected, precisely quantized conductance: values close to $e^2/h$ per edge have been reported, with rather large sample-to-sample variations, as well as fluctuations as a function of applied gate voltage. Identifying the physical origin of these deviations is proving difficult, although several mechanisms have been put forward. To progress, it is important to investigate experimentally a broader variety of 2D QSHI systems, in order to understand the influence of different microscopic parameters.

2D van der Waals (vdW) layered materials offer new opportunities, as illustrated by WTe$_2$ monolayers, whose predicted 2D QSHI nature has been confirmed by different experiments. Two-terminal edge conductance close to $e^2/h$ has been observed in hBN-encapsulated WTe$_2$ monolayers, with subsequent work reporting a more precise quantization (albeit only for contact separation shorter than 100 nm). Edge states have been directly detected by means of scanning tunneling microscopy and other types of local probes, and even the bands of WTe$_2$ monolayers could be mapped by means of angle-resolved photo-emission experiments (ARPES). The 60-70 meV band-gap –significantly larger than in previous 2D QSHI systems– and the band structure were found to be in agreement with ab-initio calculations. The improved conductance quantization, the larger band-gap, as well as the possibility to access directly the material with different experimental probes make WTe$_2$ an excellent candidate to study the physics of 2D QSHIs. These results also showcase the potential of 2D materials, and motivate the search for other atomically thin crystals, providing even better platforms for experimental work.

Among these, jacutingaite –a vdW layered material with chemical formula Pt$_2$HgSe$_3$– has been proposed to be of particular interest. In its bulk form (see the crystalline structure in Fig. 1a), jacutingaite is predicted to be a compensated semimetal hosting multiple electron and hole pockets that arise from nodal lines along which the valence (blue) and conduction (red) bands are degenerate in the absence of spin-orbit interaction (SOI) (see e.g. the KH line in Fig. 1b). SOI is, however, not negligible owing to the presence of heavy elements and lifts the degeneracy, endowing the system with a non-trivial topology protected by crystalline symmetries. The correspondingly expected surface states have been recently observed in ARPES measurements, and found to be in remarkable good agreement with theory. When the material thickness is reduced to that of an individual monolayer, jacutingaite is predicted to become a 2D QSHI realizing the original Kane-Mele model, with a search of magnitude larger than in WTe$_2$. Calculations also suggest that jacutingaite monolayers can be switched between a topological and a trivial insulating state, by applying electric fields, as achievable in 2D vdW heterostructures. If confirmed, all these predictions make monolayer jacutingaite an ideal platform to explore and possibly control 2D QSHI systems.
Despite a great interest in jacutingaite\textsuperscript{21,26–31}, no transport measurements have been performed so far, neither on atomically thin layers nor on bulk, largely because growing high-quality crystals of sufficiently large size is challenging\textsuperscript{32}. Here, we solve this problem by employing a high-pressure crystal synthesis technique enabling the growth of millimeter-sized crystals, which we exfoliate into thin layers (typically 50-to-100 nm thick) that enable the fabrication of multi-terminal devices for controlled transport experiments. The measured magnetoresistance indicates the presence of both electrons and holes, and exhibits pronounced Shubnikov-de Haas (SdH) oscillations with multiple frequencies. We discuss in detail how the Lifshitz-Kosevich (LK) formula can be adapted to the case of jacutingaite, to extract quantitative information. We find that the SdH oscillations are due to electrons, and that the observed frequencies originate from distinct pockets in the conduction band in overall good agreement with first-principles calculations. These results demonstrate that devices based on exfoliated layers can be produced that are as good as WTe\textsubscript{2} crystals of the same thickness\textsuperscript{33,34}, a very promising result for the investigation of 2D QSHI once monolayers of jacutingaite are successfully isolated.

The small size ($\leq 50\mu m$) of existing jacutingaite crystals – extracted either from aggregates of platinum based mined minerals\textsuperscript{32}, or from polycrystalline powders grown in the laboratory\textsuperscript{35} – has so far prevented the exfoliation of thin layers to realize multi-terminal devices for controlled transport experiments. To grow larger high-quality crystals we have employed a high-pressure synthesis process that allows the growth of suitable materials in a few hours (see Supporting Information, SI, for details). The process results in a polycrystalline boule a few millimeters in size, containing jacutingaite single crystals (see Fig. 1c) with dimensions up to $\sim 0.6 \times 1 \text{mm}^2$, together with crystals of a spurious phase. Indeed, X-ray powder diffraction (XRD) shows that besides Pt\textsubscript{2}HgSe\textsubscript{3} crystals, in the as-grown boule PtSe\textsubscript{2} crystals are also present in approximately 6-7 \% volume fraction, as re-
FIG. 2. (a) Top: Optical microscope image of one of our devices (the scale bar is 2 µm). Bottom: Temperature dependence of the four-probe resistivity measured on device D3. (b) Longitudinal resistivity ($\rho_{xx}$) as a function of magnetic field $B$ measured on device D1, D2, D3 at $T = 250\,\text{mK}$, and (c) corresponding MR $\eta \equiv (\rho_{xx}(B) - \rho_{xx}(0))/\rho_{xx}(0)$. (d) Transverse resistivity ($\rho_{xy}(B)$) for the three devices investigated at 250mK, showing a negative slope indicative of electron-type transport. Inset: zoom-in on the high-field transverse magnetoresistance of device D1, showing the presence of SdH oscillations (e) Taking the derivative of the longitudinal resistivity $\rho_{xx}(B)$ puts in evidence the SdH oscillations starting from 5-7 T, depending on the device. In all panels, data plotted with black, red and blue curves were measured respectively on device D1, D2, and D3.

We measured three different devices (labeled as D1, D2 and D3), based on layers that are respectively 90, 50 and 90 nm thick, all exhibiting similar behavior. The temperature dependence of the longitudinal resistivity $\rho_{xx}$ of device D3, representative of that of all devices, is shown in Fig. 2a (bottom panel). As expected for a semimetal, metallic behavior is observed, with $\rho_{xx}$ decreasing linearly from 6 $\mu\Omega\cdot\text{cm}$ at room temperature to 1.5 $\mu\Omega\cdot\text{cm}$ at 15 K, below which saturation occurs. Finding that $\rho_{xx}$ decreases upon lowering temperature indicates that down to $\sim 15$ K transport is limited by inelastic scattering and not by collisions with impurities, providing a first indication of crystal quality. The resistivity at base temperature exhibits somewhat different values in the three measured devices—respectively $\sim 5$, 8 and 1.5 $\mu\Omega\cdot\text{cm}$ in D1, D2, and D3 (see $B = 0$ values...
in Fig. 2b), indicating relatively large variations in the electronic properties of nominally identical exfoliated jacutingaite crystals, whose origin is discussed below.

The application of a perpendicular magnetic field up to 14 T, the highest field reached in our experiments (see Fig. 2b), causes the resistivity to increase. The corresponding magneto-resistance (MR; \( \eta \equiv (\rho_{xx}(B) - \rho_{xx}(0))/\rho_{xx}(0) \)), shown in figure Fig. 2c, reaches values ranging from 80 \% to 140\%, depending on the device. Notably, at high magnetic fields the MR deviates from the quadratic behavior commonly expected from classical dynamics of simple conductors, as \( \eta \) depends linearly on \( B \). A linear MR has been reported previously in systems with topological properties and/or strong SOI, but the precise mechanism at work in jacutingaite remains to be determined.

The magnetic field dependence of the transverse resistivity \( \rho_{xy} \) is shown in Fig. 2d. With the exception of device D3 (that exhibits a small, but clear, non-linearity), the \( \rho_{xy}(B) \) curve is essentially linear, with a negative slope indicative of a dominating electron contribution to transport. If interpreted in terms of a single type of charge carriers (using the relation \( \rho_{xy}(B) = R_H B \), with \( R_H = -1/n \cdot \ell \) t is the layer thickness) we find a large spread in the density values extracted from the different devices: \( n = 6.38 \cdot 10^{20} \text{ cm}^{-3} \), 1.04 \( \cdot 10^{21} \text{ cm}^{-3} \) and 1.81 \( \cdot 10^{21} \text{ cm}^{-3} \) for D1, D2 and D3. Such a large spread (as well as the fact that these values are much larger than what is expected from ab-initio calculations, \( n = 2 \cdot 10^{20} \text{ cm}^{-3} \)) cannot be attributed to unintentional doping due to impurities, since this explanation would require impurity concentrations reaching up to \( \pm 5 \% \). Such a concentration would not only have been detected in the EDS characterization of the material (see above), but would also cause a very short electronic mean free path, incompatible with the presence of well-defined SdH conductance oscillations discussed below.

Finding inconsistencies when attempting to interpret the Hall data in terms of a single type of charge carriers is not surprising, because jacutingaite is predicted to be a compensated semimetal with an equal density of carriers. The presence of multiple pockets for both types of charge carriers, each with a possibly different mobility value.

More information can be gained by analyzing the SdH conductance oscillations present in all our devices, visible in both the \( \rho_{xx}(B) \) and \( \rho_{xy}(B) \) curves (see the inset of Fig. 2d). To facilitate their analysis we look at the derivative of \( \rho_{xx} \) and \( \rho_{xy} \), shown in Fig. 2e and Fig. 3a. An estimate of the charge carrier mobility is obtained from the magnetic field \( B_{\text{onset}} \) at which the oscillations start, using the criterion \( \mu B_{\text{onset}} \sim 1 \); \( B_{\text{onset}} \) \( \sim 5 - 7 \) T depending on the specific device implies mobility values in the range \( \mu_e \sim 1400 - 2000 \text{ cm}^2/V \cdot \text{s} \). The values correspond to the electron mobility, as the sign of the Hall slope indicates that it is the electrons that give the dominant contribution to transport.

When plotted against \( 1/B \) the oscillations exhibit an irregular envelope (Fig. 3a), originating from the presence of multiple frequencies. Indeed, three peaks are present in the amplitude of the Fourier transform (FT), shown in Fig. 3b, centered at frequencies \( F_1 = 73 \text{ T} \), \( F_2 = 118 \text{ T} \) and \( F_3 = 163 \text{ T} \). The frequencies coincide in the three different devices (albeit the amplitude of the peak at \( F_2 \) nearly vanishes in device D2), indicating that the areas of the extremal Fermi surfaces responsible for the SdH oscillations are the same in all cases. It directly follows that –within the material band structure– the Fermi level is located at the same energy in the different devices, and therefore that also the charge carrier density is the same (confirming our interpretation of the Hall data, namely that the sample-to-sample dependence of the Hall slope is due to variations in the \( \mu_h/\mu_e \) ratio).

The evolution of the SdH oscillations upon increasing temperature (see Fig. 3c for data measured on device D1) is of particular interest as it allows the cyclotron mass to be determined. The analysis of the data relies on the Lifshitz-Kosevich expression for the \( T \) and \( B \) dependence of the oscillation amplitude \( \Delta \rho/\rho \): \( \Delta \rho/\rho \propto \sqrt{ \frac{\hbar e B}{m_e E_F} } \frac{X}{\sinh X} \exp\left(-\frac{\pi}{\mu B} \sin\left(\frac{2\pi F}{B}\right)\right) \) (1)

where \( X = (2\pi^2 k_B T m_e/\hbar e B) \) and \( k_B \) is the Boltzmann constant, \( T \) is the temperature. The presence of multiple SdH oscillation frequencies, however, makes the situation complex. That is why we start by discussing in detail how the LK expression can be used in the present context to extract reliable quantitative information.

The common way to extract the cyclotron mass \( m_c \) from SdH oscillations consists in plotting a resistance minimum measured at a fixed value of \( B \) as a function of \( T \), and subsequently fitting the data to the \( X/\sinh X \) dependence predicted by Eq. (1). This procedure works well in the presence of a single oscillation frequency, but it does not work if multiple frequencies are present. Without going into the microscopic physical details, that is because in the presence of multiple frequencies, the magnetic field values at which the SdH oscillation minima occur are found to vary upon changing \( T \) (contrary to the case in which a single SdH oscillation frequency is
FIG. 3. (a) Derivative of the transverse resistance $d\rho_{xy}/dB$ measured at 250 mK plotted as a function of $1/B$ (offset for clarity), exhibiting Shubnikov de Haas oscillations, and (b) corresponding Fourier Transform (black, red and blue curves represent data taken on device D1, D2, D3). Three peaks are present, located at the same frequencies $F_\alpha = 73$ T, $F_\beta = 118$ T and $F_\gamma = 163$ in the three devices (in D2, the peak at $F_\beta$ has virtually vanishing amplitude). (c) Temperature dependence of the SdH oscillations as extracted from the $d\rho_{xy}/dB$ data measured on device D1, and (d) corresponding Fourier transforms (in (c) and (d) the curves have been measured at the temperature values indicated in the legends). (e) The color dots represent the normalized amplitude of the peaks at frequency $F_\alpha$ (blue) $F_\beta$ (green), and $F_\gamma$ (red) as a function of temperature, measured on device D1. The continuous lines of the corresponding color represent the best fit obtained from the FT of the LK formula as described in the main text, using the cyclotron mass (different for every peak) as only fitting parameter. (f) Comparison of the full functional dependence of the FT of $d\rho_{xy}/dB$ data measured on device D1 at 250 mK (green triangles) with the FT calculated from the LK formula. The red curve corresponds to the case in which the carrier mobility is fixed at the value determined by the experimentally observed onset of the SdH oscillations; in the blue curve, the carrier mobility value was chosen to be different for each FT peak (see main text).

present). Hence, it is simply not possible to analyze the resistance minima at a fixed value of $B$ as a function of $T$.

The issue is well-known from past work on materials exhibiting SdH oscillations with multiple frequencies\textsuperscript{46–49}. In those systems, a commonly employed heuristic approach to analyze the temperature dependence of SdH oscillations consists in plotting the amplitude of a FT peak as a function of $T$ and fitting it with the expression $X/\sinh X$, i.e., with the same functional dependence normally used to fit the $T$-dependence of the resistivity minima. Although believed to give reasonable estimates for the cyclotron masses, this approach is problematic because the temperature dependence of the FT peak, as calculated from the LK expression, is not proportional to $X/\sinh X$. A simple way to understand the problem is to realize that since $X = (2\pi^2 k_B T m_c/eB)$–fitting the temperature dependence with the expression $X/\sinh X$ requires a specific value of magnetic field $B$ to be selected. The FT peak amplitude, however, is cal-
TABLE I. Effective masses $m_\alpha$, $m_\beta$, $m_\gamma$, extracted from the analysis of the temperature dependence of the amplitude of the $\alpha$, $\beta$ and $\gamma$ peaks in the FT, obtained from SdH resistance oscillations measured on devices D1, D2 and D3. The peak corresponding to the frequency $F_\beta$ is seen over a sufficiently large temperature range to enable the extraction of the corresponding cyclotron mass only in device D1. Note how the three different devices give nearly coinciding values for the cyclotron masses for the $\alpha$ and the $\gamma$ peaks.

| Device | $m_\alpha/m_e$ | $m_\beta/m_e$ | $m_\gamma/m_e$ |
|--------|----------------|---------------|----------------|
| D1     | 0.110          | 0.135         | 0.130          |
| D2     | 0.110          | -             | 0.127          |
| D3     | 0.103          | -             | 0.125          |

calculated by integrating the SdH oscillations over a large interval of magnetic field and not at one specific value of $B$. Therefore, the choice for the value of $B$ to be inserted in $X$ is arbitrary, which prevents a truly reliable determination of $m_c$.

We adopt a different approach, free from any ambiguity, that eliminates these problems. It consists in calculating the FT of the LK expression (Eq. (1)) and comparing the $T$-dependence of the calculated FT peak amplitude to the $T$-dependence of the corresponding experimentally measured quantity, using the cyclotron mass as a fitting parameter. The approach is free from ambiguity, because it compares the same quantity, measured or calculated.

Fig. 3d shows the FT of the SdH oscillations measured on device D1 for different values of $T$, ranging from 250 mK to 40 K. The colored dots in Fig. 3e represent the heights of the three FT peaks (normalized to the height at the lowest temperature of our experiments, i.e. 250 mK), extracted from the data in Fig. 3d. The continuous lines of the corresponding color are best fits obtained by using the FT of the LK formula, calculated in the same $B$-range used to calculate the FT of the experimental data. For each peak, the only fitting parameter is the cyclotron mass, as for all three peaks the carrier mobility (that also enters the theoretical expression) is fixed to be the value determined by the onset of the SdH oscillations. The agreement between data and the curves calculated from the LK expression is excellent and the value of the cyclotron mass extracted for each peak is given in Table I (for the peak corresponding to the frequency $F_\beta$, the quality of the data only allows us to determine the mass for device D1).

The procedure can be applied not only to the analysis of the $T$-dependence of the peak height, but also to the full functional dependence of the peaks, i.e. to their line-shape. This is useful because at sufficiently low temperature, the peak width is determined by the carrier mobility, and the comparison between calculated and measured FT can enable a more precise determination of this quantity. We illustrate the concept for device D1, for which the blue curve in Fig. 3f is calculated assuming $\mu_\alpha \approx 1800 \text{cm}^2/\text{V} \cdot \text{s}$, $\mu_\beta \approx 1000 \text{cm}^2/\text{V} \cdot \text{s}$ and $\mu_\gamma \approx 1800 \text{cm}^2/\text{V} \cdot \text{s}$ for the mobility values of the electrons in the pockets responsible for the different peaks in the FT. It is apparent that with these values the agreement with the data at high frequency is improved. Although a precise optimization is not particularly relevant in the present case, it is important to realize that using the LK formula properly to analyze the FT of the SdH oscillations offers possibilities that go beyond the heuristic approach that is commonly employed.

The information about the extremal Fermi surfaces extracted from the analysis of the SdH oscillations are largely consistent with the properties of the electron pockets of bulk jactungaite computed by density-functional theory (DFT; simulations were done using the Quantum ESPRESSO distribution, see SI for details). When SOI is neglected (see Fig. 1b), a degeneracy between the top valence and bottom conduction bands is enforced along the vertical edges of the Brillouin zone by the 3-fold rotation symmetry of the crystal (see the KH line in Fig. 1b), and gives rise to nodal lines. Additional crossings between valence and conduction bands occur away from high-symmetry directions as a consequence of a band inversion driven by the strong interlayer coupling (see the LAL line in Fig. 1b), forming two additional nodal loops. The location within the Brillouin zone of these nodal lines and nodal loops is reported in Fig. 4a. The color coding denotes the common energy of the valence and conduction bands along the nodes relative to the Fermi level (blue = above; red = below), with the dispersion resulting in multiple hole and electron pockets.

To enable comparison with experiments we perform additional DFT simulations including SOI effects. The nodal lines and loops become slightly gapped, but the overall qualitative picture does not change. Fig. 4b shows the corresponding Fermi surface, where electron and hole pockets are represented in blue and grey respectively, in good agreement (in terms of number and polarity) with the expectations from Fig. 4a. We identify four symmetry-inequivalent electron pockets and two pockets with hole character. The extremal contours of these pockets orthogonal to the magnetic field direction (z-axis) are also reported, with solid (dashed) lines corresponding to positive (negative) values of $k_z$. Their projection on the $(k_x, k_y)$ plane is shown in Fig. 4c, with the same color coding as in panel b. This projection is in very good agreement with recent ARPES measurements, with the only difference that the two types of hole pockets are merged into one in experiments. This qualitative feature is not captured by DFT, unless the Fermi energy is shifted on the hole side by few meVs (see panel d).

The area $S$ of the extremal contours of the Fermi surface that we identity provides an estimate for the expected SdH oscillation frequencies through the Onsager relation $F = \hbar/(2\pi)S \cdot \mu$, while the cyclotron mass can be computed through the derivative $m_c = h^2/(2\pi)\partial S/\partial E_F$. 

FIG. 4. (a) The two sets of nodal lines in the Brillouin zone of jacutingaite: vertical nodal lines are present at the edge of the Brillouin zone (BZ) and other closed-loop nodal lines encircle the ΓA direction. The color coding denotes the energy relative to the Fermi level. The red dashed lines show the paths over which energy bands Fig. 1b are computed. (b) Three-dimensional representation of the calculated Fermi surface in reciprocal space (the thin solid line represents the boundary of the BZ). Blue (grey) surfaces denote electron (hole) pockets, whose extremal contours are highlighted with solid (dashed) lines of different colors for positive (negative) values of $k_z$. In agreement with panel (a), we find alternating electron and hole pockets along either the vertical nodal lines or horizontal nodal loops. (c) Projections on the ($k_x, k_y$) plane of the extremal contours resulting from all different pockets, with the same colors and line styles as in panel (b). (d) Evolution of the hole pockets around the KH line, merging into a single one as seen in ARPES when the Fermi energy is slightly shifted on the hole side. (e) SdH oscillation frequencies $F$ (in Tesla) and cyclotron effective masses $m_c$ (in units of the free electron mass $m_e$) calculated using two different functionals (vdW-DF2-cx$^{51,52}$ and rVV10$^{53,54}$) as a function of the shift in Fermi energy with respect to the DFT neutral reference. Different colors correspond to different electron pockets with the same coding as in panels (b) and (c).

In summary, the results presented here demonstrate the possibility to realize nano-structured devices based on exfoliated jacutingaite ($\text{Pt}_2\text{HgSe}_3$) crystals that enable the observation of high-quality transport properties. The observed electron mobility values are comparable to those reported in WTe$_2$ layers of the same thickness. This is an extremely promising result, given that our jacutingaite devices are based on crystals grown by using a newly developed process that offers considerable margins of improvement. Crystal growth under high pressure occurs within a short time (hours) and imposes large mechanical constraints to the growing crystals, thus limiting their size and quality. Fine tuning the growth parameters and making the growth time longer is expected to provide larger crystals of even higher quality, and to allow much larger exfoliated layers to be produced, eventually enabling the realization of monolayer devices. Given that all experiments performed until now –both the transport measurements discussed here and ARPES experiments recently reported– have shown an overall excellent agreement with the predictions of first-principles calculations, we expect that jacutingaite monolayer devices will represent an extremely promising platform to investigate transport in 2D quantum spin Hall insulators.
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SUPPORTING INFORMATION

S1. DETAILS ON CRYSTAL GROWTH

Because of the high vapour pressure of mercury at low temperatures and the high reaction temperature needed for alloying platinum, the growth of Pt$_{2}$HgSe$_3$ must be carried out under extreme conditions. Single crystals of jacutingaite Pt$_{2}$HgSe$_3$ have been grown from the melt under high isostatic pressure in a hot-stage cubic-anvil press. The precursor components Pt (sponge, 99.98%), HgSe (powder, 99.9%) and Se (shot, 99.99%) have been mixed with a nominal slightly Hg-exceeding composition Pt$_{2}$Hg$_{1.1}$Se$_3$ inside a Ar-filled glove box, and pelletized to a relative mass density of > 75%. The pellet was tightly inserted into a BN crucible, covered by BN caps, and placed inside a cylinder-like graphite heater. The heater fits inside a pyrophyllite cubic cell, the pressure transmitter, on each face of which a WC piston acts, thus transforming a uni-axial force in a quasi-isostatic pressure on the sample. With this apparatus, we could process the material at temperatures of 800-1000 °C under a pressure of 1-2 GPa. After quite a short heating and cooling process (1-2h at the maximum temperature followed by cooling at 50-70 °C/h down to 650°C), crystals of Pt$_{2}$HgSe$_3$ have grown inside the pellet and could be mechanically extracted.

The largest size of the crystals was ∼ 0.6 × 1.0mm$^2$ in the cleavage plane (perpendicular to the crystallographic c-axis). Owing to the very high density of the as-grown polycrystalline bulk, the crystal size is limited and the extraction of the crystal is a delicate operation. However, the use of high pressure strongly improves the growth kinetics and allows obtaining bulk crystals within few hours, while the vapor-solid reaction technique reported$^{35}$ could only produce powder samples over a time scale of 70 days.

Single crystalline platelets were extracted from the polycrystalline as-grown boule and analysed in a Jeol JSM7600F electron microscope equipped with an Oxford AZTec X-Max EDS X-ray spectrometer. The crystal composition measured by the EDS was found to be Pt$_{2.1}$Hg$_{0.9}$Se$_3$, which is very close to the nominal one but indicates a small amount of either Hg-vacancies or Pt-Hg substitutional defects.

Part of the as-grown pellet was ground and used for powder X-ray diffraction, which confirmed the formation of the jacutingaite with lattice parameters $a = b = 7.3506(1)$ Å and $c = 5.2959(1)$ Å, in very good agreement with previously reported structural studies$^{35}$.

Reflections from the secondary phase PtSe$_2$ were often present in the powder diffraction pattern (up to 6-7% in volume) and rare single crystals of PtSe$_2$ were found as well, with a similar size and aspect to those of Pt$_2$HgSe$_3$. In fact, PtSe$_2$ can stably form in the same temperature range as Pt$_2$HgSe$_3$ and, because of its structural similarity, can even grow epitaxially next to a Pt$_2$HgSe$_3$ grain, as already reported$^{58}$. For this reason, it was mandatory to analyse carefully every single crystal prior to proceeding with device preparation and physical measurements.

FIG. S1. SEM image of a Pt$_2$HgSe$_3$ crystal extracted from the as-grown polycrystalline boule.
S2. DETAILS OF FIRST-PRINCIPLES CALCULATIONS

First-principles simulations have been carried out within density-functional theory using the Quantum ESPRESSO distribution\textsuperscript{55,56}. Several van-der-Waals compliant functionals have been adopted to relax the atomic positions until the residual force on each atom was smaller than 2.5 meV/Å, while keeping the unit cell fixed to the experimental one. At this stage spin-orbit coupling has been neglected and electron-ion interactions have been approximated using pseudopotentials from the Standard Solid State pseudopotential library\textsuperscript{59} (v1.0), with a cutoff of 50 Ry on energy and 280 Ry on the charge density. Spin-orbit coupling is then included in calculations on the relaxed structure by using fully relativistic pseudopotentials from the Pseudo-Dojo library\textsuperscript{60} with a energy cutoff of 80 Ry. The Brillouin zone is sampled using a 8 × 8 × 8 Γ-centered Monkhorst-Pack grid with a cold smearing\textsuperscript{61} of 0.015 Ry. To compute the Fermi surface, energy bands are interpolated by mapping the electronic states onto a set of maximally-localized Wannier functions\textsuperscript{62} using WANNIER90\textsuperscript{63}. As starting projections we consider s- and d-orbitals on Hg and Pt, together with p-orbitals on Se, and the wannierization is performed on a 6 × 6 × 6 regular grid over the Brillouin zone. The Fermi surface is computed from energy bands Wannier interpolated over a dense 80 × 80 × 80 k-grid. The SKEAF software\textsuperscript{64} is then adopted to extract the extremal contours, their areas, and the corresponding cyclotron mass. Wannier functions are used also to obtain the reciprocal space dispersion of the nodal loops with the help of the WannierTools software\textsuperscript{65}.

S3. COMPUTED FREQUENCIES OF RAMAN ACTIVE MODES

|        | rVV16\textsuperscript{53,54} | vdw-DF-c09\textsuperscript{51,67} | vdw-DF2-c09\textsuperscript{67,68} | vdw-DF-ex\textsuperscript{72} | optB86b\textsuperscript{69} | optB88\textsuperscript{70} |
|--------|-------------------------------|---------------------------------|---------------------------------|-------------------|------------------|------------------|
| 65.8 E_u | 62.6 E_u                       | 62.8 E_u                        | 62.7 E_u                        | 63.5 E_u          | 64.8 E_u         | 64.8 E_u         |
| 75.8 E_g | 70.8 E_g                       | 71.1 E_g                        | 70.8 E_g                        | 71.7 E_g          | 73.2 E_g         | 73.2 E_g         |
| 91.2 A_1u | 86.3 A_2u                     | 86.6 A_2u                       | 86.6 A_2u                       | 88.0 A_2u         | 89.9 A_2u        | 89.9 A_2u        |
| 91.4 A_2u | 92.0 A_1u                     | 91.6 A_1u                       | 91.9 A_2u                       | 92.0 A_1u         | 92.4 A_1u        | 92.4 A_1u        |
| 93.4 A_2u | 92.0 A_1u                     | 91.8 A_2u                       | 91.9 A_1u                       | 92.3 A_2u         | 93.4 A_2u        | 93.4 A_2u        |
| 93.7 E_u | 92.0 A_2u                     | 91.9 E_u                        | 92.2 E_u                        | 93.0 E_u          | 94.0 E_u         | 94.0 E_u         |
| 99.8 A_1g | 92.1 E_u                     | 92.5 A_1g                       | 92.2 A_1g                       | 93.7 A_1g         | 95.9 A_1g        | 95.9 A_1g        |
| 118.7 E_u | 115.8 E_u                     | 115.8 E_u                       | 115.8 E_u                       | 116.3 E_u         | 117.3 E_u        | 117.3 E_u        |
| 135.6 E_g | 129.1 E_g                     | 129.7 E_g                       | 129.5 E_g                       | 131.1 E_g         | 132.9 E_g        | 132.9 E_g        |
| 140.4 E_u | 140.0 E_u                     | 139.9 E_u                       | 140.1 E_u                       | 140.4 E_u         | 140.7 E_u        | 140.7 E_u        |
| 152.1 A_2u | 146.6 A_2u                 | 146.7 A_2u                       | 146.6 A_2u                       | 147.5 A_2u         | 149.3 A_2u        | 149.3 A_2u        |
| 173.2 E_g | 177.0 E_g                     | 176.3 E_g                       | 177.1 E_g                       | 177.4 E_g         | 176.7 E_g        | 176.7 E_g        |
| 176.5 A_2g | 184.1 A_2g                 | 183.1 A_2g                       | 184.0 A_2g                       | 182.6 A_2g         | 180.4 A_2g        | 180.4 A_2g        |
| 201.0 E_u | 196.5 E_u                     | 196.8 E_u                       | 196.9 E_u                       | 198.3 E_u         | 200.0 E_u        | 200.0 E_u        |
| 202.1 E_g | 198.7 A_1g                     | 198.8 A_1g                       | 199.1 A_1g                       | 201.4 A_1g         | 203.2 A_1g        | 203.2 A_1g        |
| 202.2 A_1g | 202.7 A_2g                 | 202.9 A_2u                       | 202.9 A_2u                       | 203.9 A_2u         | 204.6 E_g        | 204.6 E_g        |
| 207.8 A_2u | 204.0 A_1g                 | 203.9 A_1g                       | 204.1 A_1g                       | 204.7 A_1g         | 205.1 A_2u        | 205.1 A_2u        |
| 209.0 A_1g | 207.2 E_g                     | 206.3 E_g                       | 207.1 E_g                       | 206.1 E_g         | 205.8 A_1g        | 205.8 A_1g        |
| 214.1 A_2u | 219.6 A_2u                 | 218.9 A_2u                       | 219.6 A_2u                       | 218.9 A_2u         | 217.4 A_2u        | 217.4 A_2u        |
| 218.5 E_u | 221.6 E_u                     | 221.1 E_u                       | 221.7 E_u                       | 221.3 E_u         | 220.2 E_u        | 220.2 E_u        |
| 220.2 E_u | 225.3 E_u                     | 224.5 E_u                       | 225.2 E_u                       | 224.0 E_u         | 222.1 E_u        | 222.1 E_u        |
| 225.9 A_1u | 230.3 A_1u                 | 229.6 A_1u                       | 230.3 A_1u                       | 229.3 A_1u         | 227.8 A_1u        | 227.8 A_1u        |
FIG. S2. Frequencies (in cm$^{-1}$) of the Raman active modes of bulk jacutingaite computed with different van der Waals functionals. Full and empty symbols correspond to modes belonging to the $E_g$ and $A_{1g}$ representation, respectively. Experimental results are reported as horizontal lines with solid and dashed lines denoting $E_g$ and $A_{1g}$ modes, as identified by polarization-resolved Raman spectroscopy. An overall good agreement (both in terms of frequencies and symmetry assignment) is obtained with the vdw-DF-cx functional$^{52,68}$ adopted in the main text.