

Exceptional electronic transport and quantum oscillations in thin bismuth crystals grown inside van der Waals materials

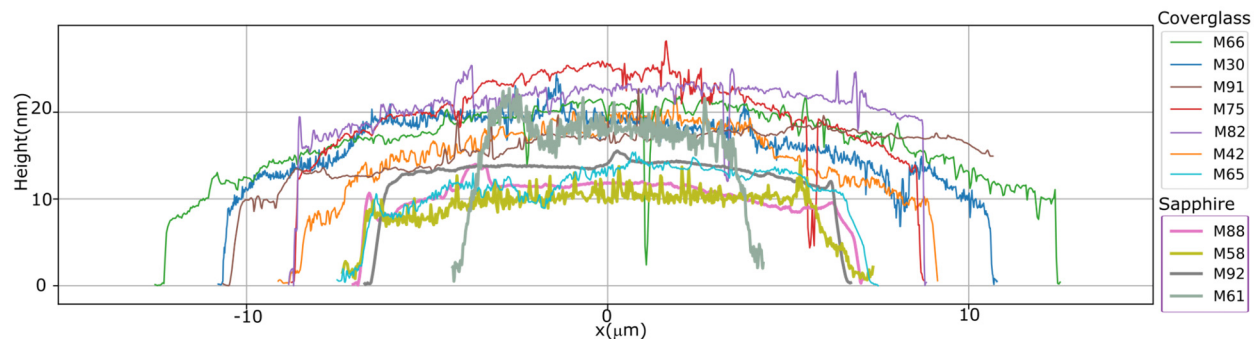
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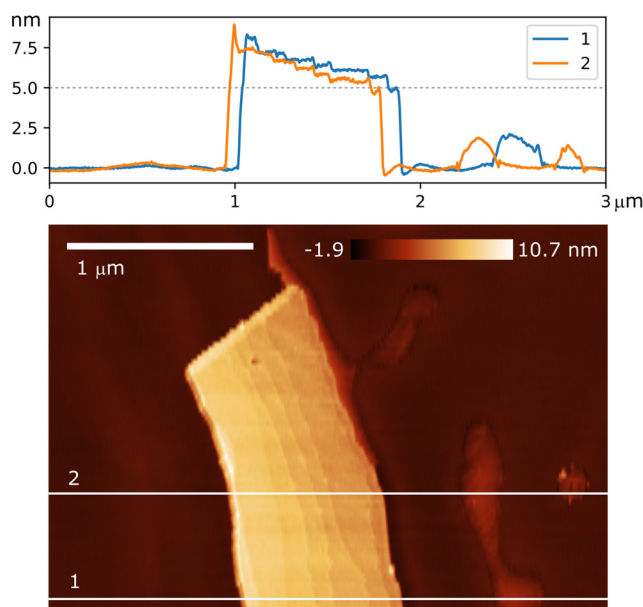
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1. Overview of vdW-molded bismuth results

In this section, we provide additional data on the vdW-molded bismuth samples we have produced. We measured 40 vdW-mold samples in AFM that produced ultraflat bismuth crystals with thicknesses ranging from 5 nm to 150 nm.



Supplementary Figure 1. AFM line profiles of vdW-molded bismuth samples squeezed by glass or sapphire top substrates showing a dome shape. Squeezing with sapphire as the top substrate leads to overall thinner bismuth with a flatter profile.

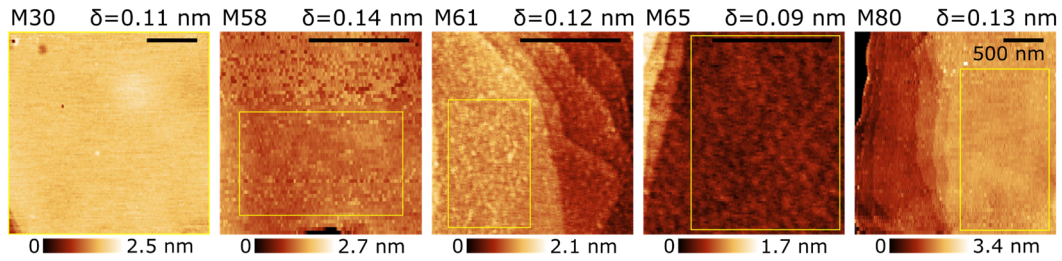


Supplementary Figure 2. AFM topography and line profile of a vdW-molded bismuth showing a 5 nm thick terrace.

Table 1: Thickness and terrace sizes for vdW-molded bismuth samples.

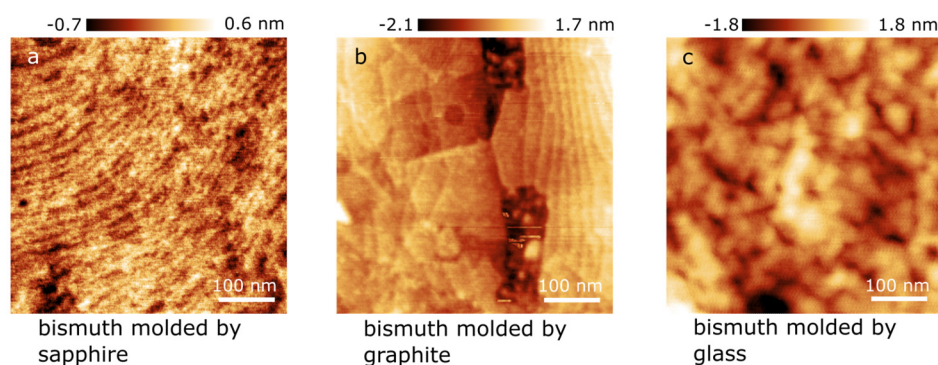
Note: This table only shows thin samples molded between two hBN layers.

Sample Name	Max thickness (nm)	Min thickness (nm)	Diameter (μm)	Substrate	Max Terrace width (μm)
M60	15	12	18	Glass	2.1
M69	16	12	11	Glass	0.4
M65	17	12	13	Glass	1.0
M48	17	12	20	Glass	
M42	19	9	21	Glass	
M77	21	12	26	Glass	0.9
M30	21	6	21	Glass	2.2
M91	22	13	32	Glass	
M66	22	9	27	Glass	0.7
M79	25	13	17	Glass	1.0
M75	26	16	24	Glass	0.3
M87	26	20	27	Glass	1.2
M76	27	13	23	Glass	0.4
M59	33	18	9	Glass	0.6
M49	39	13	24	Glass	1.0
M58	10	7	17	Sapphire	1.8
M88	11	9	18	Sapphire	2.4
M80	11	11	26	Sapphire	1.3
M92	15	11	14	Sapphire	3.8
M61	20	12	9	Sapphire	1.2



Supplementary Figure 3. Additional AFM roughness data from 5 different vdW-molded bismuth samples. The roughness shown on top of each AFM image is measured in the region inside the yellow box. Scale bar is 500 nm. All samples show low roughness comparable to the sample in Figure 2c.

2. Molding with other substrates



Supplementary Figure 4. AFM topography of bismuth molded under pressure between different substrates.

Table 2: RMS roughness of different molding substrates and the corresponding molded bismuth

Substrate	RMS roughness of the substrate	RMS roughness of the molded bismuth
Sapphire	0.63 Å	2.1 Å
Graphite ¹	0.23 Å	1.0 Å
Glass	1.3 Å	5.3 Å

To study the imprinting effect of substrates besides hBN on the molded bismuth, we did AFM analysis (Supplementary Figure 4) on bismuth molded directly between sapphire wafers (University Wafer 2" sapphire substrate wafer, 100 +/- 25 μm $\langle 0001 \rangle$ +/- 1°), graphite and microscope cover glass.

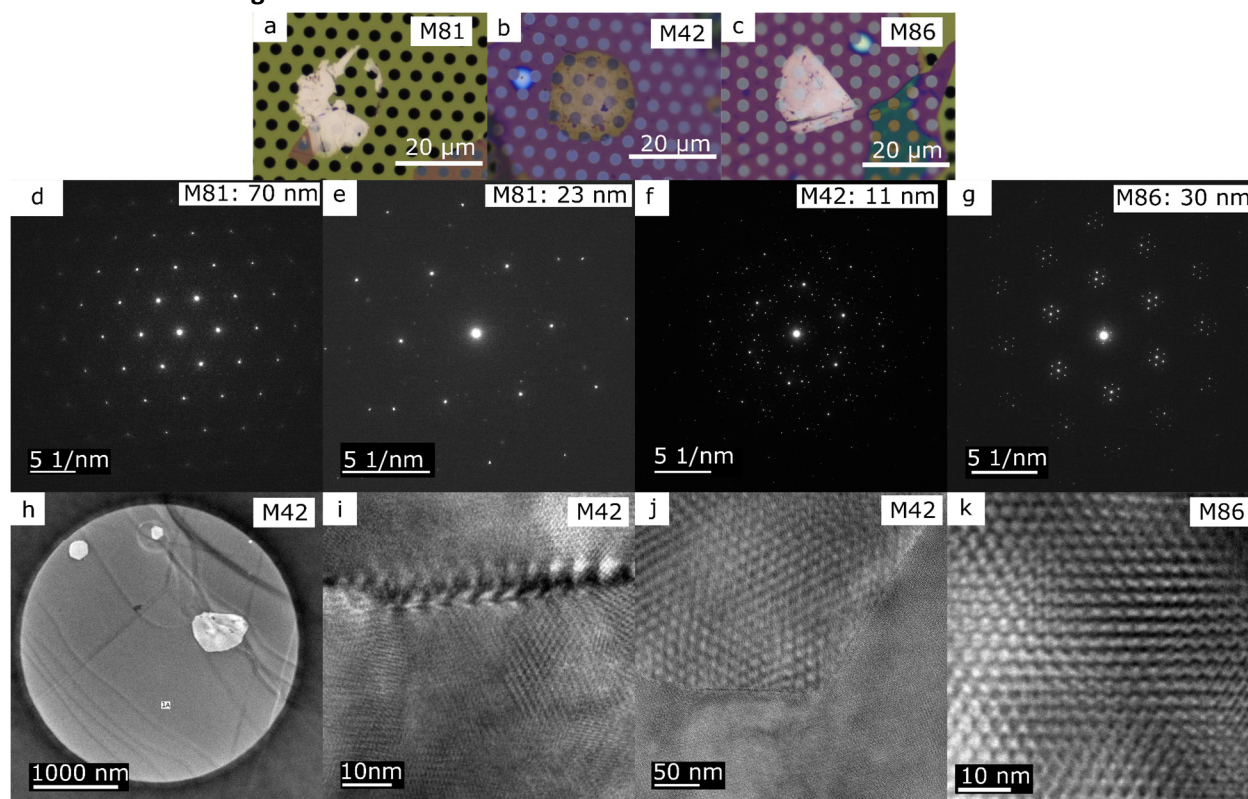
The average root mean square (RMS) roughnesses of the molding substrates and the molded bismuth are listed in Table 2. The roughness of the bismuth follows the trend of the substrates. Graphite-molded bismuth shows terrace structure similar to the hBN-molded bismuth.

3. Characterization of the crystal structure of vdW-molded bismuth

We use EBSD and TEM to characterize the crystal structure of the vdW-molded bismuth. Both measurements consistently show a (111) orientation for the thin crystals (Supplementary Figure 5-6, Extended Data Fig. 4). EBSD measurements show large internal crystal domains (Supplementary Figure 6, Extended Data Fig. 4), with only 1 to 4 domains covering a majority of a 20 μm diameter flake. The domains are differentiated by relative in-plane rotations, which will lead to identical surface state band structures except for an overall rotation of the Brillouin zone. Further discussion on the transport effects of domain walls can be found in Section 8.

The location of domain walls is highly correlated with the presence of irregular features in optical images of the vdW-molded bismuth. These irregular features are typically (1) internal voids, which arise from gases and hydrocarbons trapped inside the hBN during the bismuth encapsulation process, and (2) oxide residue, that comes from the surfaces of the source bismuth grain. Flake regions that are free of such irregularities show substantially larger single-crystal regions (Extended Data Fig. 4a). We anticipate that larger single-crystals can be obtained by processing within a vacuum chamber, which will limit oxide residue and trapped gases.

Additional TEM images and diffraction measurements



Supplementary Figure 5. 3 samples measured in TEM all exhibited rhombohedral lattice structure with the (111) axis oriented out of plane. a-c, Optical image of vdW-molded bismuth samples on TEM grids. **a,** vdW-molded bismuth with hBN flakes removed, **b,** vdW-molded bismuth encapsulated between hBN flakes and **c,** vdW-molded bismuth with bottom hBN flake. **d-g,** Selected area diffraction images for the samples shown in a,b,c. Bismuth

thickness at site of diffraction measurement is indicated in top right corners. **h-k**, Real space TEM images showing terraces (h), grain boundaries (i&j), and bismuth-hBN moire patterns (j&k).

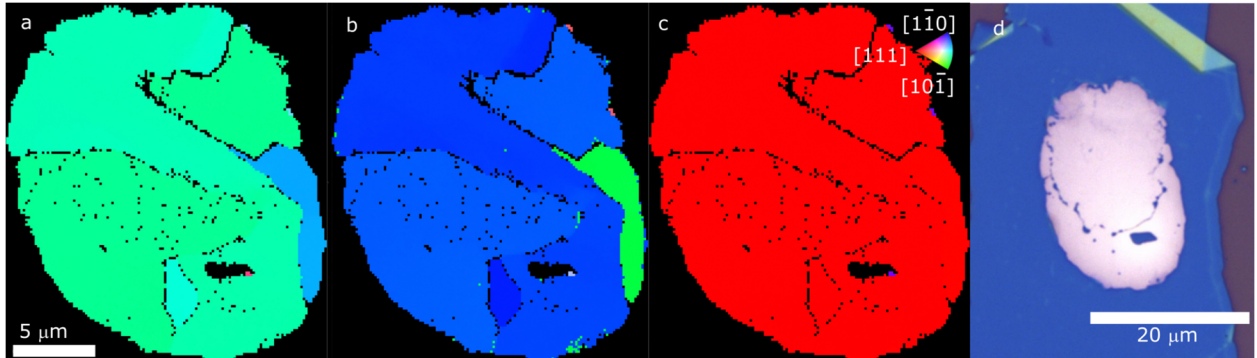
Calculation of lattice constant from diffraction images: In the selected area diffraction pattern for bismuth, we observe the $(1\bar{1}0)$ family of diffraction spots (Fig. 2f). We describe the bismuth lattice using the rhombohedral unit cell with parameters from the hexagonal close-packed structure: a is the in-plane lattice constant and c is the out-of-plane lattice constant². The reciprocal space lattice vectors of the bismuth rhombohedral unit cell can be described by the following, where $\hat{x}$, $\hat{y}$ and $\hat{z}$ are cartesian coordinate vectors.

$$\begin{aligned} b_1 &= \frac{-2\pi}{a} \hat{x} - \frac{2\pi\sqrt{3}}{3a} \hat{y} + \frac{2\pi}{c} \hat{z} \\ b_2 &= \frac{2\pi}{a} \hat{x} - \frac{2\pi\sqrt{3}}{3a} \hat{y} + \frac{2\pi}{c} \hat{z} \\ b_3 &= \frac{2\pi\sqrt{3}}{3a} \hat{y} + \frac{2\pi}{c} \hat{z} \end{aligned}$$

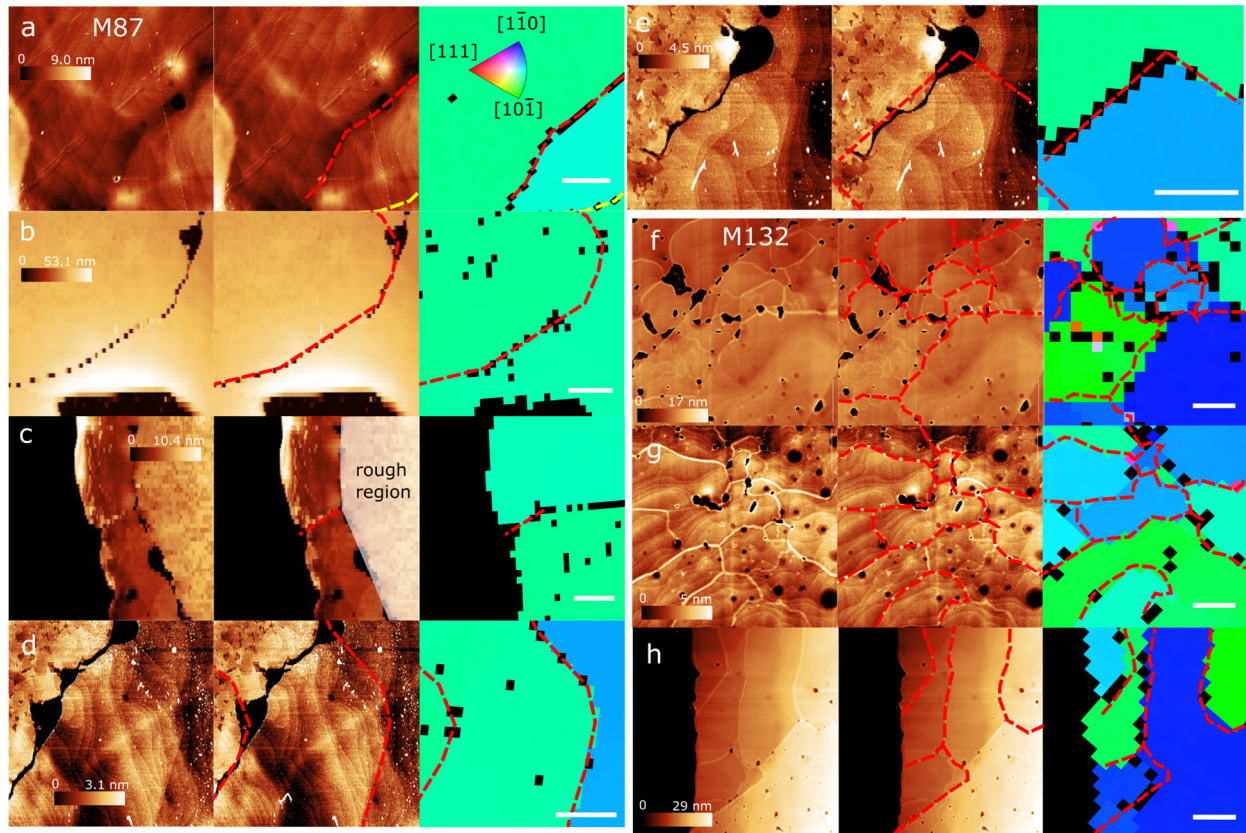
From our data, we measure the magnitude of the reciprocal space lattice vector from $(1\bar{1}0)$ to $(\bar{1}10)$ diffraction peaks. The measured magnitude is $0.434 \text{ \AA}^{-1}$, resulting in a value of $a = 4.62 \text{ \AA}$. This value is the same as is observed in real space images and is close to the bulk bismuth lattice constant of $4.54 \text{ \AA}$.

Electron beam backscattering (EBSD) measurements

EBSD measurements are performed in a Tescan GAIA3 Scanning Electron Microscope (SEM) operating at 20 kV. EBSD data is processed using the MTEX software package in Matlab³.



Supplementary Figure 6. a-c, Electron beam backscattering (EBSD) inverse pole figures (IPF) of vdW-molded bismuth sample shown in Figure 2 g&h. a, Y-axis IPF, b, X-axis IPF, c, Z-axis IPF, d, Optical image of vdW-molded bismuth used for EBSD (top hBN removed). Note that due to stage drift, the EBSD image appears distorted in size compared to the optical image.

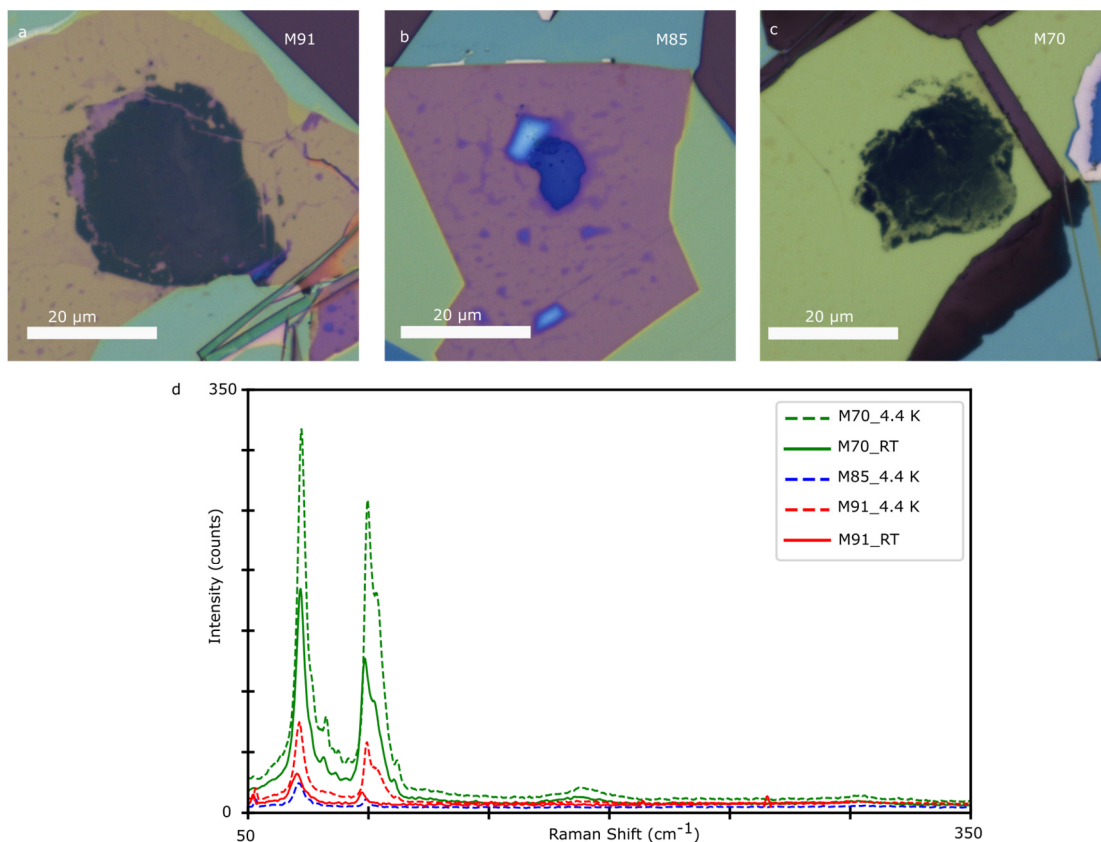


Supplementary Figure 7. Visibility of domain boundaries in AFM images. a-h, Each set of 3 adjacent images correspond to an AFM topography map (left), the same AFM map with domain wall overlay (center), and an EBSD map of the same sample region with the same overlay (right). Dashed overlay lines indicate the grain boundaries identified from EBSD. Red dotted lines indicate a domain wall observed in both EBSD and AFM. Yellow dashed line indicates a domain wall not observed in AFM (a). White shaded area is a rough surface region that we exclude. Data is taken from two samples: M87 (a-e) and M132 (f-h). The scale bar is 1 μm .

Correlating domain structure in AFM and EBSD data of vdW-molded bismuth

Domain walls observed in EBSD can also be observed in AFM. Supplementary Figure 7 shows a comparison of EBSD and AFM data from different regions on two samples. In AFM, we observe that domain boundaries typically appear along terrace edges, internal voids, valleys, or as raised ridges. Domain walls can even be identified that cross the interior of a terrace, where they appear as a 2 nm raised ridge in an otherwise flat surface (Supplementary Figure 7f and g). Within regions composed of clear terraces that are not obscured by irregular features, we found only one domain wall that is not clearly visible in AFM, as compared to the > 30 domain walls observed in both AFM and EBSD. In this way, we can use AFM images to avoid domain walls within a device (see Extended Data Fig. 5 and Section 8).

4. Raman characterization

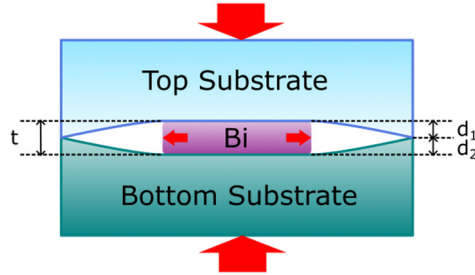


Supplementary Figure 8. **a-c**, Optical image of bismuth hBN-encapsulated samples. **d**, Raman data of M70, M91 at room temperature (solid lines) and M70, M85, M91 at 4.4 K (dash lines).

Raman data are taken in a Horiba LabRAM HR Evolution with an ultra-low frequency module (532 nm laser). The vdW-molded bismuth samples are on Si chips with 300 nm SiO₂. M91 and M85 are encapsulated stacks of vdW-molded bismuth. At room temperature, we observed two peaks in M70 at 71.9 cm⁻¹ and 98.6 cm⁻¹ (M91 at 70.3 cm⁻¹ and 97.3 cm⁻¹). These values closely match the frequencies of the normal modes observed in bulk rhombohedral bismuth at 71 cm⁻¹ (E_g) and 98 cm⁻¹ (A_{1g})⁴. In M91 and M85, we do not observe any bismuth oxide related Raman modes. In M70, we observe a faint peak at 185.9 cm⁻¹ which matches the Bi-O stretching mode at 185 cm⁻¹⁵. During the molding process of M70, most of the melted bismuth flowed out of the bottom hBN flake, leaving a higher concentration of the bismuth oxide from the source flake (Supplementary Figure 8c). We conclude that our samples are rhombohedral bismuth encapsulated between hBN flakes with minimal oxidation.

5. Model of squeezing limits and molecular dynamics simulations

Simple Continuum Model



Supplementary Figure 9. Depiction of substrate deformation during the squeezing process.

The squeezing limit model considers a balance of the pressure to squeeze the liquid bismuth due to its surface tension $P = 2S/t$ with the pressure to indent the substrate $P = 2rEd/\pi r^2 = 2Ed/\pi r$, where r is the droplet radius, E is the substrate's elastic modulus, d is the depth of indentation, and S is the difference in surface energy per unit area between the bismuth-hBN and hBN-hBN interfaces (i.e. the relative surface tension). Assuming identical top and bottom substrates, the limiting condition occurs when $t = 2d$, i.e. the bismuth is fully indented symmetrically into both substrates. From this condition we derive the thickness limit $t = \sqrt{2\pi S_T r/E}$. In the case where the substrates are different (ex: Silicon on the bottom, glass on top), then we can relate the depths of indentation into each substrate with the total thickness $t = d_1 + d_2$, where d_1 and d_2 are the depth of indent for the bottom and top substrates, respectively. The resulting derived equations are $d_1^2 = \pi r S_T / E_1 (1 + E_1/E_2)$ and $d_2^2 = \pi r S_T / E_2 (1 + E_2/E_1)$.

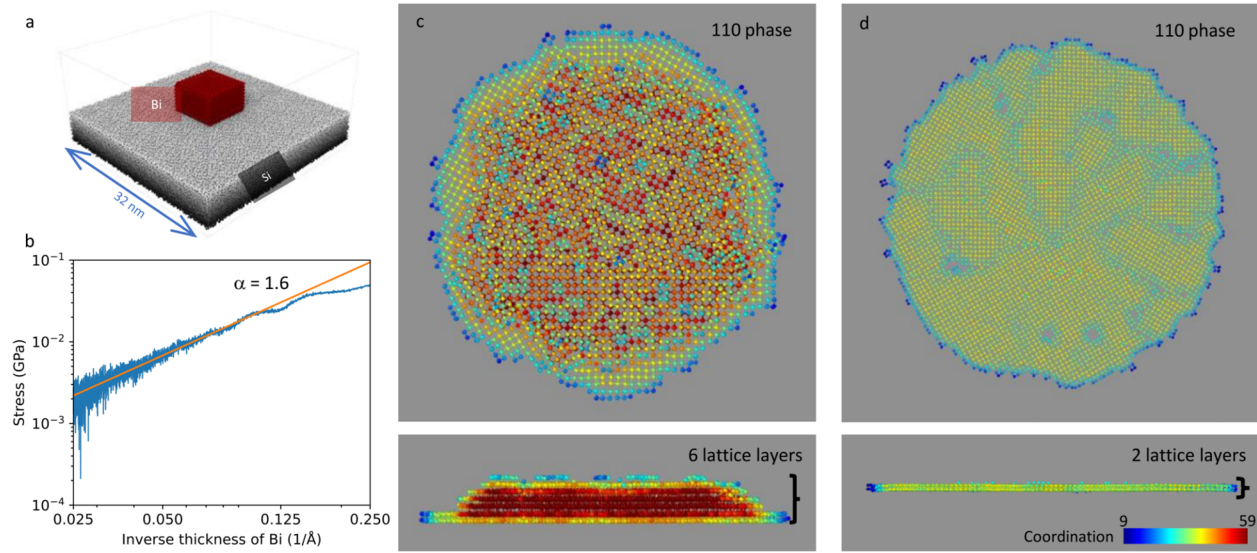
Table 3 shows calculated values for the limiting thickness under various conditions. For these estimates, we use the bare bismuth surface tension value of $S = 0.388 \text{ J/m}^2$ ⁶. Note that this neglects the interaction between the bismuth and hBN, and also neglects that growing the surface area of the bismuth requires breaking hBN-hBN interlayer bonds at the periphery of the laterally expanding droplet. The hBN-hBN cohesion energy is measured to be 0.326 J/m^2 ⁷, potentially nearly doubling the true value for S .

The primary results in this paper are for Si:Glass and Si:Sapphire squeezing, with predicted limits of 15 nm and 10.6 nm, respectively, using the above equations. This matches well the range of thicknesses we observe in our actual squeezings. Switching to dual Sapphire substrates would reduce the predicted thickness to 9 nm, and reducing the droplet radius by 4x would give an additional 2x reduction in thickness. Thinner squeezing may be achievable by circumventing the substrate deformation trapping, for example, by concentrating pressure into smaller regions.

Table 3: Limiting thickness value at max pressure for $r = 10 \mu\text{m}$ droplet following the simple deformation model

Squeezing substrates	thickness (nm)
Si:Si ($E=170\text{GPa}$)	12
Si:Glass ($170\text{GPa}:70\text{GPa}$)	15
Si:Sapphire ($170\text{GPa}:300\text{GPa}$)	10.6
Sapphire:Sapphire (300GPa)	9

Molecular Dynamics Simulations of Squeezing



Supplementary Figure 10. Molecular dynamics simulation of bismuth squeezed by silicon. **a**, 3D view of the simulation setup, consisting of a bismuth block on a silicon slab. **b**, Blue: Simulation data showing uniaxial compressive stress as a function of the inverse thickness of the squeezed bismuth in the liquid state (temperature 550 K). Orange: Power-law fitting to data with exponent α . **c**, Resulting bismuth crystal after melt-growth while squeezing with a 2 nm gap with a Si (111) substrate. Top view and side view (upper and lower panels, respectively). **d**, Same simulation as in (c) but with a 0.75 nm gap between squeezing substrates. The colors of the atoms represent the sum of coordination numbers from 1st to 5th nearest neighbors (c-d).

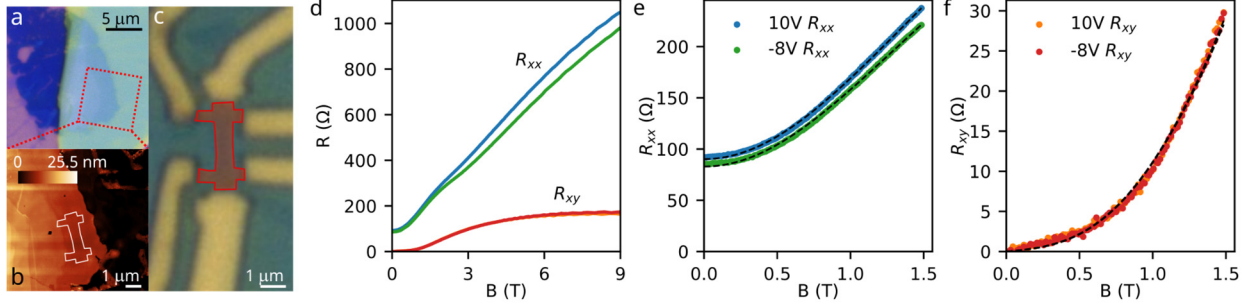
To further study the limits of the squeeze method of producing ultrathin crystals, we performed molecular dynamics (MD) simulations of a nanodroplet of bismuth squeezed between planer indenter and silicon substrates. In the simulations, we find that it is possible to achieve crystals thinner than 1 nm when using starting bismuth material that is 301 nm^3 in volume and at applied uniaxial compression of 21 MPa (Supplementary Figure 10). At this ultrathin limit, the bismuth is sensitive to the substrate interaction, in this case showing a (110)-orientated crystal when squeezed by (111)-orientated silicon (Supplementary Figure 10c&d) and an amorphous phase when squeezed by (100)-orientated silicon (not shown). Note that silicon was chosen as the squeezing substrate to simplify the simulation as compared to the more complex dynamics of a hBN crystal.

Details of Molecular Dynamics Simulations

Molecular dynamics simulations were performed using the open-source code LAMMPS⁸. The simulation consisted of a bismuth block compressed by a silicon substrate. The dimensions of the bismuth are

77.16 Å, 78.62 Å and 49.68 Å along x, y and z cartesian coordinates, respectively. The (111) plane of bismuth is perpendicular to the vertical direction, i.e., along the z-axis. The dimensions of the silicon were 325 Å, 325 Å and 55 Å, respectively. Periodic boundary conditions were applied along all three dimensions. Modified embedded-atom method (MEAM) was used to compute non-bonded interactions of bismuth⁹. Interactions between silicon atoms were modeled with a 3-body Stillinger-Weber (SW) potential¹⁰. The interactions between bismuth atoms and silicon atoms on the interface were described by the Lennard-Jones (LJ) potential, in which the well depth ϵ and size-parameter σ are 0.019996 eV and 3.225 Å, respectively¹¹. The temperature was maintained at 300 K by a Langevin thermostat with the damping parameter set to 1 ps. A planar indenter with a force constant equal to 10 eV/Å³ was used to compress the model from above the bismuth block with a constant velocity 0.1 Å/ps. The bottom of the silicon substrate was fixed to prevent movement. After the compression, the model was heated to the melting point (550 K) of bismuth for 50 ps. The planer indenter was maintained above the bismuth droplet to avoid the escape of bismuth atoms. The bismuth droplet was then cooled down to 300 K at a constant cooling rate 1 K/ps. The atomic structures were visualized by the program OVITO¹².

6. Two-carrier model fit



Supplementary Figure 11. Magnetic field-dependent measurement of the longitudinal resistance (R_{xx}) and the transverse resistance (R_{xy}) of a 8nm flat device at two different back gate voltages. a, Optical image of the bismuth where this device is fabricated. **b**, AFM image of the area marked out in a. **c**, Optical image of the fabricated device. **d**, R_{xx} and R_{xy} plotted as a function of magnetic field up to 12 T. **e**, Two-carrier model fit result under 1.5 T plotted on top of R_{xx} . **f**, Two-carrier model fit result under 1.5 T plotted on top of R_{xy} .

We are able to apply the semimetal two-carrier model¹³ on the longitudinal (R_{xx}) and transverse resistance (R_{xy}) (Supplementary Figure 11d) of M58, a flat 8 nm device (Supplementary Figure 11a-c). We corrected the R_{xy} data by subtracting the 0-field resistance, because we did not obtain resistance in the negative field to symmetrize the data.

$$\rho_{xx} = \frac{1}{e} \frac{n\mu_n + p\mu_p + (n\mu_p + p\mu_n)\mu_n\mu_p B^2}{(n\mu_n + p\mu_p)^2 + (p - n)^2\mu_n^2\mu_p^2 B^2}, \quad \rho_{xy} = \frac{1}{e} \frac{(p\mu_p^2 - n\mu_n^2)B + (p - n)\mu_n^2\mu_p^2 B^3}{(n\mu_n + p\mu_p)^2 + (p - n)^2\mu_n^2\mu_p^2 B^2}$$

Where n and p are the carrier density of electrons and holes, μ_n and μ_p are the corresponding mobility. We fit the R_{xx} and R_{xy} data under 1.5 T to the model, and the fit result is plotted on top of the data in Supplementary Figure 11e and f, and the fitted parameters are listed below.

Table 4: Carrier densities and mobilities from the two-carrier model fit

Gate Voltage (V)	n (cm ⁻²)	p (cm ⁻²)	μ_n (cm ² /Vs)	μ_p (cm ² /Vs)
10	7.63e+12	1.72e+13	1.22e+04	8.67e+03
-8	7.99e+12	1.84e+13	1.26e+04	8.84e+03

7. Effect of bismuth oxide on device transport measurements

The vdW-molded bismuth is protected from air during the growth process. However, the device nanofabrication process exposes the bismuth to some oxygen. Bismuth forms a self-limiting oxide layer about 1-3 nm in thickness as we discussed in Methods, and we can see signs of a crystalline oxide layer in our TEM measurements. As such, likely all of our electronic measurements of clean quantum oscillations in the bismuth surface states are coming from electrons near this oxide layer. Given the high quality of the resulting transport data and the agreement with STM and ARPES studies, we conclude that

this oxide layer does not significantly degrade transport. Instead, it seems that the surface roughness is the most important factor for thin bismuth transport. These observations are also supported by DFT calculations, which show that the bismuth surface state is barely affected by the presence of an oxide layer¹⁴.

8. Effect of domains and terraces on device transport measurements

In this work, we measure 12 different devices that vary in crystal uniformity. Despite wide variations in thickness and terrace structure, we observe qualitative consistent features such as larger RRR values compared to MBE-grown films, as well as clear quantum oscillations that have been previously unrealized in bismuth thin film transport. Here, we discuss in more detail the role of crystal nonuniformity in our electrical device characterization, specifically focusing on the effects of crystal domains and surface irregularities such as terraces.

Due to concerns for beam-induced damage and contamination, we do not perform EBSD or TEM diffraction measurements on our devices. As such, we cannot completely exclude the possibility of a domain wall in the device channel. But, our nanofabrication procedure is effective at minimizing domain walls. The vdW-molding process produces thin bismuth flakes with large domain sizes, typically at least $(4\text{ }\mu\text{m})^2$ in size, and some flakes are single crystal over $20\text{ }\mu\text{m}$ dimensions. Therefore, even random placement of a $1\text{ }\mu\text{m} \times 3\text{ }\mu\text{m}$ device will result in few to zero domain walls. To further minimize domains, we target regions that have flat and wide terraces. From our comparison of EBSD and AFM data, we observe that domain walls are clearly visible in AFM as topographic features (Supplementary Figure 7). In this way, we can produce devices that are single-crystal with very high certainty.

Of the 12 devices we present, 2 of the devices are made in a single flat terrace, and hence very likely to be single-crystal. These are the flat 8 nm and 13 nm devices that appear in main text Figure 3e,f,g and Figure 4c,e. We measure an additional 10 devices that are typically larger and with more non-uniform surfaces, primarily due to multiple terraces steps. Due to the surface non-uniformity, it is likely that some of these devices contain a few domain walls. As mentioned above, the non-uniform devices show the same qualitative features as the highly-uniform devices. Therefore, the presence of domain walls and terraces does not critically affect our conclusions.

These observations are consistent with the expected transport effects of a domain wall or terrace. A rotational domain wall in a (111) bismuth crystal is expected to behave as a 1-dimensional (1D) metallic wire^{2,15,16}. A domain wall that runs parallel to the device channel will have little effect on the conductance as it is a small fraction of the channel width and is shunted by parallel conduction through the metallic surface. From STM studies, we estimate that the domain wall has a width of 10 nm as compared to the $\sim 1\text{ }\mu\text{m}$ typical width of our devices¹⁷. When orientated perpendicular to the channel, the domain wall will act as a source of scattering as the domains have different in-plane orientations. This will impede electronic transport through the surface state and add extra resistance to the device. Terrace steps on the surface will behave similarly to domain walls, in that they should host 1D metallic

modes¹⁷, but should provide less resistive scattering to transverse transport when the crystal orientation is preserved across the terrace.

To summarize, the likely effect of sparse domain walls and terraces in our devices is to increase resistive scattering and, in this regard, behave similarly to other random sources of disorder, such as impurities or rough crystal edges. Disorder is expected to decrease the measured RRR value by increasing the residual resistivity at base temperatures. This is clearly observed in the comparison of the flat and rough device in Figure 3. It is also consistent with the highly-uniform devices exhibiting a greater improvement in RRR values as compared to the non-uniform devices.

Likewise, resistive scattering from disorder, including domain walls and terraces, will reduce the amplitude of quantum oscillations¹⁸, but will not change the frequency of the quantum oscillations themselves. Domain walls and terraces cannot produce quantum oscillations, as they are too narrow to support cyclotron motion. Furthermore, quantum oscillations are not sensitive to the in-plane rotations that differentiate the domains, as this simply causes an overall rotation of the electronic structure which preserves the size of the Fermi surface. Hence, a device that is made up of a few domains with different in-plane orientations will have a similar pattern of quantum oscillations as a single crystal device. We can estimate the length scale for scattering from the $B = 3$ T magnetic field where quantum oscillations typically onset, using the equations for the quantum scattering time $\tau = m_{eff}/eB$ and cyclotron diameter $d = 2m_{eff}v_F/eB$, where we use $m_{eff} = 0.16 m_0$ and $v_F = 4.7e5$ m/s¹⁹. This results in a 0.3 ps quantum scattering time that corresponds to a 280 nm cyclotron diameter. This is close enough to the ~ 1 μ m width of most of our devices that scattering from the device edges is likely an important limiting factor.

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