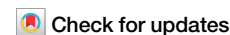


<https://doi.org/10.1038/s41699-026-00711-3>

Large area van der Waals epitaxy of high-mobility two-dimensional tellurium on silicon



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The two-dimensional form of tellurium, named tellurene, holds intriguing properties for a broad range of applications. Here, the large area growth by molecular beam epitaxy of tellurium on Sb-passivated Si(111) is demonstrated. The influence of growth temperature and film thickness on the material properties was investigated. At optimized conditions, the two-dimensional growth of Te films as thin as 4 nm is established. The epilayers possess a (10 $\bar{1}$ 0) surface orientation, with the Te chiral chains lying parallel to the surface plane. The van der Waals epitaxy occurs with the formation of a Sb₂Te₃ interlayer, as confirmed by transmission electron microscopy. Grazing incidence diffraction shows a discrete set of domains with in-plane rotation angles, which result in the alignment of Te lattice planes of neighbouring domains. The high hole mobility of epitaxial tellurium at room temperature is proven by Hall effect measurements. Optical spectroscopy measurements strongly support the electrical data. The thickness-dependent vibrational properties of the material were studied by Raman spectroscopy: by scaling down the film thickness, characteristic features of few-layer tellurium are observed. Hence, the present study opens the way for the integration of two-dimensional tellurium on silicon and the design of novel heterostructures of layered tellurides.

The exfoliation of graphene has been an outstanding achievement, contributing to the research to complement silicon in future semiconductor technology^{1,2}. Later on, a wide investigation of other two-dimensional (2D) materials has started, including transition metal dichalcogenides³ and elemental 2D materials, named Xenes⁴, such as silicene, germanene, stanene, phosphorene and borophene. While 2D compound materials are promising for high-end electronics, optoelectronics and energy harvesting, elemental Xenes are expected to be 2D topological insulators^{5,6} having potential applications in nanoelectronics and spintronics. In addition, 2D materials can be exploited to survey exotic transport and quantum effects⁷.

More recently, also 2D forms of group VI Se⁸ and Te⁹ have attracted attention. Tellurium in particular has unique physical properties with respect to other elemental 2D materials¹⁰. In its bulk form, it is a p-type semiconductor with a narrow band-gap of about 0.34 eV and strong spin-orbit coupling¹¹, featuring an inherent structural anisotropy with a pseudo-one-dimensional crystalline structure composed of parallel arranged

helical chains. Indeed, it crystallizes with one of the enantiomorphous chiral space groups P3₁21 or P3₂21: the three atoms in the unit cell are covalently bonded to their nearest neighbours within the chain, while adjacent chains are held together in a hexagonal lattice through van der Waals (vdW) interactions¹². In the following, the term “2D tellurium” is used to indicate the in-plane alignment of the chiral chains of the bulk-like phase.

Nanostructured Te exhibits compelling, thickness-dependent properties that make it a promising material for various applications¹³. For instance, it has a tunable bandgap, high carrier mobility¹⁴ and low thermal conductivity¹⁵. In addition, it has semiconducting¹⁶, photoconductive¹⁷, topological¹⁸, and thermoelectric¹⁹ properties, thus representing an engaging platform for electronics^{20,21}, optoelectronics^{14,15,22}, spintronics¹¹, as well as for the realization of infrared acousto-optic detectors, sensors and energy devices²³. Moreover, Weyl fermions were observed in n-type 2D tellurium, also named tellurene, synthesized by a hydrothermal method²⁴. Remarkably, solution-grown tellurene (substrate-free alkaline solution synthesis)

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enabled field-effect transistors with field-effect mobilities on the order of $700 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, while maintaining large switching ratios²⁰.

Theoretical studies reported in literature investigated different possible 2D phases of tellurium: (i) a bulk-like trigonal phase, comprised of helical chains with main axis along the [0001] crystallographic direction arranged in an array of centered hexagons by weak interchain interactions, as well as (ii) tetragonal²⁵, (iii) hexagonal^{25–27}, (iv) monoclinic^{26,28} and (v) buckled square²⁹ phases. The actual structure of 2D tellurium strongly depends on the growth technique and the substrate employed. The vdW epitaxy of Te by molecular beam epitaxy (MBE) has been demonstrated on the surface of graphene on 6H-SiC(0001)^{12,30}, mica³¹, and highly oriented pyrolytic graphite substrates³². In addition, 2D tellurium sheets were synthesized by physical vapour deposition and pulsed laser deposition on different substrates^{10,33}.

Besides, there are numerous examples of layered tellurides been grown on Si(111) passivated surfaces. The Si(111)–($\sqrt{3} \times \sqrt{3}$)R30°–Sb surface was used formerly by our group for the growth of Sb_2Te_3 ³⁴, $\text{Sb}_{2+x}\text{Te}_3$ ³⁵ and $(\text{GeTe})_m(\text{Sb}_2\text{Te}_3)_n$ ³⁶, whereas Kampmeier et al. grew Bi_2Te_3 on Te-terminated Si(111) substrates³⁷. Within this context, the use of silicon as a substrate for 2D tellurium would represent a key step towards its integration with Si-based technologies, bridging fundamental studies and device-oriented implementations.

In the present work, the large area vdW epitaxy of Te films on the Sb-passivated Si(111) reconstructed surface is demonstrated. The choice of this specific surface of silicon is motivated by its hexagonal surface symmetry and the absence of dangling bonds, which are essential to promote the in-plane alignment of the Te helical chains and suppress competing growth modes. The influence of growth temperature on the synthesis of Te films is discussed. After identifying the optimal growth conditions, (i) the electrical and optical properties of a Te film of about 22 nm and (ii) the structural and vibrational properties for films with thicknesses down to 4 nm are analyzed.

Results

Tellurium films were deposited by MBE at constant substrate temperature, varying between 15 °C and 205 °C (Sec. Growth study). The total deposition time was 90 minutes unless otherwise noted. After identifying the optimal epilayer quality, the electrical and optical properties of a $\approx 22 \text{ nm}$ thick Te film grown on high-resistivity Si(111) were inspected (Sec. Electrical and optical characterization). Then, the epitaxy of Te at different stages was analysed at the optimized growth temperature, i.e., 130 °C, in Sec. Thickness study. To this aim, different growth durations of 11, 22, and 45 minutes were employed. The growth experiments were followed in situ by reflection high-energy electron diffraction (RHEED). The surface morphology was probed by atomic force microscopy (AFM) and the data were analysed with Gwyddion software³⁸ to obtain the film roughness.

Growth study

Fig. 1 shows the RHEED patterns and the corresponding AFM topography images acquired for the as-deposited Te samples grown at different substrate temperatures. The RHEED images of the samples were taken along a $\langle 11\bar{2} \rangle$ azimuth of the Sb-passivated Si surface to evaluate the surface morphology and crystal quality. In the case of the film grown at 15 °C, the RHEED pattern of the substrate vanished ≈ 1 minute after the opening of the Te cell shutter, and at the end of the growth, only diffuse scattering was seen, corresponding to an overall amorphous film. Indeed, the topography image reveals only sparse features on the surface. For a substrate temperature of 80 °C, spots and faint rings are seen, which testify the deposition of both textured and misoriented three-dimensional crystalline islands, as confirmed by the AFM topography image. This finding is in agreement with Te films grown by MBE at low temperatures on (0001) sapphire³⁹. For the sample deposited at 105 °C, a superposition of streaks, sharper spots, and also very weak rings is found, the latter ascribed to a residual polycrystalline growth. For this film, the AFM measurement reveals the formation of thin crystallites elongated parallel to the substrate surface, which appear in-plane

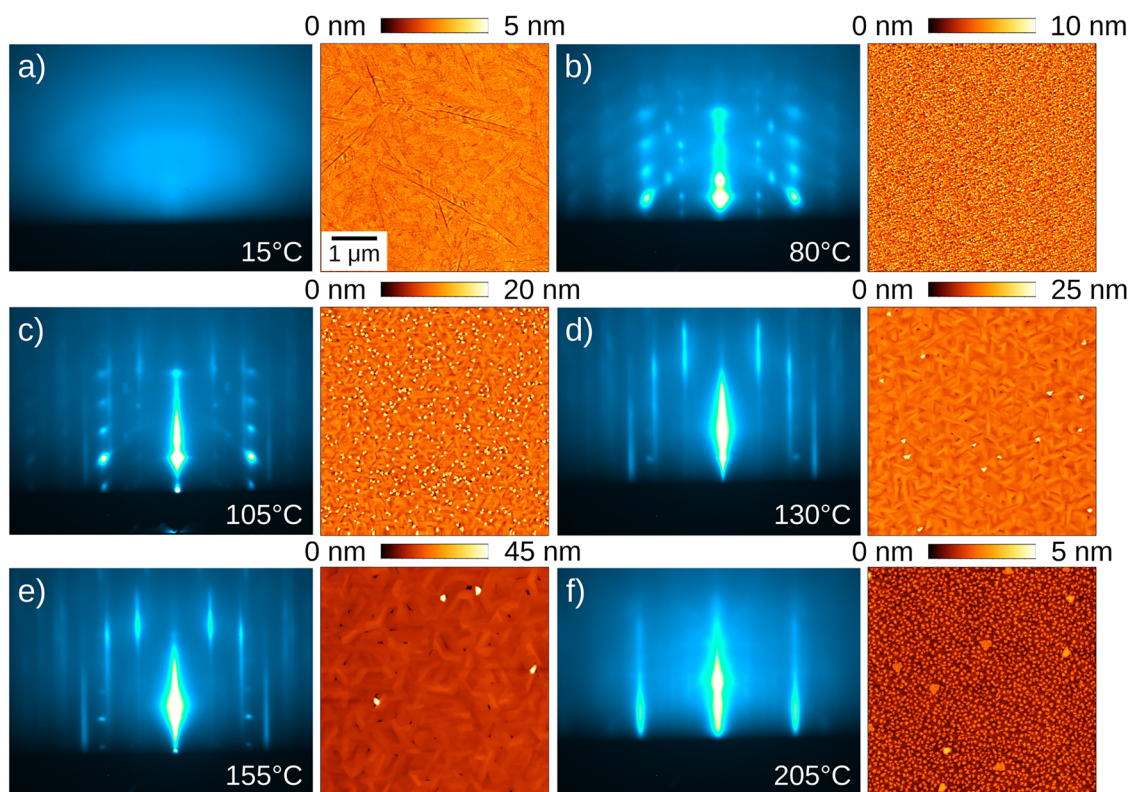


Fig. 1 | Morphological characterization of the Te films grown on Sb-passivated Si(111) at different substrate temperatures. The films were deposited at **a** 15°C, **b** 80°C, **c** 105°C, **d** 130°C, **e** 155°C and **f** 205°C. (Left) RHEED images of the as-

deposited films along a Si(11 $\bar{2}$) azimuth of the Si(111)–($\sqrt{3} \times \sqrt{3}$)R30°–Sb passivated surface and (right) AFM topography images. All the AFM images have the same size (the scale bar is reported in (a)).

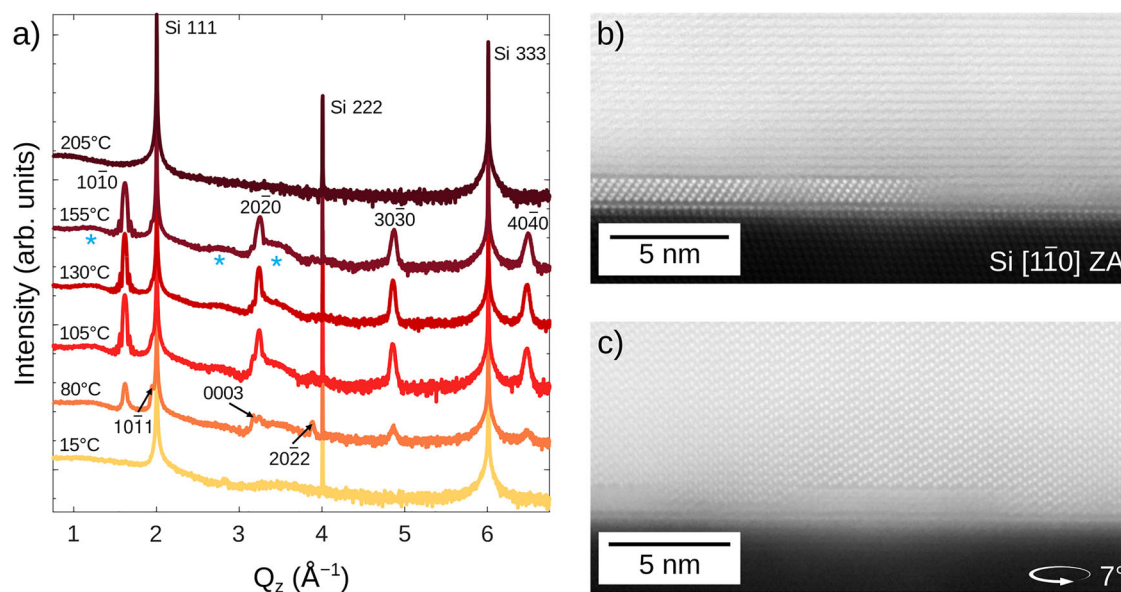


Fig. 2 | Structural characterization of the Te films grown on Sb-passivated Si(111). **a** XRD radial scans of the samples as a function of substrate temperature. The curves are plotted in logarithmic scale and vertically shifted for clarity. The black arrows indicate different families of Bragg peaks present for substrate temperatures below 130 °C. The light blue asterisks highlight broad features attributed to a very

thin Sb_2Te_3 layer formed at the interface between the Te epilayers and Si. **b** HAADF-STEM micrograph of the Te sample grown on Si(111) at 130 °C, aligned along a Si $[1\bar{1}0]$ zone axis. **c** HAADF-STEM micrograph of a different region, acquired with an azimuthal rotation of $\approx 7^\circ$ to align the sample along the $[1120]$ zone axis of the Te epilayer.

oriented, and triangle-shaped islands divided into three well-oriented domains (not shown here), which we interpret as the onset of layer-by-layer growth. In the case of the sample grown at 130 °C, after ≈ 5 minutes, the RHEED pattern turned streaky and remained unchanged until the end of the growth. The presence of a few spots with slightly different in-plane spacing was also recorded. The topography is similar to that of the previous film, although in this case the islands are much less numerous and the domains parallel to the surface are larger. The RHEED pattern of the sample grown at 155 °C is very similar to that of the film deposited at 130 °C, but the streaks are weaker, whereas the spots are sharper and more intense. In fact, in the topography image, the islands are thicker and less regular. Finally, for the deposition of Te at 205 °C, despite the RHEED pattern being streaky, only a few very thin islands are formed on the surface. The latter pattern resembles that of Sb_2Te_3 ⁴⁰, suggesting that no tellurium crystals are deposited at this temperature, in agreement with its desorption behaviour⁴¹.

From the AFM measurements and by fitting the X-ray reflectivity (XRR) curves, the values of the root mean square (RMS) roughness and XRR roughness are obtained (see Fig. S1 and Table S1 in the Supplementary Information). Both methods consistently show that, with the exception of the amorphous film grown at low temperature and of the thin islands formed at 205 °C, the sample grown at 130 °C has the lowest roughness. This analysis is in agreement with the RHEED data, suggesting that the best Te film morphology is obtained at 130 °C.

To identify the preferential orientations, crystal quality, and out-of-plane lattice spacing of the deposited Te films as a function of substrate temperature, X-ray diffraction (XRD) symmetric radial scans of the six samples in the series were performed (see Fig. 2a). The sharp peaks at $\approx 2 \text{ \AA}^{-1}$, $4 \text{ \AA}^{-1}$ and $6 \text{ \AA}^{-1}$ correspond to the 111, 222, and 333 Bragg reflections of the Si substrate. Whereas no diffraction signal from the deposited material can be identified for deposition temperatures of 15 °C and 205 °C, various diffraction peaks are found for the samples deposited at 80 °C, 105 °C, 130 °C and 155 °C. The well-defined peaks located at $\approx 1.62 \text{ \AA}^{-1}$, $3.24 \text{ \AA}^{-1}$, $4.86 \text{ \AA}^{-1}$ and $6.48 \text{ \AA}^{-1}$ correspond to the $h0\bar{h}0$ Bragg diffractions with $h = 1, 2, 3, 4$, respectively. (The Miller-Bravais four-index notation is used for the hexagonal Te, whereas Miller indices are used for the cubic substrate.) These peaks are most intense for the samples grown at 105 °C and 130 °C, in which the first diffraction order also shows well defined Laue thickness oscillations.

Furthermore, peaks at $\approx 1.94 \text{ \AA}^{-1}$ and $3.88 \text{ \AA}^{-1}$ are observed, and can be ascribed to the $10\bar{1}1$ and 2022 Bragg reflections, while the peak at $\approx 3.18 \text{ \AA}^{-1}$ arises from the 0003 Bragg diffraction (marked by black arrows in Fig. 2a).

Remarkably, the substrate temperature has a clear influence on the texture of the epilayers. In fact, while for the sample grown at 80 °C these families of crystallographic planes give fairly comparable contributions, by increasing the growth temperature the $h0\bar{h}0$ reflections become dominant, suggesting that this is the preferential out-of-plane orientation of the crystals, whereas the others are almost suppressed (see the curves for substrate temperatures of 130 °C and 155 °C). The Te $h0\bar{h}0$ peaks were fitted using pseudo-Voigt functions (not shown). By comparing the full width at half maximum (FWHM) and intensity of the peaks, we conclude that the film deposited at 130 °C has the best crystal quality, which is consistent with the outcome of the morphological analysis.

In addition, a broad signal can be seen at various positions marked by light blue asterisks in Fig. 2a. This weak feature could be attributed to a very thin layer nucleated at the interface with Si, as suggested by reducing the thickness of the epilayers (see Sec. Thickness study).

The investigation of the origin of this feature was carried out by high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) measurements of the Te sample grown on Si(111) at 130 °C (see Fig. 2b, c). First, the sample was aligned along the Si $[1\bar{1}0]$ zone axis (ZA) (panel b of Fig. 2). The typical dumbbell structure of Si can be recognised (darker region in the micrograph), along with the passivation monolayer at the Si surface. Noticeably, as anticipated by the XRD characterization, between the substrate and the Te epilayer (higher contrast in the micrograph) a block composed by five atomic layers is present. Given the use of Sb as passivation layer and the peculiar structure of the block, we conclude that this interlayer is a quintuple layer of Sb_2Te_3 , formed at growth onset upon the intermixing between Te and Sb atoms at the interface. This explanation is supported by the fact that the Sb_2Te_3 layer does not cover entirely the Si surface. In line with our findings, in a recent paper by Dragoni⁴² a novel structural model of the Te-Si(111) surface, energetically more favourable, was investigated. Note that the present result hints at the possibility to realize vdW heterostructures between tellurene and other layered tellurides.

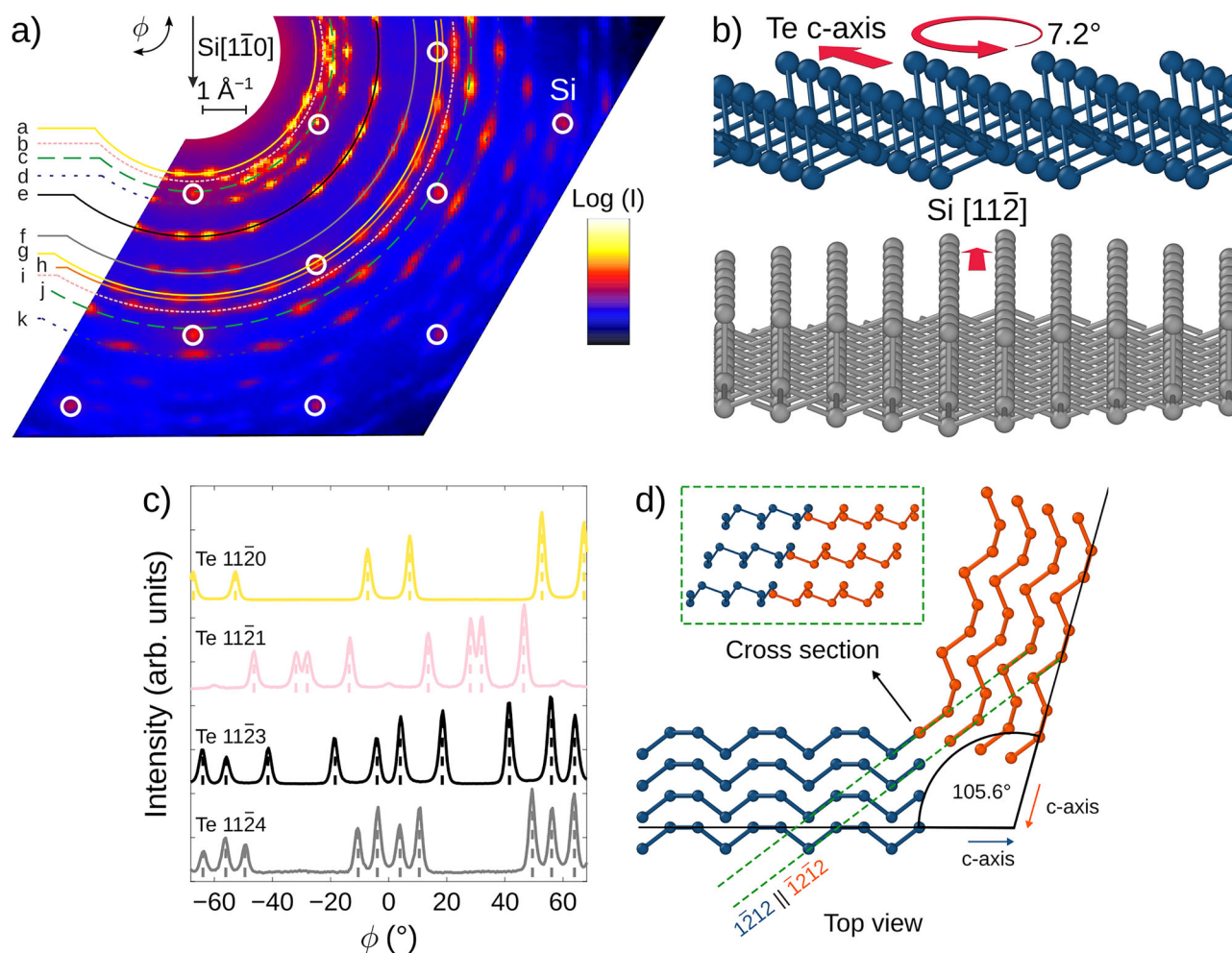


Fig. 3 | Epitaxial registry of the Te films grown on Sb-passivated Si(111). **a** GID in-plane map of the epitaxial Te film grown on Si(111) at 130 °C. The measurement covers an extended range in reciprocal space (up to $q_{\text{rad}} \approx 10 \text{ \AA}^{-1}$). Si substrate reflections, e.g. the one indicated by the black arrow originating from the $1\bar{1}0$ Bragg diffraction, are marked by white circles as reference. The Te lattice causes a very distinct pattern with several spots along a series of rings, i.e., at different q_{rad} , indicating a preferential arrangement. **b** Schematic of the orientation of one Te domain with respect to the Si substrate underneath. For better clarity, the Te c -axis is

indicated. **c** Azimuthal scans, i.e., along particular rings in the in-plane reciprocal space as graphically defined in (a). Here, the intensity profile is depicted along the ones labelled a , b , e , and f in (a), corresponding to the $11\bar{2}0$, $11\bar{2}1$, $11\bar{2}3$, and $11\bar{2}4$ Te Bragg diffractions, respectively. Vertical dashed lines indicate the calculated peak positions. **d** Atomistic representation of the twinning between Te ($1\bar{1}2$) and ($1\bar{2}1$) planes, occurring when two domains are mutually rotated by an angle of 105.6° , e.g., between the domains at $+ \delta$ and $+ 120^\circ - \delta$.

Concerning the epilayer, the layered structure of tellurium is clearly visible on top, although its atomic configuration is not well defined. This finding suggests the existence of an in-plane rotation of the epitaxial Te film with respect to the substrate underneath, causing a misalignment with the electron beam when the sample is oriented along a Si ZA. In panel c of Fig. 2, another region of the sample is shown, in which the sample was aligned with the epilayer ZA ($\approx 7^\circ$ azimuthal rotation with respect to Fig. 2b). The typical structure of the Te chains is visible, together with the high structural quality of the film, which remarkably appears to be almost defect-free within a single domain.

Grazing incidence diffraction (GID) was employed to further characterize our samples. GID provides sensitive information of in-plane lattice parameters and their azimuthal orientation in thin films, offering a powerful tool to access the epitaxial registry. This method becomes in particular surface sensitive since the X-ray beam impinges at shallow angles, i.e., close or even below the critical angle for total external reflection. Diffraction is performed on lattice planes perpendicular to the surface; consequently, GID is exclusively sensitive to in-plane information of the thin layers, complementary to the XRD radial scans discussed above. Figure 3a shows a large area GID in-plane map of the Te sample grown on Si(111) at 130 °C. White

circles indicate reflections due to the Si substrate, which arrange in a hexagonal pattern reflecting the symmetry of the (111) surface. The Te film causes a complex variety of different in-plane reflections, located at particular rings corresponding to discrete in-plane components of the scattering vector $\mathbf{q}$. Table S2 in the Supplementary Information contains a summary of these components, including their respective scattering vector length $|\mathbf{q}_{\text{rad}}|$, the measured and nominal in-plane lattice parameters $a_{\text{exp/nom}}$. Along these rings the contributions are azimuthally separated in a highly systematic way.

Interestingly, the most intense contributions (labelled a in Fig. 3(a)) are not aligned with the Si $[1\bar{1}0]$ direction but still show a narrow FWHM of $\approx 2.2^\circ$. All the observed Bragg peak positions can be reproduced by a trigonal Te crystal (with space group $P3_121/P3_221$), $a = 4.46 \text{ \AA}$, $c = 5.91 \text{ \AA}$ and a particular domain arrangement. As found by XRD radial scans, we imposed the $(10\bar{1}0)$ planes of Te to be parallel to the sample surface and varied their in-plane arrangement to obtain an agreement with the GID map. Using *xrayutilities*⁴³, we modelled the interlinked Bragg peak positions obtained for the various diffraction planes of a Te domain. To explain all the observed peak positions, we used six equivalent domains which have their a -axis direction located along the azimuths defined by Si $[1\bar{1}0] \pm \delta$, Si $[1\bar{1}0] + 120^\circ \pm \delta$, and Si $[1\bar{1}0] - 120^\circ \pm \delta$, with $\delta = 7.2^\circ$ (see also the

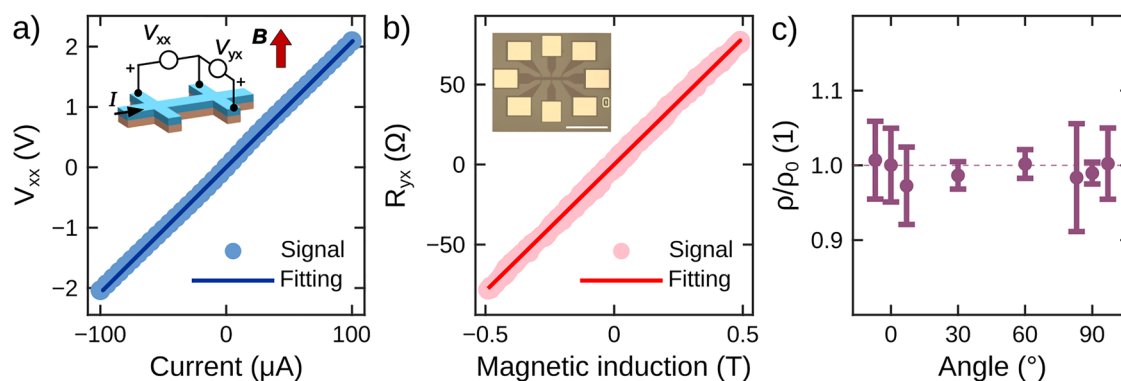


Fig. 4 | Electrical characterization of epitaxial Te on Sb-passivated Si(111).

a Longitudinal voltage V_{xx} as a function of the injected current. The sketch in the inset shows the geometry of the Hall bar and the configuration of the measurements, performed at room temperature. **b** Transverse resistance R_{yx} versus magnetic field. Inset: representative optical image of the Hall bar devices. The scale bar corresponds to 200 μm. **c** Normalized resistivity versus angle. The directions are referred to Si

[110]. Taking into account the in-plane arrangement of Te domains in the films, we measured the electrical properties also at $\pm 7^\circ$ and $90^\circ \pm 7^\circ$. For each angle, the normalized mean resistivity and standard deviation calculated for several devices with the same orientation are shown. ρ_0 is the average resistivity measured at 0° . The typical current used in both resistivity and Hall effect measurements is 100 μA.

schematic in Fig. 3b). The resulting mutual orientation of the Te Bragg peaks leads to an agreement with all observed peaks, as indicated by the vertical dashed lines in Fig. 3c (cf. The overlay of peaks in the full GID map in Fig. S2). Our model generates a pattern in which the diffraction peaks repeat every 60° , reflecting the surface symmetry of the substrate. Therefore, the presence of 60° twin domains, very typical for other telluride systems^{37,44}, would not produce additional contributions in the in-plane reciprocal space, and cannot be excluded. Moreover, a rotation angle of $105.6^\circ (=120^\circ - 2\delta)$ between the various Te domains seems to form, as this angle allows the overlap of the $(1\bar{2}12)$ and $(\bar{1}2\bar{1}2)$ lattice planes of two mutually rotated domains, as sketched in Fig. 3d. The presence of this peculiar rotation angle, as well as of 60° twin domains, is confirmed by analyzing the AFM topography image of the Te epilayer grown at 130°C (as shown in Fig. S3). Due to the limited matching of the atomic distances of Te and Si, the formation of rotated domains seems to be more favourable than aligned crystal axes. This is despite the fact that our modelling shows that a smaller coincidence lattice unit cell would result for the case of Te a -axis along Si [110], while no clear advantage is found for the rotated case (cf. Fig. S4). Therefore, the driving force to generate the observed rotated domains seems to be related to the energetic gain of the matched lattice planes in the mutually rotated domains.

Electrical and optical characterization

Following the structural characterization of the samples, the investigation of the electrical properties of epitaxial Te was carried out in analogy with previous characterization of other Te-based compounds as GeTe ⁴⁵. To this aim, a Te film with optimized quality, obtained with a substrate temperature of 130°C , was grown on high-resistivity Si(111) ($> 5000 \Omega \cdot \text{cm}$). The thickness of the epilayer (t) is 22.4 nm, as fitted from XRR measurements (not shown). Hall bar structures were patterned on the sample to measure resistivity, carrier concentration, and mobility at room temperature (RT). The Hall bars were fabricated using a combination of optical lithography and ion beam etching, achieving the structure sketched in the inset of Fig. 4a. The effective width of the fabricated Hall bars was $w = 13 \mu\text{m}$, while the distance between the two lateral contacts used to measure the longitudinal resistance ($R_{xx} = \frac{V_{xx}}{I}$) was $L = 50 \mu\text{m}$. Details about the measurements configuration are reported in Sec. Methods.

Figure 4a shows the longitudinal voltage as a function of the injected current for a representative Hall bar oriented along the Si [110] direction. The linear fit for each structure yields an average resistivity $\rho = \frac{wR_{xx}}{L} = (1.2 \pm 0.1) \times 10^4 \mu\Omega \cdot \text{cm}$. Given the remarkable agreement with optical spectroscopy measurements (see below), the contribution of the Si substrate to the electrical conduction is suggested to be negligible. In panel (b) of Fig. 4, we report the transverse resistance R_{yx} as a function of the magnetic induction field B , applied perpendicularly to the sample surface to

generate the Hall effect. The p -type character expected for this material is evident from the positive slope of $R_{yx}(B)$, and the film carrier concentration $p = (e \frac{dR_{yx}}{dB} t) = (1.8 \pm 0.1) \times 10^{18} \text{ cm}^{-3}$ is obtained⁴⁶. Combining resistivity and carrier concentration, the RT mobility of tellurium on Si(111) is obtained as $\mu = (pe\rho)^{-1} = 285 \pm 10 \text{ cm}^2/\text{V} \cdot \text{s}$.

Note that Huang et al.⁴⁷ underlined that the highest mobility values have been reported for field-effect transistors fabricated using Te flakes^{20,48–50} and nanostructures⁵¹, while the carrier mobility recorded for thin films of Te is much lower^{52–54}. Hence, the results presented here suggest the superior quality of MBE-grown 2D tellurium on Si(111), despite the presence of a non-continuous Sb_2Te_3 interlayer and of in-plane rotational domains.

Finally, Fig. 4c shows the resistivity measured along different crystallographic directions relative to Si [110] (defined as 0°), normalized to its value along the reference direction. Within the experimental error, the film resistivity does not exhibit significant in-plane anisotropy. The peculiar structure of the film, hosting multiple in-plane rotated domains in the probed volume, is likely at the origin of the overall isotropic behaviour.

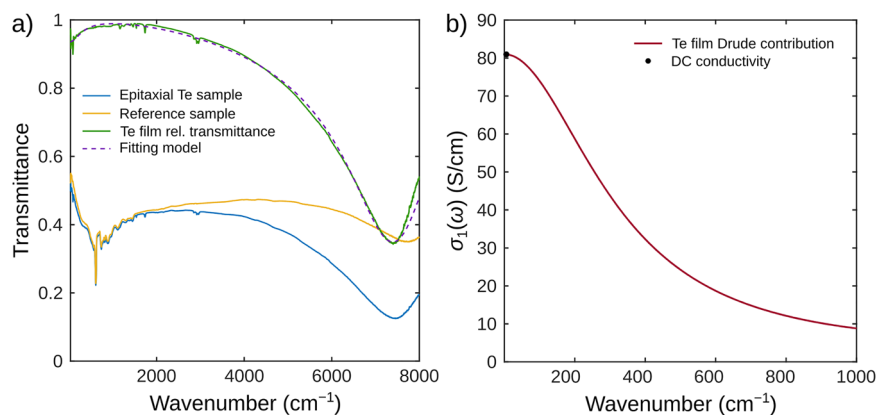
Transmittance measurements were performed on the same 22.4 nm thick Te film to gain information about the real part of the frequency-dependent optical conductivity ($\sigma_1(\omega)$) of the film, and compare its value with the direct current (DC) resistivity obtained through electrical measurements. To estimate the free charge carrier (Drude) contribution, the measurements were performed in the infrared (IR) spectral range extending from 30 cm^{-1} to 8000 cm^{-1} . Both the transmittance of the sample, a three-layer composed of a 36.5 nm $\text{ZnS}:\text{SiO}_2$ capping, a 22.4 nm Te film, and a 500 μm thick Si(111) substrate, and of a two-layer reference sample composed of a 36.5 nm $\text{ZnS}:\text{SiO}_2$ capping and a 500 μm thick Si(111) substrate were measured. The contribution of the capping layer to the transmittance was eliminated by normalizing the three-layer sample to the two-layer reference, in the framework of the thin film model, as described by the equation below^{55,56}:

$$\frac{T_{\text{sub+Te+cap}}}{T_{\text{sub+cap}}} = T_{\text{Te}} \quad (1)$$

This ratio allows us to retrieve the Te film relative transmittance, getting rid of the capping and substrate optical contributions. The contribution of the Sb_2Te_3 interface layer is negligible in the optical characterization due to its extremely small thickness.

Figure 5a shows the transmittance of the three-layer sample (light-blue curve), including the $\text{ZnS}:\text{SiO}_2$ capping, the 22.4 nm Te film, and the Si substrate, the transmittance of the two-layer sample (yellow curve), including the $\text{ZnS}:\text{SiO}_2$ capping and the Si substrate only, and the ratio between the light-blue and the yellow curves (green curve). Both the

Fig. 5 | Optical characterization of epitaxial Te on Sb-passivated Si(111). **a** Experimental transmittance in the IR spectral range, from 30 cm^{-1} to 8000 cm^{-1} , of the three-layer sample (light-blue curve), including the $\text{ZnS}:\text{SiO}_2$ capping, the 22.4 nm Te film, and the Si substrate, and of the two-layer sample (yellow curve), including the $\text{ZnS}:\text{SiO}_2$ capping and the Si substrate. The relative transmittance (light-blue curve over the yellow curve) is represented by the green curve in (a). The Drude-Lorentz fitting of the relative transmittance, as obtained by the RefFIT program⁶², is represented by a purple dashed curve. **b** Real part of the optical conductivity $\sigma_1(\omega)$ of the 22.4 nm Te film, expressed in S/cm , from 30 cm^{-1} to 1000 cm^{-1} . In this spectral range, the main contribution to $\sigma_1(\omega)$ comes from the Drude conductivity, whose extrapolation at zero frequency (highlighted by a black dot) provides a DC resistivity of $\approx 1.2 \times 10^4 \mu\Omega \cdot \text{cm}$, in very good agreement with the electrical data (see the main text). The uncertainty of the estimation is visualized by the dimension of the symbol.



transmittance of the three-layer and two-layer samples exhibit several dips from 600 cm^{-1} to 1200 cm^{-1} , associated with Si and Si oxides phonon modes, such as the two phonon Si-Si lattice vibration at 610 cm^{-1} ^{57,58} and the Si-O-Si stretching vibration at 1107 cm^{-1} ⁵⁹.

The relative transmittance of the Te film can be completely modelled (see dashed purple line in Fig. 5a), with a Drude contribution with plasma frequency $\omega_p = 1260\text{ cm}^{-1}$ and damping rate $\gamma = 320\text{ cm}^{-1}$, and a Lorentz oscillator centred at $\omega_0 = 7500\text{ cm}^{-1}$, with an oscillator strength $S = 11640\text{ cm}^{-1}$ and a damping rate $\gamma = 1460\text{ cm}^{-1}$. These two optical contributions well describe the whole transmittance between 30 and 8000 cm^{-1} . In particular, the Lorentz oscillator can be ascribed to an interband transition of tellurium, as already observed in density functional theory calculations^{60,61}. Fig. 5b reports the real part of the optical conductivity $\sigma_1(\omega)$ of the Te film, from 30 cm^{-1} to 1000 cm^{-1} , obtained from the fitting procedure with the RefFIT software⁶². In this limited spectral range, only the effect of the Drude contribution is visible, determining an increase of $\sigma_1(\omega)$ for decreasing frequency. An estimation of $\sigma_{DC} \approx 81\text{ S/cm}$ was obtained by looking at the optical conductivity value at $\omega = 30\text{ cm}^{-1}$, corresponding to a resistivity $\rho_{opt} \approx 1.2 \times 10^4 \mu\Omega \cdot \text{cm}$, which is in very good agreement with the one obtained through RT electrical measurements.

It is well known that the plasma frequency ω_p depends on the ratio between the charge density p and the effective band mass m^* . More specifically, it can be written as $\omega_p^2 = \frac{pe^2}{\epsilon_0 m^*}$. From the measured charge density data (see above) and $\omega_p = 1260\text{ cm}^{-1}$, an effective mass $m^* = 0.1m$ is obtained (m being the free electron rest mass). This value is in very good agreement with that calculated in ref.⁶³.

Thickness study

In the following, we present the analysis of epitaxial Te on Si(111) as a function of film thickness. Figure 6a–c show the AFM images acquired for the as-deposited samples having decreasing thickness. The morphology of the reference sample (thickness of $\approx 19\text{ nm}$) is overall reproduced. In fact, we note the presence of thin elongated crystals parallel to the substrate, which appear oriented in-plane, and a few triangle-shaped islands, which are divided into three areas and are also oriented. Remarkably, as the thickness of the film decreases, the lateral size of the Te domains also shrinks, ranging from $\approx 200\text{ nm}$ for the thickest sample to $\approx 130\text{ nm}$ for the thinnest one. As a result, the coverage is not complete, and the RMS roughness of the films increases with reducing thickness, as given in Table 1.

XRD radial scans of the samples with decreasing thickness show that the Bragg peaks identified for epitaxial Te on Si(111) remain the same in the thickness range under consideration (Fig. 6d). The peak width is broader for

thinner films, since it is correlated with the inverse of the material's thickness. Still, Laue thickness fringes are evident for all samples, testifying to the high epitaxial quality of the series. Remarkably, a sizable shift of the $10\bar{1}0$ Bragg peak towards lower values of Q_z occurs when decreasing the film thickness, as given in Table 1. In particular, a corresponding increase of the lattice plane spacing $d_{10\bar{1}0}$ of $\approx 0.8\%$ is recorded for the thinnest sample in the series. Hence, the out-of-plane separation between the Te layers clearly relates to the number N of layers stacked in the films.

Raman spectroscopy was demonstrated to be a suitable method to investigate the thickness-dependent vibrational properties of Te^{20,23,64}. Fig. 6e displays RT Raman spectra of epitaxial Te with decreasing thickness. The measurements exhibit for all films the three Raman-active phonon modes expected for Te, one A_1 mode ($\approx 125\text{ cm}^{-1}$) and two E modes (E_1 at $\approx 95\text{ cm}^{-1}$ and E_2 at $\approx 140\text{ cm}^{-1}$)²⁰. Moreover, the longitudinal optical E_1 -LO mode ($\approx 105\text{ cm}^{-1}$) is present together with the transverse optical E_1 -TO mode, which is the one commonly observed in bulk Te. Regarding the A_1 and E_2 modes, a blueshift with decreasing film thickness occurs. In addition, the splitting between the two E_1 modes decreases for the thinnest sample in the series. These observations resemble the behaviour reported in ref.²⁰, where the appearance of the E_1 -LO mode was explained by a thickness-dependent variation in the relative strengths of the contributing Raman scattering mechanisms. Remarkably, the spectrum recorded for the $\approx 4\text{ nm}$ thick Te film shows well-defined vibrations (see the inset in Fig. 6e), proving once again the high crystal quality of our epitaxial Te.

The thickness-dependent variation of the out-of-plane lattice spacing, unveiled by XRD measurements, allows us to tentatively interpret the observed blueshifts. In fact, the increase of the $d_{10\bar{1}0}$ for decreasing N would correspond to an in-plane compressive strain, where the Te c -axis lays. According to ref.⁶⁴, a compression along the c -axis induces a comparable hardening of both the A_1 and E_2 modes, while for a -axis strain, the Raman peak shifts were found to be negligible. Still, the shift measured for the A_1 mode ($+3.5\text{ cm}^{-1}$) is much larger than that of the E_2 mode ($+1\text{ cm}^{-1}$). Therefore, mechanical strain alone seems to provide only a partial explanation for the results found experimentally. Indeed, thickness-dependent phonon mode shifts and splittings were predicted by theory; however, only for the few-layer regime (thickness below six layers)²⁶.

Discussion

In this study, we presented the growth and characterization of epitaxial Te on $\text{Si}(111)-(\sqrt{3} \times \sqrt{3})R30^\circ\text{-Sb}$, targeting the fabrication of large-area high-quality films. RHEED gave an immediate insight on the presence of 2D-films or 3D-islands, which was confirmed by AFM topography

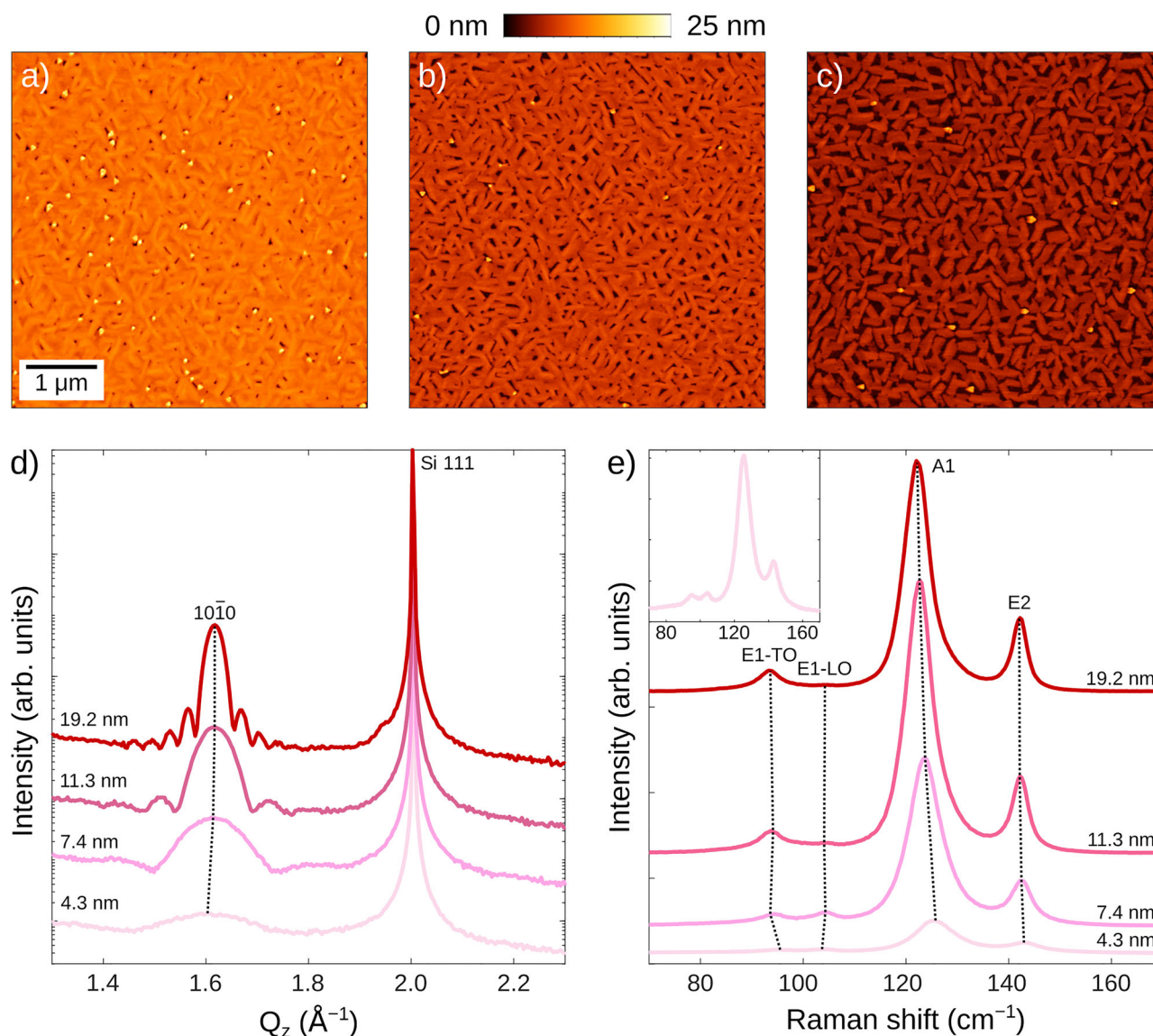


Fig. 6 | Study of epitaxial Te on Sb-passivated Si(111) as a function of the epilayer thickness. The samples were grown at 130 °C. AFM topography images of Te films with thickness equal to **a** ≈ 11 nm, **b** ≈ 7 nm and **c** ≈ 4 nm. All the images have the same size (see scale bar in **(a)**). **d** XRD radial scans of the samples with decreasing film thickness. The curves are plotted in logarithmic scale and vertically shifted for clarity. The dotted line highlights the variation of the Te 1010 peak position with film

thickness. **e** RT Raman spectroscopy measurements of the samples with decreasing thickness. The curves are vertically shifted for clarity. The dotted lines indicate the position of the Te active Raman modes. Statistical errors for the positions of the peaks are below 0.8 cm^{-1} . For better clarity, the spectrum of the thinnest sample in the series is reported separately in the inset.

Table 1 | Morphological and structural analysis of epitaxial Te films as a function of epilayer thickness

Growth time [min]	RMS roughness [nm]	Thickness [nm]	XRR roughness [nm]	Q_z [$\text{\AA}^{-1}$]	N layers [-]
90	1.3	19.2	1.0	1.617	49
45	1.3	11.3	1.4	1.617	31
22	1.6	7.4	1.7	1.614	19
11	1.7	4.3	1.9	1.603	12

The RMS roughness was obtained from AFM topography images, the thickness and roughness values were fitted from the XRR measurements (see Supplementary Information), and the Q_z value of the (1010) peak was fitted from the XRD radial scans. Statistical errors for the AFM-measured RMS roughness are less than 0.1 nm. Statistical errors for the film thickness and roughness estimated by XRR are below 0.3 nm. Statistical errors for the Q_z position of the Te (1010) peak are below $0.001 \text{ \AA}^{-1}$. The number N of layers (i.e., Te chains) of the Te samples with decreasing thickness was calculated from the film thickness and the lattice plane spacing $d_{1010} = \frac{a}{Q_z}$.

measurements. On the other hand, XRD characterization provided information regarding the crystal quality. The onset temperature for the crystallization of tellurium is ≈ 80 °C. At higher temperatures, we obtained the coexistence of in-plane-oriented crystals and islands, which affect the film roughness. We found that the optimal temperature window for the 2D growth of tellurium films on Sb-passivated Si(111) ranges from ≈ 105 °C to ≈ 155 °C. Moreover, the preferential growth plane is the Te(1010), corresponding to the Te chains parallel to the Si surface. The high structural quality of the epitaxial Te films was shown by HAADF-STEM measurements, which also allowed the identification of an interlayer of Sb_2Te_3 formed at the interface with silicon. This result is particularly relevant, since it suggests the possibility of realizing vdW heterostructures between tellurene and other layered tellurides. The GID measurements revealed a very peculiar in-plane arrangement, with rotational domains resulting from the formation of twin domains, calling for further investigations to eventually attain a single-domain epilayer. Nevertheless, the RT electrical

characterization of a ≈ 22 nm Te film, paired with IR optical measurements, unveiled the high hole mobility of the MBE-grown material, displaying an unprecedented value for thin films of 285 ± 10 cm²/V·s. This highlights the effectiveness of vdW epitaxy on Sb-passivated Si(111). We also demonstrated significant thickness-dependent differences in epitaxial Te. The reduction of the growth time results not only in thinner films, but also in a lower coverage of the substrate. This is due to the lateral growth of the Te domains over time. Moreover, a thickness-dependent change of the out-of-plane lattice spacing was measured by XRD, which we correlate to the evolution of the Raman vibrational modes as a function of the epilayer thickness. The present results constitute a significant step forward in the control of the synthesis of 2D tellurium by MBE, paving the way for its integration on silicon and the engineering of novel vdW heterostructures.

Methods

Epitaxial growth

The growth experiments were performed by MBE on Sb-passivated Si(111) substrates. The 2×2 cm² p-type Si(111) substrates (B-doped, resistivity $1\text{--}20$ $\Omega \cdot \text{cm}$) were cleaned using a standard procedure, as described by Wang et al.⁴⁴, and introduced in the MBE system. The Si(111)–($\sqrt{3} \times \sqrt{3}$)R30°–Sb surface was prepared in situ prior to the deposition of Te³⁴. The surface reconstruction process was monitored by RHEED. The substrate temperature was measured by a non-contact thermocouple. During the growth, the cell temperature was set to 334 °C and kept constant. The corresponding deposition rate at RT was ≈ 0.21 nm/min. The samples were capped in situ by a ≈ 30 nm ZnS:SiO₂ layer, sputtered at RT, to prevent oxidation in air. We have no indication of epilayer damage caused by sputtering³⁶, while intermixing of the capping layer with other Te-based materials occurs above 200 °C^{65,66}.

Atomic force microscopy

The surface morphology was probed by AFM using a Veeco Nanoscope III in tapping mode. For a measured surface area of 5×5 μm^2 (Fig. 1 and 6) and 2.5×2.5 μm^2 (Fig. S3), the scan rates were 0.2 Hz and 0.4 Hz, respectively. Reference measurements with and without the cap confirmed that the topography captured for capped samples well correlates with that of the underlying layer.

X-ray diffraction and reflectivity

All Te films were characterized by XRD and XRR using a PANalytical X'Pert PRO Materials Research Diffractometer system with a Ge(220) hybrid monochromator, employing a Cu K α_1 ($\lambda = 1.5406$ Å) X-ray radiation. A PixCel^{1D} detector was used in combination with a parallel plate collimator. Slits with $1/2^\circ$ and $1/16^\circ$ apertures were mounted at the source for XRD and XRR, respectively.

Scanning transmission electron microscopy

The crystalline structure and the morphology at atomic resolution were investigated by STEM. Cross-sectional lamellae were prepared by focused ion beam (FIB) milling in a Thermo Scientific™ Helios™ 5 UC DualBeam system using 30 keV Ga⁺ ions, followed by low-energy (2 keV Ga⁺) polishing to minimize FIB-induced amorphization. The analyses were performed by using a JEOL ARM200F Cs-corrected microscope, which is equipped with a cold-field emission gun and operating at 200 keV. Micrographs were acquired in Z-contrast mode by HAADF, with a resolution of about 0.68 Å. In each area, multiple scans were acquired and processed using SmartAlign tools, rotating 90° each scan. SmartAlign reconstruction was performed by means of non-rigid alignment^{67,68} in order to reduce any possible artifact due to sample drift or noise and to minimize the beam dwell time.

Grazing incidence diffraction

The in-plane orientations of the films were surveyed by synchrotron GID. The GID experiments were performed at the SpLine beamline BM25 at the European Synchrotron Radiation Facility (ESRF) in Grenoble (France). An incidence angle of the illuminating X-rays of 0.2° effectively suppresses

diffuse scattering by the substrate, and thus makes this method highly surface sensitive. A primary X-ray photon energy of 17 keV ($\lambda = 0.7293$ Å) was used, which facilitates the inspection of a comparatively large area in reciprocal space. This is particularly important since we intended to probe multiple reflections of the same lattice plane family.

Fabrication of the Hall bars

Hall bar structures were defined by standard optical lithography. The Hall bar was first patterned and etched by ion beam etching down to the Si substrate. Electrical pads were subsequently defined by inverse lithography, followed by selective removal of the ZnS:SiO₂ capping layer using diluted hydrofluoric acid. Ohmic contacts were then formed by deposition of Ti (10 nm)/Au⁶⁹ by Ar sputtering. The approach was validated on similar systems (GeTe⁴⁵ and Ge-rich (GeTe)_m(Sb₂Te₃)_n³⁶).

Electrical characterization

A direct current was injected in the Hall bars using a Keithley 6221 current source. The resulting voltages were acquired with a Keithley 2182A nanovoltmeter. The longitudinal voltage (V_{xx}) along the current path was measured using two contacts on the same side of the Hall bar, while the transverse voltage (V_{xy}) was probed using two contacts on opposite sides, as sketched in the inset of Fig. 4(a). The corresponding longitudinal (R_{xx}) and transverse (R_{yx}) resistances were obtained as the ratio between the measured voltages and the injected current. Note that an offset was subtracted to R_{yx} , accounting for the non-idealities of the electrodes (such as the geometrical misalignment). The uniform magnetic induction field (B) applied to the samples was generated using a GMW Associates electromagnet. The ohmic behaviour of the contacts was verified by both two-probe and four-probe I–V measurements, which showed linear and symmetric characteristics around zero bias.

Optical measurements

Transmittance measurements were performed in the IR spectral range for both the Si substrate and the Te film sample. The RefFIT software⁶² was employed to obtain the frequency-dependent behaviour of the real part of the optical conductivity $\sigma_1(\omega)$ of the 22.4 nm Te film. Measurements were carried out under vacuum conditions with a Bruker Vertex 70v Fourier Transform interferometer, using a standard sintered U-shaped SiC source (globar), a Silicon beamsplitter, and a DLaTGS detector, which allows covering a spectral window extending from 30 cm^{-1} to 8000 cm^{-1} . The spectral resolution was set to 4 cm^{-1} .

Raman spectroscopy

The vibrational properties of the samples were analyzed using RT Raman spectra acquired in backscattering $z(y,xy)\text{--}z$ geometry. The samples were excited with the 632.8 nm line of a He-Ne laser, and the scattered light was analyzed using a spectrometer equipped with a liquid N₂-cooled charge-coupled-device (CCD) detector. The emission was focused by a microscope objective with 0.9 numerical aperture, and the same objective was used for the collection of the signal. The spatial resolution achieved is 0.7 cm^{-1} , and a notch filter suppressed the stray light.

Data availability

The datasets generated and/or analyzed during the current study are not publicly available due to ongoing research, but are available from the corresponding author on reasonable request.

Received: 6 February 2026; Accepted: 22 May 2026;

Published online: 13 June 2026

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Acknowledgements

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 824957 and grant agreement No 823717 – ESTEEM3. S.C. acknowledges support by the European Union – Next Generation EU, Missione 4 Componente 2, investimento 1.1 CUP H53D23000880001 (project PRIN 2022 – B-NGO, ID 2022ZB4EFZ). C.R. acknowledges the project SORBET – ID 2022ZY8HJY – CUP D53D23002500006 for the financial support through the National Recovery and Resilience Plan (NRRP), funded by the European Union – Next

Generation EU – “PNRR – M4C2, investimento 1.1” – Fondo PRIN 2022. S.L. acknowledges support by the European Union under the Italian National Recovery and Resilience Plan (NRRP) of Next Generation EU partnership PE0000023-NQSTI. D.K. acknowledges support by the Czech Science Foundation (project No 22-22000M) and the Czech Academy of Sciences (project No LQ100102201). A.M.M. acknowledges financial support by the Italian project BEYOND NANO Upgrade (CUP G66J17000350007). The authors acknowledge the provision of beamtime under the project HC-4972 at the European Synchrotron Radiation Facility (ESRF) in Grenoble and highly appreciate the support of Dr. Juan Rubio-Zuazo. The fabrication of the Hall bar devices was carried out at PoliFAB, the micro and nano fabrication facility of Politecnico di Milano. The authors thank S. Behnke and C. Stemmler for technical support at the molecular beam epitaxy system and M. Garagiola for graphical support for Fig. 3(b).

Author contributions

S.C. designed the experiments. S.C. performed the MBE growths and RHEED characterization and analysis, together with S.I. S.I. carried out AFM, XRD, and XRR characterization, supported in the analysis by S.C. and D.K. A.M.M. performed the STEM measurements. GID measurements were done by M.H. The analysis and simulation of the GID data were carried out by S.C., D.K. and O.A.E.K. The electrical characterization was performed by G.G., F.F., and C.R. The optical spectroscopy measurements were carried out by M.C.P., I.M., S.M. and S.L. S.I. performed the Raman spectroscopy measurements with the support of M.R. All authors discussed the results. S.C. and S.I. wrote the main manuscript text and prepared the figures, with the help and contributions from all co-authors.

Funding

Open Access funding enabled and organized by Projekt DEAL.

Competing interests

The authors declare no competing interests.

Additional information

Supplementary information The online version contains supplementary material available at <https://doi.org/10.1038/s41699-026-00711-3>.

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