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Structural properties of Bi$_2$Te$_3$ topological insulator thin films grown by molecular beam epitaxy on (111) BaF$_2$ substrates

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Structural properties of topological insulator bismuth telluride films grown epitaxially on (111) BaF$_2$ with a fixed Bi$_2$Te$_3$ beam flux were systematically investigated as a function of substrate temperature and additional Te flux. A layer-by-layer growth mode is observed since the early stages of epitaxy and remains throughout the whole deposition. Composition of the epitaxial films produced here stays between Bi$_2$Te$_3$ and Bi$_4$Te$_5$, as determined from the comparison of the measured x-ray diffraction curves with calculations. The substrate temperature region, where the growth rate remains constant, is found to be the most appropriate to obtain ordered Bi$_2$Te$_3$ films. Line width of the $L = 18$ Bi$_2$Te$_3$ diffraction peaks as low as 140 arcsec was obtained, indicating high crystalline quality. Twinning domains density rises with increasing growth temperature and reducing Te extra flux. X-ray reflectivity curves of pure Bi$_2$Te$_3$ films with thickness from 165 to 8 nm exhibited well defined interference fringes, evidencing homogeneous layers with smooth surface. Our results demonstrate that Bi$_2$Te$_3$ films with very well controlled structural parameters can be obtained. High structural quality Bi$_2$Te$_3$ films as thin as only eight quintuple layers grown here are promising candidates for intrinsic topological insulator. Published by AIP Publishing.

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INTRODUCTION

Bismuth telluride has long been known to have a high room-temperature figure of merit (ZT) among the thermoelectric materials.$^{1-5}$ Recently, it has attracted intense interest due to its pronounced features as a member of a new class of materials called three-dimensional topological insulators, which are insulating in the bulk and exhibit gapless metallic surface states with linear energy-momentum dispersion shaped like a Dirac cone. Due to the strong spin-orbit coupling, these conducting surface states have electron momentum locked to the spin orientation and are protected from scattering mechanisms by time reversal symmetry. Consequently, high-mobility spin polarized surface currents can be produced without external magnetic fields, offering possibilities to new applications in spintronics or quantum computing.$^4$–$^6$

The topological order in V-VI compounds has been theoretically predicted$^7$–$^9$ and the existence of the metallic surface states has been confirmed by angle resolved photoelectron spectroscopy (ARPES) experiments in bulk single crystals of Bi$_2$Se$_3$, Bi$_2$Te$_3$, $^{10}$ Bi$_2$Se$_3$, $^{11,12}$ Sb$_2$Te$_3$, and Bi$_2$Te$_3$. $^{13,14}$ ARPES has demonstrated to be a powerful technique to characterize the conducting surface states, providing the experimental signature of a three-dimensional topological insulator. The bulk samples are cleaved inside the vacuum chamber in order to obtain a fresh surface for ARPES and, even in ultra high vacuum (UHV) conditions, aging effects have been reported. $^{15,16}$ The oxide layer formed by air exposure on the V-VI crystal surface degrades the metallic states.

In case of Bi$_2$Te$_3$, an oxide layer of 1 nm is formed after around 1 day at room conditions.$^{16,17}$ The contribution of the Dirac surface states to electrical conductivity measurements is hindered by bulk conduction due to free carriers produced by crystalline defects like vacancies and anti-sites in the V-VI alloys. Besides surface oxidation, this bulk conduction hampers electrical measurements of the metallic surface states in three-dimensional topological insulators. Counter doping, like Ca for Bi$_2$Se$_3$, $^{11}$ and Sn for Bi$_2$Te$_3$, $^{14}$ has been used to move the Fermi level inside the band gap to obtain insulating bulk samples. However, high doping affects carrier transport and conduction only through topological surface states has shown instabilities in time scale of hours during ARPES experiments. $^{18}$ In order to obtain intrinsic topological insulators, molecular beam epitaxy (MBE) has been applied to grow bismuth chalcogenide layers. Due to the control at the atomic level and reproducibility of the MBE technique, high structural quality bismuth chalcogenide films are obtained, where the bulk conduction is expected to be suppressed.

Epitaxial films of bismuth telluride have been grown by MBE on different types of substrates like (111) Si, (111) GaAs, and (0001) Al$_2$O$_3$. $^{19-22}$ In case of the Bi$_2$Te$_3$ epitaxy on (111) Si, a small lattice mismatch near to 1% is found if a supercell of both materials is considered, which enables the growth of high crystalline quality films. $^{19}$ Despite the large lattice mismatch to Bi$_2$Te$_3$ of 8.7% for both GaAs and Al$_2$O$_3$, epitaxial layers with relatively good structural quality have been achieved on these substrates as van der Waals bonds inside Bi$_2$Te$_3$ crystal structure accommodate the in-plane differences in lattice parameters. $^{22}$ Due to the small lattice mismatch of only 0.04% to Bi$_2$Te$_3$, (111) BaF$_2$ has...
been shown to be an appropriate substrate to grow bismuth telluride. The films can be grown by MBE using either separated Bi and Te solid sources or a Bi$_2$Te$_3$ compound effusion source and additional Te cell. Depending on the growth parameters, other phases of Bi$_2$Te$_3$ compounds can be obtained and the conditions to achieve single phase Bi$_2$Te$_3$ are located in a narrow window. Most recently, intrinsic conduction through topological surface states has been reported for very thin (10–50 nm) insulating Bi$_2$Te$_3$ epitaxial films, as demonstrated by ARPES and four-point probe conductivity measurements performed at room temperature in the same UHV system. However, conduction through Dirac surface states vanishes after air exposure in time scale of minutes, indicating that a capping layer is essential to protect topological state and explore these new features in room conditions. Determination of optimum growth parameters to obtain high-quality Bi$_2$Te$_3$ epitaxial layers with well-controlled structural properties is therefore crucial for practical applications.

Contrary to conventional zinc-blend and rock-salt semiconductor compounds, bismuth telluride can exist in different Bi$_2$Te$_3$ phases, depending on the growth conditions. The Bi$_2$Te$_3$ compounds constitute a homologous series of crystal structures derived from an ordered stacking of Te$_1$-Bi-Te$_2$-Bi-Te$_1$ quintuple layers along [0001] direction. The entire series can be written in terms of the Te deficit $\delta$ as Bi$_2$Te$_{3+\delta}$, where $\delta = 0$ M/(M+N). Except in the limiting cases, $\delta = 0$ and $\delta = 3$, the addition of Bi-Bi bilayers in between the quintuple layers during molecular beam epitaxial growth does not occur uniformly, but as a statistical distribution where the grown films can be considered as a random one-dimensional Bi$_2$Te$_3$ alloy rather than as an ordered homologous (Bi$_2$)$_N$(Bi$_2$Te$_3$)$_N$ structure.

Like other V-VI compounds, Bi$_2$Te$_3$ crystallizes in a tetradymite-type structure shown in Figure 1(a). The unit cell of this hexagonal lattice is described by stacking of three Te$_1$-Bi-Te$_2$-Bi-Te$_1$ quintuple layers along c-direction. Inside the QL, the neighboring Bi-Te atoms are ionic bonded and the adjacent QLs are van der Waals coupled to each other by the facing Te$_1$-Te$_1$ layers. This van der Waals gap gives the structure an anisotropic character similar to other layered materials. The lattice parameter along [0001] direction of the Bi$_2$Te$_3$ bulk material is $c = 30.474$ Å. Figure 2(b) shows the (0001) Bi$_2$Te$_3$ surface with the three topmost atomic layers, which form a hexagonal lattice with an in-plane lattice parameter $a = 4.382$ Å. The atomic rows along the symmetry azimuths [1120] and [1010], rotated by $30^\circ$ to each other, are separated by $a/2, a/3$ and $a/2$, respectively. As the in-plane lattice constant of the (111) BaF$_2$ surface $d_{BaF2} = d_{BaF2}/\sqrt{2} = 4.384$ Å is almost equal to the hexagonal lattice constant of Bi$_2$Te$_3$, the small lattice mismatch of only 0.04% between both materials makes (111) BaF$_2$ a suitable substrate for the epitaxy of bismuth telluride. In this epitaxy, the (0001) Bi$_2$Te$_3$ hexagonal planes are parallel to the (111) cubic BaF$_2$ planes and the [110] azimuth of the substrate cubic lattice is parallel to the [1120] azimuth of the film hexagonal lattice.

This work presents a systematic investigation of the structural properties of bismuth telluride thin films grown by molecular beam epitaxy on (111) BaF$_2$ substrates, using a nominal stoichiometric Bi$_2$Te$_3$ solid source and additional
Te cells. The growth conditions were varied in a wide range of substrate temperature and Te/Bi₂Te₃ beam flux ratio. The epitaxial films were characterized in situ by reflection high-energy electron diffraction (RHEED) and ex situ by high-resolution x-ray diffraction, x-ray reflectometry, and field-emission gun microscopy.

EXPERIMENTAL

Bismuth telluride epitaxial films were grown on freshly cleaved (111) BaF₂ substrates, from Korth Kristalle GmbH, in a Riber 32P molecular beam epitaxial system using an effusion cell charged with nominal stoichiometric Bi₂Te₃ solid source and two additional Te cells to offer the extra Te flux. The Bi₂Te₃ charge was synthesized in our laboratory from commercially available Bi (99.999%) and Te (99.9999%) elemental materials. For all growths, four thin substrate slices were fixed with an eutectic indium-gallium alloy to the molybdenum substrate holder. The beam fluxes were monitored by a Bayer-Alpert ion gauge moved to the substrate position before and after each growth. The beam equivalent pressure (BEP) of the Bi₂Te₃ and Te fluxes was determined. The BaF₂ substrates were pre-heated at 300°C for 10 min before starting the growth. The background pressure of the Bi₂Te₃ cell was kept constant at around 0.2 Å/s and is only determined by the Bi₂Te₃ beam flux.

To monitor in situ the film surface during growth, reflection high-energy electron diffraction (RHEED) patterns were acquired using a 35 keV RHEED system equipped with a CCD camera in front of the fluorescent screen and a data acquisition setup. The kSA 400 software package was used to analyze the recorded RHEED movies.

X-ray diffraction measurements were performed in a PANalytical X’Pert MRD high-resolution x-ray diffractometer equipped with a (220) Ge four-crystal monochromator adjusted for Cu Kα₁ radiation and an open detector with acceptance of 1°. For the twinning characterization, the same configuration of the x-ray diffractometer is used to perform azimuthal scans for a fixed Bi₂Te₃ azimuths during RHEED measurements are gyrated by 30° in relation to those shown in Figure 2(b). All RHEED patterns were measured with an energy of 16 keV.

RESULTS AND DISCUSSION

Molecular beam epitaxy

For this work, a series of samples was produced with the beam equivalent pressure of the Bi₂Te₃ cell kept constant at 5 x 10⁻⁷ Torr and varying the substrate temperature between 180°C and 310°C and the Te/Bi₂Te₃ flux ratio Φₘ from 0 to 2. The thickness of the films was measured by interference fringes in x-ray reflectivity curves and/or by field-emission gun microscopy cross section images. The growth time for all samples was 2 h, except for the series produced to x-ray reflectivity analysis.

The behavior of the growth rate as a function of substrate temperature for different Φₘ can be divided in three regions as shown in Figure 3. At low substrate temperatures near 180°C, where the re-evaporation rate of Te is too low, all extra Te atoms practically stick to the surface, leading to higher growth rates up to 0.9 Å/s. As the temperature rises to 220°C in region A, the Te re-evaporation rate increases and the growth rate resembles the sticking coefficient curve of tellurium.²⁸ In the intermediate region B, for substrate temperatures between 220°C and 280°C, the exceeding Te atoms have sufficient energy to re-evaporate and the growth dynamics allows the Bi and Te atoms to accommodate in their sites. Therefore, the growth rate remains almost constant at around 0.2 Å/s and is only determined by the Bi₂Te₃ cell temperature. For substrate temperatures higher than 280°C (region C), the growth rates start to decrease and vanish at Tₜₛₐₜ > 300°C for Φₘ = 0 and at Tₜₛₐₜ > 310°C for Φₘ = 1, due to the decrease of Bi sticking coefficient.²⁸ Contrary to region B, where the growth rate is independent of Φₘ, the reduction of the growth rates in region C is more pronounced for lower Φₘ.

Reflection high energy electron diffraction

The reciprocal of the hexagonal surface lattice shown in Figure 2(b) is also a hexagonal lattice rotated by 30° with a lattice parameter of 4π/a₀/3. In this sense, the symmetry of the Bi₂Te₃ crystal appears the same during RHEED measurements. In Figure 4(a) shows the streaky RHEED pattern of the BaF₂ substrate along the [101] ₀ C, 280 C in region C, the growth rates start to reduce due to the decreasing Bi sticking coefficient and vanish above a certain temperature depending on Φₘ. All films were grown during 2 h.
The RHEED patterns along the [2110] azimuth of the (0001) Bi₂Te₃ film after 0.2 QL, 1 QL, and 10 QLS of deposition are shown in Figures 4(b)–4(d), respectively, for a sample grown at substrate temperature of 240 °C and flux ratio \( \Phi_R = 1 \). The streaks of the Bi₂Te₃ layer pattern start to be formed already with 0.2 QL of deposition. They become more elongated with 1 QL of coverage and, for a thickness of 10 QLS, the streaky RHEED pattern is completely formed with clearly defined Kikuchi lines. RHEED images with well defined streaks confirm the layer-by-layer growth mode since the defined Kikuchi lines. RHEED images with well defined streaky RHEED pattern is completely formed with clearly defined Kikuchi lines.

For increasing substrate temperature at a fixed flux ratio, the (000L) x-ray diffraction peaks shift, become broader, or even split. This behavior is more pronounced for the Bi₂Te₃ peaks with \( L = 18 \) and \( L = 21 \) and the expanded views of the diffraction curves around these maxima are shown in Figures 5(c) and 5(d), respectively. As \( T_{\text{SUB}} \) rises from 220 °C to 300 °C for \( \Phi_R = 1 \), the (00018) Bi₂Te₃ peak slightly shifts to lower \( Q_z \) in direction to the peak position of the (00014) BiTe phase (red vertical line in Figure 5(c)), while the (00021) Bi₂Te₃ Bragg peak moves to higher \( Q_z \) values towards the (00017) BiTe peak (red vertical line in Figure 5(d)), tending to Bi-rich phases. At lower substrate temperatures (\( T_{\text{SUB}} < 250^\circ \text{C} \)), the x-ray diffraction curves do not present a significant change as a function of flux ratio. On the other hand, for higher substrate temperatures, the effect of the flux ratio variation on the x-ray curves is easily noticed. The influence of the extra Te flux is illustrated in Figures 5(e) and 5(f), where the expanded views in the vicinity of \( L = 18 \) and 21 Bi₂Te₃ peaks are represented. The large Te desorption at the elevated \( T_{\text{SUB}} \) of 280 °C is compensated by increasing \( \Phi_R \) and plain Bi₂Te₃ phase is recovered for \( \Phi_R = 2 \).

It is important to stress out that the Bi₅Te₇ films grown in this work on (111) BaF₂ substrates, using a nominal Bi₂Te₃ solid source and extra Te cell, never presented pure BiTe phase even without offering additional Te flux (\( \Phi_R = 0 \)) and at the highest \( T_{\text{SUB}} = 300^\circ \text{C} \) (for \( T_{\text{SUB}} > 300^\circ \text{C} \), no deposition is observed at all). These results are different from previously published,\(^{24,25}\) where Bi₅Te₇ epitaxial film with composition ranging from BiTe to Bi₂Te₃ was obtained by varying only the exceeding Te flux. This fact can be explained by a variation in the stoichiometry of the Bi₂Te₃ solid sources used in the different MBE machines.

To investigate the effect of the Te deficit in the \( Q_z \) scans, reflection and transmission coefficients of stacking sequences of QLSs and BLs were calculated by using a set of recursive equations\(^{29}\) that accounts for absorption, refraction, and rescattering events in the film, as well as in the substrate lattice. Figure 6 shows the calculated diffraction curves for an ordered Bi₂Te₃ film with 165 QLSs and for an ordered Bi₄Te₅ film with 33 Bi₂ BLs inserted in between 165 QLSs.
diffraction pattern of the ordered Bi$_4$Te$_5$ film is quite different from that of the Bi$_2$Te$_3$, indicating that all films have structures close to Bi$_2$Te$_3$-$d$ with $d < 0.5$. The peak near $Q_z \approx 3.1$ Å$^{-1}$ ($L = 15$ in the Bi$_2$Te$_3$ structure) shifts according to $Q_z = \frac{2\pi}{\langle \Delta z \rangle}$, where the mean inter-plane atomic distance $\langle \Delta z \rangle$ is given by

$$\langle \Delta z \rangle = 2.035 - 0.025 \delta (\text{Å}) \quad (1)$$

and accounts for contributions of both composition and strain variations for epitaxial films on (111) BaF$_2$. It implies that the Bi$_2$Te$_3$ (M:N = 0:3), Bi$_4$Te$_5$ (M:N = 1:5), and BiTe (M:N = 1:2) films have lattice periodicities of 30.525 Å ($=15 \langle \Delta z \rangle$), 54.608 Å ($=27 \langle \Delta z \rangle$), and 24.120 Å ($=12 \langle \Delta z \rangle$), respectively. The other effect that can be verified by the simulation as a consequence of small Te deficit ($\delta < 0.5$) is the splitting of the $L = 18$ peak into two peaks of nearly the same intensity, which is roughly given by

$$\Delta Q_z \approx 0.23 \delta (\text{Å}). \quad (2)$$

The $L = 18$ also splits by the same amount, but one of the peaks is weaker than the other with the stronger peak shifting to the left.

Figure 7(a) shows the $\langle \Delta z \rangle$ values obtained from the $L = 15$ peak position as a function of the substrate temperature for $\Phi_R = 0, 1,$ and 2. By comparing with the expected $\langle \Delta z \rangle$ values for epitaxial films of phases from Bi$_2$Te$_3$ to BiTe (reference lines in Figure 7(a)), it is easy to notice that the films start to deviate from Bi$_2$Te$_3$ to Bi-richer phases for substrate temperatures higher than 250°C when $\Phi_R = 0$ and 1. For higher additional Te flux ($\Phi_R = 2$), the measured $\langle \Delta z \rangle$ values indicate that stoichiometric Bi$_2$Te$_3$ epitaxial films are
Figure 8 shows the azimuthal scans for films grown at two substrate temperatures. The major peaks at $\phi = 0^\circ$, $120^\circ$, and $240^\circ$, labeled as A, correspond to the three-fold symmetry of the Bi$_2$Te$_3$ structure shown in Figure 2(b), evidencing that the [1120] azimuth of the film hexagonal lattice is parallel to the [110] azimuth of the substrate cubic lattice. The azimuthal scans also show additional peaks at $\phi = 60^\circ$, $180^\circ$, and $300^\circ$, labeled as B, corresponding to twinned domains rotated by $180^\circ$. The twinning degree can be expressed by the mean intensity ratio between both peaks, $TwD = \langle I_B \rangle / \langle I_A \rangle$. Values of $TwD$ close to zero indicate small portion of twinned domains, while $TwD$ values close to one mean that the quantity of twinned domains is practically equal to the untwinned regions. Here, all twinning analyses were made on pure Bi$_2$Te$_3$ epitaxial films ($\langle \Delta z \rangle = 2.035 \pm 0.002 \, \text{Å}$).

The crystalline quality of the grown films can be evaluated by the width of the (00018) Bi$_2$Te$_3$ x-ray diffraction peak. The $\omega$-scan around the (00018) peak was measured for all samples and its full-width at half-maximum (FWHM) is displayed in Figure 7(b) as function of substrate temperature and flux ratio. For $\Phi_R = 1$, the FWHM increases monotonically from 150 arcsec to 600 arcsec as $T_{\text{SUB}}$ rises from 220 °C to 300 °C, while without extra Te flux, the crystalline quality gets worse as it increases from 200 arcsec to values higher than 1000 arcsec in the same temperature range. As the flux ratio increases to $\Phi_R = 2$, the FWHM remains below 300 arcsec up to $T_{\text{SUB}} = 280$ °C. The crystalline quality of the grown films can be evaluated by the width of the (00018) Bi$_2$Te$_3$ x-ray diffraction peak. The $\omega$-scan around the (00018) peak was measured for all samples and its full-width at half-maximum (FWHM) is displayed in Figure 7(b) as function of substrate temperature and flux ratio. For $\Phi_R = 1$, the FWHM increases monotonically from 150 arcsec to 600 arcsec as $T_{\text{SUB}}$ rises from 220 °C to 300 °C, while without extra Te flux, the crystalline quality gets worse as it increases from 200 arcsec to values higher than 1000 arcsec in the same temperature range. As the flux ratio increases to $\Phi_R = 2$, the FWHM remains below 300 arcsec up to $T_{\text{SUB}} = 280$ °C.
The offer of extra Te atoms increases the probability of formation of Bi-Te bonds, which reduces the system energy and promotes primary domains. Figure 9(b) shows the dependence of TwD on flux ratio \( \Phi_R \) for a fixed substrate temperature (240°C), confirming that the exceeding Te flux favors single domain films. The behavior of TwD on film thickness is shown in Figure 9(c) for Bi\(_2\)Te\(_3\) layers grown at TSUB = 240°C and \( \Phi_R = 1 \). It remains near 0.4 for thickness between 100 Å and 250 Å, drops to a value of 0.1 for a thickness of 500 Å, and maintains nearly constant at this value up to 5000 Å, indicating that the twinning occurs since the initial stages of growth.

**X-ray reflectometry**

The x-ray reflectivity curves were measured with a \( \omega/2\Theta \) scan from \( \omega = 0.05^\circ \) to \( 2.0^\circ \) for all samples. The open circles in the graph of Figure 10(a) represent typical curves for samples grown in region B of Figure 3. This curve exhibits very well defined interference fringes, which indicates a homogeneous layer with a smooth surface and allows an accurate determination of the film thickness. Due to the rougher surface of the films grown in region A and to the thickness inhomogeneity of the ones grown in region C, the reflectivity curves of these films present attenuated or even no interference fringes.

Besides the films deposited during 2 h, a set of Bi\(_2\)Te\(_3\)/BaF\(_2\) films with varied thickness was produced for the reflectometry analysis. The growth parameters at fixed TSUB = 240°C and \( \Phi_R = 1 \) inside region B were chosen to ensure that pure high-quality Bi\(_2\)Te\(_3\) films are obtained for this series. The samples were grown with varied deposition time of 36, 18, and 7 min and their measured reflectivity curves are shown with open circles in Figures 10(b), 10(c), and 10(d), respectively.

In order to extract quantitative information from the reflectometry measurements, x-ray reflectivity curves were calculated using the GenX software\(^3\) and fitted to the measured ones. The main fitting parameters are the layer thickness and roughness. The real (\( \gamma \)) and imaginary (\( \beta \)) parts of the material refractive index (\( n = 1 - \gamma - i\beta \)) obtained from the GenX database are also parameters in the simulation. For the BaF\(_2\) substrate, the tabulated values (\( \gamma = 1.306 \times 10^{-5} \) and \( \beta = 0.153 \times 10^{-5} \)) were used and kept fixed. These parameters were left free for the Bi\(_2\)Te\(_3\) layer during the simulation and their values, after reaching the best fit, remained constant.
within +4.7% relative to the tabulated Bi$_2$Te$_3$ values ($\gamma = 1.972 \times 10^{-5}$ and $\beta = 0.232 \times 10^{-5}$). In the GenX simulation, the roughness is assumed to have a Gaussian distribution and is included as corrective factors to electric field amplitudes at the interfaces according to the Nevot–Croce model. The substrate roughness was left as a free parameter during the simulation process and its value after the best fit remained near 3 Å for all samples. The goodness of the fit is evaluated through the mean square deviation between the calculated and measured logarithmic intensities ($\chi^2$). The best fit for each sample is also plotted as solid line in the graphs of Figure 10 and the values of the layer thickness and roughness that came out from the best fits are displayed in the graphs. The excellent agreement between the measured and simulated curves, evidenced by the small values of $\chi^2$, demonstrates that homogeneous and smooth Bi$_2$Te$_3$ epitaxial layers can be obtained at optimized growth conditions even for a thin film of only 8 QLs.

CONCLUSION

A systematic investigation of the bismuth telluride epitaxial growth on (111) BaF$_2$ as a function of substrate temperature and Te/Bi$_2$Te$_3$ beam flux ratio was presented. RHEED analysis revealed that a layer-by-layer growth mode is observed since the early stages of epitaxy and remains throughout the whole deposition. Three regimes were determined for the growth rate dependence on substrate temperature: (i) a low temperature region, where the growth rate decreases resembling the Te sticking coefficient curve; (ii) an intermediate region, in which the growth rate remains constant independent of the additional Te flux; and (iii) a third region at higher temperatures, where it diminishes again until vanishing, due to the reduction of Bi sticking coefficient. The intermediate region is found to be the most suitable to grow high quality Bi$_2$Te$_3$ thin films.

X-ray diffraction analysis showed that the epitaxy occurs with the (000L) Bi$_2$Te$_3$ hexagonal planes parallel to the (111) BaF$_2$ surface. Comparison of the measured diffraction curves with calculations established that the composition of the Bi$_4$Te$_5$ epitaxial films produced in this work stays between Bi$_2$Te$_3$ and Bi$_4$Te$_5$, with a Te deficit from 0 to 0.3. Films with pure BiTe phase were never obtained even at the highest substrate temperature and with no additional Te flux. Line width of the $L = 18$ diffraction peak as low as 140 arcsec was measured for the Bi$_2$Te$_3$ thin films, which demonstrates the high crystalline quality obtained. Azimuthal scans for a fixed asymmetrical diffraction peak showed that twinned domains rotated by 180° are present in the films. Twinning degree rises with increasing substrate temperature and this dependence is less pronounced for higher flux ratios. It is also found to reduce as the film thickness increases.

For the pure Bi$_2$Te$_3$ films grown in the optimized conditions, the x-ray reflectivity curves exhibited well defined interference fringes, characteristic of homogeneous layers with smooth surface. Layer thickness and roughness were accurately determined by fitting the experimental data to the calculated curves.

The results presented in this work demonstrate that Bi$_2$Te$_3$ films with very well controlled structural parameters can be obtained in a reproducible manner. The high structural quality of Bi$_2$Te$_3$ films as thin as only 8 QLs grown here indicates that they are promising candidates for intrinsic topological insulator, in which the bulk conduction is suppressed and only the metallic surface states are present.

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