Kinetic limitation of chemical ordering in Bi$_2$Te$_{3-x}$Se$_x$ layers grown by molecular beam epitaxy

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Received 11 December 2015, revised 17 February 2016
Accepted for publication 18 February 2016
Published 10 March 2016

Abstract
We study the chemical ordering in Bi$_2$Te$_{3-x}$Se$_x$ grown by molecular beam epitaxy on Si substrates. We produce films in the full composition range from $x = 0$ to 3, and determine their material properties using energy dispersive x-ray spectroscopy, x-ray diffraction and Raman spectroscopy. By fitting the parameters of a kinetic growth model to these results, we obtain a consistent description of growth at a microscopic level. Our main finding is that despite the incorporation of Se in the central layer being much more probable than that of Te, the formation of a fully ordered Te–Bi–Se–Bi–Te layer is prevented by kinetic of the growth process. Indeed, the Se concentration in the central layer of Bi$_2$Te$_2$Se$_1$ reaches a maximum of only $\approx 75\%$ even under ideal growth conditions. A second finding of our work is that the intensity ratio of the 0012 and 006 x-ray reflections serves as an experimentally accessible quantitative measure of the degree of ordering in these films.

Keywords: topological insulator, epitaxy, growth model, chemical order, bismuth telluride selenide alloy

(Some figures may appear in colour only in the online journal)

1. Introduction

Bi$_2$Te$_{3-x}$Se$_x$ alloys, which continue to be of great interest in the field of thermoelectric devices [1], have also more recently been shown to be topological insulators (TIs) hosting a topologically protected spin polarized surface state [2, 3]. For use as a TI, the mixed alloy with $x \approx 1$ combines positive features of each of the binary materials Bi$_2$Te$_3$ and Bi$_2$Se$_3$. Like Bi$_2$Se$_3$, it offers a relatively large band gap of $\approx 0.3$ eV with the Dirac point of the TI surface state at an energy within the bulk band gap [4, 5]. At the same time it shares Bi$_2$Te$_3$'s property of being relatively insensitive to vacancy defects, allowing the material to be grown with lower bulk carrier density than Bi$_2$Se$_3$ [4, 6].

The ideal ternary compound Bi$_2$Te$_2$Se has an ordered tetradymite-like structure consisting of quintuple layers (QL) Te–Bi–Se–Bi–Te (see inset of figure 1). Atoms within the QL are chemically bonded to each other, whereas the QLs are bonded to each other through weaker van-der-Waals forces.

When considering growth of a real crystal it is convenient to describe the structure of the QL as VI(1)–Bi–VI(2)–Bi–VI(1), where both Se and Te can occupy either the VI(1) or VI(2) sites. The ordering is then driven by the large electronegativity of Se compared to Te which greatly favors its incorporation into the VI(2) sites where it forms six chemical bonds to Bi atoms compared to the only three chemical bonds of the VI(1) sites [7]. As this selectivity mechanism is not perfect, a method to determine the degree of ordering of a film becomes an important tool for the study of these materials. This is experimentally relevant since structural ordering maximizes the band gap while disorder enhances alloy scattering of electrons and phonons [1, 6].
In this paper we study Bi₂Te₃−ₓSeₓ layers grown by molecular beam epitaxy (MBE) on Si(1 1 1) substrates with x values ranging from 0 to 3. By combining structural and compositional analysis and a kinetic growth model, we show how the degree of ordering can be determined, and that for the case of x = 1, it is kinetically limited to a value of ≈75%.

### 2. Experimental section

MBE of epitaxial Bi₂Te₃−ₓSeₓ layers is performed by co-deposition of elemental materials (6N purity) under ultra-high vacuum (UHV) conditions (base pressure <10⁻¹⁰ mbar) on H-passivated Si(1 1 1) substrates at a substrate temperature of 300 °C. All fluxes \( f \propto \text{BEP} \sqrt{T/M/\eta} \) are determined from the source temperature \( T \), the beam equivalent pressure (BEP), the Bayard–Alpert gauge sensitivity \( \eta \) and the molecular mass \( M \) by assuming fluxes of tetramer molecules \([8, 9]\). A layer growth rate \( r \) of 1.0 QL per minute is deduced from the layer thicknesses of about 70 nm (determined by profilometry on a mesa structure) with a 70 min growth time. Given that all growths are under group VI rich conditions, the growth rate \( r \) is limited by the Bi flux. The absolute flux of Bi can be determined using the constant layer growth rate of 1 QL which contains 2 Bi monolayers (ML) per minute. In our studies, the Bi and Te fluxes are kept constant (\( f_{\text{Bi}} = 2 \text{ ML min}^{-1} \) and \( f_{\text{Te}} = 84 \text{ ML min}^{-1} \)) while the Se flux \( f_{\text{Se}} \) is varied from 0 to 250 ML min⁻¹ to obtain a series of samples covering the full composition range. Additionally a pure Bi₂Se₃ layer is deposited without Te flux. The Se content \( x \) of the layers is measured to an accuracy of about ±0.05 by energy dispersive x-ray spectroscopy (EDX) of the Bi, Te and Se emission lines. High resolution x-ray diffraction (XRD) is performed with a Panalytical X’Pert diffractometer equipped with a Cu–Kα₁ source. Raman spectroscopy is performed at room temperature with a low power laser with wavelength \( \lambda = 633 \text{ nm} \) in order to avoid heating and degradation of the layers.

### 3. Results and discussion

#### 3.1. Chemical composition and kinetic growth model

The Se/Te content ratio \( x/(3 - x) \) of the Bi₂Te₃−ₓSeₓ layers, as measured by EDX, is given as the data points in figure 1 as a function of the Se/Te flux ratio \( f_{\text{Se}}/f_{\text{Te}} \). The Se/Te content ratio is up to some 8 times higher than the flux ratio. This suggests that the incorporation of Se is much more probable than that of Te. Such a behavior is also known for MBE of other alloy systems such as zinc-blende ZnTe₁₋ₓSeₓ \([10, 11]\). It can be explained by the high electronegativity of Se compared to Te. This behavior is consistent with results from bulk crystal rods grown by the Bridgman–Stockbarger method from a stoichiometric Bi₂Te₂Se₁ melt. There, strong longitudinal gradients in Se content also indicate a preferential incorporation of Se \([12]\). The nonlinear increase of the content ratio as a function of flux ratio in figure 1 shows three distinct regions. A nearly linear increase of Se/Te content ratio at low flux ratios (see left-hand inset of figure 1) is followed by a region of increased slope for a flux ratio of \( f_{\text{Se}}/f_{\text{Te}} > 0.1 \) and a region with decreasing slope at high flux ratios.

In order to gain deeper insight into the MBE growth, and based on established models describing growth of II–VI and III–V zinc-blende materials \([13, 14]\), we developed a kinetic model that describes the details of incorporation of adsorbed Se and Te, which each compete for two non-equivalent types of sublayer sites: one VI(2) site and two VI(1) sites (as identified in the schematic of figure 1).

Our kinetic model is schematically described in figure 2(a) and considers the adsorption and desorption of Se, Te, and Bi molecules as well as the preferential incorporation of Se at non-equivalent sites VI(1) and VI(2) in the stationary state. All Bi supplied is assumed to be adsorbed at the surface and nearly instantly incubated at the Bi lattice sites with a rate \( r_{\text{Bi}} = f_{\text{Bi}} = 2 \text{ ML min}^{-1} \), implying the Bi surface coverage is negligibly small and can be set to \( n_{\text{Bi}} = 0 \). The four free parameters in the model are the desorption coefficients \( d_{\text{Se}} \) and \( d_{\text{Te}} \) for Se and Te, respectively, and the ratio \( S_i \) of the incorporation probability for adsorbed Se compared to Te on site VI(1).

The resulting stationary solutions are given in equation (1) and are described as follows. Equations (1a) and (1b) describe the equilibrium between the surface adsorption rate (left-hand side) and the sum of incorporation and desorption rate (right-hand side) for Se (Te). The growth rates of the central sublayer, site VI(2), and the two edge sublayers, sites VI(1), are also fixed at \( r_2 = f_{\text{Te}}/2 \) and \( r_1 = f_{\text{Bi}} \), respectively, due to the QL structure. The formation of point defects such as SeBi and TeBi antisites and vacancies as well as decomposition of the layer play a minor role and are not considered.

As the total surface coverage (in ML) by Se \( n_{\text{Se}} \) and Te \( n_{\text{Te}} \) is limited to 1, i.e. \( n_{\text{Se}} + n_{\text{Te}} \leq 1 \), adsorption of Se and Te from the supplied fluxes results in the left-hand terms in
In order to fit our model to the experimental results, we vary the four free model parameters $S_1$, $S_2$, $d_{Se}$, $d_{Te}$ in equations (1a)–(1d). The Se/Te content ratio, which results from numerically solving the coupled equations (1a)–(1d) (such as $n_{Se}$ and $n_{Te}$), in dependence on Se/Te flux ratio is plotted in figure 1 for a given set of parameters ($S_1 = 10$, $S_2 = 220$, $d_{Se} = 21 \text{ ML min}^{-1}$ and $d_{Te} = 15 \text{ ML min}^{-1}$) and agrees well with the EDX results. This suggests that the main kinetic aspects for MBE growth of Bi$_2$Te$_3$–Se$_x$ layers are captured by the model. These parameters imply a much higher probability of incorporation of Se compared to that of Te, especially at site VI(2), resulting in a site selectivity of Se $s = 22$.

It should be noted that the set of parameters used above is not unique to fitting the EDX data, but was chosen as follows: The desorption coefficient $d_{Te} = 15 \text{ ML min}^{-1}$ was determined from the slope of the curve at low flux ratio $f_{Se}/f_{Te} < 0.1$ (see left-hand inset of figure 1), at which, according to equation (1), adsorption and consequently incorporation of Se is limited by the surface coverage with Te ($n_{Te} = 0.66$). A desorption coefficient of Se $d_{Se} = 21 \text{ ML min}^{-1}$ was deduced from the behavior at high $f_{Se}/f_{Te}$ and describes a realistic scenario, where the vapor pressure of Se is higher than that of Te. The $S_i$ values are consistent with Boltzmann factors calculated from chalcogen antisite defect formation energies ($T_{Se}$ and $T_{Te}$) in ideally ordered Te–Bi–Se–Bi–Te [15]. The chosen model parameters, especially the site selectivity $s = 22$, are supported by the consistency of the results of the kinetic growth model with the experimental XRD and Raman results that are described in sections 3.2 and 3.3.

In addition to describing the total Se content $x$ described in figure 1, our model also yields information about the Se content on each of the sublayers. In figure 2(b) we plot the calculated Se content in sublayer sites VI(1) $x_1$ and sublayer site VI(2) $x_2$ together with the total amount of Se in a
QL $x = 2x_1 + x_2$ versus the flux ratio $f_{Se}/f_{Te}$. The data from figure 1 is replotted here for comparison. The calculated Se content $x_2$ of sublayer VI(2) quickly increases with increasing Se flux and starts to saturate at about $f_{Se}/f_{Te} = 0.1$.

Equipped with this information, the non-linear behavior seen in figure 1 can now be understood. The calculated Se surface coverage $n_{Se}$ plotted in figure 2(b) is small for low $f_{Se}/f_{Te} < 0.1$, due to the efficient Se incorporation at site VI(2), but $n_{Se}$ becomes significant as $x_2$ starts to saturate. Consequently, $n_{Te}$ decreases (see figure 2(b)) and the incorporation of Se at site VI(1) (and $x_1$) starts to increase strongly for $f_{Se}/f_{Te} > 0.1$. In plain terms, this means that for low fluxes, the Se incorporates (nearly) only in the central layer, and once the flux ratio reaches $\approx 0.1$ it gains access to the two outer sublayers.

An important element of our model is that it allows us to quantify the degree of order in the films, which we define as the fraction of Se at the central sublayer site VI(1) ($x_1$) and hence also $s = S_2/S_1$, but has minor influence on the degree of order. Variations in other model or experimental parameters (retaining $f_{Se} + f_{Te} \gg f_{Bi}$) affect the surface coverages and the Se content $x$ in dependence on the fluxes, but do not influence the degree of order at any given Se content, e.g. $x = 1$.

### 3.2. Chemical order analysis by XRD

A direct measurement of the Se content of the sublayers $x_1$ and $x_2$ is extremely difficult. Significant information can however be gained from analyzing its influence on XRD measurements. Figure 3 shows $\theta$–$2\theta$ scans of layers of various compositions. Several symmetric 00l XRD reflections are observed confirming that all layers grow with the $c$-axis parallel to the surface normal of the Si(1 1 1) substrate and have single phase tetradymite-like structure. The 0012 peak varies in intensity depending on the Se content $x$ whereas the 006 peak is nearly constant in intensity. The peaks shift to larger diffraction angles with increasing Se content. The out of plane lattice parameter $c$ of the hexagonal unit cell calculated from the peak position is plotted as the data points in Figure 4 against Se content. For $x = 0, 1$ and 3, the lattice parameter $c$, which corresponds to the height of 3 QLs, is consistent with literature values for Bi$_2$Te$_3$, ordered Bi$_2$Te$_2$Se and Bi$_2$Se$_3$, respectively [16, 17]. These values of $c$ and those of the in-plane lattice parameter $a$ determined from asymmetric reflections (not shown) confirm that the layers are relaxed and that lattice strain can be neglected. The dependence of the lattice parameter $c$ on Se content clearly deviates from linearity (Vegard’s law) and reveals a bowing for $x > 1$, as also observed for bulk crystals [18]. This bowing is assigned to the non-linear change in VI(1)–VI(1) separation $c_{vdW}$ between the QL as a function of increasing $x_1$. The van-der-Waals bonds of mixed atomic pairs Se–Te are weaker than those of Te–Te or Se–Se pairs. The deviation from Vegard’s law is thus described by a term...
The pairs and is maximal for corresponding color. 

The separations $c_{1}$, $c_{2}$ and $c_{vdW}$ between neighboring sublayers VI(1)–Bi, Bi–VI(2) and van-der-Waals bonded sublayers VI(1)–VI(1), respectively, can be calculated from the lattice parameter $c = 3(2c_{1} + 2c_{2} + c_{vdW})$ and the Wyckoff positions of the atoms in Bi$_{2}$Te$_{3}$, ordered Bi$_{2}$Te$_{2}$Se$_{1}$ and Bi$_{2}$Se$_{3}$ [16, 17]. These sublayer separations are plotted in the inset of figure 4 assuming perfect order and they reveal that all the sublayer separations depend on Se content as well as on the degree of ordering, with each of the dependences being non-monotonic.

Se replacing Te at sublayer site VI(2) reduces the separation $c_{2}$ to the neighboring Bi sublayers due to its smaller covalent radius and its higher electronegativity (2.4) than Te (2.1). This increase in ionic character of the VI(2)–Bi bond transfers electron charge into this bond from the VI(1)–Bi bond [7]. The resulting reduction of polarity of the VI(1)–Bi bond increases the separation $c_{1}$ (and $c_{vdW}$) with increasing $x_{2}$. Conversely, replacing Te by Se at site VI(1) causes a decrease of $c_{1}$ and $c_{vdW}$ (with bowing) but an increase of $c_{2}$.

We now consider the influence of the ordering on the intensity of the XRD reflections. The data points in figure 5 show the integrated intensity of the 0012 reflection as a function of Se content. These have been normalized to the intensity of the 006 reflection, which is nearly independent of Se content, in order to reduce experimental inaccuracies. The intensity ratio $I_{0012}/I_{006}$ shows an oscillatory behavior with a pronounced maximum at $x = 1$ and a smaller one at $x = 3$ (i.e. Bi$_{2}$Se$_{3}$). Minima in intensity occur for $x = 0$ and at $x \approx 2.5$ where the 0012 peak intensities are comparable to background signal. A similar behavior was observed for Bi$_{2}$Te$_{3}$–Se$_{1}$ layers grown by metalorganic vapor phase epitaxy [20].

We have calculated the structure factors $S_{0012}$ and $S_{006}$ for these reflections using the layer separations discussed above and atomic form factors of the sublayers VI($i$) determined by linear interpolation between those of Te and Se based on the composition $x_{i}$ calculated from our kinetic model [21]. The ratio of squared structure factors, which is proportional to the normalized intensity, is plotted as the red curve in figure 5. The experimental data are in good agreement with the calculated curve (to within a scaling factor$^{3}$) which unites the right and left axis in the figure and which was chosen to make the calculation for pure Bi$_{2}$Se$_{3}$ fit to the data) for partial ordering determined by the site selectivity $s = 22$ of the kinetic model.

The calculated intensity ratios for Bi$_{2}$Te$_{2}$Se$_{1}$ layers with perfect order (blue curve) and for a random alloy (black curve) are also shown for comparison. For Bi$_{2}$Te$_{2}$Se$_{1}$, perfect order gives a nearly 3-times higher intensity ratio than is experimentally observed, while the intensity ratio is small and monotonic in $x$ for the random alloy.

The inset of figure 5 again shows the ratio of squared structure factors, now calculated for Bi$_{2}$Te$_{2}$Se$_{1}$ ($x = 1$) layers with varying degree of order $x_{2}$. The intensity ratio is small for a random or nearly random alloy, and starts to increase significantly and monotonically at $x_{2} \approx 0.5$. The red dot marking the intensity ratio from our kinetic model corresponds to an ordering $x_{2} = 0.75$, where permitting an uncertainty of $\pm 1\%$ in the literature values of sublayer separations $c_{1}$ and $c_{2}$ (at constant $c$) can shift the determined degree of order by up to $\pm 0.05$. Thus, the XRD measurement of the intensity ratio $I_{0012}/I_{006}$ of a Bi$_{2}$Te$_{2}$Se$_{1}$ layer (compared to that of a Bi$_{2}$Se$_{3}$ reference layer for instrumental calibration) is a tool for analyzing the degree of structural order.

It is worth noting that due to the large slope in the inset of figure 5, assuming an uncertainty of $\pm 20\%$ on the measured peak intensity ratio still yields a result of $x_{2} = 0.75 \pm 0.04$. Even considering statistical fluctuation for the relatively weak 0012 peak and difficulties in perfectly applying XRD correction factors that vary slightly with Se content $x$ due to the shifts in Bragg angles, a $\pm 20\%$ uncertainty estimate is

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$^{3}$This scaling factor accounts for instrumental geometry factors and structural effects contributing to XRD intensities in thin films.
extremely conservative. This highlights the sensitivity of this method for characterizing the degree of ordering in at least relatively highly ordered layers.

3.3. Impact of chemical order on lattice dynamics

Further confirmation of our interpretation of chemical ordering is provided by the set of Raman spectra in figure 6(a). The spectra are observed in backscattering with parallel polarizations of the exciting and the scattered light. They show four optical phonon modes: the two $A$ modes $A_{1g}^1$, $A_{1g}^2$, the high-frequency $E_2^g$ mode and a weak additional mode. The latter is presumably a gap mode $A_{gap}$ of Te at sites VI(1) and is only observed in the range $1 \leq x \leq 2.3$. Similar phonon modes were observed by Raman scattering from bulk Bi$_2$Te$_3$.Se$_x$ crystals [22]. The Raman lines are assigned to the modes from their frequencies and polarization dependence, i.e. the disappearance of modes with $A_{g}$-symmetry for crossed light polarizations. The vibrational displacement patterns of the atoms for the different modes are sketched as insets of figure 6(a). Spring constants $k_1$ and $k_2$ in Bi$_2$Te$_3$ and Bi$_2$Se$_3$ are deduced from the measured mode frequencies in the corresponding binary layers of our series. The spring constants in Bi$_2$Te$_3$.Se$_x$ depend on the layer compositions $x_1$ and $x_2$ and are linearly interpolated. The observed frequencies of the $E_2^g$ mode and the $A_{1g}^1$ mode depending on $x$ are well described by the spring-mass model with the reasonable assumption that the spring constants $k_1$ depend linearly on the calculated composition of the neighboring layer site V(2), i.e. $k_1(x_1) = 1.067 + 0.517x_1$ and $k_2(x_2) = 1.672 - 0.036x_2$ (in units of $10^6$ amu·cm$^{-2}$) for $A$ modes. The spring constant $k_1$ considerably increases with Se content of site VI(1) $x_1$, while $k_2$ is nearly independent of $x_2$. The $E_2^g$ and the $A_{1g}^1$ mode frequencies calculated for Bi$_2$Te$_3$.Se$_x$ with partial order as predicted by the kinetic growth model are shown by red curves in figure 6(b). These curves agree well with the experimental results and reproduce the qualitative assumption of nearly constant frequencies for a high degree of order and Se contents $x \leq 1$, as most of the...
Se atoms with low mass are incorporated at the resting site VI(2). The mode frequencies should even decrease slightly with increasing Se content for x ≲ 1 in perfectly ordered Bi2Te3−xSex (blue curves). In contrast, monotonic, nearly linear shifts of mode frequencies are expected for a random alloy (black curves). Inspection of the curves shows that both mode frequencies are sensitive to the degree of order, especially around x = 1.

Compared to the A1g2 mode, the Raman peaks of the A1g2 mode at x ≈ 1 and the E1g mode at x ≈ 2 are quite broad and close in frequency to those of the Agap mode, which may reduce the accuracy of the fitted frequency values (figure 6(a)). Moreover, these modes may be shifted due to a coupling to the Agap mode with similar frequency. These aspects most probably cannot fully explain the considerable difference between measured A1g2 mode frequencies and those calculated within the simple model with spring constants depending only on the composition of the neighboring site VI sublayer. Obviously, the A1g2 mode has a more complex dependence on sublayer compositions and it shows a two-mode behavior at large Se content x ≳ 1.

Increased frequencies of the A1g2 mode may be qualitatively explained as follows: The vibrational displacement of Bi layers is in anti-phase to that of the VI(1) layers and spring k1 has a predominant influence on the A1g2 mode frequency. The higher electronegativity of Se (2.4) compared to Te (2.1) and Bi (1.8) and the dependence of the sublayer separations on the compositions of both sublayers VI(1) and VI(2), suggest that the bond strengths, i.e. the spring constants k1 and k2, also change with the polarity of bonds and thus depend on both sublayer compositions. For Bi2Se3, the effective charges (Bader charges) of the sites Se(2), Bi, and Se(1) were calculated by density-functional theory to be −0.83e, +1.0e, and −0.59e, respectively [23]. For Bi2Te3, corresponding values of effective charges −0.33e, +0.36e, and −0.19e were determined by tight binding calculations [24]. The different effective charges of sites in both materials suggest that the bond polarities in Bi2Te3−xSex depend on the Se content of the sites. An increase of the Se content x2 of sublayer site VI(2) due to ordering causes a partial transfer of valence electrons from neighboring Bi sublayers to this sublayer and the polarity of bonds Bi–VI(2) increases. Furthermore, the Bi sublayers receive a larger positive charge and attract electronic charge from the Bi–VI(1) bonds, which consequently decrease in polarity [7]. An increase (decrease) of polarity of bonds is expected to decrease (increase) the bond strength and the spring constant, as observed in semiconductors with zinc-blende structure. Consequently, the A mode spring constant k2 should decrease and k1 should increase with increasing difference in Se content between the two sites (x2 − x1). If we take spring constants k1 and k2 to have additional terms +0.26(x2 − x1) and −0.13(x2 − x1) (in units of 106 amu·cm−2), respectively, the influence of changes in the polarity of the bonds on the A mode frequencies is illustrated by the red dashed curves in figure 6(b). The A1g2 mode increases considerably in frequency, while the A1g2 mode behavior is only slightly affected. The agreement with experimental frequencies is improved for both A modes. A detailed understanding of Ag modes in (partially) ordered Bi2Te3−xSex, however, requires first-principle calculations of their structural, bonding and vibrational properties.

4. Conclusion

We have developed a kinetic growth model to describe the MBE growth of Bi2Te3−xSex layers under group VI rich growth conditions. The model includes adsorption, desorption and incorporation probabilities of Se and Te at the non-equivalent sublayer sites VI(1) and VI(2) of the QL VI(1)−Bi−VI(2)−Bi−VI(1). The kinetic rate equations quantitatively predict the Se contents x1 and x2 in the sublayers of VI(1) and VI(2). They show that despite the highly preferential incorporation of Se at the central site VI(2), the degree of chemical order in Bi2Te2Se1 as quantified by the Se content on this site is kinetically limited to x2 = 0.75 ± 0.04. The only parameter affecting the chemical order is the site selectivity of Se s.

The calculated sublayer compositions were verified by a precise description of the structural properties measured by XRD and the phonon mode frequencies measured by Raman spectroscopy for Bi2Te3−xSex layers of all compositions from x = 0 to 3. While the lattice parameter c is barely affected by the degree of chemical order, the separation of atomic sublayers depends strongly on the composition of the group VI sublayers and thus on the degree of order. The intensity of the partially destructively interfering 00120 XRD peak oscillates with increasing Se content due to these non-monotonic variations of sublayer separations and is a sensitive probe of the structural order in Bi2Te2Se1 layers.

Our main finding is that the degree of chemical order in MBE-grown V2VI3 alloys with non-equivalent chalcogen lattice sites is kinetically limited to values well below that expected from the site selectivity of the competing elements, despite this being the most important material-specific parameter for chemical ordering in these material systems. Chemical ordering in other epitaxial crystal structures with non-equivalent lattice sites for competing elements, such as Heusler or oxide compounds, may also be described by this kinetic model with modifications according to the specific material and growth properties.

Acknowledgments

We gratefully acknowledge the financial support by the EU ERC-AG Program (project 3-TOP) and the DFG (through SFB 1170 'ToCoTronics'), as well as experimental assistance from N V Tarakina. G K thanks for the support by the Polish Science Center (Grant No. 2014/14/M/ST3/00484) and by the Foundation for Polish Science by the Master program.

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