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# Relation between crystallographic orientation and the condensation coefficients of Hg, Cd, and Te during molecular-beam-epitaxial growth of $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ and $\text{CdTe}$

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We investigate here the influence of the crystallographic orientation of the  $\text{CdTe}$  substrate on the condensation coefficients of Hg, Cd, and Te during the growth of  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$  and  $\text{CdTe}$  by molecular-beam epitaxy. We show that the Hg condensation coefficient is strongly influenced by the orientation. A  $\text{CdTe}(\bar{1}\bar{1}\bar{1})B$  face requires about one order of magnitude less mercury than a  $(111)A$  face when growth occurs at  $185^\circ\text{C}$ . Whereas for a  $\text{CdTe}(100)$  face, the Hg condensation coefficient falls in between. Even though the effect is less dramatic for the condensation coefficient of cadmium, a similar tendency is observed. These results can be explained in terms of the bonding of the surface atoms and confirm once again that the surface plays an important role in the molecular-beam-epitaxial growth process.

## I. INTRODUCTION

Cadmium telluride has a zinc-blende structure which belongs to the cubic noncentrosymmetric space group  $F\bar{4}3m$ . The characteristic tetrahedral bonding of this compound is caused by  $sp^3$  hybridization.  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$  belongs to the same group as  $\text{CdTe}$  except that a fraction  $(1-x)$  of the Cd is randomly replaced by Hg. Since  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ ,  $\text{HgTe}$ - $\text{CdTe}$  superlattices, and other Hg based semiconductors are of great importance for infrared detectors it is necessary to understand and to be able to grow high-quality material.

Molecular-beam-epitaxial (MBE) growth is difficult to achieve for  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$  because evaporation of this semiconducting alloy is highly noncongruent.<sup>1</sup> The mercury tends to evaporate preferentially from the surface. As a consequence it has been experimentally shown that the condensation coefficient for Hg is on the order of  $10^{-3}$  for growth at a substrate temperature of  $190^\circ\text{C}$ .<sup>2</sup> This previous determination of the Hg condensation coefficient had been carried out for  $n$ -type  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$  layers (with  $x$  close to 0.20) grown on  $\text{CdTe}(\bar{1}\bar{1}\bar{1})B$  substrates.<sup>2</sup> The condensation coefficient for a given element is defined as the ratio of the number of atoms incorporated in the layer to the number of incident atoms impinging the surface. A temperature range of  $180$ – $200^\circ\text{C}$  is required to grow high-quality  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$  films by MBE.<sup>3</sup>

The  $(111)$  and  $(100)$  faces in the zinc blende structure are called polar faces. This polarity leads to the two types of faces in each case. A  $(111)A$  face is ideally terminated by a triply bonded Cd atom or a singly bonded Te [see Fig. 1(a)]. Hence it is called the Cd face since the stable configuration has the Cd, or possibly Hg atom in the case of  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ , at the surface. The other possible face, the  $(\bar{1}\bar{1}\bar{1})B$  face, is called the Te face since the stable configuration is terminated by triply bonded Te [see Fig. 1(a)]. The  $(100)$  faces could terminate either with a doubly bonded Te or a doubly bonded Cd [see Fig. 1(b)]. For the  $(100)$  face there is no difference in the bonding of Cd or Te at the surface, but for  $(111)A$  and  $(\bar{1}\bar{1}\bar{1})B$  either the Te or Cd atom is

triply bonded while the other atom is singly bonded. For  $(100)$ , Cd and Te are equivalent but the bonding direction is rotated by  $90^\circ$ . An important difference, in terms of polarity, is observed between the  $(111)$  and  $(100)$ . For the first one, two kinds of bonds are found for each element while only one kind of bond exists for the second.

Mercury, and to a lesser extent cadmium, is more volatile than tellurium. Because of the difference in bonding between the  $(111)A$  face, the  $(\bar{1}\bar{1}\bar{1})B$  face, and the  $(100)$  face, one can expect an influence of the crystal orientation on the growth conditions of  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$  and  $\text{CdTe}$ . Such an influence is not taken into consideration by a classical thermodynamic approach to crystal growth from a vapor phase.

In this paper it is reported for the first time that the crystallographic orientation of the  $\text{CdTe}$  substrate indeed has a strong influence on the growth parameters. An example of which is the Hg and Cd condensation coefficients during growth of  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$  and  $\text{CdTe}$  by MBE.

## II. EXPERIMENT

The different crystallographic orientations investigated are  $\text{CdTe}(111)A$ ,  $\text{CdTe}(\bar{1}\bar{1}\bar{1})B$ ,  $\text{CdTe}(111)\parallel\text{GaAs}(100)$ , and  $\text{CdTe}(100)\parallel\text{GaAs}(100)$ . The experiments were carried out in a Riber 2300 MBE machine which is capable of handling Hg.  $\text{CdTe}(111)A$  and  $\text{CdTe}(\bar{1}\bar{1}\bar{1})B$  substrates were etched in a 1% bromine methanol solution and repeatedly rinsed in methanol before being introduced into the vacuum system. The  $\text{GaAs}(100)$  substrates were etched in a solution of  $\text{H}_2\text{SO}_4\text{:H}_2\text{O}_2\text{:H}_2\text{O}$  with a 4:1:1 ratio. Following this etch, the oxide passivation was removed with concentrated  $\text{HCl}$ . The substrates were then rinsed repeatedly in deionized water and methanol and quickly introduced into the vacuum system.

$\text{CdTe}(111)\parallel\text{GaAs}(100)$  and  $\text{CdTe}(100)\parallel\text{GaAs}(100)$  can be grown on these  $\text{GaAs}(100)$  substrates by preheating the substrate at  $580$  and  $480^\circ\text{C}$ , respectively.<sup>4</sup> In order to reduce the dislocations due to the strain produced by the 14.5% mismatch between  $\text{GaAs}$  and  $\text{CdTe}$ , a buffer layer

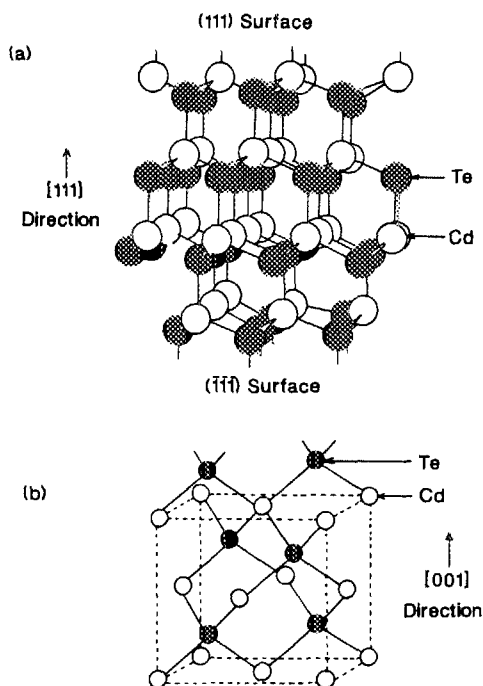


FIG. 1. Crystal structure of CdTe. (a) Viewed from (111) direction showing the triple and single bonds for this orientation. (b) Viewed from (100) direction showing the double bond for this orientation.

consisting of steps of  $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$  with graduated  $x$  values was used to grow  $\text{CdTe}(100)\|\text{GaAs}(100)$ .<sup>5</sup> If the  $x$  value of the initial step is greater than 15%, the  $\text{CdTe}(100)\|\text{GaAs}(100)$  orientation results independent of the type of preheating.<sup>4</sup> As previously reported the growth of  $\text{CdTe}(111)\|\text{GaAs}(100)$  provides a  $(\bar{1}\bar{1}\bar{1})B$  face.<sup>6</sup>

### A. $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$

The growth of  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$  and CdTe over these substrates was closely followed *in situ* by high-energy electron diffraction (RHEED). We have previously reported that RHEED can be used to see when the Hg flux is too large or too small for growth on CdTe  $(\bar{1}\bar{1}\bar{1})B$  substrates.<sup>7</sup> At a growth temperature of 185 °C, if the Hg flux is too high, double dots appear on the streaks [see Fig. 2(a)]. When the Hg flux is too large, stacking faults, which are probably anti-phase domains, are produced even though the excess of Hg atoms desorbs from the surface during growth. Then, even if the film is monocrystalline, the presence of these defects is detrimental to the electrical properties of the film. A strong correlation has been reported between the presence of these stacking faults, a decrease of the Hall mobilities, and a deterioration of the line shape profiles obtained by electroreflectance.<sup>8</sup> Figure 2(b) shows narrow and short streaks with well-defined Kikuchi lines. These show the high quality of the growth for ideal Hg flux. At the other extreme, if there is an insufficient Hg flux, an excess of Te will cause the film to be polycrystalline [see Fig. 2(c)].

The minimum amount of Hg necessary to maintain epi-

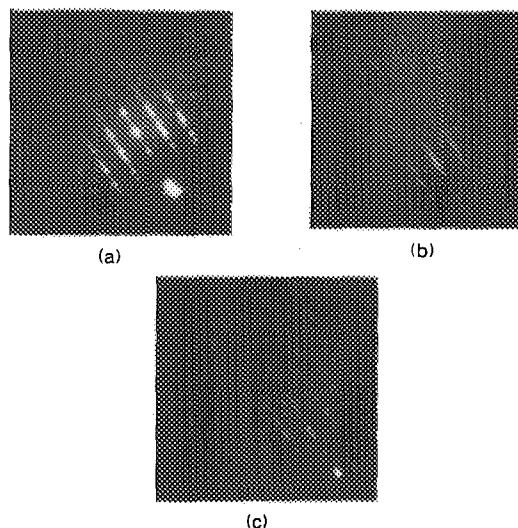


FIG. 2. RHEED pattern with  $e^-$  beam along  $[1\bar{1}0]$  for  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$  growth on a  $(\bar{1}\bar{1}\bar{1})B$  face: (a) Hg flux is too large; (b) ideal condition for monocrystalline growth; (c) insufficient Hg flux for monocrystalline growth.

taxial growth of  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$  was obtained by the following procedure. With the composition and all other conditions the same,  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$  was grown at 185 °C for about 25 min. Figure 3(a) shows the narrow and short streaks with sharp Kikuchi lines for the ideal condition of the Hg flux. The Hg flux was then slowly decreased until the difficulties in maintaining the single growth orientation were observed. For the  $(111)A$  face, when the Hg flux was insufficient, we began to see rings which indicate a polycrystalline layer [see Fig. 3(b)]. When the Hg flux was too low on the  $(\bar{1}\bar{1}\bar{1})B$  face, extra dots on the streaks appeared initially, followed by the polycrystalline rings. For the (100) case, off angle streaks followed the appearance of additional spots off the main streaks. Figure 4(a) shows the RHEED pattern for ideal growth conditions on the (100) face. Figure 4(b) shows the off angle streaks when the Hg flux is insufficient to maintain the growth on the (100) face. On the Te-rich side,  $p$ -type layers are usually grown.<sup>2</sup> Results for the minimum amount of Hg necessary to maintain a good monocrystalline growth are reported in Table I and will be discussed later.

### B. CdTe

An important difference exists between the condensation coefficient of Hg and Cd. At 200 °C the condensation

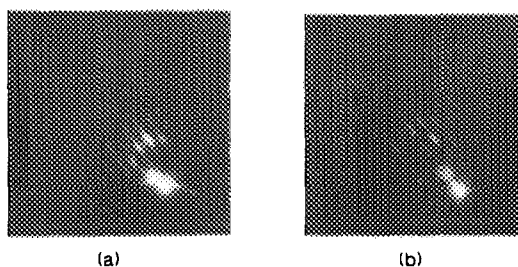


FIG. 3. RHEED pattern with  $e^-$  beam along  $[1\bar{1}0]$  for  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$  growth on a  $(111)A$  face: (a) ideal condition for monocrystalline growth; (b) insufficient Hg flux for monocrystalline growth.

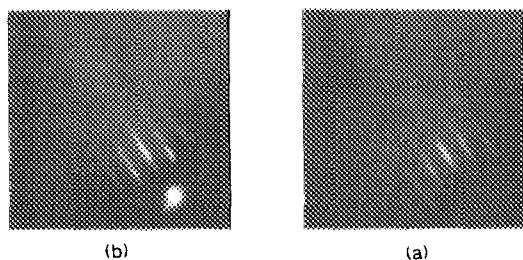


FIG. 4. RHEED pattern with  $e^-$  beam along  $[0\bar{1}1]$  for  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$  growth on a (100) face: (a) ideal condition for monocrystalline growth; (b) insufficient Hg flux for ideal monocrystalline growth.

coefficient for CdTe growth was found to be close to unity. Thus, in order to investigate the influence of the crystallographic orientation on the condensation coefficient for Cd (or Te), we had to increase the growth temperature. As a consequence, for temperatures higher than about 220 °C either free tellurium or free cadmium is reevaporated which prevented films from being polycrystalline. Hence, we were unable to use the same technique for Cd as for Hg. Thus, we investigated the change in the growth rate with substrate temperature to determine the change in the Cd condensation coefficient. This study was carried out on CdTe (111)*A*, CdTe ( $\bar{1}\bar{1}\bar{1}$ )*B*, and CdTe (100)||GaAs(100) substrates.

CdTe(111)*A* and ( $\bar{1}\bar{1}\bar{1}$ )*B* substrates were mounted on the same molybdenum substrate holder by sticking one next to the other with gallium. A single mask was used to cover part of both the (111)*A* and the ( $\bar{1}\bar{1}\bar{1}$ )*B* substrates. Hence, we were able to measure the thickness of the layer grown using a step profiler for substrate temperatures of 150, 200, 250, and 300 °C. A single CdTe cell was used to grow the CdTe films. The growth rate on CdTe(100)||GaAs(100) was determined separately for the same substrate temperatures and the same CdTe flux as in the (111) case. Also, the CdTe growth on (111)*A* and (100) orientations was performed with the addition of Cd flux from a Cd cell. The addition of Cd to ( $\bar{1}\bar{1}\bar{1}$ )*B* face creates additional dots along the streaks. We suspect this is due to the creation of anti-phase domains similar to the case of HgCdTe growth with too much Hg. Hence the growth rate observed from that case is not reported. Similarly, the growth rate of CdTe over ( $\bar{1}\bar{1}\bar{1}$ )*B* and (100) has been determined with additional Te flux. The results are presented in Table II.

### III. RESULTS

#### A. $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$

With all other conditions including the composition being constant, the minimum amount of mercury required for monocrystalline growth of each of the different orientations

TABLE I. Minimum amount of Hg flux (in arbitrary units) needed to maintain the monocrystalline growth of  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ .

Substrate	CdTe( $\bar{1}\bar{1}\bar{1}$ ) <i>B</i>	CdTe(100)  GaAs(100)	CdTe(111) <i>A</i>
Hg flux (arbitrary units)	1	4.4	9.0

TABLE II. Growth rates of CdTe at different substrate temperatures and different orientations.

Substrate temperature	CdTe(111) <i>A</i> ( $\mu\text{m/h}$ )	CdTe(111) <i>B</i> ( $\mu\text{m/h}$ )	CdTe(100)  GaAs(100) ( $\mu\text{m/h}$ )
150	0.71	0.70	0.70
200	0.62	0.63	0.58
250	0.43	0.58	0.50
290	0.31	0.50	0.43
300 (with addition of Cd)	0.64	—	0.43
300 (with addition of Te)	—	0.64	0.46

is presented in Table I. The largest amount of mercury was needed for the (111)*A* face while ( $\bar{1}\bar{1}\bar{1}$ )*B* required the least. The (100) orientation need for mercury was between the other two. The amount of mercury necessary to grow  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$  on CdTe(111)||GaAs(100) is the same as that required for CdTe( $\bar{1}\bar{1}\bar{1}$ )*B* substrates. This supports our previous finding that the CdTe(111)||GaAs(100) has a ( $\bar{1}\bar{1}\bar{1}$ )*B* face.<sup>6</sup>

It should be pointed out that at 185 °C a difference of about one order of magnitude is observed between the Hg condensation coefficient for (111)*A* and ( $\bar{1}\bar{1}\bar{1}$ )*B*.

#### B. CdTe

For all the substrate temperatures above 200 °C, the ( $\bar{1}\bar{1}\bar{1}$ )*B* and (100) orientations exhibited sharp RHEED streaks and very well-defined Kikuchi lines [see Figs. 5(a) and 5(b)]. For (111)*A*, additional dots along the original streaks followed by additional dots along an off angle line were visible for temperatures greater than 200 °C. For substrate temperatures of 300 °C, the off angle dots were connected by off angle streaks [see Fig. 5(c)]. While adding Cd to the CdTe at a substrate temperature of 300 °C the off angle streaks disappeared, but the dots remained. We believe that

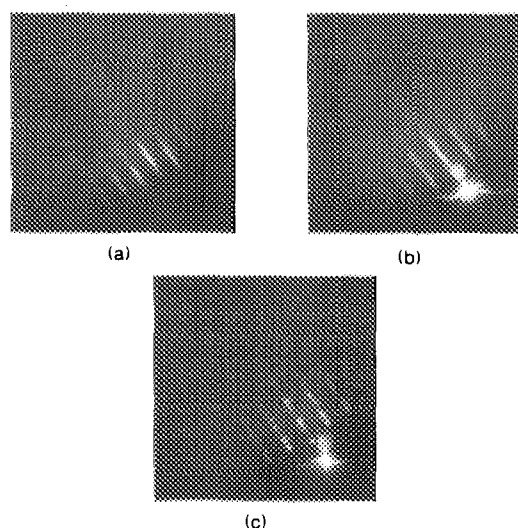


FIG. 5. RHEED pattern for CdTe growth with substrate temperature at 285 °C: (a) over (100) face,  $e^-$  beam along  $[0\bar{1}1]$  direction; (b) over ( $\bar{1}\bar{1}\bar{1}$ )*B* face,  $e^-$  beam along  $[1\bar{1}0]$  direction; (c) over (111)*A* face,  $e^-$  beam along  $[1\bar{1}0]$  direction.

these off angle streaks are due to faceting similar to that reported for GaAs.<sup>9</sup> Due to the grazing angle of incidence of the electron beam, we were able to pick up the faces of the pyramid formed on the surface.

Table II shows the growth rates of CdTe obtained over different faces at different substrate temperatures. We can see that (111)*A* shows the lowest growth rate, ( $\bar{1}\bar{1}\bar{1}$ )*B* the highest, while (100) falls in the middle. All the orientations exhibit an equal growth rate for a substrate temperature of about 150 °C. Addition of Cd to the CdTe flux at 300 °C increased the growth rate for (111)*A* orientation. This proves that the reduction in growth rate is due to insufficient Cd for the (111)*A* face, which agrees with the observation by RHEED. Similarly, addition of Te to the ( $\bar{1}\bar{1}\bar{1}$ )*B* face at a substrate temperature of 300 °C increased the growth rate. Hence, the growth rate reduction on the ( $\bar{1}\bar{1}\bar{1}$ )*B* face is mainly controlled by the Te flux. No large change in the growth rate has been observed for the (100) face when additional Te or Cd flux was added to the flux from the CdTe cell.

#### IV. DISCUSSION

A model for the MBE growth of II–VI semiconducting compounds has been proposed recently using a thermodynamic approach.<sup>10</sup> In this study where the mass action law is applied and the growth is only controlled by thermodynamics, it is found that the growth rate is controlled by the element having the lower equilibrium pressure, i.e., the Cd flux for a Te-rich material or the Te flux for a Cd-rich material. This conclusion is legitimate from a bulk crystal stoichiometry point of view. But it does not take into account the crystallographic orientation of the surface which can influence strongly the growth kinetics. Therefore, this conclusion should be revised as follows: Where the temperature is high enough to reevaporate free Te, the growth rate on (111)*A*, i.e., the Cd face, is controlled by the Cd condensation coefficient and the growth rate of ( $\bar{1}\bar{1}\bar{1}$ )*B*, i.e., the Te face, is controlled by the Te condensation coefficient.

One can explain these results by looking at the atoms of the face. Since the (111)*A* face has a triply bonded Cd, the ( $\bar{1}\bar{1}\bar{1}$ )*B* face a triply bonded Te, and the (100) face a doubly bonded Te or doubly bonded Cd, the ( $\bar{1}\bar{1}\bar{1}$ )*B* face Te acts as a cap for Hg and Cd, whereas for the (111)*A* face Hg and Cd are exposed to vacuum. Thus, Hg and Cd can more easily reevaporate from the (111)*A* face than from the ( $\bar{1}\bar{1}\bar{1}$ )*B* face. For the (100) orientation the number of atoms exposed to vacuum is smaller than that of the (111) face and also the surface atoms are doubly bonded. From our XPS (x-ray photoelectron spectroscopy) measurements the (100) CdTe|| (100)GaAs should also have Te up. For the (100) orientations Hg and Cd are more protected from reevaporation than for the (111)*A* face but less than for the ( $\bar{1}\bar{1}\bar{1}$ )*B* face. Hence, in order to maintain an epitaxial growth with the same growth rate, we need more Hg or Cd for the (111)*A* face and less for the ( $\bar{1}\bar{1}\bar{1}$ )*B* face while the amount required for the (100) orientation falls in between. The fact that the rate of growth for CdTe on the (100) face does not change significantly when additional Te or Cd is provided confirms the important difference existing between the (100) and (111) orientations in terms of polarity and chemical bonds.

For (110) orientation, we expect a behavior very close to that of the (100) orientation even though all the atoms are in the same plane.

Schetzina *et al.* have reported the observation of a difference in the CdTe growth rate on (111)*A* and ( $\bar{1}\bar{1}\bar{1}$ )*B* faces.<sup>11</sup> But, their results were contradictory to our results. This discrepancy could be due to the confusion which existed until the recent report by Fewster *et al.*<sup>12</sup> on the identification of (111)*A* and ( $\bar{1}\bar{1}\bar{1}$ )*B* face.

#### V. CONCLUSION

For III–V compounds it is established by electrical and optical measurements that the highest quality is obtained for the (100) orientation. In our experience, even for II–VI Hg related material, the highest Hall mobilities happened to be obtained for the (100) orientation. Although it is premature to draw a definite conclusion as to which orientation is most suitable for the growth of high-quality Hg<sub>1-x</sub>Cd<sub>x</sub>Te and other Hg based II–VI compounds, the (100) face seems to show higher mobilities. On the other hand, the ( $\bar{1}\bar{1}\bar{1}$ )*B* face has a very important advantage since the amount of Hg required to grow epitaxial Hg based compounds in this orientation is much less than for the other orientations that we have considered. This will reduce the difficulties that the tremendous amount of Hg causes to the MBE vacuum system. We have also demonstrated that the growth on the (111)*A* orientation of monocrystalline HgCdTe and CdTe films can be achieved by MBE even though it is more difficult than on the ( $\bar{1}\bar{1}\bar{1}$ )*B* orientation. The growth rate of CdTe at high temperatures is controlled by Te flux for the ( $\bar{1}\bar{1}\bar{1}$ )*B* face while it is controlled by Cd for the (111)*A* face.

We have shown that the amount of Hg necessary to maintain epitaxial growth on CdTe(111)||GaAs(100) is the same as that required for CdTe( $\bar{1}\bar{1}\bar{1}$ )*B*. This agrees with our previous work showing that CdTe(111) grows on GaAs(100) with a *B* face.<sup>6</sup>

These results also confirm that a classical thermodynamic approach using the mass action law and neglecting surface kinetics is not perfectly adapted for the understanding of the MBE growth even though such an approach can provide very useful information. These observations suggest that the crystallographic orientation not only has an influence on the magnitude of the condensation coefficients but could also play a role in doping. This point should be investigated in the future.

So far, this dependence of the condensation coefficient on crystallographic orientation has not been reported for the growth of III–V compounds, which also crystallize in the zinc-blende structure. We think that even if the inequality is less dramatic for As, P, or Sb condensation coefficients, the same tendency should be observed.

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