PHYSICAL REVIEW B VOLUME 56, NUMBER 15 15 OCTOBER 1997-I

Epitaxy of layered compounds: GaSe on Si(111)

N. Jedrecy*
LURE, CNRS-MRES-CEA, Bâtiment 209D, Centre Universitaire, F-91405 Orsay, France
and Laboratoire de Minéralogie-Cristallographie, Associé au CNRS et aux Universités Pierre et Marie Curie (Paris 6) et Denis Diderot (Paris 7), 4 place Jussieu, F-75252 Paris Cedex 05, France

R. Pinchaux
LURE, CNRS-MRES-CEA, Bâtiment 209D, Centre Universitaire, F-91405 Orsay, France
and Université Pierre et Marie Curie, 4 place Jussieu, F-75252 Paris Cedex 05, France

M. Eddrief
Laboratoire de Minéralogie-Cristallographie, Associé au CNRS et aux Universités Pierre et Marie Curie et Denis Diderot, 4 place Jussieu, F-75252 Paris Cedex 05, France

(Received 10 February 1997; revised manuscript received 19 May 1997)

We investigated the interface between a three-dimensional substrate, Si, and a layered compound, GaSe, in the very first steps of the growth. Atomic models, related to deposits of 1.5 and 2.5 sheets, respectively, were derived from grazing incidence x-ray diffraction. They are based on the bulk phases, that is, on Ga_2Se_2 iono-covalent sheets, stacked with van der Waals bonding. Two orientations at 180° from each other are found in nearly equal proportion. In addition, two different types of domains occur: one is strained on the substrate, the other one is strain relieved. The two types of domains are distinguished by a different treatment of their contributions to the diffracted intensity. The growth proceeds by complete sheets, preferably on the in-plane relaxed domains. As a consequence, for each sample, the final GaSe surface is expected to present steps, at least of one sheet height (7.95 Å). [S0163-1829(97)07939-3]

I. INTRODUCTION

Most epitaxial growth studies deal with covalent or metallic compounds. Recently, the epitaxy of layered structures was demonstrated, thus opening a way to achieve a different kind of heterostructure.[1–4] Gallium selenide is such a two-dimensional (2D) compound, characterized by iono-covalent sheets with stoichiometry GaSe, bound to each other by van der Waals–type interactions. All the bulk phases may be described by use of hexagonal lattices, with a common parameter a, equal to 3.74 Å.[5–7] The value of the c parameter depends on the stacking sequence of the sheets. An individual sheet consists of four (0001) atomic planes, whose sequence is Se-Ga-Ga-Se. The Ga_2Se_2 sheets are 4.8 Å thick while the intersheet spacing is 3.2 Å. The main three phases β, ε, γ, involve, respectively, 2, 2, and 3 sheets in the unit cell. One can expect the epitaxial growth of any of them on Si(111), since the misfit is -2.6%, along the azimuth $[100]^{GaAs}//[10\bar{1}]^{Si}$. According to the threefold symmetry of GaSe, two orientations are expected, mutually rotated by 180°, as shown in Fig. 1 in the case of the γ phase.

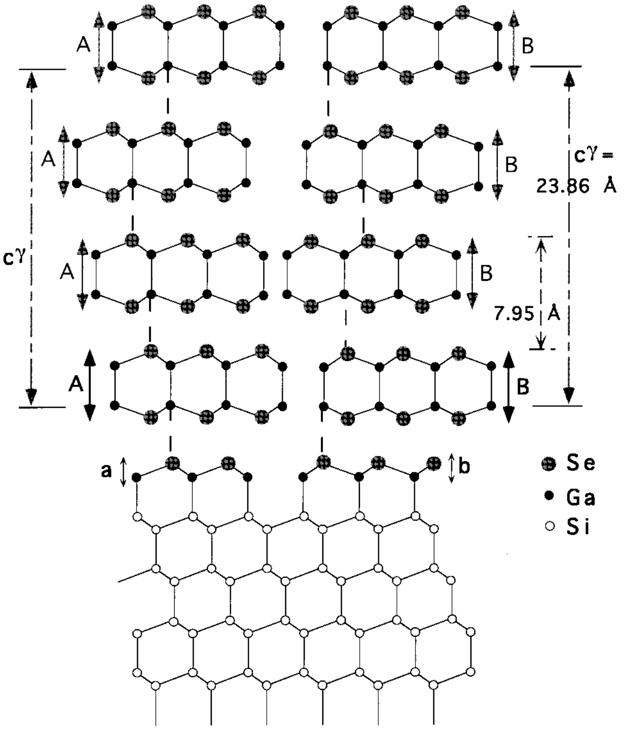

FIG. 1. Projection along $[\bar{1}10]^{Si}$ of the epitaxial γ (0001) GaSe on Si(111). Four complete sheets are depicted on top of the half-sheet interface. The two orientations, at 180° each, are shown, labeled "A," "B," in the case of a full sheet, and "a," "b," in the case of a half sheet.

Apart from the optical nonlinear properties of GaSe,[8,9] the interband transitions in the UV region,[10] and potential applications in multiple quantum well structures, the growth of GaSe on silicon may provide a way to achieve heteroepitaxy with materials that are hardly compatible with Si. The van der Waals bonding between the sheets should enable the relief of the strain produced by the lattice misfit, or the thermal expansion coefficients. Indeed, the use of GaSe as a template for GaAs growth on Si(111)-As has been reported as promising.[11] This use of layered compounds as buffers

0163-1829/97/56(15)/9583(6)/$10.00

should open new perspectives in the integration of optoelectronic functions in devices. This however requires us to get surfaces as smooth as possible, while minimizing the layers thickness. Thus, it is worthwhile to investigate the growth process of 2D compounds on 3D materials, with respect to the widely studied 3D-3D case. What effect has the van der Waals bonding on the interface stress confinement, what kind of growth is promoted, and what is the final surface state?

II. STRUCTURAL CHARACTERISTICS OF THE EPITAXIAL LAYERS

The LPS laboratory[12] (University Paris 6, France) has achieved, by molecular-beam epitaxy (MBE), the growth of GaSe on several Si(111) surfaces. A transmission electron microscopy (TEM) study has been carried out on epilayers starting from different surface states.[13] The H-terminated 1×1 surface was found to give the best interface quality and to promote the γ-type structure. We performed an x-ray analysis of such a sample with ten nominal sheets.[14] The reflectivity profile confirmed that only γ-type stacking occurred. Each γ bulk reflection was retrieved after a 180° rotation of the sample around its normal, assessing that the twinned orientations occurred in equal proportion. First, questions arose on the interface itself, that is, how the first GaSe sheet is fixed on Si. Does the epitaxy involve the sticking of Ga atoms on top of the outer Si ones, leading to a half Ga-Se sheet strained on Si, or, does the epilayer start with complete sheets bound to Si via Si-Se interactions. The half sheet arrangement was supposed in Fig. 1, taking into account the two possible orientations. Referring to the two half sheets by using the labels ''*a*'' and ''*b*,'' while the related complete sheets will be denoted by ''*A*'' and ''*B*,'' we depicted the stacking ''*aAAA*'' and ''*bBBB*,'' expected from the γ phase.

An x-ray standing wave analysis has been performed on samples, with a 0.5 sheet nominal deposit. The half layer interface model was proposed, with the Ga atoms at 2.37 Å on top of the outer Si ones, and the Se atoms rather of type ''*a*.''[15] This technique was well suited to determine the Ga and Se atomic positions in the half foil. However, investigation of the further growth stages would be complicated, particularly in the case of in-plane relaxation, or various registries of the successive sheets. We present here x-ray diffraction results on samples involving 1.5 and 2.5 nominal sheets. Our models preserve the half foil arrangement, but reveal several types of domains. The growth does not consist in a uniform stacking of γ-type sheets.

Silicon samples were cleaned using high pH HF solutions, in order to get the flat Si-H surface. Two GaSe deposits were performed. The respective samples will be referred to in the following as I and II. The growth conditions are described in detail in Ref. 3; the sample was held at 450 °C while the beam pressures of the sources gave a Se/Ga flux ratio around 8. Evaporation time was chosen in order to produce 0.5 and 1.5 equivalent sheets; however, our x-ray results suggested coverages of 1.5 and 2.5 sheets.

III. X-RAY DATA COLLECTION

X-ray diffraction was performed using a grazing incidence geometry,[16] with the six-circle diffractometer, set on the synchrotron wiggler beam line DW12 at LURE (Orsay, France).[17,18] Samples were transported from the MBE chamber of the LPS laboratory to the diffractometer, in a primary vacuum environment. Previous experiments proved that the GaSe surfaces are strongly passivated, ensuring a negligible air contamination. The incidence angle was always maintained to the critical value for total reflection. The diffracted intensity was collected by an energy dispersive detector. The wavelength was chosen at 0.8856 Å, in order to prevent possible Ga or Se fluorescence signals in the data. The rods, that is, the intensity profiles along the surface normal at different in-plane positions, were registered with the same active area in the two samples, enabling direct comparison. Each value along the rod results from the integration of the profile obtained by a Θ scan, that is, a step-by-step in-plane rotation of the sample, across the Bragg position. We used the following reciprocal-space (RS) lattice, with respect to the conventional fcc Si lattice: $\mathbf{b}_1=\frac{2}{3}$ [112], $\mathbf{b}_2=\frac{2}{3}$ [121], and $\mathbf{b}_3=\frac{1}{3}$ [111]. A RS position will be denoted (h,k,l), according to the exchanged momentum $\mathbf{q}=2\pi\ (h\mathbf{b}_1+k\mathbf{b}_2+l\mathbf{b}_3)$. For silicon, rods at $(h,k)=(1,0)$ and (1,0) are not equivalent; for the orientation of Fig. 1, the bulk allowed reflections are expected, respectively, at $l=3p+1$ and $l=3p+2$ (p integer). Intensity was collected along both rods, as well as along the rod (2,1), for which Si bulk reflections arise at $l=3p+3$. The rod (1,0) will be presented together with rod (1,0), by using negative l values, and the two rods will be shortly referred to as rod (1,0).

The experimental rods (1,0), in the two samples, are presented after proper corrections in Fig. 2(a). First, the positive and negative l parts are almost symmetric, assessing the presence of two GaSe orientations, at 180° to each other, in nearly equal proportion. Second, the peaks along the rod appear at the same positions, in the two samples. They lie on the same background level, while rising in intensity from sample I to II. We expected 0.5 sheet for sample I, but the presence of peaks along the rod imply that at least one epilayer atomic plane is reproduced along the surface normal. The analysis will lead effectively to coverages of 1.5 and 2.5 sheets in samples I and II, respectively. The small peak at $l\ \pm 3.2$ in sample II appears as a faint shoulder in sample I. Besides, it was clearly identified on the profiles issued from l scans, which consist in measuring the intensity while keeping fixed the (h,k) value. The structure in sample II appears built with units of sample I, except in the region near $l=0$.

IV. STRUCTURAL MODELS AND CALCULATION

Sample I was first considered, and different models based on the Si-Ga-Se (half sheet) or the Si bulk-terminated interfaces were tested. It came out that the rod (1,0) was more sensitive to the structure at the interface than the rod (2,1), and the discussion will be focused on it. In order to simulate the peaks along the rod, we need to consider a complete Ga_2Se_2 sheet, at least, and, to account for the experimental symmetry, we must think about the two *A* and *B* orientations (see Fig. 1). The occurrence of the two types of domains is related to the GaSe(0001) and Si(111) $3m$ symmetries. If we compare with 3D heteroepitaxy on Si(111), we could expect a single orientation.[19,20] From thicker films, we know that the γ phase is favored; then, does the *A* and *B* differentiation

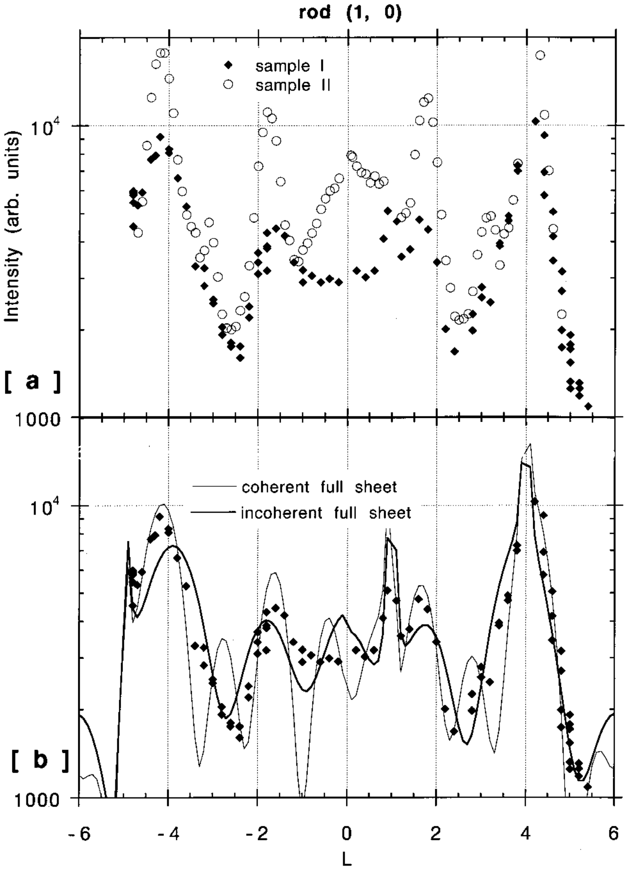

FIG. 2. (a) Experimental rods at $(h,k)=(1,0)$ for samples I and II (1.5 and 2.5 GaSe sheets, respectively). (b) Comparison between measured (sample I) and calculated intensities, according to two intermediate models based on the half sheet arrangement at the interface, with a complete sheet above. In one model, the full sheet scatters coherently with the layers underneath, while in the other one, it scatters independently.

appear after, or as soon as, the half-layer is formed? In the early growth stages, are other phases stabilized? Using the appropriate labels ''*a*'' and ''*b*'' for the half sheet, and ''*A*'' and ''*B*'' for the following complete sheet, we have to test various models, $a-A+B$, $b-A+B$, $a-A+b-B$, $a-B+b-A$, $a-A+a-B+b-A+b-B$, ... Moreover, we must examine different registries of the complete sheet in each model, that is, the in-plane displacement of the complete sheet with respect to the half sheet. Another difficulty emerges when considering that, if the half sheet is expected to be strained, the strain effect on the following foil may be relieved. We need thus to consider an in-plane accommodated, as well as a totally relaxed, complete sheet. The scattering contribution of the latter is then either coherent or incoherent with that of silicon. Of course, an intermediate situation may occur. We retained these two extreme cases only, in order to produce significant effects in the intensity. This reverts to estimating the integrated intensity by adding either the F's or the $|F|^2$'s of the two entities: the half sheet bonded to bulk Si and the next complete sheet. In the case of a single domain, F is the structure factor associated to the unit column from the ultimate surface layers to the deepest bulk ones.[21,22]

In the strain sheet hypothesis, we calculated the coordinates of the Ga and Se atoms expected from an elastic accommodation, using the interatomic distances in the γ bulk phase. The structure was supposed to be unchanged in the relaxed sheet case. We present the characteristics of the γ structure, in comparison with the β and ε phases. In the γ one, all the sheets are oriented the same and bound together according to the fact that the Ga atoms of each sheet are located at the same in-plane position than the topmost Se atoms of the sheet underneath. This is retained in the β structure, except that sheets are oriented alternatively at 180° from each other. In the ε phase, the sheets are oriented the same, but one out of two presents an in-plane displacement with respect to the γ situation.

V. DISCUSSION OF MODELS (SAMPLE I)

While preserving the structural features of the γ phase for the two successive units (half and full sheet), a coherent interface gives curves with minima too pronounced and supplementary maxima. When a unique orientation for the half sheet is assumed, the rod is clearly asymmetric. However, when considering two domains a-A and b-B in equal proportion, we produce on an average the magnitude of the peaks at $l=\pm 1.8$ and ± 4.2, as shown in Fig. 2(b). These peaks emerge from the atomic structure in a single sheet. On the other hand, the extra maxima at $l=\pm 2.8$ are related to the ''double'' sheet structure. To suppress them, one must consider the complete sheet scattering incoherently with respect to the half sheet. Actually, this hypothesis revealed itself suitable [see Fig. 2(b)]. Discrepancies remain: the peaks are slightly displaced with respect to the experimental ones, the shoulders at $l=\pm 3.2$ do not emerge, the curve near $l=0$ is badly reproduced, and the width of the peaks at $l=\pm 4$ is too large. This latter feature as well as the first one are related together and clearly demonstrate that the interface has not a totally ''incoherent'' character. We thus modified the model, while keeping the same amount of GaSe, that is, 1.5 sheets, so as to preserve the magnitude of the oscillations. The refinement was guided by the reliability factor $R=\Sigma\ [|F(\text{calc})|-F(\text{obs})]^2/\Sigma F(\text{obs})^2$. One can also consider the R' factor, given by the same formula, with the summation based on the absolute values instead of the squares. The reasons for choosing the R factors rather than the residual χ^2 are given in Ref. 21. For the latter, the systematic errors on $F(\text{obs})$ are taken into account, and estimated by the reproducibility of symmetry equivalent reflections (about 10%).

To start the fitting procedure, we considered half of the surface being covered by ''incoherent'' (i) domains, the half remaining consisting in ''coherent'' (c) domains. The two orientations at 180° (A and B) were preserved for each part of the surface. This model will be referred to as model 1. It already gives an overall R factor of 3% ($R'=12\%$, and $\chi^2=1.2$). Any modification of this base model did not significantly improve the overall R or χ^2 factors. The differences mainly affected the $[-3.5,-2.5]$ l region (and the symmetric l-positive region) of rod (1,0), while the $l=-1$ part of the rod was always unsatisfactory. The refinement shows that some structural features can be associated to some characteristics of the rod (1,0). The experimental shoulders at $l=\pm 3.2$ (that will change into sharp peaks in

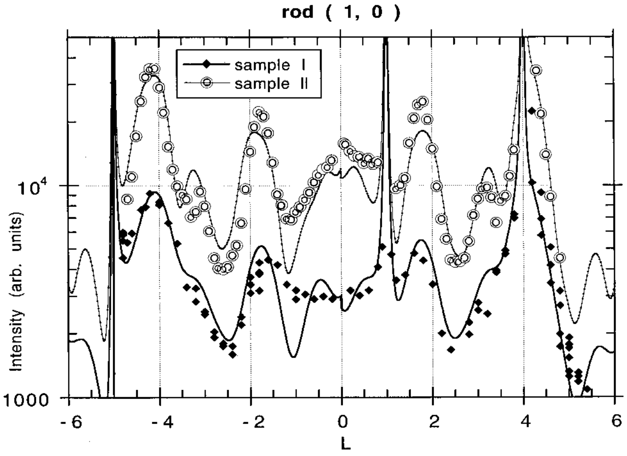

FIG. 3. Measured and calculated intensities of rod (1,0), according to the final models retained for samples I and II. The rod of sample II has been shifted at higher intensity values for sake of clarity.

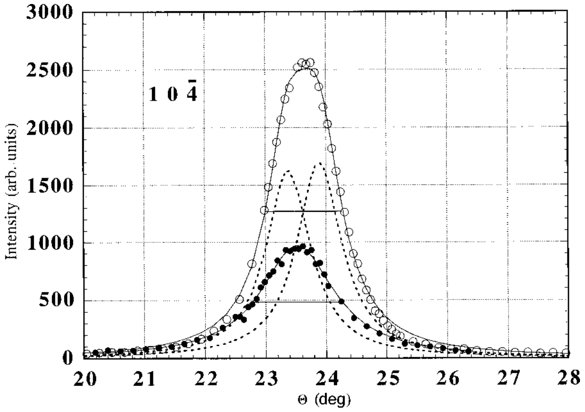

FIG. 4. Rocking curves obtained at $(h,k,l)=(1,0,\overline{4})$ for samples I (full circles) and II (open circles). The figure allows to compare the respective FWHM's, and shows the two Lorentzians used for the fit in sample II.

sample II), can be associated to a c-axis grown part, with more than 1.5 sheets (that is AA or BB). The difference in depth between minima at $l=-1$ and -2.5 is reproduced, if a second registry in the coherent part is considered (the one which is found in the ε phase). Finally, we retained the model that follows, limiting the presentation to a single orientation. In the coherent part, we have two equal contributions ''aA'' and ''a,'' while in the incoherent part we have ''aA'' and ''aAA.'' The rod is slightly improved if the half-sheet uncovered part consists entirely of type ''a.'' The final fit of rod (1,0) is presented in Fig. 3.

The presence of the AA (and BB) arrangement is also revealed by the evolution of the angular widths of the Θ-scan profiles. The correlation length acts as the inverse of the full width at half maximum (FWHM). A maximum of the FWHM was observed in the $l=\pm 3.5$ region; this is associated to the growth of the AA (or BB) structure. Another model was as satisfactory as the model retained. It does not affect the fact that the second complete sheet is found, on top of a relaxed one. This model consists in one third of the surface coherent of type ''aA,'' the remaining part being incoherent, with 45% of type ''aA'' and 25% of type ''aAA.'' We cannot exclude the presence of aA (or bB) regions, with the in-plane shift of ''A'' with respect to ''a,'' the same as in the ε phase. It must be underlined that information on the registry of the full sheets is lost, when considering the incoherent portion. The registry effects imply, thus, less than one half of the surface. Adding parameters in the calculation does not make sense, according to the experimental error bars. Besides, an intermediate situation between strain relieved and totally accommodated sheets probably occurs. As a matter of fact, we cannot extract the two contributions from the experimental Θ-rocking curves, which were fitted using single Lorentzian functions. The competition between the two strain states of the domains is however suggested by the weak values of the correlation length: from 120 to 230 Å. These values can be compared with those obtained in 3D-Si heteroepitaxy.[19] We could expect from the van der Waals bonding that, after the half-sheet is formed, the strain is relieved in the first full sheet. Our model suggests that this is mainly realized after a second full sheet forms. The questions that arise are as follows: how this second sheet will be completed over the surface, will the step formed act as a nucleation site, and what can be expected from the coherent domains in the further growth stages? Answers are provided by the analysis of sample II.

VI. THE NEXT STEPS OF THE GROWTH (SAMPLE II)

The Θ-scans at the different l values showed an increase in the correlation length compared to sample I. They could be fitted correctly only by use of two Lorentzian functions. Figure 4 shows the scans at $l=-4$ for samples I and II. Referring to a single Lorentzian fit, the FWHM changes from 1.48° to 1.2°, which leads to coherent domain sizes of 130 and 160 Å, respectively. Actually, a good fit in sample II requires two Lorentzians, each with an average FWHM of 0.88°. This gives a correlation length of 220 Å. The two center positions are shifted by 0.5°, and the lower-angle value corresponds to a lattice parameter close to the γ bulk one. Indeed, according to the diffractometer geometry, if one considers the bulk lattice values of γ-GaSe and Si, the related Θ angles should increase with l, but intermingle at $l=2.9$. The Θ values extracted from the fits behave as Si in sample I, while they are closer to GaSe in sample II, if one considers single curve fits. If one retains the Θ values of the two Lorentzians in the fits of sample II, one observes that each acts as expected from the Si or the GaSe lattice parameter. The domain width in sample II was found between 200 and 300 Å. Thus, we confirm with sample II, that domains still coexist with different lateral strain states. However, the average trend is that more GaSe layers have recovered their bulk in-plane parameter, as well as the domain width has improved.

From the structural investigation, we started from model 1 of sample I. The model consisted of a half sheet strained with Si, covered as a whole by one full sheet. The full sheet part was divided in two: one was coherent with the half layer, one was treated incoherently. Adding a second Ga_2Se_2

sheet to the incoherent portion led readily to a R factor of 2.5%. The basic feature of sample II is, one half of the surface consisting in one sheet (A and B) strained with Si-Ga-Se, the remaining half consisting in two sheets (AA and BB) completely relaxed on top of Si-Ga-Se. Finally, the best fit ($R=1.5\%$) was obtained with half of the coherent part of type aA (bB), the other half being aB (bA), and with $\frac{3}{4}$ of the incoherent part of type aAA (bBB), the last $\frac{1}{4}$ portion being $aAAA$ ($bBBB$). The rod (1,0) is shown in Fig. 3. In this model, the total coverage is 2.25 sheets, but the peaks at $l=\pm 1.8$ are underrated. We think that the coverage must be closer to 2.5, but did not find out how to place the extra portion so as to improve the fit. The occurrence of the aB (bA) stacking did not appear in refinement for sample I. This is the configuration found in the β phase. It is the only one which produces the continuous rise in intensity when approaching $l=0$ for rod (1,0). The presence of a β portion in sample II (as well as the presence of an ε one in sample I) can easily be explained in this stage of growth. Indeed, the growth is stopped before completion of the third sheet. The ε and β phases imply only two sheets in the unit cell, instead of three in the γ phase. In the low coverage regime, they can be thermodynamically stabilized with respect to the γ one. As in sample I, one could also retain a simpler model, however less satisfying: one third of the surface would be covered by coherent A (and B) sheets, the two last thirds would consist for 45% in AA (and BB) domains, and for 25% in AAA (and BBB) domains. Imaging experiments were performed on similar layer compounds.[23–25] Terraces were revealed, with steps one sheet in height, attesting that the growth proceeds before completion of the layers. In an elastic accommodation scheme, the stress is relieved by atomic relaxation along the z axis. However, the in-plane distortions from the bulk positions are a severe limit to the lateral extension of the domains. In 3D-3D heteroepitaxy, with increasing film thickness, the epilayer undergoes a transition towards plastic accommodation. If in-plane relaxation occurs in the very first steps of the growth, 3D nuclei can be formed, with eventual holes in the final film. What can be expected from the 2D-3D epitaxy presently described?

For foliated compounds, since the atoms at the surface of the sheets are in a stable configuration, flat surfaces are expected for the eventual nuclei. Besides, the number of dangling bonds on lateral sides must be minimized. This is related to the surface free energy. The mode of growth is also a function of the adhesion energies. In the beginning of growth, we must consider, on one hand, interface energies of the substrate and the crystallite with vacuum, and, on the other hand, bonding interactions between atoms of the adsorbate, as well as bonding between substrate and adsorbate. In our system, the weak bonding between the sheets compared to the intralayer one promotes lateral extension, but the size of the domains is limited by the substrate-crystallite interactions. The presence of the differently strained domains could be a way to relieve the interface stress. One can imagine a smooth transition between alternate strained-relaxed domains. The misfit is 2.6%. The two lattices are then almost commensurate with each measuring 142 Å. This is on an average as the correlation length is deduced from the x-ray reflections. If one superposes a Ga(0001) plane on top of Si(111), two regions appear, with the two types of atoms either in or out-of-phase. The substrate-overlayer interactions in the coincidence regions, by modifying the epilayer bulk positions, can produce totally accommodated portions. This will produce overlayer parts diffracting coherently with Si, the remaining parts being incoherent. The two types of domains can thus coexist. With further growth, the progressive relief of the strain should favor the lateral extension of the relaxed domains. The 2D character of the bonding is a relevant criteria for such an extension. Indeed, a distorted region can be hidden by the sheet which will grow next, thanks to the adjacent sheet. For the same reasons, the layers may rejoin between two terrace levels. As a matter of fact, TEM images gave evidence of bended (0001) planes inside the thick films.[26] Actually, the latter films revealed continuous with high crystalline quality.[13] These assertions are supported by the x-ray analysis of a 10 Ga_2Se_2 deposit.[14] It revealed that a portion strained with Si still existed, while most of the film was relaxed, with higher crystallinity. The remnant deformation was of $+0.9\%$ in the plane of the surface, and -0.2% out of plane.

VII. CONCLUSION

We manage to describe the GaSe/Si(111) interfaces involving 1.5 and 2.5 Ga_2Se_2 sheets, respectively. The models are based on the bulk GaSe phases, the transition with Si being insured by a half Si-Ga-Se layer. We can clearly argue that, first, two orientations rotated at 180° from each other take place with the same probability, and second, that a large amount of the Ga_2Se_2 sheets tend to relax laterally in the first stages of growth. The predominant structure is the γ one. The surface is found covered with domains, on an average, half being strained with Si, half being relaxed. Apart from the half sheet, the strained portion implies only one full sheet, in the two deposits, while the strain-relieved portion concerns one, two, or three sheets (with structure γ). Thus, the growth proceeds more easily on top of the relaxed domains, implying that 7.95 Å height steps are created. Last, concerning the uncovered full sheet (strained part), the presence of the β and ε arrangements is suggested.

One could expect from the van der Waals bonding, that after the half sheet interface is formed, growth proceeds by a 2D sheet by sheet growth, with total relaxation of the sheets. Evidence of differently strained domains, with the thicker ones being relaxed, as well as the presence of other phases than the γ one, show that the growth process is less ideal. One question remains: how the strained domains will develop in the further stages of the growth. Concerning the twinned orientations, we cannot speculate on the kind of junction that will be formed. They can limit the quality of the GaSe epilayers. In this sense, the use of III-V compounds as templates could be interesting, for instance GaAs(111), for which the $3m$ symmetry could promote a single orientation.

Our analysis shows that the surface diffraction technique is powerful, not only for determination of surface atomic configuration, but also for investigation of a complex interface, up to 10 atomic planes in the overlayer.

[*] Author to whom correspondence should be addressed. Electronic address: JEDRECY@LURE.U-PSUD.FR

[1] A. Koma, K. Sunouchi, and T. Miyajima, J. Vac. Sci. Technol. B **3**, 724 (1985).

[2] A. Koma, K. Ueno, and K. Saiki, J. Cryst. Growth **111**, 1029 (1991).

[3] L. T. Vinh, M. Eddrief, C. Sebenne, A. Sacuto, and M. Balkanski, J. Cryst. Growth **135**, 1 (1994).

[4] D. Fargues, L. Brahim-Otsmane, M. Eddrief, C. Sebenne, and M. Balkanski, Appl. Surf. Sci. **65**, 661 (1993).

[5] K. Schubert, E. Dörre, and M. Kluge, Z. Metallkd. **46**, 216 (1955).

[6] F. Jelinek and H. Hahn, Z. Naturforsch. **166**, 713 (1961).

[7] S. Bebazeth, Nguyen-Huy Dund, M. Guittard, and P. Laruelle, Acta Crystallogr. Sect. C **44**, 234 (1988).

[8] A. Bourdon, E. Bringuier, M. T. Portella, M. Vivieres, and N. Piccioli, Phys. Rev. Lett. **65**, 1925 (1990).

[9] M. Gauthier, A. Polian, J. M. Besson, and A. Chevy, Phys. Rev. B **40**, 3837 (1989).

[10] J. Petalas and J. A. Kalomiros, Phys. Rev. B **44**, 8694 (1991).

[11] J. E. Palmer, T. Saitoh, T. Yodo, and M. Tamura, J. Appl. Phys. **74**, 7211 (1993).

[12] The MBE and surface activities of the LPS Laboratory are presently realized in the LMCP Laboratory (Laboratoire de Minéralogie-Cristallographie, Universités Paris 6 et 7, France).

[13] A. Koëbel, Y. Zheng, J. F. Pétroff, M. Eddrief, L. T. Vinh, and C. Sebenne, J. Cryst. Growth **154**, 269 (1995).

[14] C. Trocellier and N. Jedrecy (unpublished).

[15] Y. Zheng, A. Koëbel, J. F. Pétroff, J. C. Boulliard, B. Capelle, and M. Eddrief, J. Cryst. Growth **162**, 135 (1996).

[16] R. Feidenhans'l, Surf. Sci. Rep. **10**, 105 (1989).

[17] M. B. Veron, Ph.D. thesis, Université P. et M. Curie, Paris, 1996; N. Jedrecy (unpublished).

[18] M. Lohmeier and E. Vlieg, J. Appl. Crystallogr. **26**, 706 (1993).

[19] N. Jedrecy, A. Waldhauer, M. Sauvage-Simkin, R. Pinchaux, and Y. Zheng, Phys. Rev. B **49**, 4725 (1994).

[20] H. von Känel , Mater. Sci. Rep. **8**, 193 (1992).

[21] E. Vlieg, J. F. Van der Veen, S. J. Gurman, C. Norris, and J. E. MacDonald, Surf. Sci. **210**, 301 (1989).

[22] Y. Garreau, M. Sauvage-Simkin, N. Jedrecy, R. Pinchaux, and M. B. Veron, Phys. Rev. B **54**, 17 638 (1996).

[23] F. S. Ohuchi, T. Shimada, B. A. Parkinson, K. Ueno, and A. Koma, J. Cryst. Growth **111**, 1033 (1991).

[24] J. E. Palmer, T. Saitoh, T. Yodo, and M. Tamura, J. Cryst. Growth **147**, 283 (1995).

[25] O. Lang, R. Schlaf, Y. Tomm, C. Pettenkofer, and W. Jaegermann, J. Appl. Phys. **75**, 7805 (1994).

[26] Y. Zheng (private communication).