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X-Ray Topographic Identification of Dislocation Nucleation Mechanisms in the Heteroepitaxial System GaAs/Ge

By

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The very low mismatched heteroepitaxial system GaAs/Ge has been studied by X-ray transmission and reflection topography, and the detailed steps of misfit dislocation formation are observed. Relation with growth rates and cooling rates are proposed and specific mechanisms are suggested.

1. Introduction

GaAs/Ge is a model system for heteroepitaxy. Actually, the lattice parameters are very close ($a_{GaAs} = 0.56533$ nm, $a_{Ge} = 0.56567$ nm) so the misfit (i.e. deformation at the interface $\varepsilon_{xx} = \varepsilon_{yy}$) is notably low. This allows high critical thickness for the epilayers, and the formation of a misfit dislocation (MD) network is progressive. Moreover, very perfect epitaxial films can be obtained even when the theoretical critical thickness is exceeded (metastability).

Under these conditions, the defect density is low and TEM observations are not well adapted to study the first stages of MD nucleation. On the contrary, X-ray topography is a large scale technique, allowing to observe each individual dislocation even with a near-to-zero density. So, in spite of its poor spatial resolution (≈ 5 mm), X-ray topography is a well suited technique to observe the successive steps of deformation in the film and in the substrate [1]. Of course, TEM observations are superior to XRT when the MD density is high compared to the XRT resolution.

In this paper we report X-ray topographic studies of the GaAs/Ge heteroepitaxial system grown by close-spaced vapor transport (CSVT). We identified two types of defects compared to two steps of MD formation and we try to correlate their occurrence with the growth rate and cooling rate of samples.

2. Experimental

The samples studied were 0.3 to 4 μm thick layers deposited on 200 to 260 μm (100) Ge substrates by the close-spaced vapor transport (CSVT) [2]. This growth technique is based on the thermal dependence of the chemical reaction [3]

$$2\,GaAs\,(s) + H_2O\,(g) \leftrightarrow Ga_2O\,(g) + As_2\,(\text{or } 1/2\,As_4\,(g)) + H_2\,(g)\,. \tag{1}$$

The GaAs source and the Ge substrate are enclosed in a reactor and separated by a thin spacer (1 mm in general). Graphite susceptors held them at different temperatures: 800 °C

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Table 1
Sample characteristics

samples series	growth rate (GR) (μm/min)	cooling rate (CR) (K/min)	thickness range (μm)
1	0.08	40	0.7 to 4,0
2	0.04 to 0.05	50	0.3 to 0.8
3	0.04	5 to 50	constant 0.8

for GaAs and 750 °C for Ge. At 800 °C, H_2O reacts with GaAs according to reaction (1). The reverse reaction occurs on Ge. The driving force of the deposition is the temperature difference of 50 K which allows a net transport of material from the source to the substrate. The growth rate (GR) can be adapted by acting on the H_2O gas flow. The cooling rate (CR) is controlled by the thermal sequence applied to the substrate after the reactor was closed [2].

Several sets of samples have been studied; their characteristics are given in Table 1.

Two topographic techniques were applied, giving complementary information:

– XR transmission topography (XRTT), using planes perpendicular to the interface (Lang setting, Fig. 1A; radiation AgKα, $\lambda = 0.056$ nm). As the images correspond to a summation of film and substrate contributions, they are depth-dependent, so we get information about the whole sample;

– XR reflection topography (XRRT), using planes slightly inclined relative to the interface (Berg-Barrett setting, Fig. 1B). With a large enough wavelength (we used CuKα radiation, $\lambda = 0.154$ nm), diffracted beams on (*hkl*) planes from the film and the substrate can be separated, and two different images can be obtained.

3. Results and Discussion

As it has been reported in [1], two types of defects were observed in our samples, corresponding to two steps of MD formation:

(i) threading dislocations which can give an initial network of interfacial MD with a maximum developed length of 10^3 cm in a 1 cm^2 interface;

(ii) short edge segments which seem to be related to a second MD network, the development of which can exceed 10^4 cm in a 1 cm^2 sample.

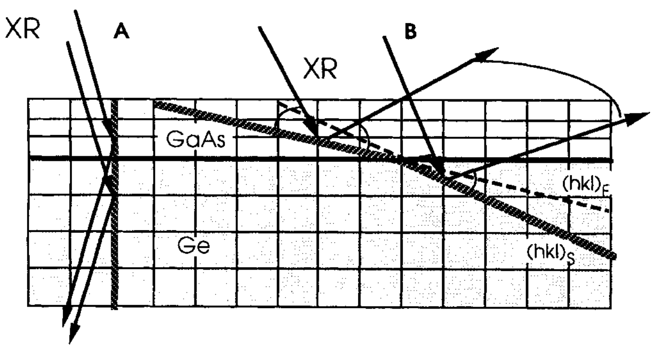


Fig. 1. Epitaxial system with transmission (A) and reflection (B) positions of diffracting planes

3.1 Threading dislocations

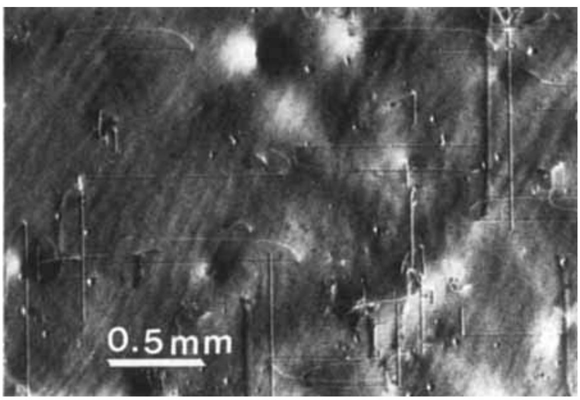
The first process is the one described by Matthews et al. [4]:

– below a critical value t_c, no dislocation at all is developed at the interface;

– beyond t_c, a few dislocations are nucleated from threading ones and straight arms elongate in the interface; the maximum development of this step is reached when each threading dislocation has produced a long interfacial line throughout the whole sample; but the as-grown dislocation density in Ge substrates is very low, so sources of this type are not very numerous and interactions between MD are exceptional; therefore multiplication processes are not efficient and no noticeable relaxation can be obtained, as could be expected. The measured critical thickness is about 1.2 to 1.3 μm, i.e. four times larger than the theoretical value $t_c = 0.3$ μm obtained from the model by Matthews et al. [4]; it depends on experimental conditions and chiefly on the cooling rate: the value of 1.2 μm is correlated with usual "quick" cooling (about 40 K/min). Such an experimental critical value was already noticed by Frigeri et al. [5] in samples grown by MOVPE with high growth rates (higher than 0.1 μm/min), which is close to the GR of our first series (Table 1).

In most samples observed by X-ray transmission topography, it can be noticed that these interfacial MDs present an unusual configuration: they are very often connected with arms developed in the substrate (Fig. 2a). So the defect has a "hairpin" shape which is closely related to stress field variations undergone by the film–substrate system (Fig. 2b).

This particular configuration was also observed in the Ge(B)/Ge system by Prokhorov et al. [6]. These authors have demonstrated, following Matthews, the existence of critical



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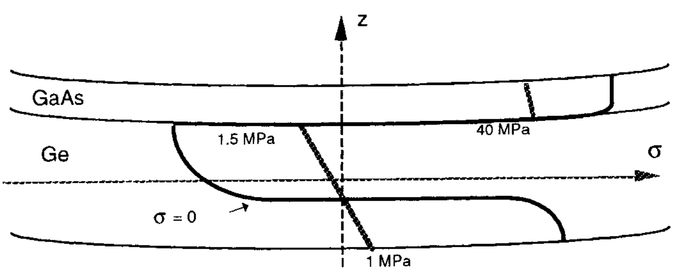

b

Fig. 2. a) Hairpin dislocations (XRT topography, 220 reflection vector); b) stress field through the sample (the z-axis is the [001] growing direction) and shape of associated dislocation

film thicknesses t_c^{II} and t_c^{III} corresponding to the dislocation development in the compression and tension parts of the substrate. This mechanism was rarely reported because it requires high thicknesses and metastable layers to be obtained, which exist only in very low misfit systems. It has been calculated [1] that a thickness of 1.2 μm is required in GaAs/Ge for the complete development of a hairpin.

Nevertheless, Prokhorov et al.'s calculations showed that hairpin configurations are energetically possible, but no kinetic argument was proposed. As the lengths of the two arms are often the same, the hairpin shape suggests that the dislocation velocities are similar in GaAs and Ge. However, it must be taken into account that stresses acting on interfacial GaAs arm, on one side, and Ge internal arm, on the other, are very different: 40 and 1 MPa, respectively, at room temperature (close to the deposition temperature the lattice parameters are not known with enough precision to determine the applied stresses, but the stress ratio must be the same). Unfortunately precise velocity measurements are not available for high temperatures and low stresses in Ge, and extrapolations are tricky: one can only say that velocities are expected to be quite higher in GaAs under 40 MPa than in Ge under 1 MPa. This would not imply a hairpin shape, so the most probable reason is that the velocity in GaAs would be limited by the „length effect" [7]: as the layer thickness is lower than or at least equal to 1 μm, the leading threading segment in the layer is no more long. In that case the dislocation velocities would not be controlled by kink collisions: they would be length dependent and could be appreciably smaller than the bulk velocity invoked above. Although the limit of this effect is not exactly known in GaAs, it was estimated to be higher than 1 μm [8], which means that this length effect might be effective in our case.

A last series of samples has been studied, in which the cooling rate was 5 K/min and the layer thicknesses in the range of 0.3 to 0.8 μm. The saturation of the first step (i) (as far as it can be concluded from XRRT images, since transmission images are not available for these samples) is not obtained when the second step (ii) occurs. It may imply that the development of the substrate arm of the hairpin takes an active part in this first mechanism, so that it cannot occur when the layer thickness is under t_c^{III}.

A study of the residual contrast from Berg-Barrett images has revealed that these segments would be, very likely, 90° Shockley partials. This result has been confirmed by the observation of stacking faults in GaAs layer by TEM.

3.2 Short edge segments

Small edge segments parallel to the $[0\bar{1}1]$ direction were observed in samples of various layer thicknesses (Fig. 3). Because of the total extinction observed on a $(0\bar{2}2)$ transmission topograph, it was obvious that these defects had a pure edge Burgers vector; so only three possibilities were credible: $(a/2)\,[011]$ (Lomer-Cottrell locks), $(a/3)\,[111]$ ($(a/3)\,[0\bar{1}\bar{1}]$) (Franck loops) or $(a/6)\,[211]$ ($(a/6)\,[2\bar{1}\bar{1}]$) (Shockley partials). The Lomer-Cottrell sessile defect can be formed only by interaction of two glissile 60° dislocations as the long straight segments produced by the first stage of deformation: but their density is so weak and their average length so high that this mechanism cannot be taken into account for the nucleation of these short segments. Franck loops are often produced by accumulation of interstitials or vacancies, but were never observed in GaAs films. A study of the residual contrast from Berg-Barrett images has revealed that these segments would be, more probably, 90° Shockley partials. This result was confirmed by our own TEM observations of stacking faults bounded by Shockley partials in GaAs layers, and now by the TEM observations of Frigeri et al.

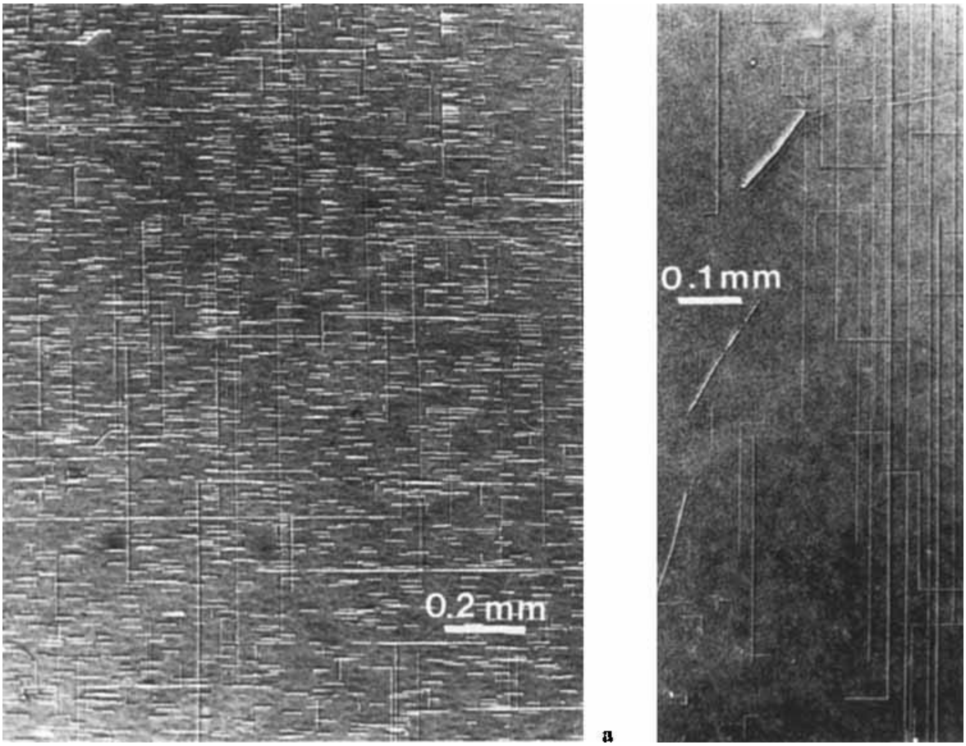

Fig. 3. a) Edge segments (XRR topography, 422 reflecting vector); b) nucleation of MD from edge segments (XRRT, 422 reflecting vector)

[5], who established that stacking faults (SF) existed only in low growth rate samples (<0.1 μm/min). We noticed such defects with a density of $\approx 4\times10^4$ cm^{-2} in low growth rate samples (series 2 and 3, GR $\approx$ 0.04 μm/min) and a few of them (density $\approx 10^3$ cm^{-2}) in first series samples. So we consider that these short defects are more likely stacking faults emerging at the surface and limited at or near the interface by $(a/6)\,[211]$ Shockley partials.

In our samples, these defects seem to be related to long straight segments lying in a normal [011] direction (Fig. 3b); this suggests the occurrence of a second MD process. Frigeri et al. [5] also observed by TEM final configurations associating stacking faults and long glissile dislocations; they suggested that these faults were created by dissociation of the dislocation lines. However, as we observed the short segments by XRT in samples free of MD, we think the most probable mechanism would be the reciprocal one: nucleation of long straight dislocations from the short segments (SF). The very precise interaction giving rise to this special configuration is of course impossible to deduce from XRT images, but one possible mechanism might be based on the nucleation and glide of an $(a/2)\ \langle 110\rangle$ dislocation produced by the decomposition of the edge partial bounding the stacking fault in the layer, as follows (Fig. 4):

$$\frac{a}{6}[2\bar{1}\bar{1}] \rightarrow \frac{a}{2}[1\bar{1}0] + \frac{a}{6}[\bar{1}2\bar{1}]\,.$$

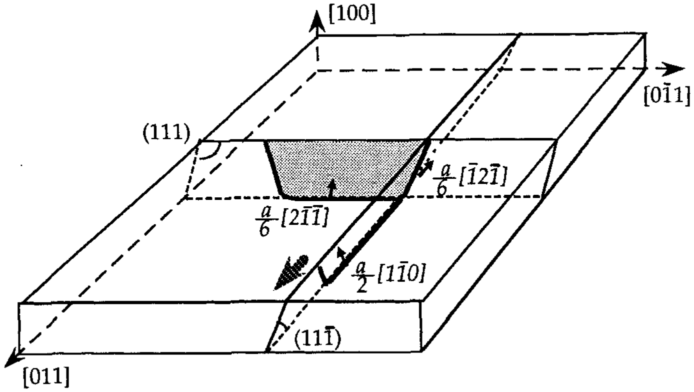

Fig. 4. The second nucleation mechanism: dislocation nucleation from the edge bounding partial of a stacking fault

This dissociation could not be spontaneous, since it requires a substantial amount of energy. However, as the layer is in a metastable state, the stored elastic energy might be large enough to allow such a mechanism to occur. However, due to the poor resolution of XRT, the actual occurrence of the correlation between short edge segments and MD can also be questioned. In this case, the nucleation of long straight dislocations should be found in another mechanism, such as half loop nucleation from the free surface, but this will require a similar energy as involved in the case of SF decomposition. Anyway, the occurrence of the stacking faults would be explained.

Despite of the fourfold symmetry, in all samples these defects were observed in only one direction (for example [0$\bar{1}$1]): none was developed along [011]. This is highly coherent with the stacking fault hypothesis. As a matter of fact, as it was shown in several papers (for example [5]), because of the stress acting on partial dislocations, the dissociation is favourable in one $\langle 011 \rangle$ direction of the (100) plane and unfavourable in the other one.

An attractive explanation of the presence of stacking faults ([1, 5]) correlates their formation with the 2D–3D growth transition: in three-dimensional growth, stable nuclei are formed by deposition of atoms on facets. Errors would be easy when stacking on $\{111\}$ facets because the energy difference between correct and incorrect atomic positions is very small. Such errors would give rise to stacking faults as soon as neighbouring nuclei join. This process is not valid in two-dimensional growth, which is the case of high growth rates.

4. Conclusion

It was shown that XRT allows to get a lot of information about the early stages of misfit dislocation formation in epitaxial systems. We suggest the existence of two distinct nucleation processes for MD in GaAs/Ge. The first one is based on heterogeneous nucleation from Ge threading dislocations; the various steps were observed, and original "hairpin" configurations were analysed. The maximum density obtained from this mechanism is actually too low to get any stress relaxation. The second one is suggested to imply edge segments localised in the layer, which are interpreted as stacking faults. Their dissociation would give rise to glissile MD network whose density is much too high to be resolved by X-ray images.

More work must be done to give a full analysis of these mechanisms:

– about the stacking fault hypothesis a forthcoming argument would be obtained from quantitative contrast analysis and related calculations (presently in progress); the validity of this hypothesis is not definitely proved, but until now it seems to be the "less bad" one.

– Strain measurements of the system as a function of temperature are planned in order to determine the variations of misfit and metastability with temperature.

References

[1] N. Burle, B. Pichaud, N. Guelton, and R. J. Saint-Jacques, Thin Solid Films, accepted.

[2] G. Lalande, N. Guelton, D. Cossement, R. G. Saint-Jacques, and J. P. Dodelet, Canad. J. Phys. **72**, 225 (1994).

[3] D. Côté, J. P. Dodelet, B. A. Lombos, and J. I. Dickson, J. Electrochem. Soc. **133**, 1925 (1986).

[4] J. W. Matthews, A. E. Blakeslee, and S. Mader, Thin Solid Films **33**, 253 (1976).

[5] C. Frigeri, G. Attolini, C. Pelosi, and F. Longo, 1st Conf. Materials for Microelectronics, Barcelona, October 17 to 19, 1994, to be published in Mater. Sci. Technol.

[6] I. A. Prokhorov, B. G. Zakharov, V. S. Mashin, and I. L. Shulpina, J. Phys. D **26**, A76 (1993).

[7] A. George and J. Rabier, Rev. Phys. appl. **22**, 9, 941 (1987).

[8] D. Caillard, N. Clement, A. Couret, A. Androussi, Y. Lefevre, and G. Vandershaeve, Inst. Phys. Conf. Ser. No. 100, 403 (1989).

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