Critical thickness of Ge / GaAs(001) epitaxial films

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Epitaxial single crystal films of Ge, with thickness from 0.2 to 2.4 μm, were grown on GaAs (001) by rf sputtering. These layers were characterized by High Resolution X-Ray Diffraction (HRXRD). Measured rocking curves show that pseudomorphic samples with good structural quality can be obtained by this growth technique. Asymmetrical reflections (115) and (-1-15) are used to determine the in-plane and in-growth lattice parameters of the grown films. From the behavior of these parameters and Ge diffraction peak broadening, with the layer thickness, an experimental value of 1.8 μm is obtained for the critical thickness of Ge grown on GaAs (001).

Keywords: Heterostructure IV/III-V; critical thinks; High resolution X-ray diffraction

1. Introduction

Recently, it has been experimentally observed the existence of two critical layer thicknesses when growing homoepitaxial films of Ge(001) by MBE (molecular beam epitaxy) at low temperatures: h_1 (that from this point we will call h_c), that is the film thickness at which bulk structural defects are first observed, and h_2 , the film thickness at which the entire layer transforms from epitaxial to amorphous [1]. Even though Ge epitaxy has been widely studied, there is a renewed interest in understanding the growth process of Ge. It is expected that the obtained information aids to solve unclear growth aspects of more complex materials, such as the IV-IV alloy $Ge_{1-x}Sn_x$, which has to be grown under non-equilibrium conditions. This alloy has promising applications for mid-infrared optoelectronics , and has been grown on Ge and GaAs. However, device quality samples can still not be obtained, because of the large lattice mismatch between Ge and Sn and the possible appearance of two different layers in the same grown film as mentioned above for homoepitaxial Ge. Therefore, the growth of homo and heteroepitaxial Ge thin films is still of great interest, since, it has been said, that there is a relation between the epitaxial critical thickness (h_2) and the critical thickness (h_c), [2] first introduced by Van der Merwe, at which misfit dislocations appear to release the strain caused by the lattice constant mismatch between the layer and its substrate.

In this work, we obtain the experimental critical thickness h_c of Ge / GaAs(001) films grown by rf-sputtering at 540°C. Although Ge/ GaAs films have been widely studied, as mentioned above, very different values for this h_c have been reported, from 1.2 to 1.6 m, [3-5] and to our knowledge, an accurate value of this critical thickness, systematically obtained by High Resolution X-Ray Diffraction (HRXRD), has not been reported.

Our films were analyzed by HRXRD to obtain their lattice parameters and to use the "full width at half maximum (FWHM)" of the diffraction profiles to estimate this critical thickness. The determination of h_2 for Ge/ GaAs films is the next step in this work.

2. Theory

GaAs and Ge are crystalline cubic materials with room temperature lattice constants 5.6535 Å and 5.6577 Å respectively [6]. When growing Ge on GaAs, the lattice mismatch produces a compressive bi-axial stress in the growth plane, which is compensated with an enlargement of the Ge lattice in the growing direction (001).

In order to obtain the film lattice parameters, the diffraction of the asymmetric planes (115) and (-1-15) was measured by HRXRD. These reflections were chosen over the other commonly used asymmetric planes (224), because they are more intense and allowed us to accurately determine the position and width of the Ge layer peak even for the thinner films.

The parameters were obtained using the Macrander formulas, which use asymmetric planes reflections [7]:

$$a_{\perp} = a_0 \frac{\cos\gamma}{\cos(\gamma + \Delta\tau)} \frac{\sin\theta_0}{\sin(\theta_0 + \Delta\theta)} \quad (1)$$

$$a_{//} = a_0 \frac{\sin\gamma}{\sin(\gamma + \Delta\tau)} \frac{\sin\theta_0}{\sin(\theta_0 + \Delta\theta)} \quad (2)$$

where a_o represents the GaAs bulk lattice constant, $a_{\parallel}$ and $a_{\perp}$ are the in-plane and in-growth Ge layer lattice parameters, θ_o is the Bragg angle, γ is the angle between the reflection direction (i.e. (115), (-1-15)) and the growing plane, $\Delta\tau$ and $\Delta\theta$ are given by

$$\Delta\tau = \frac{\Delta\omega^- - \Delta\omega^+}{2} \quad (3)$$

and

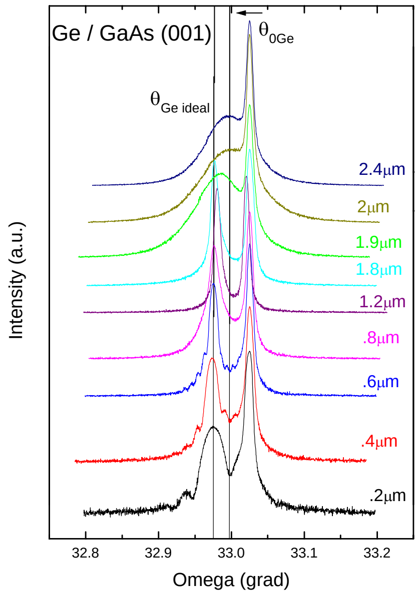

Figure 1. Rocking curves for Ge/GaAs at different thicknesses.

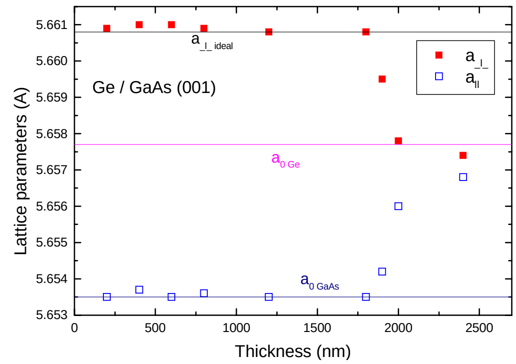

Figure 2. Thickness vs lattice parameters.

$$\Delta\theta = \frac{\Delta\omega^{-} + \Delta\omega^{+}}{2} \quad (4)$$

with $\Delta\omega^{-}$ and $\Delta\omega^{+}$ representing the angular separation between the diffraction peaks of the film and its substrate, in ω^{-} and ω^{+} geometries, respectively.

3. Experimental procedure

The samples used in this study were all grown in a RF-Sputtering system. Before the Ge layers were grown, the surface oxides of the GaAs substrates were desorbed at 600°C for 2 minutes. Then the temperature was decreased to 540°C to start the growing process. Argon (Ar) plasma was used as the pulverization agent using an RF power of 50 W and a pressure of $2x10^{-2}$ mbar. Nine films of different thicknesses were grown under these conditions. Once the films were grown, they were analyzed by HRXRD, to obtain the lattice parameters. The interference "Pendellösung" fringes [8] of the diffraction profiles were used to determine the thinner films thickness.

Due to the missing Pendellösung fringes for films of thicknesses larger than 0.4 μm, we had to estimate their thickness based on a linear interpolation, considering the 200 A/min (3.33 A/seg) growth rate of the three thinner films. This interpolation is further supported by the fact that the use of a cantilever profilemeter, to measure the layer thickness of the thicker films, gives values within +-0.5 % of the thickness estimated by the interpolation.

4. Results

In Fig.1 the rocking curves of (004) reflections from Ge films grown with different thicknesses are shown. We can clearly observe the Bragg peaks for the Ge and the GaAs substrate. In the first three layers we can observe a Pendellösung interference pattern, which is characteristic of high crystalline quality. We can notice that the Pendellösung fringes disappear for the thicker layers (h> 0.6 μm). The vertical lines, θ_{Ge} and θ_{Ge_ideal}, indicate the diffraction angle of bulk and pseudomorphic Ge (on GaAs) peaks respectively. We can also observe that the separation (Δθ) between the layer and substrate peaks decreases when the film thickness increases (h>1.8 μm). This is due to the relaxation process and indicates that the film has overcome its critical thickness. Table I shows growing time, lattice parameters, relaxation coefficient "r" and the thickness for each grown film. This coefficient "r", [7] gives a measure of the average relative difference of the in-plane and in-growth parameters respect the substrate lattice constant during the relaxation process and is obtained from the experimental lattice parameters by:

$$r = \frac{a_{\parallel} - a_{O}}{a_{\perp} - a_{O}} \quad (5)$$

As we can see in table I, the first six films are strained (r=0). Meanwhile, the relaxation process has started in sample 36, (r>0) which indicates the presence of dislocations. The two last samples are quite relaxed, which means they have surpassed the critical thickness h_c.

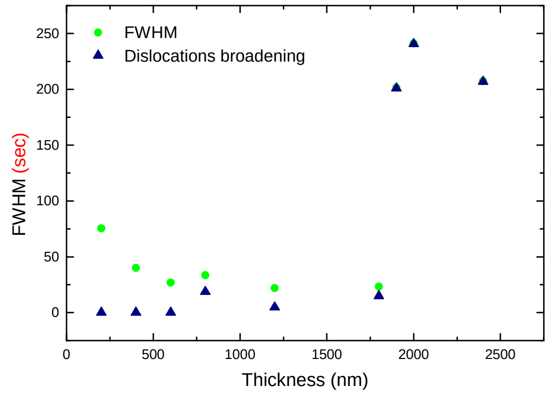

Figure 3. FWHM and dislocations broadening vs layer thickness.

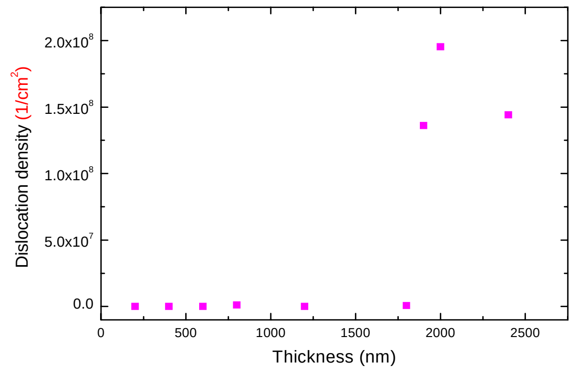

Figure 4. Dislocation density vs. layer thickness.

Figure 2 shows the behavior of the in-plane and in-growth lattice parameters with the layer thickness. It can be seen that for small thicknesses, $a_{||}$ and $a_{\perp}$lie around the values of the substrate (a_o) and ideal pseudomorphic Ge on GaAs ($a_{\perp}$) lattice constants, respectively, marked with horizontal lines in the figure. As the thickness is increased, both parameters approach to the value of bulk Ge, however they do not take the same value, as even the thicker film is not fully relaxed (r=0.79). From these data, we have that h_c is 1.8 ± 0.05 μm, for it is when the Ge lattice parameters change radically as the film relaxes generating dislocations.

To support this result, we present figure 3, which shows with full circles, the measured FWHM and with full triangles the broadening associated with dislocations present in the layer, as it is commonly described in literature [9,10]. By carefully observing fig. 3 we have that the critical thickness value lies around the one mentioned above. Finally, in figure 4, we can see how the dislocation density highly increases once the layer thickness goes over 1800 nm.

5. Conclusions

As the HRXRD rocking curves show, good structural quality Ge films can be grown by rf-sputtering. From the behavior of the in-plane and in-growth lattice parameters with the thickness of the Ge grown layer, a value of 1.8 ± 0.05 μm is found for the critical thickness of Ge / GaAs (001) films.

Further experiments have to be performed in order to establish if there is a relation between the value found for the critical thickness (h_c) and the epitaxial critical thickness (h_2) that it is observed in Ge homoepitaxy.

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Table I. Main data for the nine films grown in this work. The first row shows ideal lattice parameters of a pseudomorphic Ge/GaAs film.

Sample	**Growth time** (min)	+/- 0.0002 (A)	+/- 0.0002 (A)	**r**	**Layer Thickness** (A)
Ideal	-	5.6608	5.6535	-	-
27	10	5.6609	5.6535	0	2000
28	20	5.6610	5.6537	0	4000
29	30	5.6610	5.6535	0	6000
26	40	5.6609	5.6536	0	8000
17	60	5.6608	5.6535	0	12000
33	90	5.6608	5.6535	0	18000
36	95	5.6595	5.6542	0.17	19000
35	100	5.6578	5.6560	0.71	20000
34	120	5.6574	5.6568	0.79	24000