Study of Surface Kinetic Effects in the MBE Growth of III-V Compounds by RHEED Analysis

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A comparative surface kinetic study at MBE on (100) GaAs, AlSb, GaSb and InSb surfaces has been presented. The growth mode evolution was determined in all cases by measurements of RHEED oscillation intensity. The temperature dependence of surface diffusion length for Al, Ga and In adatoms on the GaAs, AlAs, AlSb, GaSb and InSb was founded. The result shows that interface roughness in heterostuctures depends strongly not only on substrate temperature but also on growth rate.

I. Introduction

Reflection high energy electron diffraction (RHEED) is one of the mostly useful methods for *in situ* monitoring growth processes in molecular beam epitaxy (MBE). Such characterisation is helpful in several aspects, including determination of growth rate and molar fraction, adatom diffusion length, diffusion coefficients, micromorphology and growth mode transitions[1-3]. These characteristics are usually extracted by measurements of oscillation frequency and temperature transition between the growth modes. Despite the great field of qualitative applications, just few reports apply quantitative RHEED intensity analysis to investigate surface kinetic.

In this work we investigate the substrate temperature (T_s) and growth rate (V_G) effects on RHEED oscillation amplitude. We have performed a comparative study of the growth kinetic for GaAs, AlAs, GaSb, AlSb and InSb, grown on same substrate conditions. The surface diffusion length, for all compounds, has been presented.

II. Theoretical considerations

Recently, several articles have been published describing theoretical predictions of elementary growth process in MBE[4-7]. Such predictions show the possibility of explaining RHEED oscillations as function of surface supersaturation ratio $\alpha = n/n_0$ (n is surface concentration of Ga adatoms and n_0 is equilibrium Ga concentration) and diffusion length of adatoms (λ). The behaviour of RHEED oscillations during the growth can be easily described by a damping harmonic oscillatory equation expressed by:

$$A = A_0 \exp(-\sigma) \cos(\omega t + \psi) \qquad (1)$$

where A_0 is the first oscillation amplitude, σ the amplitude decay rate, ω the oscillation frequency corresponding to the effective incorporated flux. The parameter ψ is the phase shift of the first oscillation. This can be related with changes of lattice periodicity of surface atoms (surface superstructure) or surface disordering, at beginning of growth process.

In the case of the formula 1, A_0 should be related with differences of the square distance between terrace and 2D nuclei[1]. According to MBE growth theory[4-7], a number of nuclei with radius r_c and average distance between centres 2λ is expected to be present on the central part of the terrace at the growth beginning, when α is more than the critical value (α_c) (see Fig. 1a). At this moment RHEED intensity corresponds to a maximum. A minimum of RHEED signal will correspond to the moment of nuclei coalesces (see Fig. 1b.), at deposition of half monolayer. Since the adatoms are incorporated also in the terrace steps, the

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area nearby in a distance of 2λ will be free of 2D nuclei. Modifications of substrate temperature and growth rate affect λ and α values, and consequently the square distance between terrace and 2D nuclei. Therefore the amplitude of RHEED oscillation is related with λ value. The surface diffusion length (λ) is given in Ref. 4. by equation:

$$\lambda = a\exp((E_{des} - E_D)/2kT_s) \quad (2)$$

where a is the adatom jump distance, E_{des} is the desorption energy and E_D is the activation energy for surface diffusion. It is clear by formula above that the diffusion length is an exponential function of substrate temperature T_s.

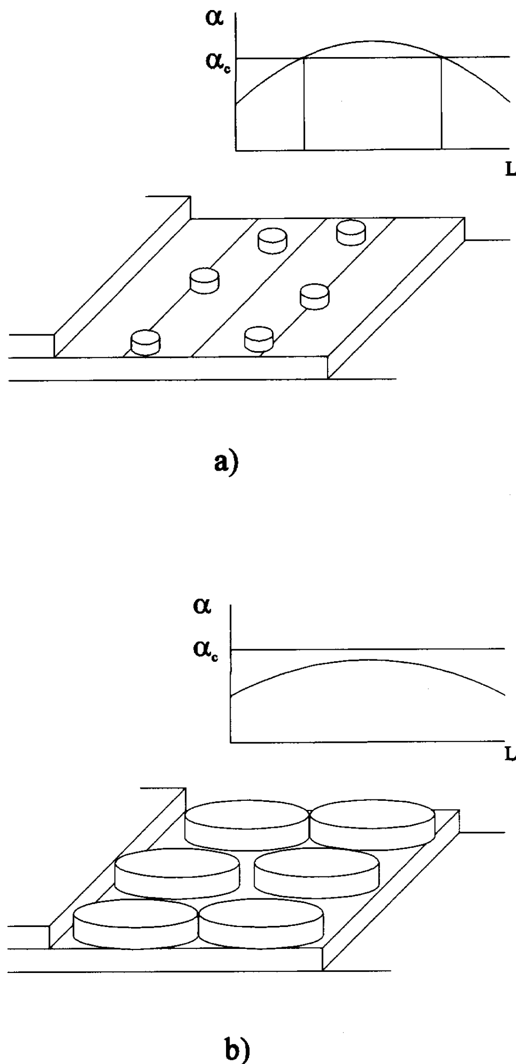

Figure 1. Schematic diagram of surface morphology showing 2D nucleation at cases a) at growth beginning and b) half monolayer deposition. The insets show the respective dependence of a along terrace at the beginning of growth process (a) and at half monolayer deposition (b).

According to this growth model, RHEED oscillation amplitude is should depend on λ and α. This relation between RHEED intensity and the main growth parameters in MBE gives the possibility of studying the growth mode evolution by determination of $A_0 = f(T_s)$ and $A_0 = f(V_g)$.

III. Experimental proceedings and results

All set of experimental results in this letter were produced into one growth cycle on the GaAs substrate ((100) plane), misoriented 20' (corresponded terrace length L= 48.5nm). The small substrate misorientation is necessary to satisfy the condition for observation of RHEED oscillations, when $2\lambda < L$. RHEED analysis were performed in several stages during the growth of GaAs, AlAs, AlSb, GaSb, and finally InSb in sequence. To obtain an atomic smooth morphology and reduce dislocation density, we have grown each layer with a thickness of ~ 1.5 μm. In order to measure the growth mode evolution, a difractometer (possible to accelerate electrons up to 30 KeV) and microphotometer were also connected to the system.

The first set of experiments were basically the observation of RHEED oscillations as a function of T_s, for a constant III/V flux ratio and growth rate (1μm/hour). For all cases the III/V flux ratio were corresponded to group V element stabilised surfaces. In this case surface kinetic and growth rate are determined by the group III element. This approach simplifies the growth system, reducing the analysis to a single component. Therefore it is possible to determine the surface diffusion length of metal adatoms on the surface of $A_{III}B_V$ compounds.

Fig. 2a shows the evolution of RHEED oscillations during growth of GaAs for different T_s values. It is observed a strong dependence of A_0 with T_s. In addition, we have noticed two other phenomena. First, when T_s is reduced below 515°C it is possible to observed a phase shift in the amplitude oscillation, corresponding to surface structure transition from (2x4) to c(4x4). A partial transition of surface structures, on wide terraces, produces a phase shift in the RHEED oscillations detected earlier then would be possible by pattern observations. The same phase shift was observed also during the growth of GaSb and AlSb, in a T_s range near the temperature of superstructure transitions. A second remarkable event is observed on the

plot of RHEED oscillations showing an interference like structure. We believe that such interference of RHEED signal is evidence of surface step interactions.

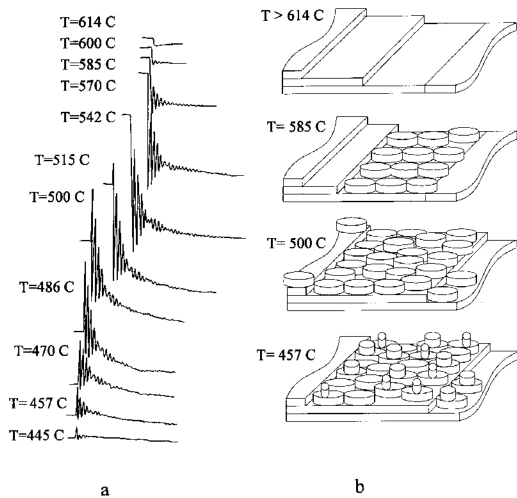

Figure 2: a) RHEED oscillation dependence on substrate temperature for GaAs. b) Surface morphology evolution as function of substrate temperature. Such morphologies are sketched at the moment of alfmonolayer deposition.

Fig. 2.b sketchs the surface morphology at the deposition of half monolayer (maximal roughness on heterointerface) for different T_s. Qualitatively, it is possible to identify five different kinds of growth modes and relate them to different surface morphologies. We have seen that for $T_s > 614°C$ oscillations are absent and growth mode keeps the step flow propagation. A 2D nuclei appear at a critical $T_s \cong 610°C$, when $\lambda = L/2$, due to the fact that α value is higher than α_C on the centre of terraces. For such kind of mixed growth mode, exists a competition of adatom incorporation between nuclei and steps. Reducing T_s further, the 2D nuclei area enlarges on all terraces and growth turns to be a 2D nucleation. At this moment the RHEED oscillation intensity has a maximum. Reducing even more T_s, α value increases so much that on the top of first level nuclei starts to be created a second level, and intensity goes down. This mode can be regarded as a many level mode one. Finally, for lower T_s, λ decreases down to zero and the growth mode turns to be a 3D mode and the oscillations disappear. In our experiments and for GaAs, this temperature was determined as $T_s \cong 407°C$.

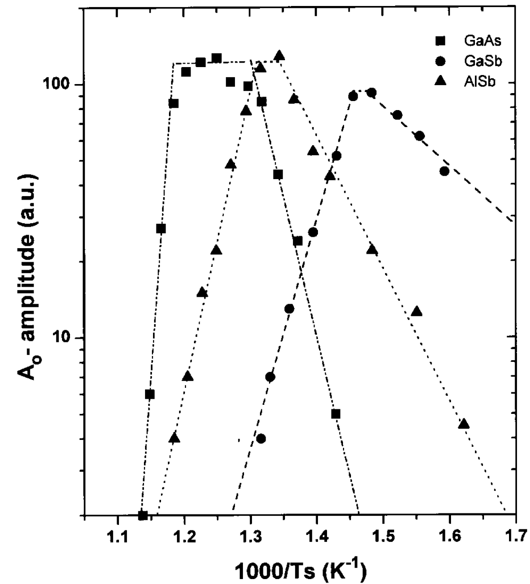

Figure 3: RHEED oscillation amphtude (A_0) as function of substrate temperature, for the GaAs, GaSb and AlSb compounds.

As we have seen before, the evolution of mode can be identified from the amplitude of RHEED oscillations as function of T_s. The A_0 dependence as a function of T_s, for the case GaAs, AlSb and GaSb is shown in Fig. 3. We have also performed measurements on AlAs and InSb but they are not shown here. The results in Fig. 3 show also evidence of the five different growth modes. The experimental data, with good accuracy, can be fitted in log plots. From Fig. 3, by transition from step flow propagation to mixed growth mode, it is possible to extract the value $Q = E_{des} - E_D$ for surface diffusion of Ga and Al on GaAs, GaSb and AlSb. In the case of AlAs, and T_s range 400-600°C, A_0 has shown a monotonous dependence with substrate temperature, corresponded to the many level mode. Contrary, for the InSb a transition from step flow to 2D nucleation is observed at $T_s = 374°C$, due to a small Q value. Table I presents the experimental results obtained for $Q = E_{des} - E_D$. We found a limit for the growth of InSb, GaSb and AlSb at low temperature ($Ts < 345°C$), due to stebium surface accumulation. These phenomena produce the growth of amorphous layers. According to our experimental results we can estimate λ value as function of substrate temperature for GaAs, GaSb, AlSb and InSb as $\lambda[nm] = a \exp(Q/kT)$, where a and Q are given in Table I. Unfortunately, it is not possible to find in the current literature the suitable data for E_{des} in the case of GaSb, AlSb and InSb (100) surfaces. Due

to this fact it is not possible to determine the surface diffusion energy for such compounds.

Table 1: Data for the jump distance (a), the mode transition temperature (T_s) and the $Q = E_{des} - E_D$ values for GaAs, GaSb, AlSb and InSb compounds.

	GaAs	GaSb	AlSb	InSb
a(nm)	0.39975	0.43105	0.43385	0.45816
T_c(K)	879	787	864	647
Q(eV)	0.622	0.553	0.611	0.457

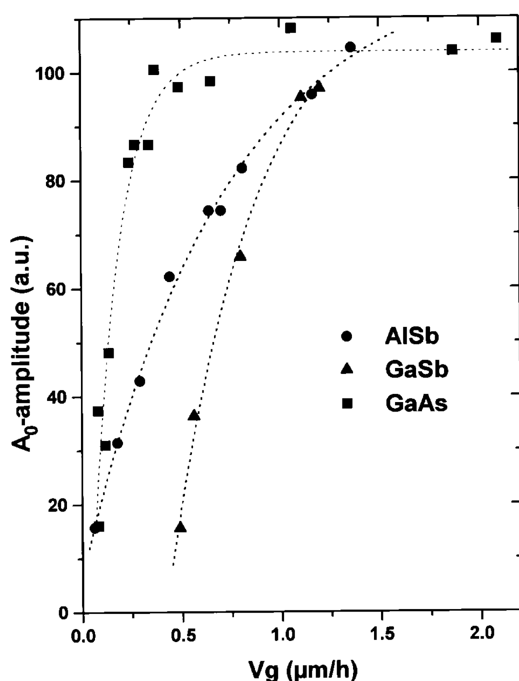

Figure 4: RHEED oscillation amplitude (Ao) as function of growth rate, for the GaAs, GaSb and AlSb compounds.

Fig. 4 presents $A_0 = f(V_G)$ for GaAs, GaSb and AlSb. For each compound, the substrate temperature were chosen to be in the middle of 2D nucleation growth mode. The figure shows that A_0 value depends exponentially on V_G. Such dependence evidences that reducing V_G, for a typical growth rate 0.1-1 μm/hour, growth mode phase diagram is shifted into the direction of step flow mode. In supperlattice growth, heterointerface roughness is usually determined by one of the high temperature stable compounds.

IV. Conclusions

In conclusion, we have compared the substrate temperature and growth rate effects on mode evolution of GaAs, AlAs, GaSb, AlSb and InSb. It was seen a strong effect of T_s and V_G on growth kinetic at usual MBE conditions. By means of changes in T_s and V_G, it was possible to observe five different growth modes. The temperature dependence of surface diffusion length for Al, Ga and In on GaAs, AlSb, GaSb and InSb were also presented. The experimental results presented in this paper indicate that it is possible to smooth the interfaces at heterojunctions, by lowering the growth rate.

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