Beating in the RHEED Intensity Oscillations during Surfactant Mediated GaAs Molecular Beam Epitaxy: Process Physics and Modeling

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In a recent work, beating in the reflection high energy electron diffraction (RHEED) intensity oscillations were observed during molecular beam epitaxial (MBE) growth of GaAs with Sn as a surfactant. The strength of beating is found to be dependent on the Sn submonolayer coverage with strong beating observed for 0.4 monolayer coverage. For a fixed temperature and flux ratio (Ga to As), the period of oscillation decreases with increasing Sn coverage. In this work, we have developed a rate equation model of growth to investigate this phenomenon. In our model, the GaAs covered by the Sn is assumed to grow at a faster rate compared to the GaAs not covered by Sn. Assuming that the electron beams reflected from the Sn covered surface and the rest of the surface are incoherent, the results of the dependence of the RHEED oscillations on Sn submonolayer coverages for various Sn coverages were obtained and compared with experimental data and the agreement is good.

Keywords: Surfactant, GaAs, MBE, RHEED, Modeling, Rate Equation.

1. INTRODUCTION

The presence of impurities on the surface is known to influence the flow of steps during the growth and modify the surface roughness, the composition of the epilayer and the degree of ordering [1-3]. In the growth of GaAs, Sn, which is an amphoteric dopant is known to ride the surface due to much larger atomic radius of Sn (1.42Å) compared to that of Ga (1.22Å) and As (1.18Å). Petrich et al [4] have experimentally studied the effect of submonolayer concentrations of Sn on the surface and observed the occurrence of beating in the RHEED intensity oscillations of the specular spot. They observed that the Sn submonolayer coverage required for strong beating decreases with temperature. They also observed that at 600°C, the period of oscillation decreases with Sn coverage.

In this article, we propose a kinetic rate equation model to explain the observed beating in the RHEED intensity oscillations of Ref.[4]. The details of the model are presented in in Section 2. Our results are compared with that of experiments of Ref.[4] in Section 3. Conclusions are presented in Section 4.
2. THE PROPOSED MODEL

2.1 Physics Of The Model

The presence of beats in the RHEED intensity oscillations (RO's) is assumed to be a result of two different intensity oscillations of slightly different frequencies adding up incoherently. A possible physical growth model which will yield two RO's of different frequencies is as follows. In a surfactant mediated growth with submonolayer coverage of the surfactant there are two distinct surfaces, one covered with the surfactant and the other not so. If the island size of the surfactant covered area is much larger than the coherent length of the incident electron beam, the resultant reflected intensity will be a sum of three components, intensities from surfactant covered surface, interface and surface not covered by the surfactant. The intensity components from the two surfaces will be incoherent if the Sn island size is larger than the coherent length of electron beam (about 100 Å for a typical 10 kV RHEED system). If the growth rates for the Sn-covered and non Sn-covered surfaces are different, then the RHEED intensities from these two areas will oscillate at different frequencies with respect to time. If these intensities interfere incoherently, beats will be observed in the resultant intensity. The interfacial component of intensity will be the coherent component which will contribute insignificantly to the total intensity for large island sizes because as the island size increases, the interfacial area (coherent component) to bulk area (incoherent component) ratio decreases.

2.2 The Rate Equation Model

The only elementary surface kinetic processes included in our model are: adsorption and migration. The time rates of change of concentration of Ga and As under the Sn-covered and non - Sn-covered surfaces can be described in terms of the rates of individual kinetic processes. Thus, the time rate of change of Sn-covered Ga concentration in the \(2n^{th}\) layer, \(C_{\text{Ga}}^{\text{Sn}}(2n)\), can be written as:

\[
\frac{dC_{\text{Ga}}^{\text{Sn}}(2n)}{dt} = S_{\text{Ga}}J_{\text{Ga}} [C_{\text{As}}^{\text{Sn}}(2n-1) - C_{\text{Ga}}^{\text{Sn}}(2n)] + \left[R_{\text{Ga,Sn},(2n-1)} J_{\text{Ga}} - C_{\text{Ga}}^{\text{Sn}}(2n) \right] \times \left[R_{\text{Ga,Sn},(2n-2)} J_{\text{Ga}} - C_{\text{Ga}}^{\text{Sn}}(2n-1) \right] \times \left[R_{\text{Ga,Sn},(2n)} J_{\text{Ga}} - C_{\text{Ga}}^{\text{Sn}}(2n+1) \right]
\]

The term \(A1\) represents the adsorption of Ga atoms on the Sn-covered surface. The flux and sticking probability of Ga are denoted as \(J_{\text{Ga}}\) and \(S_{\text{Ga}}\), respectively. The term \(A2\) describes the rate of addition of Ga from adjacent Ga layers by migration through Arrhenius type rate equation with activation energy, \(E_{\text{Ga}}\), and frequency factor, \(R_{\text{Ga,Sn}}\). The activation energy for migration of Ga atoms from the \((2n+2)^{th}\) layer is given by:

\[
E_{\text{Ga}}^{\text{Sn}}(2n+2) = E_{\text{Ga}}^{\text{iso}}(2n+2) + zE_{\text{Ga}}^{\text{Sn}}(2n+2)
\]

and a similar equation is assumed for the \((2n-2)^{th}\) layer. In Equation 2, the \(E_{\text{Ga}}^{\text{iso}}(2n+2)\) and \(E_{\text{Ga}}^{\text{Sn}}(2n)\) define the activation energy of migration of an isolated Ga on the surface and interaction energy for second nearest neighbor Ga – Ga pairs, respectively. \(z\) is the inplane coordination number for the (100) plane which is 4. The term A3 in the equation represents the rate of deletion of atoms from \(2n^{th}\) layer due to migration of Ga atoms to adjacent layers. Similar time evolution equations can be written for Ga and As on non – Sn-covered and As on Sn-covered surface. Additionally, the total concentration of Ga in the \(2n^{th}\) layer satisfies the following condition:

\[
C_{\text{Ga}}^{\text{Sn}}(2n) + C_{\text{Ga}}^{\text{non-Sn}}(2n) \leq 1.0
\]

where \(C_{\text{Ga}}^{\text{non-Sn}}(2n)\) is the concentration of Ga in on the non – Sn-covered area of the \(2n^{th}\) layer. The frequency factor, \(R_{\text{Ga}}\) for Ga and As were assumed to be \(10^{13}/\text{sec}\). \(E_{\text{iso}}\) for Ga and As for all sur-
faces were assumed to be 1.3 eV. \( E_{GaGa} \) and \( E_{AsAs} \) for both the surfaces is assumed equal to 0.17 eV. The sticking probabilities of \( Ga \) for the two surfaces were assumed to be different and used as fitting parameters of the model.

3. RESULTS AND DISCUSSION

The 2\( n \) coupled first order non-linear differential equations for 2\( n \) layers were solved numerically with a time step of \( 10^{-6} \) seconds. Instantaneous RHEED intensities of specular spot were computed using the kinematical theory of electron diffraction with a 10 kV electron beam at 1.5° grazing incidence. The fitting parameters (sticking probability of \( Ga \) on Sn-covered and non-Sn-covered surfaces) were obtained by matching the experimental and theoretical RHEED intensity oscillations (RO’s).

Simulations were performed for various submonolayer coverages of Sn for a temperature of 600°C at a flux rate of 1 monolayer/sec. The RHEED intensity versus time plot is shown in Figure 1. The qualitative agreement between the RO’s reported in Ref.[4] with Figure 1 is fairly good. The RO’s are dependent on the Sn coverage with low and high coverages yielding typical RO’s observed in MBE growths and medium coverages resulting in beating behavior as shown in Figure 1. The beating behavior is a result of RHEED intensities from the Sn-covered and non-Sn-covered surfaces adding up incoherently. For low and high Sn coverages, the RHEED intensity in the specular beam contains predominantly a single frequency component from either the non-Sn-covered or the Sn-covered GaAs with both coherent and incoherent components. Since, the frequencies of the coherent and incoherent components are the same, there is no beating observed. However, for medium coverage of Sn, the RHEED intensity contains three components: intensities from within Sn-covered and non-Sn-covered surfaces respectively and electron beam reflected from the interface between these surfaces which is coherent. The reflected electron beams from the Sn-covered surfaces and from the non-Sn-covered GaAs will not be coherent with each other if the island sizes of Sn are larger than the coherent length of the electron beam (which is typically of the order of 100Å). The third coherent component arising from the interface will be insignificant for large island sizes as the interfacial area decreases in comparison to the Sn-covered or non-Sn-covered areas. Thus, for medium Sn coverages, the addition of intensities from incoherent electron beams from the Sn-covered and non-Sn-covered surfaces results. Since these two areas grow at different growth rates, the frequencies of oscillation of these two components will be slightly different which will yield beats in RO’s. Additionally, the amplitude of RO’s decreased slightly with Sn submonolayer coverage as shown in Figure 2 unlike experiments in which the amplitude increased slightly which may be a result of assumptions in the RHEED computation such as exclusion of multiple reflections, atomic scattering factor differences and effect of surface reconstruction on the RHEED intensity.

A plot of time period of the RO’s versus Sn submonolayer coverages at 600°C is shown in Figure 2. As the Sn submonolayer coverage increases, the period of RO decreases which is in good agreement with the experimental data of Ref.[4]. Noting that the period of RO’s is inversely related to the growth rate and that the Sn-covered are a grows at a faster rate compared to non-Sn-covered area, it is expected that for large Sn coverages, the average growth rate will be...
larger and hence a smaller period of RO's and vice versa.

4. CONCLUSION

In this work, a rate equation model is proposed and utilized to investigate surfactant mediated MBE growth of GaAs. The model reproduces semi-quantitatively most of the experimental observations of RHEED oscillations and their dependence on the surface conditions such as the Sn coverage. The key assumption of the model is that the growth rates of GaAs on Sn-covered and non-Sn-covered surfaces can be different and that electron beam intensities reflected from these two surfaces, if separated by more than the coherent length of the electron beam, will interfere incoherently. It was found that the GaAs on Sn-covered surface grows at faster rate compared to GaAs on non-Sn-covered GaAs.

References


Biographies

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