

RHEED oscillations of arsenic-controlled growth conditions to optimize MBE growth of III/V heterostructures

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RHEED intensity oscillations of arsenic-controlled growth conditions have been used to measure the sticking coefficient of As_2 on InGaAs, GaAs, and AlGaAs surfaces. The sticking coefficient shows two types of discrete behavior: (1) a substrate temperature independent region where the sticking coefficients are maximized and of the same value for the three surfaces; (2) a substrate temperature dependent region where the sticking coefficient decreases as the substrate temperature is increased. The onset of the temperature dependent regions differ for the three surfaces. The onset occurs at the highest temperature for AlGaAs, at a lower temperature for GaAs, and at a still lower temperature for InGaAs. The arsenic sticking coefficient not only changes with substrate temperature but also with the group III species and the group III/III combination. Measurements of how the arsenic sticking coefficient changes at a given substrate temperature for different III–III/V compounds provides an understanding for improving growth conditions of heteroepitaxial films and their interfaces.

1. Introduction

The influence of growth conditions in molecular beam epitaxy (MBE) on the properties of heterostructure devices is of great importance. The optimum performance of device structures such as: surface emitting lasers (SELs), pseudomorphic high electron mobility transistors (PHEMTs), and multiple quantum well spatial light modulators (MQWSLMs) depend on obtaining atomically flat interfaces with excellent electrical and/or optical characteristics in the individual layers. In order to realize the optimum growth conditions, it would be desirable to routinely measure a parameter during growth set-up that is sensitive to the fluxes, the effects of substrate temperatures and the type of layers that create an interface. A parameter that fulfills these conditions can be found in reflection high energy electron diffraction (RHEED) oscillations of arsenic-controlled growth conditions [1]. Arsenic-controlled growth conditions occur when excess group III atoms (not incorporated in the crystal lattice) are induced on GaAs surfaces in the

presence of an arsenic flux. RHEED intensity oscillations occur as the excess group III elements are incorporated in the substrate lattice. Excess group III species can be produced on substrates in at least two ways. While the arsenic shutter is open, one may open the group III shutters with a combined flux greater than the incident arsenic flux can directly incorporate into the GaAs lattice. This is shown for excess gallium in ref. [1]. Another method involves closing the arsenic shutter and opening the group III shutters with normal growth fluxes, allowing the accumulation of several monolayers of group III species, and then opening the arsenic shutter without terminating the group III fluxes. The reason for not closing the group III shutters is to maintain the background arsenic flux contribution low during the arsenic-controlled RHEED oscillation measurements. In ref. [1], the increase flux from the background arsenic after closing the gallium shutter is shown as a change in the arsenic controlled growth rate. Furthermore, the period of the arsenic controlled RHEED intensity oscillations are shown to depend on the ar-

senic flux and the substrate temperature. Hence, the growth rates with excess gallium atoms are referred to as the arsenic-controlled incorporation rates or group-V-controlled conditions.

Since the first reported observation [2] of arsenic-controlled RHEED intensity oscillations, the growth conditions producing these oscillations have been shown to be useful for: (A) growing InAs/GaAs superlattices with periods larger than the critical thickness for misfit dislocations [3], (B) setting reproducible growth conditions independent of ion gauge measurements [4,5], and (C) measuring the sticking coefficient of As_4 and As_2 , and their fluxes during MBE of GaAs on GaAs [1]. The arsenic-controlled incorporation rates can be used to measure the incident arsenic flux. Once the arsenic flux is known, the sticking coefficient of arsenic can be extracted for any growth condition, as shown in ref. [1] and later in this work. The sticking coefficient of arsenic is defined as the fraction of the incident arsenic flux that is incorporated in the substrate lattice. The above definition for the arsenic sticking coefficient is the same as in the mass spectrometry studies in ref. [6].

MBE growth of III/V compounds can be accomplished in at least two ways: (1) conventional MBE growth performed with excess arsenic at relatively high growth temperature; (2) growth performed with excess group III atoms at relatively low substrate temperatures, as was done in ref. [3]. In symmetry with group-V-controlled conditions, growth rates with an excess arsenic flux will be referred to as the group-III-controlled growth rates or group-III-controlled conditions. This second growth category has been studied and refined, and is now known as migration enhanced epitaxy (MEE) [7]. The refinement consists mainly of depositing alternating layers, each one monolayer thick. First one monolayer of group III atoms is deposited in the absence of a direct arsenic flux. The excess gallium atoms undergo enhanced migration because they are not incorporated in the substrate lattice. This is followed by exposing the surface to an arsenic flux just long enough to incorporate all free group III atoms into the substrate lattice. Incorporation rates of the group III and group V atoms are

controlled by growth temperature and the arrival rate of arsenic molecules. Growth is carried out by iterating this procedure to obtain the desired film thickness.

Implementation of the above two growth processes can easily be characterized by measurements of arsenic controlled incorporation rates as described in refs. [1,5,8] for GaAs. Arsenic controlled incorporation rates in units of monolayers/s, $\mu\text{m/h}$, or $(\text{Ga or As})/\text{cm}^2 \cdot \text{s}$ can be obtained from the period of the arsenic-controlled RHEED intensity oscillations. Characterization and better understanding of growth conditions come about in that arsenic-controlled incorporation rates determine the time required to incorporate excess group III atoms for MEE as a function of the substrate temperature and the arsenic flux. For group-III-controlled growth conditions (excess arsenic), improved characterization and understanding can be realized by using the temperature dependent ratio of arsenic-controlled to group-III-controlled growth rates. The temperature dependent incorporate rate ratios paired with a group-III-controlled growth rate can be linked to optimum characteristics of MBE layers. Growth conditions characterized in this manner can be accurately reproduced in any MBE system. In the present work, As_2 -controlled incorporation rates are shown to depend also on the group III–III combination. As_2 sticking coefficient measurements under arsenic controlled conditions are extended to the InGaAs and AlGaAs systems. These results highlight important considerations for optimizing growth conditions of heterostructures.

2. Experimental procedure

Measurements were performed in a Varian Modular Gen II MBE system using the Varian RHEED intensity detection system, and a solid arsenic cracker operated in the dimer mode. (100) GaAs wafers were used throughout in these measurements. Arsenic-controlled incorporation rates were measured by closing the arsenic shutter and simultaneously opening the group III shutters. After allowing accumulation of 6 to 10

monolayers of group III atoms, the group V shutter was opened. While the group III shutters were held open, arsenic-controlled RHEED intensity oscillations of the specular spot were measured. After incorporation of the excess group III atoms on the surface, the growth rates change to that of group-III-controlled conditions. The group III shutters are then closed. The measurements were all performed from low to high substrate temperatures, and on the two fold azimuth of the $\text{As}(2 \times 4)$ RHEED pattern. The electron beam energy was 8 kV and the angle of incidence was set to satisfy $S_{\perp}d = 2\pi$ diffraction conditions. S_{\perp} is the electron momentum normal to the surface, and d is the Ga to Ga (or As to As) adjacent atomic plane distance.

3. Results and discussion

Fig. 1 shows RHEED intensity oscillations for gallium-controlled MBE growth at two different substrate temperatures. When the gallium shutter is opened, the RHEED intensity increases for the low substrate temperature conditions, while for

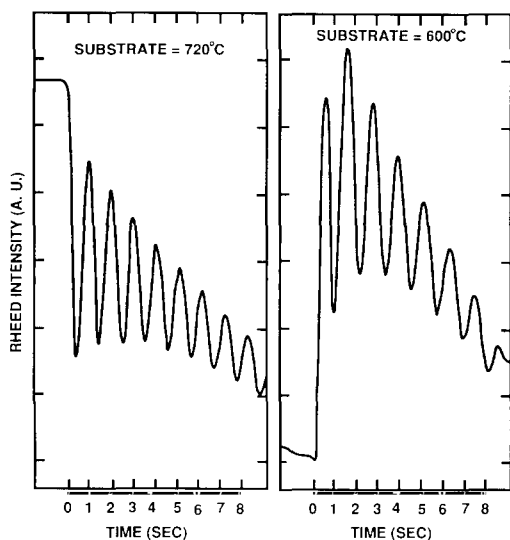


Fig. 1. Gallium-controlled RHEED intensity oscillations. Substrate temperatures given in the figure are thermocouple-indicated values. A tentative temperature calibration of: $\text{Pyro.} (^{\circ}\text{C}) = 0.62 \times \text{TC} (^{\circ}\text{C}) + 152 ^{\circ}\text{C}$, can be used for approximate wafer surface temperatures.

the high temperature case the intensity decreases. For $t < t_0$, the time at which the gallium shutter is opened, the static RHEED intensity is lower at low substrate temperatures, and intensities during oscillations increases above the $t < t_0$ value. As the substrate thermocouple temperature is reduced from 720 to 600 $^{\circ}\text{C}$, the static RHEED intensity decreases and the pattern changes from an $\text{As}(2 \times 4)$ to an $\text{As}(2 \times 2)$ reconstruction, consistent with results in refs. [9,10]. The surface reconstruction at low temperatures appears to be different from that reported in the literature; however, As_2 was used in these experiments. The static intensity drop is probably related to an increase in the surface arsenic composition as the temperature is reduced. The arsenic surface composition should increase as the substrate temperature is lowered because the arsenic sticking coefficient increases. Refs. [1] and [5] indicate that at relatively low temperatures during arsenic-controlled incorporation rate measurements, RHEED intensity oscillations are initially controlled by the gallium arrival rates before gallium accumulation initiates, and then subsequent oscillations are controlled by the arsenic arrival rate. These initial gallium-controlled intensity oscillations are indicative of a relatively high arsenic surface content, or an enhanced (transient) sticking coefficient of arsenic at the very beginning of growth at the low temperatures. That surface compositions change with temperature is well known from studies of surface reconstructions [11,12].

Fig. 2 shows typical RHEED intensity oscillations for arsenic controlled MBE growth of GaAs at various substrate temperatures. The curves have been displaced vertically to reduce confusion. However, the static RHEED intensity for curve 2 is higher than that of curve 1, but lower than those of curves 3 and 4 which have the same intensity. The initial increase in intensities at low substrate temperatures, curves 1 and 2, following the closing of arsenic and the opening of gallium shutters might be explained as follows: Arriving gallium atoms lower the surface arsenic composition by incorporating the surface arsenic into the GaAs lattice. At the peak of the initial intensity increase, the surface compositions reach condi-

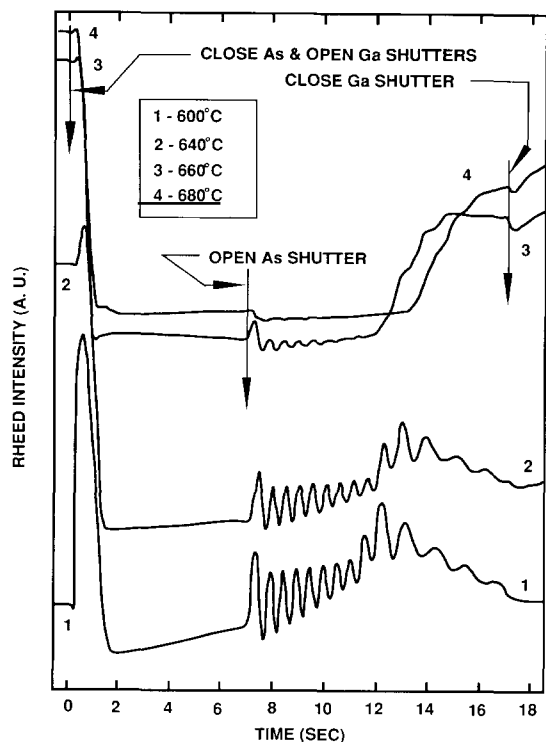


Fig. 2. Arsenic-controlled RHEED intensity oscillations on GaAs. Gallium and arsenic fluxes were held constant. See fig. 1 caption for substrate temperature calibration.

tions similar to that present at high temperatures before the shutters are operated, since similar RHEED intensities are observed. When the surface arsenic composition has been incorporated, gallium atoms begin to accumulate producing a state of roughness and disorder which reduces the RHEED intensity. During the initial intensity increase and decrease, the RHEED pattern for curve 1, fig. 2, is observed to change from $As(2 \times 2)$ to (1×3) to $Ga(2 \times 2)$. After sufficient gallium atoms have accumulated, the arsenic shutter is opened while the gallium shutter is held open to reduce the effects of the background arsenic flux as described earlier. When the arsenic shutter is opened, RHEED intensity oscillations occur and incorporation rate values are determined by the arsenic flux and the substrate temperature. If there is more than one group III species incident and accumulating on the surface, then the arsenic-controlled incorporation rates are also determined by the group III combination, via a

characteristic change in the arsenic sticking coefficient to be discussed below. In fig. 2, after excess gallium is incorporated, oscillations continue but are then gallium-controlled and are observed as a change in incorporation rates (period of the oscillations). For curve 2, the initial increase in intensity is relatively small compared to curve 1 because the static RHEED intensity is higher at higher temperatures as explained in reference to fig. 1. During these measurements, the static RHEED intensities were found to increase with increasing temperature to a saturation value as in ref. [10] for As_4 . More curves showing this trend are not presented in the figure to also reduce confusion.

In fig. 3, As_2 -controlled incorporation rates and arsenic sticking coefficients are plotted versus inverse substrate thermocouple temperatures for InGaAs, GaAs, and AlGaAs. From modulated beam mass spectroscopy (MBMS) results [13], it is suggested that the maximum sticking coefficient of As_2 approaches a value of unity. However, data showing arsenic sticking coefficients higher than 0.75 are not presented. From RHEED results in ref. [1], it appears that the maximum As_2 sticking coefficient saturates at 0.75. Therefore, in fig. 3, a value of 0.75 has been taken as the maximum sticking coefficient corresponding to the maximum As_2 -controlled incor-

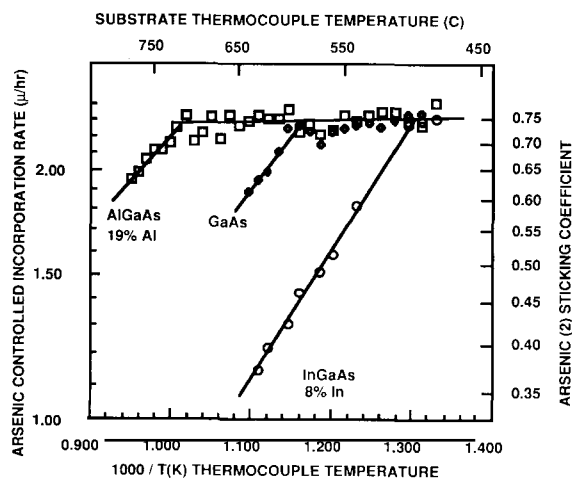


Fig. 3. Arsenic-controlled incorporation rates and sticking coefficients for InGaAs, GaAs, and AlGaAs as a function of substrate temperature.

poration rates. The arsenic-controlled arsenic sticking coefficients can be calculated from:

$$\epsilon_{\text{As}}(T) = \epsilon_{\text{As}}^{\text{LT}} \text{IR}_{\text{As}}(T) / \text{IR}_{\text{As}}^{\text{LT}}. \quad (1)$$

$\text{IR}_{\text{As}}^{\text{LT}}$ is the As_2 -controlled incorporation rate in the low substrate temperature region, where the As_2 sticking coefficient has the maximum value and is independent of the temperature. $\text{IR}_{\text{As}}(T)$ is the As_2 -controlled incorporation rate in the substrate temperature dependent region. $\epsilon_{\text{As}}^{\text{LT}}$ is the maximum sticking coefficient of As_2 at low temperatures, 0.75.

The group III and the arsenic fluxes in the present study are for approximately $1 \mu\text{m/h}$ group-III-controlled (excess group V) growth rates. This is in contrast to the MBMS studies, where arsenic fluxes of 2 to 3 order of magnitude lower were typical. In the MBMS studies, the sticking coefficient of As_2 on GaAs surfaces was found to increase as the substrate temperature increases. In fig. 3, the As_2 sticking coefficient decreases as the substrate temperature increases. In the MBMS studies, the sticking coefficients are obtained from desorption rates as one minus the fraction of the incident arsenic flux that is not incorporated, while in the RHEED studies, the sticking coefficient of arsenic as obtained from in eq. (1), is the fraction of the incident arsenic flux which is incorporated. The definitions from the two techniques are identical. $\text{IR}_{\text{As}}^{\text{LT}} / \epsilon_{\text{As}}^{\text{LT}}$ gives the incident arsenic flux when the group-V-controlled incorporation rates are calculated as $\text{As}/\text{cm}^2 \cdot \text{s}$. The different results with substrate temperature for the two techniques arise from the presence of different surface conditions. In the MBMS studies, gallium excess or accumulation (group-V-controlled conditions) to the extent induced in the RHEED studies apparently did not occur at the high temperatures. As implied in the MBMS study, the increase of the As_2 sticking coefficient with increasing temperature reflects a decrease in the surface arsenic composition through loss of As_2 from decomposition of GaAs. However, desorption of sufficient arsenic to produce excess gallium was not induced. The As_2 -controlled arsenic sticking coefficients are easy to measure with the RHEED technique, and qualitatively

exhibit the expected temperature dependence of surface arsenic compositions for group III (excess arsenic) growth conditions.

In fig. 3, the As_2 sticking coefficient has substrate temperature independent regions in which the maximum value is the same for the three materials, and each material has temperature dependent arsenic sticking coefficients in different temperature ranges. The onset for the AlGaAs temperature dependent region occurs at a higher temperature than for GaAs, which occurs at a higher temperature than for InGaAs. This is qualitatively consistent with the AlAs bond strength being higher than that of GaAs, which is higher still than that of InAs.

Fig. 3 reflects qualitatively what is well known on how growth conditions should be adjusted at the various heterostructure interfaces in order to optimize the MBE film growth during group-III-controlled conditions (excess arsenic). For group-V-controlled growth of GaAs on GaAs at temperatures lower than 590°C , the As_2 -controlled arsenic sticking coefficient has reached the maximum value. Therefore, for the above temperature region, we should expect the group V to group III flux ratio for optimum group-III-controlled growth to be the smallest possible. The ratio should be small compared to higher temperatures, because the surface arsenic composition is higher at lower temperatures. MBE growth with an excess arsenic flux is the norm and yields stoichiometric GaAs. However, this stoichiometry is only approximate [14]. In ref. [14], small deviations in the stoichiometry of MBE-grown GaAs films have been measured and are shown to depend on the substrate temperature and the arsenic to gallium flux ratios.

At a substrate temperature of 550°C , fig. 3 shows that for group-V-controlled growth of InGaAs, the As_2 -controlled arsenic sticking coefficient is lower than that of GaAs. Therefore, for MBE of group-III-controlled growth of the inverted interface, InGaAs on GaAs, the group V to group III flux ratio should be increased compared to the growth of GaAs on InGaAs. The implication is that during group-III-controlled growth conditions, the surface arsenic composition on InGaAs is lower than on GaAs. This is

supported in the case of As_4 in ref. [15]. There it is shown that the sticking coefficient of As_4 on InAs also increases with increasing substrate temperature, as is typical of the MBMS studies and explained earlier. At a given temperature, the sticking coefficient of As_4 on InAs is higher than that on GaAs, but less than 0.5. Hence, InGaAs surfaces would be expected to have lower surface arsenic compositions than GaAs surfaces at the same temperature.

At a substrate temperature of 650 °C, fig. 3 shows that for group-V-controlled growth of AlGaAs, the As_2 -controlled arsenic sticking coefficient is at the maximum value. Consequently, for group-III-controlled growth of the normal interface, AlGaAs on GaAs, the group V to group III flux ratio should be decreased to the lowest value possible compared to the growth of the inverted interface, GaAs on AlGaAs. The same arguments about the surface arsenic compositions as discussed earlier are also applicable in this case.

Typically we assume that MBE grown films are stoichiometric, except for films grown at very low temperatures [16,17]. For films grown at normal temperatures, results in ref. [14] suggest that variations in the stoichiometry may be very common, though very small, $C_{\text{As}} - C_{\text{Ga}} \approx 10^{-4}$. In view of this result and the present results suggesting different surface arsenic compositions for the different MBE layers grown at a fixed temperature, we may speculate that the initial surface arsenic composition likely plays an important role in the stoichiometry near heterostructure interfaces. Particularly, for the inverted high electron mobility transistor (I-HEMT), where GaAs is grown on AlGaAs without growth interruption. Since we expect the surface arsenic composition to be higher on AlGaAs than on GaAs surfaces, then for the initial stages of GaAs growth on the AlGaAs surface, gallium atoms arrive on a surface with too much arsenic. This could induce a relatively large deviation in the stoichiometry of the GaAs layer near the interface. Therefore, not only the inherent surface roughness present on the AlGaAs surfaces [18], but also a relatively high stoichiometric deviation near the interface in the GaAs layer, if present, affects the electrical characteristics of the two-dimensional (2D) elec-

tron gas. However, the surface roughness at the inverted interface may be the more important factor. Growth interruption after a monolayer of GaAs on the AlGaAs surface, at or near the 2D channel, would not only improve the surface roughness as reported in refs. [19,20], but also allow time for the surface composition to change and produce surface arsenic compositions that improve the GaAs stoichiometry near the interface after growth is resumed.

4. Conclusions

RHEED intensity oscillations of arsenic-controlled growth conditions were used to measure the sticking coefficient of As_2 for different III-III/V compounds. The changes observed in the sticking coefficient of As_2 , when they are qualitatively associated with surface arsenic compositions, provide a better understanding for improving growth conditions of heterostructures and their interfaces.

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