Reflection high-energy electron diffraction studies of the growth of InAs/$Ga_{1-x}In_xSb$ strained-layer superlattices

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We have used reflection high-energy electron diffraction to study the surface periodicity of the growth front of InAs/GaInSb strained-layer superlattices (SLSs). We found that the apparent surface lattice spacing reproducibly changed during layers which subsequent x-ray measurements indicated were coherently strained. Abrupt changes in the measured streak spacings were found to be correlated to changes in the growth flux. The profile of the dynamic streak spacing was found to be reproducible when comparing consecutive periods of a SLSs or different SLSs employing the same shuttering scheme at the InAs/GaInSb interface. Finally, when the interface shuttering scheme was changed, it was found that the dynamic streak separation profile also changed. Large changes in the shuttering scheme led to dramatic differences in the streak separation profile, and small changes in the shuttering scheme led to minor changes in the profile. In both cases, the differences in the surface periodicity profile occured during the parts of the growth where the incident fluxes differed.

I. INTRODUCTION

Recent developments in the growth of arsenide/antimonide (As/Sb) heterostructures have led to a wide range of devices including a variety of novel InAs/AlGaSb tunnel structures,[1-4] extremely high frequency oscillators,[5] and far infrared (IR) detectors grown in both the InAs/$Ga_{1-x}In_xSb$,[6] and the InAs/InAsSb[7] material systems. These efforts have resulted in a large number of promising devices but in order for these structures to reach their full potential, a basic materials issue must be understood: How to control the structural and chemical properties of the As/Sb interface?[8] Because the vapor pressures of the group V's (As and Sb) are substantially larger than those of the relevant group III's (In, Ga, and Al) their sticking coefficients are very different at typical substrate temperatures. As a result, it is necessary to evaporate 4–10 group V atoms for each group III atom in order to grow a stoichiometric crystal. In structures involving a common anion, such as AlAs/GaAs heterostructures, this is not an issue. However, in As/Sb heterostructures the question of what to do with the excess anion becomes very important. There are two distinct problems. The first is controlling cross contamination of the group V's: As incorporation in the GaSb layers when growing an InAs/GaSb superlattice for instance. The second is the composition of the interface. For example, when switching between InAs and GaSb layers, one could imagine preparing an interface that consisted of InAs/In/Sb/GaSb or one with InAs/As/Ga/GaSb or some intermediate composition. There is no *a priori* reason to believe that these different kinds of interfaces would lead to similar electrical and optical properties. In addition, some of these structures have rather large lattice mismatches between their constituent materials, further complicating growth.

In order to study the details of the As/Sb interface, an *in situ*, time resolved measurement of the crystal during growth is needed. In this work, we report measurements of the dynamics of the growth surface of InAs/$Ga_{1-x}In_xSb$ strain-layer superlattices (SLSs) using electron diffraction. These SLSs have been shown to be promising candidates for optical detectors in the 8–12 μm wavelengths range.[6] We find that the streak spacing of the reflection high-energy electron diffraction (RHEED) pattern changes during growth. The dynamics of the streak spacing was found to be reproducible between growths using the same shuttering sequence at the GaInSb/InAs interface. The streak separation dynamics of growths employing different interface shuttering schemes was found to vary markedly. In Sec. II, we detail the crystal growth and the experimental apparatus used to obtain the data. Section III contains dynamic measurements of the RHEED streak spacing showing both the reproducibility of the measurement and the effect of changing the shuttering scheme at the As/Sb interface. Section IV is a discussion of the results which are summarized in Sec. V.

II. EXPERIMENTAL

In order to extract information from the RHEED pattern, we have developed a technique for digital data acquisition. First the diffraction pattern is videotaped using a charge coupled device (CCD) camera and an S-VHS video cassette recorder. We focus on the specular and first order streaks in order to most accurately measure the streak separation, but any portion of the RHEED pattern could be videotaped (see Fig. 1). The tape is then played back and digitized into a 640×480 array of single-byte data with a RasterOps framegrabber installed in SPARC 2 workstation. To increase the data acquisition rate, often only a

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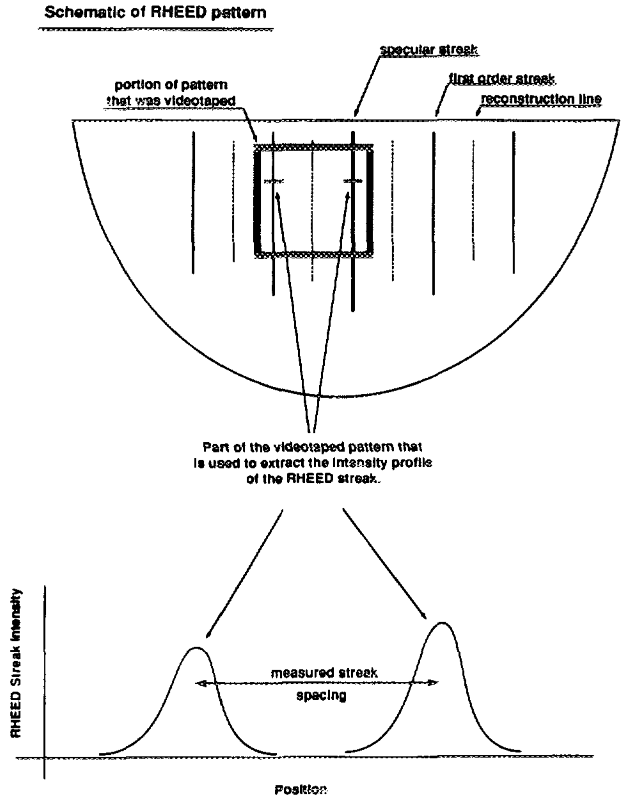

FIG. 1. Schematic representation of the data acquisition method.

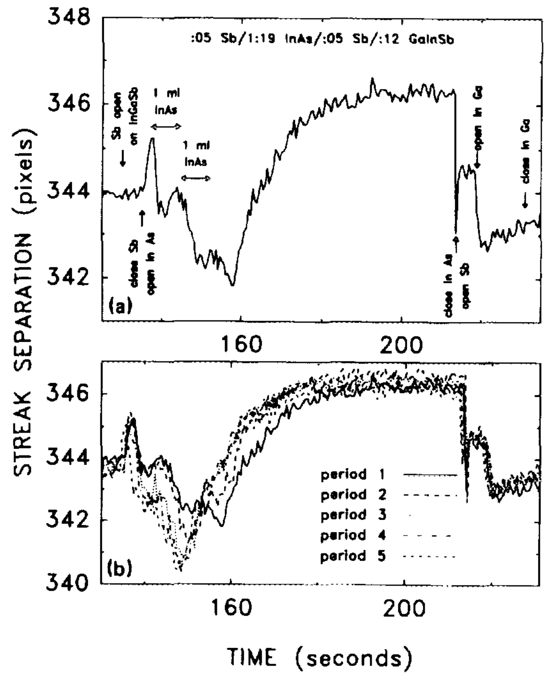

FIG. 2. Separation between the specular streak and first order diffraction streak in the RHEED pattern of an InAs/$Ga_{1-x}In_xSb$ SLS. (a) Notes the times when the shutters are either opened or closed, and (b) is an overlay of four consecutively grown periods of the SLS showing the reproducibility of the effect.

portion of the videotaped pattern is digitized. The system can digitize between 2 and 10 frames/s depending on the size of the portion of the pattern that is being examined. After digitization the data can be either integrated to obtain intensity variations or fit with a Lorentzian plus a linear function to determine the streak positions. Recording large portions of the diffraction pattern on videotape provides a great deal of flexibility. One of the goals of this project is to identify which portions of the RHEED pattern are relevant to understanding the nature of the As/Sb interface. The ability to examine different parts of the diffraction pattern of the same growth is crucial for realization of this goal.

All of the samples studied here were grown in a Perkin–Elmer 430 molecular-beam epitaxy (MBE) system equipped with cracked Sb and As sources. The SLSs were grown on semi-insulating GaAs substrates and particular care was taken to use substrates from different boules in order to decrease the likelihood that the peculiarites of a particular lot of wafers would affect the data. The SLSs were grown on a thick stress relaxed GaSb buffer at a substrate temperature of ~385 °C. The growth rate of the InAs was 0.12 monolayer (ML)/s and that of the GaInSb was 0.64 ML/s. The In mole fraction in the antimonide layers was 0.24. The SLSs are coherently strained to the thick GaSb buffer with the InAs layers under 0.6% tensile stress, and the GaInSb layers under 1.5% compressive stress. A more detailed description of the growth has been previously reported.[9]

III. RESULTS FROM InAs/$Ga_{1-x}In_xSb$ SUPERLATTICES

Figure 2 shows the measured spacing between the specular streak and the first order streak during the growth of an InAs/GaInSb SLS. Each period consists of 28 Å of InAs and 23 Å of $Ga_{0.76}In_{0.24}Sb$. At the end of each layer, before switching materials, the growth surface is soaked in Sb. Because of this soak the internal interfaces of the SLS are termed to be "Sb-like." The data were taken on the [110] azimuth at a rate of 2.5 data points per second. Figure 2(a) shows the dynamic streak spacing for a single period of the SLS. The times at which the oven shutters are opened and closed are marked with arrows. Figure 2(b) is an overlay of four consecutive periods of the SLS where the profile labeled "period 1" is the one shown in Fig. 2(a). There are no vertical offsets in the data shown, and the timing of the shutter actuations were determined from the clock on the VCR and period of the SLS. Three things stand out in the data. First, the apparent lattice spacing (ALS), which is inversely proportional to the streak separation, of the surface of the crystal changes during growth. Second, abrupt changes in the ALS are correlated with either opening or closing a shutter. Third, Fig. 2(b) shows that different periods of the same SLS have very similar dynamic streak spacings. Subsequent growths showed that when the same interface shuttering scheme was used, the profiles of the ALS matched those shown in Fig. 2.

To ensure that the features in the ALS that are correlated to the shutter openings are due to changes in the

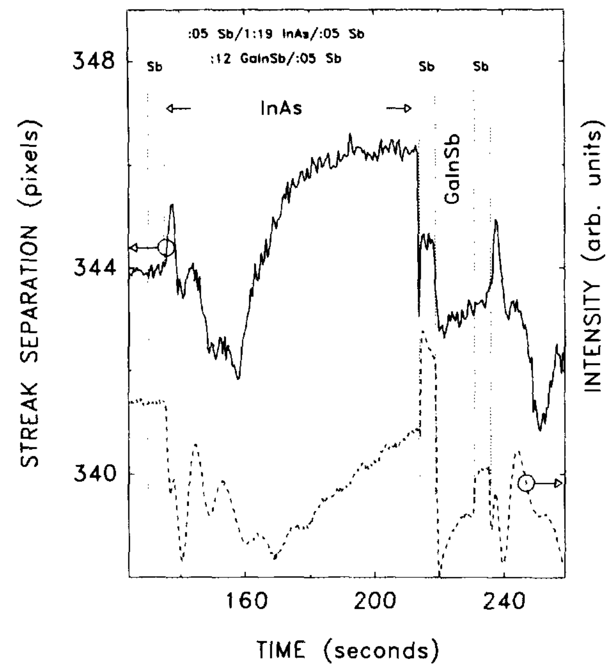

FIG. 3. Comparison of the measured streak separation with the intensity profile of the specular streak. The upper solid line is the streak separation and the lower dashed line is the RHEED intensity. The extent of the Sb soaks are marked by the pairs of dotted lines.

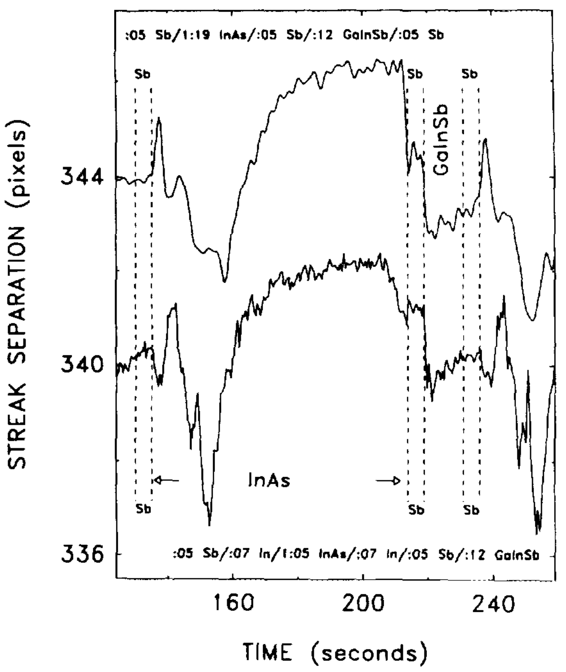

FIG. 4. Comparison of the measured streak separation of two superlattices with the same layer thickness and compositions both of which were grown under the same conditions. The extent of the Sb soaks are marked by the pairs of dotted lines.

incident fluxes and not an electrical or mechanical artifact of the growth chamber, we measured the dynamic position of the specular and first order streaks while opening and closing the shutters of ovens that were at their idle temperature of 300 °C. The measurements were done on GaAs surfaces along both the [110] and [1$\bar{1}$0] azimuths while growing GaAs and during As-flux soaks. To within our experimental resolution of about 0.5 pixels out of a streak spacing 420 pixels, neither the specular streak nor the first order streak moved. This indicates that the measured changes in the streak spacing when the shutters are actuated are due to changes in the incident flux and hence changes in the growth surface. It should be noted that the size of the measured shifts is small. In Fig. 2(a), the streak separation increases by about one pixel after the Sb shutter is closed, and the In and As shutters are opened. This translates into ~0.3% change in the ALS.

We compare the measured streak spacing of the SLS, shown in Fig. 3, with the intensity variation of the specular streak for slightly more than one period of the structure. The solid upper line is the data shown in Fig. 2(a), and the lower, dashed line is the intensity variation of the specular streak. The pairs of dotted vertical lines denote the extent of the Sb soaks. The two curves have similar shapes, and the plot of RHEED intensity has sharp features when the growth fluxes are changed just as in the case of streak separation profile. These intensity variations are reproducible when comparing different periods of the SLS. The features of importance in this graph are the oscillations in the intensity at the start of the InAs layer (second vertical line). These oscillations have the same period as the oscillations in the streak separation with a period equal to the monolayer deposition time. These types of intensity oscillations are routinely used by many investigators to determine growth rates and are believed to be caused by periodic changes in the roughness of the growth front.[10,11] The strong correlation between the ALS profile and the RHEED intensity profile indicates that the streak separation modulation is sensitive to changes in the texture of the growth front.

We have examined the RHEED characteristics of roughly a dozen different interface shuttering schemes. In every case, abrupt changes in the ALS were correlated with changing the growth flux, and the changes in the ALS were reproducible between different periods of the SLS and different growths using the same or similar interface schemes.

Figure 4 compares the measured streak spacing of two Sb-like, SLSs grown with similar, but not identical interface shuttering schemes. The two structures have the same compositions and layer thicknesses and were grown under the same conditions. The data are vertically offset by four pixels numerically. The extent of each Sb soak is marked by a pair of vertical dashed lines. The upper curve is the same as shown in Fig. 2(a). The difference between the shuttering schemes is that in the lower curve a single monolayer of In is deposited at the beginning and end of the InAs layer without an accompanying group V flux. The two curves are qualitatively similar; however, the two profiles are not identical. At the start and end of the InAs layer (the second and third vertical lines, respectively) the two curves are markedly different. These differences occur at precisely the times when the fluxes incident on the growth surface are different. This observation indicates that the measurement is sensitive to subtle changes in the

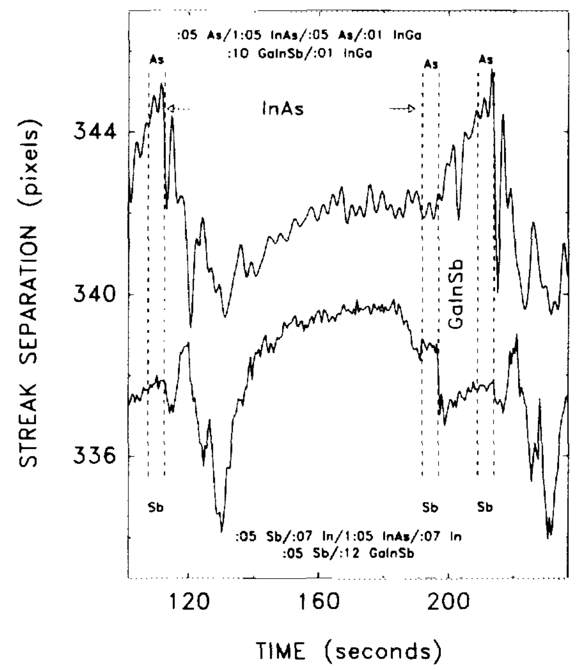

FIG. 5. Comparison of the measured streak separation of two superlattices with the same layer thickness and compositions both of which were grown under the same conditions. The extent of the group V soaks are marked by the pairs of dotted lines, Sb for the lower curve and As for the upper curve.

growth surface. This similarity in ALS profiles when comparing similar interface schemes was observed in all the SLSs that were studied.

Figure 5 compares the ALS profiles of two SLSs with the same layer thicknesses and compositions which were grown under the same conditions. The lower curve is the same as the lower curve in Fig. 4 (an Sb-like SLS.) For the upper curve, each layer is terminated in an As soak, and 0.64 ML of $Ga_{0.76}In_{0.24}$, without an accompanying group V flux, are deposited at the beginning and end of the GaInSb layer. This SLS is termed "As-like." The two curves are vertically offset by five pixels for clarity and the positions of the group V soaks are marked by the pairs of vertical lines, Sb in the case of the lower curve and As for the upper curve. In this case the interface shuttering schemes are very different as are the ALS profiles. While both curves show decreasing streak separation, with oscillations having a period equal to a monolayer deposition time, at the start of the InAs layer, the relative sizes of the streak separation at the beginning and end of the InAs layer are different in the two curves. In the upper curve (As-like) the separation decreases over the course of the InAs layer while in lower curve (Sb-like) the streak separation increases after growing the InAs layer. In addition, the response of the ALS to the group V soaks is very different. In the upper curve, the ALS does not change during the As soaks on the InAs layer (see the third vertical line), while at the interrupts on the GaInSb layers (the first and fifth lines) the streak separation rises very slightly. In the lower curve, the ALS is not affected by the Sb soaks on the GaInSb layers (first and fifth vertical lines), while the ALS changes markedly at the Sb soak on the InAs layer and at the start of the GaInSb layer. The sharp dip in the streak separation in the upper curve during the GaInSb layer (between the fourth and fifth lines in the upper curve) is a reproducible feature and not a noise spike. Figure 5 shows that the ALS profiles of structures grown with very different interface shuttering schemes are also very different.

These two type of interface schemes shown in Fig. 5 are especially interesting to compare. We have found that undoped SLSs grown with Sb-like interfaces have a *p*-type background carrier concentration as determined by low temperature, four-point Hall measurements and that undoped As-like, SLSs have *n*-type background carrier concentrations. It is our hope that careful study of the RHEED characteristics of these different shuttering schemes will help us understand this behavior.

IV. DISCUSSION

Interpreting the change in streak separation of these SLSs is complicated. The greatest difficulty is understanding the relationship between the surface periodicity we are measuring and the final bulk lattice constant. X-ray diffraction measurements on these structures are consistent with the SLSs being coherently strained to the GaSb buffer layer, yet the periodicity of the growth front is clearly changing. One possibility is that as the strained material is grown, it nucleates in islands which have interatomic spacings that are intermediate between those of the coherently strained structure and the natural lattice constant of the material. If this were the case, it would only be after the islands coalesced and were buried under subsequent layers that the material would reach its final, coherently strained-lattice constant. This explanation is consistent with oscillations in streak separation having a period equal to the monolayer deposition time. A second difficulty is that the RHEED pattern moves during growth. We have observed rotations of the diffraction pattern about an axis parallel to the direction of electron travel and translations normal to the direction of electron travel or both. Without corrections for these effects, quantitative measurements are impossible. Finally, the dearth of techniques, which are capable of probing the chemistry and smoothness of individual, buried interfaces complicates analysis of the final structures. Nevertheless, we are hopeful that this technique will prove to be useful in understanding mixed anion interfaces at least to the level of providing an empirical tool for growth of high quality, reproducible SLSs.

In order to understand the experimental results reported in Sec. III, we have begun the study of the growth of GaAs on GaAs in the hope that this simpler system will be more tractable. Many of the phenomena seen in the InAs/Ga$_{1-x}$In$_x$Sb SLSs were also seen during GaAs growth: variations in the period of the RHEED intensity oscillations along the RHEED streaks, oscillations in the positions of RHEED streaks having the same period as the growth rate, translations of RHEED pattern normal to the direction of travel of the incident electrons and rotations of RHEED patterns about an axis parallel to the direction of travel of the incident electrons. However, when the rotations of the diffraction pattern are accounted for there is no change in the streak separation, to within the sensitivity of our measurement. Due to space limitations, the results of this study can not be given here and will be published elsewhere.[12]

V. SUMMARY

In conclusion, we have used RHEED to study the surface periodicity of the growth front of InAs/GaInSb SLSs. We found that the apparent surface lattice spacing reproducibly changed during growths which subsequent x-ray measurements indicated were coherently strained. Abrupt changes in the measured streak spacings were found to occur when the oven shutters were either opened or closed. Care was taken to check that these changes in the RHEED pattern were due to changes in the fluxes incident on the growth surface and not electrical or mechanical artifacts of the shutter actuator mechanism. The profile of the dynamic streak spacing was found to be reproducible when comparing consecutive periods of a SLSs or different SLSs employing the same shuttering scheme at the InAs/GaInSb interface. Finally, when the interface shuttering scheme was changed, it was found that the dynamic streak separation profile also changed. Large changes in the shuttering scheme led to dramatic differences in the streak separation profile, and small changes in the shuttering scheme led to minor changes in the profile. In both cases, the differences in the surface periodicity profile occured at the parts of the growth where the incident fluxes differed.

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