Reflectance anisotropy spectra from Si δ-doped GaAs(001): Correlation of linear electro-optic effect with integrated surface field

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Reflectance anisotropy spectroscopy (RAS) has been employed *in situ* to investigate the overlayer growth of GaAs onto submonolayer to one monolayer coverages of Si δ layers deposited on the GaAs(001)-$c(4\times4)$ surface. The intensity of RAS features, thought to arise from the linear electro-optic (LEO) effect, is found to vary with both the number of atoms in the Si δ layer and the position of the δ plane from the GaAs surface. Self-consistent solutions to Poisson's equation are made to calculate the electric field in the near-surface region of the samples. The results show a direct correlation between the LEO intensity and the surface field averaged over the penetration depth of the incident radiation, in confirmation of the LEO model. [S0163-1829(97)00648-6]

I. INTRODUCTION

Considerable effort has been employed, over many years now, to characterize fully the nature of the reconstructions obtained from GaAs surfaces prepared under different growth conditions. More recently, similar attention has become focused on the way in which these reconstructions become altered after planes of dopant atoms, such as Si or Be, are deposited on the GaAs surface. The majority of these studies have employed either reflection high-energy electron diffraction (RHEED), or a combination of RHEED and scanning tunneling microscopy as tools to probe the atomic bonding which gives rise to the observed surface reconstructions.[1–3] Although these investigations have explored a wide manifold of possible reconstructions, using a broad range of growth conditions, only a small number of studies have been aimed specifically at probably the most technologically relevant Si/GaAs interfaces, i.e., those prepared at low temperature (400 °C and below).[4–6] At these growth temperatures, the spread of Si atoms away from the δ plane remains small enough for practical δ-doping applications.

A combination of reflectance anisotropy spectroscopy (RAS) and RHEED measurements has been used recently to study submonolayer coverages of Si on the GaAs(001)-$c(4\times4)$ surface, at a substrate temperature of 400 °C.[7,8] One of the observations to come out of this and other RAS studies[9,10] of the GaAs(001) surface is that there are characteristic spectral features that vary in a systematic manner with the reconstruction of the surface. The most well known of these features occurs at an energy of $\sim$2.65 eV and changes in both shape and sign from a pronounced minimum for the $c(4\times4)$ reconstruction, to a maximum for the (2×4) reconstructed GaAs(001) surface.[7,10] This change reflects an alteration in surface bond orientation from along $[\bar{1}10]$ to $[110]$, and contributes to making RAS measurements extremely sensitive to the degree of dopant coverage. To date, a sensitivity to overlayer coverages of as little as 0.005 ML of either Si or Be has been demonstrated.[8]

More recently, the overlayer growth of GaAs on top of different submonolayer coverages of Si on the GaAs(001)-$c(4\times4)$ surface has been studied.[11] This is a subject of direct relevance to the practical inclusion of δ layers within III-V semiconductor devices. The interpretation of the RAS spectra has been shown to be somewhat more complicated in this case since the measured anisotropy reflects not only changes in the surface order, but is also influenced by the thickness of the overlayer and is correlated with the electric field at the surface of the sample.[11] This field, perpendicular to the surface, is simply that arising from the surface depletion region of the semiconductor, due to pinning of the Fermi level at the surface.[11(a)]

The dependence on surface field has been studied previously for *uniformly* doped GaAs (Refs. 12 and 13) and has been shown to give rise to a feature in the RAS spectrum at around 3 eV, attributable to the linear electro-optic (LEO) effect associated with the E_1 and $E_1+\Delta_1$ interband transitions. However, the present lack of a quantitative theory relating the LEO effect to the RAS spectrum, and limited experimental data, merits further investigation of this phenomenon. A more detailed understanding of the LEO effect is required when employing δ doping, and this forms the main thrust of the experiments we report here. By δ doping, it is possible to alter the strength of the depletion electric field in two ways: either by changing the *concentration* of Si atoms in the δ layer, or by varying the *position* of the δ layer with respect to the surface. Thus we can examine, in a controlled manner, the influence on RAS of the depletion electric field. Moreover, we shall show that our experiments demonstrate that the RAS anisotropy is influenced by the electric field in the *whole* of the near-surface region, not only that exactly at the surface.

In this paper, we focus on the development of the LEO feature as a function of both GaAs overlayer thickness and Si concentration. We make calculations of the depletion electric field using a simple self-consistent Poisson solver, and show that there is a direct quantitative relation between the integrated RAS spectrum intensity and the electric field in the near-surface region, in agreement with the LEO effect.

II. EXPERIMENT

All growth took place on GaAs(001) on-axis substrates within a VG Semicon V80H molecular-beam epitaxy (MBE) reactor, fitted with a VG LEG 110 RHEED system. Experimental procedures such as substrate preparation, growth temperature, and MBE flux calibration details can be found elsewhere.[14] The in-house constructed RAS system was positioned at a strain-free pyrometer viewport situated on the MBE reactor growth chamber. A description of the integrated MBE/RAS system can also be found elsewhere, the design being based upon that due to Aspnes *et al.*[15,16] The RAS system, which has a working spectral range from 1.5 to 5.5 eV, measures the difference (Δr) between the anisotropic complex reflectance (r) along the $[\bar{1}10]$ and $[110]$ optical eigenaxes within the (001) surface crystallographic plane, normalized to the mean reflectance ($\bar{r}$):

$$\frac{\Delta r}{\bar{r}} = 2\,\frac{r_{[\bar{1}10]} - r_{[110]}}{r_{[\bar{1}10]} + r_{[110]}}.$$

Only the real part of the RAS signal was investigated, since even small residual strain effects, associated with the pyrometer viewport, significantly affect the imaginary component of the RAS signature.[15]

Following the complete thermal desorption of the GaAs surface oxides at ~600 °C under an As_4 flux of $F_{As} \sim 5\times10^{14}$ molecules cm^{-2} s^{-1}, a sharp, clear GaAs(001)-(2×4)-β reconstruction was observed by RHEED. A 1 μm, undoped GaAs buffer layer was then grown at a rate of 1 μm h^{-1}, with an As_4/Ga flux ratio of ~0.8, at a temperature of 580 °C. The sample was cooled to 400 °C for both the deposition of Si and subsequent overgrowth with GaAs. The Si was deposited from a standard 40 cm^3 VG Mk. II effusion cell, which had been rigorously calibrated from a large series of Hall effect and electrochemical C-V profiling measurements. The effusion cell temperature was set to T_{Si}=1120 °C, which yielded a flux of $F_{Si} \sim 2.8\times10^{11}$ atoms cm^{-2} s^{-1}, such that a single monolayer would be deposited in ~0.64 h. To replicate the growth conditions encountered during δ doping, the As_4 flux was incident onto the sample surface at all stages throughout the experiment. Si coverages of 0.001, 0.0025, 0.005, 0.01, 0.1, and 1.0 ML were investigated in this study. GaAs overlayer coverages started at 1 ML and doubled with each successive deposition, up to a maximum of 512 ML. RAS spectra were recorded for the clean GaAs(001)-$c(4\times4)$ surface, at 400 °C, and after each deposition. The RAS intensities presented here differ from those in our previous publications,[7,8,15] as noted in Ref. 17.

III. RAS SPECTRA FOR GaAs-Si-GaAs

RAS and RHEED data indicate that, under the growth conditions employed here, low-temperature (400 °C) growth results in a partially disordered surface which, when growth is terminated, recovers slowly. As discussed previously,[11] a time scale of >1 h is required to recover fully the intensity of the 2.65-eV minimum at this growth temperature. This is a somewhat unrealistically long time to use when studying GaAs overgrowth on Si/GaAs by a cycle of sequential growth and RAS measurements, under UHV conditions.

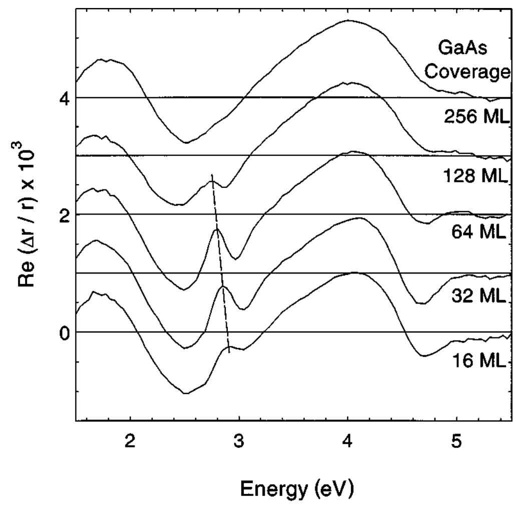

FIG. 1. RAS spectra for increasing GaAs coverage on top of 0.01 ML Si/GaAs, showing the development of the LEO feature at ~2.9 eV. Dashed line indicates redshift.

Hence, we have chosen to systematically record RAS spectra 5 min (300 s) after each deposition. This has been gauged to result in an uncertainty of ~8% in the intensity of the 2.65-eV minimum, which we have taken to be an acceptable systematic uncertainty in our study of GaAs overgrowth on Si/GaAs.

Figure 1 shows a series of RAS spectra following overgrowth with 8, 16, 32, 64, 128, and 256 ML GaAs on 0.01 ML Si/GaAs. Each spectrum is plotted using the same absolute scale, but displaced vertically for clarity. The position of the zero line has been included in each case. The RAS spectrum for 8 ML GaAs shows an inflection at ~2.9 eV, the signature of an LEO-related feature. The intensity of the LEO feature is found to increase initially with GaAs coverage, reach a maximum at an overlayer thickness of 64 ML GaAs, and then decrease with additional GaAs coverage. At the same time, there is a corresponding redshift (maximum value ~100 meV) in energy of the LEO feature (as indicated by the dashed line in Fig. 1). One might expect the decrease in LEO intensity for thicknesses >64 ML GaAs to be explicable simply by the electric field at the surface decreasing as the δ layer is buried more deeply, or the δ layer moving beyond the penetration depth of the light, but this does not explain the behavior for thicknesses less than 64 ML. Therefore, we suggest the most likely explanation for the observed trend is that the LEO intensity depends, in fact, on some average *near-surface* field. We develop this model in Secs. IV and V.

In Fig. 2 we display RAS spectra for 64 ML GaAs overgrowth, the thickness at which the LEO feature is fully developed, on all the Si submonolayer coverages we have studied. Once again all spectra have been plotted with the same absolute scale, but displaced vertically for clarity. The overall shapes of the RAS spectra are remarkably similar, excluding the contribution of the LEO-related feature, considering that the Si coverages span three orders of magnitude from 0.001 to 1.0 ML. It is to be noted, from our previous RAS measurements for Si on GaAs (001),[7] that a Si coverage of 0.1 ML corresponds to a crossover in the behavior of the 2.65 eV feature. At coverages of <0.1 ML Si, both

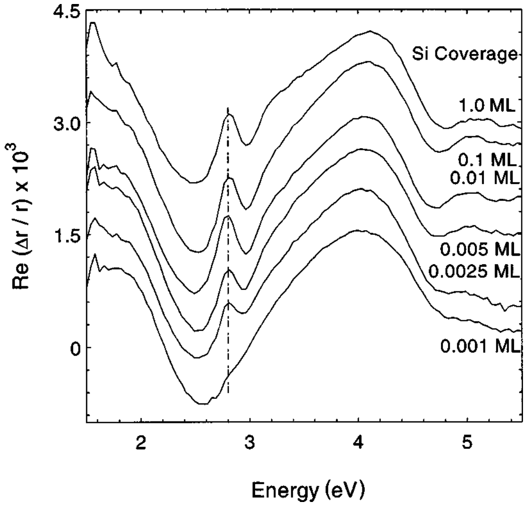

FIG. 2. RAS spectra for 64 ML GaAs deposited on Si/GaAs, where the Si coverages vary from 0.001 to 1.0 ML. Dashed line indicates absence of redshift.

$c(4\times4)$ and (2×1) reconstructions appear to coexist and so the overall RAS signal contains contributions from both surface phases. For Si coverages >0.1 ML, the (1×2) reconstruction dominates and the 2.65 eV feature shows a positive rather than a negative peak. Hence, the similarity in the overall shape of the RAS spectra indicates that overgrowth with 64 ML GaAs is sufficient to restore the surface order, even for GaAs overgrowth on 1 ML Si/GaAs.

It is evident that the intensity of the LEO-related feature increases with Si submonolayer coverage up to 0.01 ML, and then decreases slightly with further coverage up to 1.0 ML Si. A similar correspondence has been reported between the density of Si_{Ga} (i.e., Si on donor sites) and the total Si coverage, up to a coverage of $\sim10^{13}$ cm^{-2} (0.016 ML).[18] In that case, the measured density of Si_{Ga} then remained approximately constant up to a coverage of $\sim4\times10^{14}$ cm^{-2} before beginning to decrease, in good agreement with the results presented here. Figure 2 also indicates that, in this case, there is no observable variation in energy of the LEO feature with Si submonolayer coverage.

IV. CALCULATION OF SURFACE AND NEAR-SURFACE FIELDS

For uniform doping, to a good approximation the electric field in the surface depletion region of the GaAs is a maximum at the surface of the sample and decreases linearly with distance into the sample, reaching zero at the depletion width. Introducing a δ layer produces a more complex behavior. Very approximately (if the δ layer is not too far from the surface compared with the depletion width) the electric field is constant from the surface to the δ layer, and then decreases linearly with distance beyond the δ layer (reaching zero at a distance less than the depletion width without δ doping). For our samples, this behavior happens on a length scale comparable to the penetration depth[19] of the incident radiation at 3 eV (~17 nm or 60 ML GaAs).

The influence of the electric field, normal to the sample surface, on the RAS feature is known to be linear in field amplitude.[12] The data of Fig. 1 cannot be interpreted using the electric field exactly at the surface, however, since this decreases monotonically with depth of the δ layer. We show here that a suitably defined *near-surface average* field experienced by the light, correlates very well with the RAS signal. We proceed to define this near-surface average, and show how it is calculated for our samples.

We denote the local field [strictly speaking, $-E(x)$] by du/dx, where $u(x)$ is the electrostatic potential and x is the distance into the sample. The field at the surface is then $du(x=0)/dx$. The mean electric field averaged over any distance L could be defined from the local field $E(x)$ in the following way:

$$\langle E\rangle=\frac{\int_0^L E(x)d(x)}{\int_0^L dx}=\frac{1}{L}\int_0^L\frac{du}{dx}\,dx=\frac{1}{L}\,[u(L)-u(0)],$$

which is essentially just the voltage drop across any region of length L. But this is not, of course, an appropriate average to calculate the influence on the incoming light, which decreases in intensity as it penetrates the sample. Instead we need a quantity that is independent of the region of integration (that is, converges as $L\to\infty$) and with an appropriate weighting for the light intensity. We therefore define the quantity

$$\langle E\rangle=\frac{\int_0^L e^{-x/\lambda}E(x)dx}{\int_0^L e^{-x/\lambda}dx}=\frac{1}{\lambda(1-e^{-x/\lambda})}\int_0^L e^{-x/\lambda}E(x)dx$$

$$\to\frac{1}{\lambda}\int_0^\infty e^{-x/\lambda}E(x)dx,$$

which reflects the *average* field as experienced by the light. We shall refer to this last integral as the ''*integrated surface field*.'' Here λ in these calculations is the experimentally determined penetration depth of 17 nm, for GaAs at a photon energy of 3 eV.[19] It is of course implicit in this expression that the RAS feature of interest is caused by a *linear* electro-optic effect.

For uniform (bulk) doping, it is easy to obtain an analytic expression for the field from the surface into the bulk and hence evaluate the integrated surface field. With nonuniform (δ) doping, however, we need to find the exact solution of Poisson's equation in the near-surface region numerically. This was done by solving self-consistently the finite-difference representation of Poisson's equation using a straightforward shooting method. In this method, Poisson's equation is integrated from the surface into the bulk using an initial trial for the surface electric field. This trial value is adjusted until the correct boundary condition deep in the bulk region (that is, zero electric field) is obtained. The resultant potential profile $u(x)$ can be used to calculate the electric field $E(x)$.

In the calculation, the incorporated Si atoms are assumed to form an ideal uniform delta sheet of donors (i.e., all electrically active and not spread in the x direction) within the GaAs crystal. Background bulk doping levels of

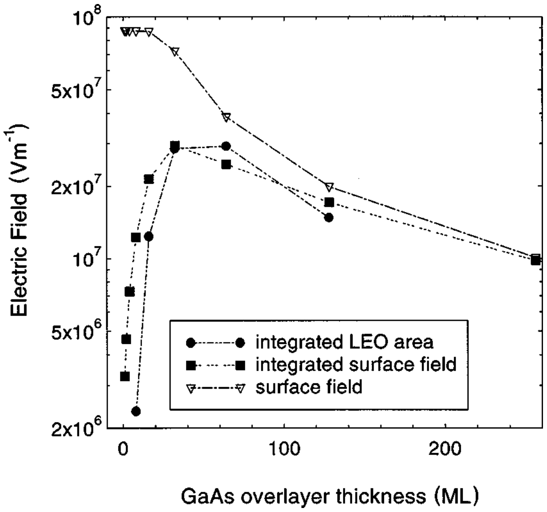

FIG. 3. Calculated values of surface and integrated surface fields for GaAs overgrowth on 0.01 ML Si/GaAs, together with experimentally determined LEO intensities.

2×10^{15} cm^{-3} (n-type, donor ionization energy 0.0053 eV) and 1×10^{15} cm^{-3} (p-type, acceptor ionization energy 0.020 eV) were assumed. Electron and hole effective masses were taken as 0.067 and 0.41 times the free-electron mass, respectively. The GaAs band gap was taken as 1.42 eV, and the relative permittivity as 13.1. The surface Schottky barrier was taken as 0.72 eV. One monolayer of GaAs is taken to have a thickness of 0.2825 nm. For the Si doping, a surface concentration of one monolayer is equivalent to 6.265×10^{14} atoms cm^{-2}.

V. COMPARISON OF CALCULATED FIELD WITH LEO INTENSITY

In previous studies,[12,13] an accurate determination of the integrated LEO area was facilitated by being able to subtract the RAS spectrum for an undoped GaAs(001) sample from the spectra obtained for GaAs layers with different degrees of bulk doping, but similar surface structure. In the present case (as discussed earlier), the RAS spectra for GaAs overgrowth on 0.01 ML Si/GaAs are not identical to that for the clean GaAs(001)-$c(4\times4)$ surface, hence the integrated LEO area cannot be obtained by a similar process of spectral subtraction. The approach taken here has been to interpolate linearly between two points on either side of the LEO feature, and to integrate numerically the deviation of the RAS data over this region.

Figure 3 contains a comparison between the calculated surface field and integrated surface field values for GaAs overgrowth on 0.01 ML Si/GaAs. The superimposed experimental data has been multiplied by a scale factor to facilitate comparison with both calculated curves. It is evident that the observed decrease in LEO intensity for overlayer thicknesses both above and below 64 ML GaAs is reproduced extremely well by the behavior of the integrated surface field. The maximum in integrated surface field can be explained qualitatively as follows: The electric field between the surface and the δ layer can be fairly high compared with the ordinary depletion field (particularly when the layer is close to the surface and heavily doped), and so will contribute strongly to the integral for the integrated surface field. However, when the δ layer is close to the surface, the *range* of its contribution to the integral is small (zero, in the limit that the δ layer is *at* the surface) and increases as it moves further from the surface. As the δ layer moves still further from the surface, however, the reduced field between the δ layer and surface decreases its contribution to the integral. Eventually, when it is buried by more than the depletion width, the δ layer has no effect on the field in the depletion region. The variation in energy of the LEO feature with increasing GaAs thickness seen in Fig. 1 is also consistent with the position of the δ layer moving through the penetration depth of the incident radiation.[20]

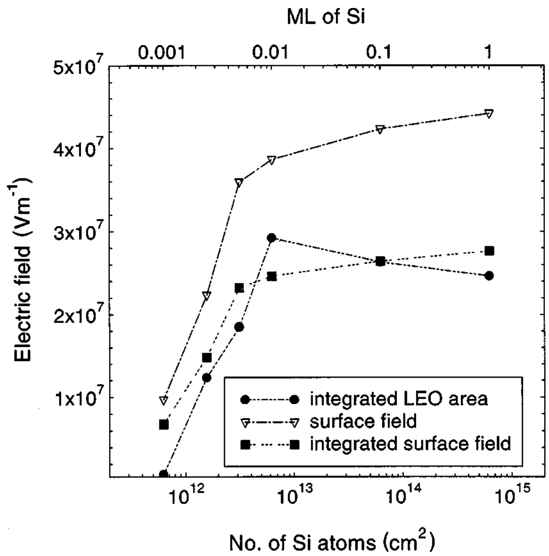

FIG. 4. Calculated values of surface and integrated surface fields for 64 ML GaAs overgrowth on Si/GaAs, and comparison with measured LEO intensities, where the Si content ranges from 0.001 to 1.0 ML (6.27×10^{11} to 6.27×10^{14} atoms cm^{-2}).

Figure 4 shows a similar comparison between the calculated surface field and integrated surface field for 64 ML GaAs overgrowth on all the Si submonolayer coverages we have studied. Once again, the LEO intensities have been multiplied by a scale factor, *identical* to that used in Fig. 3. In this case, there is little to choose between the two calculated dependencies of field on Si content, since both curves follow the slope of the experimental data for low Si concentrations before reaching a knee around 6×10^{12} atoms cm^{-2} (0.01 ML Si). From this point, the calculated curves continue to show an increase with Si content, while the LEO intensity decreases. Such a disparity is to be expected, however, since the solution of Poisson's equation in these cases assumes all the Si atoms to be electrically active, i.e., does not allow for the saturation in the number of Si donors which is known to occur[18] and is apparent in the integrated LEO area data.

VI. CONCLUSIONS

In situ reflectance anisotropy measurements in Si δ-doped GaAs(001) have been explained in terms of the linear electro-optic effect. The use of Si δ layers has enabled the strength of the electric field in the near-surface region to be altered either by changing the concentration of Si atoms in the δ layer, or by varying the position of the δ layer with respect to the surface. In both cases, self-consistent solutions to Poisson's equation indicate a quantitative correlation between the integrated RAS intensity and the surface field averaged over the penetration depth of the incident radiation. This correlation also extends qualitatively to the redshift observed in peak energy, for a fixed Si δ-layer concentration, when the thickness of the GaAs overlayer is varied. However, the absence of any variation in LEO energy with Si submonolayer coverage, at a fixed GaAs overlayer thickness of 64 ML, is difficult to interpret within the framework of the current model.

Finally, the sensitivity of RAS as an *in situ* probe of electronic properties has been emphasized further by the use of the LEO intensity as a direct indication of the level of activity of Si donors within a single δ plane.

ACKNOWLEDGMENTS

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