Optical and Structural Properties of Low Temperature GaAs Layers Grown by Molecular Beam Epitaxy

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A detailed photoluminescence, photoreflectance and X-ray study has been performed on a series of undoped and n-type low-temperature GaAs layers grown by molecular beam epitaxy at different substrate temperatures. Intense deep level emission bands were obseved in the photoluminescence spectra of samples grown below 450°C. These emissions were related with an excess concentration of arsenic antisite and/or interstitial defects and with complexes involving gallium vacancies. At very low substrate temperatures the excess of arsenic causes a lattice expansion of the epilayer of about 0.16%. Several features were observed in the photoreflectance spectra below, close to and above the fundamental GaAs band gap. They were attributed to impurity related, optical etalon effects, exciton transition and Franz-Keldysh oscillations. From the lineshape changes observed in the modulated reflectance experiments we identified a transition substrate temperature at 350°C, below which the insulating character of the epilayers is strongly enhanced due to a higher incorporation of defects. We also performed photoreflectance experiments at E_1 and $E_1 + \Delta_1$ spectral region for these samples. The observed results indicate that the electric field modulation has contributions not only from the sample surface but also from the epilayer/substrate interface.

I. Introduction

Low temperature (LT) GaAs epitaxial layers grown by Molecular Beam Epitaxy (MBE) have attracted considerable interest due to its technological importance such as for semi-insulating buffers in GaAs based electronic devices as well for high speed photodetectors[1,2]. The understanding and control of the insulating properties of these LT-GaAs layers are very important for future development of electronic and optoelectronic devices using this kind of material. Several works were dedicated to study optical, electrical and structural properties of both as-grown and annealed LT-GaAs layers[3-9]. However, a systematic study of LT-GaAs optical properties obtained at different growth conditions has not been done. In this work we report optical and structural investigation on a series of undoped and silicon doped LT-GaAs layers obtained at different substrate temperatures (T_s). Photoluminescence (PL), photoreflectance (PR) and X-ray measurements were used to study the properties of these layers. The combination of these techniques provides significant information about the band gap, deep radiative defects, built-in electric field strength and lattice constant. Our results indicate that there is a critical substrate temperature below which the insulating properties of the epilayers is enhanced.

II. Experimental details

The samples analysed in this work were grown in a single chamber MBE machine (MECA-2000) on semi-insulating [100] and [311] and Si-doped [100] oriented GaAs substrates. Two sets of samples were grown: undoped and silicon doped (n $\cong 5 \times 10^{17}$ cm^{-3}) LT-GaAs layers. The growth rate, defined from RHEED pattern oscillations, was kept constant at 1.0 μm/h. The nominal thickness of all the epitaxial layers is 1 μm and they were obtained with substrate temperatures ranging from 200-550°C.

Double-crystal X-ray diffraction measurements were performed with CuKα_1 radiation. The symmetric 400 reflection was used to obtain the rocking curves.

Photoluminescence measurements were performed at 77K using the 514,5 nm line of an Ar$^+$ laser as the excitation source. The spectra were obtained with a 1/4 meter spectrometer and the PL signals were detected with a liquid-nitrogen cooled Ge detector. Photoreflectance spectra were taken at room temperature. The probe and pump beams consist of a 55W tungsten lamp and the 633 nm line of a He-Ne laser, respectively. In all the experiments both beams are defocused and their power density were kept below 100 μW/cm^2 to avoid possible photovoltaic effects[10]. The spectra were taken with a 1/4 meter spectrometer and the reflected probe beam was detected with a Si photodiode. In both techniques synchronous detection, at a typical frequency of 200 Hz, were used with lock-in amplifiers.

III. Results and discussion

Fig. 1 shows representative X-ray rocking curves for undoped LT-GaAs/GaAs-n$^+$ layers grown in the temperature range 200-300°C. At very low substrate temperature ($T_s = 200$°C) it was observed two peaks. The dominant X-ray signal comes from the GaAs substrate. The second peak was attributed to large amounts of excess arsenic that were incorporated in the epilayer, as interstitial defects, giving rise to a perpendicular lattice expansion[9] From the difference between these two peaks we determinated a lattice mismatch of 1.6×10^{-3}. In the case of the LT-GaAs layer grown at 250°C it was observed that the X-ray signal from the layer and from the substrate are almost coincident and a very small asymmetry is present at the left side of the X-ray peak. For samples grown at 300°C, or even at higher temperatures, the X-ray signal from the GaAs substrate and the epilayer coincide and only one symmetric peak was observed.

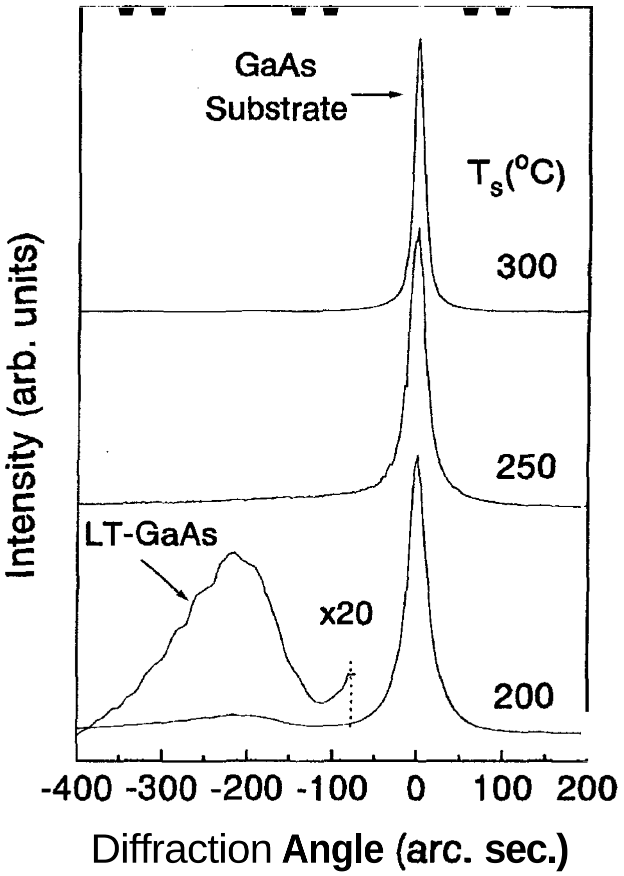

Figure 1: X-ray rocking curves of undoped LT-GaAs layers grown at different substrate temperatures.

The 77M PL spectra of silicon doped LT-GaAs/GaAs(S.I.) and undoped LT-GaAs/GaAs(n$^+$) are shown in Figs. 2a and 2b, respectively. When the substrate temperature was lowered from 550 to 350°C the PL intensity becomes weaker and below 350°C no emission line could be detected for these two sets of as-grown LT-GaAs layers. This behavior can be attributed to the presence of large amounts of nonradiative recombination centers formed in the LT-GaAs layers. We observed that the PL efficiency is higher in the case of Si doped samples. In the temperature range 450-550°C the dominant feature at around 1.5 eV is due to the near band-gap emission. The full width at half maximum (FWHM) is 40 meV and 12 meV for Si doped and undoped LT-GaAs layers, respectively. In the case of Si doped LT-GaAs samples it was observed a small shoulder below the GaAs band gap (1.4-1.455 eV). This emission line is completely absent in the PL spectra of undoped LT-GaAs layers grown in the same substrate temperature range. This peak can be associated with transitions involving Si acceptor species[11]. At T_s = 350°C a third PL peak (at ≅ 1.32 eV) was observed in the Si doped LT-GaAs sample. The origin *of this emission is not clear at the present*. For samples grown at 350 and 400 °C the PL spectra are dominated by deep-radiative emission lines. The insets shown in Fig. 2 are representative of the data taken on both sets of samples that exhibited similar behavior. These broad emission bands are centered at around 1.0 and 1.15 eV, with FWHM of order of 200 meV. for Si doped and undoped LT-GaAs layers, respectively. The deep-level emission close to 1 eV can be attributed to a transition involving the electronic trap EL2 and the valence band[6]. The emission bands observed at around 1.15 eV can be assigned to a transition from the conduction band edge to gallium vacancy-donor complexes[12]

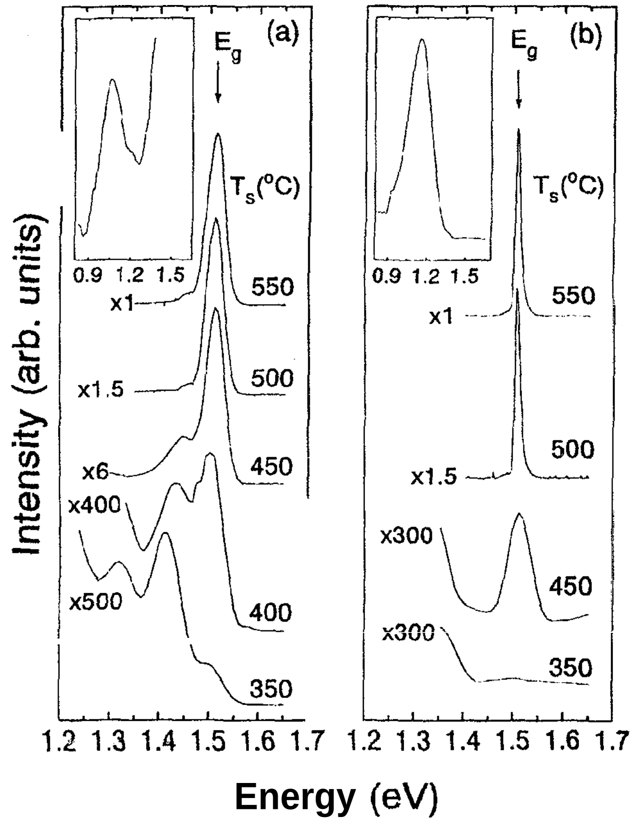

Figure 2: 77K PL spectra of (a) Si doped LT-GaAs layers on *semi-insulating* GaAs *substrate* and (b) *undoped* LT-GaAs layers on n^+-GaAs substrate, grown at different temperatures. In the insets it is shown representative PL spectra of the observed deep-level emission bands for both sets of samples.

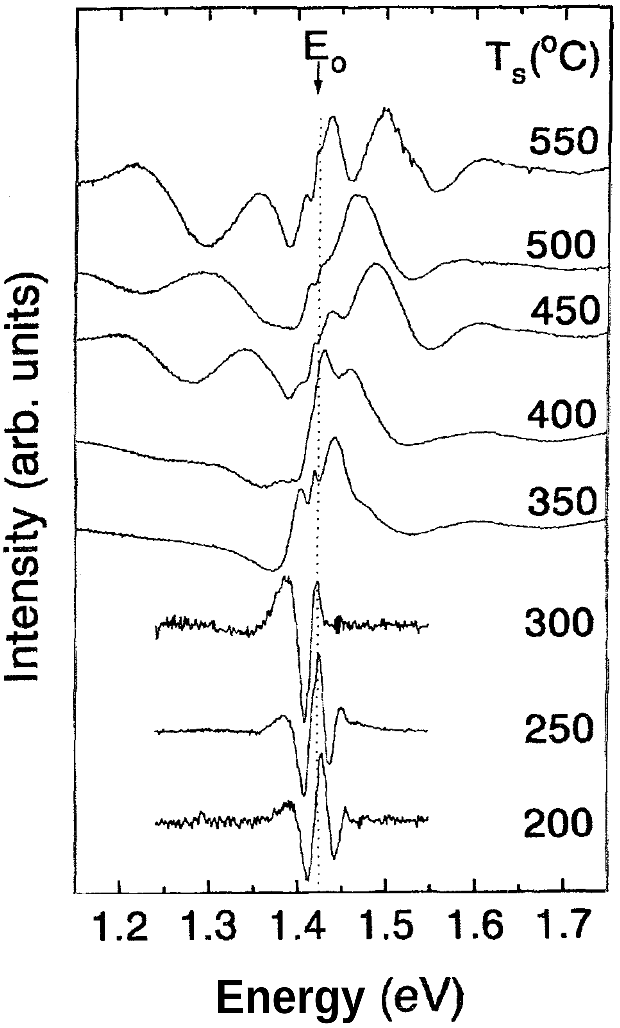

Figure 3: 300K PR spectra at the fundamental E_0 transition energy of silicon doped LT-GaAs layers on semi-insulating GaAs substrate grown at different temperatures.

Room temperature PR spectra of silicon doped LT-GaAs layers obtained at different substrate temperatures are shown in Fig. 3. The dotted line corresponds to the fundamental energy transition (E_0) of the GaAs band gap at 300K temperature[13]. The PR spectra of samples grown in the temperature range 350-550°C exhibited complex lineshapes and several features can be clearly observed below, close to and above the GaAs band gap. The structures above the band gap are attributed *to the Franz-Keldysh oscillations (FKO)*[13]. In the spectral region close to the fundamental gap sharp features are superimposed to this transition. We interpret this result as an excitonic transition from a sample region where the bands are almost flat with a built-in electric field strength not large enough to quench the exciton[14]. The features observed below the GaAs band gap are attributed to impurity related transitions[15]. As the substrate temperature was lowered from 550 to 350°C the PR signal from these structures becomes smaller, disappearing at 350°C. In the same temperature range the intensity of the sharp lines also decreases becoming negligible at low substrate temperatures. The observed results suggest a change of the layer resitivity due to the incorporation of a high density of defects as the growth temperature was reduced. This effect is enhanced for samples grown at lower substrate temperatures ($T_s < 350°C$). As an opposite to the PL measurements, the PR technique provides higher sensitivity allowing the observation of optical structures for LT-GaAs layers obtained at lower values of T_s. The PR spectral lineshape for samples obtained in the range $200 < T < 350°C$ are very different from those observed at higher T_s values. They are third-derivative like[16] with small broadening factors. The PR signal are very similar to those obtained for semi-insulating GaAs substrates[17]. All these results indicate the existence of a critical substrate temperature below which the incorporation of defects is so high that the layers become high resistive, confirming PL observations.

Fig. 4 shows the PR spectra of undoped LT-GaAs/GaAs(n$^+$) obtained at different substrate temperatures. As can be observed, the spectra shows less broadening effects and Franz-Keldysh oscillations as compared to the data shown in Fig. 3. Most of the samples exhibited third-derivative function signal. For samples grown at higher temperatures ($T_s \geq 450°C$) it was observed exciton transitions superimposed to the fundamental band gap. When the substrate temperature was lowered these features disappeared and only the E_0 transition can be observed. In some of the analysed samples (T_s = 500, 300 and 250°C) oscillatory structures were observed below the GaAs band gap. These features were attributed to interference of two reflected beams[18]: one from the sample surface and the other from the substrate/epilayer interface. The other three samples did not exhibit this behavior. We attributed this result to differences in the epilayer/substrate interface quality that unable constructive interference between these two reflected beams. The film thickness d can be derived employing a simple model of light interference between the film and substrate using the following expression:

$$d = \frac{m}{\left[2(n^2 - sin^2\theta)^{1/2}\left(\frac{1}{\lambda_{l+m}} - \frac{1}{\lambda_l}\right)\right]} \qquad (1)$$

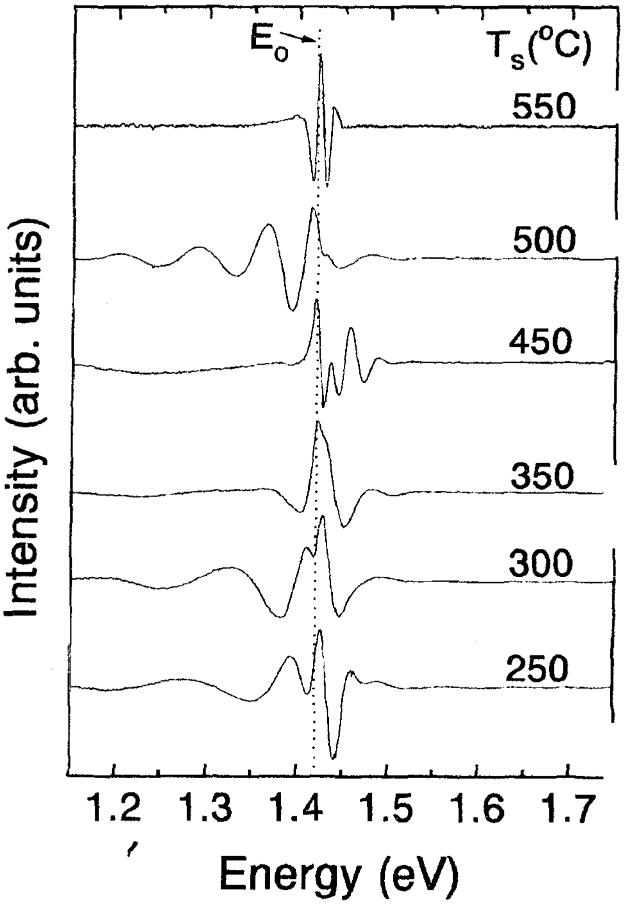

Figure 4: 300K PR spectra at the fundamental E_0 transition energy of undoped LT-GaAs layers on n^+-GaAs substrate grown at different temperatures.

where n is the GaAs refractive index, θ is the incident angle, and λ_{l+m} and λ_l are the probe wavelength of the lth and $(l+m)$th extrema, respectively. The oscillation periods for samples grown at 250 and 300°C are very similar but they are very different when compared with the sample grown at 500°C, indicating differences in the film thicknesses. The calculated thicknesses for samples grown at 250 and 300°C and for the sample grown at 500°C are 0.61 μm and 1.04 μm, respectively. The calculated thickness for the former samples is smaller than the expected value of 1μm. This can be attributed to variations in the growth rate at very low substrate temperatures.

In order to further improve informations about LT-GaAs layers we performed PR measurements at the E_1, $E_1+\Delta_1$ spectral region, where the penetration of light is about 10-20 nm. In this case the PR signal reflects the strength of the electric field close to the surface region. Fig. 5 shows the room temperature spectrum of a LT-GaAs layer grown at 350°C at the optical transition E_1 and $E_1+\Delta_1$. This spectrum is representative of the data taken on the other samples grown at higher temperatures that exhibited similar behavior. Dotted lines at 2.91 and 3.11 eV corresponds to the E_1, $E_1+\Delta_1$ optical transition, respectively. These energies were obtained by fitting the experimental data with a third derivative functional lineshape for a critical point $M_1^{[16]}$. For the samples grown at substrate temperatures lower than 350°C the PR signal in this spectral region was too small to be detected. This indicates that the Fermi level is kept close to midgap to make a flat band, due to the large incorporation of defects at lower substrate temperatures. As a consequence, the observed PR signal at the fundamental E_0 transition for samples grown in the temperature range 200-300°C have contributions not only from built-in surface electric field but also from the substrate/epilayer interface.

Finally we show in Fig. 6 the room temperature PR spectra of an undoped LT-GaAs layer grown at 300°C simultaneously on [100] and [311] oriented GaAs semi-insulating substrates. The data exhibited large number of Franz-Keldysh oscillations due to the high field and small broadening parameters. Oscillations up to the 6th and 13th extrema were observed for samples grown on [100] and [311] oriented substrates, respectively. The large number of FKO in the sample grown on [311] oriented substrate indicates a uniform electric field over a large region. The extrema in the FKO are given by[13]

$$m\pi = \phi + (4/3)[(E_m - E_0)/\hbar\theta]^{3/2} . \qquad (2a)$$

The above relation can be used to determine the built-in electric field **F**. The quantity m is the index of the *mth* extrema. ϕ is an arbitrary phase factor and E_m is the photon energy at the mth extrema. The electro-optic energy $\hbar\theta$ is[13]

$$(\hbar\theta)^3 = e^2\hbar^2F^2/2\mu , \qquad (2b)$$

where μ is the reduced interband effective mass (for the electron and the heavy hole pair) in the direction of the electric field F. Plotted in Fig. 6b is the quantity $(4/3)(E_m - E_0)^{3/2}$ as a function of index m for the spectra shown in Fig. 6a. The solid lines are linear fittings to Eq. (2a). The slopes yield electric fields of 41.3 and 39.1 kV/cm for the samples grown on [100] and [311] oriented GaAs substrates, respectively.

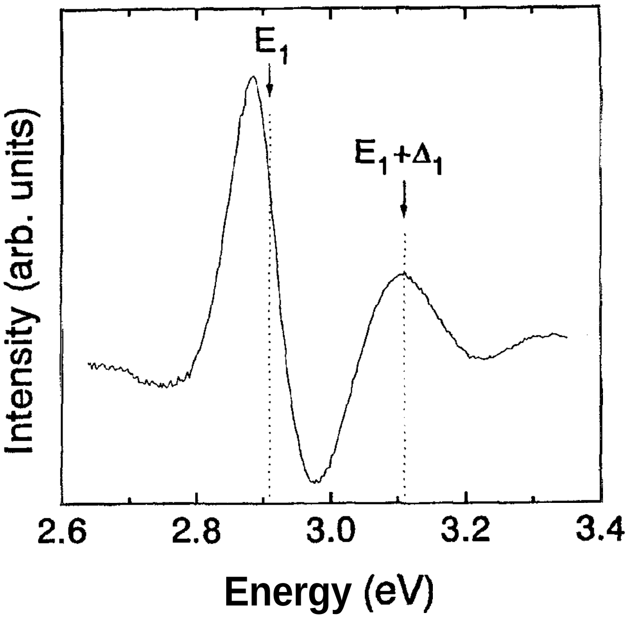

Figure 5: 300K PR spectrum at the E_1, $E_1+\Delta_1$ optical region of a silicon doped LT-GaAs layer on semi-insulating GaAs substrate grown at 350°C.

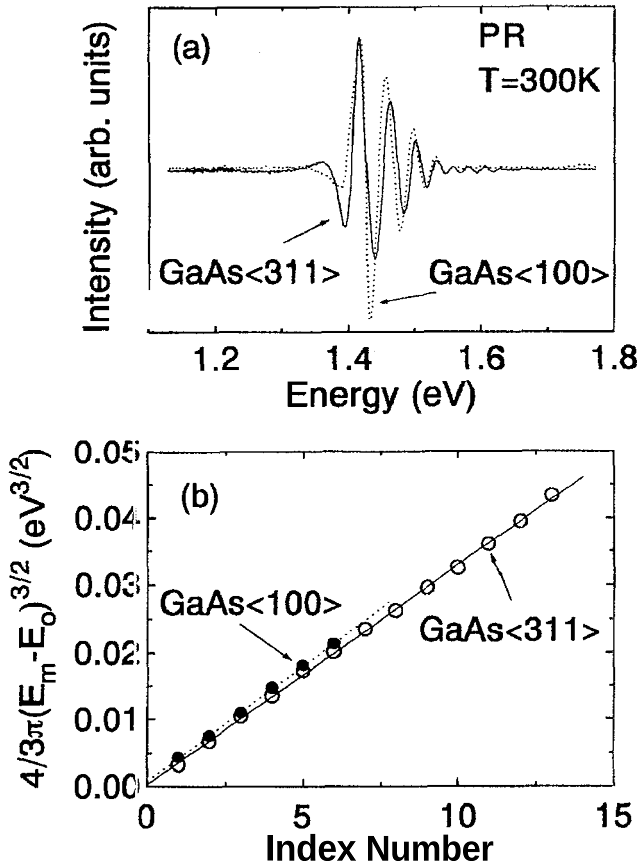

Figure 6: (a) 300K PR spectra at the fundamental E_0 transition energy of undoped LT-GaAs layers on [100] and [311] oriented semi-insulating GaAs substrate grown at 300°C, (b) Plot of $4/3\pi(E_m - E_0)^{3/2}$ against the index number m for both samples.

IV. Conclusions

We have performed a systematic study of the optical and structural properties of undoped and Si doped LT-GaAs samples grown by MBE at different substrate temperatures. The combination of photoluminescence, photoreflectance and X-ray diffraction experiments allowed us to evaluate the semi-insulating character and lattice distortion of these epilayers. From the PL and PR measurements we identified a critical substrate temperature at 350°C, below which the incorporation of defects is enhanced. In the PR spectra of some analysed samples it was observed optical interference effects that were used to measure the epilayer thickness. The LT-GaAs sample grown on [311] oriented GaAs substrate exhibited a very uniform electric field over a larger extension.

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