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Determination of surface lattice strain in ZnTe epilayers on {100}GaAs by ion channeling and reflectance spectroscopy

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We report on the direct measurements of surface lattice strain in ZnTe epitaxial layers on {100}GaAs substrates by ion channeling Rutherford backscattering spectrometry and low-temperature (10 K) reflectance spectroscopy measurements. The measured ZnTe strain is the superposition of the expected thermal (tensile) strain and a thickness-dependent residual compressive strain. Our data indicate that the removal of this residual strain is slower than the rate predicted by the equilibrium theory, following an apparent $h^{-1/2}$ power-law dependence on the epilayer thickness h , above ~ 100 nm.

High quality ZnTe epitaxial layers on GaAs substrates are of great interest for a large number of applications, ranging from blue-green light emitting devices to the use as buffer layers for CdTe and $\text{Cd}_x\text{Zn}_{1-x}\text{Te}$ epitaxy. However, the occurrence of both high lattice and thermal mismatches between ZnTe and GaAs affects the structural properties of the ZnTe epilayers, resulting in the generation of a high density of misfit dislocations (MDs) at the ZnTe/GaAs interface and of, respectively, compressive or tensile elastic strain fields in the epilayers. Optimal growth temperatures for molecular beam epitaxy (MBE) and metalorganic vapor phase epitaxy (MOVPE) of ZnTe are in the 300–350 °C range,^{1,2} giving rise to lattice mismatch values between ZnTe and GaAs $\sim -7.4\%$. Although the removal of such a high lattice mismatch by MD nucleation is expected to start since the very early stage of the epilayer growth, ZnTe layers whose thicknesses are much greater than the critical thickness can still exhibit some residual compressive strain.³ However, the experimental determination of this residual strain is made rather difficult by the high thermal mismatch between ZnTe and GaAs, which introduces relatively large additional tensile strains into the ZnTe epilayers when the structure is cooled down from the growth temperature to the measurement temperature.

Several reports in the literature^{4–7} have addressed the occurrence of strain in MBE and MOVPE grown ZnTe epilayers on {100}GaAs substrates, although to our knowledge only optical (i.e., indirect) methods have so far been used for this purpose.

In this letter, we report on the direct measurements of surface lattice strain in MOVPE grown ZnTe epilayers on {100}GaAs by ion channeling Rutherford backscattering spectrometry (RBS), whose results are then compared with low-temperature reflectance spectroscopy measurements performed on the same samples. The present work shows that the ZnTe measured surface strain is the superposition of a tensile (thermal) strain and a thickness-dependent residual compressive strain. These two contributions have been discriminated by a careful investigation of the surface strain as a function of the ZnTe thickness,

allowing us to determine the residual strain for each epilayer.

ZnTe layers have been grown at a nominal 350 °C on (100) oriented semi-insulating GaAs substrates by atmospheric pressure MOVPE, obtaining the epitaxial relationships: (100)ZnTe|| (100)GaAs and <011>ZnTe|| <011>GaAs.⁸ Different samples have been grown for the present measurements at about 2.1 $\mu\text{m}/\text{h}$ growth rate, their thicknesses ranging between 80 nm and 3.0 μm . A 2.0 MeV $^4\text{He}^+$ beam has been used to record random and channeling spectra for each ZnTe epilayer. Moreover, angular yield curves for a surface region have been recorded for several lattice directions inclined to the surface normal. To this purpose, a goniometer having both a repeatability and an overall precision of 0.01° has been used for the channeling measurements.⁹ Low-temperature (10 K) reflectance measurements have been performed in backscattering geometry by using a 150 W tungsten-halogen lamp as a light source and a 1 m monochromator equipped with a cooled photomultiplier tube and a lock-in amplifier. A monochromator entrance slit width of 30 μm has been used for the measurements, achieving a spectral energy resolution of about 0.1 meV.

Ion channeling RBS measurements have shown that the surface crystalline quality of the ZnTe epilayers is relatively high for thickness values above about 300 nm. This clearly appears in Fig. 1, where the value of χ_{\min} (i.e., the [100] aligned minimum yield just behind the Te surface peak normalized to the random yield) is reported as a function of the epilayer thickness. Above 300 nm we found constant χ_{\min} values at about 5% which compare well with the semiempirical estimate¹⁰ of $\chi_{\min} = 4.1\%$ expected for an ideal ZnTe crystal. Below 300 nm an increase of χ_{\min} is observed with decreasing epilayer thickness up to a value of about 18% for the thinnest (80 nm) of the ZnTe samples. This fact is related to the occurrence of lattice defects (most probably dislocations) in a 300 nm thick region close to the ZnTe/GaAs interface. Also, our ion-channeling spectra indicate that a higher defect density zone, extending for about 100 nm from the heterointerface, can be identified within this defected region. This can be

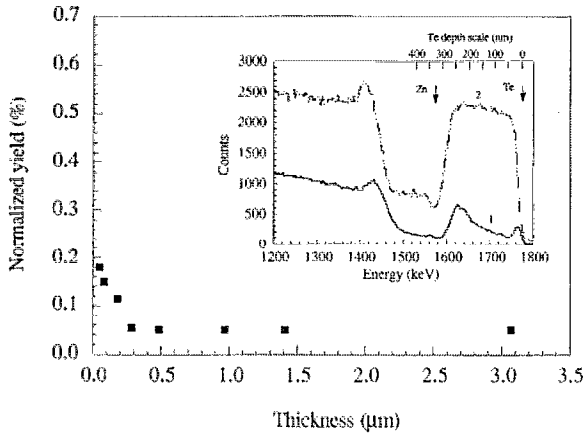


FIG. 1. The values of the [100] aligned minimum yield χ_{\min} as a function of the ZnTe epilayer thickness. Typical 2.0 MeV $^4\text{He}^+$ RBS spectra are shown in the insert for a 284 nm thick sample: (1) [100] aligned and (2) random geometry.

inferred by the relatively high in-depth dechanneling rate observed in the aligned RBS yield shown in the insert of Fig. 1. Despite this high dechanneling rate, the crystalline quality remains fairly good even for the thinnest layers, allowing us to perform meaningful ion channeling strain measurements on all the present samples. Therefore, values of the tetragonal distortion $\epsilon_T = a_{\perp} / a_{\parallel} - 1$, where a_{\perp} and a_{\parallel} are, respectively, the ZnTe lattice parameters normal and parallel to the ZnTe/GaAs interface, have been obtained by measuring the angular deviations of lattice directions inclined to the surface normal with respect to the corresponding directions in the unstrained lattice.⁹ The strain parallel to the interface has been then calculated from the relationship $\epsilon_{\parallel} = \epsilon_T / (1 + \alpha)$, the value of $\alpha = 2C_{12} / C_{11}$ being about 1.14 for ZnTe.¹¹

Ion channeling data have been compared with the corresponding parallel strain values derived from the strain-induced energy splitting of the light and heavy hole exciton resonances in the 10 K reflectance spectra of ZnTe epilayers thicker than 300 nm. Below such value the occurrence of extended defects deteriorates the optical quality of ZnTe and prevents reflectance measurements from detecting any excitonic resonance in the spectra. A typical reflectance spectrum is reported in Fig. 2. As a general trend, two sharp resonances associated with the heavy-hole (hh) and light-hole (lh) excitons can be resolved ~ 2.38 eV. The positions and the energy splitting of the hh and lh resonances are a signature of the type (tensile or compressive) and of the amount of strain present in the layers and can be compared to the calculated strain-induced shifts based on the Hamiltonian of Picus and Bir¹² to get a quantitative estimate of the crystal strain. The results of this calculation for ZnTe are shown in the insert of Fig. 2 (a constant exciton binding energy of 13 meV has been assumed). In order to determine the excitonic eigenstates, a detailed line-shape analysis of the reflectance spectra has been performed in the framework of the harmonic oscillator model.⁷ Lorentzian shape for the dielectric function around the exciton resonances and the formation of an electric-field-induced dead layer at the ZnTe crystal sur-

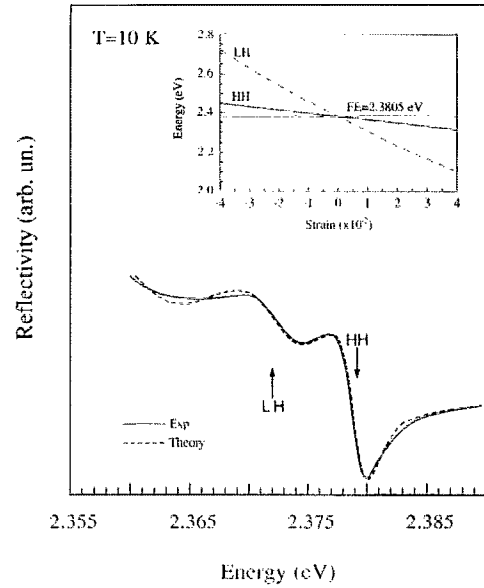


FIG. 2. A typical 10 K reflectance spectrum (solid line) obtained for a 2.04 μm thick ZnTe epilayer is reported together with the result of the line shape fitting (dashed line). The positions of the heavy (hh) and light (lh) hole exciton resonances are indicated. Their expected positions are reported as a function of the parallel strain in the insert, where FE indicates the free-exciton energy for bulk ZnTe.

face have been assumed, allowing to determine the hh and lh eigenstate energies within 1 meV precision.

The surface strain values determined by the present ion channeling and reflectance measurements are shown in Fig. 3. The reflectance data in the figure have been scaled from 10 K to room temperature by subtracting a 0.057% value to account for the thermal strain contribution between the two temperatures. Our data indicate that the two techniques are consistent in the epilayer thickness range where they can be compared. For layers thinner than 300 nm the measured strain is compressive. However, above ~ 300 nm the surface strain is tensile and it slowly increases with the ZnTe thickness, although it remains below the expected built-in thermal strain value. This suggests that the thermal strain is partly balanced by a thickness-

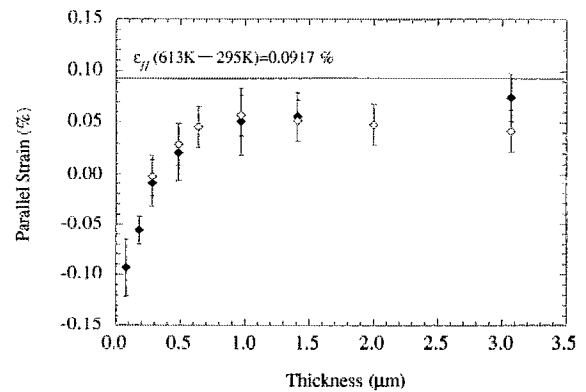


FIG. 3. The values of ϵ_{\parallel} obtained by ion channeling (full points) and reflectance (light points) measurements are reported as a function of the epilayer thickness. The horizontal line indicates the value of the expected in-plane thermal strain when the samples are cooled from the growth temperature (613 K) to room temperature (295 K).

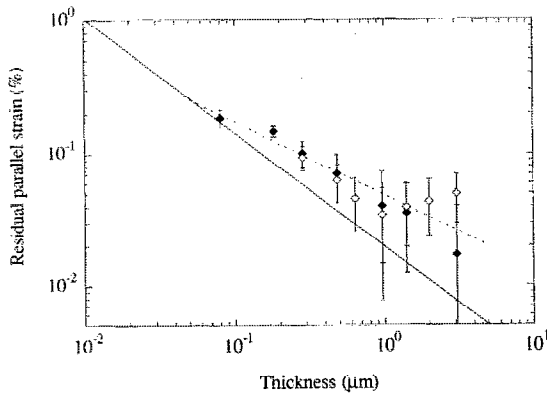


FIG. 4. The residual parallel strain obtained by ion channeling (full points) and reflectance (light points) data is shown as a function of the ZnTe epilayer thickness h . The solid line in the figure represents theoretical values obtained from the equilibrium model (Ref. 3). The dashed line represents the best-fit of the ion channeling values according to a h^{-m} power-law dependence, where $m=0.552 \pm 0.059$.

dependent residual compressive strain $\epsilon_{\parallel, \text{res}}$, which has not been totally relaxed by the formation of extended defects at the ZnTe/GaAs interface.

The expected thermal strain contribution to the total in-plane surface strain has been calculated to be $\epsilon_{\Delta T} = (0.0917 \pm 0.0092)\%$ on the basis of the currently available ZnTe and GaAs linear expansion coefficient data.¹³⁻¹⁵ For ZnTe thickness above 300 nm, our ion-channeling data show a continuous increase of the measured strain with the epilayer thickness toward the expected thermal strain value, suggesting that no relaxation actually occurs down to room temperature. On the contrary, the present reflectance data show a slow decrease of the measured tensile strain with the ZnTe thickness above 1.0 μm , which reduces to about 50% of the expected value for a 3.0 μm thick layer, also in accordance with Ref. 7. However, strain relaxation at low temperatures (below 300 K) is unlikely, in consideration of the activation energy involved in the MD nucleation and/or multiplication.¹⁶ Indeed, low temperature (1.7 K) PL measurements by Zhang *et al.*⁶ indicate that no thermal strain relaxation occurs for thick ZnTe layers, further supporting the above conclusion. In this respect, a badly resolved splitting of the hh exciton resonance due to polariton effects (about 1.5 meV) is observed in the best optical quality (i.e., thicker) samples, which could explain the discrepancy between reflectance and ion channeling data.

The ZnTe residual compressive strain $\epsilon_{\parallel, \text{res}}$ has been obtained by subtracting the calculated thermal strain $\epsilon_{\Delta T}$ from the measured in-plane values ϵ_{\parallel} for each epilayer. This is shown in Fig. 4, where the amount of residual strain at the growth temperature is reported as a function of the ZnTe epilayer thickness for both ion channeling and reflectance data. Our residual strain data show a monothonic decrease with increasing ZnTe thickness, although again reflectance data deviate from this behavior above about 1.0 μm . The solid line in the figure represents theoretical $\epsilon_{\parallel, \text{res}}$ values calculated following the equilibrium model of Ref. 3 for the ZnTe/GaAs heterostructure and assuming that 60° dislocations along [011] and $[\bar{0}\bar{1}\bar{1}]$ directions are formed at

the ZnTe/GaAs interface. It appears that all our strain data are systematically higher than the corresponding theoretical values, but for the thinnest of the measured samples, whose residual strain coincides with the expected one, within the experimental errors. A regression analysis has shown that our ion channeling strain values follow an apparent h^{-m} power-law-dependence on the ZnTe layer thickness h , whose best-fit exponent is $m=0.552 \pm 0.059$, the uncertainty of the calculated thermal strain affecting the best-fit value by less than ± 0.054 . The equilibrium curve in the same thickness range shows a value of about 0.89. This clearly indicates that above ~ 100 nm the strain relaxation of the ZnTe layers proceeds at a slower rate than the one predicted by the equilibrium model. It is noteworthy that this thickness also corresponds to that of the near-interface region, where a high density of defects is indicated by the dechanneling rate.

To our knowledge, this is the first detailed study of strain relaxation in ZnTe/GaAs or other II-VI based highly mismatched heterostructures ever reported in the literature. However, similar results have been reported for $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ heterostructures,¹⁷ for which the amount of residual strain above the epilayer critical thicknesses was shown to follow a typical $h^{-1/2}$ power-law dependence. This peculiar behavior can be explained in terms of a reduced MD generation rate, variously attributed by different authors either to MD interactions¹⁸ or to work hardening effects,¹⁹ which prevent the epilayer residual strain from relaxing according to the equilibrium theory.

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