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High resolution spectroscopy of free-standing GaAs films prepared by epitaxial liftoff

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Free-standing layers of high purity GaAs prepared by epitaxial liftoff are investigated using high resolution photoluminescence, optical transmission, and x-ray diffraction techniques. Low temperature (1.5 K) optical measurements of these thin, strain-free films yield spectra rich in structure, revealing much about the fundamental properties of the materials. X-ray diffraction analysis of layers as thin as 2000 Å produce well-resolved Pendellösung fringes, in excellent agreement with dynamical theory simulations. Once removed from their underlying substrates, these thin semiconductor cavities constitute unique systems for a variety of novel spectroscopic studies not possible in as-grown heterostructures. © *1998 American Institute of Physics.* [S0021-8979(98)00222-9]

I. INTRODUCTION

The epitaxial liftoff (ELO) technique has received widespread attention since its introduction a decade ago.[1] Many novel electronic and optoelectronic devices have sprouted from this process which would have, otherwise, been impossible to fabricate. Recently, advances with compliant substrates[2,3] have further increased the interest in ELO due to the essential role the process promises to play in the development of ''universal'' growth platforms for the deposition of semiconductor materials of virtually any lattice constant and crystal structure.

While considerable progress has been made on developing applications for ELO technology, surprisingly little attention has been given to the fact that the layers prepared using this method may constitute unique and interesting spectroscopic systems in their own right. In fact, only a handful of low resolution studies have focused on examining the structural, and/or, optical properties of thin film materials produced using this technique.[4–8] In all of these prior investigations the ELO structures, once fabricated, were subsequently bonded via van der Waals forces[9] or adhesives to other planar substrates (i.e., glass, Si, SiO_2, etc.) prior to undertaking measurements. While some form of post-ELO attachment would be an undeniable requirement in most device applications, the affixing process inherently introduces significant and undesirable strain if it is the properties of the microcavity itself that are of interest. This point is considerably compounded where high resolution, low temperature studies are concerned.

Photoluminescence (PL), optical transmission, and x-ray diffraction are powerful, nondestructive characterization techniques which can be used to reveal much about the properties of thin semiconductor films. In this article, these versatile tools are used to investigate high purity thin films of GaAs, prepared by ELO. High resolution, low temperature PL and transmission results are presented and indicate that it is possible to produce unstrained free-standing microcavities which exhibit spectra rich in sharp structure (some of which is not observable in the epilayers prior to liftoff). In addition, x-ray diffraction studies of ELO films as thin as 2000 Å are undertaken and spectra clearly exhibiting well-defined Pendellösung fringes are obtained in these materials.

II. EXPERIMENT

The GaAs material which formed the focus of this study was grown by molecular beam epitaxy (MBE) and typically consisted of ~2.3 μm of GaAs deposited on a 250 Å AlAs/1000 Å GaAs buffer layer grown on semi-insulating (SI) or *n*-doped GaAs substrates. A 2000-Å-thick sample was also grown by metalorganic chemical vapor deposition (MOCVD) for the x-ray diffraction measurements. In both cases the ELO samples were typically 0.25–1.0 cm^2 in size and were prepared using the technique first developed by Yablonovitch *et al.*[1] The free-standing layer used for the PL studies was passivated with 100 Å GaAs/250 Å $Al_{0.2}Ga_{0.8}As$ caps deposited on either side of the film during the initial MBE growth. Surface passivation of GaAs is well known to play an important role in increasing PL efficiency by reducing surface recombination velocities (see Ref. 10 and references therein). As these effects can drastically influence the PL intensity associated with ''normal'' epitaxial surfaces, it is not surprising that the issue is of paramount importance in the case of free-standing layers possessing two exposed surfaces (particularly when layer thicknesses are reduced).

High resolution (0.02 cm^{-1}) PL results were obtained using a BOMEM DA8 Fourier transform interferometer in conjunction with a cooled Si avalanche photodiode detector and Ar^+ laser excitation. In transmission studies a high intensity tungsten lamp was used in combination with a narrow

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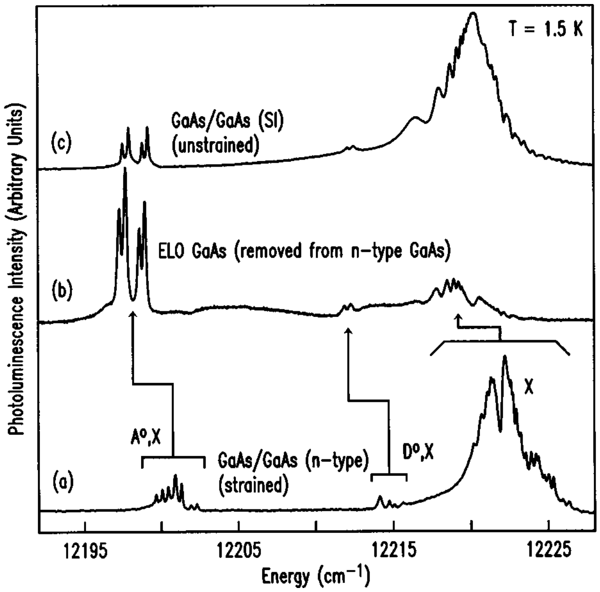

FIG. 1. PL signal obtained at 1.5 K from (a) a 2.3 μm GaAs epilayer grown on *n*-type GaAs, (b) the same epilayer following ELO, and (c) an unstrained, as-grown GaAs/GaAs (SI) heterostructure. The complicated series of sharp features associated with donor and acceptor bound excitons in (a) are greatly simplified in (b) once the substrate induced biaxial strain is removed via ELO.

band pass interference filter centered near the GaAs band gap. During optical investigations the samples were mounted in a strain free manner in a small vessel and held in the tail of a liquid He immersion cryostat. Care was taken to prevent damage to the fragile layers while He liquid was introduced to the dewar and pumped below its lambda point.

X-ray rocking curve data were collected using a Bede D^3 triple axis crystal x-ray diffractometer. Two Si channel-cut crystals were used to obtain a low divergence ($\sim$0.5 arcsec) x-ray beam. A third Si channel-cut crystal was placed in front of the detector to achieve higher resolution. (004) Bragg peaks with full widths at half maximum (FWHM) of as low as 8 arcsec from typical low dislocation SI GaAs substrates have been obtained using this arrangement. Computer simulations of the x-ray rocking curve data were carried out using the commercial software package ‘‘RADS’’ supplied by Bede Scientific.

III. RESULTS AND DISCUSSION

A. Photoluminescence results

Figure 1 presents high resolution (0.02 cm^{-1}), low temperature (1.5 K) PL data from Figs. 1(a) and 1(c) as-grown and Fig. 1(b) post-ELO GaAs epilayers. The bottom spectrum, due to a 2.3 μm epilayer grown on *n*-type GaAs, displays clear evidence of as-grown, substrate-induced biaxial strain. Three groups of features owing to the radiative recombination of acceptor bound excitons (A^0,X), donor bound excitons (D^0,X), and free excitons or polaritons (X) are easily identified. In particular, at least seven sharp ($\leqslant$0.15 cm^{-1}) peaks are clear in the A^0,X region of the trace and another four are apparent in the D^0,X region. The origin of these components is well understood and related to the presence of biaxial strain due to the small difference in lattice constant between the pure epilayer and the doped substrate material (for details see Ref. 11).

The polariton section of the spectrum is also rich in sharp structure. The ‘‘ripple’’-like features superimposed on the relatively broad underlying component are due to the interference of polaritons which coherently propagate across the thickness of the film. Observance of these effects are an indication of high quality epitaxial material and have been discussed elsewhere.[12]

The spectrum of the same epilayer following ELO is shown in Fig. 1(b). While three groups of features still persist in the ELO result, the relative intensities of the various components have changed quite noticeably from that of the pre-liftoff spectrum. The higher energy luminescence associated with the polariton, and to a lesser degree with D^0,X, are reduced in intensity compared to the A^0,X features. This behavior is likely the result of a lower effective excitation level, brought about by changes to the epitaxial surfaces encountered during ELO. The interference effects are still evident in Fig. 1(b) indicating that the polariton mean free path has not changed substantially as a result of the liftoff procedure.

The other noticeable difference between the PL spectra in Figs. 1(a) and 1(b) is the simplification of the fine structure associated with bound excitons in the latter. The biaxial substrate-induced strain present in the epilayer of the as-grown structure in Fig. 1(a) is relaxed upon liftoff, effectively removing the stress-induced splitting and hence lowering the number of spectral components recorded in Fig. 1(b). It is noted that a broad (FWHM $\sim$10 cm^{-1}) PL background underlying the sharp bound exciton structure in both the pre- and post-ELO spectra was theoretically subtracted to permit easier comparison of the traces. The energy, spectral width, and intensity of this feature were highly dependent upon both excitation density and sample temperature and believed to be related to charging effects at the GaAs/$\mathrm{Al}_{0.2}\mathrm{Ga}_{0.8}\mathrm{As}$/GaAs interfaces.

The remaining spectrum in Fig. 1 is due to an unstrained, as-grown, 2–3 μm epilayer deposited on (SI) GaAs. While the relative intensities of the excitonic features in this result are in good agreement with those of the strained as-grown sample shown in Fig. 1(a), the fine structure associated with the bound excitons is very similar to that found in the ELO spectrum of Fig. 1(b). This supports the earlier notion that the former effect is correlated with surface changes sustained during the liftoff process and also that it is possible to produce, and hence examine, free standing, essentially unstrained layers using this approach.

B. Optical transmission results

The real strength of ELO spectroscopy is exemplified in Fig. 2 where the results of optical transmission studies performed at 1.5 K on thin, strain-free ELO samples are presented. These measurements, which are altogether impossible to perform on as-grown heterostructures due to highly absorbing SI or strain-inducing doped substrates, unveil an impressive wealth of detail. The spectra at the top and bot-

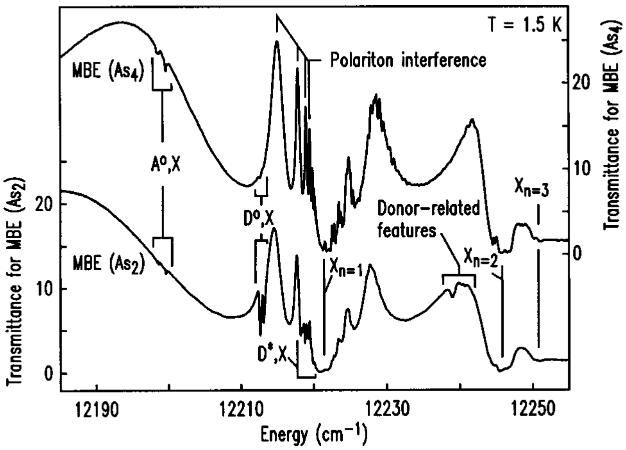

FIG. 2. Transmission spectra obtained at 1.5 K from free-standing GaAs films fabricated using ELO and grown using AS_2 (bottom) and As_4 (top) sources. Transmission minima associated with the $X_{n=1,2,3}$ energies are indicated. Numerous sharp absorption features due to donor and acceptor bound excitons are evident in both films. The donor concentration in the As_2 film is believed to be $\sim 10\times$ higher than in the As_4 layer resulting in stronger D^0, X features and heavily damped polariton interference effects.

tom of Fig. 2 correspond to films grown with As_4 and As_2 sources, respectively. On the low energy side of Fig. 2, series of sharp and discrete lines associated with donor and acceptor bound excitons are evident. Transmission through the D^0,X region of the sample grown with As_2 is heavily attenuated suggesting that the donor concentration (N_d) in this material is considerably higher ($\sim 10\times$) than in the As_4 film. This source dependent N_d has been observed before, and appears strongly correlated with the observance (or lack of in the case of As_2 grown material) polariton-related interference effects in the reflectivity spectra of high purity GaAs epilayers.[12] This observation is also consistent with the large polariton neutral donor scattering cross section observed previously.[13]

The same behavior is also evident in the transmission spectra of Fig. 2. While the As_4 result shows a large number of interference fringes on either side of the $X_{n=1}$ polariton resonance (though only four are denoted), the amplitude of the oscillations in the As_2 spectrum are considerably dampened. Subsequent transmission minima associated with higher energy (i.e., $X_{n=2}$ and $X_{n=3}$) polariton states are readily observable in both traces.

Two additional groups of components marked ''D^*,X'' and ''donor-related features'' are evident only in the As_2 spectrum and as such, are also related to the elevated N_d value in this material. The former, a series of sharp lines due to excited donor bound excitons, have been investigated in some detail in earlier PL studies.[14] The origin of the latter high energy features are not presently understood, though it is believed that they may be the result of resonant ''bound'' exciton levels below the $X_{n=2}$ polariton electronic excited state. While it is beyond the scope of this article to probe further into the nature of the diverse phenomena presented herein, it is clear that the transmission spectra of these unique spectroscopic cavities provide considerable insight into the properties of these thin film materials.

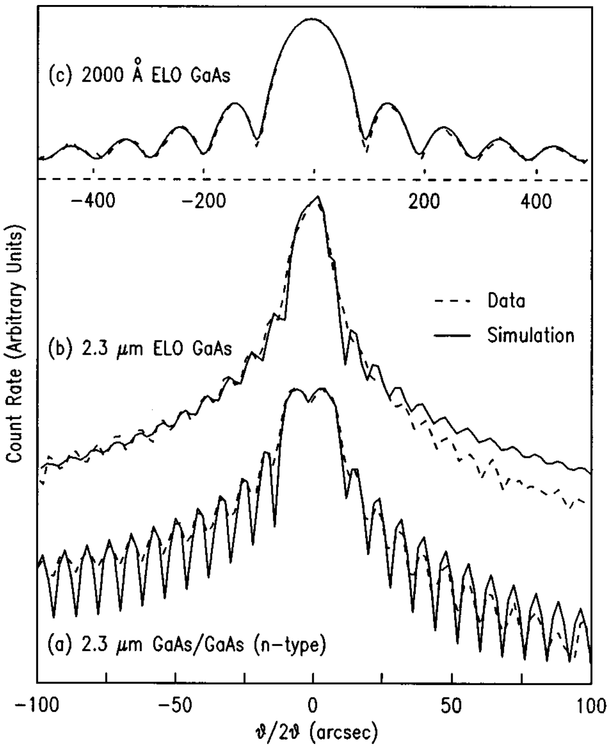

FIG. 3. X-ray diffraction spectra obtained from (a) an as-grown 2.3 μm GaAs/250 Å AlAs/GaAs (n-type) heterostructure, (b) the same heterostructure after removal of the AlAs layer and bonding to the original substrate, and (c) a 2000 Å ELO layer attached only to liftoff wax. Experimental and theoretical results are represented by the dotted and solid lines, respectively.

C. X-ray results

X-ray diffraction studies have also been used to reveal a great deal regarding the quality of epitaxial semiconductor layers (see, e.g., Ref. 15). The character of crystals can be inferred, to some degree, by the shape of the Bragg peaks and Pendellösung fringes in their rocking curve data. The Pendellösung or thickness fringes are ordinarily much weaker than the main Bragg peak and highly sensitive to the thickness uniformity and flatness of the film. These fringes (particularly the higher order ones) are generally difficult to observe in thicker (>1 μm) as-grown epitaxial layers and are altogether absent in crystals of inferior quality.

The x-ray rocking curve data for (a) a 2.3 μm epilayer grown on n-type GaAs, (b) the same epitaxial layer following liftoff and subsequent bonding to its original substrate, and (c) an ELO layer attached only to the liftoff compound, are shown by the dashed lines in Fig. 3. All of the experimental spectra clearly exhibit well defined Pendellösung fringes, in good agreement with those generated by dynamical theory simulations (solid lines). This is interpreted as strong evidence that the crystal quality of the epilayers remains intact throughout the entire ELO process. Upon comparing the results in Figs. 3(a) and 3(b) it is evident that the two well-resolved Bragg peaks in Fig. 3(a), due to the epitaxial layer and the substrate, merge closer together in Fig. 3(b) as the biaxial strain is reduced through the liftoff/bonding procedure. The agreement between the experimental and calculated results are somewhat less satisfactory for this film, presumably since the simulation neglects the van der

Waals forces between the epitaxial layer and the substrate.

The top spectrum, Fig. 3(c), due to the 2000 Å film supported only by the liftoff wax is intriguing in its own right. The x-ray curve for this sample shows only a single Bragg peak since the layer is entirely removed from the substrate material. The period of the Pendellösung fringes in Fig. 3(c) is much larger than that of the films shown in Figs. 3(a) and 3(b) since the layer thickness is approximately 10× less than for the other two films. In addition to being what is believed the first result of its kind, this spectrum demonstrates the fact that the structural properties of these thin films are not adversely affected during ELO and that they remain highly uniform following the procedure. While the single layer analyzed in Fig. 3(c) is quite clearly the simplest of structures, it follows that the ELO approach to thin film x-ray sample preparation could easily be extended to more complicated heterostructures where the addition of a substrate could potentially introduce unwanted strain and/or diffraction signal.

IV. CONCLUSIONS

In conclusion, thin free-standing layers of GaAs prepared by ELO have been studied using high resolution PL, transmission, and x-ray diffraction measurements. Initial results indicate that the films suffer no appreciable degradation as a result of the liftoff process. Once removed from their underlying substrates, these thin semiconductor cavities constitute unique systems for a variety of novel spectroscopic studies not possible in as-grown heterostructures. In particular, free-standing ELO films may prove altogether invaluable in studies where substrates introduce complications due to their influence upon epitaxial layers (i.e., via strain), their own optical or electrical properties (i.e., conductivity or opacity) or when they act as sources of unwanted background signals. In any case, it seems certain that these novel spectroscopic systems will find many uses in the study of materials science in the near future.

ACKNOWLEDGMENT

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