Epitaxial lift-off of ZnSe based II–VI structures

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The epitaxial lift-off technique is applied to II–VI based structures. Epilayers of 255 nm thickness containing quantum wells are lifted off their substrates and redeposited onto polyimide coated GaAs. The technique has also been applied to II–VI samples onto which dielectric films had been deposited. Photoluminescence measurements show that the material quality has not been degraded during the processing. The success of this technique with II–VI's opens up many possibilities for the integration of these materials with metals and dielectrics in vertical structure devices. © *1995 American Institute of Physics.*

Epitaxial lift-off (ELO) technology has been developed since 1987 in response to the need for integration of different semiconductor materials or structures which are difficult to grow monolithically.[1,2] This technique has achieved considerable success with GaAs based materials in which successful transplantation of light emitting diode (LED),[3] laser diode,[4,5] photodetector,[6,7] and integrated circuit devices[8] onto Si, InP, and other substrates without degradation of the device characteristics has been achieved. In this letter we outline the first application of the ELO technique to II–VI semiconductors: a ZnSe based quantum well structure is lifted-off and deposited on a new host substrate. The success of the process is evident in the preservation of the optical quality of the materials during the processing. The compatibility of such transplantation with these II–VI materials opens up the possibility of incorporating II–VI materials with multilayer dielectric stacks, metal films, and III–V semiconductors, in various vertical structure devices such as vertical cavity lasers, microcavity structures, or visible light modulators. In this way the difficulties associated with fabricating II–VI mirrors can be overcome.

Figure 1 is a schematic of the structure of the samples used in this work. A 1 μm thick GaAs buffer layer is grown by gas source MBE on a (001) GaAs substrate, followed by a 500 Å thick AlAs layer and a further 800 Å of GaAs. The AlAs layer is the sacrificial layer in the ELO process. After III–V growth the sample is coated with As_4 and transferred, out of vacuum, to the II–VI MBE chamber, where the As_4 layer is removed by heating the substrate before II–VI growth commences. ZnSe and ZnSSe buffer layers precede a CdZnSe/ZnSSe three quantum well region. Double crystal x-ray diffraction is used to calibrate the S concentration in order to achieve lattice matching to the substrate. Both doped and undoped structures were grown.

The ELO process is based on the release of layer structures from their growth substrates by the undercutting of a sacrificial layer. Traditionally the process is used for layers of the $Al_xGa_{1-x}As$ system, using a hydrofluoric acid (HF) based etchant and an AlAs layer as a sacrificial layer. In our experiments it has been observed that the II–VI layers of interest (ZnSe, ZnSSe, and CdZnSe) can easily withstand the HF etchant, allowing the standard ELO procedure developed for GaAs based materials to be applied. Chips approximately 1.5×2 mm^2 in size, covered with a $\approx$300 μm thick protective wax layer, are defined by cleaving. The sacrificial AlAs layer is undercut in HF: de-ionized water (1:5) at room temperature. After the etchant is diluted in de-ionized water, the wax-covered ELO films are picked up gently by means of a vacuum pencil and transplanted to the polyimide coated host substrates on which they are van der Waals bonded by a slight pressure. To avoid dust particles getting trapped between the film and host substrate, the entire transplantation process has to be performed under water. After drying of the ELO films at room temperature for approximately one day, the wax layer is removed in trichloroethylene. To enhance the adhesion of the van der Waals bonded films a 1 h bake-out step at 150 °C is performed. A more detailed description of this procedure can be found in previous publications.[2]

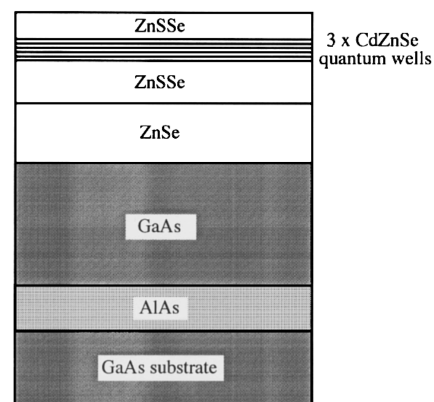

FIG. 1. Schematic of samples used in this work. The AlAs layer is the sacrificial layer in the ELO process.

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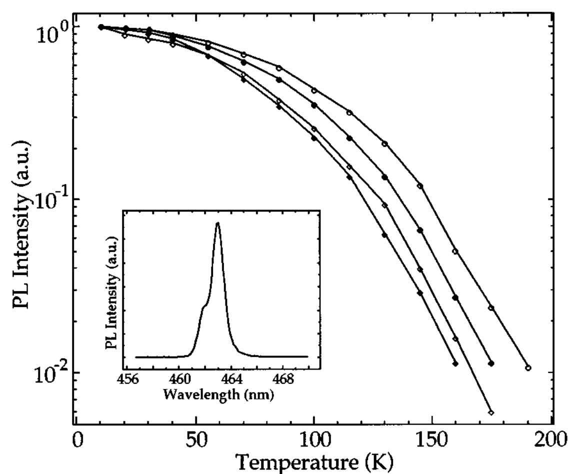

FIG. 2. Temperature dependence of the integrated PL intensity for pump powers of 30 μW (♦), 150 μW (◇), 300 μW (○), and 1 mW (●). Inset shows PL spectrum at T=10 K from an unprocessed sample.

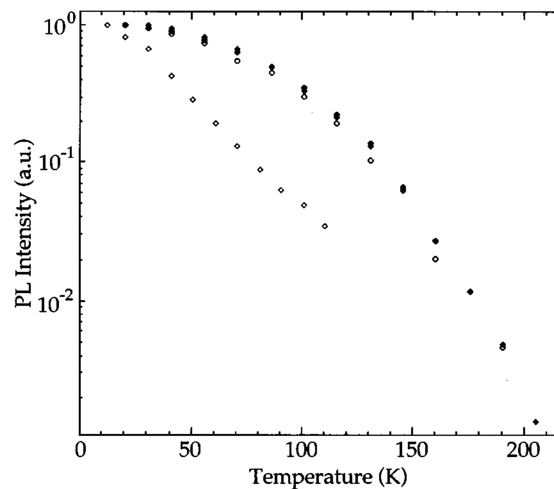

FIG. 3. Temprature dependence of the integrated PL intensity in unprocessed (●), ELO processed (○), dielectric coated and processed (♦), and bulk (◇) sample.

Following optical characterization (described below) of these materials which show that the optical quality has been preserved during processing, the same process was applied to sections of the sample onto which dielectric films had been deposited.

A dielectric multilayer MgF_2(180 nm)/HfO_2(220 nm)/MgF_2(180 nm) was selectively deposited by e-beam evaporation. The pattern consisted of 400×600 μm^2 rectangles, each ELO film, 1.5×1.9 mm^2 in size, carrying one rectangle. Note that these dielectric coatings were not designed to be mirrors but were used as a test structure to see whether or not the ELO process would work with coated samples. In all cases the host substrates were GaAs substrates spin-coated with approximately 300 nm of polyimide. ELO of both the coated and the uncoated II–VI films resulted in bonded films with good morphology.

Intermediate steps of the processing described above involves handling of delicate structures and the impact of such processing on the microscopic material quality needs to be characterized. One way to measure the material quality is by examination of the photoluminescence (PL).[9] In general good quality samples have stronger PL signals.

The inset in Fig. 2 shows a PL spectrum at T=10 K from a sample before ELO processing. The PL was excited with the 355 nm line of an Ar-ion laser, which excites the buffer layers in addition to the quantum wells with subsequent relaxation of the carriers into the well resulting in very strong PL signals. There is a double peak structure in the PL with the two peaks separated typically by ≈5 meV and the relative magnitude of the two lines varying from point to point on the sample. This structure appears in all the samples and is likely related to fluctuations in the plane of the quantum wells. The double peak structure disappears as the temperature is raised to 50 K with only the higher energy peak remaining. Exciton linewidths are ≈5.5 meV (FWHM) due to alloy broadening and well thickness fluctuations. The shape and energy of the PL from the ELO processed sample, and the sample with the dielectric coating is the same as that from the sample before processing. Exact comparison of the PL intensities of these different samples is difficult since the PL intensity varies by up to 20% from point to point even on a given sample. It is clear however that there is no dramatic drop in the PL efficiency of the materials after ELO processing, indicating a preservation of material quality. Other samples which were doped n type show defect luminescence as well as the blue exciton emission, and again the spectra from these samples are the same before and after processing.

As the temperature is raised nonradiative processes become more important since the increased mobility of the excitons means that more nonradiative centers, which may capture the exciton, are encountered. This leads to a drop in the PL efficiency as the temperature increases. The temperature at which the PL efficiency starts to drop off can be taken as some measure of the same quality.

Figure 2 shows the integrated PL intensity of the unprocessed sample as a function of temperature for constant pump intensities in the range 30 μW–1 mW. It can be seen on this figure that the fall off of the PL occurs at higher temperatures as the pump intensity is increased. This is due to greater saturation of nonradiative centers as the pump intensity increases. Room temperature luminescence can be observed on pumping with larger intensities.

Figure 3 shows the temperature dependence of the PL efficiency in the unprocessed, processed, and dielectric coated samples, for the same pump intensity. The PL intensity at the lowest temperature is normalized so that the various samples may be compared exactly, independent of the variations which occur on the wafer. There is no noticeable difference in the manner in which the PL efficiency falls off in the three samples. Again this indicates that there has been no measurable increase of the nonradiative defect density in the materials due to the ELO processing. For comparison the temperature dependence of the PL efficiency of a bulk ZnSSe sample is also shown on Fig. 3 and this can be seen to fall off

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much more rapidly with increasing temperature than in the case of quantum wells. This is expected due to the relative ease of encountering nonradiative centers in bulk material because of lack of confinement.

In conclusion, the epitaxial lift-off technique has been used to remove ZnSe based II–VI epilayers containing quantum wells from their III–V substrates and redeposit them onto polyimide coated GaAs substrates. Photoluminescence characterization of the materials before and after transplantation shows that there is no noticeable degradation of the microscopic material quality. Transplantation of II–VI films onto which dielectric layer structures had been deposited has also been successful using the same technique.

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