High-quality GaAs on Si substrate by the epitaxial lift-off technique using SeS$_2$

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In this letter, we demonstrate the realization of strong bonding between GaAs epilayers on Si substrates by using selenium sulphide (SeS$_2$) compound. After bonding, the sample has been transplanted to Si substrate using the epitaxial lift-off process. Such a transplanted film was found to be very smooth and adhered well to Si. The resulting chemical bond was covalent in nature, robust, and withstood clean room processing steps. The film bonded in this manner exhibited very good photoluminescence and high crystal quality by double crystal x-ray diffraction. The double crystal x-ray diffraction had a low full width at half maximum of 44 arcsec, and the strain was absent in these types of heterostructures. The interfacial chemical reaction and bonding were studied by depth profile x-ray photoelectron spectroscopy. It was concluded that Ga–Se and Si–S phases such as Ga$_2$Se$_3$ and SiS$_2$ were responsible for the strong bonding between GaAs and Si. © *1999 American Institute of Physics.* [S0003-6951(99)01150-X]

There has been a growing interest in the integration of semiconductor devices with silicon electronic circuits for quite a number of applications. Though silicon possesses very attractive properties for electronic devices, its indirect band gap limits the desirable optical properties. On the contrary, GaAs, InP, and their alloys exhibit outstanding optical properties and enjoy a matured fabrication technology in the electronic circuits. Thus, the monolithic integration of III–V's on Si substrates seems to be the ideal approach. The investigations had been mainly carried out by epitaxially growing III–V layers on Si substrate, but the crystalline quality of the layers was degraded by the high density of threading dislocations generated due to the large material mismatches.[1,2] A superior method to reduce the problems in III–V layers is yet to be established.

The direct wafer bonding technique[3] has received tremendous attention because of its potential to overcome the problems with epitaxial growth. Investigations on this technique, have lead to the result that material-mismatched wafers can be successfully sandwiched without degrading the crystalline quality.[4–6] The results had been especially satisfactory and promising when InP was bonded to GaAs.[7] III–V layers bonded to Si substrates have also been reported,[8] however, the employment of high-pressure, high-temperature, ambient gas, and crystallographic alignment was very much essential.

Recently the idea of lattice engineered compliant substrates has been proposed in which on a universal substrate (of Si or GaAs), any III–V or related semiconductor compound could be grown without dislocations.[9]

In this letter, we investigate a process in contrast with the conventional wafer bonding technique. In this approach, GaAs thin films were bonded to Si by SeS$_2$ at low temperature and low pressure and the latter was lifted off from lattice-matched GaAs growth substrate by means of an ultrathin AlAs release layer.

We used metal-organic chemical vapor deposition (MOCVD) to grow GaAs homoepitaxy on silicon-insulator (SI) GaAs substrate. Initial growth was proceeded by growing a GaAs buffer layer of ~20 nm thickness at 750 °C, at a growth rate of 8.3 Å/s. A release layer of AlAs was grown on the buffer layer at a growth rate of 8.8 Å/s. The release layer thickness was around 10 nm. Following this growth, a 3 mm GaAs layer was grown which was later bonded to the Si substrate using a bonding assisting layer of SeS$_2$ in the following way.

The epitaxially grown layer was treated in SeS$_2$ solution dissolved in CS$_2$ for 5 min. A thin orange film was visible on the GaAs surface. The substrate was taken out and dried in N$_2$ gas, and then sandwiched with the Si substrate. The sandwiched specimen was transferred to a furnace set at 350 °C under N$_2$ ambient and kept for 10 min without applying pressure on the wafer after which it was cooled down to room temperature. The specimen removed from the furnace exhibits an intimate contact between the GaAs epilayer and Si substrate. The sample was then immersed in HF:H$_2$O (1:2) for 24 h to undergo the epitaxial lift-off (ELO) process. After removing the GaAs substrate and the AlAs layer by this selective chemical etch, a 3 μm layer of GaAs was left bonded to the Si substrate.

The photoluminescence measurement was recorded at 77 K using a 514.5 nm Ar-ion laser as an excitation source and

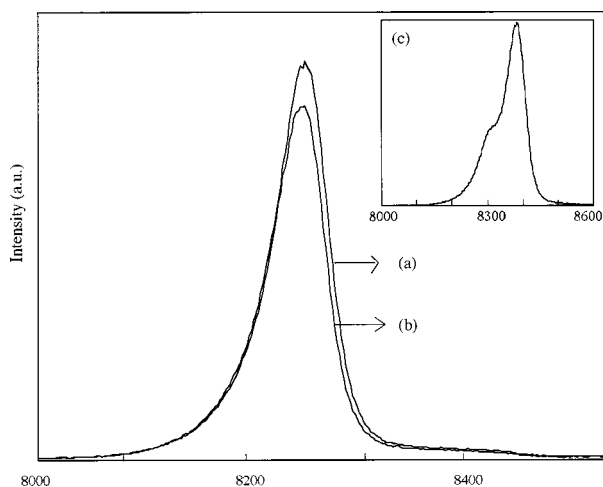---

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FIG. 1. Photoluminescence spectra at 77 K for (a) GaAs bonded to Si, (b) GaAs epilayer grown on Si, and (c) GaAs grown on GaAs.

FIG. 2. Double crystal x-ray diffraction spectrum of the bonded sample.

GaAs photomultiplier tube as a detector. The x-ray rocking curve was recorded for the (400) reflection using the Cu $K\alpha$ radiation by Rigaku double-crystal x-ray diffractometer. The photoemission experiments were carried out using SSX-100 (Surface Science instruments).

In contrast to many other techniques, the advantage of this method is that it is a low temperature process, and more importantly, does not involve precise crystallographic alignment of Si and GaAs. We were able to obtain strong bonding between Si and GaAs for any arbitrary alignment of the two crystal orientations. These are the important parameters to be considered for the eventual integration of GaAs devices on Si substrates. The bonding strength was so high that the bonded structure could be cleaved right after the bonding, before removing the SI GaAs substrate, without causing any detachment.

The photoluminescence spectra of the bonded sample recorded at 77 K are shown in Fig. 1. The spectra consist of (a) GaAs bonded to Si by epitaxial lift off using SeS_2, (b) GaAs epilayer grown on GaAs substrate, and (c) GaAs epilayer grown on Si (shown as insert in the figure). The shapes and peak wavelengths of the two spectra *a* and *b* were nearly identical without degradation of the bonded film. This clearly indicates that the photoluminescence (PL) peak shift due to stress, which was observed in the heteroepitaxial GaAs on Si (spectrum *c*) was not observed in the ELO film obtained by this bonding process. The crystalline quality itself does not seem to be degraded, suggesting the effectiveness of this bonding method. Further from the results of the double-crystal x-ray diffraction, it is evident that the full width at half maximum (FWHM) of 44 arcsec was the lowest obtained so far as shown in Fig. 2.

The mechanism of bonding is proposed as follows. After the desorption of native oxide, the surface of GaAs, treated with SeS_2, becomes strongly reactive at 350 °C. To reduce the surface energy, surface reconstruction takes place. However, when the two surfaces are close enough, new chemical bonds are formed during the cooling process since the energy is lower than that for the separately reconstructed surfaces. Since only the surface atoms in each material participate in the reaction, the rest of the material is not affected by the bonding process. In order to further study the interface chem-
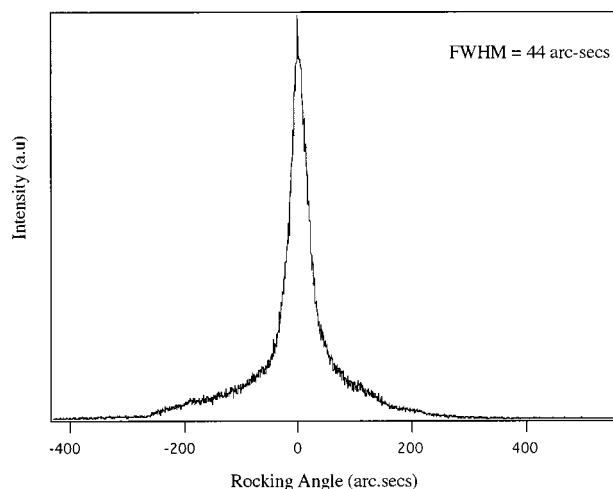
istry and have more clear insight of the bonding process depth profile photoemission spectra of Ga $3d$, As $3d$, Se $3d$, S $2p$, and Si $2p$ core levels were recorded individually with high instrumental resolution.

The Ga $3d$ spectrum is shown in Fig. 3(a). In addition to the main Ga–As peak, two peaks at higher energy levels of 0.8 and 1.4 eV were observed. The data presented later for S $2p$ spectra clearly indicated that no Ga–S based peaks were observed. These peaks are ascribed to Ga–Se bonds and labeled as Ga–Se (1) and Ga–Se (2) respectively. The Ga–Se (2) bonds are assumed to be Ga_2Se_3 like and Ga–Se (1) bonds to be $GaSe_x$ ($x > 1.5$) like bonds.[10]

From the As $3d$ spectrum, Fig. 3(b), as the depth profile was performed, in addition to the main As–Ga peak two more peaks were observed. The first peak which has a chemical shift from the Ga–As based peak of 2.5 eV is assigned to As–S (1) and the second peak with a chemical shift of 0.8 eV to As–Se. The chemical shifts of these peaks with respect to the As–Ga peak were very close to the previously reported values.[11]

In Se $3d$ photoemission spectrum, Fig. 3(c), only one peak was observed with a binding energy of 55.8 eV. No peak relevant to As–Se was seen. The amount of charge transfer from Ga to Se was considered to be larger than that from As to Se since the electronnegativity difference between Ga and Se is larger than that between As and Se. Therefore, this peak corresponds to Se–Ga bonds. It has been reported in the literature that at 350 °C the exchange of As to Se increases until complete exchange takes place.[12] The binding energy for this peak is in close agreement with the previously reported values.[11]

Figure 3(d) shows the core level photoemission spectrum of Si $2p$. Three peaks were observed in addition to the main Si peak, located at higher binding energy. These peaks are attributed to Si–S based bonds and are indicative of the formation of the compound SiS_2.[13]

The S $2p$ photoemission spectrum for the bonded sample shown in Fig. 3(e) consists of two peaks. The peak with the lower binding energy is assigned to S–As and the higher energy peak to S–Se.

The atomic concentration ratios of different elements changed with respect to depth as shown in Fig. 4. The progressive diffusion of S and Se atoms as a function of depth
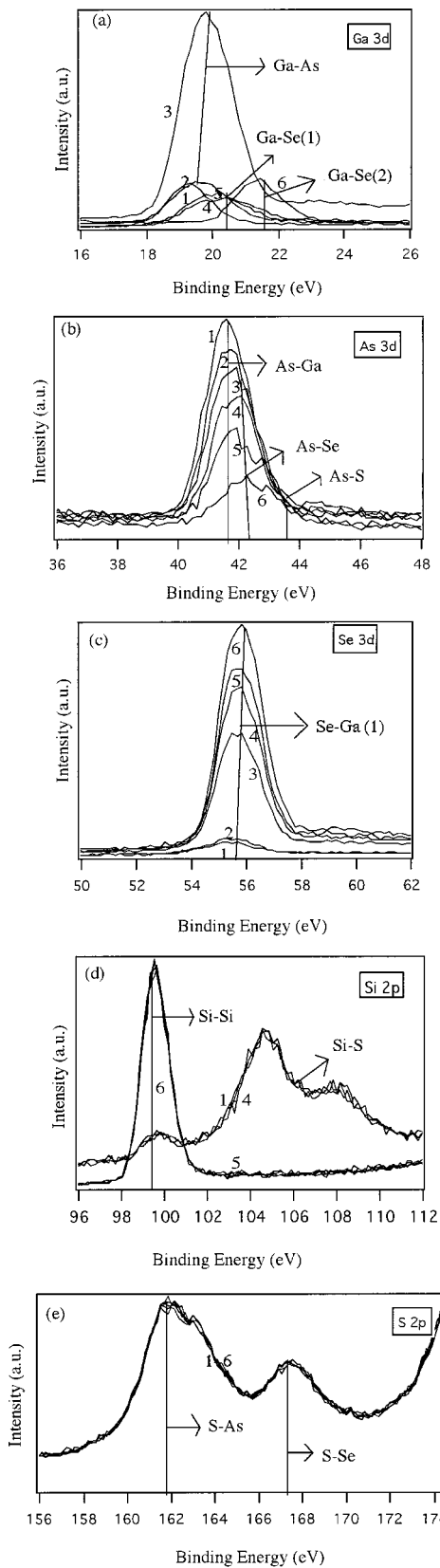
FIG. 3. XPS core level spectra of (a) Ga $3d$, (b) As $3d$, (c) Se $3d$, (d) Si $2p$, and (e) S $2p$.
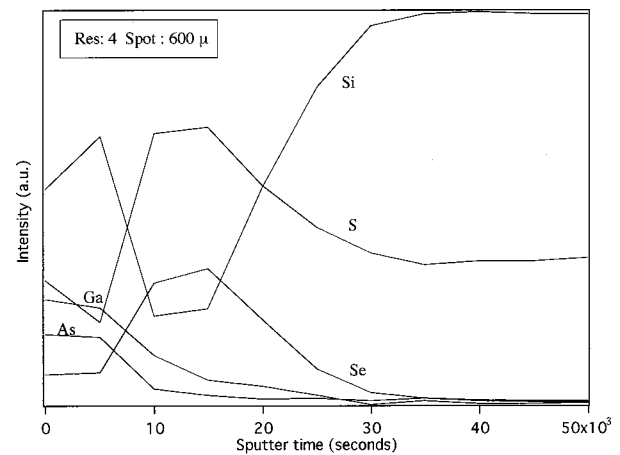

FIG. 4. Depth profile of different elements as a function of time.

and the topmost region is made up of larger part of S atoms bound to Se atoms [Fig. 3(e)]. Thus in this case, a S atom forms a bridge bond between Se and Si atoms. Then Si is bonded to S in the form of SiS_2. These results reveal a rich surface-derived substructure that is due to changes in the near-interface chemical bonding environment. Thus our results show that by this chemical treatment a very robust and permanent chemical bonding between GaAs and Si can be obtained.

In conclusion, we have demonstrated by this bonding process that GaAs thin films can be bonded to Si substrates without degrading the crystalline quality of GaAs epilayer. No shift in PL peak wavelength and low full width at half maximum of 44 arcsec provide persuasive evidence for the good quality of the epilayers. By XPS the mechanism of bonding has been speculated. The bonding achieved by this means it proves to be robust and could overcome the problems in heteroepitaxy growth.

was estimated from the intensity of the Se $3d$ and S $2p$ core-level peaks. The atomic concentration study indicates that the interface consists of a chemically stratified structure. It was quite evident that the immediate surface of GaAs is primarily of Ga based selenides. The next uppermost region is made up of As atoms bonded to S and Se atoms [Fig. 3(b)]

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