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Band alignment studies in InN/p-Si(100) heterojunctions by x-ray photoelectron spectroscopy

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The band offsets in InN/p-Si heterojunctions are determined by high resolution x-ray photoemission spectroscopy. The valence band of InN is found to be 1.39 eV below that of Si. Given the bandgap of 0.7 eV for InN, a type-III heterojunction with a conduction band offset of 1.81 eV was found. Agreement between the simulated and experimental data obtained from the heterojunction spectra was found to be excellent, establishing that the method of determination was accurate. The charge neutrality level (CNL) model provided a reasonable description of the band alignment of the InN/p-Si interface and a change in the interface dipole by 0.06 eV was observed for InN/p-Si interface. © *2011 American Institute of Physics*. [doi:10.1063/1.3596520]

INTRODUCTION

Indium nitride (InN) has attracted extensive studies for its distinctive characteristics, such as small effective mass,[1] large electron drift velocities,[2] surface electron accumulation layer,[3] and its controversial band gap.[4,5] These unique properties prompted serious scientific questing for wide possibilities in device applications,[6] for example, in high speed high frequency electronics,[7] electron emitters, detectors,[8] solar cells,[9] etc. From the perspective of the devices, the Si substrates offer several advantages such as ease of cleaving, availability of conducting substrates in large size wafers at very low cost, and suitability in device processing, the growth of InN on Si substrates is of significant academic and commercial interest. Recently several studies have been reported on the growth of InN epilayers and nanostructures on Si (111) substrates.[10,11]

The band alignment at the interface of the semiconductor heterostructures is an important part of electronic and optoelectronic devices because it governs efficient carrier transfer across the heterostructure. However, to date there is a lack of experimental reports on the interface band alignment parameters of the InN/p-Si heterojunction system. Recently our report[12] on the n-InN nanodot/p-Si heterostructures established the band offset values by capacitance- voltage measurements. However, no systematic study has been found on the determination of valence band and conduction band offsets of an InN/p-Si heterojunction with well accepted InN bandgap value of 0.65–0.8 eV by using x-ray photoelectron spectroscopy (XPS). We have used the method of charge neutrality levels (CNLs) to cross check the band offset values[13,14] which gave a reasonably good agreement with the experimental determinations.

EXPERIMENTAL

The InN thin films were grown on p-Si(100) substrates by nitrogen plasma assisted molecular beam epitaxy system (PAMBE). The general set of growth conditions including the beam equivalent pressure (BEP) of indium, substrate temperature, nitrogen flow rate, and RF-plasma power, were kept at 3×10^{-7} mbar, 410 °C, 0.5 sccm, and 350 W respectively. The duration of growth was 5 min for ~8 nm thick InN film. An InN film of thickness ~200 nm was also grown and the duration of growth was 2 h. To avoid the unintentional nitridation of the Si substrate, which results in a Si_xN_y layer, an ultrathin layer (~5 ML) of indium was deposited prior to the InN growth. It is known that the InN surface is susceptible to oxidation after exposure to air. To reduce the contamination effect, all the samples were subjected to a surface cleaning procedure by Ar+ bombardment in a vacuum chamber attached to the XPS instrument and reduced by a thickness of 3–4 nm, estimated by the sputtering time. The band offsets of the InN/p-Si heterojunction were estimated by the results of XPS, obtained for the conditions of a 200 nm InN film, cleaned Si substrate (using hydrofluoric acid (HF)) and 4 nm thin InN/p-Si heterojunction.

RESULTS AND DISCUSSION

The VBO (valence band offset) ($\Delta E\nu$) value can be evaluated by the energy difference between the In and Si core-levels (CLs) (ΔEcl) acquired from the InN/p-Si heterojunction sample and the CL energies of the respective valence band maximum (E_{VBM}) from the InN epilayer and Si substrates. All the CL spectra were fitted to Voigt (mixed Gaussian-Lorentzian) line shape by employing a Shirley background. The relation between $\Delta E\nu$ and photoelectron spectroscopy measurement results is given by,

$$\Delta E\nu = \Delta E\mathrm{cl} + \left(E^{\mathrm{InN}}_{\mathrm{VBM}} - E^{\mathrm{InN}}_{\mathrm{In3d}}\right) - \left(E^{\mathrm{Si}}_{\mathrm{VBM}} - E^{\mathrm{Si}}_{\mathrm{Si2p}}\right) \quad (1)$$

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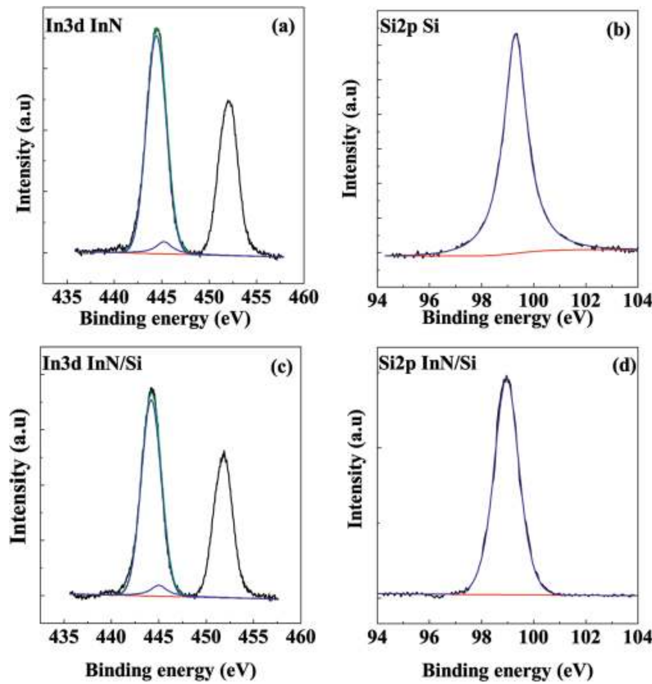

FIG. 1. (Color online) In3d XPS spectra for (a) InN and (c) InN/p-Si samples and Si 2*p* XPS spectra for (b) Si and (d) InN/p-Si samples. Experimental data points are fitted by Voigt (mixed Lorentzian-Gaussian) line shapes after the application of a Shirley background (BG).

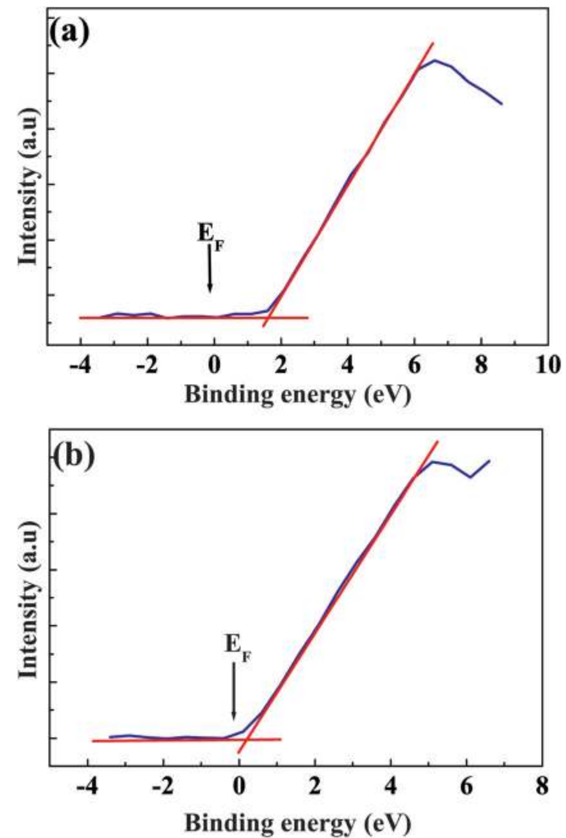

FIG. 2. (Color online) VBM spectra for (a) InN film and (b) HF cleaned Si substrates.

where ΔEcl denotes the difference in the energy between In3d5/2 and Si2p core level peaks in the InN/Si heterojunction. The In3d5/2 and Si2p XPS spectra of the samples are shown in Figs. 1(a)–1(d). Since considerable accordance of the fitted line to the original measured data has been obtained, the uncertainty of the CL position should be lower than 0.01 eV, as evaluated by numerous fittings with different parameters. The only In3d5/2 component located at 444.1 eV is attributed to the In-N[15] bond along with the extremely low intense oxygen related peak of In-O bond. $(E_{\rm VBM}^{\rm InN} - E_{\rm In3d}^{\rm InN})$ is the bulk constant which was obtained from the 200 nm thick InN film and $(E_{\rm VBM}^{\rm Si} - E_{\rm Si2p}^{\rm Si})$ was obtained from the HF cleaned Si substrate. The valence band maximum (VBM) positions in the VB spectra were determined by linear extrapolation of the leading edges of valence band (VB) spectra to the base lines in order to account for the instrument resolution induced tail.[16] Figure 2 shows the XPS VB spectra of InN epilayer and Si substrates. VBM values of 0.2 eV and 1.6 eV were deduced from the VB spectra for Si and InN respectively, by the linear fitting procedure depicted above. The obtained CL and VBM parameters are summarized in Table I. The CL and VBM positions of InN as well as Si are comparable to the values reported in Refs. 15 and 17.

By comparing the spectra recorded on the InN and Si samples, it was found that the In3d5/2 peak of In-N bond in InN/Si shifted $\sim$0.32 eV to a binding energy of 444.10 eV, and the Si2p CL of Si-Si bond is shifted to 98.99 eV. The VBO value is calculated to be 1.39 ± 0.01 eV by substituting those values obtained in experiments into Eq. (1).

As XPS measurements are spatially averaged due to the finite mean free path of elastic electrons, band bending could induce a systematic error in our measurements. The deviation in the band alignment occurs due to the band bending has not been considered in the present analysis. Another factor that can affect the measured value is the strain existing in the InN overlayer of the heterojunction, which will induce a piezoelectric field as explained by Martin *et al.*[18] The nitrides are piezoelectric materials, so the strain-induced static electric fields are observed via the piezoelectric effect. By using the constants and equation from the work of Martin *et al.*,[18] we estimated the field magnitude, which was in the order of 10^7 V/m. Assuming typical heterojunction InN overlayer thickness of $\sim$4 nm (after Ar^+ bombardment), the error of VBO induced by lattice mismatch is less than 10 meV. Furthermore, the strain in the epifilms mostly relaxes, which means the "residual" effect of piezoelectric field is greatly reduced due to the dense networks of threading defects extending from the substrates to the surfaces which is known for all nitride

TABLE I. The XPS CL spectra fitting results and VBM positions obtained by linear extrapolation of the leading edge to the extended base line of the VB spectra.

Samples	States	B.E (eV)	Bonding
InN	$In3d_{5/2}$	444.42	In-N
		445.46	In-O
	VBM	1.6	
Si	Si2p	99.3	Si-Si
	VBM	0.2	
InN/Si	$In3d_{5/2}$	444.1	In-N
	Si2p	98.99	Si-Si

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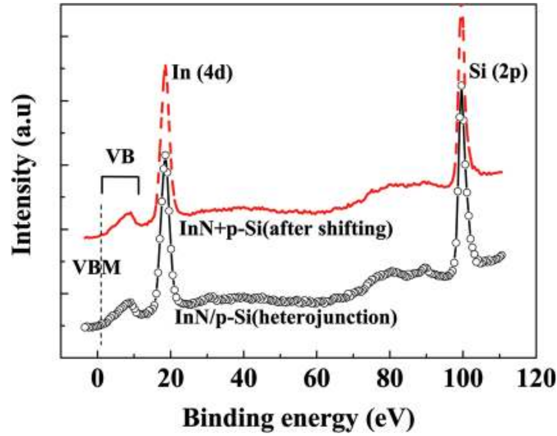

FIG. 3. (Color online) Experimental (circles) and simulated (solid line) photoelectron spectra for the InN/p- Si heterojunction sample. The simulated spectrum is obtained by combining the experimental photoemission spectra from InN and Si.

epitaxial layers.[18] In addition to this, the heterojunction underlayer Si is thick enough to be completely relaxed and the (0001)//(100) configuration for the interface of these two materials produces a lattice mismatch between the Si(100) and the InN(0001) by 8%. Such large mismatch results in few monolayers of critical thickness. Further, due to the small linear pressure coefficient of InN (~0.06 meV/GPa),[19] we could neglect the bandgap variation by the interface strain. Considering all these facts, the 4 nm thick InN grown on Si should be relaxed and, thus, the strain-induced piezoelectric field and the bandgap variation is not considered in the present work. The photoelectron spectrum contains information from the near surface region of the sample within the photoelectron probing depth. The photoelectron spectrum from InN/Si heterojunction may include signals from both InN film and Si. Hence, for a check of the method, we have simulated the valence band spectra of the heterojunctions by shifting and summing the appropriately weighted spectra for clean InN and Si. The clean surface spectra were shifted in energy, and the In 4d and Si 2p match with those of the heterojunction. Figure 3 shows the comparison of experimental photoelectron spectrum (circles) and simulated spectrum (solid line) of InN/Si heterojunction. The simulated spectrum is simply the weighted summation of InN and Si photoelectron spectra with a relative energy shift (between the valence band edges of InN and Si) using the estimated VBO value of 1.39 eV (without any piezoelectric field correction). The relative contribution from InN and Si were determined by the In 4d and Si 2p CL peak intensities in the InN/Si heterojunction photoelectron spectrum. An excellent agreement between the line shapes of experimental and simulated spectra confirms both the accuracy of the measured ΔEv value of InN/Si heterojunction and the strain free nature as well in the InN film.

Finally, the conduction band offset (ΔEc) was estimated by the relation: $\Delta Ec = \Delta Eg - \Delta Ev$, where ΔEg is $(Eg^{\mathrm{Si}} - Eg^{\mathrm{InN}})$. The bandgap values of 0.7 eV and 1.12 eV were considered for InN and Si, respectively. ΔEc was calculated to be 1.81 eV and, as a result, a type-III band alignment for the InN/p-Si heterojunction has been proposed, as shown in Fig. 4. The InN/p-Si band offsets of these results can be found

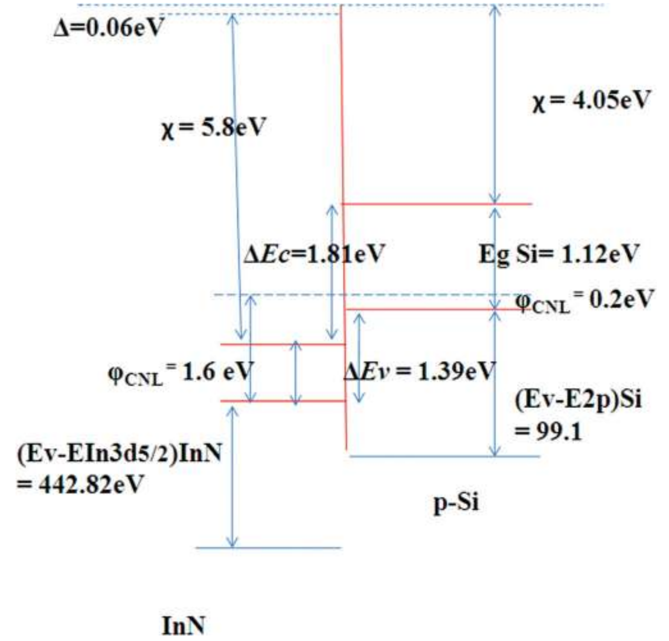

FIG. 4. (Color online) Schematic illustration of Type III band alignment of InN/p-Si. The top dashed line corresponds to the electron affinity of Si. The deviation in the band alignments from the electron affinity model is indicated by Δ.

significantly different from the band offset values, in particular for the CBO reported by Yoshimoto *et al.*,[20] who reported a type-II band alignment for InN/p-Si heterojunction. The factor that could play a major role in this difference arises from the reduced bandgap of InN. A higher bandgap value of 1.8–2 eV was considered for InN by Yoshimoto *et al.*

The existence of interface dipoles determines the band alignment between two semiconductors, just as in the case of Schottky barriers. According to the electron affinity model, CBO is given by the difference in the electron affinities in the limit of no charge transfer dipoles, equivalent to the Schottky limit. The charge transfer across the interfacial bonds creates a dipole, which modifies the band lineup given by the electron affinity rule. This model, i.e., Schottky barrier interface defect model, has been adapted to interpret the band alignment of InN/p-Si, which was originally presented by Cowley and Sze.[21] This model was used to analyze silicon/oxide and SiN/GaN interfaces by Robertson[13] and Cook[14] *et al*, respectively. The model also seems to be consistent with our experimental result, and is given by,[14]

$$\varphi_{\mathrm{CBO}} = (\varphi\mathrm{CNLInN}\ \text{-}\ \varphi\mathrm{CNLSi}\ ) - (\mathrm{Eg_{InN}} - \mathrm{Eg_{Si}}) + S\{(\chi_{\mathrm{InN}} - \chi_{\mathrm{Si}}) + (\mathrm{Eg_{InN}} - \mathrm{Eg_{Si}}) - (\varphi_{\mathrm{CNLInN}} - \varphi_{\mathrm{CNLSi}})\} \quad (2)$$

where φ_{CBO} is the CBO and χ and φ_{CNL} are the electron affinities and charge neutrality levels of each semiconductor (InN and Si), respectively. CNL is the branch point of the semiconductor surface (or interface) states since they are related to the valence or conduction band and also CNL is the balance point of the valence and conduction band density of states. Charge can transfer between the interface states of the two materials, causing an interface dipole. In other words, the branch point

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energy, which constitutes a charge neutrality level[22] for the semiconductor and is thought to be universal on an absolute energy scale, marks the energy where surface states change their character from predominantly donor type (below) to predominantly acceptor type (above). In general, the branch point determines the favorable charge state of all gap states, including surface states, metal and interface induced gap states, and defect induced gap states. Interface states exist in the present InN/Si interface. At the energy of charge neutrality, their dominant character changes from donor type to acceptor type. Charge transfer occurs across the interface, creating an interface dipole, which tends to align the InN Fermi-level toward the CNL of Si (pinning).

The S factor modified matching of the charge neutrality levels (CNLs) of each semiconductor would result in the band lineup. The parameter S in equation (2) is a dimensionless pinning factor of the wider-gap semiconductor, which describes whether the barrier is "pinned" or not. S varies between the limits $S = 1$ for unpinned Schottky barriers and $S = 0$ for Bardeen barriers which are pinned by a high density of interface states. S depends on the electronic part of the dielectric constant (ε_∞), empirically given by[13]

$$S = 1/\left(1 + 0.1(\varepsilon_\infty - 1)^2\right).$$

By substituting the factor $\varepsilon_\infty = 12$ for Si, an S value of 0.08 has been obtained, indicating significant (strong) pinning of the Fermi-level to the φ_{CNL} at the interface. For strong pinning, the alignment is just given by the alignment of two CNLs. The CNL energy below the vacuum level is a measure of the mean electronegativity of the semiconductor, in the similar way that the work function of a metal is proportional to the metal's electronegativity. Thus, equation (2) illustrates that the band alignment is the difference in electronegativity screened by the S factor.[13]

XPS was also employed to determine the pinning position of the Fermi level at the InN surface. The surface Fermi level position can be obtained by extrapolating the leading edge of the valence band photoemission to the baseline in order to take account of the finite resolution of the spectrometer. The values determined by this way are given in Table I. King *et al.*[22] suggested that the linear extrapolation method of analysis allows a direct experimental determination of the CNL position without referring to theoretical calculations or detailed knowledge of the bulk Fermi level in the samples. There are various reports on the value of CNL of InN, given as 1.83,[22] 1.6,[23] and 1.51 eV.[24] It was also reported that an increase in the doping shifts the leading edge of the valence band photoemissions to higher binding energies, indicating an increase in the VBM to Fermi level separation at the surface.[22] The XPS data presented above reveal the surface Fermi level at 1.6 eV. The branch point energy is therefore determined as lying at 1.6 eV above the VBM.

The estimation of CNL of Si has also been carried out by the above method. It depicts that the CNL is slightly above the valence band maximum, i.e., at 0.2 eV. The values of CNLs for Si and InN are in close agreement with the reports by Robertson *et al.*[13] and King *et al.*,[22] respectively. Pinning of the surface Fermi-level of InN to the CNL of Si also infers the pinning of the InN surface Fermi-level to the CNL of InN itself, resulting in the alignment of the CNLs of the both materials. The Fermi level pinning is expected to be caused by the charge distribution at the interface.[25] The interface state density (D*it*) in the bandgap of a semiconductor can roughly be estimated from the S factor by following the relation proposed by Cowley and Sze,[21]

$$Dit = 1.1 \times 10^{13} \times (1 - S)/S \text{ States cm}^{-2}\text{eV}^{-1}$$

From the above equation, D*it* for InN/Si interface was obtained to be $\sim 10^{14}$ states cm^{-2} eV^{-1}. When $S \rightarrow 0$, the Fermi level at the interface is pinned by the surface states at the energy value measured from the valence bandedge at the semiconductor surface and specifies the level below which all surface states must be filled for charge neutrality at the interface of Si. The considerable amount of interface states are attributed to broken bonds, strained bonds, mismatched bonds, or physically damaged layer formation with some interfacial variations.

Furthermore, the band offsets would be determined by the relative position of the CNLs of the two materials if the density of states is high or if the CNLs of the two materials are similar in relative energy.[14] By taking account of bandgap and CNL values of InN and Si followed by aligning the CNLs, one can deduce the CBO of 1.82 eV in the proposed band alignment diagram shown in Fig. 4. The conduction and valence band offsets based on CNL are compared with our experimental results. To our knowledge the value of S of InN/p-Si interface has not been reported so far. By substituting the CBO value obtained from the XPS results (i.e., 1.81 eV) in equation (2), one can obtain the S value of 0.1 ($\sim$0.08). In other words, by using the S value of Si i.e., 0.08 (S is the pinning parameter of wider-gap semiconductor),[26] the CBO of 1.814 eV can be obtained. Hence the experimental results are in agreement with the CNL model within the experimental measurement error.

A comparative interpretation of the experimentally derived (or predicted from the CNL model) band alignment with the band alignment obtained from the electron affinity model (EAM) has been carried out by constructing the band alignment diagram (shown in Fig. 4) as follows. Initially the diagram was aligned to the vacuum level at Si by the electron affinity of $\chi_{Si} = 4.05$ eV. The bandgap of each material is indicated. Then the CBs and VBs are aligned by substituting the CBO and VBO values obtained from the XPS results or CNL model. Since both the methods resulted in reasonable matching of band offset values, no significant difference was observed in the band alignments. Experimental results as well as the results from the CNL model show a deviation from the EAM of 0.06 eV. The EAM of heterojunction formation assumes that the vacuum levels would align at the interface so that conduction band minima of the materials would align with respect to their electron affinities ($\chi_{Si} = 4.05$ eV and $\chi_{InN} = 5.8$ eV). The basic assumption in the EAM is that the interface is formed without perturbation of the surface electronic states of either of the two materials. The measured difference between the prediction of the EAM and the experimentally observed band offset represents a change in the interface dipole.[14]

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CONCLUSIONS

The band offsets of InN/p-Si heterojunctions were estimated using XPS data. A type-III band alignment with a valence band offset of $\Delta Ev = 1.39$ eV and conduction band offset of $\Delta Ec = 1.81$ eV was identified. The reduced bandgap of InN (1.8–2 eV to 0.65–0.8 eV) leads to the variation of band offsets from the previous report on the experimental determination of band offsets. Agreement between the simulated and actual heterojunction spectra is excellent with regard to the positions and relative intensities of all features, establishing the accuracy of the method. The charge neutrality level model provided a reasonable description of the band alignment of the InN/p-Si interface. The interface dipole deduced by comparison with the electron affinity model was 0.06 eV.

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