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Electrical Characteristics of n-ZnO/p-Si Heterojunction Diodes Grown by Pulsed Laser Deposition at Different Oxygen Pressures

Authors: R.S. Ajimsha · M.K. Jayaraj · L.M. Kukreja

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	Heterojunction diodes of *n*-type ZnO/*p*-type silicon (100) were fabricated by pulsed laser deposition of ZnO films on *p*-Si substrates in oxygen ambient at different pressures. These heterojunctions were found to be rectifying with a maximum forward-to-reverse current ratio of about 1,000 in the applied voltage range of −5 V to +5 V. The turn-on voltage of the heterojunctions was found to depend on the ambient oxygen pressure during the growth of the ZnO film. The current density–voltage characteristics and the variation of the series resistance of the *n*-ZnO/*p*-Si heterojunctions were found to be in line with the Anderson model and Burstein-Moss (BM) shift.	
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Electrical Characteristics of *n*-ZnO/*p*-Si Heterojunction Diodes Grown by Pulsed Laser Deposition at Different Oxygen Pressures

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Heterojunction diodes of *n*-type ZnO/*p*-type silicon (100) were fabricated by pulsed laser deposition of ZnO films on *p*-Si substrates in oxygen ambient at different pressures. These heterojunctions were found to be rectifying with a maximum forward-to-reverse current ratio of about 1,000 in the applied voltage range of –5 V to +5 V. The turn-on voltage of the heterojunctions was found to depend on the ambient oxygen pressure during the growth of the ZnO film. The current density–voltage characteristics and the variation of the series resistance of the *n*-ZnO/*p*-Si heterojunctions were found to be in line with the Anderson model and Burstein-Moss (BM) shift.

Key words: Heterojunctions, ZnO, *p*-Si, pulsed laser deposition

INTRODUCTION

Currently there is significant interest in ZnO as a candidate for various future optoelectronic devices. ZnO is a rugged semiconductor with direct wide band gap and it exhibits significant *n*-type conductivity even without any intentional doping. This *n*-type conductivity can be further enhanced by doping with Al or Ga.[1–3] This property and the transparency in the visible spectral region have prompted extensive investigations of ZnO films as transparent electrodes in flat-panel displays,[4] *p*–*n* heterojunction diodes,[5–7] thin-film transistors,[8] multiple-quantum-well structures,[9] and solar cells.[10] Recently we have reported ZnO based all-transparent conducting *p*–*n* heterojunction diodes with *p*-type $AgCoO_2$.[11,12] Although ZnO films can be grown by a variety of methods, including radiofrequency (RF) and direct-current (DC) sputtering,[3,13,14] chemical vapor deposition,[15] spray pyrolysis,[16] and electron cyclotron resonance-assisted molecular-beam epitaxy,[17] we used pulsed laser deposition (PLD)[1,18,19] to deposit high-quality ZnO films because of its effectiveness and amenability to different growth conditions.[20] For the present study we fabricated heterojunctions of *n*-type ZnO on *p*-type Si, which has many advantages such as low cost, large wafer size, and the possibility of integrating oxide semiconductors with already highly matured silicon technology.

The growth of ZnO on Si substrates has been studied extensively including the epitaxial growth of ZnO on Si (100) substrates,[21] ZnO/*p*-Si diodes,[22–24] ZnO:N/*p*-Si heterostructures[25] etc. Studies on the electrical transport properties of ZnO/*p*-Si heterojunctions with different dopants in the *p*-Si[26] and ZnO[27] have also been reported recently. However, due to the complex nature of the carrier transport across the interfaces of the *n*-ZnO/*p*-Si heterojunction, the transport properties of these heterostructures are not yet well understood and are even debatable. We have furthered these studies on *n*-ZnO/*p*-Si heterojunction diodes fabricated by pulsed laser deposition at different oxygen pressures. These heterojunction diodes are found to have highly favorable forward-to-reverse current ratio. We have also studied the parametric dependence of the electrical characteristics of these heterojunctions. The results of these studies are presented and discussed in this communication.

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EXPERIMENTAL

The pulsed laser deposition (PLD) of the ZnO films was carried out in a growth chamber, which was first evacuated to a base pressure of 10^{-6} mbar. A polycrystalline, stoichiometric, sintered (for 5 h at 1200°C) pellet of ZnO with a purity of 99.999% was used as the target for PLD. The third harmonics (355 nm) of a Q-switched Nd:YAG laser with a repetition rate of 10 Hz, pulse width of 9 ns, and fluence of about 3 J/cm^2 per pulse was used for ablation of the ZnO target. Cleaned *p*-type silicon (100) wafers with a carrier concentration of about 1×10^{15} cm^{-3} were used as substrates. The growth chamber was filled with flowing oxygen ambient and its pressure was varied from 0.003 mbar to 0.007 mbar during the growth of different samples. The substrate-to-target distance was kept at about 4.5 cm. The ZnO films were deposited for about 30 min on the Si substrates at room temperature. To measure the conductivity and band gap of the ZnO films those were separately deposited on silica substrates under identical experimental conditions as those used for the growth on the Si substrates. For electrical measurements, indium metal contacts were made on both *p*-type silicon surface and *n*-type ZnO films, which were found to be ohmic in nature. The room-temperature electrical measurements of the ZnO thin films grown on the silica substrates were carried out using the four-probe van der Pau configuration in the Hall geometry.

RESULTS AND DISCUSSION

The thickness of the deposited ZnO films, measured using a stylus profiler (Dektak 6 M Stylus profiler) was found to be about 250 nm. The X-ray diffraction patterns of all the ZnO films showed only (002) peaks along with the Si (200) peak. A typical XRD pattern of these films is shown in Fig. 1a. This confirmed the highly *c*-axis-oriented growth of the ZnO films. The full-width at half-maximum (FWHM) of the (002) X-ray diffraction peak of the ZnO films was found to be about 0.34°, indicating a reasonably good crystalline quality of these films.

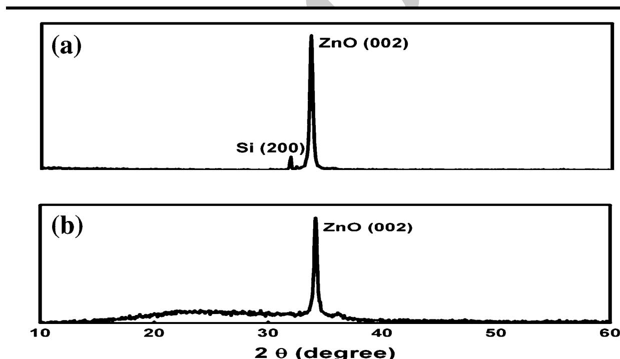

Fig. 1. XRD pattern of ZnO films deposited on (a) *p*-silicon (100) and (b) silica substrates.

X-ray diffraction pattern of the ZnO films deposited on the silica substrates is shown in Fig. 1b. This also showed only a (002) peak of ZnO, confirming the same *c*-axis-oriented growth as in the case of ZnO films grown on the *p*-Si substrates. However the FWHM of this peak was found to be about 0.36°, which is slightly higher than that of the films grown on the Si substrates, as expected.

Figure 2a shows the band gap of the ZnO thin films grown on silica substrates, estimated from the plot of $(\alpha h\nu)^2$ versus $h\nu$. It can be seen from this figure that the band gap decreased from 3.36 eV to 3.257 eV with an increase of the oxygen pressure from 0.003 mbar to 0.007 mbar. Series resistance, an inherent resistance of the depletion region in *N*-ZnO/*p*-Si heterojunction of all the diodes grown at different oxygen pressures was calculated from the plot of log (*I*) versus *V*,[28] which is also shown in Fig. 2a. As can be seen in this figure the series resistance increased from 3.45×10^5 ohm to

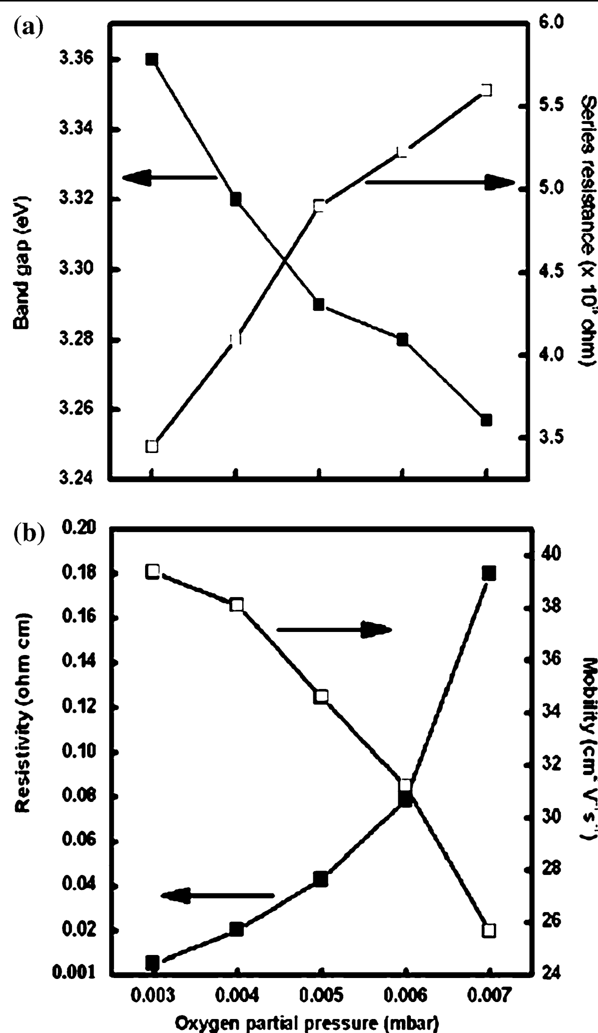

Fig. 2. (a) The series resistance and the variation of the optical band gap with oxygen pressure and (b) a plot of resistivity and mobility with oxygen pressure.

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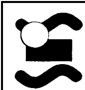
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145 5.6×10^5 ohm with increasing oxygen partial pres-
146 sure from 0.003 mbar to 0.007 mbar. Figure 2b
147 shows the variation of resistivity and the electron
148 mobility for the ZnO thin films with respect to the
149 oxygen pressure. It can be seen from this figure
150 that, while the resistivity increased, the mobility
151 decreased when the oxygen pressure used during
152 the deposition was increased. Hall measurements
153 confirmed the n-type conductivity of the ZnO films.
154 Using these Hall measurements, the carrier con-
155 centration was found to decrease from about
156 3.2×10^{19} cm^{-3} to 1.32×10^{18} cm^{-3} when the oxygen
157 pressure was increased from 0.003 mbar to
158 0.007 mbar, as shown in Fig. 3. A theoretical curve
159 based on the calculated values of the carrier con-
160 centration from the Burstein-Moss (BM) shift[29] is
161 also shown in this figure. With a small gap between
162 the two curves, the trend of experimental data and
163 that of the calculated ones coincide reasonably well.
164 As seen from Fig. 2a the band gap of the ZnO
165 films decreased with increasing oxygen pressure
166 during growth, as did the electron concentration.
167 This means that films grown at lower oxygen pres-
168 sure had a larger band gap due to the enhanced
169 carrier concentration in the film. This increase in
170 the band gap accompanied by an enhanced carrier
171 concentration can be explained using the BM
172 shift.[29] As is well known, this model relies on the
173 effective mass approximation (EMA) in which the
174 wavefunctions are represented by plane waves and
175 the conduction and valance bands are taken to be
176 parabolic near the Brillouin zone. The BM shift in
177 band gap, ΔE_g according to this model[29] is given by:

$$\Delta E_g = \frac{h^2}{8\pi^2}\left(\frac{1}{m_e} + \frac{1}{m_h}\right)\left(3\pi^2 n\right)^{2/3} \quad (1)$$

180 where m_e = 0.28 m_e, m_h = 0.59 m_e, h, and n are the
181 effective electron mass, effective hole mass, Planck
182 constant, and electron density per unit volume,
183 respectively.
184 This leads to a total band gap of

$$E_g = E_{go} + \Delta E_g \quad (2)$$

186 We took the band gap of ZnO without BM shift
188 to be E_{go} = 3.25 eV, which is that of the ZnO bulk
189 crystal at room temperature.[30] The BM shift in the
190 band gap (ΔE_g) was obtained from Eq. 2 using the
191 total band gap (E_g) estimated from the optical
192 transmission spectra. Then electron concentrations
193 (n) were calculated using Eq. 1. These calculated
194 values of the electron concentration are plotted as
195 a function of the oxygen partial pressure in Fig. 3.
196 Experimental values of the electron concentrations
197 obtained from the Hall measurements are also
198 shown in Fig. 3. It can be seen in this figure that
199 the electron concentrations obtained from the Hall
200 measurements match well with those obtained
201 from the theoretical BM shift except at the lowest
202 oxygen pressure. This might be due to the strain
203 resulting from the increased oxygen vacancies in
204 the film.
205 The physical basis for the concentration of oxygen
206 incorporation in the ZnO films was investigated by
207 X-ray photoelectron spectroscopy (XPS) of the films
208 grown at oxygen pressures of 0.003 mbar and
209 0.007 mbar using an Al K_α radiation source
210 (1486.6 eV). The results are shown in Fig. 4. The
211 intensity of the oxygen 1s XPS peak showed greater
212 oxygen incorporation in the ZnO films grown at
213 0.007 mbar oxygen pressure. It was also observed
214 from the XPS data that increase of oxygen pressure
215 during deposition enhanced the O/Zn ratio in the ZnO
216 thin films. From the XPS and Hall measurement data

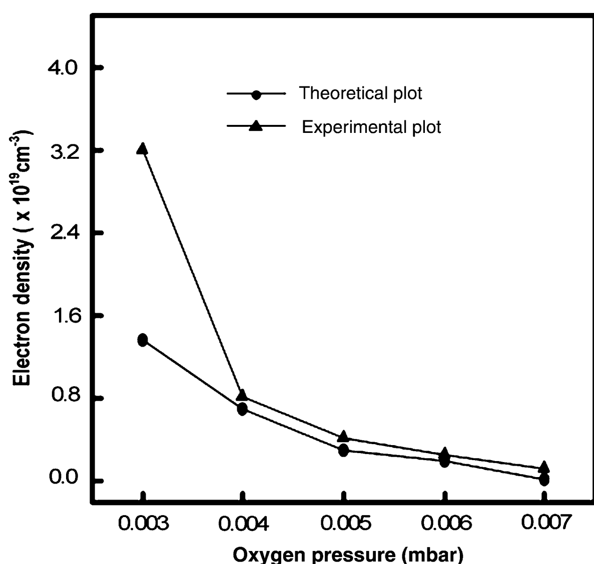

Fig. 3. The variation of the electron concentration in the ZnO films (obtained from the Hall measurement and theoretical model using the BM shift) with oxygen pressure.

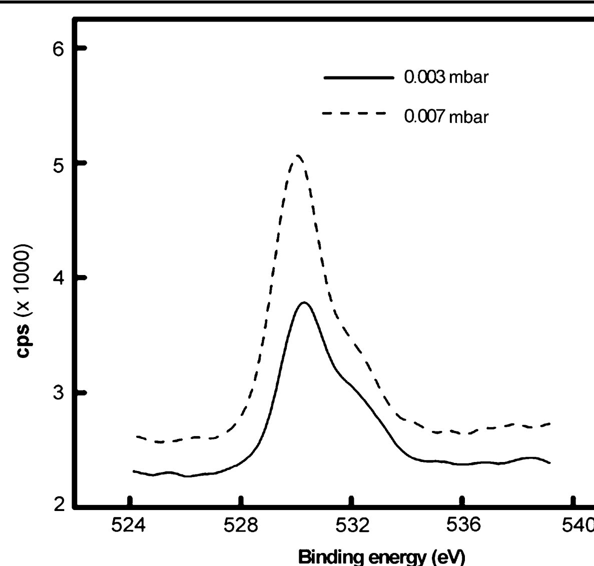

Fig. 4. XPS of O 1s ZnO thin films deposited at 0.007 mbar and 0.003 mbar oxygen pressures.

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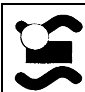
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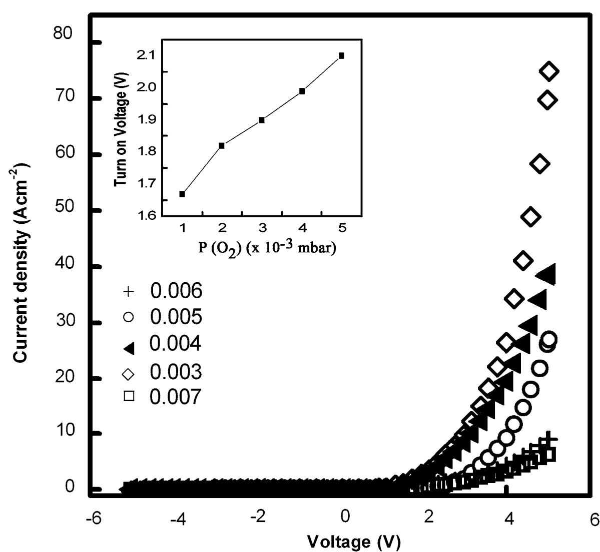


Fig. 5. Current density–voltage (J–V) plot of ZnO/p-Si heterojunctions. The inset shows the variation of the turn-on voltage with oxygen pressure, $P(O_2)$.

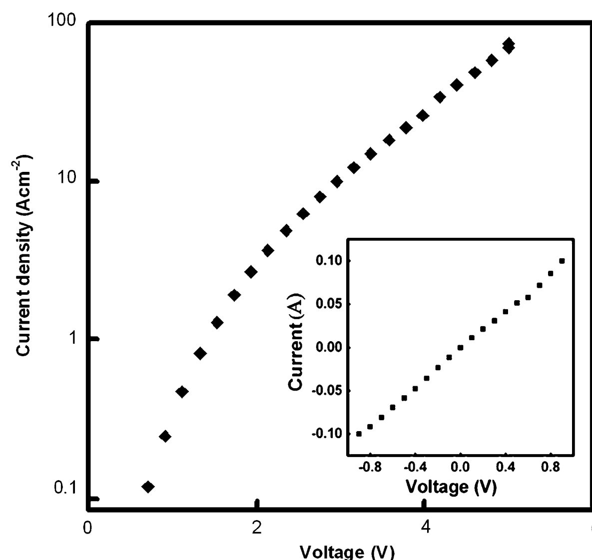
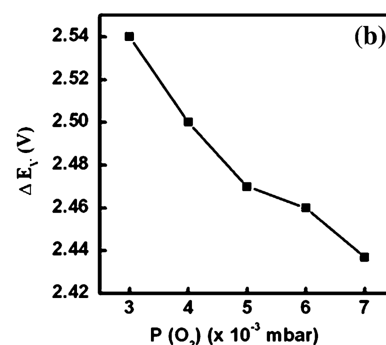


Fig. 6. Current density–voltage (J–V) plot of ZnO/n-Si heterojunctions on a logarithmic scale. The inset shows the current–voltage (I–V) plot of the In/ZnO contact.

it can be elicited that, the greater the level of oxygen incorporation in the films, the lower the electron concentration. This is also in agreement with the earlier study of Look et al.[31]

Figure 5 shows the J–V characteristics of five different n-ZnO/p-Si heterojunctions with ZnO films grown at different oxygen pressures. All of the five heterojunctions were found to be rectifying and the turn-on voltage of the heterojunctions increased as shown in the inset of Fig. 5 with increasing oxygen pressure during the growth of the ZnO films. The J–V characteristics of the n-ZnO/p-Si heterojunction diode with the lowest turn-on voltage is plotted on a logarithmic scale in Fig. 6. The maximum forward-to-reverse current ratio was found to be about 1,000 in the range of applied voltage from –5 V to +5 V. The inset to Fig. 6 shows the ohmic nature of the In/ZnO contact. The room-temperature leakage current at –5 V was of the order of 10^{-7} A. The ideality factor was found to be greater than 10 for all the heterojunctions fabricated.

The band structure of n-ZnO/p-Si at the heterojunction can be constructed using the Anderson model[32] by assuming continuity of vacuum levels and neglecting the effects of dipole and interfacial states. A similar band structure has been suggested for doped and pure ZnO/Si heterojunction by P Chen et al.[26,33] Figures 7a and 8 show the constructed band structure of the n-ZnO/p-Si heterojunction fabricated at the 0.007 mbar oxygen pressure under zero and forward bias, respectively. Values of the band gaps of E_g (ZnO) = 3.257 eV and E_g (Si) = 1.12 eV, and of the electron affinities of χ(ZnO) = 4.35 eV and χ(Si) = 4.05 eV, were used.[26] The valance-band offset (ΔE_v) and conduction-band offset (ΔE_c) are equal to 2.43 eV and 0.3 eV



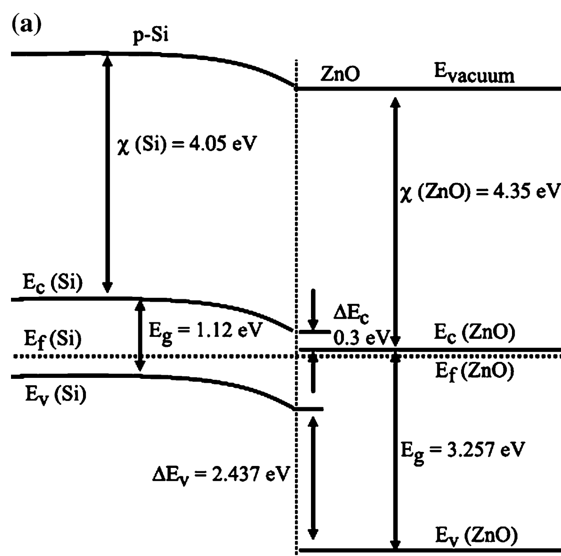


Fig. 7. (a) The band structure of the ZnO/p-Si heterojunction (grown at 0.007 mbar oxygen pressure) under zero bias. (b) The variation of ΔE_v with oxygen pressure during PLD of ZnO films.

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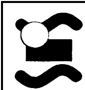
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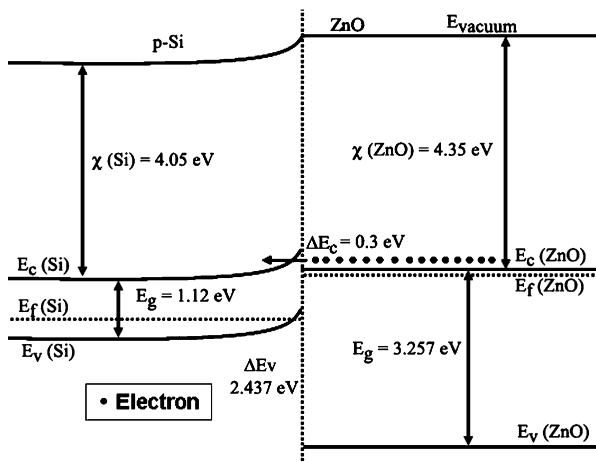

Fig. 8. The band structure of the ZnO/p-Si heterojunction (grown at 0.007 mbar oxygen pressure) under forward bias.

respectively. The variation of ΔE_v with oxygen pressure during PLD of ZnO films is shown in Fig. 7b. Both ΔE_v and ΔE_c are emerging out of the difference in the electron affinities and band gaps of the two materials forming the junction. It can be noted that the valance-band offset ΔE_v is much higher than the conduction-band offset ΔE_c.

Since the carrier concentration in the p-Si side is about 3 orders of magnitude lower than that in the ZnO side, all the depletion region within the p-Si/ZnO heterojunction is extended into the p-Si side. Figure 7a shows that the bottom of the conduction band on the ZnO side lies lower in energy than that on the p-Si side. Hence under relatively low forward bias, the chance of electron flow from the ZnO side to the p-Si side is negligible due to the higher barrier difference felt by the electrons at the bottom of the conduction band on the ZnO side. This resulted in a higher turn-on voltage for the p-Si/ZnO junction grown at 0.007 mbar oxygen pressure. However, under higher forward bias, the barrier difference is lowered and the injection of electrons from the bottom of the conduction band on the ZnO side to the p-Si increased considerably (as shown in Fig. 8). Thereby the forward current rapidly increased under a higher voltage bias. When the oxygen pressure during the deposition of ZnO was decreased, the carrier concentration increased and hence the Fermi level shifted towards the bottom of the conduction band. This means that, upon decrease of the oxygen pressure, the Fermi level may even move into the conduction band, resulting in the easy flow of electrons from the ZnO side to the p-Si side. Hence the forward voltage required for considerable forward current decreased and thereby the turn-on voltage decreased. This seems to explain the decrease of the turn-on voltage for the n-ZnO/p-Si heterojunction fabricated at the lower oxygen pressure.

The variation of the turn-on voltage with oxygen pressure can also be explained with calculated values of series resistance. Due to series resistance, a part of the applied voltage is effectively wasted and hence a larger applied voltage is necessary to achieve the same level of current compared to the ideal value. Hence the turn-on voltage will increase with the increase of series resistance in the quasi-neutral region of p-Si/ZnO. It is noticed that the calculated values of series resistance thus obtained increased with increasing oxygen pressure, thereby increasing the turn-on voltage.

CONCLUSION

In conclusion c-axis-oriented crystalline ZnO films deposited on p-type Si (100) at different oxygen pressures using PLD form effective n-ZnO/p-Si heterojunctions, which were found to be rectifying. The maximum forward-to-reverse current ratio was found to be 1000 in the applied voltage range from –5 V to +5 V. The variation of the turn-on voltage with oxygen pressure was modeled with the Anderson model and the BM shift, which is in agreement with the values of the series resistance calculated across the n-ZnO/p-Si heterojunction.

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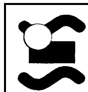

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