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The effect of high-energy electron irradiation on ZnO-based ohmic and Schottky contacts

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Abstract
A systematic study of radiation effects on the major parameters of ohmic and Schottky contacts based on n-ZnO is introduced. Al and Au metals were used as contact elements in order to fabricate the ohmic and Schottky structures, respectively. The transmission line method (TLM) measurements on Al/n-ZnO have revealed that high-energy (6, 9, 12 MeV) and relatively low-dose (3×10^{12} e$^-$ cm^{-2}) electron irradiation produced lower specific ohmic contact resistivity values as compared with the reference sample. The current–voltage (I–V) and capacitance–voltage (C–V) measurements on the Au/n-ZnO structures are shown to increase in ideality and to decrease in the Schottky barrier heights with increasing electron energy. These findings have been interpreted based on the assumption that the atoms of the contact elements diffused into the semiconductor material, thus turning the rectifying character to ohmic behaviour with the influence of radiation–matter interaction and subsequent annealing effects.

1. Introduction

ZnO exhibits a unique combination of very interesting piezoelectric, electrical, optical and thermal properties compared to other compound semiconductors. These properties are already successfully applied in the fabrication of piezoelectric transducer, optical wave-guides, selective gas sensors, acoustic-optic devices and conductive transparent electrodes [1]. ZnO having a band gap of 3.4 eV at room temperature is a promising candidate for ultra-violet light emission and well-known UV laser material. Its useful properties include the existence of highly stable excitons, a high electron saturation velocity, the availability of large-area substrates, amenability to wet chemical etching, relatively low materials' costs, and a high radiation resistance [2, 3]. With regard to the radiation properties, it has been shown that room-temperature bombardment by electrons [4, 5], protons [6] and heavy ions [7] causes much less damage in ZnO than in other semiconductors, e.g., Si, GaAs or GaN. Although its radiation hardness is still not completely understood theoretically, our recent electron irradiation studies show that significant defect annihilations take place, even at low temperatures, thus showing why ZnO is so resistant to radiation effects [8, 9]. This fact suggests that these materials should be useful for space applications in which particle irradiation is strong. It is however important to determine how the contact parameters, both ohmic and the Schottky, give a response to the high-energy electron irradiation since all semiconductor-based devices communicate with each other over the metal/semiconductor (MS) contacts in an electronic circuit. It is for this purpose that we have investigated the effects of high-energy (6, 9, 12 MeV) and relatively low-dose (3×10^{12} e$^-$ cm^{-2}) electron irradiation on n-ZnO-based ohmic and Schottky contact parameters.

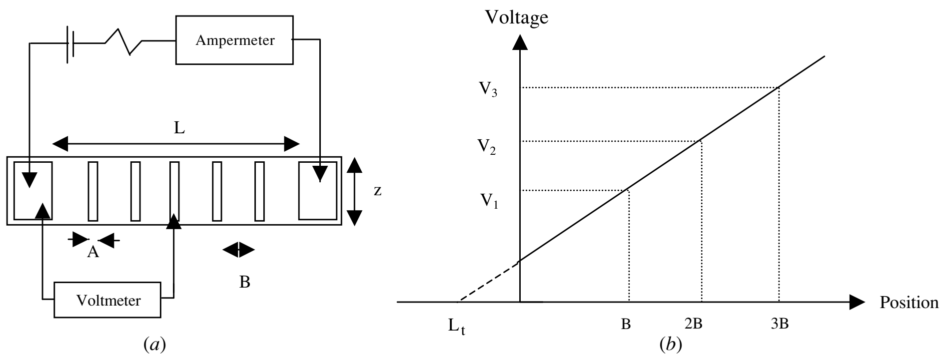

Figure 1. (*a*) Ladder network pattern and (*b*) its possible voltage-position representation for specific ohmic contact resistivity measurements by TLM [14].

2. Experiment

In this work, n-ZnO bulk samples from Eagle–Picher were used. In order to ensure the sample homogeneity, four adjacent pieces with dimensions $5 \times 5 \times 0.5$ mm^3, two of them are the reference samples and the other two are the samples to irradiate, were cut out from the same wafer. All samples were cleaned with trichloroethylene, methanol, acetone and deionized (DI) water, respectively. 6N Al [10] and Au [11] metals were used as contact elements in order to fabricate the ohmic and Schottky structures, respectively. Both metals were evaporated through shadow masks (SMs) produced before [12], in an *Edwards Pumb* system with a base pressure of 10^{-5} Torr. The ohmic contact samples and the ohmic sides of Schottky samples were annealed in a homemade furnace [13] under dry N_2 gas flow at 300 °C for 5 min.

The specific contact resistivity of ohmic contacts to ZnO was measured by applying the transmission line model (TLM), as illustrated in figure 1(*a*). This method was originally proposed by Shockley in 1964. In this technique, a constant current is applied between two large-area ohmic contacts and then two voltage probes, connected to a high-impedance voltmeter (Keithley 171 Digital Multimeter), are placed, respectively, on a current pad and one of the other pads spaced at B, 2B, 3B, etc. This allows us to plot potential versus distance, as shown in figure 1(*b*). In general, zero potential appears at some extrapolated distance L_t (transfer length). Shockley has shown that the specific contact resistivity, ρ_c, is given by [14]

$$\rho_c = L_t^2 R(\Omega\ \mathrm{cm}^2), \tag{1}$$

where L_t is the intercept and R is the slope of the voltage versus position graph. A SEM image of the SM used in transferring the ladder pattern to the sample surface was taken using a JSM 6400 scanning electron microscope and is shown in figure 2 [12].

The major parameters of the Schottky diodes such as Schottky barrier height (SBH), ideality factor, donor concentration and Fermi level energy have been calculated using the well-known thermionic emission (TE) theory formulation which can be found elsewhere [15, 16]. Electron irradiation was achieved by Siemens–Primus electron accelerator which allows one to accelerate the electrons up to 21 MeV energy.

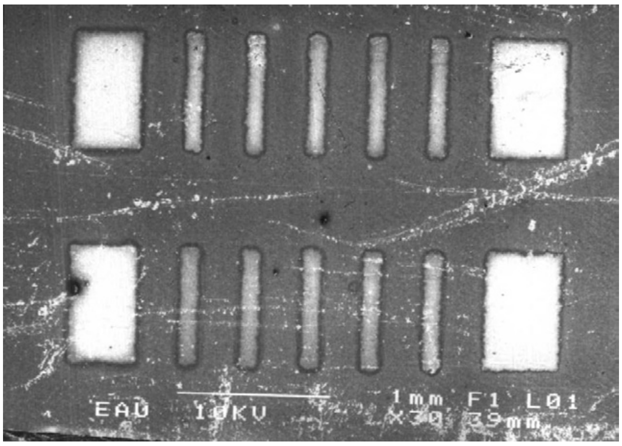

Figure 2. SEM image of the SM used in measuring ohmic contacts by TLM. The sizes of the large current contacts and narrow voltage contacts are 692×384 and 692×76 μm^2 (length × width), respectively [12].

3. Results and discussion

3.1. Ohmic contacts

In order to investigate the radiation effects on Al/n-ZnO ohmic contact structures, two samples fabricated at the same conditions were prepared, and one of them (called RO1) was kept to be a reference sample. Electron irradiation (EI) studies were carried out on the other sample (IO1) so as to understand the possible radiation and subsequent annealing effects. The electrons were directed onto the ohmic facet of the samples lest they lose their kinetic energies by colliding with the atoms through the material. The total sequence of irradiations and anneals, for the sample called IO1, is designated as follows: (1) as-grown (before EI); (2) 6 MeV (after first EI at 6 MeV); (3) 9 MeV (after second EI at 9 MeV); (4) 12 MeV (after third EI at 12 MeV); (5) A 400 °C (after annealing at 400 °C for 10 min). All EIs were carried out at room temperature (RT) and lasted about 4 min. The total fluence, F, for each EI was around of 3×10^{12} e cm^{-2}. Also, annealing process at 400 °C was done under dry nitrogen flow with a sensitivity of ±1 °C. Figure 3 shows the ohmic behaviour of the current–voltage (I–V) characteristics taken right after each event, using HP-4140 B picoammeter. As seen in the figure,

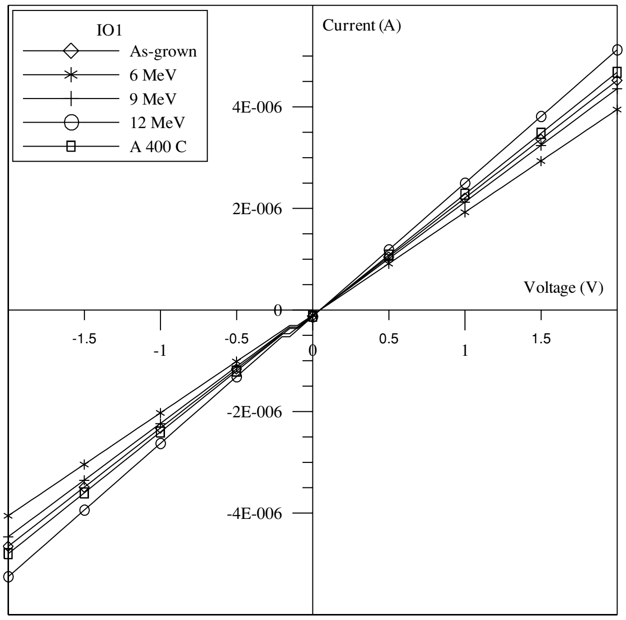

Figure 3. The effect of electron irradiation and subsequent annealing procedure on the I–V characteristics of the Al/n-ZnO ohmic structures. Each line was taken right after the related event.

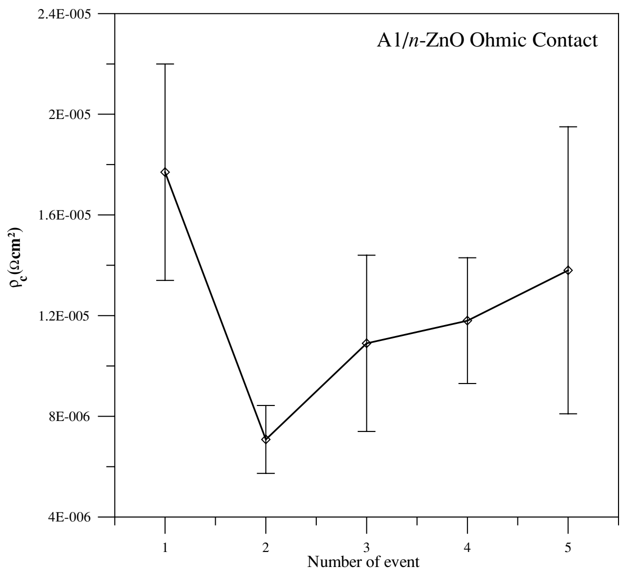

Figure 4. The specific ohmic contact resistivity values for each event. The events are designated as follows: (1) before EI (as-grown); (2) after EI1 ($E = 6$ MeV); (3) after EI2 ($E = 9$ MeV); (4) after EI3 ($E = 12$ MeV); (5) after annealing at 400 °C for 10 min.

each I–V curve exhibits a clear ohmic behaviour. Irradiation at 6 MeV diminishes the current at the same voltage value as compared to the reference sample. However 9 and 12 MeV electron irradiations improve the I–V characteristics and then subsequent annealing at 400 °C for 10 min brings it closer to the reference sample. This behaviour should suggest that the bulk resistance of the sample is increased by first EI and then is shown to decrease at further processes. In order to get additional evidence about the effect of electron irradiation on Al/n-ZnO ohmic contact structures we calculated the specific ohmic contact resistivity of the samples by TLM after each event. Figure 4 shows the variation of ρ_c with respect to the event number. The huge error bars could be resulted from the instrumentation problems. When investigated the variation, in accordance with the above interpretation, one can see that ohmic contact resistivity decreased after the first irradiation. This is expectable because Al atoms diffuse into the semiconductor with the influence of electron–Al atoms collision. Furthermore, heat which emerged due to the electron–lattice interaction can cause a kind of annealing effect and thus diminishes the ohmic contact resistance drastically. However, further irradiations and anneal do not result in any improvement in ohmic contact resistivity of the Al/n-ZnO structure. What is likely occurring during these processes is that first irradiation at 6 MeV energy creates the V_{Zn}–Zn_I Frenkel pairs [17] with the influence of displacement damage and then sample heating during subsequent irradiations and anneal at 400 °C dissolve the Frenkel pairs and turn them into an atom in a lattice point. Thus, after first electron irradiation, the bulk resistance of ZnO increases whereas the specific ohmic contact resistivity decreases due to the defect formation and then inversely the bulk resistance decreases whereas the specific ohmic contact resistivity is shown to increase due to the defect annihilation. This could be a satisfying explanation when considered the reverse relation between ρ_c and defect concentration [18].

Table 1. Variation of the major parameters of Au/n-ZnO Schottky diodes with respect to the incident electron irradiation energy.

	Φ_b (eV)					
Events	I–V	C–V	n	N_d (cm^{-3})	V_{dif} (eV)	E_f (eV)
As-grown	0.53	0.62	2.63	2.96×10^{16}	0.50	0.124
6 MeV	0.51	0.58	5.07	2.41×10^{16}	0.45	0.29
9 MeV	0.50	0.56	5.99	2.41×10^{16}	0.43	0.129
12 MeV	0.49	0.52	7.62	2.23×10^{16}	0.39	0.131

3.2. Schottky contacts

Radiation effects on Au/n-ZnO Schottky structures have been investigated on two samples fabricated at the same conditions. The sample RS1 has been kept to be a reference sample and electron irradiation studies are carried out on the sample IS1. The electrons are directed onto the Schottky face of the samples for this time in order to understand the radiation effects on Au/n-ZnO Schottky structures. All radiation parameters are the same as ohmic contact study given before. Figure 5 shows the current–voltage (I–V) and capacitance–voltage (C–V) characteristics of Au/n-ZnO Schottky diodes taken right after each event, using HP-4140 B and HP-4192 A impedance analyzsr at 500 kHz, respectively. Also, table 1 gives the variation of the major parameters of Au/n-ZnO Schottky diodes for each event. In calculation of the parameters, the dielectric constant, $\varepsilon_s\varepsilon_0$, the density of states for conduction band, N_c and the Richardson constant, A^* have been taken to be 1.96×10^{-11} F m^{-1}, 3.5×10^{18} cm^{-3} and 0.15 A cm^{-2} K^{-2}, respectively [19]. As seen in table 1, the ideality factor increased with increasing radiation

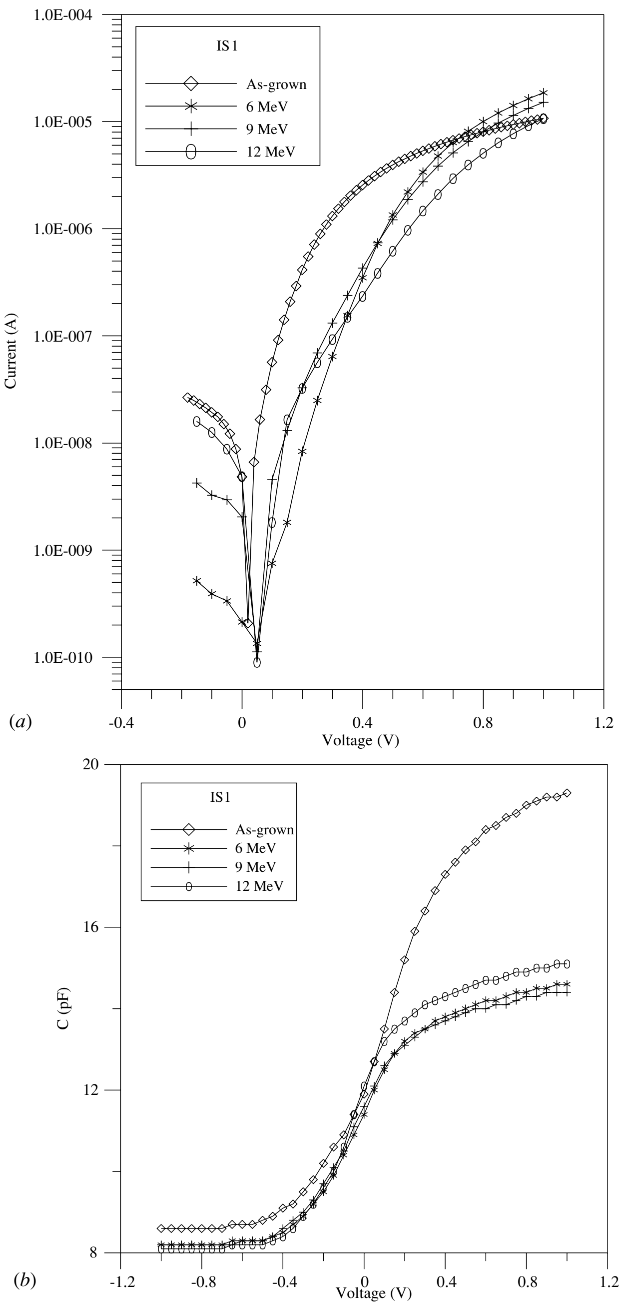

Figure 5. (*a*) The current–voltage (*I*–*V*) and (*b*) capacitance–voltage (*C*–*V*) characteristics of Au/n-ZnO Schottky diodes.

energy, whereas the SBHs obtained by both *I*–*V* and *C*–*V* measurements are shown to decrease. This fact suggests that the electron irradiations turn the rectifying behaviour of the Au/n-ZnO Schottky structure into the ohmic contact behaviour and thus removes the contact from the ideality. It is frequently observed experimentally that the Schottky barrier height calculated from *I*–*V* characteristics does not agree with the barrier height extracted from *C*–*V* measurements [20–22]. Some general causes for these differences in barrier height have been mentioned in the literature, such as surface contamination at the interface, deep impurity levels, an intervening insulating layer, quantum mechanical tunnelling, image force lowering, and edge leakage currents [23–25]. Also in [21], the authors attributed the difference observed for Si and GaAs to the influence of thermionic field emission on the charge transport through the interface. Similar observations have been reported for several metal/InP Schottky barriers [22], and the discrepancy observed in the values obtained for the BH was attributed to an interfacial layer and/or interface states. In general, these kinds of interface states affect the *I*–*V* behaviour because they may act as recombination centres or as intermediate states for trap-assisted tunnel currents. Therefore this mechanism can increase the ideality factor and lower BH [26]. *C*–*V* measurements are less prone to such defects because they are based on ac technique and thus, measure the average barrier height, regardless of the change in current. Thus, these states can contribute to the difference observed for the two techniques. In our case, what is probably occurring is that the atoms of the contact elements diffuse into the semiconductor material with the influence of radiation–matter interaction and create the recombination centres and traps at the metal/ZnO interface. These states give rise to the trap-assisted tunnel currents and thus diminish the SBH and increase the ideality. Also these traps that prevail at the interface can compensate the pre-existing donors at the interface by lowering the N_d [9] and dislocates the position of Fermi level with respect to the conduction band (see table 1). In recent years, similar observations have been reported in proton irradiated ZnO-based Schottky diodes as well [27, 28]. These results may have practical as well as fundamental significance, because the high energetic particles are often encountered in space applications.

In conclusion, the effects of high-energy (6, 9, 12 MeV) and relatively low-dose (3×10^{12} e$^-$ cm^{-2}) electron irradiation on Al/n-ZnO ohmic contact and Au/n-ZnO Schottky structures have been investigated. TLM measurements have shown that the ohmic contact resistivity decreased after the first irradiation at 6 MeV and then increased slightly at further irradiations and anneals. The current–voltage (*I*–*V*) and capacitance–voltage (*C*–*V*) measurements on the Au/n-ZnO structures are shown to increase in ideality and to decrease in the Schottky barrier heights with increasing electron energy. These results have been interpreted based on the assumption that the atoms of the contact elements diffused into the semiconductor material and thus turning the rectifying character to the ohmic behaviour with the influence of radiation–matter interaction and subsequent annealing effects.

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