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Analysis of junction properties of gold-zinc oxide nanorods based Schottky diode by means of frequency dependent electrical characterization on textile

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Abstract:

Present work is an effort to reveal the junction properties of gold (Au) / Zinc oxide (ZnO) nanorods based Schottky diode by using the frequency dependent electrical properties. The most important electrical parameters such as conductance (G), resistance (R), capacitance (C) and impedance (Z) were studied as function of frequency across the series of AC voltages. Moreover, current density-voltage (J - V) was measured to know the performance of present Schottky diode. The effect of native defects at the interface was also studied by using cathodoluminescence spectroscopy by varying different accelerating voltage. The textile substrate was used for the growth of ZnO nanorods by using the aqueous chemical growth (ACG) method and Schottky diode fabrication. Diode fabrication on textile fabric is a step forward towards the fabrication of electronic devices on non-conventional, economical, soft, light weight, flexible, wearable, washable, recyclable, reproducible and nontoxic substrate.

Keywords: Textile fabric; ACG method; ZnO nanorods; Schottky diode; frequency dependent electrical characterization.

1. Introduction

Textiles are an essential and fundamental component of our everyday lives for hundreds of years. At one extreme, it is an attractive material by means of composition, construction, and/or finishing; while on the other side textile has been utilized as a substrate for fabrication of sensors, output devices, and printed circuit boards. Since last few years researcher are working hard to integrate the desired functionalities inside the textile architecture [1], like introduction of electronic or optical sensors and output devices in the textile architecture. That means development in this field is focused on creating devices and logic circuits in order to achieve electronic functions based textiles. But the challenge in creating such textiles is to preserve the comfort ability and wash ability of textile in the presence of electronic circuits, so it can be used easily in daily life [2-4]. At commercial level we need such textiles that fulfill the requirements of both the textile and electronics. Subsequent development in this field has already seen considerable applications in this regard including healthcare, fashion, energy, sports, wellness, safety and security, automotive and transport construction, home and interior textiles [1-10].

ZnO is a well-known and an interesting compound semiconducting material with several worthy properties including wide direct band gap (~ 3.37 eV), relatively high exciton binding energy (~ 60 meV) at room temperature, large piezoelectric coefficient, high thermal / chemical stability, and additionally being bio-safe and biocompatible. Therefore ZnO is a useful material for the fabrication of different electronic devices especially on textile fabric. ZnO also possesses self-organized growth properties and due to this ZnO nanostructures can be grown on any substrate such as GaN, SiC, Si, glass, plastic/fiber, paper and textile fabric etc. [11-17]. This property is advantageous to facilitate ZnO nanostructures as a material for development of simple and cost effective nanodevices on cheap substrates. In addition, several morphologies of ZnO nanostructures have been synthesized like one dimension nanorods, nanowires, nanoneedles, nanotubes, and two dimension nanoflakes and nanowalls etc. But one-dimensional (1D) morphologies are widely used for the development of different nanodevices [11]. ZnO nanostructures have been used for the fabrication of optical, electrical, and piezoelectric devices e.g. light emitting diodes [11], solar cells [18], laser diodes [19], and UV photo detectors [20] etc., a recent comprehensive review of variety of issues related to Schottky contacts on ZnO thin film has been published [21].

The study of frequency dependent electrical characterization of Schottky diodes on nanostructures is of interest, as the miniaturizing of nanodevices can affect the performance of nanodevices [22-24]. Moreover, Schottky diodes are associated with quicker switching and lower turn on voltages compared to p-n junction diodes. There has been less work reported on the frequency dependent electrical characterization of metal-semiconductor Schottky nanocontacts [25-26]. Faraz et al. fabricated Au/ZnO Schottky diode to investigate the interface properties [27]. Hussain et al. also develop Au/ZnO based Schottky diode to investigate the interface properties [28]. Park et al. (2003) fabricated Schottky nanocontacts based on Au/ZnO/ITO heterostructures with improved electrical properties like reverse bias breakdown voltage from -3 to -8 volts by capping the Au layer on the nanorods tip [29]. Cheng et al. (2007) demonstrated the role of Schottky contact for improving the electronic properties of ITO/ZnO based Schottky diode using atomic force microscopy (AFM) [30]. Brillson et al. (2012) deeply investigated defects at interface of metal-semiconductor/ZnO Schottky barrier and Ohmic contact with different materials [31-33]. Panda et al. (2013) investigated the electrical properties of Schottky nanocontacts on single crystalline ZnO nanorods by using AFM in contact mode for PtIr tip/ZnO Schottky nanocontacts [34].

Recently, we have developed ZnO nanorods based Schottky diode on silver (Ag) coated textile fabric substrate. The choice of textile substrate is advantageous over other conventional substrates due to various properties of textile such as being cheap, abundant, soft, light weight, wearable, washable, recyclable and non-toxic. While aqueous chemical growth method used for growth of ZnO nanorods which is an environment friendly and non-toxic method. The performance of Schottky diode was investigated by means of frequency dependent electrical characterization across the series of AC voltages. Moreover, analysis of junction properties and grain boundaries effect was investigated by using frequency dependent G , R , C and Z to understand the present transport mechanism in the fabricated Schottky diode along with I-V characteristics, which is also an effective alternative technique to analyze the device performance. Moreover the cathodoluminescence (CL) spectroscopy was also performed to discuss the effect of ZnO native defects on metal-semiconductor interface.

2. Experimental Section

2.1. Growth of ZnO nanorods:

The sample preparation is very important step in order to achieve the desired performance of the fabricated device. Therefore all experimental work has been conducted in clean room environment. First of all the textile sample was washed by detergent powder and dried in air, then washed by acetone, isopropanol and deionized water for 5, 5 and 10 minutes respectively in electronic sonicator to avoid any dust particles or any other unseen contaminants. Now the clean textile sample surface was treated by a homogeneous thin layer of polyimide (PI) by using a spin coater at 4000 rpm to fill the space between the textile fibers and to reduce the surface roughness. Finally the prepared substrate was heated for few minutes on hotplate at a temperature of 80°C for proper stacking. For the surface to be conducting a thin layer of Ag was used as bottom electrode. Optical microscopic images of the surface preparation are shown in figure 1 (a-c). Now zinc acetate mediated seed solution was deposited three times by spin coating technique at 4000 rpm for 40 s (the ZnO seed solution was prepared by dissolving zinc acetate and potassium hydroxide (KOH) in methanol such as, “An amount of 274 mg zinc acetate was dissolved in 125 ml methanol at a temperature of 60° C and 109 mg potassium hydroxide (KOH) was dissolved in 65 ml methanol at room temperature separately. Then solution containing KOH was drop wise mixed into other solution at a temperature of 60° C, while beaker was continuously on magnetic stirrer until the solution will be transparent). Afterwards the seed layer coated substrate was annealed at 100°C for several minutes to have good adhesion of the seed layer. Annealing at 100°C will not effect on Ag because its melting point is more than 900°C, only it caused a minute diffusion into the PI/textile substrate. ZnO nanorods were then grown by using the low temperature ACG method. The growth solution (aqueous solution) was prepared by dissolving zinc nitrate and hexamethylenetetramine in 200 ml deionized water in equilmolar concentration. The substrate was affixed into a Teflon sample holder facing downward and dipped into the growth solution contained beaker. Now the beaker was fully covered by the aluminum foil and placed in a preheated oven at temperature of 90°C for 6 hours. After this period the substrates with grown ZnO nanorods were taken out and washed with deionized water in order to clean the residue on the substrate and then dried by nitrogen.

2.2. Device fabrication:

Prior to the growth of ZnO nanorods a small portion of Ag coated textile substrate was covered by heat resistant plastic and scotch tape to be used later as a bottom contact to the ZnO nanorods. Short circuit was prevented by filling the gaps between ZnO nanorods with

homogeneous layer of (Shipley-1805) photoresist, (which is an insulating polymer) by spin coating technique at 2000 rpm. The sample was heated on the hot plate at 120°C for 30 s to dry / proper sticking of insulting layer. Now to expose the top surface of the grown ZnO nanorods, reactive ion etching (RIE) plasma was applied and a shadow mask of (1.5 mm diameter) circular shape was used for making the Au top contacts (front Schottky contacts) by using thermal evaporation technique in Satis CR 725 evaporator at a pressure of 2×10^{-7} torr. Schematic diagram of the grown ZnO nanorods is shown in figure 2 (a) and schematic diagram of fabricated Schottky diode is shown in figure 2 (b).

3. Results and Discussion

3.1. Structural characterization

The crystalline quality of grown ZnO nanorods was analyzed by using the X-ray diffraction (XRD) technique. In figure 3(a), all diffraction peaks such as (100), (002), (101) and (102) belongs to the ZnO as reported in JCPDS card no (36-1451). As the grown ZnO nanorods are perpendicularly oriented along the c-axis direction therefore the (002) peak is much higher than the other diffraction peaks. While the two additional peaks indicating the existence of Ag. Figure 3(b) represents low resolution SEM image of grown ZnO nanorods on the threads of the textile, while figure 3 (c) is the high resolution SEM image of grown ZnO nanorods. These SEM images confirm that the grown ZnO nanorods are uniform, well aligned and highly dense along with hexagonal wurtzite structure. The diameters of the grown ZnO nanorods were in range of 150 - 200 nm and the length is $\sim 1.8 - 2 \mu\text{m}$.

3.2. Current density-voltage characteristics

The J - V characteristic of Au-ZnO nanorods based Schottky diode was measured at room temperature. In figure (4), the J - V characteristics exhibits non-linear rectifying behaviour with threshold voltage of 2.1 V and the soft breakdown voltage starts at around -5 V. Inset in figure (4) represents the semi-log plot of J - V characteristics. The estimated values of the barrier height and the ideality factor were found to be 0.61 eV and 2.34 respectively. These are the average values in agreement with other published works [26-29, 35-38]. Following fundamental equations of a Schottky diode were used for the calculation of electrical parameters.

$$I = I_s \left\{ e^{\left(\frac{qV}{\eta kT} \right)} - 1 \right\} \quad (1)$$

$$I_s = AR^*T^2 \left\{ e^{\left(-\frac{q\phi_b}{kT}\right)} \right\} \quad (2)$$

Where A is the active area of Schottky diode, R^* is the Richardson constant, ϕ_b is the barrier height, k is the Boltzmann constant, T is the absolute temperature and I_s is the reverse saturation current. The value of $R^* = 32 \text{ A/cm}^2\text{K}^2$ for n-ZnO obtained theoretically by using $R^* = 120 m_n^*/m_o \text{ A/cm}^2\text{K}^2$, where $m_n^* = 0.27 m_o$, m_n^* is the effective mass of electrons [27]. The reverse saturation current density was observed to be $\sim 199.4 \mu\text{A/cm}^2$ at -4 V . The estimated value of ideality factor indicates that, the thermionic emission is not the dominant transport mechanism. This implies that other mechanisms like recombination-generation, tunneling, and interface states assisted transport are present [27].

3.2. Frequency dependent impedance analysis

The room temperature frequency dependent electrical characterization of the Au-ZnO based Schottky diode was investigated by using the impedance spectrometer at zero DC bias.

Frequency dependent G is a very common tool to analyze the hopping dynamics of the charge carrier [24]. Normally, G is increased due to the carrier charges flow from ZnO grains towards grain boundaries [23]. The figure 5 (a) exhibits a non-linear exponential increasing behaviour of G at higher frequencies ($>4 \text{ MHz}$) and linear/constant behaviour at lower frequencies ($< 4 \text{ MHz}$) having no influence of the AC signals. The magnitude of G is constant at different AC voltages. The G is directly proportional to the frequency as the G of the diode is increasing with increase in frequency and vice versa. It means that, the G has strong frequency dependence and showing the proper charge carrier at the junction of metal and ZnO nanorods [23-24]. This behaviour between metal and semiconductor is very important to analyze the device performance and influence on the transport properties of Schottky diode. Often this kind of dependence attributed to the presence of Schottky barrier and impurities level [26, 28, 39-40]. Figure 5 (b) represents R as a function of frequency in linear scale. The analysis of R in figure 5 (b) confirms that, the R is decreasing exponentially with increasing frequency ($<4 \text{ MHz}$), while linear (zero) at the higher frequencies ($> 4 \text{ MHz}$). The flow of charges at the junction is directly influenced by the R . It means that, the R of the Schottky diode has frequency dependence [25, 39-40].

Figure 6 displays the semi log plot of capacitance-frequency (C-f) measurement of present Schottky diode over a series of AC voltages (0.1 to 3 volts). The graph is showing

exponential decay for $f < 1$ Hz and then it becomes linear for $f > 1$ Hz. The effect of AC signals in terms of variation in C magnitude caused by the dispersion in grain-grain boundaries at the interface of metal-semiconductor [22]. In general, the measured capacitance for the Schottky junction depends on the reverse bias potential and frequency. There are some distinct features behind the AC voltage and frequency dependency such as impurity levels, Schottky barrier, interface states, series resistance and interface layer at the junction of Au and ZnO [41]. At the lower frequencies capacitance has the higher values because of excess capacitance originating from the interface states, which are in equilibrium with the ZnO to follow the AC signals properly. Therefore the capacitance at the lower frequencies for Schottky junction is dominated by the depletion capacitance, which is independent of frequency and dependent of bias [41]. Hence with the increasing frequency the diode capacitance can be affected by the bulk resistance as well as by the depletion capacitance. The frequency dependent bulk resistance of the Schottky diode occurred due to the electron emission responding from the deep impurities levels slowly. Moreover, at the higher values of frequencies interface states cannot follow the AC signals because interface states have no contribution to the capacitance. This is why at the higher values of the frequencies the diode capacitance is low as compare to the lower values of the frequency [40]. The obtained results are closed to those obtained for metal–semiconductor contacts [34-42].

Figures 7 (a) and 7 (b) are semi-log plots of frequency vs real (Z') and imaginary components (Z'') of the Z . Figure 7 (a) describe that, Z' is linear at low frequencies and decaying at the higher frequencies. The variation in magnitude of the Z' at different AC voltages is due to the negligible effect of the R at the junction, thus a proper charge injection between metal and semiconductor has been observed. It is observed that, the Z' of the Z is following the AC signals. The disturbance in graph caused by the surface roughness of textile substrate or it can be due to grain boundaries effect of the inherent properties of ZnO nanorods [23-24, 43]. It is also the indication of presence of deep level defects in the depletion region, which was formed at the grain boundaries of ZnO nanorods [23]. Figure 7 (b) displays semi elliptical profile of imaginary component of the Z . These semi-elliptical peaks are formed in response to combined effect of the C and R . The exponentially increasing curve at the lower frequencies (<50 kHz) is due to the C and the exponential decay at higher frequencies (>50 kHz) is because of R . These results are consistent with the published reports [23, 39-40].

In figure 8, Nyquist plot ascribes a semi-circular behaviour, which is asymmetric at the different AC voltages with different diameters. These semi-circles describing the charge transfer process at the interface of the metal and semiconductor [43]. This semi-circular shape of Nyquist plot is due to the electrical properties of the grain and grain boundaries of ZnO nanorods [39-40]. The process of injected charge transfer inside the ZnO nanorods and the injected charge transfer with in the metal exhibited large semi-circles at the lower frequency in reverse order over the series of AC voltages [43]. This trapping occurred due to the high R effects on carrier flow between the metal and semiconductor or it can be caused due to the surface roughness or lattice mismatch at the interface of metal-semiconductor [22-24]. Usually, in n-type ZnO a large amount of electrons tends to diffuse away to reach the interface layer but trapped by the trapping states by a positive ionized donor charges in the neighborhood of interface of metal-ZnO nanorods [23-28]. Therefore a large amount of charges moved along the metal and semiconductor interfaces, so trapping states occurred in the interface layers due to the various metal-oxides addition [23-25]. In our Schottky diode a typical Nyquist exhibited the proper charge injection of electrons between the metal and semiconductor interface. The magnitude of the diameter at the different AC voltages indicates that, the Z has frequency dependence and R is involved in carrier flow between the metal and semiconductor. When AC voltage is increased then the diameter of semi-circle is decreased, this indicates that the R of Schottky diode is increasing with increase in AC voltage [22-28, 39-40 and 43-44].

3.3. Cathodoluminescence spectroscopy

CL technique was performed to investigate the native defects in the grown ZnO nanorods. CL spectra were captured by Mono CL 4 system which was integrated with a LEO 1550 Gemini SEM by using a single channel photomultiplier tube and nitrogen cooled charged coupled device camera calibrated in SEM under the high vacuum ($< 10^{-5}$ torr) with accelerating voltage from 5-20 kV. Figure 9 (a) shows the room temperature CL measurement in semi-log scale of the grown ZnO nanorods measured at different acceleration voltages ranging from 5-20 kV. It is observed that, the near band emission (NBE) peaks at different accelerating voltages were less in intensity than the corresponding deep level emission (DLE) peaks [33]. Both peaks intensities were increasing with increasing the accelerating voltage. The DLE peak centered at around 2.0 eV in figure 9 (a) which indicates the existence of the native defects in the grown ZnO. As the

accelerating voltage is increasing the depth of penetration of the beam is increasing. The DLE peak intensity of the ZnO nanorods was observed to increase linearly as the accelerating voltage is increased and at relatively high accelerating voltages it starts to saturate [45]. The same is also observed regarding the NBE peak intensity (see Figure 9(b)). The linear increase of the DLE peak intensity with increasing the accelerating voltage indicates that the distribution of native defects is along the whole horizontal length of the nanorods. This means that these native defects also exists at the Au/ZnO nanorods junction, and hence they do influence the transport properties. To obtain quantitative information on the native defects at the interface, we have attempted to perform deep level transient spectroscopy (DLTS) measurements on our Schottky samples. Unfortunately, our substrate, namely, textile fabric was not thermally stable and hence it was not possible to perform DLTS measurements.

4. Conclusion

Well aligned ZnO nanorods arrays were grown on Ag coated textile fabric substrate. The Au/ZnO-based Schottky diode exhibited nonlinear and asymmetric J - V characteristics. The estimated values of the ideality factor, barrier height and the reverse break down voltage were 3.9, 0.67 eV and -5 V respectively. Moreover, the junction properties of diode were characterized by means of frequency dependent Z . The accumulation of charges at the grain boundaries and the junction of metal-semiconductor have been discussed in $(G-f)$, $(R-f)$, $(C-f)$, $(Z'-f)$, $(Z''-f)$ and $(-Z''-Z')$. CL spectroscopy was performed at room temperature by varying the accelerating voltage from 5-20kV; it demonstrates that the proposed device has native defects which affects the device performance. This study will possibly help to integrate electronic devices on non-conventional substrates like textile. Moreover, this study will also contribute to develop non-toxic chemical based future generation devices with several features like environment friendly, abundant, soft, flexible, wearable, washable, recyclable, and reproducible.

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Figures captions:

Figure 1: Microscopic images of textile fabric substrate **(a)** before surface treatment **(b)** after surface treatment by polyimide film **(c)** after deposition of Ag layer.

Figure 2: **(a)** Schematic diagram of growth of ZnO nanorods on pre-treated textile and **(b)** Schematic diagram of Schottky diode.

Figure 3: **(a)** Typical XRD pattern of grown ZnO nanorods on pre-treated textile fabric substrate **(b)** SEM image of grown ZnO nanorods at low magnification and **(c)** at high magnification.

Figure 4: Typical J - V characteristics of Au-ZnO Schottky diode in linear scale, while insert showing the semi-log plot of J - V characteristics.

Figure 5: A plot of frequency dependent **(a)** G - f in linear scale and **(b)** R - f also in linear scale over the series of AC voltages.

Figure 6: Typical semi-log plot of C - f at room temperature over a series of AC voltages.

Figure 7: Typical semi-log plot of frequency dependence **(a)** $\hat{Z}'$ - f and **(b)** $-Z''$ - f over a series of AC voltages.

Figure 8: Nyquist plot ($-Z''$ - Z') of Au-ZnO nanorods based Schottky diode.

Figure 9: **(a)** A typical room temperature depth dependent CL spectra of grown ZnO nanorods captured at accelerating voltages varies from 5 – 20 kV **(b)** ratio of DLE and NBE as function of accelerating voltage.

Figure 1:

Figure 2:

Figure 3:

Figure 4:

Figure 5:

Figure 6:

Figure 7:

Figure 8:

Figure: 9