Electrical conductivity of Cu/ZnO/Si heterostructures

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Abstract. The electrical conductivity studies of the Cu/ZnO/Si thin film heterostructures were carried out by the current-voltage (*I-V*) characteristics. It was found that the *I-V* characteristics were asymmetric and showed weak rectifying properties. The most probable mechanism of electrical conductivity was determined. The concentration of trapping levels and the carrier mobility were calculated.

1. Introduction
Zinc oxide possesses a unique combination of physical and chemical properties: a high melting point and thermal conductivity, photosensitivity [1], piezoelectric [2] and pyroelectric [3] effects, the adsorption of gases on the surface [4]. This material is widely used in microelectronics, chemistry and medicine [5]. Single crystals, thin films and diode structures based on the ZnO material are the subjects for scientific studies and applications.

An anisotropic crystal structure, a non-stoichiometric composition, a wide band gap, semiconductor parameters varied by doping and other properties make this material attractive for comprehensive research. Material characteristics largely depend on preparation conditions and various external factors. Such dependences are especially significant in the case of ZnO thin films, which arouse now great interest, for example, in the semiconductor sensor technology. Recently, the preliminary measurements of electrical conductivity were carried out on ZnO films deposited on silicon wafers by the RF-magnetron sputtering [6]. In this paper, we present further research of the electrical properties of ZnO films and perform a more detailed analysis of their parameters. Thus, the purpose of this work is to produce ZnO thin film structures and to study the electrical conductivity by determining their characteristics.

2. Experiment
The samples were Cu/ZnO/Si heterostructures. A ZnO film was deposited on a *p*-type silicon substrate (100) by atomic layer deposition. Argon (99.998 %) was used as a purging and carrier gas. The process temperature was 150°C. Diethylzinc and deionized water were used as precursor. The pulse length for diethylzinc and water was 200 ms, the purge time was 1 s. Figure 1 presents the results of structure and composition investigations of ZnO films.

The thickness of the ZnO film on a Si substrate is equal to 200 nm. Copper circular electrodes with a diameter of 3 mm were sputtered on the film surface through a shadow mask by thermal evaporation. According to the data obtained by scanning electron microscopy, the films have a crystalline structure.
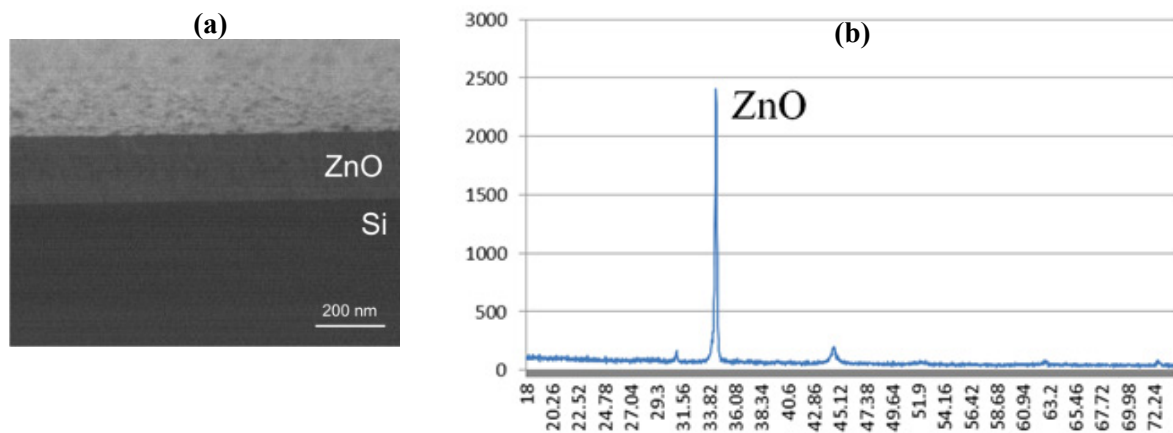
Figure 1. Scanning electron microscopy image (a) and x-ray diffraction spectra (b) of a ZnO film formed on a silicon substrate.

The conductivity measurements were carried out by the current-voltage (*I-V*) characteristics method using an immittance meter E7-20. This device allows applying the bias to the sample from −40 to +40 V in steps of 0.02 V in the range of −4 to +4 V and 0.2 V at higher voltages. The voltage applied to the film is accepted positive if a positive potential is applied to the top electrode, and vice versa.

3. Results and discussion

I-V characteristics were obtained at room temperature to determine the conductivity mechanisms of the ZnO film deposited on Si. Figure 2 shows a typical dependence of electric current on a voltage applied to a Cu/ZnO/Si heterosructure.

In case of a positive potential applied to the top electrode, the electric current increases nonlinearly with the voltage except for the initial voltage range from 0 to 4 V. Moreover, the *I-V* characteristics are asymmetric and show weak rectifying properties with a rectification coefficient $K = 5$ (at $U = 2.5$ V) and the non-ideality coefficient of diode structure $n = 5.55$. On the basis of the linear section of the current-voltage characteristics, the electrical conductivity of a ZnO layer is calculated according to the relation:

$$\sigma = \frac{I \cdot d}{U \cdot S}, \tag{1}$$

where I is the current, d is the layer thickness, U is the applied voltage, S is the top electrode area. The value of the ZnO layer conductivity is equal to $4.5 \cdot 10^{-6}$ $\Omega^{-1}\text{m}^{-1}$.

Rectifying properties of the structure are associated with the presence of the potential barrier at the interface ZnO/Si. The asymmetry of the current-voltage characteristics of the barrier is typical for such structures. A dependence of current on voltage in these structures is caused by a change in the number of majority carriers taking part in charge transfer processes. A role of external voltage is to change the number of free carriers transferred from one to another part of the barrier structure. However, in this case, the barrier height is not uniform over the boundary of the zinc oxide due to the presence of surface states. Thus, current may leak through the lowered barrier areas. This explains the nonideal rectifying properties and the presence of the high reverse current.
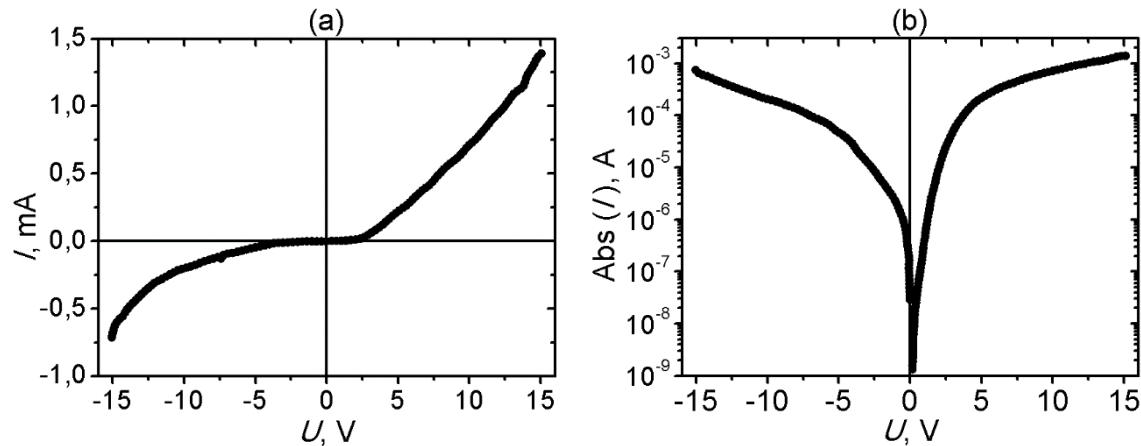
Figure 2. Current-voltage characteristics of the ZnO film deposited on silicon on: (a) a linear scale, (b) a semi-logarithmic scale.

It should be noted that the forward and reverse trajectories of current-voltage characteristics are practically the same that indicates the absence of the resistive switching effect in the present ZnO films.

To explain the observed behavior of the current-voltage characteristics, several conduction mechanisms of film structures can be considered. It is well known [7, 8] that the leakage current in dielectric or semiconductor films may be attributed to several conduction mechanisms including the Poole-Frenkel emission, the Schottky emission, the Fowler-Nordheim tunnelling and the space charge limited current (SCLC).

In the voltage region from 5 to 14 V, the current observed in the ZnO films increases in accordance with a quadratic law, i.e. $I \sim U^2$ (figure 3). Such a behavior can be ascribed to the SCLC mechanism. For the space charge limited current in the case when a single discrete trap level exists in the band gap, the current density is given by equation (2) [9]:

$$j = \frac{9}{8}\varepsilon\varepsilon_0\mu\frac{U^2}{d^3}, \tag{2}$$

where j is the current density, ε is the material dielectric permittivity, ε_0 is the vacuum dielectric permittivity, μ is the carrier mobility.

As previously mentioned, the linear segment of the I-V characteristics preceded the quadratic region that corresponded to the SCLC model. At low voltages, the injection level is low and a concentration of injected carriers does not exceed the concentration of free equilibrium carriers. In this case, not all traps are filled, so the current obeys Ohm's law

$$j = en_0\mu\frac{U}{d}, \tag{3}$$

where n_0 is the concentration of free equilibrium carriers, e is the elementary charge.

The transition from linear to quadratic parts of I-V characteristics occurs when the concentration of injected carriers becomes comparable to the concentration of free equilibrium carriers. In this case, all traps are filled and do not affect the flow of space-charge-limited current. On the one hand, the voltage of the transition from linear to quadratic parts of I-V characteristics is given by

$$U_{tr} = \frac{4}{3}\frac{en_0d^2}{\varepsilon}. \tag{4}$$

On the other hand, the voltage is

$$U = \frac{eN_t d^2}{2\varepsilon}, \tag{5}$$

where N_t is the concentration of trapping levels.

Taking into account that the dielectric permittivity of ZnO was equal to 8.5 and the transition occurred at a voltage of ≈ 4.5 V, we used equation (5) and found the concentration of trapping levels $N_t \approx 5.3 \cdot 10^{18}$ cm^{-3}. We determined $n_0 \approx 2 \cdot 10^7$ cm^{-3} from equation (4). We substituted the n_0 concentration in equation (3) and found $\mu \approx 190$ cm^2/(V·s).
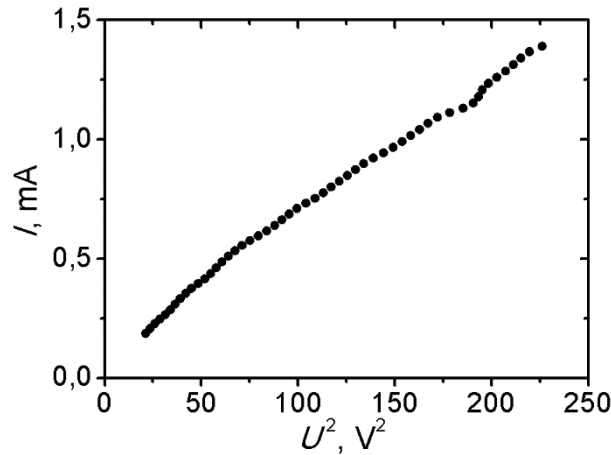

Figure 3. Dependence of the current on square voltage applied to Cu/ZnO/Si.

For the negative voltage part of *I-V* characteristics, the most probable mechanism of electrical conductivity is the Poole-Frenkel emission. The current dependence on the applied voltage modulus is shown in figure 4 in the Poole-Frenkel representation.
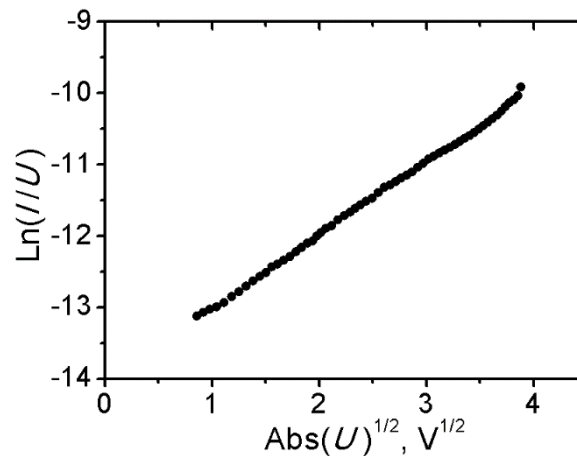

Figure 4 Current–voltage characteristic of the structure on the base of ZnO according to the Poole-Frenkel mechanism.

This dependence is nearly linear that gives evidence of the Poole-Frenkel emission contribution to the current. According to this mechanism, a field applied to the sample changes the shape of potential barriers for charge carriers between atoms in the lattice. It leads to an increase in the number of free carriers in the sample due to overcoming the potential barrier.

4. Conclusion

The ZnO thin films were fabricated on the *p*-type silicon substrate (100) by atomic layer deposition to make the ZnO/Si heterogeneous structure. For the electrical response measurements, Cu-electrodes were deposited on a free film surface by means of thermal sputtering. To determine conductivity mechanisms of the ZnO films, the current-voltage characteristics were measured at room temperature. It was found that the *I-V* characteristics are asymmetric and nonlinear with the exception of the initial voltage range from 0 to 4 V. The conductivity of the ZnO layer calculated for a weak field is equal to $4.5 \cdot 10^{-6}$ $\Omega^{-1} m^{-1}$. The main mechanisms of the electrical conductivity in the Cu/ZnO/Si heterostructure were revealed. For the positive voltage region ($\sim 4 - 15$ V), the electric current obeyed the quadratic law that corresponded to the space-charge-limited current model. At the negative potential applied to the top Cu-electrode, the *I-V* characteristic is described by the Poole-Frenkel emission. On the basis of the *I-V* characteristic measurements, the concentration of free equilibrium carriers, the concentration of trapping levels and the free carrier mobility were calculated. The value of n_0 is nearly equal to $2 \cdot 10^7$ cm^{-3}, N_t is about $5.3 \cdot 10^{18}$ cm^{-3} and $\mu \approx 190$ $cm^2/(V \cdot s)$. To study the properties associated with the presence of the barrier in the heterostructure, additional measurements are needed such as capacitance-voltage characteristics, dielectric dispersion etc. This is the subject of further research.

References

[1]   Lee J, Jung J, Kang M and Koo S 2013 *J. Nanosci. Nanotech.* **13** 7033
[2]   Xua T, Wub G, Zhang B and Haob Y 2003 *Sensors and Actuators A: Physical* **104** 61
[3]   Heiland G and Ibach H 1966 *Solid State Comm.* **4** 353
[4]   Shao C, Chang Y and LongY 2014 *Sensors and Actuators B: Chemical* **204** 667
[5]   Coleman V and Jagadish C 2006 *Zinc Oxide Bulk, Thin Films and Nanostructures* ed Jagadish C and Pearton S (Amsterdam: Elsiever)
[6]   Egorov A V, Solnyshkin A V, Bogomolov A A, Silibin M V, Gavrilov S A 2012 *Proceedings of the International Conference* "INTERMATIC-2102" Moscow: MIREA, *in Russian* **2** 164
[7]   Solnyshkin A V, Troshkin A S, Bogomolov A A, Raevski I P, Sandjiev D N and Shonov V Yu 2009 *Int. Ferroelectrics* **106** 61
[8]   Kamenshchikov M V, Solnyshkin A V, Bogomolov A A and Pronin I P 2011 *Physics of the Solid State* **53** 2080
[9]   Kao K S and Hwang W 1981 *Electrical Transport in Solids* (Oxford, New York, Toronto: Pergamon Press)