THERMOELECTRICALLY CHARACTERISTICS OF ZNO: AL FILMS OBTAINED BY THERMAL AND MAGNETRON SPUTTERING

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In this paper, we consider the temperature dependences of the electrical characteristics of a conducting and transparent ZnO: Al film obtained by two methods: thermal sputtering in vacuum and magnetron ion-plasma sputtering. The temperature dependences of the resistivity, the concentration, the mobility of the charge carriers, and the field dependence of the magnetic resistance were measured. It is shown that the aluminum doping of a ZnO film by various sputtering processes leads to a change in the transfer of charge carriers by defects in impurity atoms and the grain boundary, in addition, with increasing impurity concentration, the resistivity of the film remained constant. Measurement by the Seebeck effect showed that the magnetic resistance for all the samples under study is negative and decreases with increasing temperature and an increase in the level of doping. Therefore, the ZnO: Al film is electrically conductive. The absolute value of the magnetic resistance does not exceed 2.5%. Thus, films obtained by magnetron sputtering can be used as a film as an antireflection and stable coating for textured and silicon nanowires.

Keywords: Temperature dependences, magnetron sputtering, Seebeck effect, magnetic resistance, resistivity, concentration, mobility, Hall effect, impurity.

Introduction

As is known, a transparent conductive film based on zinc oxide ZnO, doped aluminum of various concentrations is of great interest among researchers, because of the wide-gap semiconductor material for the Schottky barrier in solar cells [1] and in light-emitting diodes [2]. Previous work [3] [4], as a TCO film on the surface of silicon nanofilms, a transparent antireflective coating based on ZnO: Al, the so-called (AZO) film was used. We studied zinc oxide films made by thermal and magnetron evaporation using a different level of Al doping.

In this paper, we will consider the electrical characteristics of the ZnO: Al film, such as the resistivity, charge carrier concentration and mobility with the help of the Hall measurement, and also their temperature dependence.

1. Experimental description

The investigated zinc oxide films were obtained using two methods: thermal evaporation and magnetron sputtering. In thermal evaporation, we investigated films of zinc oxide 0.3 μm thick. Different levels of doping with aluminum were used [5]. The temperature dependences of the resistivity and magnetic resistance of Hall effects were experimentally investigated in the range from 2 to 300 K in magnetic fields up to 8 Tesla. The investigated films had n-type conductivity in the range from $2 \times 10^4$ to $2 \times 10^5$ S / m, depending on the level of doping.

The temperature dependence of the conductivity, electron density, and mobility is weak [6] (values at helium and room temperatures differ by less than 5%). At low temperatures, the films exhibit a small negative magnetoresistive effect (a limitation of up to -2.5% at 4 K), which decreases with an increase in the level of aluminum doping and temperature.

Glass was used as substrates and single-crystal silicon plates obtained by the Czochralski method, coated with a layer of SiO₂. The synthesis was carried out at 225°C. The target was pure
zinc oxide or zinc with the addition of aluminum (in a ratio of 30: 1 or 20: 1). The thickness of the deposited films was (0.29 ± 0.1) μm. In this work, the temperature dependence of the resistivity from 0 °K to room temperature 300°K will be considered for the deposition of a ZnO film with doped and without doped aluminum impurities. The experiments will be carried out on quartz glass at different temperatures, magnetron-sputtering methods (vacuum unit VUP-4) and thermal evaporation in vacuum (VUP-5). We will also determine other parameters, in particular, the concentration and mobility of charge carriers in a transparent ZnO film of Al doped and without it.

2. Results and discussion

As shown by our experiments, the highest resistance is observed in films deposited by magnetron sputtering without doping with aluminum (Fig. 1). It can be seen that at low temperatures the resistivity of films synthesized by magnetron sputtering is approximately five orders of magnitude higher than for films obtained by thermal evaporation; at room temperature, this difference is reduced to 3 orders of magnitude.

![Fig.1. Temperature dependence of the resistance for doped zinc oxide films: (a) magnetron sputtering, (b) thermal evaporation](image)

An increase in temperature from 4 to 300 K is accompanied by a decrease in the resistivity of films deposited by magnetron sputtering by a factor of 100, while the resistance of films obtained by thermal evaporation changes by less than 10% in the entire investigated temperature range. Thus, films obtained by magnetron sputtering show significantly higher resistivity and sensitivity to temperature, while thermally sputtered films have a low resistivity, which depends little on temperature.

Aluminum doping of zinc oxide films obtained by thermal evaporation reduces their resistance by approximately an order of magnitude and reduces the sensitivity of the resistance to temperatures (Figure 2). For thermally sputtered in vacuum films, the resistivity decreases monotonically with the level of doping by aluminum atoms. At the same time, the effect of doping on the resistivity of films obtained by magnetron sputtering was not observed.

As can be seen from Fig. 3, the magnetic resistance (MR) for all the films under study is negative and decreases with increasing temperature. The absolute value of MR does not exceed 2.5% and decreases with increasing doping level. Experiments have shown that the Hall coefficient is negative throughout the temperature range studied, which indicates the n-type conductivity for all types of films.
Fig. 2. Temperature dependence of the resistivity for films of zinc oxide doped with aluminum and obtained by thermal evaporation.

Fig. 3. Field dependence of the magnetic resistance of zinc oxide films at different temperatures for samples: (a) obtained by magnetron sputtering, without doping; (b) thermally deposited, without doping; (c) thermally deposited, doped at 1:30; (g) thermally deposited, doped at 1:20.

The electron concentration N was estimated from the results of measurements of the Hall effect taking into account the fact that the Hall factor is close to unity:
\[ R_H = \frac{1}{eN}, \]  

(1)

where \( e \) is the electron charge.

The temperature dependence of the electron concentration calculated from the expression for \( R_H(T) \) by the formula (1) is shown in Fig. 4. As can be seen from the figure, regardless of the type of films, the electron concentration is practically constant in the investigated temperature range (4-300 K). This fact is naturally explained by the high level of doping of the films, which leads to the formation of an overlap of the impurity band with the zinc oxide conductivity zone.

The Hall mobility of electrons \( \mu \) can be expressed as follows

\[ \frac{1}{\rho} = \mu Ne, \]  

(2)

\[ \rho = \frac{R_H}{\mu}. \]  

(3)

The temperature dependences of the mobility calculated by the formula (3) are shown in Fig. 5. It can be seen from the figure that the mobility in films obtained by the magnetron sputtering method is much lower and depends strongly on temperature.

Fig.4. Temperature dependence of the electron concentration in zinc oxide films: (a) obtained by magnetron sputtering, undoped; (b) thermally deposited, undoped; (c) thermally deposited, alloyed with aluminum at 1: 30; (d) thermally deposited, alloyed with Al at 1: 20.
Fig. 5. Temperature dependence of electron mobility in zinc oxide films obtained by:
(a) magnetron sputtering, undoped; (b) thermal evaporation, undoped; (c) thermal evaporation, doped with aluminum at 1: 30; (d) thermal evaporation, alloyed with aluminum at 1:20

The increase in electron mobility with temperature for films obtained by magnetron sputtering, as well as its insignificant change with temperature for films obtained by thermal evaporation, is probably due to the fact that electron scattering is mainly determined by defects (impurity atoms, grain boundaries), then As scattering by phonons plays an insignificant role.

Conclusion

As a result of the experimental measurement of the temperature dependence of the resistive characteristic of the ZnO film obtained by magnetron sputtering, the resistivity and sensitivity to temperature are much higher, while the thermally sputtered films have a low resistivity that is weakly temperature dependent. For thermally sputtered in vacuum films, the resistivity decreases monotonically with the level of doping by aluminum atoms. At the same time, the effect of doping on the resistivity of films obtained by magnetron sputtering was not observed.

In addition, the magnetic resistance (MR) for all the films under study is negative and decreases with increasing temperature. The absolute value of MR does not exceed 2.5% and decreases with increasing doping level. The temperature dependence of the electron concentration, calculated from the expression for \( R_H(T) \), showed that, regardless of the type of films, the electron concentration is practically constant in the investigated temperature range (from 4 to 300 K). This
fact is naturally explained by the high level of doping of the films, which leads to the formation of an overlap of the impurity band with the zinc oxide conductivity zone.

The temperature dependences of mobility of charge carriers calculated from the Hollow measurement and obtained by the magnetron sputtering method are much lower, and strongly depend on temperature. An increase in the mobility of electrons with temperature is due to the scattering of electrons in the volume of the film by defects (impurity atoms, grain boundaries), whereas scattering by phonons plays an insignificant role.

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REFERENCES


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