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INFLUENCE OF PREPARATION CONDITIONS ON THE ELECTRICAL CONDUCTIVITY OF GaN THIN FILMS

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ABSTRACT

Background. Gallium nitride (GaN) is a promising material for the developing of LEDs, lasers, ultraviolet detectors, and high-frequency transistors. The properties of GaN significantly depend on the type of defects and the parameters of thin film production. In particular, substrates, buffer layers, sputtering temperature, and partial pressure of the working gas have an impact. This led to studying the electrical conductivity of GaN thin films obtained by radio-frequency (RF) ion-plasma sputtering under different technological conditions.

Materials and Methods. GaN films with a thickness of 0.3..1.0 μm were deposited on sapphire and quartz substrates, with/without buffer layers of $MgAl_2O_4$ or ZnO. RF sputtering was carried out in a N_2 atmosphere at a pressure of 5×10^{-3} .. 5×10^{-2} Torr and temperatures of 400-650 °C. The structure was analyzed by X-ray diffraction, and the electrical conductivity was determined in the temperature range of 100..450 K by the method of two-point contacts using ohmic carbon contacts.

Results and Discussion. It was found that GaN films have a polycrystalline structure with different orientations, depending on the type of substrate and the presence of a buffer layer. The resistivity varies from 10^5 to 10^{10} ohm-cm. High-impedance samples are characterized by a thermal activation energy of 0.34 eV, and the donor type of conductivity in the films has been established. In the films on quartz substrates with a ZnO sublayer, lower activation energy values (0.004..0.05 eV) were recorded, indicating a different conduction mechanism. The conductivity of GaN increases with increasing substrate temperature and decreasing partial pressure of N_2 in the sputtering atmosphere, which leads to the formation of nitrogen vacancies – the main donor defects.

Conclusion. The conditions of their preparation largely determine the electrical conductivity of GaN thin films. The most important factors are the type of substrate, buffer layers, sputtering temperature, and gas pressure in the sputtering atmosphere. The obtained dependences of the change in the electrical conductivity of GaN thin films on the partial pressure of the working nitrogen gas and the temperature of the substrate during sputtering show that the most likely defects of the donor type in gallium nitride are nitrogen vacancies V_N.

Keywords: thin films, gallium nitride, electrical conductivity, donor defects.

INTRODUCTION

Gallium nitride (GaN) and compounds based on it are a promising material for the production of semiconductor light diodes and lasers emitting in the blue and ultraviolet regions of the spectrum, as well as ultraviolet radiation detectors [1-3]. Powerful high-speed transistors based on GaN, which are superior to their silicon counterparts in terms of operating frequencies and power densities, have also become widely used [4, 5]. At the same time, an important task is to obtain GaN thin films with the required composition of intrinsic defects. Intrinsic atomic defects largely determine the optical and electrical properties of gallium nitride, such as absorption and emission spectra, type and value of electrical conductivity. Another important task is to find optimal substrates that match GaN in terms of lattice parameter and thermal expansion coefficient.

The basic task in the development of technology for electronic or optoelectronic devices based on gallium nitride is to obtain homogeneous thin film layers of high quality. One of the ways to solve this problem is to grow epitaxial GaN layers on sapphire substrates by using thin buffer layers. In this case, it is necessary to match the characteristics of the deposited buffer layer with the modes of high-temperature growth of the main GaN layer. The properties of the buffer layer in such a system are determined only indirectly, through the properties of the thin film layer deposited on it. On the other hand, the optimization of the deposition regime of the corresponding films is possible only if a suitable buffer layer is available. On suboptimal buffer layers, GaN thin films have poor quality regardless of the deposition method. Therefore, the procedure for determining the optimal parameters for depositing gallium nitride thin films is complex and complex. For this purpose, the electrical conductivity of thin films on sapphire and quartz substrates with buffer sublayers of $MgAl_2O_4$ and ZnO was studied in this work. The films were obtained by radio-frequency (RF) ion-plasma sputtering, which is considered optimal to produce semiconductor and dielectric films [6].

MATERIALS AND METHODS

Thin films of gallium nitride with a thickness of 0.3..1.0 μm were obtained by RF ion-plasma sputtering on sapphire substrates (Al_2O_3) and quartz substrates (SiO_2). In some cases, buffer sublayers of $MgAl_2O_4$ and ZnO were used. The RF sputtering was carried out in a nitrogen atmosphere at pressures from 5×10^{-3} to 5×10^{-2} Torr. The target for sputtering was metallic Ga. The temperature of the substrates during sputtering varied from 400 to 650 °C, and the RF discharge power was from 100 to 150 W.

The structure and phase composition of the obtained films were studied by X-ray diffraction analysis (Shimadzu XDR-600). The characteristic diffractograms of the obtained films are shown in Fig.1. The analysis of the diffractograms shows that the structure of the obtained films corresponds to the hexagonal crystal structure of GaN wurtzite.

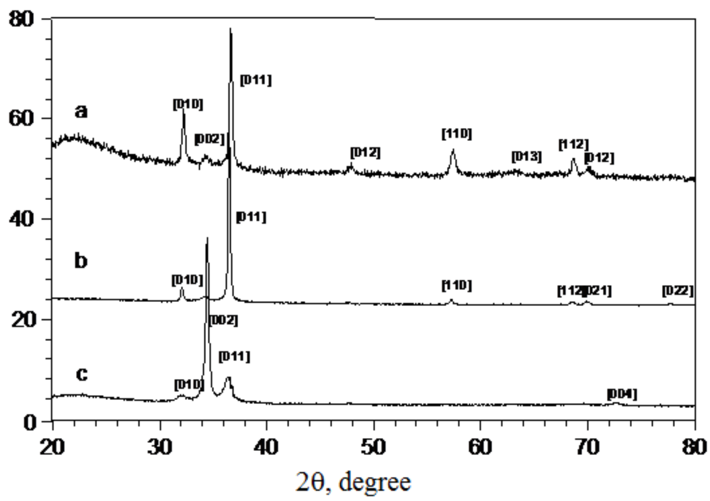

Fig. 1. X-ray diffractograms of GaN thin films deposited on a sapphire substrate with a $MgAl_2O_4$ sublayer (a), on a pure sapphire substrate (b), and on a quartz substrate with a ZnO sublayer (c).

The results obtained indicate that the films are characterized by a polycrystalline structure. At the same time, when deposited on sapphire substrates, the predominant orientation of the films is observed in the (011) plane, and when deposited on quartz substrates, in the (002) plane. The use of a sublayer does not change this pattern but leads to a slight increase in the orientation of the films in other planes. For example, the use of a $MgAl_2O_4$ sublayer on sapphire substrates leads to a slight increase in the orientation of GaN films in the (010), (110), and (112) planes (Fig. 1, *a* and *b*).

Conduction currents in the temperature range of 100-450 K were measured on an automated setup. An electric voltage of 10-100 V was applied to two-point contacts with a diameter of 1 mm, which were applied at a distance of 1 mm. When measuring the current flowing in GaN thin films, the main requirement is the use of ohmic, non-rectifying contacts that do not create additional barriers at the interface. The ohmic contact to the studied films is created by materials that, when displaced directly, provide electron injection into the film and have an output work of ~4.5 eV. The polycrystalline carbon (Aquadag) we use meets these requirements and has been used in numerous publications to study diamond, garnet, spinel, and other high-resistivity samples [7-9].

RESULTS AND DISCUSSION

The studies show that GaN thin films deposited on sapphire substrates, depending on the preparation conditions, have a rather high resistivity from 10^5 to 10^{10} Ohm×cm. The temperature dependence of the electrical conductivity for such films in the coordinates ln$I$= f(1000/T) is shown in Fig. 2 (curve 1). As can be seen from the dependence, the experimental results obtained in the temperature range of 100..450 K are well approximated by a linear dependence. This situation indicates the activation nature of the electrical conductivity of these films and allows us to determine the energy of thermal activation of electrical conductivity, which is E_T = 0.34 eV, from the slope of the line.

It should be noted that the decrease in resistivity in gallium nitride films deposited on a ZnO sublayer may be due to the interaction between the sublayer material and the film material. In particular, on the one hand, the possible diffusion of excess Ga^{3+} from the film can lead to an increase in the conductivity of ZnO sublayers, and on the other hand, the

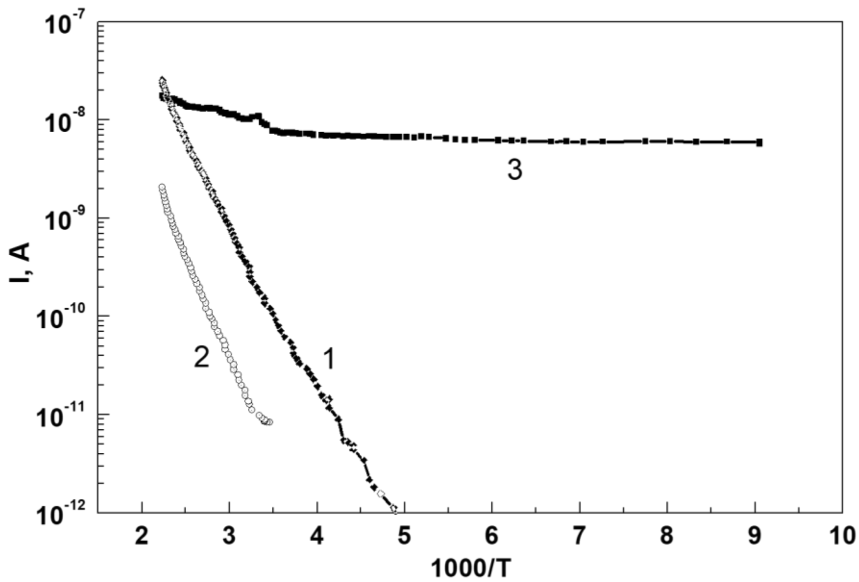

Fig. 2. Temperature dependence of the electrical conductivity of GaN thin films deposited on a sapphire substrate without a buffer sublayer (1) and with a MgAl2O4 sublayer (2), films on quartz substrates with a ZnO sublayer (3).

diffusion of Zn^{2+} into GaN can lead to an increase in conductivity due to acceptor centers in gallium nitride. It is known that GaN can have both impurity electronic and hole conductivity [10-12]. The donors in GaN films can be substitutional atoms Si_{Ga}, Ge_{Ga}, O_N, interstitial gallium Ga_i, and nitrogen vacancies V_N. Such donors create low energy levels in the band gap near the bottom of the conduction band.

For a more detailed study of the electrical conductivity of GaN thin films, the effect of the technological conditions for obtaining the films, in particular the pressure of the working gas N_2 and the temperature of the substrate, was investigated. The dependence of the specific electrical conductivity of GaN thin films on sapphire substrates on the partial pressure of N_2 in the working chamber during the sputtering of the films is shown in Fig. 3.

As can be seen from Fig. 3, with an increase in the partial pressure of nitrogen from 0.006 to 0.03 Torr, the value of the specific electrical conductivity of GaN films decreases by several orders of magnitude from 10^{-5} to 10^{-9} $Ohm^{-1}cm^{-1}$. Such a significant dependence of the electrical conductivity of the obtained films on the partial pressure of N_2 indicates that the most dominant charge carriers in gallium nitride thin films under RF ion-plasma sputtering are nitrogen vacancies, which play the role of electron donors. A decrease in the partial pressure of nitrogen in the sputtering atmosphere leads to an increase in nitrogen vacancies in the films and, accordingly, to an increase in the donor electrical conductivity.

It has been found that the temperature of the substrate largely determines the electrical conductivity of gallium nitride thin films during film deposition. The results of measuring the specific electrical conductivity at 300 K for GaN films obtained at different substrate temperatures are shown in Fig. 4. As can be seen from the figure, an increase in the substrate temperature from 500°C to 630°C almost linearly leads to a significant increase in the specific electrical conductivity from 10^{-9} to 10^{-5} $Ohm^{-1}cm^{-1}$. Such a significant increase in the specific electrical conductivity with increasing substrate temperature indicates the formation of electrically active point defects.

The obtained dependences of the specific electrical conductivity σ of the obtained gallium nitride thin films on the partial pressure of nitrogen and the temperature of the substrate can be explained if we consider that the most likely donor-type defects in these

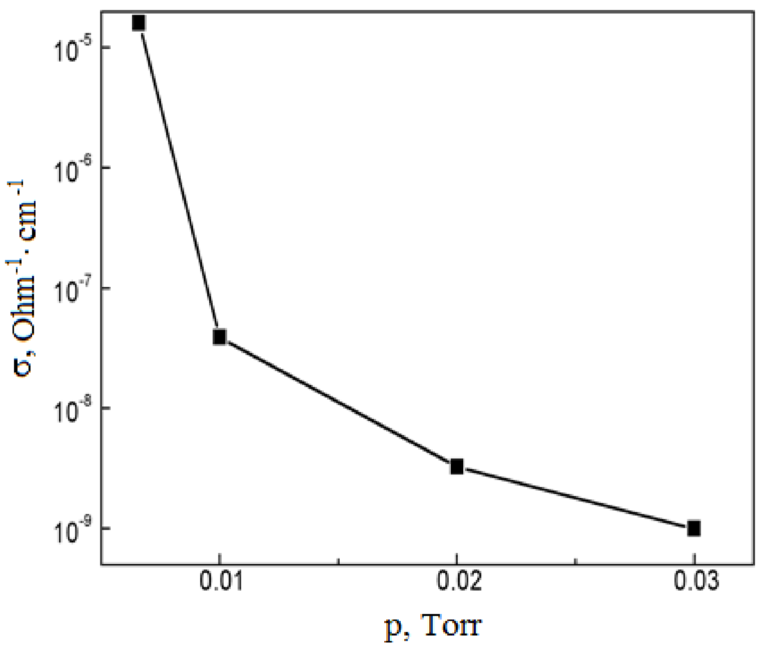

Fig. 3. Dependence of the specific electrical conductivity (at 300 K) of GaN thin films on sapphire substrates on the partial pressure of N2 during film sputtering (substrate temperature at 800 K).

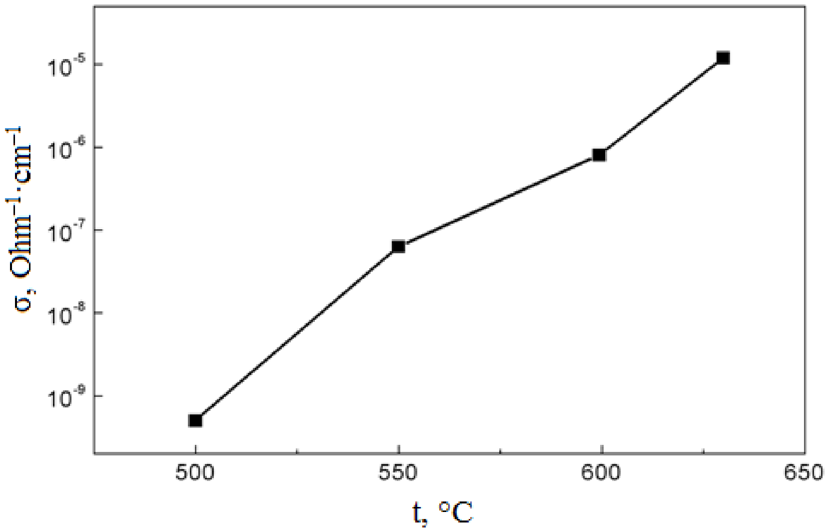

Fig. 4. Dependence of the specific electrical conductivity of GaN thin films on sapphire substrates at 300 K on the temperature of the substrate during sputtering.

films are nitrogen vacancies V_N. According to [13], epitaxial GaN films have an electron concentration that can vary from 10^{17} to 10^{20} cm^{-3} depending on the preparation conditions. The experimental values of the energy levels of donor centers in pure GaN films and crystals are conditionally divided into 4 ranges. This is the range of very low energies of 0.01..0.04 eV, intermediate energies of about 0.1 eV, and average energies of 0.2..0.4 eV [14-17], as well as in the region of 0.33..0.39 eV for high-resistivity gallium nitride samples [17]. Such differences in the values of the activation energy of conductivity in gallium nitride are associated with the existence of several types of donor defects and the interaction of point defects with the formation of defect complexes. Our experimental values for the thermal activation energy E_T = 0.34 eV, for GaN thin films on sapphire substrates are consistent with the literature data [17] for the energy depth of donor levels in high-resistivity gallium nitride films. For GaN films on quartz substrates with a ZnO sublayer, the determined values of E_T = 0.004 eV and E_T = 0.05 eV fall into the range of very low energies and are characteristic of low-impedance gallium nitride samples.

In [14, 18, 19], theoretical calculations of the formation energies of the most probable point defects in GaN were performed. According to these calculations, when thin films are deposited in a low-pressure nitrogen atmosphere, there are two dominant types of defects: nitrogen vacancies V_N, and inter-nodal gallium Ga_i. It is shown that the energy of nitrogen vacancy formation is several times lower than the energy of gallium vacancy formation and other point defects. Therefore, given that the energy of formation of nitrogen vacancies in gallium nitride is much lower than the energy of formation of other point defects, the most likely donor defects in this semiconductor should be considered anionic vacancies V_N.

CONCLUSION

The studies have shown that the RF ion-plasma sputtering of GaN thin films in a nitrogen atmosphere produces films whose electrical conductivity is determined by the type of substrate, the presence of a buffer sublayer, the pressure of the working gas, and the temperature of the substrate during sputtering. The thermal activation energy of electrical conductivity of high-resistance films is 0.34 eV. For low-impedance gallium nitride films, this value ranges from 0.004 eV to 0.05 eV, depending on the temperature range. The obtained dependences of the change in the electrical conductivity of GaN thin films on the partial pressure of the working gas nitrogen and the temperature of the substrate during sputtering show that the most likely donor-type defects in gallium nitride are nitrogen vacancies V_N.

COMPLIANCE WITH ETHICAL STANDARDS

The authors declare that they have no competing interests.

AUTHOR CONTRIBUTIONS

Conceptualization, [O.B., I.Ku., M.P., I.M., I.Ko., Zh. T.]; methodology, [O.B., I.Ku.]; validation, [M.P., I.M.]; investigation, [I.Ku., M.P., I.M., I.Ko.]; writing – original draft preparation, [M.P., I.M., I.Ko., Zh.T.]; writing – review and editing, [O.B., I.Ku.]; visualization, [O.B., I.Ku., M.P., I.M., I.Ko., Zh.T.].

All authors have read and agreed to the published version of the manuscript.

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ВПЛИВ УМОВ ОДЕРЖАННЯ НА ЕЛЕКТРОПРОВІДНІСТЬ ТОНКИХ ПЛІВОК GaN

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АНОТАЦІЯ

Вступ. Нітрид галію (GaN) — перспективний матеріал для створення світлодіодів, лазерів, детекторів ультрафіолету й високочастотних транзисторів. Властивості GaN суттєво залежать від типу дефектів та параметрів одержання тонких плівок. Зокрема, вплив мають підкладки, буферні шари, температура напилення та парціальний тиск робочого газу. Це обумовило дослідження електропровідності тонких плівок GaN, отриманих методом високочастотного (ВЧ) іонно-плазмового розпилення за різних технологічних умов.

Матеріали та методи. Плівки GaN товщиною 0,3–1,0 мкм осаджували на сапфірові та кварцові підкладки, з/без буферних шарів $MgAl_2O_4$ або ZnO. ВЧ розпилення проводилось в атмосфері N_2 при тиску $5\cdot10^{-3}$–$5\cdot10^{-2}$ Торр та температурах 400–650 °C. Структуру аналізували за допомогою Х-променевої дифракції, електропровідність визначали в температурному діапазоні 100–450 К методом двоточкових контактів з використанням омічних контактів із вуглецю.

Результати. Встановлено, що GaN-плівки мають полікристалічну структуру з різною орієнтацією, залежно від типу підкладки та наявності буферного шару. Питомий опір змінюється від 10^5 до 10^{10} Ом·см. Високоомні зразки характеризуються енергією термічної активації 0,34 еВ та встановлено донорний тип провідності у плівках. У плівках на кварцових підкладках із підшаром ZnO зафіксовано менші значення енергії активації (0,004..0,05 еВ), що свідчить про інший механізм провідності. Провідність GaN зростає при збільшенні температури підкладки і зниженні парціального тиску N_2 в розпилювальній атмосфері, що приводить до утворення вакансій азоту — основних донорних дефектів.

Висновки. Електропровідність тонких плівок GaN значною мірою визначається умовами їх одержання. Найбільший вплив мають тип підкладки, буферні шари, температура напилення та тиск газу в розпилювальній атмосфері. Отримані залежності зміни величини електропровідності тонких плівок GaN залежно від парціального тиску робочого газу азоту та температури підкладки при напиленні показують, що найбільш імовірними дефектами донорного типу в нітриді галію є вакансії азоту V_N.

Ключові слова: тонкі плівки, нітрид галію, електропровідність, дефекти донорного типу.

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