Transparent for microwave electrodes for applications in nonlinear devices based on ferroelectric ceramic

A G Alltynnikov¹, R A Platonov¹, R A Komlev¹, A V Tumarkin¹, A M Sosunov¹, P M Trofimov¹ and A A Tcymbaliuk¹

¹Saint Petersburg Electrotechnical University "LETI", Saint Petersburg 197022, Russia

*Corresponding author e-mail: raplatonov@etu.ru

Abstract. This article provides the results of an experimental investigation of characteristics of thin-films transparent electrodes deposited on ferroelectric ceramic plates. The experimental investigation of parameters of ZnO and resistive alloy of Si-Ti-Ce deposited on ferroelectric BSTO plates was performed. The influence of transparent electrode deposition on properties of ferroelectric ceramic based on a solid solution of barium-strontium titanate was investigated. The measured low insertion loss of the Si-Ti-Ce thin-film deposited on BSTO ceramic substrate allows considering this material as the perspective for the transparent for microwaves electrodes realization.

1. Introduction

The term "transparent electrodes" is commonly used in optical electronics [1,2]. Such electrodes are produced based on a thin-film or thick-film material that meets two main requirements: relatively high electrical conductivity and high transparency in the optical wavelength range. The ITO (indium tin oxide) is the typical material to manufacture such electrodes [3]. However, in addition to optical electronics, the term "transparent electrodes" is applicable in microwave techniques. As a rule, such electrodes are used to construct lens devices and tunable nonlinear elements [4,5]. These electrodes must be transparent for radiation in the microwave frequency range, and therefore, in contrast to electrodes used in optoelectronics, they should demonstrate a high resistance. ZnO material is suitable for transparent (in the microwave and optical ranges) electrode applications. Another perspective class of materials for microwave transparent electrode applications is resistive alloys. These materials are usually used for thin-film resistors manufacturing. Notice that these materials are not optically transparent but produce thin-film coating with high sheet resistance values up to 10 MOhm. Such high resistance values allow suggesting small insertion loss induced by electrode layers made of resistive alloys. Of particular interest is choosing the material of transparent electrodes and the technology of their deposition for use in devices based on nonlinear materials. The transparent electrode deposition procedure directly to the surface of a nonlinear material can change its nonlinear properties, which is a negative effect. This work is devoted to the investigation of the influence of transparent electrode deposition on properties of ferroelectric ceramic based on a solid solution of barium-strontium titanate [6,7]. The ZnO and resistive alloy based on Si-Ti-Ce were chosen as a material for transparent electrodes [8]. Despite a great number of articles devoted to preparing thin-films on different substrates, the compatibles of ZnO and resistive alloys with BSTO ceramic plates are unclear from the point of view of transparency in the microwave and millimeter-wave ranges. This article provides the results of an experimental investigation of characteristics of thin-films transparent electrodes deposited on ferroelectric ceramic plates. The thin
films of ZnO and resistive alloy of Si-Ti-Ce were deposited on ferroelectric BSTO plates for experimental investigations.

2. Samples preparation

The resistive alloys such as silicides of chromium, nickel, iron as well as binary and ternary systems on their base are traditionally used for deposition of resistive thin films. A wide range of thin-film surface resistivity is achieved by the variation of Si concentration from 15% to 95% in the resistive alloys. Such variation allows for providing control over permissible dissipation power and temperature coefficient of resistance of deposited resistive thin films.

The methods of thermal vacuum resistive evaporation and ion-plasma sputtering can be used for deposition of resistive thin films. The use of magnetron sputtering has the several advantages over resistive evaporation. The significantly higher adhesion of films to the substrate, associated with the energy of the condensing particles and additional activation of the substrate surface caused by the plasma. The higher accuracy in controlling the thin film's thickness (including films thinner than 100 nm). It is quite important as far as the film's thickness determines its resistance.

In this work, the properties of two transparent electrode materials deposited on a ferroelectric substrate were investigated. The first of them is the resistive alloy containing Si-76%, Ti-20%, and Ce-4% (Si-Ti-Ce). The deposition of thin-film Si-Ti-Ce transparent electrodes was performed by the vacuum magnetron sputtering method. The residual pressure in the chamber did not exceed 6×10^{-4} Pa. High-purity argon at a pressure of 0.1 Pa was used as a plasma-forming gas. The sputtering of the target was carried out at a constant current at a power of 0.25 kW. The deposition temperature was kept constant at 300 °C. The growth rate was 50 nm/min. As a result of the experiments, the samples of high-resistive films with a thickness of 40-100 nm were deposited on BSTO ceramic plates.

The second material for transparent electrodes was ZnO. ZnO films were deposited by the method of high-frequency ion-plasma magnetron sputtering of ceramic ZnO target. The residual pressure in the chamber did not exceed 10^{-3} Pa. High-purity oxygen at a pressure of 1 Pa was used as a plasma-forming gas. The target-to-substrate distance was 25 mm. The substrate holder is heated from below by a system of heaters made of nichrome. Plates of polycrystalline aluminum oxide and bulk ceramics BaSrTiO₃ were used as substrates. The substrate temperature was varied in the range of 600-800°C.

3. Experimental results

The first stage of measurements was devoted to observing the transparent electrode's sheet resistance and its variation with time. Measurements show that Si-Ti-Ce thin-films deposited on BSTO ceramic plates demonstrate higher sheet resistance (~1.5 MOhm/square) than the ZnO films (~1.1 MOhm/square). Each sample was tested to observe the thin-film coating quality, which was observed that sheet resistance variation per sample does not exceed 10% along the sample surface. This result shows a high homogeneity of the obtained thin films on BSTO substrates. During the experimental investigations, the change of sheet resistance value in time was observed in the case of the ZnO thin-films deposited on the BSTO ceramic plate. At the same time, Si-Ti-Ce did not demonstrate such a negative tendency. The obtained experimental results of changing sheet resistance in time are presented in Figure 1.
The comparison of thin films’ sheet resistance in time for ZnO and resistive alloy of Si-Ti-Ce. 

One can see that the ZnO thin-film electrode demonstrates a significant decrease in sheet resistance value for 10 days after manufacturing. The decrease down to 85% of the value measured in the first day was observed for ZnO thin films. In comparison, Si-based resistive alloy thin film is more stable in time and demonstrates a slightly lower deviation of sheet resistance per ceramic sample (see error bars in Figure 1). Note that observed sheet resistance degradation with time is not typical for ZnO thin-films widely used in electronic devices. In our case, the nature of this effect can be explained by oxygen-containing compounds in the substrate. Films of ZnO were deposited on BSTO ceramic at relatively high temperatures, and this could lead to formation of an oxygen-defective interface BSTO/ZnO. This defective surface can be the reason of sheet resistance variation with time. It should be noted that the presented explanation is hypothetic, and the observed effect requires additional investigation.

Since the substrate is a BSTO ceramic, it is important to investigate the influence of transparent electrode deposition procedure on ceramic nonlinear properties. The characterization of BSTO ceramic nonlinearity is provided by capacitance-voltage (CV) dependency measurements of BSTO capacitors under dc control voltages. Because of the demonstrated degradation of the sheet resistance of ZnO thin-films, further measurements were focused on Si-Ti-Ce thin-film electrodes. The ceramic parallel plate capacitors were manufactured on the base of BSTO with deposited Si-Ti-Ce film and without it. Estimation of the Si-Ti-Ce film deposition procedure influence on ceramic properties was performed by comparison of CV characteristics of manufactured capacitors. The silver ink was used to form the electrodes of parallel plate capacitors. Experimental results of measured CV characteristics at 1 MHz are presented in Figure 2. In accordance with measured CV-characteristics presented in Figure 2, one can conclude that the Si-Ti-Ce deposition procedure influence on nonlinear properties of BSTO ceramic is negligibly low.
Figure 2 Measured CV-characteristics of parallel plate capacitor based on BSTO with Si-Ti-Ce film and without it.

One of the main parameters of transparent electrodes from the microwave application point of view is the insertion loss value per electrode layer. This parameter was measured at the frequency of 30 GHz by the method of two-port waveguide measurements. The results show that the insertion loss of deposited Si-Ti-Ce thin-film is about 0.1-0.3 dB.

4. Conclusion

The ZnO and Si-Ti-Ce materials were considered as transparent for electrodes for microwave applications in nonlinear devices based on BSTO ceramic. The samples of both thin-film materials were prepared on the BSTO ceramic plates to measure sheet resistance and insertion loss values. The decreasing of sheet resistance in time of ZnO electrodes deposited on the BSTO ceramic was observed in contrast to Si-Ti-Ce material. The experimental investigations showed that there is no degradation of nonlinear properties of BSTO ceramic after deposition of Si-Ti-Ce thin-film. The measured low insertion loss of the Si-Ti-Ce thin-film deposited on BSTO ceramic substrate allows considering this material as the perspective for the transparent for microwaves electrodes realization.

Acknowledgments

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References

[1] Ellmer K 2012 Nature Photonics 6 12 p 809
[2] Cao W et al. 2014 Journal of Photonics for energy 4 1 p 040990
[3] Hofmann A I, Cloutet E, Hadzioannou G 2018 Advanced Electronic Materials 4 10 p 1700412
[4] Platonov R, Altynnikov A, Kozyrev A 2020 Coatings 10 2 p 180
[5] Scheele P et al. 2007 IEEE transactions on microwave theory and techniques 55 2 p 383
[6] Nadaud K, Borderon C, Gillard R, Fourn E, Renoud R, Gundel H W 2015 Thin Solid Films 591 p 90
[7] Altynnikov A G et al. 2019 Technical Physics Letters 45 6 p 540
[8] Kolesnikova I G, Korotkov V G, Kuz'mich Y V 2016 Russian Metallurgy (Metally) 2016 5 p 472