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## Investigation of conductivity mechanisms in ferroelectics based on the doped barium titanite

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Abstract. The results of studies of the current-voltage characteristics of ceramic structures based on manganese doped barium titanate were presented. It was found that the electrical conductivity of ceramic samples based on barium titanate at high field strengths is determined by the Schottky emission. Moreover, the addition of manganese increases the resistance of the samples.

Ferroelectrics with a perovskite structure are widely used both in developing of microwave microelectronic devices [1] and switching devices for accelerating technology [2] due to the nonlinear field strength and temperature dependences of the permittivity, and possibility of varying properties with change the stoichiometric composition. Thin ferroelectric films are not compatible to use in high power tunable devices. Therefore, it requires the use of bulk ceramics. Ceramics based on barium titanate (BTO) have despite the high tunability but have high dielectric permeability, high dielectric losses and significant dielectric hysteresis in the microwave region [3]. Synthesis of heterophase materials with BTO doped with manganese can reduce dielectric permeability, integral losses and hysteresis of the material while maintaining controllability of the dielectric constant. The doping of Mn leads to the influence of the magnetic field on the composite structure, i.e. the properties of multiferroics appear [4].

Leakage currents and charge carriers transport mechanisms are an important aspect of the use of manganese (Mn) doped ferroelectrics ceramic for high-power microwave devices and for the fabrication of multiferroic materials. Therefore, the aim of this work was to study the electrical conductivity of BTO ceramics with different manganese content.

This paper presents the results of studies of the current-voltage characteristics (CVC) of the planeparallel Ag/BTO/Ag structures with a Mn impurity concentration variation from 0 to 10 mol%, namely: 1) pure BaTiO<sub>3</sub> ceramics (BTO); 2) BaTiO<sub>3</sub> with 5 mol. % Mn (BTO5M), 3) BaTiO<sub>3</sub> with 10 mol. % Mn (BTO10M). Samples were prepared using ceramic technology [5] at a sintering temperature about 1350 °C. The composition and content of the manganese-containing impurity were chosen in such a way that at the sintering temperature it is necessary to ensure a weak interaction of the components with each other and to preserve the ferromagnetic properties of the Mn impurity. Silver-electrodes was fabricated on the plane-parallel polished surfaces of the ceramic discs, the disks thickness varied within h = 0.25 - 1.35 mm, the discs area was S = 25-120 mm<sup>2</sup>. The CVC measurements were made at room temperature (T = 283 K).

The current-voltage dependences shows linear and nonlinear regions. The regions of ohmic conductivity corresponded to weak electric fields (up to  $E \sim 5 \cdot 10^4 \text{ V/m}$ ), for which the sample resistance

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was estimated. The resistance was decreased with an increase the Mn impurity concentration from  $\sim 10^{12} \Omega$  for pure BTO to  $\sim 10^{11} \Omega$  for BTO10M samples (at sample thickness 1mm).

Figure 1 shows the current-voltage characteristics for BTO10M samples at different thicknesses. Nonlinear regions on the CV characteristic was started at field intensities ~  $5 \cdot 10^6$  V/m. The nonlinearity of the CVC in dielectrics and ferroelectrics is usually associated with the following mechanisms: Schottky emission, field emission and the Frenkel-Pool effect.



**Figure 1.** The current-voltage characteristics for different thickness samples BTO10M.

The conduction mechanism due to Schottky emission is described by the following expression [6]:

$$J = AT^{2} \exp\left(\frac{1}{kT} \sqrt{\frac{q^{3}E}{4\pi\varepsilon_{0}\varepsilon_{r}}}\right),$$
(1)

where A is the Richardson constant, T is the absolute temperature, k is the Boltzmann constant, q is the electron charge, E is the field strength,  $\varepsilon_0$  is the electric constant  $\varepsilon_r$  is the relative permittivity.

Figure 2 shows the BTO10M samples CVC of different thickness in  $\ln(J/T^2)$  from  $E^{1/2}$  coordinates. All the curves are well approximated by linear dependences at field intensities  $\geq 5 \cdot 10^6$  V/m. That presupposes the presence of a conduction mechanism associated with Schottky emission, while the values of the Pearson agreement ( $\chi$ ) were sufficiently high (in Figure 2, values of the Pearson test).

Thermal ionization of traps and its ionization by an electric field depending on concentration of the traps and their depth affects on current throw thin dielectric layers. The thermal ejection of trapped electrons into the conduction band, known as the Frenkel-Pool effect, gives the following expression for the current-voltage characteristic [6]:

$$J \approx \exp \frac{1}{kT} \left[ \left( 57.7 \frac{eV}{\varepsilon S} \right) - E_l \right], \tag{2}$$

where  $E_l$  is the depth of traps, and S is the gap between the electrodes. The presence of a linear section on CVC at ln (J/E<sup>2</sup>) = f(1/E) coordinates indicates the presence of Frenkel-Pool effect. All the experimental CVCs shown in Figure 1 were rearranged in the ln (J/E<sup>2</sup>) = f(1/E) coordinates and showed



**Figure 2.** The current-voltage characteristics for different thickness samples BTO10M in Schottky coordinates.

the dependencies close to linear. However, the values of the Pearson criterion for the approximation corresponding to the Frenkel-Pool effect were lower than for the Schottky emission.

Field emission from the traps begins to be observed with a large electric field strength or with heating during the CVC recording. It consists of trapping through a potential barrier. This mechanism is described by the following expression:

$$J = C_t E^2 \exp\left(-\frac{\alpha \varphi_t^{3/2}}{E}\right), \ \alpha = \frac{8\pi \sqrt{2m_{ox}q}}{3h},$$
(3)

where  $m_{ox}$  is the effective mass of the electron in the oxide,  $C_t$  is the concentration density of the traps, and  $\varphi_t$  is the depth of the traps. In fact, the expression describing the process of field emission from traps, like the well-known Fowler-Nordheim tunneling [7]. The linear approximations of the CVC of the samples in the coordinates  $\ln(J/E^2) = f(1/E)$  were also characterized by Pearson's criterion.

In figure 3 shows the values of Pearson's agreement criterion for all the approximations for different thicknesses of the samples. At large sample thicknesses, the values of  $\chi$  for the Schottky emission and for the field emission practically coincide, and the values of  $\chi$  for the Frenkel-Pool effect are much smaller. Consequently, for large sample thicknesses, electrical conductivity is mainly determined by Schottky emission and field emission. At sample thicknesses  $\leq 0.5$  mm, the mechanism associated with Schottky emission is most likely to predominate. At a thickness of 0.25 mm, the Frenkel-Pool effect can also make a significant contribution to the electrical conductivity.

A plane-parallel structure is the series connection between the volume sample resistance and the resistance of the contact region. Therefore, for resistances of unit area we can write the expression:

$$R \cdot S = \frac{h}{\sigma} + R_k \,, \tag{4}$$

where *R* is the measured resistance of the sample,  $R_k$  is the unit contact area resistance, *S* is the sample area, *h* is the sample thickness, and  $\sigma$  is the electrical conductivity of the ceramic.



Figure 3. Dependence of the Pearson's agreement criterion for various conduction mechanisms on the thickness of

Figure 4 shows the experimental dependence  $R \cdot S(h)$ . After its approximation by the method of least squares, the extrapolation resulting straight line to the ordinate axis gives us the resistance value of the unit area of the contact area  $R_k$  and the angle value of the resulting line gives us the volume sample resistance. The obtained value of the electrical conductivity of the ceramic BTO10M is  $\sigma = (2\pm 2) \cdot 10^{-10}$   $\Omega^{-1} \cdot m^{-1}$ , the value of the contact area resistance is  $R_k = (9\pm 9) \cdot 10^5 \Omega \cdot m^2$ .



**Figure 4.** Resistance per unit area for structures Ag/BTO10M/Ag different thicknesses.

Thus, the CV characteristics of the all studied samples has the initial sections corresponding to the fulfillment of Ohm's law, and for field intensities ~  $5 \cdot 10^6$  V/m, Schottky emission and field emission from traps contribute significantly to the through conductivity currents. When the thickness of the samples is lower than 0.5 mm, the main contribution to the conductivity is made by contact phenomena due to Schottky emission.

The estimation of the electrical conductivity of the ceramics BTO10M is  $\sigma = (2\pm 2) \cdot 10^{-10} \Omega^{-1} \cdot m^{-1}$ , and the resistance of the unit of the contact area Ag/BTO10M/Ag is  $R_k = (9\pm 9) \cdot 10^5 \Omega \cdot m^2$ .

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