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Tensoelectric properties of $(Bi_xSb_{1-x})_2Te_3$ films under the influence of a microwave field

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This paper presents the results of an experimental study of the tens metric and dielectric properties of polycrystalline $(Bi_xSb_{1-x})_2Te_3$ films in the temperature range of 280−480 K and at microwave frequencies. The temperature dependences of the specific conductivity, impedance, and permittivity under the action of uniaxial static deformation are analyzed. The deformation phenomena detected in polycrystalline films under the action of a microwave field are qualitatively interpreted based on the effective medium theory.

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1. Introduction

As is known, solid solutions of bismuth and antimony chalcogenides are effective thermoelectric [1-6] and tensoelectric [7,8] materials. They are used not only for thermoelectric cooling and heating, thermoelectric conversion of thermal energy, and also as a topological insulator, effectively shielding electromagnetic interference, but also for the creation of fatigue damage accumulation sensors. At present, sufficient attention is paid to establishing the patterns of structure formation, phase composition of polycrystalline films based on the solid solution $(Bi_xSb_{1-x})_2Te_3$ and studying their mechanisms of electrical conductivity and dielectric properties. Thus, in works [6, 8] the “metallic” and “semiconductor” nature of electrical conductivity was discovered depending on temperature. In our previous work [7] it was shown that the resistance of a porous polycrystalline $(Bi_{0,3}Sb_{0.7})_2Te_3$ film grown by thermal vacuum evaporation at a substrate temperature $T_{sub} \leq 363$ K, sharply decreases near the threshold frequency $\omega_0 \approx 10^5$ Hz of alternating current to the resistance values of dense films with $T_{sub} \approx 423$ K. After exposure to $N \approx 10^5$ cycles of mechanical deformation with an amplitude of $\varepsilon = \pm 1 \cdot 10^{-3}$ relative units, the film resistance increases by 1.5 times, and the critical value ω0, determined by the film inhomogeneity, decreases by almost 10^2 times. Unlike bulk crystals and polycrystals, in thin semiconductor polycrystalline films, as the film thickness decreases, their properties change due to a noticeable contribution from the surface and near-surface conductivity of carriers due to adsorption, diffusion of impurities, and surface electron states. The influence of the transition layer between the film and the substrate, thickness inhomogeneity caused by the production technology, and quantum size effects at very small film thicknesses are significantly manifested. It is also necessary to take into account the quality of the film obtained, its porosity, the presence of multiphase inclusions, numerous point and extended structural defects, etc. This paper presents the results of an experimental study of the tensometric and dielectric properties of polycrystalline $(Bi_xSb_{1-x})_2Te_3$ films in the temperature range of 280−480 K and at microwave frequencies in order to detect the mechanisms of the influence of inhomogeneities on the working parameters of the samples. The temperature dependences of the specific conductivity, impedance and permittivity under the action of uniaxial static deformation are considered. The deformation phenomena detected in polycrystalline films under the action of a microwave field are qualitatively analyzed based on the effective medium theory.

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2. Experimental technique

Impedance is a value characterizing the electrical properties of polycrystalline films at a certain frequency of an alternating field ($\tilde{Z} = R + iR_{LC}$, where R – and $R_{LC} = \omega L - (1/\omega C)$ are the active and reactive resistances, ω=2πν is the cyclic frequency, L – and C are the inductance and electrical capacity of the film as a whole). Therefore, measuring this value with high accuracy is of great importance for recording the strain gauge characteristics of samples in an alternating field. In particular, studying the impedance of inhomogeneous films based on the solid solution $(Bi_xSb_{1-x})_2Te_3$ depending on the level of relative tensile strain $\varepsilon = \Delta\ell / \ell_0$ allows us to determine the complex coefficient of strain sensitivity at microwave frequencies

$$\tilde{K} = K_R + iK_{LC} \ , \tag{1}$$

where

$$K_R = \frac{\Delta R}{R_0 \cdot \varepsilon} \quad , \quad K_{LC} = \frac{\Delta R_{LC}}{R_{LC}^0 \cdot \varepsilon} \ , \tag{2}$$

strain gauge coefficients for active and reactive resistances $\Delta R = R(\varepsilon) - R_0$, a $R(\varepsilon)$ and R_0 - active resistances of the film with and without deformation.

We have obtained and investigated films from the solid solution $(Bi_{0,3}Sb_{0.7})_2Te_3$. The choice of this composition was due to the fact that the data available in the literature [7-10] indicate that in the system of solid solutions $(Bi_xSb_{1-x})_2Te_3$ the composition $(Bi_{0,3}Sb_{0.7})_2Te_3$ has the highest strain sensitivity. Polycrystalline films with a thickness of 3-4 μm and dimensions of 5×30 mm on a polyamide substrate were produced by the method of thermal evaporation in a vacuum $P = 10^{-2}\,Pa$ at a substrate temperature of $T_n = 363\,K$ and a deposition rate of $W \approx 200$ Å/c [9,10].

The studied polycrystalline films $(Bi_{0,3}Sb_{0.7})_2Te_3$ were placed in a measuring cell with a waveguide section, which was equipped with a heater allowing the temperature to be changed from 77 to 480 K and the microwave field frequency in the range of 10^8 -10^{10} Hz. The impedance spectroscopy measurement technique was used, as in [11-12].

Specific conductivity was defined as,

$$\sigma = \frac{\ell}{b \cdot d} \frac{I}{V_\sigma} \quad , \tag{3}$$

where ℓ, b, d, are the length, width and thickness of the sample, respectively, I -is the current through the sample, V_σ- is the voltage drop between the electrodes for measuring electrical conductivity.

The deformation was created by bending the cantilever plate of a conventional equal resistance deformation device made of titanium alloy. Film samples glued to it with a special adhesive were subjected to one-sided stretching deformation by bending the substrate. The magnitude of the relative deformation ε was calculated using the well-known expression [13]

$$\varepsilon = \frac{4d}{\ell^2} \cdot y \ , \tag{4}$$

where y- is the deflection of the free end of the plate at the point of application of the force. The strain values varied in the range from ε=0 to ε = + 6.10^{-3} rel. units. The study of dielectric permittivity under mechanical deformation was carried out mainly at a fixed frequency ω=5,1·10^{10} Hz.

3. Results and discussion

The temperature dependence of the conductivity of $(Bi_{0,3}Sb_{0.7})_2Te_3$ films is shown in fig. 1. it is evident from the figure that the real part of the electrical conductivity increases with increasing temperature. And the imaginary part of the electrical conductivity decreases and then increases with increasing temperature up to 350 K. The electrical conductivity of $(Bi_{0,3}Sb_{0.7})_2Te_3$ films at direct current weakly depends on temperature in the studied range. It should also be noted that the real part of the electrical conductivity measured by the microwave method differs from the electrical conductivity measured at direct current. Naturally, the conductivity of polycrystalline layers of $(Bi_{0,3}Sb_{0.7})_2Te_3$ at microwave frequencies is significantly determined by the electronic and structural heterogeneities of the layers. Comparison of the conductivity measured in the microwave field and at direct current confirms the presence of these heterogeneities in the studied samples.

Polycrystalline films $(Bi_{0,3}Sb_{0.7})_2Te_3$ are a heterogeneous system consisting of individual crystalline grains with different local electrical conductivities at a sharp crystallite boundary. Such heterogeneous systems have high permittivity, measuring which one can calculate the effective and true values of the electrophysical parameters of the components.

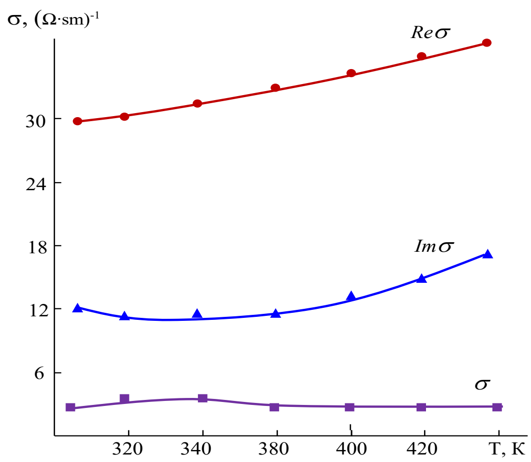

Fig. 1. Dependence of specific electrical conductivity of films $(Bi_{0,3}Sb_{0.7})_2Te_3$ on temperature.

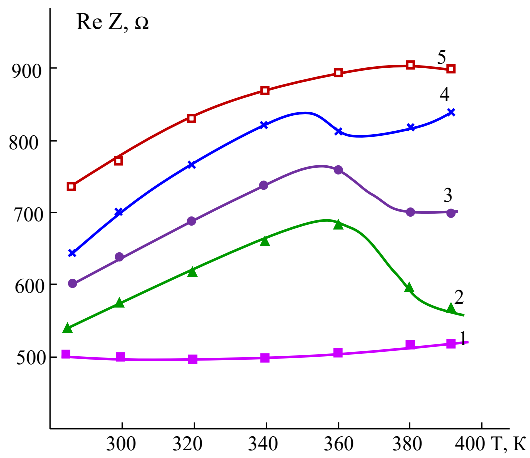

Fig. 2. Dependence of the real part of the impedance $(Bi_{0,3}Sb_{0.7})_2Te_3$ on temperature at different levels of tensile strain $\varepsilon 10^3$: 1-0, 2-1.12; 3-2.27; 4-3.4; 5-4.52.

The issue of shunting barrier regions at the boundaries of microcrystals by capacitances at high frequencies has been discussed in the literature [14]. However, this idea has not been verified in the study of the strain properties of polycrystalline films $(Bi_xSb_{1-x})_2Te_3$ in the microwave range.

It turned out that the microwave method has a number of advantages in measuring some quantities in comparison with measurements on direct current. On microwaves, some bulk properties of the semiconductor have a number of interesting features related to the fact that the period of microwave oscillations has a value of the order of the collision frequency of charge carriers. Microwave measurements make it possible to obtain additional information about their properties that cannot be established in measurements on direct current, and allow one to determine the real and imaginary parts of the impedance, with the help of which it was possible to judge their contribution to the strain sensitivity of films.

Figure 2 shows the dependence of the real part of the impedance of n-type polycrystalline $(Bi_{0,3}Sb_{0.7})_2Te_3$ films on temperature at different levels of relative tensile strain. It is evident that with an increase in the level of relative strain and temperature, the impedance of the films increases and then decreases significantly rapidly, especially at low strain levels (the real part of the impedance of undeformed films depends weakly on temperature).

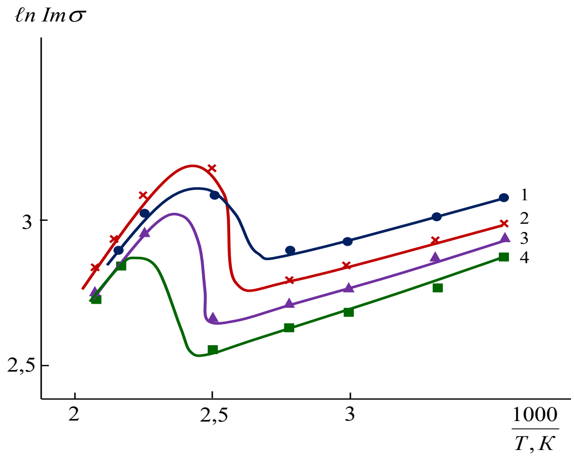

Fig. 3. Dependence of $\ell n\ Im\sigma$ on 1/T of $(Bi_{0,3}Sb_{0.7})_2Te_3$ films at different levels of relative deformation $\varepsilon.10^3$: 1-0.2; -1.4; 3 - 2.8; 4 -4.2.

The dependence of the imaginary part of the electrical conductivity of n-type inhomogeneous $(Bi_{0,3}Sb_{0.7})_2Te_3$ films on temperature at different levels of uniaxial tensile strain in coordinates $\ell n\,\mathrm{Im}\,\sigma - 1/T$ is shown in fig. 3, where it is evident that the imaginary part of the electrical conductivity of $(Bi_{0,3}Sb_{0.7})_2Te_3$ films decreases with increasing temperature and level of uniaxial tensile strain. This pattern is violated at T>360 K, whereby an increase in the imaginary part of the electrical conductivity of the films is first observed with increasing temperature, and then its decrease.

The sensitivity of the impedance of $(Bi_{0,3}Sb_{0.7})_2Te_3$ films in the microwave range to external mechanical deformations allows us to judge the strain properties of these films. Studies of the strain effect in polycrystalline films using the microwave method allow us to determine the mechanism of high strain sensitivity. Analyzing the experimental data, we can note that abnormally high strain sensitivity is not observed in all polycrystalline film samples.

In the simplest case, film samples used as strain gauges must consist of two phases whose conductivity differs in certain respects. The effective conductivity of a heterogeneous medium is a function of the conductivity of the components and their volume fraction. With an increase in the volume fraction of the second phase, the effective conductivity will change according to a certain pattern (from σ_1 to σ_2). If a smooth change in the effective conductivity is observed with an increase in the volume fraction of the second phase, these samples will have a strain property, but not high sensitivity. If an abrupt change in the effective conductivity is observed in a certain interval of the volume fraction of the second phase, then the strain sensitivity in these samples can reach a maximum value. In this narrow region of the volume fraction of the second phase, the fluctuation in the polarizability of crystallites under the action of uniaxial tensile strain reaches a maximum value.

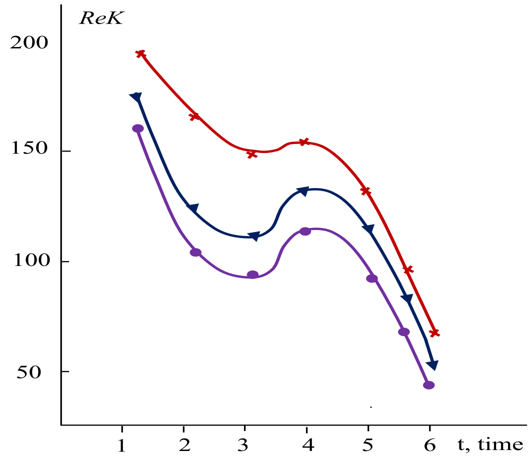

Fig. 4. Dependence of the real part of the strain-sensitivity coefficient $(Bi_{0,3}Sb_{0.7})_2Te_3$ *on the annealing time at relative deformation levels ε10³: 1-3.3; 2-4.5; 3-5.6.*

The contactless microwave method serves as a powerful tool for studying the complex strain sensitivity of $(Bi_xSb_{1-x})_2Te_3$ films heat-treated in air. Therefore, the regularities of the change in the real and imaginary parts of the strain sensitivity of $(Bi_{0,3}Sb_{0.7})_2Te_3$ films at different temperatures and annealing times were studied.

Figure 4 shows the dependence of *ReK* on the annealing time at different relative deformations, when the annealing temperature was constant and equal to t_{refl}=500 K. It is evident that with an increase in the annealing time and relative tensile deformation, *ReK* decreases. In the range from 2.5 to 4.5 hours, the linear dependence of *ReK* on the annealing time is disrupted.

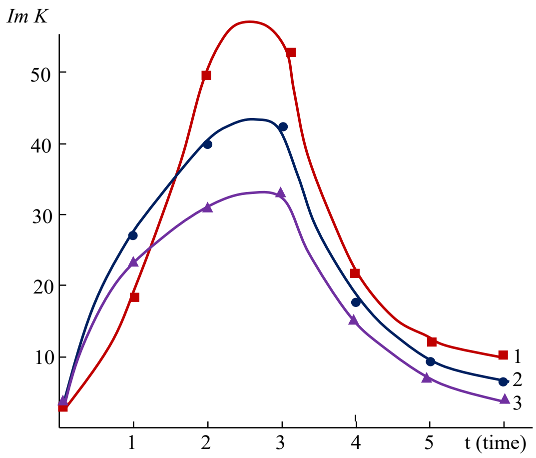

Fig. 5. Dependence of the imaginary part of the strain-sensitivity coefficient $(Bi_{0,3}Sb_{0.7})_2Te_3$ *on the annealing time at different levels of relative deformation ε10³: 1-1.12, 2-2.27, 3-3.30.*

Figure 5 shows the dependence of *ImK* on the annealing time at different strain levels. It is evident that with increasing annealing time, *ImK* increases and after reaching its maximum, it decreases exponentially. *ImK* is positive over the entire range of annealing times.

It is assumed that the change in the strain-sensitivity coefficient in the microwave range during heat treatment in air is due to a change in charge carriers in the $(Bi_xSb_{1-x})_2Te_3$films. The microwave method allows recording changes in the concentration of charge carriers.

Deformation phenomena in polycrystalline films under the influence of a microwave field can be considered on the basis of the effective medium theory [14]. During heat treatment of films in air, the concentration of current carriers in the boundary regions of crystallites will change. When the crystallite sizes are commensurate with the diffusion length of oxygen, the medium mainly consists of the second phase. Anomalous strain effects cannot be expected in such films. If the films do not contain various types of inhomogeneities, i.e. they are perfect films, then strain sensitivity in these samples can be minimal.

The increase in the strain-sensitivity coefficient is due to the fluctuation of the current density in the spatial coordinates of the films. Due to thermal inhomogeneities, the emerging surface conductivity of the crystallites, by nature, differs significantly from the bulk conductivity. The surface conductivity is a function of the diffusion length of oxygen. If the volume of these layers is equal to the volume of the base material, then the effective complex conductivity changes significantly quickly, and the fluctuation of the current density reaches its maximum value. Observation of the maximum value of the imaginary part of the strain-sensitivity coefficient with increasing annealing time does not contradict the above explanations.

It should be noted that the dielectric properties of bismuth tellurides are characterized by large static and optical permittivity ε_s, ε_∞; low frequencies of transverse optical phonons [15]. The permittivity of Bi_2Te_3 films has a high value (50÷100) arb. units and at a frequency of 10^{10} Hz in the range from 193 to 400 K depends weakly on temperature. The authors of [14] found that the permittivity ε in $Pb_{1-x}Sn_xTe$ solid solutions reaches 10^4, and the temperature dependence of $\varepsilon(T)$, described by the Curie-Weiss law, brings this substance closer to ferroelectrics.

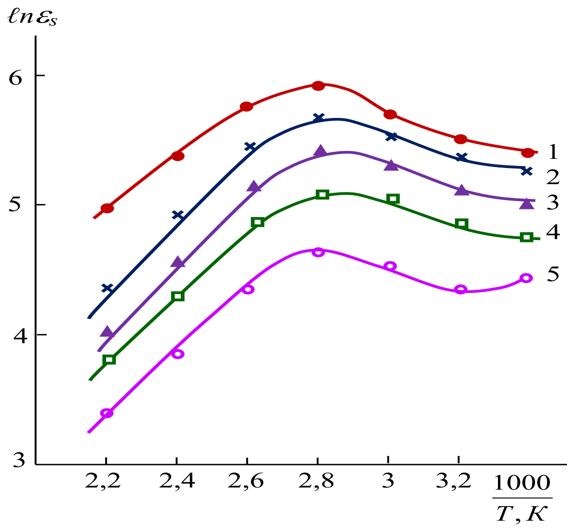

Fig. 6. Dependence of the real part of the permittivity $(Bi_{0,3}Sb_{0.7})_2Te_3$on temperature at different levels of deformation $\varepsilon 10^3$: 1-0; 2-0.68; 3-1.36; 4-2.04; 5-3.4.

Fig. 6 shows the dependence of $\ell n\ \varepsilon_s$ on the inverse temperature at different deformations. It is evident that the permittivity of $(Bi_{0,3}Sb_{0.7})_2Te_3$ films has an activation character. With an increase in temperature to 400 K, the permittivity increases and then decreases, i.e. there is a flat maximum.

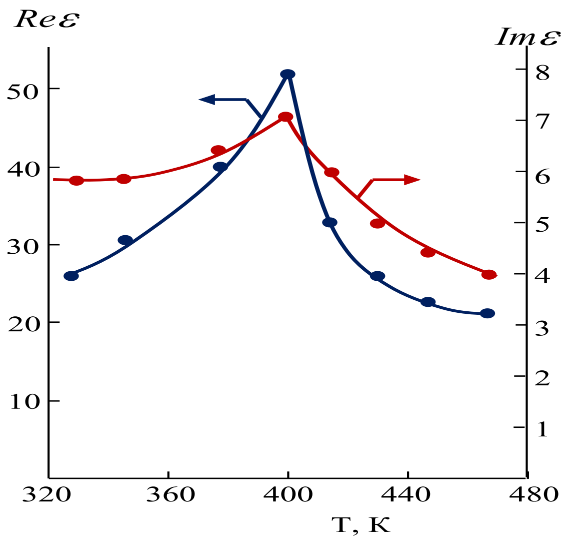

Fig. 7. Dependence of the dielectric constant $(Bi_{0,3}Sb_{0.7})_2Te_3$ on temperature.

This pattern is also observed when studying the imaginary part of the permittivity of $(Bi_{0,3}Sb_{0.7})_2Te_3$ films as a function of the inverse temperature at different levels of tensile strain in the microwave range. Its absolute value is an order of magnitude smaller than the real part of the permittivity. It was experimentally found that the real part of the permittivity of polycrystalline $(Bi_{0,3}Sb_{0.7})_2Te_3$ films reaches 5.3×10^4 units, and the imaginary part is 7.1×10^3 units (fig. 7). These data, in our opinion, are caused by the interaction of the film with oxygen under the combined effect of temperature and deformation. In this case, a certain contribution is made by localized states formed at the boundaries of crystallites. The dynamics of local inhomogeneities and inclusions of the second phase with increasing temperature leads to a significant change in the permittivity of polycrystalline films $(Bi_{0,3}Sb_{0.7})_2Te_3$. The intensity of microwave fields takes a random value due to the inhomogeneity of the material under study. This is due to the fact that in each section of the films under study, components of a heterogeneous medium with different electrical and dielectric properties can be located. In this case, the fluctuation of electric fields in spatially random areas differs from zero. Uniaxial deformation, acting on an inhomogeneous medium, leads to a redistribution of localized carriers and a change in the concentration of free current carriers. Consequently, the electronic part of the permittivity decreases and qualitatively coincides with the experimental results.

The high value of the permittivity may be due to the formation of heterogeneous systems during condensation of films in a vacuum. In polycrystalline lead selenide films, the real part of the impedance is responsible for the conductive region, and the imaginary part is responsible for the non-conductive region. This conclusion can be reached based on the theory of effective media, which sufficiently describes the electrical conductivity and permittivity of conducting and non-conducting systems if $R_e\sigma$ and $I_m\sigma$ differ from each other by no more than an order of magnitude. If the heterogeneous medium under study consists of two phases, then in the case of $\sigma_1 >> \sigma_2$, the theory of effective media gives correct results not for all values of the volume fraction of the low-resistance component, but only starting with those that correspond to the percolation threshold $\Theta = \Theta_c$ [16].

Due to the fixed frequency, the dependence of the permittivity and electrical conductivity of $(Bi_{0,3}Sb_{0.7})_2Te_3$ on the inverse temperature coincides with the results of work [17]. The experimental results indicate that by changing the volume fractions of inclusions, it is possible to get into the percolation threshold region.

4. Conclusion

Thus, the study of the temperature dependence of the complex permittivity of solid solutions $(Bi_{0,3}Sb_{0.7})_2Te_3$ revealed the presence of features in the form of a change in the sign of the temperature coefficient of permittivity, as well as in the form of impedance maxima and minima in the temperature range of 350–450 K and frequency ω=5,1·10^{10}Hz. These features are interconnected and are due to independent resonance mechanisms: the coincidence of the frequency of the measuring alternating voltage with the frequency of electron jumps during the implementation of hopping conductivity; the proximity of the period of the measuring signal to the time of formation of the barrier layer at the grain boundary; the approach of the frequency of the external microwave field to the frequency of the structural elements of the sample. The obtained results can be used to create functional devices based on solid solutions $(Bi_xSb_{1-x})_2Te_3$.

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