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THERMOELECTRIC PROPERTIES OF THIN FILMS OF BISMUTH AND BISMUTH-ANTIMONY SOLID SOLUTION

The temperature dependences of the resistivity and thermoEMF were investigated by the method that excludes the occurrence of external strain in the film-substrate system, and the thermoelectric power factor was calculated in the temperature range of 77 to 300K for bismuth-antimony solid solution films on substrates with different thermal expansion coefficients. It has been found that to get the maximum thermoEMF, the ratio of the crystallite size and film thickness is critically important, which is due to the different confinement of electrons and holes mobility by the surface and crystallite boundaries. The maximum thermoEMF and power factor correspond to thick block films of $Bi_{0.88}Sb_{0.12}$ *on mica.The research was supported by the Ministry of Education of the Russian Federation as part of a state assignment (project No. FSZN-2020-0026). Bibl. 19, Fig. 7, Tabl. 1.*

Key words: bismuth, bismuth-antimony, thermoEMF, size effect, power factor

Introduction

The bismuth-antimony solid solution is known as the most effective low-temperature (temperatures below 200 K) thermoelectric material. At the same time, works of recent years show the possibility of using quantum and classical size effects in electronic phenomena, as well as internal strains to increase the thermoelectric figure of merit of materials [1 – 5].

The thermoelectric figure of merit (Z) in low-dimensional structures and nanostructures in comparison with homogeneous bulk materials can increase both due to an increase in the power factor (P), due to the peculiarities of the densities of states in the vicinity of the bottom of the lower dimensional quantization subband [5, 6] and due to a decrease in thermal conductivity due to the scattering of phonons at the interfaces [7]. Another mechanism for changing the thermoelectric power can be a change in the ratio of the contribution of electrons and holes to the thermoEMF due to different restriction of their mobilities by the surface and crystallite boundaries in a thin film [8, 9].

Straintronics offers another possibility in the task of increasing the thermoelectric figure of merit [10]. Straintronics (from the English “strain” - tension) is a new scientific direction of condensed matter physics, using physical effects in solids caused by strains arising in micro- and nanolayers and heterostructures under the action of external control fields, leading to changes in the band structure, electric, magnetic, optical and other properties of materials [11]. The possibilities of straintronics become obvious if we pay attention to the fact that some theoretical calculations and some experimental results show that the use of high pressures can significantly increase the ZT of some materials [12]. However, this approach is not widely used due to the technological complexity of creating high pressures in finished devices. It is deformation that can be an analogue of high pressures, which, in the case of thin-film materials, can be easily created in several ways, in particular, by using substrates with different lattice parameters and coefficients of thermal expansion, deposition of films on bent substrates or their controlled bending directly during operation, and much more. In low-dimensional structures located on substrates, it is possible to create record elastic strains, for example, vitrified bismuth wires of submicron size withstand relative elongations of 2-3% [13]. This is equivalent to the value of elastic strains in bulk crystals corresponding to mechanical stress up to 1 GPa, which approximately corresponds to the values used in the study of bulk crystals of this type. At present, active research in the field of straintronics is just beginning.

Within the framework of this work, the possibilities of increasing the thermoelectric figure of merit of thin films of bismuth and bismuth-antimony solid solution are experimentally investigated using the above approaches.

Experimental procedure

Bismuth films with a thickness of 10 nm to 1 μm and bismuth-antimony films with an antimony concentration from 3 to 15 at. % Sb were investigated. Plates of monocrystalline mica (muscovite) and a polyamide film were used as substrates. The coefficient of linear thermal expansion (CLTE) of these materials is 8×10^{-6} K^{-1} and $45*10^{-6}$ K^{-1}, respectively. The CLTE of bismuth in the trigonal plane is 10.5×10^{-6} K^{-1}. Thus, mica substrates cause in-plane tensile strain, while polyimide substrates cause in-plane compression strain of the film at temperatures below the film formation temperature.

Various methods were used to obtain films with different structural perfection. The main method for the preparation of thin-film samples was thermal deposition in high vacuum (10^{-5} Torr). In this case, for the films of the bismuth-antimony solid solution, the method of discrete evaporation was used, which makes it possible to obtain a homogeneous distribution of the solid solution components throughout the volume. Using this method, under optimal production conditions [14], it is possible to obtain films with crystallite size more than an order of magnitude larger than the film thickness (for bismuth) and several times larger than the film thickness (for bismuth – antimony solid solution). To obtain films with a single crystal structure, the method of zone recrystallization of a film under a protective coating was used [8]. In order to obtain films with block sizes of the order of the film thickness, we used a technique based on growing a film in a high vacuum on preformed nanoclusters [15]. The structure of the films was monitored by atomic force microscopy and X-ray structural analysis. All films were oriented in the (111) plane parallel to the substrate plane.

On the obtained films, the temperature dependences of the thermoEMF and resistivity were investigated in the temperature range of 77 to 300 K at a stepwise temperature change with temperature stabilization at the measurement point. To measure the thermoEMF, we used a technique that excludes distortion by the installation components of natural strain in the film – substrate system, presented in [16].

Experimental results and their discussion

As indicated in the introduction, in bismuth films, the crystallite boundaries and the surface restrict the mobility of electrons and holes in different ways, which leads to significant changes in the value of the Hall coefficient depending on the ratio between the thickness and crystallite size. The mobility of electrons is to a greater extent restricted by the surface at low temperatures and the mobility of holes – by crystallite boundaries. In order to study the influence of the above phenomenon on the thermoEMF, which, like the Hall effect, is a difference effect, bismuth films on mica with significant differences in crystallite size have been investigated. We studied films with crystallite size of the order of the film thickness (No. 1,4 in Figs. 1 and 2), obtained using bismuth nanoclusters in accordance with the method developed in [15], films with crystallite size more than an order of magnitude larger than their thickness (No. 2,5 in Figs. 1 and 2), obtained by thermal evaporation in high vacuum under optimal conditions [14] with subsequent annealing, and single-crystal films obtained by zone recrystallization (No. 3,6 in Figs. 1 and 2) [8].

Figs. 1 and 2 show the temperature dependences of the thermoEMF and resistivity of bismuth films with a thickness of 300 nm (No. 1, 2, 3) and 1000 nm (No. 4, 5, 6). It can be seen from the presented dependences that for films of the same thickness, with a decrease in the crystallite size, the absolute value of the thermoEMF increases at low temperatures, which, as in the case of the Hall effect, is due to the greater restriction of the hole mobility by the crystallite boundaries in comparison with the electron mobility. For film No. 4, the absolute value of the thermoEMF at 77 K exceeds the analogous value for bulk bismuth (α_{11} = -45 μV / K). In this case, the effect of crystallites on the mobility of charge carriers for films obtained using bismuth nanoclusters can vary significantly from sample to sample, which can be seen from a comparison of the resistivity of these films at low temperatures: for a film 1000 nm thick, the resistivity at 77 K is almost 2 times higher in comparison with a film with a thickness of 300 nm, obtained in a similar way (Fig. 2).

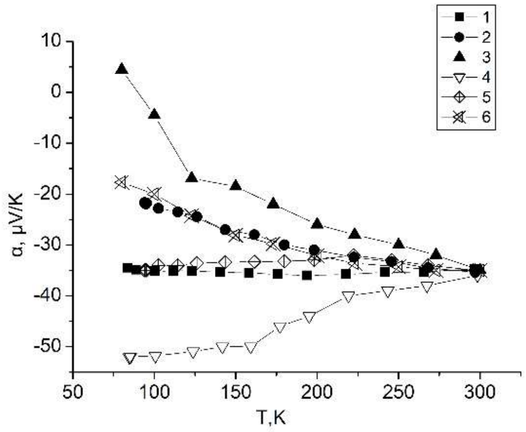

Fig.1 . Temperature dependence of thermoEMF for bismuth films on mica substrate

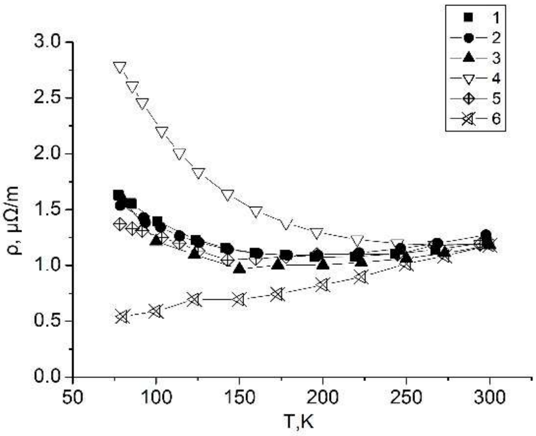

Fig. 2 Temperature dependence of resistivity for bismuth films on mica substrate

In [1, 2], it was theoretically shown for the first time that the quantization of the energy of charge carriers in thin films and wires can lead to an increase in the thermoEMF and a significant increase in ZT. A significant thermoEMF increase in thin films due to the quantum size effect should occur at thicknesses h commensurate with the de Broglie wavelength of charge carriers $= 2\pi h/\sqrt{2E_F m^*}$. In bismuth single crystals, charge carriers have a sufficiently large λ value, which in the direction of the C_3 axis is 67 nm for electrons and 11 nm for holes at a temperature of 77 K. In this work, an attempt is made to experimentally discover the influence of the quantum size effect on the thermoelectric properties of thin films of bismuth on mica.

Figure 3 shows the temperature dependences of the thermoEMF of bismuth films with a thickness of 10 nm to 1 μm, obtained by thermal spraying under optimal conditions [14] with annealing. It can be seen from the presented dependences that at low temperatures for films with a thickness of 1 μm to 27 nm, the absolute value of the negative thermoEMF decreases with decreasing film thickness, and for the thin film itself, the thermoEMF at low temperatures goes into the positive region. In this case, due to the peculiarities of the formation of thin-film structures, with a decrease in film thickness, an increase in the ratio between crystallite size and the thickness of the film (*D/h*) occurs. As indicated above, in this case, with a decrease in the film thickness, the electron mobility is more significantly restricted with respect to the hole mobility, which leads to a decrease in the contribution of electrons to the thermoEMF and a decrease in its absolute value for thinner films. However, for films with a thickness of less than 27 nm at low temperatures, an increase in the absolute value of the thermoEMF begins (inset in Fig. 3), while the dependence on the thickness *D/h* remains the same as for films with a greater thickness; therefore, the change in the character of the thickness dependence of the thermoEMF for films thickness less than 27 nm cannot be due to various restrictions of the mobility of electrons and holes by the surface and boundaries of crystallites. Probably, an increase in the absolute value of the thermoEMF with a decrease in the thickness of the bismuth films is associated with a change in the electronic energy spectrum due to the quantum size effect.

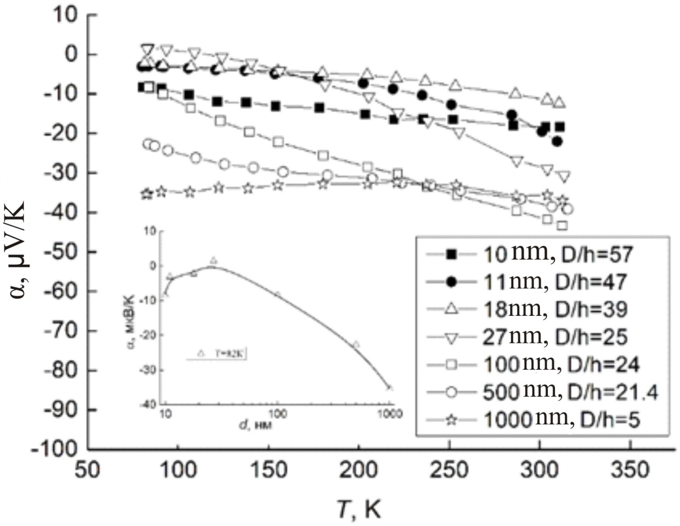

Fig. 3. Temperature dependences of thermoEMF of bismuth films of thickness from 10 nm to 1 μm. D/h is the ratio of crystallite size to film thickness

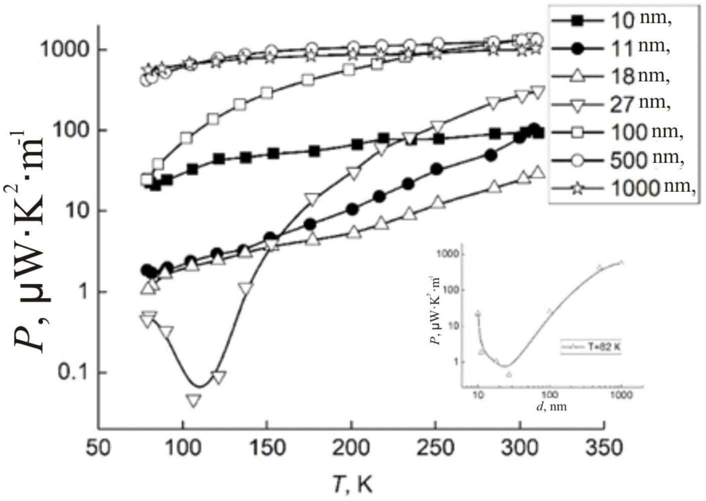

Fig. 4. Temperature dependences of thermoelectric power factor of bismuth films of thickness from 10 nm to 1 μm. D/h is the ratio of crystallite size to film thickness

For bismuth-antimony films this effect was not found, apparently, due to the lower efficiency of annealing in the task of increasing the crystallite size (Table 1) for thin films of bismuth-antimony solid solution and, as a result, smaller values of the coherence length of charge carriers as compared to films of pure bismuth, the large values of which are critical for observing coherent phenomena [17].

Using the measured temperature dependences of resistivity and thermoEMF for the studied films, the thermoelectric power factor P was calculated (Fig. 4). The highest value of thermoelectric power for all temperatures is observed for films with a thickness of 500-1000 nm. However, its thickness dependence is non-monotonic at low temperatures. For films less than 27 nm thick, the power factor begins to grow with a decrease in the film thickness, which, like an increase in the absolute value of the thermoEMF, is caused by a change in the band structure of the films due to the manifestation of quantum coherence of charge carriers.

In order to experimentally study the possibility of using internal mechanical stresses to improve the thermoelectric properties of thin films of the bismuth-antimony system, in this work, the thermoelectric properties of thin films of a bismuth-antimony solid solution on substrates with different thermal expansion coefficients: polyimide and mica (muscovite) are investigated. Under the influence of the difference in thermal expansion of the film and substrate materials, bismuth films on polyimide are in a state of in-plane compression, and bismuth films on mica, in a state of in-plane tension at a temperature below the film formation temperature. When analyzing the results, we used the values of the average crystallite size of the films of the bismuth-antimony system obtained by the methods developed in [18, 19] and presented in Table.

An increase in the concentration of antimony in the film is accompanied by an increase in the absolute value of the thermoEMF at low temperatures, which reflects a change in the thermoEMF in single crystals with a change in their composition (Fig. 5 and Fig. 6). A decrease in the film thickness in films of a bismuth-antimony solid solution leads to a decrease in the thermoEMF in absolute value in the low-temperature region, while a decrease in the crystallite size leads to an increase in its absolute value, in complete analogy with pure bismuth films.

The effect of film strain, due to the difference in the thermal expansion of the film and substrate materials, leads to a different type of the temperature dependences of the thermoEMF in bismuth-antimony films on mica and polyimide substrates.

Table

Crystallite size of bismuth and bismuth-antimony solid solution films, μm.

Substrate material	Thickness, μm / Composition, at.% Sb	1	0.5	0.5
Mica	0	5.4	10.7	3.6
	3	6.8	5.6	4.3
	5	8.2	1.8	3.2
	8	2.2	3.5	–
	12	3.8	3.2	2
	15	2.7	–	1.6
Polyimide	0	2.0	1.4	1.3
	3	1.0	0.8	0.6
	5	–	–	0.9
	8	1.2	0.7	–
	12	1.1	0.7	–
	15	1.0	–	–

The use of films on substrates with a high thermal expansion leads to a decrease in the absolute value of the thermoEMF, especially in the low-temperature region. The maximum values of the thermoEMF and power factor correspond to $Bi_{0.88}Sb_{0.12}$ films on mica and polyimide (Fig. 6, 7).

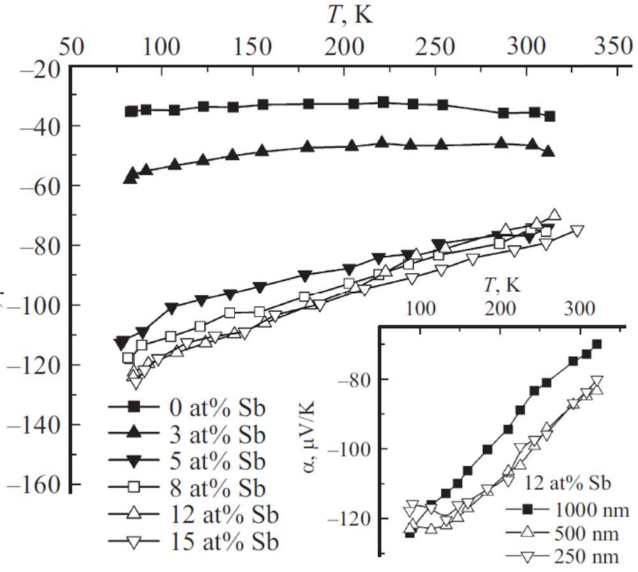

Fig. 5 ThermoEMF of films 1000 nm thick of different composition on mica. On the inset — thermoEMF of $Bi_{0.88}Sb_{0.12}$ films of different thicknesses.

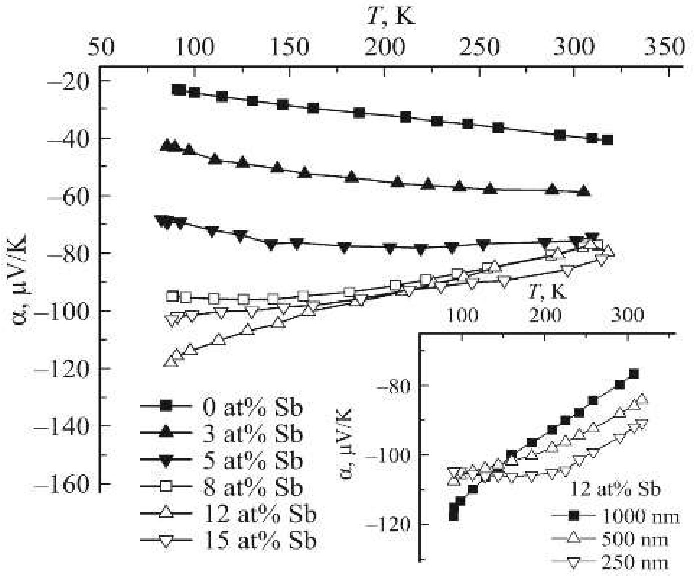

Fig. 6. ThermoEMF of films 1000 nm thick of different composition on polyimide. On the inset — thermoEMF of $Bi_{0.88}Sb_{0.12}$ films of different thicknesses.

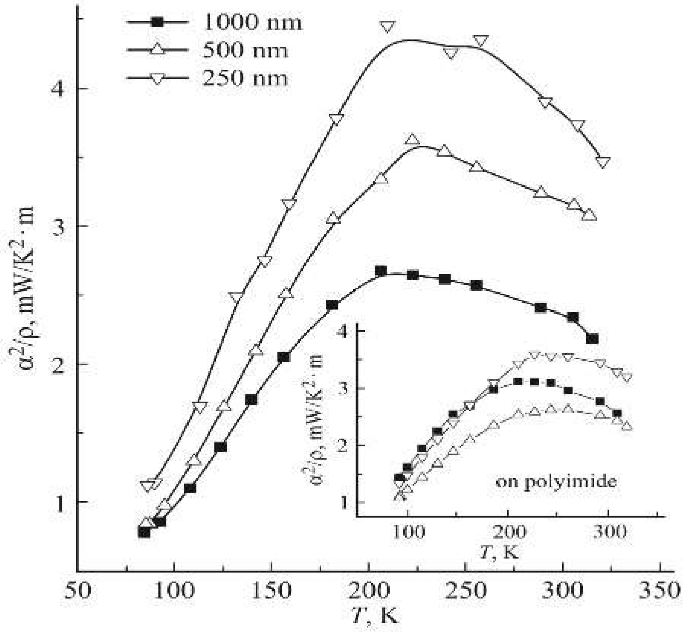

Fig. 7. Power factor of $Bi_{0.88}Sb_{0.12}$ films of different thicknesses on mica.
On the inset — power factor of $Bi_{0.88}Sb_{0.12}$ films of different thicknesses on polyimide.

Conclusions

It has been established that the use of films on substrates with a high thermal expansion leads to a decrease in the thermoEMF, especially in the low-temperature region. The maximum thermoEMF and power factor correspond to thick block films of $Bi_{0.88}Sb_{0.12}$ on mica. In these films, the maximum power factor of 4 * mW / K^{2}m was obtained at temperatures of 200-250 K. The study of ultrathin monocrystalline bismuth-antimony films with an achievable minimum defectiveness and a high surface perfection, which provides a long coherence length of charge carriers with a predominance of specular reflection from the film surfaces, seems to be promising for achieving high values of thermoelectric power. However, the technology for creating such films has not been developed yet.

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ТЕРМОЕЛЕКТРИЧНІ ВЛАСТИВОСТІ ТОНКИХ ПЛІВОК ВІСМУТУ І ТВЕРДОГО РОЗЧИНУ ВІСМУТ-СУРМА

Методом, що виключає виникнення зовнішніх деформаційних впливів на систему плівка-підкладка, були досліджені температурні залежності питомого опору і термоЕРС, розрахований фактор термоелектричної потужності в інтервалі температур 77-300К для плівок твердого розчину вісмут-сурма на підкладках з різним коефіцієнтом температурного розширення. Встановлено, що для отримання максимальної термоЕРС критично важливим є співвідношення розміру кристалітів і товщини плівки, що обумовлено різним обмеженням рухливостей електронів і дірок поверхнею і межами кристалітів. Максимальне значення термоЕРС і фактора потужності відповідає товстим блоковим плівкам $Bi_{0.88}Sb_{0.12}$ на слюді. Робота виконана в рамках державного завдання за фінансової підтримки Міносвіти Росії (проект № FSZN-2020-0026). Бібл. 19, Рис. 7, Табл. 1.

Ключові слова: вісмут, вісмут-сурма, термо, розмірний ефект, фактор потужності

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ТЕРМОЭЛЕКТРИЧЕСКИЕ СВОЙСТВА ТОНКИХ ПЛЕНОК ВИСМУТА И ТВЕРДОГО РАСТВОРА ВИСМУТ-СУРЬМА

Методом, исключающим возникновение внешних деформационных воздействий на систему пленка−подложка, были исследованы температурные зависимости удельного сопротивления и термоэдс, рассчитан фактор термоэлектрической мощности в интервале температур 77-300К для пленок твердого раствора висмут-сурьма на подложках с различным коэффициентом температурного расширения. Установлено, что для получения максимальной термоэдс критически важным является соотношения размера кристаллитов и толщины пленки, что обусловлено различным ограничением подвижностей электронов и дырок поверхностью и границами кристаллитов. Максимальное значение термоэдс и фактора мощности соответствует толстым блочным пленкам $Bi_{0.88}Sb_{0.12}$ на слюде.
Работа выполнена в рамках государственного задания при финансовой поддержке Минпросвещения России (проект № FSZN-2020-0026). Библ. 19, Рис. 7, Табл. 1.

Ключевые слова: висмут, висмут-сурьма, термоэдс, размерный эффект, фактор мощности

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