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Linear Positive Magnetoresistivity of the $Bi_{1.9}Lu_{0.1}Te_3$ Alloy with Inhomogeneous Micrograined Structure

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Positive nonsaturating transverse magnetoresistivity (*MR*) has been observed below room temperature in the $Bi_{1.9}Lu_{0.1}Te_3$ alloy with inhomogeneous micrograined structure. A crossover from parabolic $MR \sim B^2$ dependence in low magnetic fields to linear $MR \sim B$ dependence in high fields was found in the magnetoresistivity curves. The crossover field is shifted to lower magnetic fields as temperature decreases. Within the temperature range of "metal" type of conductivity, the temperature dependences of the specific electrical resistivity and *MR* magnitude are determined by the temperature dependence of the carrier mobility due to acoustic phonon scattering. Linear *MR* can be associated with microstructural inhomogeneity leading to strong electrical disorder of the $Bi_{1.9}Lu_{0.1}Te_3$.

Keywords: Linear Magnetoresistance, $Bi_{1.9}Lu_{0.1}Te_3$ alloy, Inhomogeneous Microstructure, Carrier Mobility.

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1. INTRODUCTION

The positive linear magnetoresistivity, LMR, takes place in a variety materials such as elemental metals, disordered and inhomogeneous semiconductors, Dirac materials including the graphene, topological insulators $Bi_{1-x}Sb_x$, Bi_2Se_3 and Bi_2Te_3, layered systems and so on [1-6].

At present, there are two approaches in a theoretical explanation of the LMR phenomenon. The first of them is the quantum approach proposed by Abrikosov [7]. This approach is based on the quantum theory of possible changes of spectrum properties of semimetals or narrow gap semiconductors.

The second approach is classical and takes into account a contribution of the structural and compositional inhomogeneities in the magnetoresistivity of materials with LMR. This approach is applicable for solids where a mean size or some characteristic size of inhomogeneities $l_c \gg l_o$ (here l_o is a free path length of the charge carriers) and at moderate temperatures.

In this work, we report on the observation the LMR effect in the $Bi_{1.9}Lu_{0.1}Te_3$ alloy with inhomogeneous micrograined structure.

The Bi_2Te_3 alloy is known to be one of the best materials for various low-temperature thermoelectric applications [8]. Recently it was found that doping Bi_2Te_3 with rare earth elements (Lu, Ce, Sm, Er, etc.) can improve thermoelectric properties of the Bi_2Te_3-based materials [9-12].

So, detailed research of transport properties including the magnetoresistivity is extremely important in order to find key material parameters responsible for improving the thermoelectric properties of Bi_2Te_3 doped with rare earth elements.

2. EXPERIMENTAL PROCEDURE

The microwave assisted solvothermal synthesis was applied to prepare the starting $Bi_{1.9}Lu_{0.1}Te_3$ powder.

Analytically pure chemicals were used for the synthesis (bismuth oxide, Bi_2O_3, tellurium oxide, TeO_2, lutetium oxide, Lu_2O_3, ethylene glycol, nitric acid and N,N-dimethylformamide). The Bi_2O_3, TeO_2 and Lu_2O_3 oxides taken in a stoichiometric ratio were dissolving in a mixture of concentrated nitric acid and ethylene glycol. Then N,N-dimethylformamide was added in mixture after dissolving. The microwave assisted reaction was carried out in a MARS-6 microwave reactor with a power of 1000 W at 2.45 MHz working frequency. Synthesis was carried out for 15 min at temperature of 463 K and pressure of 40 bar.

The powder after synthesis was spark plasma sintered (SPS) at temperature of 673 K and pressure of 40 MPa to prepare a bulk grained material.

Scanning electron microscope (SEM), the Nova Nano SEM 450, was used to study microstructure of the bulk material.

Cryogenic free system was used to measure the specific electrical resistivity, ρ, at the dc current density of $0.68 \cdot 10^3$ A/m^2. Magnetic field up to 5 T can be applied perpendicular to a current direction during the ρ measuring.

The magnetoresistivity, *MR*, as a function of applied magnetic field was determined from the field-dependent transverse resistivity

$$MR = \frac{\Delta\rho(B)}{\rho(0)} = \frac{\rho(B) - \rho(0)}{\rho(0)} \times 100\% . \qquad (1)$$

3. RESULTS AND DISCUSSION

The *MR*(*B*) dependences for the $Bi_{1.9}Lu_{0.1}Te_3$ alloy taken at various temperatures are shown in Fig. 1. One can see that the specific electrical resistivity increases as *B* increases. So, the $Bi_{1.9}Lu_{0.1}Te_3$ alloy is characterized by positive magnetoresistivity. The *MR* magnitude arises gradually as temperature decreases. It is important to note that the *MR*(*B*) dependences do no saturate up to maximum magnetic field at 5 T.

For high temperatures at ≥ 220 K, the *MR*(*B*) dependences obey the parabolic law that is *MR* grows quadratically with *B* as shown in inset in Fig. 1.

The field $MR \sim B^2$ dependence is typical for a lot of conventional metals and semiconductors and associated

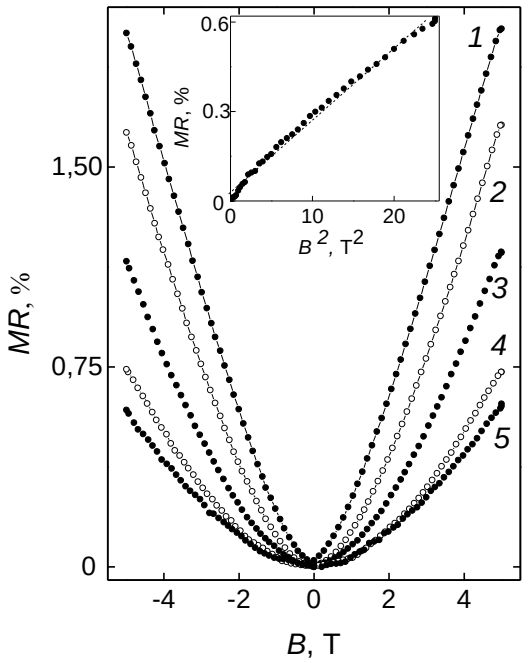

Fig. 1 – The *MR*(*B*) dependences for the $Bi_{1.9}Lu_{0.1}Te_3$ alloy at temperatures of 2 (curve 1), 60 (2), 130 (3), 160 (4) and 240 K (5). The inset shows the *MR*(*B*²) dependence at 240 K

with cyclotron motion of the charge carriers under external magnetic field.

The $\rho(T)$ curves taken at various magnetic fields were further used to extract the temperature dependence of *MR* (inset to Fig. 3). The ρ values at 0 and 5 T for the same temperature were used to calculate the *MR* magnitude. The *MR* magnitude was found to be decreased at cooling.

It is known that the Lu atoms substituting for Bi site in the Bi_2Te_3 compound behaves as donors [9]. So, electrons are major charge carriers and the specific electrical resistivity can be expressed as

$$\rho = \frac{1}{en\mu_n}, \qquad (2)$$

where e, n and μ_n are the unit charge, concentration and mobility of electrons, respectively.

For metals and generated semiconductors with "metal" type of conductivity the $\rho(T)$ dependence will be determined by the temperature dependence of the carrier mobility because n is T-independent for these solids.

The temperature dependence of the electron mobility due to acoustic phonon scattering can be written as [13]

$$\mu_n = \frac{2\sqrt{2\pi}e\hbar^2 d\upsilon_s^2}{3m^{*5/2}(kT)^{3/2}D_{ac}^2} \qquad (3)$$

where $\hbar$ is the Plank constant, d is the mass density, υ_s is the sound velocity, m^* is the effective mass of electron, k is the Boltzmann constant and D_{ac} is the deformation potential.

The $\rho(T)$ curves in Fig. 3 were replotted for the $\rho - T^{3/2}$ coordinates (Fig. 4). It was found that above some temperature $T^* \approx 33$ K up to 70 K the temperature dependence of the specific electrical resistivity is changed in accordance with the $\rho(T) \sim T^{3/2}$ law.

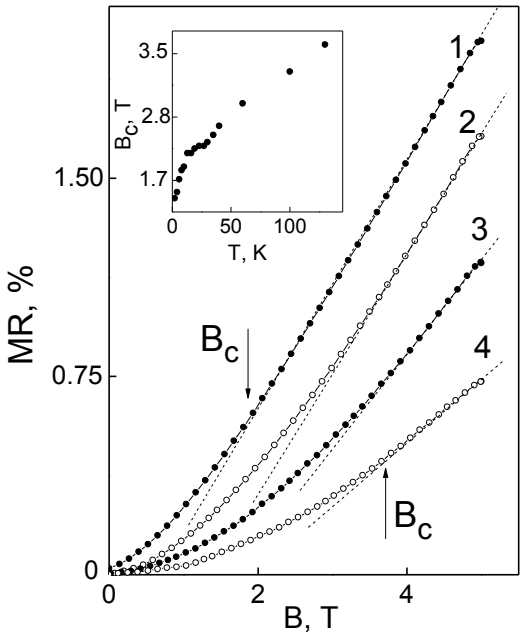

Fig. 2 – Crossover in the *MR*(*B*) dependences from parabolic behaviour to linear one at temperatures of 2 (curve 1), 60 (2), 130 (3) and 160 K (4). The inset shows the temperature dependence of the crossover field

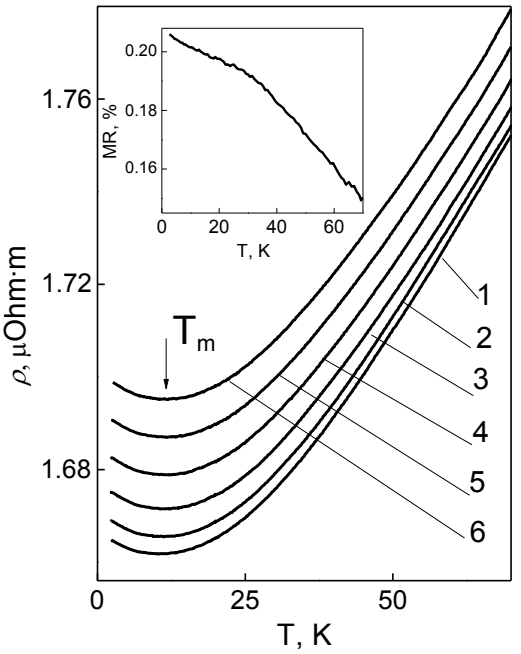

Fig. 3 – The MR(T) dependence for the Bi1.9Lu0.1Te3 alloy at magnetic field of 0 (curve 1), 1 (2), 2 (3), 3 (4), 4(4) and 5 T (5). The inset shows the MR(T) dependence

It means that acoustic phonon scattering is responsible for the $\rho(T)$ dependence within the temperature 30-70 K interval. It is important to note that for the same temperature interval the $MR(T)$ dependence is also linear for the $\rho - T^{3/2}$ coordinates (inset to Fig. 4). Linear fitting the experimental $MR(T^{3/2})$ dependence allowed us to recover the temperature dependence of *MR* as

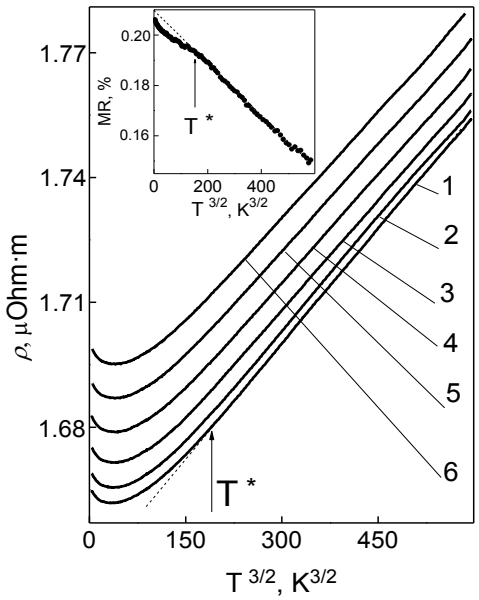

Fig. 4 – The $\rho(T^{3/2})$ curve for the $Bi_{1.9}Lu_{0.1}Te_3$ alloy at magnetic field of 0 (curve 1), 1 (2), 2 (3), 3 (4), 4(4) and 5 T (5). The inset shows the $MR(T^{3/2})$ dependence

$$MR(T) = 0.021 - 1.046 \times 10^{-5} \left[K^{-3/2} \right] \times T^{3/2}. \qquad (4)$$

Thus, the temperature dependence of the carrier mobility determines the temperature dependences of ρ and MR for the $Bi_{1.9}Lu_{0.1}Te_3$ alloy.

Below temperature T^*, the $MR(T)$ behavior becomes rather complicated. Obviously, this behavior is associated with change of the conductivity mechanism occurring around temperature T_m. So, in order to analyze the magnetoresistivity below T^*, the conductivity mechanism of "semiconductor" type should be established.

According to preliminary results, the variable-range hopping conductivity mechanism is responsible for the changes of the specific electrical resistivity within the temperature $\rho(T)$ range of "semiconductor" type. This mechanism takes into account the electron hops from one localized state to another one via a tunneling process. The localized states are corresponding to the different Lu atoms for $Bi_{1.9}Lu_{0.1}Te_3$.

As was mentioned above, the LMR effect can be observed in the structurally and compositionally disordered solids.

It is obviously that the structural and compositional inhomogeneities can exist in the $Bi_{1.9}Lu_{0.1}Te_3$ alloy.

For instance, Fig. 5 shows the SEM-image of the $Bi_{1.9}Lu_{0.1}Te_3$ alloy surface.

One can see that material under study has inhomogeneous micrograined structure consisting of dense grained layers divided by loose grained layers. According to Fig. 5, width of these layers is equal to ~20 μm and typical size of grains in these layers is a few microns.

It should be noted that the layers in Fig. 5 are oriented in the direction of the pressure application to a green body during the SPS-process. So, the layered structure of $Bi_{1.9}Lu_{0.1}Te_3$ can be formed at sintering the bulk grained material.

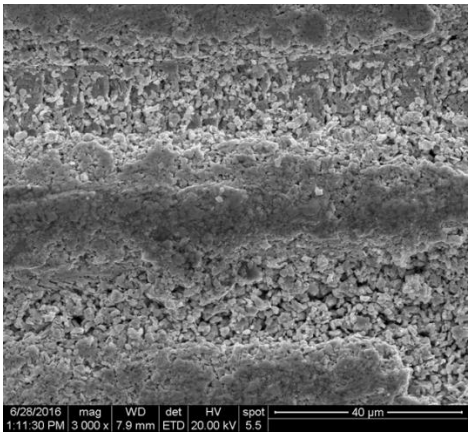

Fig. 5 – SEM-image of the structural inhomogeneties in the $Bi_{1.9}Lu_{0.1}Te_3$ alloy

Inhomogeneous volume distribution of the Lu atoms can be considered as another origin leading to the structural and compositional disorder in the $Bi_{1.9}Lu_{0.1}Te_3$ alloy. The variable-range hopping conductivity is characteristic of disordered semiconductors [14, 15]. In this case, the variable-range hopping conductivity mechanism supposed for the $\rho(T)$ behaviour at low temperatures can be taken as an evidence of the structural and compositional disorder in $Bi_{1.9}Lu_{0.1}Te_3$.

Classical model developed by Parish and Littlewood is often applied to describe of the LMR phenomenon in disordered semiconductors [16, 17]. This model takes into account that structural disorder leads to strong electrical disorder. In disordered systems a strong magnetic field forces a significant portion of the dc current to flow in a direction perpendicular to the applied voltage and thereby provides a linear-in-B Hall resistance contribution to the total magnetoresistance.

4. CONCLUSION

It was found that has the $Bi_{1.9}Lu_{0.1}Te_3$ alloy with inhomogeneous micrograined structure demonstrates positive nonsaturating transverse magnetoresistivity below room temperature. The crossover from quadratic field $MR \sim B^2$ dependence in low magnetic fields to linear field $MR \sim B$ dependence in high fields was observed. The crossover field decreases as temperature decreases, too. The temperature dependences of the specific electrical resistivity and MR magnitude are controlled by the temperature dependence of the carrier mobility within the temperature range of "metal" type of conductivity. The microstructural inhomogeneity leading to strong electrical disorder can be responsible for the linear magnetoresistivity of the $Bi_{1.9}Lu_{0.1}Te_3$ alloy.

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