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Low-temperature Minimum in the Electrical Resistivity of the $Bi_{1.9}Lu_{0.1}Te_3$

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The temperature dependence of the specific electrical resistivity, ρ, of the $Bi_{1.9}Lu_{0.1}Te_3$ alloy has been studied within the temperature 2 ÷ 230 K interval. Minimum in the resistivity was found at temperature $T_m \approx 11$ K. This minimum is originated from a change of conductivity mechanism. Above T_m, the resistivity ρ increases as temperature increases. This behavior is due to the electron mobility decrease via an acoustic phonon scattering at heating. Below T_m, the variable-range hopping conductivity based on electron tunneling takes place. In this case, ρ increases as temperature decreases. Two electric field regimes of the hopping conductivity were observed in the resistivity versus electric field strength dependences.

Keywords: Electrical resistivity, $Bi_{1.9}Lu_{0.1}Te_3$ alloy, Electron mobility, Hopping conductivity

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1. INTRODUCTION

The Bi_2Te_3 alloy is known to be one of the best materials for various low-temperature thermoelectric applications [1]. 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 [2 - 5]. So, properties of these materials should be studied in detail.

It should be noted that rare earth dopants can sufficiently effect on the electrical properties of the Bi_2Te_3 semiconductor via changes of the concentration and mobility of charge carries. A few mechanisms like the Kondo effect [6], an electron scattering by neutral or ionized impurity atoms [7], a hopping conductivity [8, 9] can control the electrical properties of the doped Bi_2Te_3. All of these mechanisms are effective for low-temperature range.

The aim of this paper is to study and analyze low temperature behavior of the specific electrical resistivity of the $Bi_{1.9}Lu_{0.1}Te_3$ alloy.

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 synthe-sis (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 at temperature of 653 K and pressure of 40 MPa.

Cryogenic free system was used to measure of the spe-cific electrical resistivity by a voltmeter-ammeter method at various values of the dc current. In this method, voltage drop across the sample and current through the sample were measured. Electrical contacts were made via silver paste. No effects from self-heating were observed.

3. RESULTS AND DISCUSSION

Figure 1 shows the $\rho(T)$ dependence for the sample under study taken at heating from 2 up to 230 K at the dc current density of $0.68 \cdot 10^3$ A/m^2. Clear $\rho(T)$ minimum is observed at temperature $T_m \approx 11$ K as is shown in the inset. Above this temperature ρ increases as temperature increases. This behavior is characteristic of metals. Below T_m the $\rho(T)$ dependence is typical for semiconductors that is ρ increases at the sample cooling.

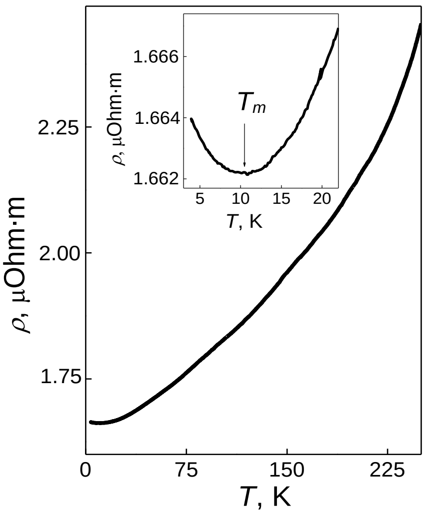

Fig. 1 – The $\rho(T)$ curve for the $Bi_{1.9}Lu_{0.1}Te_3$ alloy. The inset shows the details of the curve around the temperature T_m

Appearance of the $\rho(T)$ minimum can be associated with a change of the conductivity mechanism from "metal" type to "semiconductor" one.

It was found that above T_m up to temperature $T_1 \approx 190$ K the temperature dependence of the specific electrical resistivity is changed in accordance with the $\rho(T) \sim T^{3/2}$ law .

It is known that the Lu atoms substituting for Bi site in the Bi_2Te_3 compound behaves as donors [5]. So, electrons are major charge carriers. In this case for the temperature range under consideration the specific electrical resistivity can be expressed as [10]

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

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

Then the $\rho(T) \sim T^{3/2}$ dependence will be determined by the temperature dependence of the electron mobility due to acoustic phonon scattering [11]. The corresponding mobility can be written as

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

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

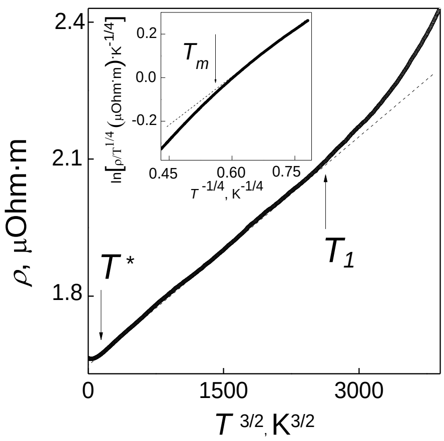

Fig. 2 – The $\rho(T^{3/2})$ curve for the $Bi_{1.9}Lu_{0.1}Te_3$ alloy. The inset shows the $\ln(\rho/T^{1/4})$ vs. $T^{-1/4}$ dependence

It should be noted that no $\rho(T)$ minimum observed in the non-doped Bi_2Te_3 alloy. So, doping Bi_2Te_3 with the Lu atoms will be responsible for appearance of this minimum.

A few main mechanisms can be considered to explain the $\rho(T)$ behavior in $Bi_{1.9}Lu_{0.1}Te_3$ below T_m:

(a) The Lu atoms are thermally ionized at heating. However, the change of ρ around T_m is too small and disagrees with a large Lu concentration.

(b) Conduction electrons are scattered by magnetic impurities due to the Kondo effect [6]. However, The Kondo effect is usually observed for far smaller concentration of magnetic impurities in comparison with our $Bi_{1.9}Lu_{0.1}Te_3$ alloy. Besides, a magnetoresistance of the Kondo alloys is negative, while the specific electrical resistivity of the $Bi_{1.9}Lu_{0.1}Te_3$ alloy increases under magnetic field.

(c) Electrons can be scattered by ionized impurities due to an electrostatic attraction between the electrons traveling in the lattice and the impurity. In this case the electron mobility increases as temperature increases in the accordance with the $\mu_n \sim T^{3/2}$ law [11]. This temperature behavior is connected with increase of the kinetic energy of electrons at heating that will just decrease time of interaction between the conductivity electrons and ionized impurities.

(d) The hopping conductivity can be realized in heavily doped semiconductors. According to this mechanism electron hops from one localized state to another one via a tunneling process. Generally, a probability of tunneling increases as the kinetic energy of electron increases, too. So, the tunneling conductivity will also increase at heating.

According to Ref. [8] the hopping conductivity in three dimensional doped crystalline semiconductors can be realized via different mechanisms given by a universal equation

$$\rho(T) = AT^p exp\left[\left(\frac{T_0}{T}\right)^p\right], \qquad (3)$$

where A is the constant, T_0 is the characteristic temperature and p is the exponent depending on the hopping conductivity mechanism.

The case of $p = 1$ corresponds to the regime of nearest-neighbor hopping conductivity and $p = 1/4$ and $1/2$ to the Mott and the Shklovskii-Efros types of variable-range hopping conductivity (VRH), respectively [8]. Generally, the VRH conductivity sets in when the internal microscopic disorder is high enough to make tunneling between the nearest sites energetically unfavorable.

The inset to Fig. 2 shows that the experimental $\rho(T)$ dependence for temperatures below T_m is linear for the the $\ln(\rho/T^{1/4}) - T^{-1/4}$ coordinates. It means that the VRH conductivity is responsible for the changes of the specific electrical resistivity within this temperature range.

In order to study the electrical behavior of the $Bi_{1.9}Lu_{0.1}Te_3$ alloy within the temperature VRH conductivity range, the $\rho(T)$ curves were taken at various values of the dc current density (Fig. 3).

One can see that the $\rho(T)$ minimum is depressed and shifted to lower temperatures as the dc current density increases. Besides, the voltage-current dependences were plotted to extract the dependences of the specific electrical resistivity versus electric field strength, E, around T_m.

Fig. 4 shows that ρ steadily decreases as E gradually increases. Such $\rho(T)$ change decreases as temperature increases and totally disappears at ≥ 27 K. The $\rho(T)$ (the inset to Fig. 2) and $\rho(E)$ (Fig. 3) dependences are in agreement with the tunneling conductivity.

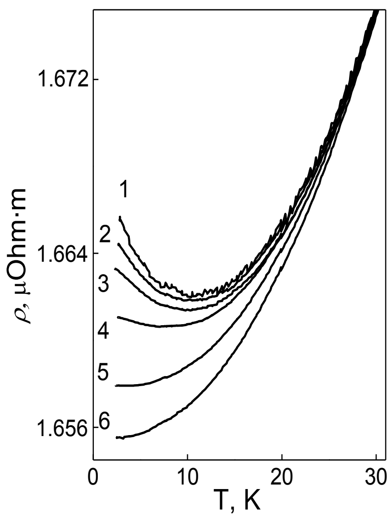

Fig. 3 – The $\rho(T)$ curves for the $Bi_{1.9}Lu_{0.1}Te_3$ alloy at various values of dc current density: $0.68 \cdot 10^3$ (curve *1*), $2.27 \cdot 10^3$ (*2*), $3.4 \cdot 10^3$ (*3*), $6.8 \cdot 10^3$ (*4*), $13.6 \cdot 10^3$ (*5*) and $22.7 \cdot 10^3$ A/m^2 (*6*)

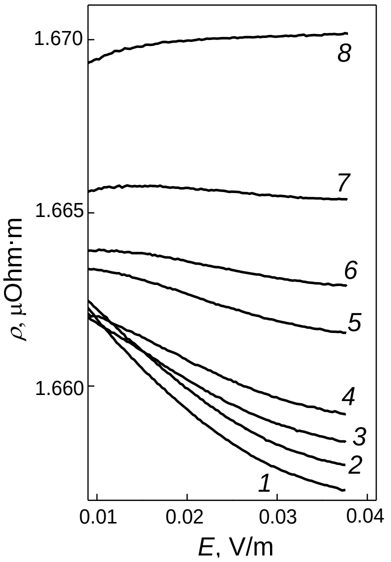

Fig. 4 – The $\rho(E)$ curves for the $Bi_{1.9}Lu_{0.1}Te_3$ alloy at various temperatures: 2 (curve *1*), 4 (*2*), 8 (*3*), 10 (*4*), 13 (*5*), 19 (*6*), 23 (*7*) and 27 K (*8*)

Two electric field regimes are considered separately for the hopping conductivity in the electric field. The weak-field regime, with $e\varepsilon_r Er << kT$, and the strong-field regime, where $e\varepsilon_r Er >> kT$ (where ε_r is the relative dielectric constant and r is the average hopping distance).

There are different models describing the $\rho(E)$ dependences of the semiconductors with the hopping conductivity [12]. All these models show the $\rho(E) \sim \exp(E^{-1/3})$ dependence for the strong electric fields. For the weak electric fields, the different models are not conclusive.

The experimental $\rho(E)$ dependences shown in Fig. 4 were replotted for the $\ln\rho - E^{-1/3}$ coordinates (Fig. 5). At least, two electric field regimes of the hopping conductivity can be observed for the $Bi_{1.9}Lu_{0.1}Te_3$ alloy at temperatures of 2, 4, 8, 10, 13, 19 and 23 K. In fact, the specific electrical resistivity for the strong electric fields above ~ 0.018 V/m is proportional to $\exp(E^{-1/3})$ as shown by dashed lines in Fig. 5. This behavior is in agreement with the models of the hopping conductivity.

CONCLUSION

The Bi_2Te_3 alloy was prepared by the microwave assisted solvothermal synthesis and the spark plasma sintering. The temperature dependence of the specific electrical resistivity of the alloy has been studied in detail within the temperature 2 ÷ 230 K interval. Minimum in the $\rho(T)$ dependence observed at temperature

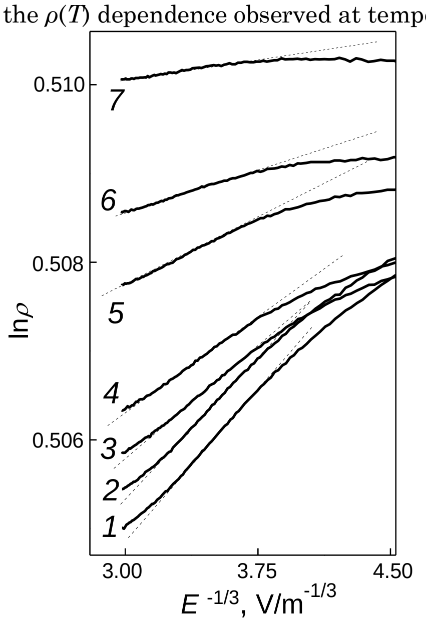

Fig. 5 – The $\ln\rho(E^{-1/3})$ curves for the $Bi_{1.9}Lu_{0.1}Te_3$ alloy at various temperatures: 2 (curve *1*), 4 (*2*), 8 (*3*), 10 (*4*), 13 (*5*), 19 (*6*) and 23 K (*7*)

$T_m \approx 11$ K. This minimum is associated with the change of the conductivity mechanism. Above T_m, the resistivity increases as temperature increases. This "metal" behaviour is due to the electron mobility decrease via an acoustic phonon scattering at heating. Below T_m, the variable-range hopping conductivity based on electron tunneling takes place. In this case, ρ increases as temperature decreases. Two electric field regimes of the hopping conductivity were observed in the specific electrical resistivity versus electric field strength dependences.

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Низкотемпературный минимум электрического сопротивления $Bi_{1.9}Lu_{0.1}Te_3$

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Температурная зависимость удельного электрического сопротивления ρ $Bi_{1.9}Lu_{0.1}Te_3$ изучено в интервале температур 2 ÷ 230 K. Минимум сопротивления обнаружен при температуре $T_m \approx 11$ K. Этот минимум появляется из-за изменения механизма проводимости. Выше T_m сопротивление увеличивается с увеличением температуры. Это поведение обусловлено уменьшением подвижности электронов из-за рассеяния фононов при нагревании. Ниже T_m прыжковая проводимость с переменной длиной прыжка, основанная на туннелировании электронов, имеет место. В этом случае ρ увеличивается с понижением температуры. Два полевых режима прыжковой проводимости обнаружены в зависимостях сопротивления от напряженности электрического поля.

Ключевые слова: Электрическое сопротивление, Сплав $Bi_{1.9}Lu_{0.1}Te_3$, Подвижность электронов, Прыжковая проводимость

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