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Fig. 1

Figure 1: Fig. 1

Abstract

Full Text

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ON THE INFLUENCE OF THE "DRAG" EFFECT ON THERMOMAGNETIC PHENOMENA IN BISMUTH SELENIDE

Bismuth selenide belongs to the class of well-conducting semiconductors, to which, in recent years, many investigations have been devoted in connection with their broad practical application. The electrical properties of Bi_2Se_3 were studied by Koality and Konorov ([1,2]). The study of thermomagnetic effects has made it possible, as will be shown below, to clarify certain new features of the interaction of current carriers with the crystal lattice. We investigated the electrical conductivity, Hall effect, thermopower, and the transverse and longitudinal Nernst–Ettingshausen effects (N.–E.) on 10 polycrystalline bismuth selenide specimens prepared by hot pressing or by slow cooling of the melt. The measurement method was analogous to that described previously ([3,4]); the measurements were carried out in the temperature interval 120—700°K. Below are given the results of measurements on 6 specimens. According to the sign of the Hall effect and of the thermopower at low temperatures, specimens Nos. 1 and 3 are hole-type, the others electron-type. The concentrations of holes (electrons), calculated from the Hall effect at $T < 200$°K (in cm^{-3}): for specimen No. 1 $1.2 \cdot 10^{19}$, No. 2 $1.6 \cdot 10^{18}$, No. 3 $1.9 \cdot 10^{19}$, No. 4 $1.2 \cdot 10^{18}$, No. 5 (cast) $1.5 \cdot 10^{19}$, No. 6 $1.7 \cdot 10^{19}$. Figure 1 presents plots of the temperature dependence of the coefficient of the transverse N.–E. effect.

Fig. 1

$$Q = E_y / \frac{\partial T}{\partial x} H_z.$$

(E_y is the field of the N.–E. effect, $\partial T/\partial x$ is the temperature gradient, H_z is the magnetic-field strength.)

Fig. 2

Figure 2: Fig. 2

Fig. 3

Figure 3: Fig. 3

For specimen No. 2 (and for two other specimens, the data for which are not given), in the region 120—200°K, $|Q| \sim T^{-1.3}$; for specimen No. 4 (and for one other analogous specimen), $|Q| \sim T^{1.3}$ (Fig. 2). Such a temperature dependence of the N.–E. effect cannot be explained with the aid of the usual formula for the case of weak magnetic fields ($uH/c \ll 1$, u is the mobility of the current carriers, c is the speed of light):

$$Q_{\mathrm{e}} = \frac{1-n}{2}\, a_n \frac{k}{e}\, u, \tag{1}$$

where n is determined by the relation between the mean free path of a current carrier l and its velocity v: $l = l_0(T)v^n$, $a_n \sim 1$.

In crystals with a predominantly homeopolar bond, such as, apparently, bismuth selenide also is, when current carriers are scattered by

acoustic vibrations of the lattice, according to the theory $n = 0$, and the N.–E. effect should be positive. The negative sign of the effect at low temperatures could be attributed to scattering by ionized impurity atoms, for which $n = 4$. But in this case we encounter two difficulties:

1) The mobilities calculated from formula (1) and from the Hall effect differ considerably from one another: for example, at 200° K, $u_x \simeq 10u_{\mathrm{meas}}$.

2) According to (1), the temperature dependence of Q is determined by the temperature dependence of the mobility. However, according to the Hall-effect and electrical-conductivity data, in samples Nos. 2, 4, and 5 below 450° K, $u \sim T^{-0.5} \div T^{-1.0}$.

Fig. 2

Fig. 3

It may be supposed that in the low-temperature region studied the N.–E. effect in the Bi_2Se_3 sample No. 2 is determined mainly by the dragging of electrons by phonons [5]. According to the calculation of Gurevich and Obraztsov [6], in nondegenerate semiconductors the drag should affect the N.–E. effect more strongly than the thermoelectric power. This was experimentally confirmed in the work of Mochan, Obraztsov, and Krylova [7], in which high-resistance p-Ge was studied in detail.

The assumption that drag influences the N.–E. effect in sample No. 2 is confirmed by measurements of the thermoelectric power (Fig. 4). From 120° K to 160° K the thermoelectric-power coefficient α decreases in absolute value according to the law: $|\alpha| \sim T^{-1.5}$. A decrease of $|\alpha|$ with increasing T at low temperatures was observed on single crystals of gray tin, Ge, Si, and InSb by a number of authors [9–13], who explained it by the drag effect. The dependence $\alpha(T)$ for polycrystalline Bi_2Se_3 No. 2 can also be explained by the influence of electron drag by phonons. When electrons are scattered only by phonons, the thermoelectric power can be represented approximately as the sum of the electronic part α_e and the phonon part (due to drag) α_ϕ [12]:

$$\alpha = \alpha_e + \alpha_\phi = \pm\frac{k}{l}\left(2 - \frac{\mu}{kT}\right) \pm \frac{w^2\tau_\phi}{uT}, \tag{2}$$

where μ is the chemical potential, τ_ϕ is the phonon relaxation time, w is the speed of sound; the signs + and − refer respectively to hole and electron semiconductors. From the experimental curve $\alpha(T)$ we found the temperature dependence of α_ϕ. Assuming that in the interval 170-300° K $\alpha \approx \alpha_e$, one can, using the Hall-effect data, calculate the effective mass of the electrons m_-. It turned out to be $0.18\,m_0$ (m_0 is the mass of a free electron). Then, using the value found for the effective mass, we calculate α_e in the interval 120-160° K. The calculated

Thus the values of α_{ph} obey the law: $|\alpha_{ph}| \sim T^{-2.8}$. Using the temperature dependences of u and α_{ph}, we find from (3): $\tau_{ph} \sim T^{-2.3}$.

The weaker dependence $Q(T)$, observed for specimen No. 4 with an electron concentration even somewhat smaller than that of specimen No. 2, can be explained by the influence of phonon scattering at grain boundaries (at a magnification of 15-20 times it is clearly seen that specimen No. 4 is considerably more fine-grained than specimen No. 2) and by a less steep temperature dependence of the mobility, which should lead to a decrease of the exponent in the dependence $\tau_{ph} \sim T^{-r}$.

In Ref. (10) it was shown, using Ge as an example, that at 100–150°K an increase in impurity concentration from $\sim 10^{14}$ cm^{-3} to $\sim 10^{18}$ cm^{-3} does not lead to a decrease in α_{ph}. The same should, naturally, be true for Q_{ph}. Our experiments with bismuth selenide show that, at carrier concentrations $N \sim 10^{18}$ cm^{-3}, the influence of drag on the Hall–Ettingshausen effect and on the thermopower is decisive. As for specimens with $N \sim 10^{19}$ cm^{-3}, it may be asserted that the higher the concentration, the smaller the role played by drag (in Bi_2Se_3 No. 6 it is not manifested at all), and that in hole specimens the influence of drag is greater than in electron specimens (in all hole specimens, only negative regions of Q are observed at low temperatures).

Fig. 4

Fig. 4

Figure 4: Fig. 4

For Bi_2Se_3 No. 2 the longitudinal Hall–Ettingshausen effect (the change of thermopower in a magnetic field) was also investigated. The effect proved to be comparatively small, but it was established that, with increasing temperature from 120°K to 160°K, $\Delta\alpha = \alpha(H) - \alpha(0)$ decreases in agreement with the theory (6).

The positive part of the curve $Q(T)$ for specimens Nos. 2, 5, and 6 is determined by ordinary scattering of current carriers by phonons and is described by formula (1). Calculation of the electron mobility from (1) at $n = 0$ (scattering by acoustic vibrations) near the maximum of $Q(T)$, where neither the drag effect nor the appearance of the second kind of current carriers substantially affects the Hall–Ettingshausen effect, gives values of u_{HE} in good agreement with the Hall mobilities. Thus, for example, at 360°K in Bi_2Se_3 No. 2, $u_{\mathrm{HE}} = 760\ \mathrm{cm}^2/(\mathrm{V}\cdot\mathrm{sec})$, while $u_H = 780\ \mathrm{cm}^2/(\mathrm{V}\cdot\mathrm{sec})$. At temperatures above 450°K, at which, according to the data for R, σ, and α, holes begin to participate in the conductivity, the Hall–Ettingshausen constant would have had to change sign. The absence of a sign reversal in the region of mixed conductivity can be explained by the large value of the ratio of the mobilities of electrons and holes. This assumption is confirmed by comparison of the Hall mobilities in electron and hole specimens and of the effective masses calculated from the thermopower, by estimation of b from the Hall–Ettingshausen effect, etc. For temperatures below 200°K the ratio of the mobilities of electrons and holes, estimated from the Hall effect for specimens Nos. 2 and 3, is $\cong 13$. From the ratio of effective masses we find $b \cong (m_+/m_-)^{5/2} \cong 12$. If one assumes that, at the temperatures where Q changes sign in specimens Nos. 1 and 3, $N_+ \cong N_-$, then, using the forbidden-band width $\Delta E = 0.27$ eV, determined from the dependence of $\ln RT^{3/2}$ on $1/T$ up to $T = 800$°K, it is easy to find $b \cong 16$ for specimen Bi_2Se_3 No. 3 and $b \cong 17$ for specimen No. 1. One can also estimate b from the ratio of the maximum value of the Hall constant for hole specimens in the region of mixed conductivity to the value of R in the impurity ···

regions: $|R_{\max}|/R_{\mathrm{straight}} = (b-1)^2/4b$. For sample No. 1 we obtain $b \simeq 13$, for sample No. 3 $b \simeq 12$.

From the data for R and σ, and also from the high-temperature part of the $Q(T)$ curves for sample No. 2 and for other samples above 450°K, the law of variation of the mobility $u \sim T^{-0.5} \div T^{-1.0}$ is replaced by the law $u \sim T^{-1.5}$. The stronger dependence $u(T)$ for a small forbidden-band width leads to the fact that above $\simeq 500$°K σ begins to decrease (Fig. 3).

The dependences $Q(T)$ for samples No. 1 and No. 3 are similar. The differences consist in the fact that the minimum of $Q(T)$ for sample No. 3 is located at a higher temperature than for sample No. 1, and that at low temperatures $|Q|$

for sample No. 1 is about 4 times larger than for No. 3. This difference can be explained by the fact that the hole concentration in sample No. 1 at low temperatures is 1.5-2 times smaller than in No. 3. A minimum associated, as in the case of Ge, with the appearance of current carriers of the other kind, at larger carrier concentrations must be shifted toward higher temperatures. At low temperatures the electron gas in both samples is degenerate. If one uses the estimate of the effective mass of holes ($m_+ = 0.5\, m_0$), made from thermopower data in the region 200-280°K, where Pisarenko' s formula is applicable, then it is easy to calculate the reduced chemical potential μ/kT. At 130°K in sample No. 1 this potential is $\simeq 3$, and in sample No. 3 $\simeq 5$, i.e. in the second case the electron gas is more strongly degenerate. Such a difference in the quantities μ/kT should, according to Wright' s calculations ([14]), correspond to a twofold difference in the constants Q_e. The observed difference in the quantities Q can be explained if, in addition, the drag mechanism is taken into account, which should manifest itself more strongly in sample No. 1. The negative sign of Q at low temperatures may also be explained, as already mentioned, by the influence of the drag effect. It should be noted that at a high concentration of ionized impurity atoms, scattering of current carriers by them also leads to a negative sign of Q. The absence, in the $Q(T)$ curves for hole samples Nos. 1 and 3, of a positive region corresponding to scattering of current carriers by acoustic phonons is connected with the appearance, at comparatively low temperatures, of conduction electrons (in sample No. 1 R changes sign from positive to negative at 300°K, in sample No. 3 at 370°K), to which the N.-E. constant is very sensitive. One can also understand the reason for the "dragging out" of the negative part of the $Q(T)$ curves toward high temperatures. In the region of mixed conductivity the concentration of low-mobility holes is greater than the concentration of high-mobility electrons, as a result of which $Q < 0$. On passing to intrinsic conductivity, when the concentrations become equal, the predominance of the electron mobility over the hole mobility leads to a change in the sign of Q. In conclusion we note that the discrepancies between the values of the mobilities from the Hall effect and from the N.-E. effect, observed at low temperatures in PbS, PbSe, and PbTe ([15]), are also apparently determined by the influence of drag on the N.-E. effect.

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Note: Figure translations are in progress. See original paper for figures.