Soviet-era science, translated into English

Crystallography

1965

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Abstract

Full Text

Crystallography

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Comparative Study of the Electrical and Thermal Properties of Single Crystals and Pressed Specimens of Lead Sulfide

(Presented by Academician A. V. Shubnikov, 12 IX 1964)

Studies of the properties of compounds of elements of Groups IV and VI of the periodic system are carried out both on single crystals and on polycrystalline–pressed–specimens. The aim of the present work is to clarify the difference in the thermal conductivity and electrical conductivity of single-crystalline and pressed specimens, using lead sulfide with electronic type of conduction as an example, and to estimate the errors that arise in calculating the thermal conductivity of the crystal lattice and the mobility of current carriers when working with pressed specimens.

Fig. 1. 1 –dependence of the thermoelectric emf on the concentration of current carriers; 2 –dependence of electrical conductivity on the thermoelectric emf. *a* –single crystals, *b* –pressed specimens.

Single crystals of lead sulfide were grown by slow cooling of the melt. In obtaining the single crystals, lead chloride $PbCl_2$ with an excess of lead was used, as proposed by T. L. Koval' chik for obtaining lead telluride (ˆ1). Specimens for measurements, of dimensions $10 \times 10 \times 20$ mm, were cut from the single crystals. From the ground single crystals, pressed specimens were prepared by the cermet method at a sintering temperature of 400°. The specimens were annealed for 70 hours at 650° and then for 150 hours at 300°. On the annealed specimens the electrical conductivity, thermoelectromotive force, Hall effect, and thermal conductivity were measured. All measurements were carried out at room temperature.

Figure 1 presents the dependences $\alpha = f_1(n)$ and $\alpha = f_2(\sigma)$, where α is the

Fig. 2

Figure 2: Fig. 2

absolute thermoelectric emf; n is the concentration of current carriers, calculated from the Hall effect on the assumption that PbS is an atomic semiconductor (ˆ2); and σ is the specific electrical conductivity.

As is seen from Fig. 1, all pressed specimens have some additional ohmic resistance. Apparently, it may be attributed to interlayers between individual grains.

The total resistance of one cubic centimeter of a pressed specimen may be represented in the form

$$\frac{1}{\sigma_{\text{pr}}} = \frac{1}{\sigma_{\text{cr}}} + r, \tag{1}$$

where σ_{pr} is the electrical conductivity of the pressed specimen, σ_{cr} is the electrical conductivity of the crystalline part of the pressed specimen, and r is the resistance of the interlayers. It turned out that the resistance of the interlayers in pressed lead sulfide specimens amounts to 10 to 40% of the resistance of single crystals, i.e., it is quite considerable. Since it is obvious that the thickness of the interlayers is small, their resistivity must be very large.

As is known, the Hall mobility u is expressed by the relation

$$u = A\sigma R, \tag{2}$$

where A is a coefficient depending on the degree of degeneracy of the electron gas and on the mechanism of scattering of charge carriers in the substance; R is the Hall coefficient.

As follows from the data we obtained (Fig. 1), the electrical conductivity at a given concentration of charge carriers is a material constant only for single crystals. Consequently, the mobility must be calculated only from the results of measurements carried out on single-crystal specimens. Indeed, the mobility u for single-crystal specimens with a charge-carrier concentration of $3 \cdot 10^{18}$ cm^{-3} was equal to 600 $\text{cm}^2/\text{V} \cdot \text{s}$, which agrees with the results of work (3). At the same time, u for pressed specimens with the same concentration is on average 400 $\text{cm}^2/\text{V} \cdot \text{s}$.

Fig. 2. 1 –dependence of the reciprocal of the total thermal conductivity on the concentration of charge carriers; 2 –dependence of the thermal conductivity of the crystal lattice on the concentration of charge carriers. a –single crystals, b –pressed specimens

Let us proceed to a discussion of the thermal conductivity. Figure 2 presents the dependence of the thermal resistance of single-crystal and pressed specimens on the concentration of charge carriers. It is seen that the reciprocal of the total thermal conductivity of single crystals is smaller than that of pressed specimens at identical charge-carrier concentrations. At the same time, the thermal conductivity of undoped pressed specimens agrees well with the values obtained by Yu. A. Dunaev for analogous PbS specimens (4).

The difference between the thermal resistance of pressed and single-crystal specimens is apparently determined by the thermal resistance of the interlayers. Indeed, the thermal resistance of 1 cm^3 of a pressed specimen can be represented as follows:

$$\frac{1}{\chi_{\mathrm{pr}}} = \frac{1}{\chi_{\mathrm{cr}}} + g, \tag{3}$$

where χ_{pr} is the thermal conductivity of the pressed specimen; χ_{cr} is the thermal conductivity of the crystalline part of the pressed specimen, equal

thermal conductivity of the crystalline specimen at the same concentration of charge carriers as in the pressed specimen being measured; g is the thermal resistance of the interlayers, which remains approximately the same at all concentrations of charge carriers (see Fig. 2).

The thermal conductivity of the crystal lattice is calculated, as is known, from the equation

$$\chi_p = \chi_{\mathrm{tot}} - \chi_e, \tag{4}$$

where χ_{tot} is the total thermal conductivity; χ_p is the thermal conductivity of the crystal lattice; χ_e is the electronic component of the thermal conductivity, $\chi_e = L\sigma T$; L is the Lorenz number, which depends on the mechanism of scattering of charge carriers and on the degree of degeneracy of the electron gas; T is the absolute temperature.

Assuming that lead sulfide is an atomic semiconductor[2], we calculate the thermal conductivity of the crystal lattice both for single-crystal and for pressed specimens. In this case the thermal conductivity of the crystal lattice, calculated for single-crystal specimens, decreases with increasing concentration of charge carriers (Fig. 2).

However, when calculating the thermal conductivity of the crystal lattice from the experimental data for pressed specimens, it was found that the thermal conductivity of the crystal lattice increases with increasing electron concentration (see Fig. 2). Thus, for 3 pressed specimens with an electron concentration of $2.5 \cdot 10^{20}$ cm^{-3}, the average calculated lattice thermal conductivity is $6.6 \cdot 10^{-3}$ cal/cm $\cdot$ s $\cdot$ deg, whereas for a single crystal with the same electron concentration it is only $4.6 \cdot 10^{-3}$ cal/cm $\cdot$ s $\cdot$ deg.

Substituting relations (3) and (1) into expression (4), we obtain

$$\chi_p = \frac{\chi_{\rm pr}}{1-g\chi_{\rm pr}} - LT\,\frac{\sigma_{\rm pr}}{1-r\sigma_{\rm pr}}. \tag{5}$$

As is seen from relation (5), the error in calculating the thermal conductivity of the crystal lattice from the results of measurements on pressed specimens arises because we neglect the additional thermal and ohmic resistance of the interlayers. At high concentrations of charge carriers, when, in calculating the lattice thermal conductivity, a greatly underestimated value of the electronic component is subtracted, the errors in determining the lattice thermal conductivity become very substantial. As was indicated above, they lead to an increase in the calculated lattice thermal conductivity with increasing concentration of charge carriers.

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Received
9 IX 1964

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