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1957

SovietRxiv

Abstract

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PHYSICS

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THERMAL CONDUCTIVITY OF TELLURIUM

Among the numerous works on tellurium there is not a single one devoted to the study of thermal conductivity. In the papers of Wold ([1]), as well as of one of the authors ([2]), only the value of the thermal-conductivity coefficient of tellurium at room temperature and its change in a magnetic field are given.

In the present article we set forth the results of investigations of the temperature dependence of the thermal conductivity λ of pure tellurium in the interval from 10° to 500°. Fine-crystalline samples, prepared in the form of pellets by cold pressing under a pressure of 4000 kg/cm^2 and by hot pressing—at a temperature of 400° under a pressure of 360 kg/cm^2 for 6 hours—were investigated.

Beforehand the tellurium was purified by threefold fractional distillation in vacuum at 10^{-4} mm Hg. Spectrally pure samples were obtained in this way.

The thermal conductivity was determined by the flat compensation method ([3]), using vacuum in order to protect the sample from oxidation at the melting temperature.

The error of the measurements did not exceed 1-2% at temperatures close to room temperature and 4-5% at the melting temperature. The results are shown in Fig. 1. The curve of the change of the thermal conductivity λ as a function of temperature has a minimum at 250°, at which the value of λ decreases to 2/3 of its value at room temperature. On passing through the melting point of the sample, the thermal-conductivity coefficient rises from the value $5 \cdot 10^{-3}$ cal/cm $\cdot$ s $\cdot$ deg, preceding melting, to $4.8 \cdot 10^{-2}$ cal/cm $\cdot$ s $\cdot$ deg, and with further heating decreases rapidly.

In addition, special determinations of the temperature variation of the heat capacity were carried out on an adiabatic microcalorimeter of the type described in ([9]). These investigations showed the absence of any structural changes in tellurium up to the melting temperature, in particular—the polymorphic transition at 250° mentioned in some works ([1]).

To interpret the results of the experiments, one should proceed, first, from the presence of a number of mechanisms in the thermal conductivity of semiconductors ([4]), and, second, from the fact that tellurium above room temperature is a semiconductor with mixed conductivity and with an energy-gap width $\Delta E = 0.38$ eV ([7]).

Fig. 1. Temperature dependence of the thermal conductivity of tellurium in the interval 10–500°

Figure 1: Fig. 1. Temperature dependence of the thermal conductivity of tellurium in the interval 10–500°

Fig. 2. Temperature dependence of the change in various components of the thermal conductivity of tellurium in the solid state

Figure 2: Fig. 2. Temperature dependence of the change in various components of the thermal conductivity of tellurium in the solid state

The presence of two signs of charge carriers in tellurium further complicates the already complex general picture of thermal conductivity in it, as is evidenced by our experimental data.

The curves shown in Fig. 2 indicate the temperature variation of the different components of the thermal conductivity of tellurium. Experimental curve 1 is drawn from our data up to 400°. Curve 2 represents the temperature variation of the phonon part of the thermal conductivity, calculated according to Eucken' s law $\lambda T = c$. The value of the constant c was determined from our experimental data in the interval 10–100°, where $\lambda \sim T^{-1}$. Here $c = 1.1$. For higher temperatures the value of c was extrapolated. Comparison of curves 1 and 2 gives curve 3.

The values of the electron-hole thermal conductivity, calculated from the formula of Davydov and Shmushkevich [8],

$$\lambda_{\text{e.h.}} = L(\sigma_+ + \sigma_-) + L\frac{2\sigma_+\sigma_-}{\sigma_+ + \sigma_-}\left(\frac{\Delta E}{2kT} + 2 + r\right)^2, \qquad (1)$$

which takes into account other conditions of charge motion in semiconductors with mixed conductivity, as well as recombination of the electron-hole pair during their motion in a thermal field, fit curve 3 well. The factor before the second bracket, found from work [7], is equal to $0.5\,L$ over the entire temperature interval investigated.

Fig. 1. Temperature dependence of the thermal conductivity of tellurium in the interval 10–500°
Fig. 2. Temperature dependence of the change in various components of the thermal conductivity of tellurium in the solid state

Finally, curve 4 was obtained from the Wiedemann-Franz relation $\lambda_e = L\sigma$ and from our electrical-conductivity data.

The data of our experiment make it possible to conclude that the thermal conductivity in tellurium throughout the entire region of intrinsic conductivity is provided, in addition to quasi-elastic lattice vibrations and thermal diffusion of

charge carriers, also by thermal conductivity caused by diffusion and recombination of electron–hole pairs.

The sharp increase in the coefficient of thermal conductivity upon passing through the melting point is evidently explained by an increase in the concentration of current carriers. This view is supported by works devoted to electrical conductivity and the Hall effect [5,6].

The electrical conductivity of tellurium, upon passing through the melting temperature, increases to $\sigma = 2.4 \cdot 10^3\ \Omega^{-1} \cdot \mathrm{cm}^{-1}$. The Hall constant falls from 0.4 to 0.02.

The data presented indicate that, upon passing through the melting temperature in tellurium, dissociation of an enormous number of atoms occurs, as a result of which the number of free charge carriers increases sharply.

Thus, the phenomenon we have observed of an increase in the specific thermal conductivity of tellurium upon passing through the melting temperature by almost a factor of 10 in comparison with its value before melting not only does not contradict other experimental facts (increase in electrical conductivity, decrease of the Hall effect), but, on the contrary, supplements them.

It may be assumed that the increase in thermal conductivity associated with a change in the aggregate state of the semiconductor is a complex process,

in which not only electron–hole heat conduction but also phonon heat conduction takes part. However, the contribution of the phonon component to the sharp increase in the total thermal conductivity of tellurium on passing through the melting temperature may be considered insignificant, since the volume of tellurium changes smoothly in this process, without a jump, and in its liquid state immediately near the melting temperature, according to the concepts of A. F. Ioffe and A. R. Regel [4,5], the chain structure is preserved to a considerable extent, being completely destroyed only near 550°.

The very rapid fall of the coefficient of thermal conductivity above the melting temperature can be explained by the fact that the change in the structure of tellurium is accompanied by metallization of the bonds in the melt, where the mixed conductivity characteristic of this semiconductor in the crystallization region changes over into conductivity of one sign. Here that part of the thermal conductivity which is due to diffusion and recombination of electron–hole pairs disappears; consequently, the thermal conductivity is due mainly to thermal diffusion of the current carriers.

Thus, our experiments with molten tellurium cover a small temperature interval (460—490°) between two limiting states of molten tellurium, occurring continuously: the semiconducting state at the first moment after melting (452°) and the metallic state (550°).

For the case of molten tellurium in the semiconducting state (452°), the thermal conductivity, calculated by formula (1) with allowance for the transfer of heat

by diffusion and recombination of electron-hole pairs, has the value $1.38 \cdot 10^{-1}$ cal/cm · sec · deg. In the metallic state (550°), the thermal conductivity was calculated by the same formula without allowance for the second term. In this case $\lambda = 10^{-2}$ cal/cm · sec · deg. Both these values of the thermal conductivity fit well on our experimental curve when it is extended in both directions.

In addition to pure tellurium, we have studied solid solutions Te–Se. The curves of the temperature dependence of the change in thermal conductivity in these solutions are analogous to that obtained for pure tellurium, but differ in the magnitude of the minimum of thermal conductivity, observed at a temperature different for each solution, and in the character of the growth of λ on melting and its decrease after melting.

The dependence of the thermal conductivity of the solid solution Te–Se on the selenium content makes it possible to calculate the mean free path of phonons when they are scattered by phonons and impurity centers. This quantity is of the order of six interatomic distances.

At the present time, investigations of thermal conductivity in a broader temperature interval are continuing.

Received
16 VII 1957

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