INFLUENCE OF DIMENSIONS ON THE THERMAL CONDUCTIVITY OF CRYSTALLINE He^4 *SAMPLES*

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A study was made of the dependence of the thermal conductivity of crystalline He^4 samples on their dimensions. The crystals were grown, under a constant pressure of 85 atm, in glass ampoules having diameters of 4.2, 2.47, and 1.6 mm. It was found that the critical dimension of a crystal, at which the Poiseuille flow of the phonon gas began to exert a significant influence on the thermal conductivity, was about 1.3 mm for these crystals. The effective mean free path of phonons in the case of normal collisions was calculated from the measurements of the thermal conductivity in the Poiseuille phonon flow region. At the very lowest temperatures, the thermal conductivity of the samples was affected considerably by the specular scattering of phonons on the walls of the glass ampoule.

SOLID He^4 is a very convenient substance for the investigation of transport processes in dielectric crystals. It contains no chemical impurities and it can be prepared in an isotopically pure form (the concentration of He^3 can be less than 10^{-6}% in the initial gas). Therefore, the purity of He^4 crystals depends mainly on the method of growth employed and on the possibilities of controlling the quality of the samples obtained. The growth of crystals under constant pressure and under continuous visual and temperature control makes it possible to prepare perfect single-crystal samples of sufficiently large dimensions.[1-3] Consequently, several new quantum phenomena have been observed in the processes of energy transport in the crystal lattice.[3-5] In the best of samples, the effective mean free path of phonons l_{eff}, calculated from the measurements of the thermal conductivity, has been found to be of the order of the dimensions of a sample at the very lowest temperatures and l_{eff}, corresponding to the thermal conductivity maximum, has been found to be almost an order of magnitude greater than the diameter of the sample. This effect can be explained satisfactorily within the framework of a model first proposed by Gurzhi.[6]

It is known[7] that the thermal resistance of the crystal lattice is due to the inelastic scattering of phonons by phonons (umklapp processes) or by lattice defects (crystal boundaries, point defects, etc.,). Normal scattering of phonons by phonons, without any loss of momentum, does not result directly in thermal resistance. However, such scattering may alter considerably the value of the thermal conductivity and its dependence on temperature and on the dimensions of a crystal in that range of temperatures where the thermal resistance is due to the scattering of phonons by the boundaries of the crystal.

In fact, we can have a situation in a crystal when

$$l^N \ll d \ll l^U, \tag{1}$$

where l^N is the mean free path of phonons for normal collisions; l^U is the mean free path of phonons for inelastic phonon-phonon scattering; d is a characteristic dimension (for example, the diameter) of a sample. In this case, a phonon, which moves like a Brownian particle, travels a distance of the order of $l_{eff} \approx d^2/l^N$ between two collisions with the boundaries, i.e., the effective mean free path of phonons, which governs the thermal conductivity of a crystal, may exceed the characteristic dimensions of a sample (Poiseuille flow of phonon gas).

The existence of the Poiseuille flow is a sufficient condition for the possibility of exciting the second sound in crystalline substances. Direct observations of the second sound in He^4 crystals, grown under a constant pressure of 54.2 atm, have been reported recently by Fairbank et al.[5]

Clearly, the range of temperatures in which the condition (1) may be satisfied depends strongly on the internal properties of crystals (for example,

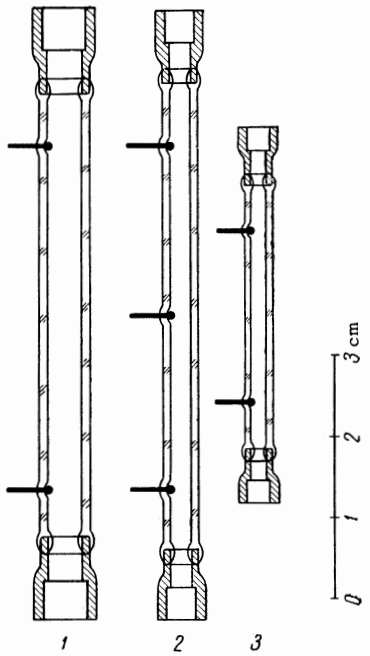

FIG. 1. Longitudinal sections of the ampoules in which samples were grown: 1) internal diameter 4.2 mm, distance between platinum wires 40 mm; 2) diameter 2.47 mm, distance 21 mm; 3) diameter 1.6 mm, distance 21 mm.

the Debye temperature and the orientation of the sample) as well as on the degree of imperfection and dimensions of the sample. It is difficult to obtain a theoretical estimate of the critical dimensions at which the Poiseuille flow of phonons begins to affect the properties of a sample; therefore, it is necessary to investigate the influence of dimensions on the thermal conductivity of crystalline He^4 samples.

Figure 1 shows a scale drawing of three ampoules, in which the investigated samples were grown. The construction of the apparatus and the measurement method have already been described.[3] The latter paper also contains the results of measurements of the thermal conductivity of samples grown in an ampoule with an internal diameter of 2.47 mm (ampoule 2). The measurements of the thermal conductivity were repeated on crystals grown under a constant pressure of 85 atm in ampoules having diameters of 4.2 and 1.6 mm (Nos. 1 and 3 in Fig. 1). The distance between the thermometers was 40 and 21 mm, respectively.

The results of the measurements in the wide ampoule were practically identical with those obtained earlier. An attempt to grow crystals against the force of gravity (the cold zone on top) also failed to improve markedly the quality of the crystals. This system was intended to suppress the influence—on the growing crystal—of foreign particles entering the ampoule together with gaseous helium. A marked improvement in the quality of such long samples was not obtained because of the incorrect construction of the ampoule: the platinum wires to which thermometers were attached extended too far into the interior of the ampoule so that stresses appeared in a crystal when the temperature was lowered.

In the present study, platinum leads in the narrow ampoules were designed more carefully and were placed flush with the glass wall. Typical results of the measurements of the thermal conductivity of samples grown in a glass ampoule of 1.6 mm diameter are given in Fig. 2. The same figure includes, for comparison, the results for the best of crystals grown under the same pressure in an ampoule of 2.47 mm diameter. It is evident from these curves that when the diameter was reduced the range of temperatures in which the influence of the normal phonon-phonon scattering appeared became considerably narrower so that it had practically no effect on the behavior of curves d and e, which represent the less perfect samples.

Thus, we could estimate that in the case of He^4 crystals grown under a pressure of 85 atm ($\rho = 0.218$ g/cm^3, $\Theta_D = 34$°K) the critical diameter was about 1.3 mm. If we assume that the shift of the exponential branch of the thermal conductivity curve from sample to sample is due to the different orientations of the crystals with respect to the ampoule axis, then this value should be the smallest dimension of a crystal, corresponding to the direc-

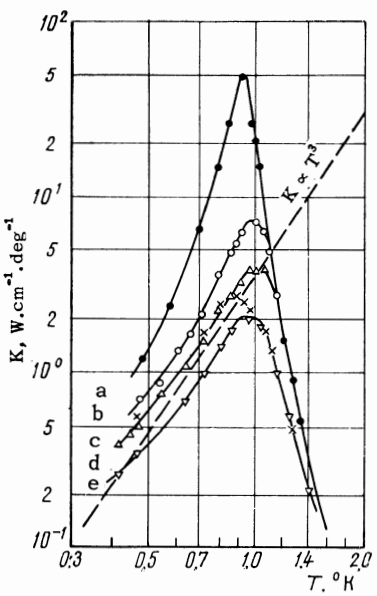

FIG. 2. Typical results of measurements of the thermal conductivity of samples grown under a pressure of 85 atm: a) the best of the crystals grown in an ampoule of 2.47 mm diameter; b)-e) crystals grown in an ampoule of 1.6 mm diameter.

tion of the highest thermal conductivity.

In a discussion of the results of the measurements on samples grown in an ampoule of 2.47 mm diameter, we suggested that at the very lowest temperatures the specular scattering of phonons on the walls of a glass ampoule could be important. Since the fraction of specularly reflected phonons increased when the temperature was lowered, there should be an increase in l_{eff}. If the specular reflection begins already in the Poiseuille phonon flow region, this could extend the region of the existence of the effect (represented in relative units), alter the temperature dependence and increase considerably the thermal conductivity at the very lowest temperatures.

For crystals grown under a pressure of 85 atm, the average wavelength of phonons at 0.5°K is about 10^{-6} cm, i.e., it may be greater than the characteristic dimensions of the roughness of the walls made of fused glass. Figure 3 shows the temperature dependences of the effective mean free path of phonons, calculated from the thermal conductivity of the same crystals as in Fig. 2. As before, the thermal conductivity was expressed in the form

$$K = {}^1/_3 Cvl_{\text{eff}} \qquad (2)$$

and, instead of the average velocity of sound, we used in these calculations the value of the velocity of longitudinal vibrations in solid helium,[8] which was about 1.8 times greater than the average velocity calculated in the Debye approximation; the specific heat was assumed to be proportional to $(T/\Theta_D)^3$. While the accuracy of the earlier measurements was found to be insufficient and the observed deviations from the law $K \propto T^3 d$[9] were within the limits of the experimental error, the accuracy in the present measurements reached ± 10% at the very lowest temperatures and l_{eff} increased approximately by a factor of 1.5. From the curves given in Fig. 3, it is evident that the contribution of the specular reflection of phonons from the walls of an ampoule becomes significant already at a temperature of the order of $(T/\Theta_D) \approx 1.5 \times 10^{-2}$, and in less imperfect crystals the specular reflection is masked initially by a stronger reduction in l_{eff} when the temperature is lowered in the Poiseuille phonon flow region.

Measurements of the thermal conductivity of crystals of sufficiently large dimensions make it possible to calculate directly l^N,[10] which is the effective mean free path in the case of normal collisions;[1)] indirect estimates of l^N can be ob-

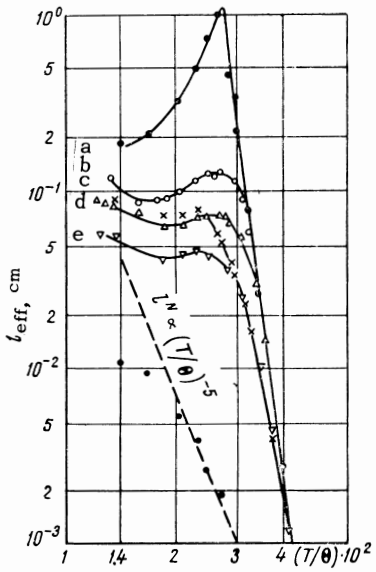

FIG. 3. Dependence of the effective mean free path of phonons on the reduced temperature for the same crystals as in Fig. 2. The results of calculations of the mean free path of phonons for normal collisions, using the measurements of the thermal conductivity of the best of the crystals (curve a), are shown below the other curves. For comparison, the calculated points are joined by a dashed line, representing the dependence $l^N \propto (T/\Theta)^{-5}$.

tained, for example, from the attenuation of the second sound. In fact, when the condition (1) is satisfied, the Gurzhi solution, based on an expansion in terms of small parameters l^N/d and l^N/l^U, is exact. For a circular cylinder, it has the form

$$l_{\text{eff}} = l^U\left[1 - \frac{2I_1(z)}{zI_0(z)}\right], \qquad z = \frac{d}{2\sqrt{l^U l^N}} \qquad (3)$$

where I_0 and I_1 are Bessel's functions with an imaginary argument; d is the diameter of the cylinder. For convenience, this expression can be rewritten in the form $l_{\text{eff}} = l^U f(z)$. Hence

$$f(z) = l_{\text{eff}} / l^U = K_{\text{meas}} / K^U.$$

The value of K^U can be calculated easily by extrapolating the exponential branch of the thermal conductivity curve. We can calculate z and determine l^N from the known ratio K_{meas}/K^U.

However, it is simpler to proceed as follows. It is evident from Fig. 4 that, with a high degree of accuracy, we can use $f(z) = z^2/8$ for $z < 1$. Hence, we find that for $f(z) < 0.1$

$$l^N = d^2/32 l_{\text{eff}}, \qquad (4)$$

i.e., for cylindrical samples the hydrodynamic approximation is valid for $l^N < d/32$ [cf. inequality (1)].

[1)] This was pointed out to us by I. M. Khalatnikov in a discussion of the results of previous measurements.

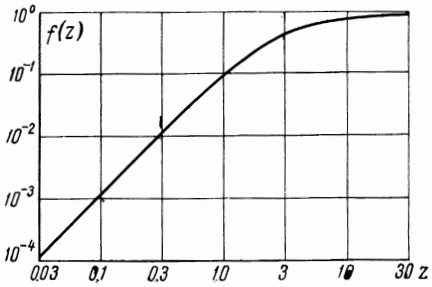

FIG. 4. Plot of the function $f(z) = 1 - 2I_1(z)/zI_0(z)$.

The results of calculations of l^N from the measurements of the thermal conductivity of the best crystal, grown in an ampoule of 2.47 mm diameter, are shown in Fig. 3. It follows from this figure[2)] that the dependence obtained is close to $l^N \propto (T/\Theta)^{-5}$. Similarly, we can find the dependence of l^N on T/Θ for crystals of other densities.

It is interesting to note that when the Debye temperature increases, the positions of the l^U and l^N curves in the figure shift not proportionally to Θ_1/Θ_2, which could be expected from the usual considerations of the dimensions of the quantities in these dependences, but rather as $(\Theta_1/\Theta_2)^3$. The reason for such a strong dependence is not clear but it may be associated with a reduction in the influence of the zero-point vibrations on the properties of solid helium when the solidification pressure is increased.

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Note added in proof, (January 12, 1967). Theoretical treatments of the processes of heat transport in dielectrics, allowing for the possibility of the diffuse motion of phonons, have been recently published by Gurevich and Éfros [11] and Guyer and Krumhansl. [12] Guyer and Krumhansl [12] make a detailed comparison of the theory and the experimental results. Although the results of Gurzhi reported in [6] and those of Guyer and Krumhansl [12] are in full agreement, numerical estimates of l^N from the measured thermal conductivity in the Poiseuille flow region using Eq. (10) in [13] result in values of l^N which are an order of magnitude greater than the values reported by us.

2)Within the limits of the experimental accuracy we could not distinguish between the exponents 4 and 5; we preferred 5 because $f(z) \sim 1$ near the maximum and if the calculations were more accurate the point on the extreme right-hand side in the figure could only be lower than the position obtained.

[1] A. I. Shal'nikov, JETP 41, 1056 (1961), Soviet Phys. JETP 14, 753 (1962).
[2] L. P. Mezhov-Deglin, JETP 46, 1926 (1964), Soviet Phys. JETP 19, 1297 (1964).
[3] L. P. Mezhov-Deglin, JETP 49, 66 (1965), Soviet Phys. JETP 22, 47 (1966).
[4] A. I. Shal'nikov, JETP 47, 1727 (1964), Soviet Phys. JETP 20, 1161 (1965).
[5] C. C. Ackerman, B. Bertman, H. A. Fairbank, and R. A. Guyer, Phys. Rev. Letters 16, 789 (1966).
[6] R. N. Gurzhi, JETP 46, 719 (1964), Soviet Phys. JETP 19, 490 (1964); Dissertation, Khar'kov Physico-technical Institute, 1965.
[7] R. E. Peierls, Quantum Theory of Solids, 1955 (Russ. Transl., IIL, 1955).
[8] J. H. Vignos and H. A. Fairbank, Proc. VIII Intern. Conf. Low Temp. Phys., London, Butterworths, 1963, p. 31.
[9] H. B. G. Casimir, Physica 5, 495 (1938).
[10] C. Herring, Phys. Rev. 95, 954 (1954).
[11] V. L. Gurevich and A. L. Éfros, JETP 51, 1693 (1966), Soviet Phys. JETP 24, 1146 (1967).
[12] R. A. Guyer and J. A. Krumhansl, Phys. Rev. 148, 766, 778, (1966); R. A. Guyer 148, 789 (1966).

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