The thermal conductivity of perfect and plastically deformed ^{4}He crystals grown in a narrow bent capillary

A. A. Levchenko and L. P. Mezhov-Deglin

Institute of Solid State Physics, Academy of Sciences of the USSR, Moscow

(Submitted 21 July 1981)
Zh. Eksp. Teor. Fiz. **82**, 278–292 (January 1982)

The thermal conductivity of perfect ^{4}He crystals and of crystals deformed by bending, has been studied. They were grown in a 1 mm diameter capillary, bent into a ring, at constant pressures of $P = 84$, 31 and 26 atm. By lowering the pressure from 84 to 26 atm, the diameter sufficient for observing fully developed Poiseuille flow of the phonon gas is reduced from 2.5 to 1 mm. Single or repeated bending of crystals at 0.45 K leads to a reduction in their thermal conductivity, and the magnitude of the additional thermal resistance at this temperature increases with increasing solidification temperature or with a reduction in the quality of the crystals for the same pressure. Annealing of the deformed crystals was observed at temperatures from 0.3 to 0.8 T_m. It was sufficient to heat the specimen up to a temperature $T_0 = 0.6$ to 0.8 T_m for returning the thermal conductivity of a specimen to its initial value within time $\leq 10^3$ s.

PACS numbers: 67.80.Gb, 66.70. + f, 81.40.Lm

§1. INTRODUCTION. STATEMENT OF THE PROBLEM AND THE MAIN THEORETICAL CONSIDERATIONS

1. Solid helium is an ideal material for studying the role of various energy transfer mechanisms in a dielectric solid.[1] Its large compressibility at small pressures enables the characteristic properties of the crystals (Debye temperature Θ, molar volume V_m) to be changed significantly from experiment to experiment in one and the same apparatus, by varying the solidification pressure. Quantum effects can greatly influence the properties of crystals at pressures close to the minimum of $P = 25$ atm in ^{4}He (see Ref. 2 for quantum crystals). In particular, the atomic oscillations in a quantum crystal lattice are highly anharmonic, i.e., the frequency of phonon interactions in such a crystal is much greater than in a classical dielectric at the same reduced temperatures T/Θ. As the pressure is increased, the contribution of quantum effects should rapidly decrease and helium crystals differ from normal dielectrics in the density which is an order of magnitude smaller and in the smaller velocity of sound, i.e., the elastic moduli of solid helium are two or three orders of magnitude lower than in ordinary crystals.

Helium-4 can easily be obtained with extreme chemical and isotopic purity (less than $10^{-6}\%$ of impurities and isotopes), by passing the superfluid through a finely dispersed filter.[3] It is not surprising that study of the thermal conductivity of perfect ^{4}He crystals[4] led to the discovery of a new hydrodynamic heat conductivity mechanism in solids.[5] These studies stimulated the appearance of a whole series of investigations devoted to the study of kinetic phenomena in solid helium. One can now find in the literature information on the orientation dependence of the thermal conductivity of ^{4}He single crystals,[6] results of studying the influence of ^{3}He impurity and thermal shock on the thermal conductivity of ^{4}He (Ref. 7) and papers on the increase in thermal conductivity of specimens with the maximum molar volume, produced by rapid cooling of compressed liquid at constant volume, as a result of annealing at temperatures $T < 1$ K (Ref. 8).

In the present work we present and discuss the results of studying the thermal conductivity of perfect crystals and of crystals of pure ^{4}He plastically deformed by bending at $T \leqslant 1$ K, which were grown in a 1 mm inside-diameter cylindrical capillary bent into a ring, from normal liquid at $P = 84$ and 31 atm and from superfluid helium at 26 atm. The following sections are devoted to a description of the experimental method, the results of the measurements and discussion of them. We should note here that it was not clear in advance whether it was possible to grow in a bent narrow tube a perfect single crystal with constant orientation relative to the longitudinal axis of the cylinder. When planning the investigations it was not clear whether it was possible in principle for fully developed Poiseuille flow to be observed in specimens grown in a tube of diameter $d = 1$ mm. It followed from the experience of all previous measurements that for pressures $P \geqslant 80$ atm a tube of diameter $d \geqslant 2.5$ mm is required.[4]

The effect of plastic deformation on the thermal conductivity had also not been studied previously. Only experiment could therefore provide the answers to the following questions: does bending influence the thermal conductivity of narrow helium crystals; what defects can accumulate on plastically deforming helium; can the annealing of defects introduced by the bending be observed at temperatures appreciably below T_m and, moreover, what mechanism of phonon scattering by the defects should predominate. The answers to these questions are also important in interpreting the results of many other experiments. For example, it has long been known that on cooling from T_m to $\sim 0.3 T_m$ the pressure in a container filled with solid helium falls by $\sim 1\%$ (for $P \geqslant 30$ atm) because of the great difference in thermal expansions of the helium specimen and the container. This is equivalent to hydrostatic tension of a highly anisotropic crystal under conditions such that the applied mechanical stresses can be comparable with the limiting strength of the crystal.

Evidence for the possible presence in helium of large mechanical stresses comes from observation of ex-

0038-5646/82/010166-09$04.00

plosive cracking of crystals and breaking away of wire thermometers when the temperature or pressure is varied in the neighborhood of the bcc-hcp transition in ^{4}He (Refs. 4 and 9). Why can helium crystals in closed cylindrical containers, as distinct from crystals of other noble gases, remain of high quality when specimens are slowly cooled and heated (less than 10^5 dislocations$\cdot$ cm^{-2} in the most perfect crystals grown in a bare cylindrical tube at constant pressure[4])? How does the structure of such crystals change with rapid cooling? Does the texture of specimens with metal electrodes or quartz crystals frozen in them change as the temperature is varied, in experiments with induced charges or in ultra-sonic investigations,[10,11] when changes in current or in sound velocity are observed at a given temperature during cycles of cooling to below 0.5 T_m and heating up to $T \sim T_m$?

2. It is usual to estimate the importance of different phonon relaxation mechanisms in a dielectric from the behavior of the effective phonon mean free path, l_{eff}, which is calculated from the thermal conductivity, $\varkappa$, measured in the experiment, by the well known kinetic equation[1]

$$\varkappa = {}^1/_3 C s l_{\rm eff}, \tag{1}$$

where C is the specific heat and s the mean velocity of sound. In the Debye approximation one can derive from this

$$l_{\rm eff}\,[\mathrm{cm}] \approx 4\cdot 10^{-6}\,\Theta^2 V_m^{2/3} \varkappa / T^3, \tag{2}$$

where V_m is the molar volume of the crystal in cm^3 $\times$ mole^{-1}, $\varkappa$ is the thermal conductivity in W$\cdot$cm$^{-1}\cdot$K^{-1} and T is the Kelvin temperature.

Above the temperature of the thermal conductivity maximum $T_{\rm max}$, the value of l_{eff} for a perfect crystal is limited by phonon-phonon umklapp processes (U-processes), in other words for $T > T_{\rm max}$

$$l_{\rm eff} = l_U \sim \exp(\Theta/\gamma T).$$

In an anisotropic crystal the value of the parameter γ depends strongly on orientation. The thermal conductivity of helium specimens, which according to Eq. (2) is proportional to the product $\varkappa \sim T^3 l_{eff}$, increases exponentially with decreasing temperature from T_m and the slopes of the high-temperature parts of the $\varkappa(T)$ curves depend on the orientation of crystals relative to the longitudinal axis of the tube.

As several experimenters have shown (e.g., Ref. 6), the $\varkappa(T)$ dependence in the region $T > T_{\rm max}$ for hcp ^{4}He crystals in the pressure range from 31 to 290 atm with the trigonal C_3 axis parallel or perpendicular to the tube axis (the direction of heat flow), can be described by single expressions of the form

$$\varkappa_\parallel \sim \exp(\Theta/4.6T), \quad \varkappa_\perp \sim \exp(\Theta/2.5T). \tag{3}$$

If β is the angle between the C_3 axis of a single crystal and the direction of heat flow, then the thermal resistance of the crystal, $W \equiv \varkappa^{-1}$, is correspondingly

$$W(\beta) = W_\parallel \cos^2\beta + W_\perp \sin^2\beta, \tag{3a}$$

and for a polycrystalline specimen

$$W = {}^2/_3 W_\perp + {}^1/_3 W_\parallel, \tag{3b}$$

since the thermal conductivity $\varkappa_\perp$ of a hcp crystal in the basal plane perpendicular to C_3, is isotropic. The structure of specimens can be judged by comparing the slopes of the high-temperature parts of the experimental $\varkappa(T)$ curves with the dependences of Eqs. (3) to (3b).

The maximum values of l_{eff} and $\varkappa$ and the slopes of the $l_{eff}(T)$ and $\varkappa(T)$ curves below $T_{\rm max}$ depend on the degree of perfection of the specimens and on their dimensions, and for perfect large single crystals of diameter d, also on the relationship between l_U, d and l_N, the phonon mean path for normal phonon-phonon interactions (N-processes which conserve total quasimomentum). If the hydrodynamic conditions[5]

$$l_U/l_N \gg 1, \quad d^2/l_U l_N \gg 1 \tag{4}$$

are fulfilled, the motion of the phonon gas in the crystal under the action of an applied temperature gradient is analogous to Poiseuille flow of a viscous liquid or gas along a tube under the action of a pressure difference. In the region where Poiseuille flow is developed

$$l_{\rm eff} \approx 0.1\, d^2/l_N \gg d \tag{5}$$

and it rapidly decreases as the temperature is lowered:

$$l_{\rm eff} \sim l_N^{-1} \sim T^{3-5}.$$

Fully developed Poiseuille flow can therefore only be observed in sufficiently large and perfect single crystals over a relatively narrow temperature range near $T_{\rm max}$. As numerical calculations showed,[12] Eq. (5) only holds if stricter conditions are satisfied than Eq. (4), namely

$$d/l_N \gtrsim 30, \quad l_R/l_N \gtrsim 3\cdot 10^3. \tag{6}$$

Here l_R is the effective mean free path for resistive scattering of phonons by phonons, defects, and impurities within the volume of the crystal, and is determined from the relation

$$l_R^{-1} = l_U^{-1} + l_{pi}^{-1}$$

(l_{pi} is the mean free path for scattering of phonons by defects of a given type). If conditions (6) are satisfied, then

$$\varkappa(T) \sim T^{6-8} d^2,$$

while the phonon mean free path for normal process interactions, l_N, can be estimated directly from the $l_{eff}(T)$ curve; this is difficult to calculate from first principles.

As the temperature is reduced, the phonon mean free paths in a perfect crystal increase rapidly and the Poiseuille flow of the phonon gas is replaced by Knudsen flow (the flow of an extremely rarefied gas along a tube). In the Knudsen limit at $T \ll T_{\rm max}$

$$l_R,\, l_N \gg d, \quad l_{\rm eff} \approx d \tag{7}$$

provided that the phonons are scattered diffusely at the boundaries of a cylindrical crystal.[1,13]

In single crystals which are not sufficiently large or perfect, when conditions (6) cannot be satisfied but the hydrodynamic conditions of Eq. (4) are satisfied, a transition region in the flow of the phonon gas from Poiseuille to Knudsen should be observed:

$$\begin{aligned} &\varkappa(T)\sim T^n, \quad n\geqslant 3, \\ &0.1d^2/l_N > l_{\rm eff} \geqslant d. \end{aligned} \tag{8}$$

Strictly speaking, it is just this regime which is observed in most work on solid helium.

3. The cross section for scattering of phonons by defects depends on the nature of the defect and the frequency (energy) and polarization of phonons.[1,13,14] If normal processes can be neglected, the mean free path for scattering by defects which enters into the thermal conductivity is $l_{pi}\sim T^{-4}$ for scattering by point defects and proportional to T^{-1} for scattering by the strain field around static dislocations, while for diffuse scattering at the surfaces of misoriented polycrystals the value of l_{pi} is independent of temperature and is determined by the mean characteristic dimensions of the crystals. Thus in the Knudsen limit $l_{\rm eff} \ll d$ and with decreasing temperature it can increase for scattering by static defects or remains constant for diffuse scattering at the boundaries, i.e., $\varkappa(T)\sim T^n$, where $n \leqslant 3$.

The flutter effect[13,15] i.e., dynamic phonon scattering by "vibrating" dislocations or, in other words, by local phonon modes associated with dislocations, can be important in plastically deformed crystals. The effective radius for scattering of thermal phonons by vibrating dislocations can be an order of magnitude greater than the scattering radius for static dislocations, especially for resonance scattering, so that when the flutter effect arises in the temperature range $T < T_{\rm max}$ when $l_{\rm eff} < d$, either a reduction or an increase of $l_{\rm eff}$ with decreasing temperature can be observed, depending on the relation between the frequencies of the localized phonon modes and thermal phonons.

In the general case, for example, when $l_N \ll l_{\rm pi} \leqslant l_U$, the contribution of different scattering mechanisms to the total thermal resistance W is not additive, i.e., Matthiessen's rule

$$W=\sum_i W_i$$

is not obeyed. For example, the effectiveness of scattering of phonons by point defects within the crystal increases by almost two orders of magnitude when frequent normal processes are taken into account.[14] In the temperature region where the hydrodynamic conditions of Eq. (4) are satisfied it is, therefore, rather difficult to separate the partial contributions of various scattering mechanisms, when not even the temperature dependences and orders of magnitude of the effective cross sections for scattering of phonons by the defects are given. This problem requires further theoretical study.

§2. EXPERIMENTS

The crystals were grown from specially purified ^{4}He (Ref. 3) at constant pressures $P = 84$, 31 and 26 atm. The melting temperatures of the specimens were $T_m = 3.05$, 1.80 and 1.46 K and the corresponding Debye temperatures $\Theta = 36.5$, 27.6 and 26.3 K. The design of the lower part of the apparatus is shown schematically in Fig. 1. Tube T, in which the crystals were grown, was made from a 15 cm long cold-drawn nickel capillary of diameter 1.2 mm and wall thickness 0.1 mm. The capillary was initially bent on a 7 cm diameter template. A cold finger from which the crystal started to grow was soldered below the broad cap in which the tube ended. The cold finger connected the tube to a copper vessel with liquid ^{3}He. A narrow coiled capillary C fitted into the top of the tube, leading from the external high-pressure line of the filling system. The tube and liquid ^{3}He vessel were contained in a vacuum jacket in a metal helium cryostat. The external setup, similar in design to that described earlier,[4] enabled temperatures $T \leqslant 1$ K to be maintained inside the jacket continuously for several days.

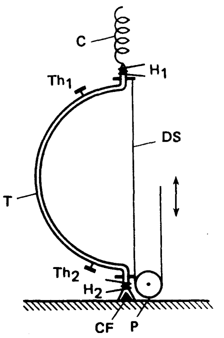

FIG. 1. Diagram of lower part of the apparatus: T—tube, CF—cold finger, C—capillary of filling system, DS—drawstring, P—cooled pulley of the pulling system, H_1, H_2—heaters, Th_1, Th_2—thermometers.

The upper end of the tube could be moved vertically over a distance of ~3 cm by means of the drawstring DS which connected the tube to a pulling system controlled from outside. As the pull on the string was reduced, the upper end of the tube tended to return to its initial position, i.e., the tube itself acted as a flat spring. The strength (and stiffness) of the tube at low temperatures was quite sufficient for studying the effect of repeated deformation of the bending-unbending type on the properties of helium crystals. The extent of the deformation of a helium crystal for a single movement of the upper end of the tube, calculated from the change in the radius of curvature of the tube, was ~1%, i.e., the dislocation density produced within the body for a single bending was 10^6 to 10^7 cm^{-2}. The thermometers Th_1 and Th_2 and heaters H_1 and H_2, placed along the tube, enabled the thermal conductivity of the crystals grown to be measured in the temperature range from 0.4 to 2 K with a relative error of $\leqslant 5\%$. The thermometers were positioned on the convex part of the tube separated by a distance of 10 cm.

The upper end of the tube was kept fixed during crystal growth, its position being fixed by the pulling system. The mean rate of crystal growth from normal liquid at 84 and 31 atm was 1 to 3 mm·min^{-1}, and from superfluid liquid at 26 atm it was an order of magnitude higher. After the thermal conductivity of the initially grown specimen had been measured, the effect of deformation on its thermal conductivity was studied for a single bending of the tube or as a result of a cycle of repeated bending-unbending movements. The rate of moving the upper end of the tube was, on average, 3×10^{-2} cm·s^{-1}. The temperature of the liquid ^{3}He was maintained constant during the deformation process, but the temperature of the tube was raised noticeably by 0.03 to 0.1 K, depending on the thermal conductivity of the specimen and the rate of movement, so that the first reliable point in thermal conductivity measurements could only be obtained after 15 to 30 min following the end of the deformation, when thermal equilibrium was reestablished in the apparatus. Bending at temperatures $T\geq1$ K affected the thermal conductivity of specimens weakly, so that most of the measurements were obtained on specimens deformed at $T=0.45\pm0.05$ K.

§3. RESULTS OF THE MEASUREMENTS

As was shown by the measurements, the magnitude of the additional thermal resistivity ΔW, determined from the difference between the thermal resistances of the deformed, $\varkappa_d^{-1}$, and initial, $\varkappa_i^{-1}$, specimen $\Delta W=\varkappa_d^{-1}-\varkappa_i^{-1}$ depends not only on the temperature at which the deformation was carried out, but also to a considerable extent on the properties of the crystals, the pressure at which the specimen was grown and the degree of perfection of the initial specimen. It is, therefore, more convenient to consider separately the results corresponding to different pressures.

a. $P=84$ atm. The temperature dependences of the thermal conductivity of three of the crystals grown at 84 atm are shown in Figs. 2 to 4. The great differences in the high-temperature parts of the $\varkappa(T)$ curves indicate, according to Eqs. (3), that the specimens studied consisted of one or several extended large single crystals whose orientations relative to the longitudinal axis of the tube remained constant over fairly large distances. The dominant orientation of the crystals could change appreciably from specimen to specimen. The scatter in the slopes of the high-temperature parts of the $\varkappa(T)$ curves in Fig. 2 to 4 correlates well with the results of earlier observations.[4,6]

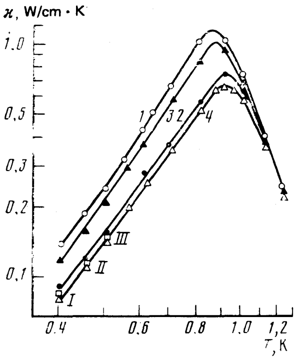

FIG. 2. Temperature dependences of the thermal conductivity of a specimen grown at 84 atm, before (curve 1) and after (2–4) plastic deformation at 0.45 K. Curve 2—a single bending, 3—after annealing of the bent specimen at 2.5 K; curve 4—after two bending and unbending cycles. Points I, II, III (□) show the change in thermal conductivity of the annealed specimen after bending.

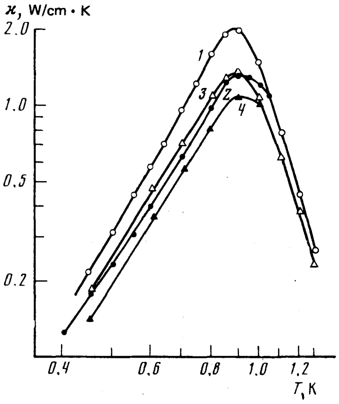

FIG. 3. The effect of plastic deformation and thermal shock on the thermal conductivity of the best of the crystals grown at 84 atm. Curve 1—the initial crystal, 2—after bending at 0.45 K; curve 3—thermal conductivity of the same specimen after thermal shock and then, 4—after a bending and unbending cycle.

Curve 1 in Fig. 2 corresponds to the thermal conductivity of the initial specimen. Below the maximum, the slope of the low-temperature part of the curve is close to $\varkappa(T)\sim T^3$. Curve 2 shows the $\varkappa(T)$ dependence after bending the initial specimen at $T=0.45$ K, measured as the temperature was raised in steps from 0.4 to 1.1 K. Subsequent continuous heating to $T\sim2.5$ K for 9 hours led to a noticeable increase in $\varkappa$ (curve 3). After cooling to 0.4 K, the tension in the string was gradually reduced and the tube unbent to its initial radius. The thermal conductivity of the specimen was again reduced (points I, II and III under curve 2), i.e., bending and unbending of the tube had roughly the same influence on the thermal conductivity (they introduce the same num-

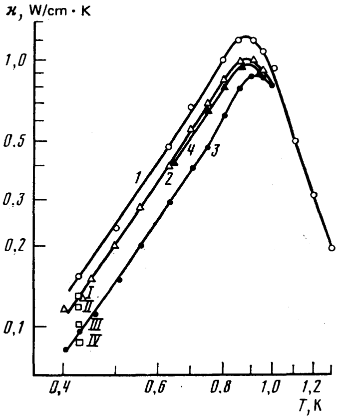

FIG. 4. The effect of the degree of deformation on the thermal conductivity of a specimen at $P=84$ atm. Curve 1—initial specimen. Points I to IV (□) show the changes in thermal conductivity at 0.43 K after, respectively, 1, 2, 5, and 10 bending and unbending cycles. Curve 2 was measured after heating up to 0.95 K; curve 3—after repeated deformation (10 cycles at 0.45 K) with step-wise heating from 0.4 to 1 K, curve 4—the behavior on cooling from 1 to 0.4 K.

ber of defects into the helium crystal). Subsequent additional deformation (bending and unbending at 0.45 K) led to only an insignificant reduction in thermal conductivity— curve 4.

The behavior of the thermal conductivity of the most perfect of the specimens grown at 84 atm is shown in Fig. 3 (curve 1). Curve 2 [the $\varkappa(T)$ relation after bending] was measured as the temperature was raised in steps from 0.4 to 1.05 K. For this specimen, unlike the specimen shown in Fig. 2, the slope of the $\varkappa(T)$ curves of the original and deformed crystal at $T \leqslant T_{\max}$ are steeper than cubic [and are close to $\varkappa(T) \sim T^4$ for the initial crystal], which is connected with the development of the transition regime in the phonon gas flow near $T_{\max}$. The diameter of the tube is quite insufficient for observing developed Poiseuille flow at this pressure, according to the conclusions of previous work.[4,12]

A rapid rise in the temperature of the specimen (thermal shock) to near the melting point T_m led to a reduction in the thermal conductivity over the whole temperature range (curve 3). The difference between the slopes of the high-temperature parts of curves 1 and 3 shows that severe reorientation of the crystallites occurs in the specimen as a result of thermal shock. The reduction in the values of $\varkappa$ for $T < T_{\max}$, when the phonon mean free paths are limited by scattering at the crystal boundaries and by defects, also points to this. It could be expected that more severe deformation would be required for a noticeable reduction in thermal conductivity of such an imperfect specimen that is the case for the initial perfect single crystal. However, it turned out that even a single bending and unbending at 0.45 K led to an appreciable reduction in its thermal conductivity (curve 4), i.e., the additional thermal resistance ΔW in a polycrystalline specimen resulting from the same degree of deformation is several times greater than the additional resistance arising on deforming a perfect crystal.

It was interesting in this connection to explain how the thermal conductivity of a fairly perfect initial crystal will change at constant temperature with an increasing number of bending and unbending cycles. The results of these investigations are shown in Fig. 4. The temperature dependence $\varkappa(T)$ of the initial crystal was close to cubic below $T_{\max}$. Points I, II, III, and IV show how the thermal conductivity of the specimen at 0.43 K changes after 1, 2, 5 and 10 consecutive bending and unbending cycles at 0.45 K. The additional thermal resistivity calculated from the change in thermal conductivity at 0.43 K, where hydrodynamic effects in thermal conductivity can be neglected, increases nonlinearly with increasing degree of deformation. When the number of bending and unbending cycles is increased to 18, the specific thermal resistivity referred to a single cycle decreased by ~3-fold compared with the first cycle. In what follows we shall therefore limit ourselves to 1 to 10 cycles at 0.45 K.

Slow warming of a strongly deformed specimen from 0.4 to 0.95 K over 9 hours led to a significant increase in its thermal conductivity: curve 2, measured at $T = 0.43$ K on cooling from 0.95 to 0.4 K, passes through point I, corresponding to a single bending and unbending of the initial crystal. A subsequent long anneal at $T \leqslant 0.95$ K also led to partial recovery of the thermal conductivity. In order to elucidate how much the annealing time at these temperatures affect the thermal conductivity behavior of a deformed specimen, we again deformed it at 0.45 K (10 cycles) and carried out measurements of $\varkappa(T)$ for stepwise heating to $T \approx 1$ K and subsequent cooling (curves 3 and 4). On average, the time spent in measuring one point was 10 min, so that the whole of curves 3 was measured in 2 h, while the crystal was at temperatures of 0.95 to 1 K, where the most intensive annealing occurred, for about 30 min. Curve 4 practically coincided with curve 2, i.e., it was sufficient for reducing the additional thermal resistivity several fold to maintain the crystal deformed by bending for tens of minutes at $T \approx 0.3\ T_m$.

We should note that the slopes of the low-temperature parts of curves 1, 2 and 4 in Fig. 4 are close to $\varkappa(T) \sim T^3$. The bend in curve 3 near $T_{\max}$ is explained by the partial annealing of defects during the measurement as the temperature of a severely deformed specimen is raised near 1 K. Since it is natural to suppose that the rate of annealing falls exponentially with decreasing temperature, it would be more correct when discussing the temperature dependences $\varkappa(T)$ for deformed crystals to use curves obtained on cooling, or during heating in the temperature range where annealing for the duration of the measurement of the curve is negligible. Complete return of the thermal conductivity of the deformed specimen of Fig. 4 to the initial value took place after raising its temperature to 1.7 K, i.e., holding the specimen at $T = 0.6\ T_m$ for 10 to 20 min was quite sufficient for practically complete annealing of the defects introduced by bending.

b. $P = 31$ atm. Figure 5 shows the results of studying the behavior of the thermal conductivity of one of the specimens grown at 31 atm. Curve 1 of Fig. 5a shows the thermal conductivity of the initial crystal. The $\varkappa(T)$ dependence below $T_{\max}$ is stronger than cubic and this is related to the presence of a transition phonon gas flow region: in the region $T \leqslant T_{\max}$ the value of $l_{\text{eff}} > d$ and the temperature dependence of $\varkappa(T) \sim T^{4.5}$. Judging by the temperature dependences $\varkappa(T)$ above and below $T_{\max}$ and Eqs. (3) to (3b), this specimen was a perfect single crystal with the C_3 axis at an acute angle to the longitudinal axis of the tube.

The behavior of the thermal conductivity of the deformed specimen (bent and unbent once at 0.45 K) with stepwise increase in its temperature is shown in curve 2. The specimen was then cooled again to 0.4 K and annealed by slow heating from 0.4 to 0.92 K over 9 h. Heating produced complete recovery of the thermal conductivity to its initial value (the points coincided with points on curve 1).

The annealed crystal was severely deformed again (10 cycles at 0.45 K) and measurements of $\varkappa(T)$ were made during subsequent raising of the temperature from 0.4 to 0.7 K (curve 3) and lowering of it (curve 4). It can be seen that partial annealing took place during measurements of curve 3. The results of subsequent

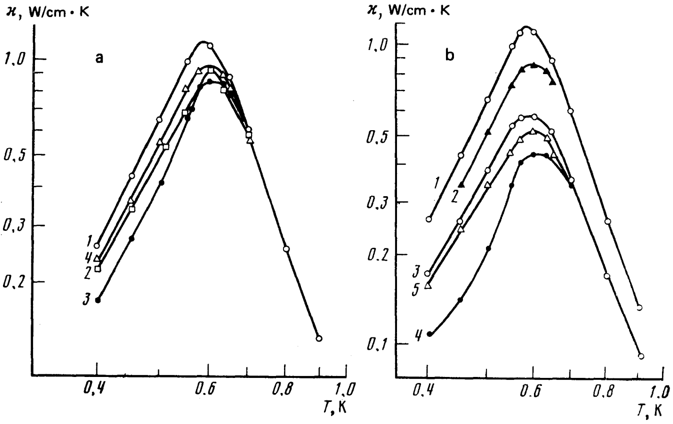

FIG. 5. The effect of bending and thermal shock on the thermal conductivity of a crystal grown at 31 atm. a) Curve 1—the initial crystal, 2—after a single bending and unbending cycle at 0.45 K (slow raising of temperature); curves 3 and 4—after severe deformation (10 cycles at 0.45 K) on raising and lowering the temperature in the range 0.4 to 0.7 K. b) Curve 1—after annealing at 1.4 K coincides with curve 1 of Fig. 1a; curve 2—after 10 cycles of bending and unbending at 0.45 K and heating to 0.70 K; curves 3 to 5—thermal conductivity of a specimen after thermal shock and subsequent deformation (10 cycles) going up (4) and down (5) in temperature in the range 0.4 to 0.7 K.

measurements of thermal conductivity going up and down in temperature over the range from 0.4 to 0.7 K practically coincided with the points on curve 4, i.e., after annealing for 10 to 20 min at $T = 0.7$ K, the thermal conductivity of a deformed specimen below 0.7 K changes slowly with time. It appeared to be sufficient for practically complete recovery of the thermal conductivity to the initial value to maintain the specimen at $T = 1.4$ K for tens of minutes.

The next cycle of measurements of the properties of the same crystal is shown in Fig. 5b. Curve 1 (the thermal conductivity of a well annealed specimen) coincides with curve 1 of Fig. 5a. The specimen was then severely deformed again (10 cycles at 0.45 K) and the changes in thermal conductivity with time at the three fixed temperatures 0.55, 0.60 and 0.65 K were tracked. It was found that after a time ≈ 50 min the thermal conductivity at each temperature increased by less than 20%, i.e., the annealing rate for the crystal at $T \leqslant 0.65$ K is negligbly small compared with annealing for $T \geqslant 0.70$ K. One should therefore judge the $\varkappa(T)$ dependence at $T \leqslant T_{\max}$ for a deformed specimen either from a heating and cooling curve in the range 0.4 to 0.65 K or from the $\varkappa(T)$ dependence measured on cooling after heating to $T \geqslant 0.70$ K, for example curve 2 measured on cooling the specimen after heating to 0.7 K. It can be seen that the $\varkappa(T)$ dependence for $T \leqslant T_{\max}$ in the weakly deformed specimen of Fig. 5a and 5b is stronger than cubic, as for the initial state.

Thermal shock led to a reduction in the thermal conductivity of the specimen (Fig. 5b, curve 3), as for a perfect 84 atm crystal (Fig. 3), and to changes in the slopes of the high-temperature and low-temperature parts of the $\varkappa(T)$ curve. Slow warming of the specimen over 10 h from 0.4 to 1.05 K did not change the form of curve 3, which was different from the heating of bent specimens, i.e., appreciably higher temperatures are required to anneal the polycrystalline specimen which is formed as a result of shock.

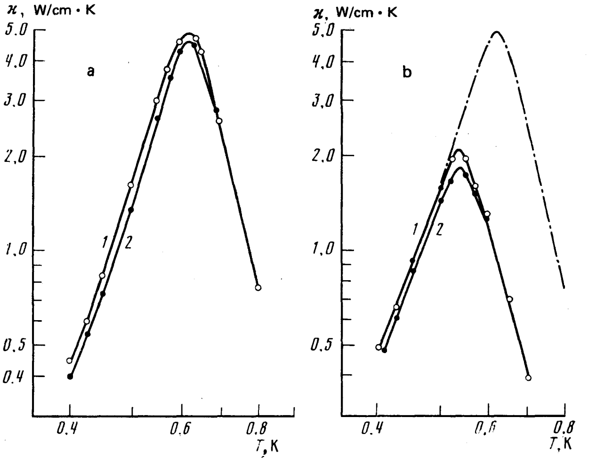

FIG. 6. The thermal conductivity of two specimens grown at 26 atm: 1—initial, 2—crystal deformed by bending (10 cycles at 0.45 K). The dashed curve in Fig. b shows the thermal conductivity of specimen *a*.

Plastic deformation of a polycrystalline specimen (10 bending and unbending cycles at 0.45 K) produced a considerable reduction in its thermal conductivity. Curve 4 was measured on heating from 0.4 to 0.7 K and curve 5 on subsequent cooling. The additional thermal resistance ΔW arising on plastic deformation of a polycrystalline specimen is much greater than the resistance produced by similar deformation of the initial crystal, as for deforming specimens grown at 84 atm.

c. $P = 26$ atm. The behavior of two specimens grown from superfluid liquid is shown in Fig. 6. Judging by the slopes of the low-temperature sections, $\varkappa(T) \sim T^6$, and also by the calculated $l_{\rm eff}$ (for a crystal with the largest thermal conductivity $l_{\rm eff} = 4$ mm $= 4d$ at the maximum), both specimens were perfect single crystals and the variation in their thermal conductivities in the range $T \leqslant T_{\max}$ is determined by the appearance of Poiseuille flow of the phonon gas when a temperature gradient is applied. The differences in the slopes of the high-temperature parts of the $\varkappa(T)$ curves and in the maximum values of $l_{\rm eff}$ are, according to calculations using Eq. (3), related to differences in the crystal orientation relative to the axis of the tube.

It can be seen from Fig. 6 that plastic deformation of the specimens (10 cycles of bending and unbending at 0.45 K) has little effect on their thermal conductivity (curve 1 is for the initial crystal and curve 2 after deformation, measured on raising the temperature from 0.4 K). Thermal shock, as a result of which the lower part of the crystal, shown in Fig. 6a, even melted, also had practically no effect on its thermal conductivity. This appreciably differentiates the properties of 26 atm crystals from the properties of crystals grown at 31 and 84 atm.

§4. DISCUSSION OF THE RESULTS

We brought out, when presenting the problems for investigation, a number of questions which could only be answered by experiment. We shall discuss them in order.

1. Can a perfect ^{4}He crystal be grown in a bent tube? Can one, in principle, observe the onset of Poiseuille flow of the phonon gas in a 1 mm diameter specimen. Both these questions can now be answered in the affirmative. The temperature dependences of the effective phonon mean free path, $l_{eff}(T)$, for the three best crystals grown at pressures $P = 26$, 31 and 84 atm are shown in Fig. 7 (curves 1, 2 and 3). It can be seen that near T_{max} the effective length $l_{eff} \geqslant d$ and decreases with decreasing temperature, i.e., the best of the specimens consisted of only one or of several perfect large crystals of diameter $d = 1$ mm. The appearance of the hydrodynamic mechanism of thermal conductivity[5] could be observed in all these specimens: from fully developed Poiseuille flow at $P = 26$ atm ($l_{eff} \gg d$ at the maximum) to something closer to the Knudsen transition regime for $P = 84$ atm ($l_{eff} \gtrsim d$). The slope of the high-temperature parts of the $\varkappa(T)$ curves can differ markedly from specimen to specimen grown at the same pressure (Figs. 2 to 6). The range over which the slopes of the $\varkappa(T)$ curves vary is no less than for specimens grown in straight tubes at the same pressure in earlier work.[4,6] According to Eqs. (3) to (3b) this indicates that in a tube bent into a circle, as in straight tubes, rather perfect single crystals can be obtained with constant orientation relative to the longitudinal axis of the tube.

We showed that according to earlier estimates,[4,12] fully developed Poiseuille flow in perfect crystals for $P \geqslant 80$ atm could only be observed in specimens with diameter $d \geqslant 2.5$ mm, and the phonon mean free path for normal processes, $l_N = 3 \times 10^{-9}(T/\Theta)^{-4}$ cm in crystals grown at 85 atm is almost independent of crystal orientation, unlike the phonon mean free path l_U for umklapp processes. These calculations agree well with the $l_{eff}(T)$ dependences observed in the present work for $P = 84$ atm (curves 3 of Fig. 7).

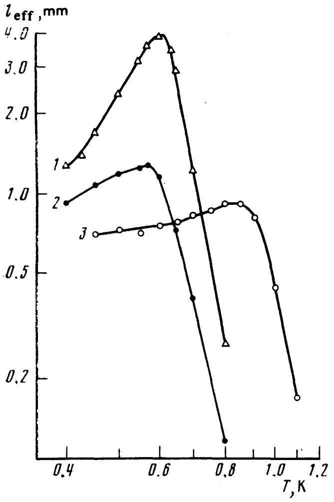

FIG. 7. The temperature dependence of the effective phonon mean free path for the three best crystals grown at pressures of 26 atm (curve 1), 31 atm (2) and 84 atm (3).

With a reduction in pressure to 26 atm, Poiseuille flow of the phonon gas could even be observed for $d = 1$ mm (curves 1 of Figs. 6a and 7). Assuming that in the region $T \approx T_{max}$

$$l_{eff}=0.1\,d^2/l_N,$$

i.e., conditions (6), which are sufficient for the appearance of developed Poiseuille flow, are satisfied and expressing $l_N \sim (T/\Theta)^{-4}$ as earlier, we obtain for $P = 26$ atm

$$l_N=0.5\cdot10^{-9}\,(T/\Theta)^{-4}\ \text{cm}$$

is 6 times less than for $P = 84$ atm.

The increase in the degree of anharmonicity of the lattice of a quantum crystal with decreasing pressure manifests itself in kinetic phenomena by an increasing frequency of phonon-phonon interactions, i.e., in a reduction in the corresponding phonon mean free path for the same reduced temperatures T/Θ. This makes it easier for hydrodynamic effects to be observed in solid helium. Near P_{min} in the region of developed Poiseuille flow and then in the region of Knudsen flow of the phonon gas (Fig. 6), the thermal conductivity of perfect crystals is close to the thermal conductivity of the superfluid in a millimeter capillary[16] at temperatures $T \leqslant 0.6$ K, where phonon-roton scattering and the contribution of the roton component can be neglected, i.e., the frequency of normal phonon-phonon interactions in solid helium for $P \approx P_{min}$ is close to the frequency of normal processes between phonons in liquid He II at the same reduced temperatures.

2. The experiments carried out showed unambiguously that plastic deformation of hcp ^{4}He crystals, brought about by single or repeated bending of the tube, can reduce the thermal conductivity of specimens appreciably at temperatures close to and below T_{max}. For the same degree of deformation and a fixed temperature of measurement, the additional thermal resistivity ΔW depends appreciably on the temperature at which the deformation was carried out, on the subsequent temperature regime (maintenance at a constant temperature, heating or cooling of the specimen) and on the properties of the initial specimen (solidification pressure, degree of perfection). The magnitude of ΔW increases with decreasing deformation temperature, increasing pressure at which the crystal was grown and with worsening specimen quality.

Obviously, differences in the thermal conductivity of the initial specimens grown at constant pressure can be connected with differences not only in orientation, but also in the degree of perfection of the crystals comprising the specimen, or even on the quality of the upper and lower parts of one and the same specimen.[4] But heating such specimens to a temperature $T = 0.8$ to $0.9\ T_m$ at a rate of 1 K per hour did not lead to annealing of the defects formed during growth. Unlike such defects, the defects introduced into the body of the crystal in the plastic deformation process could be annealed at a noticeable rate at much lower temperatures. As can be seen from Figs. 2 to 4, the thermal conductivity of specimens grown at the same pressures as before,[4] and deformed by bending at 0.45 K, is raised appreciably by heating to 1 K ($T \leq 0.3\ T_m$) and subsequent cooling. Moreover, for almost complete recovery of the thermal conductivity to the initial (before deformation) value, it turned out to be sufficient to maintain the specimen for $\leq 10^3$ s at a temperature $T_0 = 0.6$ to $0.8\ T_m$, i.e., the mobility of the defects introduced by the bending was considerably greater than the mobility of defects produced during growth.

Bending crystals at temperatures $T \geq 0.8\ T_m$ did not affect their thermal conductivity since the crystal manages to anneal during the time for the measurement.

Non-uniform mechanical stress arising when the temperature is varied rapidly with an amplitude of the order of 1 K (thermal shock) leads, as was known,[7] to a reduction in the thermal conductivity of crystals grown at pressures $P \geq 31$ atm over the whole temperature range (Figs. 3, 5b). It follows from Eqs. (3) to (3b) that this implies that instead of perfect initial crystals, specimens of several misoriented crystallites of mean cross-section $\lesssim 0.3d$ are formed. As before,[4] we could not observe annealing of the polycrystalline specimens on heating above $0.8\ T_m$. At the same time, bending of polycrystalline specimens (curves 3, 4 and 5 in Figs. 3 and 5b) produced defects which annealed completely at temperatures $T_0 \leq 0.8\ T_m$, i.e., the mobility of the defects introduced by bending was much greater than the mobility of the large-angle boundaries between the polycrystals.

The properties of crystals were radically altered by lowering the pressure to 26 atm. Bending at 0.43 K led to only a small change in thermal conductivity. It was sufficient to heat specimens to 0.65 K for complete annealing (Fig. 6). The thermal conductivity of the crystal in Fig. 6 after thermal shock practically coincided with the initial values, although the lower part of the crystal was melted as a result of the shock, so that the jump in pressure in the tube amounted to a few atmospheres. (The annealing of a polycrystalline specimen obtained at a pressure close to $P_{\min}$ by the blocked capillary method, on raising the temperature from 0.02 to 0.8 K was observed by Armstrong *et al.*[8]) All this shows that the mobility of both defects introduced by bending and of grown-in defects in solid helium near $P_{\min}$ is an order of magnitude greater than the mobility of defects in crystals grown at $P \geq 31$ atm.

It is reasonable to suppose that the principle diffusion mechanism for defects introduced by bending is thermally activated diffusion (at least for $P \geq 31$ atm and $T > 0.4$ K), so that the mobility of the defects introduced increases exponentially and the concentration falls with increasing temperature. This agrees qualitatively with the observed dependence of ΔW on the temperature of deformation and on the time for which the specimen is maintained at constant temperature. We hope to carry out soon detailed quantitative measurements of the annealing dynamics and derive the characteristic activation energies of diffusion of the defects.

3. We can now try to answer the question of what defects accumulate in the specimen in the bending process and what mechanism of phonon scattering by the defects is dominant. As was pointed out in Sec. 3 of the Introduction, analysis of the $\varkappa(T)$ behavior of deformed crystals under conditions when normal phonon-phonon interactions are the most frequent, is complicated and ambiguous since the momentum exchange between phonons with different wave vectors under normal collisions considerably influences the effectiveness of phonon scattering by defects. We shall, therefore, consider the effectiveness of bending on the thermal conductivity of specimens grown at $P = 84$ atm when $l_N \geq d$ below $T_{\max}$ and normal processes can be neglected (Figs. 2 to 4). Bending reduced the thermal conductivity in the region $T \leq T_{\max}$, the slopes of the high-temperature parts of the $\varkappa(T)$ curves then remained unchanged [except for the change in $\varkappa(T)$ on thermal shock], while the mobility of the defects introduced by bending is much higher than the mobility of grain boundaries. This indicates that the change in conductivity is produced by an increase in the concentration of vacancies or dislocations (single or forming low-angle boundaries). For scattering by static defects[13]

$$l_{pi} = 5 \cdot 10^{-2} a x^{-1} (T/\Theta)^{-4} \text{ cm}$$

for scattering of phonons by vacancies (a is the interatomic distance and x the relative concentration of vacancies) and

$$l_{pi} = 0.6 \cdot 10^{6} N_g^{-1} (T/\Theta)^{-1} \text{ cm}$$

for scattering by single dislocations with density N_g and Burgers vector close to the value of a. Substituting here $T = 0.5$ K, $l_{pi} = 3 \times 10^{-2}$ cm, we obtain $x \geq 0.5$, $N_g \geq 10^9$ cm^{-2}, which is much higher than any sensible estimate of the concentration of vacancies and dislocations introduced by bending a crystal on a 3 cm radius template ($N_g \sim 10^7$ cm^{-2}), and also the temperature dependences $l_{\text{eff}}(T)$ below $T_{\max}$ in the deformed specimens are appreciably different from those indicated.

Both in the initial and in the crystals deformed by bending, the $\varkappa(T)$ dependences below $T_{\max}$ in Figs. 2 to 4 are close to cubic (or slightly steeper), i.e., the additional thermal resistivity $\Delta W \sim T^{-3}$ while the phonon mean free paths limited by scattering at the defects are constant or decrease slightly as the temperature is reduced to 0.4 K. It follows from what was said in Sec. 3 of the Introduction that dynamic scattering of phonons by vibrating dislocations (flutter effect) or scattering by the walls of dislocations, if the mean distance between dislocations in the wall is much less than 10^{-6} cm, must constitute the dominant effect in the given specimens.

The density of dislocations introduced by bending could be sufficient to form small-angle boundaries, distributed at distances of 0.3 mm from one another and consisting of dislocation networks with a mean mesh size $\sim 10^{-6}$ cm. Most likely, in view of the features of the behavior of the thermal conductivity on annealing carried out at 0.43 K, the dislocations are at first distributed chaotically throughout the volume, and as the temperature is raised an ordering takes place and less mobile small-angle boundaries are formed. In fact, annealing of deformed crystals takes place in two stages for $P \geqslant 31$ atm. In the first stage on heating from 0.4 K to T_{max} only partial annealing takes place: $\varkappa(T)$ curves measured on going up to $T \approx T_{max}$ and on subsequent lowering of the temperature differ noticeably (curves 2 and 3 of Figs. 2 and 3 and curve 4 of Figs. 4 and 5a). After heating to a temperature $T \approx T_{max}$ the thermal conductivities of a singly and multiply deformed specimen almost coincide (curves 2 and 4 of Fig. 4). Heating to a higher temperature, $T_0 = 0.6$ to $0.8\ T_m$, depending on pressure, is essential for complete return of the thermal conductivity to its initial value (curve 1). The same picture is observed on repeating the deformation and also for bending polycrystalline specimens.

The difference between the behaviors of freshly introduced and 'aged' dislocations on partial annealing can be explained by phonons in the first case being scattered by vibrating single dislocations and in the second case by networks of pinned dislocations accumulating in the slip planes and forming low-angle boundaries, i.e., on partial annealing at temperatures $T \sim T_{max}$ it is not so much the density of introduced defects which changes as their configuration and the corresponding mechanism of phonon scattering by defects. The results of measuring the dependence of ΔW on the number of bending and unbending cycles at a constant temperature of 0.43 K (Fig. 8) also supports this suggestion. On raising the total number of cycles from 1 to 18 the specific thermal resistivity at 0.45 K calculated per cycle decreases by ~3-fold. This can be explained within the framework of the theory of the flutter effect[15] either by partial pinning of the oscillating dislocations as a result of their mutual intersection, or because of conservation laws only transversely polarized phonons interact effectively with vibrating dislocations, so that for a dislocation density above a certain limit, increasing their number does not lead to a growth in resistivity.

The study of the influence of plastic deformation on the thermal conductivity of ^{4}He crystals also makes it possible to understand the reasons for the differences in behavior of crystals grown in a bare cylindrical tube and in a tube with leads brought inside or with quartz crystals etc.[10,11] The mechanical stresses arising inside on changing the temperature are especially great near T_m, but the mobility of dislocations introduced by plastic deformation is high in this region so that the stresses can rapidly relax. Unlike grown-in defects, the mobility of defects introduced by deformation is fairly high down to a temperature $T \approx 0.3\ T_m$, so that for sufficiently slow cooling (or heating) the defects arising because of the differences in coefficients of thermal expansion of the tube and the specimen are able to anneal, and the degree of perfection of the crystal is unchanged. The rate of change of temperature of a tube with electrodes inside must be much less than the rate of cooling or heating of a bare tube, since the nonuniform mechanical stresses which arise at the edges of the electrodes promote an appreciable increase in the concentration of defects introduced. Correspondingly, for the same time of maintenance, the annealing temperature for such specimens must be noticeably higher. This agrees well with the results of observations of the creation of defects at $T \leqslant 0.5\ T_m$ and their annealing at $T \geqslant 0.8 T_m$ in experiments with ions,[10] and ultrasonic measurements[11] with the temperature changing at a rate of ~1 K per hour.

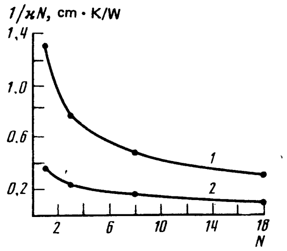

FIG. 8. The dependence of the magnitude of the additional thermal resistivity per cycle on the number of bending and unbending cycles N at $T = 0.43$ K. Curve 1—$P = 84$ atm, 2—$P = 31$ atm.

The authors are grateful to V. N. Khlopinskiĭ for help in preparing and carrying out the experiments.

[1] R. Berman, Thermal Conduction in Solids, Oxford University Press (1976).

[2] A. F. Andreev, Usp. Fiz. Nauk **118**, 251 (1976) [Sov. Phys. Usp. **19**, 137 (1976)].

[3] L. P. Mezhov-Deglin, Prib. Tekh. Eksp. No. 3, 217 (1971); [Instrum. & Exp. Tech. (USA) **14**, 911 (1971)].

[4] L. P. Mezhov-Deglin, Zh. Eksp. Teor. Fiz. **49**, 66 (1965), **52**, 866 (1967), **71**, 1453 (1976) [Sov. Phys. JETP **22**, 47 (1966), **25**, 568 (1967), **44**, 761 (1976)].

[5] R. N. Gurzhi, Usp. Fiz. Nauk **94**, 689 (1968) [Sov. Phys. Usp. **11**, 255 (1968)].

[6] R. Berman, C. R. Day, D. P. Goulder, and J. E. Vos, J. Phys. C **6**, 2119 (1973).

[7] A. A. Golub and S. V. Svatko, Fiz. Nizk. Temp. **6**, 957 (1980), **6**, 1112 (1980), **7**, 413 (1981); Sov. J. Low Temp. Phys. **6**, 465 (1980), **6**, 540 (1980), **7**, 203 (1981).

[8] G. A. Armstrong, A. A. Helmy, and A. S. Greenberg, Phys. Rev. **B20**, 1061 (1979).

[9] V. B. Efimov and L. P. Mezhov-Deglin, Fiz. Nizk. Temp. **2**, 652 (1976) [Sov. J. Low Temp. Phys. **2**, 322 (1976)].

[10] V. B. Efimov and L. P. Mezhov-Deglin, Fiz. Nizk. Temp. **8**, 5 (1982); [Sov. J. Low Temp. Phys. **8**, No. 1 (1982)].

[11] I. Iwasa, K. Araki, and H. Suzuki, J. Phys. Soc. Jpn. **46**, 1119 (1979).

[12] L. P. Mezhov-Deglin, Fiz. Tverd. Tela (Leningrad) **22** 1748 (1980) [Sov. Phys. Solid State, **22**, 1018 (1980)].

[13] J. M. Ziman, Electrons and Phonons, Clarendon Press, Oxford (1960).

[14] V. S. Oskotskiĭ and I. S. Smirnov, Defekty v kristallakh i teploprovodnost' (Defects in Crystals and Thermal Conductivity), Nauka, Leningrad (1972).

[15] F. L. Madarasz and P. G. Klemens, Phys. Rev. **B23**, 2553 (1981).

[16] R. W. Whitworth, Proc. R. Soc. London Ser. A **246**, 390 (1958).

Translated by R. Berman