Heat transfer in solid parahydrogen and possibility of observing a Poiseuille flow of phonons

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The thermal conductivity of solid parahydrogen has been measured over the temperature interval 1.5–8 K. The temperature dependence of the thermal conductivity and the size of the effective mean free path of the phonons indicate that a Poiseuille flow of phonons can be observed in solid parahydrogen.

Two phenomena observed in high-quality crystals illustrate the quantum-mechanical nature of excitations of a crystal lattice (phonons). These two phenomena, the Poiseuille flow of phonons and second sound, can be observed only in high-quality crystals, in which resistive phonon-scattering processes result from only the boundaries of the crystal and from the phonon-phonon interactions accompanied by a change in quasimomentum (U-processes). A theory for the Poiseuille flow of phonons was derived in Refs. 1–3. The idea is that in crystals of exceptionally high quality the normal three-phonon scattering processes (N-processes), which occur without a loss of quasimomentum, may dominate the picture to such an extent that the motion of phonons becomes analogous to a laminar flow of a gas of colliding molecules. As a result, phonons colliding with each other can traverse a distance substantially greater than the distance between the boundaries. The phonons "no longer see the wall." The conditions for the observation of the Poiseuille regime are the inequalities

$$l_N \ll D, \qquad l_N l_R \gg D^2,$$

where l_N is the mean free path of the phonons in normal scattering processes, l_R is that in resistive processes, and D is a characteristic dimension of the crystal. A completely concrete consequence of this condition is that the thermal conductivity increases faster than T^3 with increasing temperature. The characteristic temperature dependence of the regime of a Poiseuille flow of phonons was first observed in a study of solid helium.[4] This phenomenon has yet to be observed in crystals of other substances, despite high quality of the samples. The reasons for this difficulty stem from the low intensity of the normal processes, itself a consequence of the inadequate degree of anharmonicity of the molecular interaction potential of these substances. Solid hydrogen follows helium on the list in order of values of the translational de Boer parameter. This parameter characterizes the quantum nature of the behavior of a crystal. However, most studies of solid hydrogen have been aimed at determining the behavior of molecules with a nonzero rotational angular momentum (orthohydrogen) in a crystal; the quality of the crystals has not been a matter of importance.

The study which we are reporting here demonstrates that it is possible to observe second sound and the Poiseuille flow of the phonon gas in solid parahydrogen. The thermal conductivity of a sample of solid parahydrogen was measured by the plane steady-state method. The experimental apparatus is described in Ref. 5. The sample was prepared in the following way: Chemical impurities were removed from the hydrogen by passing the gas through a palladium filter. The hydrogen was then exposed to the catalyst $Fe(OH)_3$ at liquid-hydrogen temperature. The hydrogen was then evaporated into a special glass flask, from which it was transferred to the measurement cell. The sample was crystallized from the gas phase at a pressure of 35 Torr. The birefringence revealed that the sample was a single crystal. The error of the measurements of the thermal conductivity was less than 20%.

Figure 1 shows the temperature dependence of the thermal conductivity according to our measurements. The maximum value of the thermal conductivity is roughly three times the value found in Ref. 6; this result is testimony to the high quality of the crystal. Figure 2 shows the effective phonon mean free path calculated as a function of the temperature. In these calculations we used the simple gas-kinetic expression

$$K = \frac{1}{3} C_V v l,$$

where K is the thermal conductivity, C_V is the specific heat,[7] and v is the sound velocity. To the left of the maximum in the thermal conductivity, the phonon mean free path assumes a constant value ~1.5 mm. The only cryocrystal for which such large mean free paths (and such values of the thermal conductivity) have been ob-

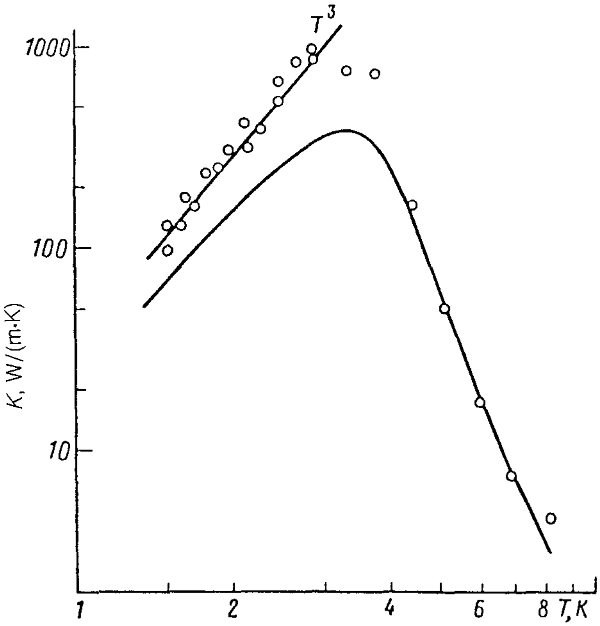

FIG. 1. Temperature dependence of the thermal conductivity of samples of crystalline parahydrogen. Lines—Data of Ref. 6; O—data of the present study.

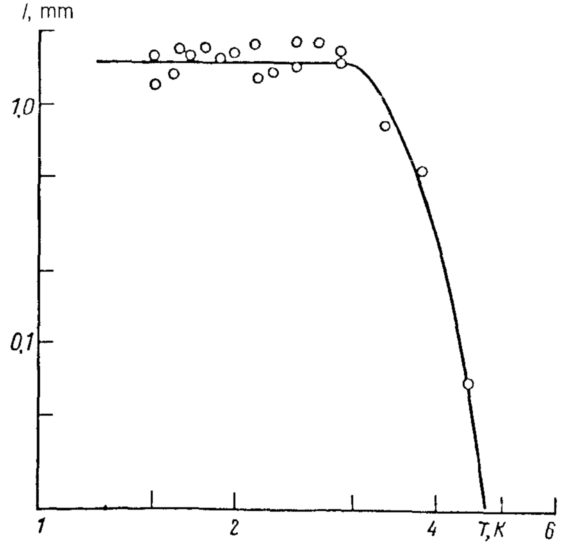

FIG. 2. Temperature dependence of the effective phonon mean free path (according to the results of the present study).

served previously is helium. A distinctive feature of the Poiseuille regime is the presence of a peak on the $l(T)$ curve. This peak stems from the small value of the mean free path for N-processes, which is on the order of 0.1 mm for parahydrogen at 3 K (Ref. 5). If this regime is not realized, then we know that the mean free path assumes a constant value as the temperature is lowered. This constant value corresponds to the cross-sectional size of the sample. In other words, the resistive processes in the sample in our case stem from boundary scattering alone. Since the mean free path is smaller than the diameter of the crystal, the boundary scattering is probably a consequence of a substructure of the crystal. We were not able to determine the impurity composition, in terms of either isotopic composition or orthomolecules, in the crystal which we used. Nevertheless, estimates of the scattering by an isotopic impurity, for the natural isotopic abundance of hydrogen, lead to a thermal resistance about two orders of magnitude higher than that found for our test sample. The scattering of phonons by orthohydrogen molecules at a concentration of 0.21% (Ref. 8) is equivalent, in terms of the mean free path, to the dimensions of the test crystal (which was 7 mm in diameter).

When a high structural quality is achieved in a parahydrogen crystal (through a decrease in the transverse dimensions of the crystal, for example), there can thus be a regime of a Poiseuille flow of phonons. Values $l_R \sim 10$ mm and $D \sim 1$ mm are necessary here. The results reported here demonstrate that achieving these values is completely feasible.

This problem is extremely attractive for further studies, since parahydrogen crystals differ from helium in that they are not under pressure and in this sense are less artificial.

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