response is attributed to an infrared-divergentlike response of dielectric excitations. Ngai also concludes²⁰ that other dynamic phenomena are a consequence of the same universal law.

¹M. Barmatz and R. Farrow, in *Proceedings of the Ultrasonics Symposium*, 1976, edited by J. de Klerk and B. McAvoy (Institute of Electrical and Electronics Engineers, New York, 1976), p. 62.

²R. E. Walstedt, R. S. Berg, J. P. Remika, A. S. Cooper, B. E. Prescott, and R. Dupree, in *Fast Ion Transport in Solids*, edited by P. Vashishta, J. N. Mundy, and G. K. Shenoy (North-Holland, Amsterdam, 1979), p. 355.

³J. H. Simmons, A. D. Franklin, K. F. Young, and M. Linzer, J. Am. Ceram. Soc. <u>63</u>, 78 (1980).

⁴D. P. Almond and A. R. West, in Proceedings of the Third International Meeting on Solid Electrolytes, Solid State Ionics and Galvanic Cells, Tokyo, 1981 (to be published).

 $^{5}\mathrm{R.}$ J. Grant, M. D. Ingram, and A. R. West, unpublished.

⁶A. K. Jonscher, Nature (London) <u>267</u>, 673 (1977). ⁷A. K. Jonscher, Nature (London) <u>256</u>, 566 (1975).

A. I. Souscher, Mature (London) 250, 500 (1915).

- ⁸A. K. Jonscher, J. Phys. D <u>13</u>, L89 (1980).
- 9 R. J. Grant, I. M. Hodge, M. D. Ingram, and A. R. West, Nature (London) <u>266</u>, 42 (1977).
- ¹⁰A. K. Jonscher, Colloid and Polym. Sci. <u>253</u>, 231 (1975).
- ¹¹T. J. Higgins, L. P. Boesch, V. Volterra, C. T.
- Moynihan, and P. B. Macedo, J. Am. Ceram. Soc. <u>56</u>, 334 (1973).

¹²P. B. Macedo, R. Bose, V. Provenzano, and T. A. Litovitz, in *Amorphous Materials*, edited by R. W.

- Douglas and B. Ellis (Wiley, New York, 1972), p. 251.
- ¹³M. S. Whittingham and R. A. Huggins, J. Chem. Phys. <u>54</u>, 414 (1971).

 $^{-14}$ C. Zener, Elasticity and Anelasticity of Metals (Univ. Chicago Press, Chicago, 1948).

¹⁵S. J. Allen, Jr., and S. P. Remeika, Phys. Rev. Lett. <u>33</u>, 1478 (1974).

¹⁶L. L. Chase, C. H. Hao, and G. D. Mahan, Solid State Commun. <u>18</u>, 401 (1976).

¹⁷A. K. Jonscher, J. Mater. Sci. <u>13</u>, 553 (1978).

- ¹⁸J. C. Wang, M. Gaffari, and Sang-il Choi, J. Chem. Phys. 63, 772 (1975).
- ¹⁹K. L. Ngai and C. T. White, Phys. Rev. B <u>20</u>, 2475 (1979).

²⁰K. L. Ngai, Comments Solid State Phys. <u>9</u>, 141 (1980).

Heat Transfer between Phonons and Quasiparticles in a Dilute Mixture of ³He in ⁴He

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A description is given of a novel technique for measuring the thermal resistance $(R_{\rm ph-qu})$ between phonons and quasiparticles in a bulk sample of dilute mixture of ³He in liquid ⁴He. For 0.1% ³He we find $VR_{\rm ph-qu}T^{6\cdot5} = 3.7 \times 10^{-11}$ m³ K^{7·5} W⁻¹ for 35 < T < 60 mK, in fair agreement with known interactions between phonons and quasiparticles. The discrepancy, however, suggests an additional, unidentified, inelastic scattering process.

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One of the outstanding questions concerning the thermal resistance between liquid helium and solids (the Kapitza resistance) is the apparent absence of the expected large resistance (R_{ph-qu}) between the ³He quasiparticles and the phonons in a dilute mixture of ³He in liquid ⁴He at millikelvin temperatures.¹ This resistance should be in series with the boundary resistance between the phonons in the helium and the phonons in the solid and with the thermal resistances within the solid. The absence of a large phonon-quasiparticle resistance has had important practical significance in that dilution refrigerators perform better than they should; it has also allowed the

measurement of the properties of dilute mixtures down to the millikelvin temperature range. However the anomaly is of interest in its own right, and is the subject of this Letter, in which we present the first measurement of this resistance alone in bulk helium.

It is expected that heat transfer between phonons at temperature $T_{\rm ph}$ and quasiparticles at temperature $T_{\rm qu}$ in a dilute mixture is brought about by two processes, phonon absorption and inelastic phonon scattering. The phonon absorption process has been considered in the context of phonon thermal conductivity and ultrasonic attenuation by Baym and Ebner (BE).² They also include phonon scattering, but this scattering can be regarded as essentially elastic in their work. For comparison with our experiment, we have constructed a theory of the thermal resistance $R_{\rm ph-qu}$ using the approach of Leggett and Vuorio³ and the phonon-quasiparticle interaction of BE. The scattering processes are strongly frequency dependent; hence the resultant heat transfer between phonons and quasiparticles is strongly temperature dependent, and relatively unimportant below about 50 mK, as is shown below. The absorption process is associated with the viscosity of the dilute mixture; the absorption rate is $\Gamma \propto P^2/\eta$, where *P* is the osmotic pressure and η the viscosity. For $T \leq T_F$, $\Gamma \propto T^2$; for $T \geq T_F$ with $P \propto T$ and $\eta \propto T^{0.81}$ for a 0.1% mixture,⁴ then $\Gamma \propto T^{1.2}$. When the absorption rate is included in a Debyetype summation over all phonon frequencies,^{5,6} the heat-transfer coefficient is proportional to T^{5} ($T^{4\cdot 2}$) for $T < T_{\rm F}$ ($T > T_{\rm F}$) or $R_{\rm ph-qu} \propto T^{-5}$ $(T^{-4}).$

There have in the past been many measurements of the total resistance between the quasiparticles in dilute mixtures and the cell that contains the helium.⁷ This total resistance has been compared with the expected resistance^{1,6} and found always to differ in magnitude and temperature dependence (too small and generally $\propto T^{-3}$). A universal feature of the experiments was that the dilute mixture was confined within the pores of either sintered metal powder or cerium magnesium nitrate crystallites and the pore sizes were comparable to or smaller than the mean free path for quasiparticle-quasiparticle scattering.

The present experiment was designed to measure R_{ph-qu} alone for a dilute mixture in a cell with all dimensions larger than the quasiparticle mean free path (typically 0.5 μ m at 30 mK). The sample cell was fitted with three thermometers to separately measure $T_{\rm c}$, $T_{\rm ph}$, and $T_{\rm qu}$, the temperatures of the cell wall, the phonons, and the ³He quasiparticles in the mixture. Heat was applied to the outside of the cell and the three temperatures monitored as the cell and dilute mixture approached equilibrium. Figure 1 shows an electrical analog. C_{qu} represents the heat capacity of the quasiparticles. The phonon and cell heat capacities can be neglected. The total thermal resistance between the guasiparticles in the cell and the dilution refrigerator is the sum of $R_{\rm ph-qu}$, the Kapitza boundary resistance $R_{\rm K}$, and the resistance of the link to the mixing chamber R_L . Also shown schematically in Fig. 1



FIG. 1. Electrical analog of the cell and the temperature-time curves following heat \dot{Q} sufficient to cause a step increase in T_c . \dot{Q}_i represents the heat leak or heat input to the mixing chamber.

are temperature-time curves following heating of the cell sufficient to cause T_c to rise by $\Delta T_c'$. T_{qu} rises with a time constant $\tau = C_{qu} (R_K + R_{ph-qu})$ and T_{ph} rises immediately by $\Delta T_{ph} = R_{ph-qu} \Delta T_c / (R_K + R_{ph-qu})$ and then rises to T_c' with the time constant τ . Following a step change in the heat supplied to the cell, as in this experiment, the behavior of T_c , T_{ph} , and T_{qu} is more complicated but R_{ph-qu} can be deduced from R_{ph-qu} $= \Delta T_{ph} / \dot{Q}_0$, where $\dot{Q}_0 = C_{qu} (dT_{qu}/dt)_{t=0}$.⁸

Typically, above 30 mK R_{ph-qu} is much smaller than $R_{\rm K}$ for 1.5% and 5% mixtures and hence $\Delta T_{\rm vh}/$ $\Delta T_c \ll 1$. $\Delta T_{\rm ph}$ would then have been difficult to measure. This was avoided by working with a more dilute mixture and with an enhanced boundary area in the cell. The mixture used was ⁴He +0.1%-³He. The cell was a copper cylinder of 6.6 mm i.d. The interface area was increased to 45 cm² by pressing 113 disks, 6.6 mm o.d. \times 4.4 mm i.d. \times 40 μ m, into the cylinder with an average spacing of 30 μ m. The quasiparticle thermometer was the second-sound velocity in the mixture; this was determined with a time-of-flight technique using standard Nuclepore porous-membrane transducers.⁹ These transducers were mounted on epoxy caps which sealed the two ends of the cell. The phonon thermometer was a 2×1 $\times 0.15$ -mm slice from a Matsushita $\frac{1}{8}$ -W 56- Ω carbon resistor suspended in the center of the cell by two 5-mm-long \times 75- μ m-diam Constantan

VOLUME 47, NUMBER 6

wires. Upon the basis of models for heat flow from solids into phonons and into quasiparticles in a dilute mixture we believe that the thermometer was in far better contact with the phonons than the quasiparticles. There is no known or postulated significant interaction that couples excitations in a solid to excitations of the quasiparticles in a dilute mixture.^{5,6} The cell thermometer was a Matsushita 56- Ω resistor.

Great care was taken, when admitting mixture to the cell, that the concentration of the mixture in the cell was the same as that of the original gas mixture. This included working with a helium sample volume (0.51 cm^3) that was less than the cell volume to keep the fill line empty. After the cell had been filled equilibrium values of second-sound velocity and phonon, cell, and calibrated Speer carbon thermometer resistances were made over the temperature range 35 mK to 1 K or so. The cell was then recooled and heating pulses applied while the thermometers were monitored.

The following technique was used to deduce R_{ph-qu} and to reduce the effect of calibration error:

$$R_{ph-qu} = \frac{\Delta T_{ph}}{C_{qu} (dT_{qu}/dt)_0} = \frac{\Delta r (dT/dr)}{C_{qu} (dT_{qu}/dt)_0}$$
$$= \frac{\Delta r (dT/dS) (dS/dr)}{C_{qu} (dT/dS) (dS/dt)_0},$$

where r is the phonon thermometer electrical resistance and S is the time of flight of the second-sound pulse. The calibration slopes (dT/dS)cancel. Thus $R_{\rm ph-qu}$ is derived from the initial rise in the phonon thermometer resistance Δr , the initial slope of time of flight versus time following the step in heat input, and the almosttemperature-independent heat capacity of the dilute mixture. Typically $\Delta T/T < 10\%$ and the time constants were ~ 1 min.

The measured values of $R_{\rm ph-qu}$ are shown in Fig. 2. The error bars reflect the uncertainty in the temperature calibration and its influence upon $R_{\rm ph-qu}$ as well as errors in the temperaturetime curves. The measured $R_{\rm ph-qu}$ can be described by $VR_{\rm ph-qu}T^{6.5} = 3.7 \times 10^{-11} \text{ m}^3 \text{ K}^{7.5} \text{ W}^{-1}$. The uncertainty in the exponent is given by 6.5 ± 1 .

The predictions of the theory are shown as the solid lines. The heat-transfer rate was calculated with use of the "golden rule"³ and the phononquasiparticle interaction of BE.¹⁰ The resulting thermal conductance $R_{\rm ph-qu}$ ⁻¹ consists of two terms. The curve labeled "Absorption" results



FIG. 2. The closed circles are R_{ph-qu} for ${}^{4}\text{He} + 0.1\%$ - ${}^{3}\text{He}$ as a function of temperature. The solid lines are discussed in the text. The open circles are the boundary resistance for ${}^{4}\text{He} + 5.4\%$ - ${}^{3}\text{He}$ and are compared with $ART^{3} = 2.5 \times 10^{-2} \text{ m}^{2} \text{ K}^{4} \text{ W}^{-1}$ (dashed line).

from the absorption/emission of single phonons with the excess momentum distributed among the quasiparticles via quasiparticle-quasiparticle interactions. The measured viscosity (n = 1.4) $\times 10^2 T^{0.81} \ \mu\text{P})^4$ for a 0.1% solution has been used to obtain, following BE, the necessary quasiparticle density correlation function. The curve labeled "Inelastic" results from the energy transfer involved in the quasielastic phonon scattering from the quasiparticles as described by the Tmatrix of BE. The resulting resistance from both processes acting in parallel is shown as the heavy line. The theory, which contains no free parameters, predicts a heat-transfer rate which is close to the experimental results at ≤ 40 mK but which is too small at higher temperatures. We conclude that an alternative and as vet unknown mechanism also contributes to the heat transfer. While the cell was designed to have no dimensions smaller than the quasiparticle mean free path at these temperatures we note that the experimental phonon attenuation length is $\sim 1 \text{ m}$ at 50 mK.

This experiment should be placed in context

with other experiments that have been used to study phonon-quasiparticle interactions in dilute mixtures. Thermal conduction in a dilute mixture above about 30 mK is by phonons and is limited by boundary and quasiparticle scattering. Below and above about 0.1 K the absorption and elastic scattering terms dominate, respectively. The agreement with BE seems satisfactory.^{11,12} The attenuation of second sound is related to the interaction between guasiparticles and phonons through a dependence upon the thermal conductivity, but also depends upon the viscosity and diffusion coefficient of the mixture.¹³ Some agreement with BE has been obtained above 0.3 K where elastic scattering determines the conductivity.¹⁴ Below 0.2 K the attenuation is not understood.¹⁵ None of the above work relates directly to the inelastic scattering of phonons. However, de Voogt et al.¹⁶ measured heat-pulse propagation through a series of dilute mixtures and from a pulse-shape analysis determined the thermal conductivity and rate of energy loss from the phonons. The thermal conductivity agreed with the earlier work¹² and by assuming a magnitude and temperature dependence of τ_n the energy transfer was fitted with the BE absorption process. There was evidence for two contributions to the phonon-quasiparticle coupling, proportional to x^2 and x, respectively (x is the concentration). We could now view these terms as absorption and inelastic scattering, respectively. Unfortunately it is hard to interpret the experiment since the initial heat pulse was probably ~1 K, presumably introducing nonlinearities. The phonon temperature was not measured. Finally, Kummer, Narayanamurti, and Dynes made heat-pulse measurements with a short path between heater and bolometer and with heater temperatures close to mixture sample temperatures.¹⁷ Ballistic phonon propagation at low temperatures changed to diffusive phonon and finally to second-sound propagation as the temperature was increased. A pulse-shape analysis gave the same phonon mean free path as would be measured in a thermal conductivity experiment, with good agreement.

We also performed experiments with 0.3% and 5.4% mixtures. For the 0.3% sample $\Delta T_{\rm ph}$ steps were observed only at the lowest temperatures indicating $R_{\rm ph-qu} \ll R_{\rm K}$ over most of the temperature range and so detailed work was not pursued. The 5.4% experiment was to obtain the Kapitza resistance $R_{\rm K}$ from the time constant τ . The results, shown in Fig. 2, are compared with the dashed line given by $ART^3 = 2.5 \times 10^{-2} \text{ m}^2 \text{ K}^4 \text{ W}^{-1}$. Although the slope is less than expected the magnitude is reasonable for a liquid-helium-copper interface.¹⁸

To conclude, we have made a direct measurement of the thermal resistance between phonon and quasiparticles in a dilute mixture and find a strong dependence upon temperature over the range 35-60 mK. Although the results agree reasonably well with our understanding of phononquasiparticle interactions there is a discrepancy that suggests that an inelastic process has been overlooked.

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¹F. Guillon and J. P. Harrison, in *Phonon Scattering in Condensed Matter*, edited by H. Maris (Plenum, New York, 1980), p. 157.

 ${}^{2}G.$ Baym and C. Ebner, Phys. Rev. <u>164</u>, 235 (1967). ${}^{3}A.$ J. Leggett and M. Vuorio, J. Low Temp. Phys. <u>3</u>, 359 (1970).

⁴K. A. Kuenhold, D. B. Crum, and R. E. Sarwinski, in *Low Temperature Physics*, *LT-13*, edited by K. D. Timmerhaus, W. J. O'Sullivan, and E. F. Hammel (Plenum, New York, 1974), p. 563.

 $^5 J.$ C. Wheatley, O. E. Vilches, and W. R. Abel, Physics (N.Y.) 4, 1 (1968).

⁶J. P. Harrison, J. Low Temp. Phys. <u>37</u>, 467 (1979). ⁷For references, see Sections 4.2 and 4.4 of Ref. 6. See also D. D. Osheroff and L. R. Corruccini, Phys. Lett. <u>82A</u>, 38 (1981); H. Godfrin, G. Frossati, B. Hebral, and D. Thoulouze, J. Phys. (Paris), Colloq. <u>41</u>, C7-275 (1980).

 $^{8}C_{\rm qu}$ was taken from R. Radebaugh, U. S. National Bureau of Standards Technical Note No. 362 (U. S. GPO, Washington, D. C., 1967).

⁹R. A. Sherlock and D. O. Edwards, Rev. Sci. Instrum. <u>41</u>, 1603 (1970); D. S. Greywall and G. Ahlers, Phys. Rev. A <u>7</u>, 2145 (1973).

¹⁰Details of the theoretical calculations will be published elsewhere.

¹¹W. R. Abel and J. C. Wheatley, Phys. Rev. Lett. <u>21</u>, 1231 (1968).

 $^{12}R.$ L. Rosenbaum, J. Landau, and Y. Eckstein, J. Low Temp. Phys. $\underline{16},\ 131\ (1974).$

¹³I. M. Khalatnikov, Zh. Eksp. Teor. Fiz. <u>23</u>, 265 (1952).

 $^{14}W.$ J. P. de Voogt and H. C. Kramers, Physica (Utrecht) $\underline{85B},~73$ (1977).

¹⁵D. S. Greywall, Phys. Rev. Lett. <u>42</u>, 1758 (1979).

¹⁶W. J. P. de Voogt, J. B. M. de Haas, J. Wiebes, and H. C. Kramers, Physics (N.Y.) 84B, 315 (1976).

¹⁷R. B. Kummer, V. Narayanamurti, and R. C. Dynes, Phys. Rev. B 16, 1046 (1977).

¹⁸J. T. Folinsbee and A. C. Anderson, J. Low Temp. Phys. <u>17</u>, 409 (1974).