²B. McCarrol, J. Chem. Phys. <u>50</u>, 4758 (1969).
³L. E. Brus and J. Comas, J. Chem. Phys. <u>54</u>, 2771

(1971).
⁴E. E. Huber, Jr., and C. T. Kirk, Jr., Surface Sci.
<u>5</u>, 447 (1966).

⁵Wm. H. Kreuger and S. R. Pollack, Surface Sci. <u>30</u>,

263 (1972).

⁶E. E. Huber, Jr., and C. T. Kirk, Jr., Surface Sci. <u>9</u>, 217 (1968).

⁷G. Herzberg, Molecular Spectra and Molecular Structure: I. Spectra of Diatomic Molecules (Van Nostrand, Princeton, N. J., 1950), 2nd ed. pp. 400-405.

⁸B. Kasemo and L. Wallden, to be published.

Evidence for the Role of Conduction Electrons in the Kapitza Resistance*

F. Wagner, † F. J. Kollarits, ‡ and M. Yaqub Department of Physics, The Ohio State University, Columbus, Ohio 43210 (Received 11 February 1974)

Measurements of the Kapitza conductance by the ac technique between 1.2 and 1.8 K are reported for thin oriented single-crystal disks of high-purity gallium. The data show convincing evidence that the conduction electrons play a dominant role in the transmission of heat across the metal-He-II interfaces.

Theoretical investigations of Kapitza conductance $h_{\rm K}$ between metals and liquid He are invariably treated on the basis of the Khalatnikov¹ model, which explains the resistance in terms of the acoustic transmission and reflection of thermal phonons and, above 1 K, obtains numerical values 10 to 50 times smaller than those observed experimentally. In an attempt to remove the disagreement, Little² suggested that the electrons in a metal interact with the surface phonons to provide an additional mechanism for heat transfer. Although their calculations of this mechanism did not alter the disagreement appreciably, Little³ and Andreev⁴ independently showed that in a free-electron metal the electronic contribution to h_{K} is comparable to that of the phonons. At the same time $Little^3$ pointed out that the calculations are based on several simplifying assumptions and therefore cannot agree quantitatively with real metals, in which band-structure effects can alter absolute magnitudes considerably. Nevertheless, experiments should be able to verify the calculations by measuring the change in $h_{\rm K}$ when a metal is magnetized from the superconducting to the normal state. Well below the transition temperature, electrons take no part in heat flow and a reduction in $h_{\rm K}$ should occur. The largest change is predicted for Pb, which has been studied by several investigators⁵ who have found $h_{\mathrm{Kn}}/h_{\mathrm{Ks}}$ to vary from 1.3 to 15. Hg, In, and Sn in general gave smaller ratios but these also differed from one experiment to another.⁵ Originally this was considered evidence for the electronic contribution. Subsequent work by

Cheeke,⁶ however, has shown that strain at the metal surface can result in spuriously large values for $h_{\rm Kn}/h_{\rm Ks}$ and that the differences in the two states are mainly due to this rather than to real differences in the transmission coefficient.

In his calculations Little³ also shows that the electronic contribution to $h_{\rm K}$ comes from two different mechanisms. In one of these the electronic coupling with the surface waves is very sensitive to the curvature of the electron orbits and their angle of incidence at the surface. Thus $h_{\rm K}$ should alter in a magnetic field H applied parallel to the metal surface. Attempts to observe this in Pb, Hg, and Cu have proved inconclusive.⁵ From all these results, the generally accepted conclusion⁷ is that the electrons, in spite of being the main carriers of heat in the bulk, are unable to absorb energy directly from the helium phonons striking the metal surface. It is for this reason that the problem is always treated on the basis of phonon transmission for both metals and dielectrics. In this paper we show that the above conclusions are not of general validity and that under favorable conditions electrons can indeed play a dominant role in the transfer of heat across the surface.

We have measured $h_{\rm K}$ of oriented single-crystal disks of gallium between 1.2 and 1.8 K by an improved version of an ac technique first successfully used by Challis and Sherlock⁸ for polycrystal-line metal foils. It determines the coupling between two second-sound resonating cavities separated by a thin metal foil whose thickness $l \ll \kappa / h_{\rm K}$, where κ is the bulk thermal conductivity.

p. 305.

Such a foil offers no obstacle to the passage of thermal waves at low frequencies (~ 200 Hz), and the attenuation of second sound is entirely due to the Kapitza resistance on either side. Ga was chosen for the following reasons:

(1) It can be grown into single-crystal disks with mirror-smooth oxide-free surfaces which require no further treatment. Single crystals were necessary to observe the predicted magnetic field effect.

(2) Between 1 and 2 K gallium has one of the highest κ 's for a metal which is practically all electronic. The mean free paths l of its electrons are so large^{9,10} that specimens with t < l over the entire temperature range covered could be prepared easily. This was considered essential in order to keep the scattering of electrons in the bulk to a minimum.

(3) Its electron trajectories are drastically altered by H as shown by a complex magnetoresistance.¹¹

Special precautions (details to be published elsewhere) were observed to avoid strains and surface contamination, which affect absolute values of $h_{\rm K}$. They were successful to the extent that our absolute values of $h_{\rm K}$ were reproducible to within 5% for different specimens. Measurements were made for heat current \dot{Q} along the three principal axes of the orthohombic lattice. The samples were disks 2.54 cm in diameter with thicknesses varying from 0.820 to 0.295 mm. For all specimens $\kappa/h_{\rm K}$ was >1000 mm. $h_{\rm K}$ was also measured as a function of an applied *H*, up to 4 kG. *H* was parallel to the specimen surfaces and perpendicular to the direction of \dot{Q} .

Figure 1 shows $h_{\rm K}$ as a function of H at 1.6 K for \dot{Q} along the three principal axes. The directions of H are given in the figure. Apart from orientation, all the other characteristics of the three specimens were identical. For \dot{Q} along the a and the b axes, $h_{\rm K}$ initially increases with H and starts to decrease after reaching a maximum. For the c axis there is no initial increase. We believe that these observations are the first convincing evidence that the conduction electrons can interact directly with the surface modes generated by the impinging He phonons and thereby transmit heat across a metal-liquid-He interface.

A careful examination of Fig. 1 suggests the following mechanisms. The initial rise in $h_{\rm K}$ is presumably due to an increased coupling between surface phonons and electrons, which results from the change in curvature of their orbits as



FIG. 1. $h_{\rm K}$ versus *H* for *a*-, *b*-, and *c*-axis samples at 1.6 K. The thickness of each sample is 0.295 mm.

predicted by Little.³ This can manifest itself only for small H when the cyclotron radius is greater than t and the energy gained by the electrons from one surface can be transmitted directly to the other on collision. As soon as the orbit fits inside the sample, scattering of the electrons at the surface begins to diminish and a corresponding reduction in $h_{\rm K}$ starts to take place. As H increases, the electrons are trapped in helical paths of decreasing radii, with the result that they can reach the other side only after repeated collisions in the bulk. This increases the thermal isolation between the two surfaces and a further drop in $h_{\rm K}$ occurs. Thus, at a given temperature, the rate of decrease of the flow of electrons across the specimen, which determines the change in $h_{\rm K}$, will be governed by the type of orbit allowed by H. For \dot{Q} parallel to the c axis and H in the ab plane the electrons apparently move in highly convoluted closed orbits,¹² causing a much more rapid fall in $h_{\rm K}$. On the other hand, if the field makes the electrons move along open orbits, they are able to go from one surface to the other with only slight modifications in their trajectories and the flow of energy re-



FIG. 2. Rotation diagrams of $h_{\rm K}$ at 1.4 K for magnetic fields of 1 and 4 kG for $\hat{Q} \| b$. The sample thickness is 0.820 mm and the field is rotated in the *ac* plane. Data points are omitted for clarity.

mains essentially unimpeded. The geometrical configuration of H for curve 1 in Fig. 1, in which the high-field value of $h_{\rm K}$ shows only a slight decrease, is that for an open orbit.¹³ This is perhaps the most compelling evidence that electrons can carry heat from one surface to the other. The conclusion is further confirmed in Fig. 2, which shows $h_{\rm K}$ for $\dot{Q} \parallel b$ in fields of 1 and 4 kG as they are rotated in the *ac* plane. The sudden large increase for $H \parallel a$ is again for an open orbit. Subsidiary maxima at 30 and 53° are due to extended orbits whose trajectories are less sensitive to H.

It can be argued that our experiment only measures the net transfer of heat across the metal and cannot distinguish between changes in $h_{\rm K}$ and changes in κ due to bulk magnetoresistance. The following observations, however, suggest that bulk magnetoresistance cannot cause the observed variations in $h_{\rm K}$.

(i) Transverse magnetoresistance is always positive and cannot be responsible for the initial increase in $h_{\rm K}$.

(ii) Our data, for those orientations of *H* in which bulk magnetoresistance^{11,13,14} varies as H^2 , could always be fitted by an expression of the form $h_K = \alpha + \beta / H$ as shown by the continuous lines in Fig. 3. This suggests that variations in



FIG. 3. $h_{\rm K}$ versus *H* for $\dot{\mathbf{Q}} \| b$ at different temperatures. The field is in the *ac* plane at an angle of 17° from the *a* axis. The sample thickness is 0.820 mm. Solid lines, result of a least-squares fit using the expression $h_{\rm K} = \alpha + \beta / H$ for H > 1.5 kG.

 $h_{\rm K}$ are governed by the orbit radius of the electrons, and not by bulk magnetoresistance.

(iii) From Fig. 3 one can also see that $\Delta h_{\rm K}/h_{\rm Ko}$ at 4 kG is smaller at lower temperatures. If changes in $h_{\rm K}$ were caused by bulk magnetoresistance, the opposite would be true.¹⁴

(iv) Thermal¹⁴ and electrical^{11,13} magnetoresistances for Ga show the largest relative change for H in the ac plane when thermal or electrical current is along the b axis, and the smallest change for H in the ab plane when current is along c. In our data the reverse is true for these two configurations.

(v) The most compelling argument against bulk resistance is as follows: In a solid the decay length of a temperature wave is $\delta = (\kappa/S\pi\nu)^{1/2}$, where S is the (specific heat)/volume, κ the thermal conductivity, and ν the frequency. In a specimen of thickness t this would cause a phase shift of t/δ between the two cavities. For the thicker specimens it should be about 20°, on the assumption that the change in $h_{\rm K}$ at 4 kG is enVOLUME 32, NUMBER 20

tirely due to bulk resistance. No phase change could be observed in any specimen, with the phase-sensitive detector, indicating an absence of measurable resistance. This was further confirmed by driving the cavities in higher harmonics. A finite bulk resistance caused by the field would result in a larger decay for the frequencydependent thermal wave, and the reduction in the transmitted amplitude would be enhanced. This too could not be detected. From several independent estimates we were able to conclude that, even for the thickest samples, the bulk contribution could not exceed 10%. Our final conclusion, therefore, is that the main reason for the variation in energy flow is a change in $h_{\rm K}$ caused by the electrons.

*Work supported by the National Science Foundation under Grants No. GH 33385 and No. GH 40901.

†Work supported in part by a Fellowship from The University of Ohio and in part by the Deutsche Forschungsgemeinschaft.

‡Now at the Department of Metallurgy.

¹I. M. Khalatnikov, Zh. Eksp. Teor. Fiz. <u>22</u>, 687

(1952).

²W. A. Little, Can. J. Phys. <u>37</u>, 334 (1959).

³W. A. Little, Phys. Rev. <u>123</u>, 435 (1961).

⁴A. F. Andreev, Zh. Eksp. Teor. Phys. <u>43</u>, 1535

(1962) [Sov. Phys. JETP 16, 1084 (1963)].

⁵N. S. Snyder, *Thermal Conductance at the Interface* of a Solid and Helium II (Kapitza Conductance), U. S. National Bureau of Standards Technical Note No. 385 (U. S. GPO, Washington, D. C., 1969).

⁶J. D. N. Cheeke, in *Proceedings of the Eleventh International Conference on Low Temperature Physics*, *St. Andrews*, *Scotland*, 1968, edited by J. F. Allen, D. M. Finlayson, and D. M. McCall (St. Andrews Univ.

Press, St. Andrews, Scotland, 1969), Vol. 1, p. 567.

⁷L. J. Challis, J. Phys. C: Proc. Phys. Soc., London <u>7</u>, 481 (1974).

⁸L. J. Challis and R. A. Sherlock, J. Phys. C: Proc. Phys. Soc., London <u>3</u>, 1193 (1970).

 9 R. I. Boughton and M. Yaqub, Phys. Kondens. Mater. 9, 138 (1969).

 $^{10}\mathrm{R.}$ J. von Gutfeld and A. H. Nethercot, Jr., Phys. Rev. Lett. $\underline{18},\ 855$ (1967).

¹¹W. A. Reed and J. A. Marcus, Phys. Rev. <u>126</u>, 1298 (1962).

¹²W. A. Reed, Phys. Rev. <u>188</u>, 1184 (1969).

¹³J. C. Kimball and R. W. Stark, Phys. Rev. B <u>4</u>, 1786 (1971).

¹⁴F. W. Gorter and L. J. Noordermeer, Physica (Utrecht) 46, 507 (1970).

Picosecond Ellipsometry of Transient Electron-Hole Plasmas in Germanium

D. H. Auston

Bell Laboratories, Murray Hill, New Jersey 07974

and

C. V. Shank Bell Laboratories, Holmdel, New Jersey 07733 (Received 28 February 1974)

An ellipsometer has been used with picosecond pulses to measure the time evolution of optically generated plasmas in intrinsic Ge. By measuring the change in optical ellipticity of a weak probe beam following the absorption of an intense excitation pulse, the time evolution of the plasma density can be determined with a precision of a few picoseconds. At a density of 1.7×10^{20} cm⁻³, the ambipolar diffusivity of the plasma was observed to be a factor of 3 greater than the low-density value, consistent with a simple theoretical model of diffusion in the degenerate regime.

In this Letter we report measurements on optically generated electron-hole plasmas in germanium using a new technique which combines ellipsometry and optical picosecond pulses to measure small changes in index of refraction $(\delta n/n < 10^{-3})$ on a picosecond time scale. The physical properties of semiconductor plasmas produced by interband optical absorption have been measured previously by a number of techniques, including infrared¹ and millimeter-wave reflectivity² and free-carrier absorption.³ The ellipsometric technique allows a direct time-resolved measurement of the plasma contribution to the optical refractive index. In addition, both