be conducted with a cw Nd: YalG laser of moderate power and that in this material a stepwise four-photon process involving two doublephoton transitions is operative.

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## TEMPERATURE DEPENDENCE OF HEAT-PULSE PROPAGATION IN SAPPHIRE

R. J. von Gutfeld\* and A. H. Nethercot, Jr. IBM Watson Research Center, Yorktown Heights, New York (Received 31 August 1966)

Results of heat-pulse measurements between 4 and 54'K are reported for sapphire and show that the propagation is ballistic below  $18^{\circ}$ K and diffusive above  $40^{\circ}$ K. In the intermediate range, the propagation is a superposition of the two.

Data have been reported earlier on the propagation of heat pulses in quartz and sapphire at  $3.8^{\circ}$  and  $\sim 8.5^{\circ}$ K.<sup>1</sup> Other reports have since appeared on the propagation of heat pulses in sapphire and other solids. $2-5$  Heat-pulse data, taken as a continuous function of temperature, can give information on phonon-phonon scattering phenomena. A wave-like propagation of heat pulses (second sound) may result from a high rate of normal-process events compared to all other scattering events.<sup>6</sup> The experiment in solid helium<sup>4</sup> has been interpreted as evidence of second sound. We wish to report results on heat-pulse propagation in sapphire. The material is particularly well suited for this investigation since its thermal conductivity is high and its elastic properties are well known. Also, the acoustic velocity is quite isotropic and the singleness of the crystals can be assured. We have observed the superposition of several modes of thermal transport but none corresponding to second sound. It appears that if indeed second sound has been observed in solid helium, the phenomenon is not as general as recent theory predicts. '

We have extended our earlier low-temperature work as a continuous function of temperature up to 54'K. This is well above the temperature  $(\sim 30^\circ K)$  of the thermal conductivity per ature (50 K) or the thermal conductivity<br>maximum,<sup>8</sup> the temperature near which a maximum number of N-process compared with  $U$ -process collisions should occur, and therefore near which second sound is most likely to occur.

The findings of the present experiment are that (l) the observed onset time of the arriving heat pulses for the longitudinal phonons is essentially constant for the temperature range 4-40'K and for the transverse phonons this time is almost constant, increasing by only  $\sim 5\%$  over this temperature range; (2) as the temperature increases the amplitude of the sharp transverse pulse relative to the longitudinal pulse decreases; also by 18'K appreciable phonon-phonon scattering is present which gives rise to an additional diffuse maximum at times substantially later than either the acoustic-energy transport time or that expected for second sound; (3) from 40 to  $54^{\circ}K$ , no observable heat arrives at the acoustic velocity, but

the heat arrives instead by diffusion at a very much later time, in agreement with thermal conductivity results.

As before, a heat pulse was generated on one face of a  $z$ -cut sapphire crystal and detected on the opposite face. Light from a giant-pulse ruby laser (half-power width -40 nsec) was absorbed in an evaporated alloy film  $(In_{0.94}Sn_{0.06})$ . With masks to define the light beam, 0.4- and O.l-in. diameter heat sources were available. The detector was an evaporated thin film of pure indium. It consisted of closely spaced 0 2-mil lines -2000 <sup>A</sup> thick in a zigzag array covering 0.060 in. x0.090 in. This film was used as a bolometer with ~20 mA bias current. The voltage change was proportional to incident heat-pulse power. The small heat capacity of the detector provided a fast thermal response. $9,10$  The sapphire crystals were supplied by the Valpey Crystal Company with 1.00 and 0.50 cm lengths and 0.59 cm diameter.

The shapes of the received pulses at various temperatures are shown in Figs. 1 and 2 for



FIG. 1. Observed heat pulses (upper trace) after propagating through a  $\frac{1}{2}$ -cm sapphire crystal at four different temperatures. The initial pulse is due to laser light falling directly on the detector; the second and third pulses represent heat arriving with the longitudinal and transverse phonon velocities. The lower trace in these figures is the direct response of a phototube to the laser light. The time scale is  $0.5 \mu$  sec/cm (major division).

the  $\frac{1}{2}$ -cm crystal and the 0.4-in. heat source. The first pulse is caused by stray laser light reaching the detector directly. Its decay indicates that the response speed of the over-all system is limited mainly by the electrical circuits to ~25 nsec. The velocities corresponding to the onset times of the transverse and longitudinal pulses are listed in Table I and are quite independent of temperature. These values are in good agreement with the energy velocities calculated from the phase velocities with the aid of calculations by  $\text{Farnell.}^{1,11}$  The observed rise time of  $\sim 0.2$   $\mu$  sec of the modes can be accounted for by the finite size of the heater and detector. At the lowest temperatures, the detector response does not return to zero after the sharp pulses have passed as was reported previously.<sup>1</sup> This is a consequence of the high power input used and has been observed previously, but no detailed explanation can be given.

The observed decrease in the ratio of the unscattered transverse to longitudinal heatpulse amplitude with increasing temperature is consistent with the larger increase of the shear-wave attenuation with temperature than the longitudinal attenuation as observed with 1 the longitudinal attenuation as observed with  $1$ -<br>kMc/sec ultrasonic waves.<sup>12</sup> The absolute peak heights are affected by the change in detector sensitivity with temperature,  $9,10$  but clearly less heat must arrive in the sharp pulse above 10'K since appreciable heat is scattered from the direct beam and arrives at a later time. The unexpected feature is that the sharp pulses are not continuously broadened and do not shift appreciably to later arrival times. Instead, the diffuse maximum due to scattered phonons appears at a considerably later time and grad-



FIG. 2. The heat pulse observed at 38°K, indicating a broad temperature maximum characteristic of heat flow by diffusion.





ually increases in height and in arrival time in the temperature range  $~18-40\text{°K}$ . Little, if any, additional heat flow is found in the region between the sharp maxima and the broad maximum. The structure of the broad maximum at 38'K is shown in Fig. 2.

Above 40'K the unscattered modes were essentially absent, leaving only heat transport by diffusion. A thermal-conductivity mean free path can be calculated (Table II) fromthe heat-pulse data by using the special one-dimensional solution<sup>13</sup> for a  $\delta$ -function heat-flux excitation at  $x = 0$ :

$$
\Delta T \propto \frac{\Delta T_0}{\sqrt{t}} \exp(-x^2/4\kappa t). \tag{1}
$$

Here  $\kappa$  is the diffusivity of the sapphire and  $\Delta T$  the temperature above ambient at a distance x from the excitation. Although Eq. (1) is not expected to hold exactly for a sample of finite length, it should be obeyed reasonably well especially for the initial stages of the diffusion pulse. A comparison of phonon mean free paths from heat-pulse data using Eq. (1) evaluated at  $\Delta T/\Delta T_0$  = 0.25 and those calculated from thermal-conductivity  $(l = 3K/C\overline{v})$  data<sup>8</sup> are in quite good agreement. (Here  $\Delta T_0$  is the maximum temperature reached by the detector. )

It might also be mentioned that for high input powers at the lowest temperatures, an additional heat-pulse arrival time is observed which corresponds to an echo at three times the onset time of the unscattered transverse mode. This pulse was only observable near 4.2 $\mathrm{K}$ . The disappearance of this echo at higher temperatures is related to the increase in the observed attenuation of the transverse mode already discussed.

Thus, the effects observed in the study of heat-pulse propagation in sapphire at both the higher and the lower temperatures are as expected from the theory of diffusive and ballistic heat flow, respectively. At the intermedi-

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ate temperatures between 18 and 40'K, the persistence of the sharp unshifted pulses while the heat flow is becoming mainly diffusive is rather unexpected, since one might have expected a gradual broadening and shift of the sharp pulse into the more diffusive type of behavior. Instead, the sharp pulse persists even at temperatures where the bulk of the heat arrives considerably later due to diffusion. This may be related to a rather abrupt and unexpected difference in behavior of the lower frequency and higher frequency phonons comprising the heat pulse.

A possible explanation of these observations might be the following: The higher frequency phonons may suffer frequent large-angle scattering processes (probably predominantly  $U$ processes and perhaps some point-defect scattering) and thus contribute to the broad maximum. The low-frequency phonons may either be unscattered or suffer only small-angle Nprocess collisions. These would directly interfere only slightly with the thermal current and heat-pulse shape. The intermediate-frequency phonons must then be either too few in number or their rate of large-angle N-process collisions must be too low to produce a significant contribution to the heat flow at the velocity corresponding to that expected for "second sound." It has recently been suggested that the  $N$ -process events may be predom-

Table II. The phonon mean free path determined from the present heat pulse data and thermal conductivity data.<sup>8</sup>

Heat-pulse	Thermal conductivity
data	data $(l = 3K/C\bar{v})$
(c <sub>m</sub> )	(c <sub>m</sub> )
$1.5 \times 10^{-2}$	$2.4 \times 10^{-2}$
$7.8 \times 10^{-3}$	$8.8 \times 10^{-3}$
$2.2 \times 10^{-3}$	$2.1 \times 10^{-3}$

inantly small-angle collisions.<sup>14-16</sup> For such normal-process collisions, second sound cannot be expected to occur, and, in fact, no indication of second sound or even of an approach to second sound is seen in the data.

The data for both the 0.4-in. diameter and the O.l-in. diameter heat source show no essential difference. The larger heat source would be the more likely one for which second sound might be observed. The ratios of input pulse width to transit times in the  $\frac{1}{2}$ -cm crystal were  $\sim 0.1$  and 0.05 for the longitudinal and transverse pulses, respectively. Thus most of the external conditions for this experiment were rather similar to those employed in the recent experiment on solid helium for which the observation of second sound has been reported.<sup>4</sup> Even though there is not necessarily a direct conflict between these two results (since the role of phonon collisions may be quite different in the two materials), it would be of considerable interest to extend the solidhelium results to lower temperatures to investigate the transition from the reported behavior to the expected ballistic flow at the ordinary sound velocity.

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## de HAAS-van ALPHEN EFFECT IN FERROMAGNETIC NICKEL\*

D. C. Tsuit and R. W. Stark<sup>‡</sup>

Department of Physics and Institute for the Study of Metals, The University of Chicago, Chicago, Illinois (Received 8 September 1966)

We report here an investigation of the de Haas-van Alphen effect in nickel. The results are discussed in terms of the models which have been recently proposed for the band structure of ferromagnetic nickel.

We wish to report in this Letter the preliminary results of our de Haas —van Alphen (dHvA) investigation of the Fermi surface of nickel. Several models have recently been proposed for the ferromagnetic nickel band structure. $1-5$ In general, these models are quite similar in their gross features but differ in detail. This similarity is expected since rather stringent limitations are placed on any proposed model. The exchange interactions are small compared with the crystal potential so that one assumes that the ferromagnetic band structure can be obtained from the paramagnetic band structure<sup>6,7</sup> by considering the exchange splitting as a perturbation. In addition, the resulting model

must meet the dual criteria of having an excess of about 0.55 spin-up  $(*)$  electrons<sup>8</sup> and a Fermi surface enclosing a net volume corresponding to one electron per atom.<sup>9</sup> It must also have one open sheet, similar to the copper Fermi surface, to satisfy the magnetoresistance experiments of Fawcett and Reed<sup>10</sup> and the dHv*l*<br>experiments of Joseph and Thorsen (JT).<sup>11</sup> experiments of Joseph and Thorsen (JT). The dHvA data which we present here offer the first independent confirmation of this general approach to the ferromagnetic nickel band structure.

The single-crystal nickel samples which were used in this experiment were prepared by electron-beam zone refining in ultrahigh vacuum.



FIG. 1. Observed heat pulses (upper trace) after propagating through a  $\frac{1}{2}$ -cm sapphire crystal at four different temperatures. The initial pulse is due to laser light falling directly on the detector; the second and third pulses represent heat arriving with the longitudinal and transverse phonon velocities. The lower trace in these figures is the direct response of a phototube to the laser light. The time scale is  $0.5 \mu \sec/cm$ (major division).



FIG. 2. The heat pulse observed at  $38^{\circ}\mathrm{K},$  indicating a broad temperature maximum characteristic of heat flow by diffusion.