Propagation of sound across an interface between liquid helium and polycrystalline gold

K. N. Zinov'eva and I. N. Shvetsov

P. L. Kapitsa Institute of Physical Problems, Russian Academy of Sciences, 117973 Moscow, Russia

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The angular distribution of the energy transmission coefficient has been studied for a monochromatic sound wave passing from liquid ^{4}He into polycrystalline gold with a grain size of 5–10 μm. The measurements were carried out at frequencies of 13, 39, and 65 MHz at temperatures of 150–250 mK. The absorption spectra are governed by the high attenuation of sound in the polycrystalline gold, which results from the scattering of the sound by grain boundaries. At 39 and 65 MHz, the absorption is one or two orders of magnitude stronger than that by conduction electrons. This huge attenuation can be described qualitative by a model of Rayleigh scattering of sound by irregularities of the elastic medium.

1. INTRODUCTION

This paper reports a study of the energy transmission coefficient as sound enters polycrystalline gold from liquid ^{4}He. The flux of heat (phonons) incident from ^{4}He on the surface of a solid experiences a Kapitsa resistance at the interface.[1] A temperature difference arises between the liquid helium and the solid. This effect has been explained theoretically by Khalatnikov[2] on the basis of an acoustic mismatch of the media.

The phonon flux crossing an interface is determined by the probability for the transmission of a phonon or by the transmission coefficient for acoustic energy, $\alpha(\theta,\omega)$, where θ is the angle of incidence, and ω is the phonon frequency. In order of magnitude, the coefficient $\alpha(\theta,\omega)$ is equal to the acoustic impedance ratio of the media: $\alpha\sim\rho c/Dc_l$, (where ρ and D are densities, and c and c_l are the sound velocity in the ^{4}He and the longitudinal sound velocity in the solid). For an interface between liquid ^{4}He and a solid we have $\alpha\sim10^{-3}$–10^{-2} within a critical cone $\theta_c=c/c_t=5$–$10°$, where c_t is the transverse sound velocity in the solid. Outside the critical cone we have $\alpha=0$ because of total internal reflection. The heat flux affects the energies of all vibrational modes of the solid, including the energy of a surface Rayleigh wave.

It has now been solidly established that the theory of Ref. 2 sets an upper limit on the Kapitsa resistance and holds better, the higher the quality of the crystal and its surface. For example, experimental values of the Kapitsa resistance for single crystals with carefully prepared surfaces, at temperatures $T<0.3$–4 K, are very close to the theoretical Khalatnikov limit.

Numerous experiments have shown that the significant deviations of the Kapitsa resistance from the theoretical values which have been observed are due to enhancement of heat transfer as the result of phonon dissipation in the solid. Causes of this dissipation might be conduction electrons in metals, lattice defects, surface roughness, the edges of the sample, etc.

Andreev[3] was the first to point out that absorption of phonons in a solid has a strong effect on the magnitude and angular distribution of the transmission coefficient $\alpha(\theta)$. He showed that the scattering of sound (acoustic phonons) by conduction electrons in a metal leads to resonant absorption of a surface Rayleigh wave in a supercritical Rayleigh angle $\theta_R=c/c_R$, where c_R is the velocity of the Rayleigh wave. A sharp peak of height $\alpha\approx1$ appears in the transmission coefficient at θ_R; the width of this peak is a few angular minutes. The reflection coefficient correspondingly vanishes at θ_R. The contribution of Rayleigh waves to the heat flux is roughly equal to the contribution of bulk waves, in agreement with the conclusion reached by Khalatnikov.[2]

The resonant absorption of sound by the surface of a metal was first observed in a tungsten single crystal in Ref. 4. The acoustic theory has been generalized to the case of the attenuation of phonons in a solid in theoretical treatments.[5–7]

The first measurements of the coefficient $\alpha(\theta)$ for thermal phonons in a NaF single crystal[8] at $T\sim1$ K showed that the absorption and emission spectra were identical and consisted of two regions: a central peak with a width equal to the width of the critical cone and some wide wings, which decay according to Lambert's law with increasing distance between θ and the normal and which make the major contribution to the Kapitsa conductivity. It was suggested[9] that the wide wings are due to diffuse inelastic scattering of a certain fraction of the phonons by surface defects.

The coefficient $\alpha(\theta,\omega)$ was studied in more detail in Refs. 4 and 10–13, where the angular distributions and frequency spectra of the absorption of sound by metal single crystals of cubic symmetry (tungsten, copper, and aluminum) were studied. The measurements were carried out at the frequencies 10–300 MHz, at temperatures from 60 mK to 0.4 K. The general behavior found in those studies can be summarized as follows.

The $\alpha(\theta)$ absorption spectra have a complex angular distribution, which reflects the distribution of the energy transmitted into the crystal over the solid angle. In an

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acoustically isotropic single crystal, only bulk longitudinal and transverse waves are excited inside the critical cone θ_c. The transmission coefficients of these waves are small (10^{-3}–10^{-2}) and essentially constant, depending slightly on the attenuation. The longitudinal and transverse waves have transmission zeros at the critical angles c/c_l and c/c_t. Outside the critical cone, a sharp peak is observed in the resonant scattering of a Rayleigh wave, with $\alpha \sim 1$ at the Rayleigh angle of incidence. The width and height of this peak depend strongly on the attenuation in the crystal.

In an acoustically anisotropic single crystal, two resonant modes are excited by virtue of the anisotropy at the (001) and (111) surfaces: a Rayleigh surface wave and a pseudosurface wave. The transmission coefficient for each of these modes is $\alpha \sim 1$. Only the Rayleigh wave is excited at the (110) surface. The critical cone is more complicated. The primary result of the studies reported in Refs. 4 and 10–13 was the first attainment of good agreement between experimental curves of $\alpha(\theta)$ and theoretical values. The calculations used a phenomenological attenuation parameter $p \sim 10^{-4}$–10^{-3} ($p = \gamma c/2\omega$, where γ is the attenuation in e-folds per unit length). Measurements on an Al single crystal in the normal and superconducting states[12] showed that the attenuation of the Rayleigh wave is indeed due to conduction electrons. In the superconducting state, the Rayleigh peaks become vanishingly small ($p < 10^{-4}$) and make essentially no contribution to the heat flux.

After studies of single crystals with high-quality surfaces, which confirmed the validity of the dissipative acoustic theory, the following questions naturally arose: How does the transmission coefficient for acoustic energy, $\alpha(\theta,\omega)$, vary in the case of lower-quality ("poor") crystals, e.g., polycrystalline samples, whose surfaces have not been subjected to preliminary processing? What is the mechanism for the attenuation of sound in this case? Are the peaks in the absorption for a Rayleigh wave still present? What is their contribution to the heat flux?

There has been no study of the acoustic properties of the interface between ^{4}He and a polycrystalline sample, even though the bulk of Kapitsa's resistance measurements were devoted to specifically polycrystalline samples.

2. EXPERIMENTAL PROCEDURE AND TEST SAMPLE

The method for determining $\alpha(\theta)$, which is described in detail in Ref. 4, consists of measuring the heating of a test sample by the acoustic energy which passes into it from liquid ^{4}He.

The transmission coefficient $\alpha(\theta)$ and the heating of the sample, ΔT, are related by

$$\alpha(\theta) = \frac{\Delta T}{N R_k} \frac{\sigma}{\sigma_0}, \qquad (1)$$

where N is the energy flux density of a monochromatic acoustic wave, R_K is the Kapitsa resistance, σ is the total surface area of the sample, and σ_0 is the area on which the sound is incident. By measuring the heating ΔT as a function of the angle of incidence θ of the sound and the parameters in Eq. (1), we can determine $\alpha(\theta)$.

Measurements were carried out in ^{4}He at saturation vapor pressure and at temperatures of 150–250 mK. The measurement chamber was cooled in a ^{3}He–^{4}He dissolution cryostat. The acoustic cell in the chamber consisted of two parallel disk-shaped mounts connected by a vertical support. An x-cut quartz radiator, 15 mm in diameter, was mounted on the lower disk; the test sample was mounted on the upper one. The quartz and the sample were separated by a distance of 1 cm.

Measurements were carried out at the fundamental frequency of 13 MHz and at the two harmonics 39 and 65 MHz. A rotatable rack holding the sample was mounted on supports extending from the upper disk, on four plane springs. The rotation angle θ was measured with the help of a variable capacitor, connected in the LC resonant circuit of a frequency oscillator. The movable plates of the capacitor were mounted on the rack, while the fixed plates were mounted on the upper disk of the acoustic cell. As the angle was varied from $-\theta$ to $+\theta$, the capacitance of the capacitor varied monotonically in one direction. This circumstance made it possible to avoid an uncertainty in the angle near $\theta = 0°$. The frequency ν of the LC oscillator was varied over the range 935–995 kHz. These frequencies provided good relative accuracy in the angle measurements.

The temperatures of the test sample and the liquid were measured by semiconductor thermometers. One contact of carbon and germanium thermometers was soldered to thin strips of the sample ($d \sim 0.2$ mm) separated from the edges by an electric-arc method. The heater was cemented to the back of the sample with BF cement. Similar thermometers and a similar heater were immersed in the liquid ^{4}He.

The resistances of the thermometers were detected by sensitive Cryobridge S-72 and R3310 ac bridges. The output signals from these bridges were sent to digital voltmeters and were recorded on either a two-pen x, y chart recorder or a computer. The measurements were carried out in a shielded room. The power dissipated in the thermometers by the measurement current was less than 1 nW.

For accurate measurements of the slight heating of the sample, the temperature of the liquid helium in the measurement chamber was regulated within 10^{-6} K. The time required for one recording was of order 1.5–2 h.

The test sample, of high-purity polycrystalline gold ($R_{300}/R_{4.2} = 36\,000$), was prepared from the melt. It was a parallelepiped with dimensions of $13 \times 10 \times 1.9$ mm. In the preparation process, the test sample was sent through rollers, flattened under a press between polished quartz plates, and annealed in vacuum. The surface quality was monitored with the help of a Linnik interferometer. The roughness and deviations from a planar surface were less than 0.5 μm. Since the gold was not oxidized, the surface of the sample was not mechanically polished.

From the nature of the rings on Laue diffraction patterns (Fig. 1) recorded at two different points on a sample (the lines are continuous and equal in thickness), we can estimate the average size of the grains to be $\bar{a} \lesssim 10\ \mu$m.

In addition, after the experiments, the surface was subjected to fine mechanical polishing and etching and then

FIG. 1. Laue diffraction pattern of polycrystalline gold with a grain size < 10 μm.

studied under a microscope at high magnification. The typical grain size as a result of the recrystallization during annealing turned out to be 5–10 μm.

3. EXPERIMENTAL RESULTS AND DISCUSSION

Experimental curves of $\Delta T(\theta)$ of two types are shown below. The recordings shown in Fig. 2 were made on the two-pen x, y recorder. They contain the simultaneous readings of the sample temperature and the liquid temperature in the field of a plane acoustic wave as θ is varied. Figure 3 shows corresponding recordings made by the computer, in this case of only the sample temperature. Again in this case, the temperature of the liquid was held constant, at the same precision. The difference between the temperatures of the liquid ^{4}He at the beginning and end of the recording can be inferred from the vertical shift of the outer regions of the $\Delta T(\theta)$ curve. Over the recording time, the temperature of the liquid ^{4}He remained constant within ± 5 μK. We verified that in the absence of the sound the thermometers on the sample and in the liquid showed the same temperature as the sample was rotated.

Let us examine the results in more detail. The recordings for each of the three frequencies are quite reproducible. On all the curves we can clearly see a critical angle $\theta_c = 12°$, within which the sample is heated to a maximum extent by the incident sound. The angle θ_c was found from the ratio of the sound velocities in helium to the average

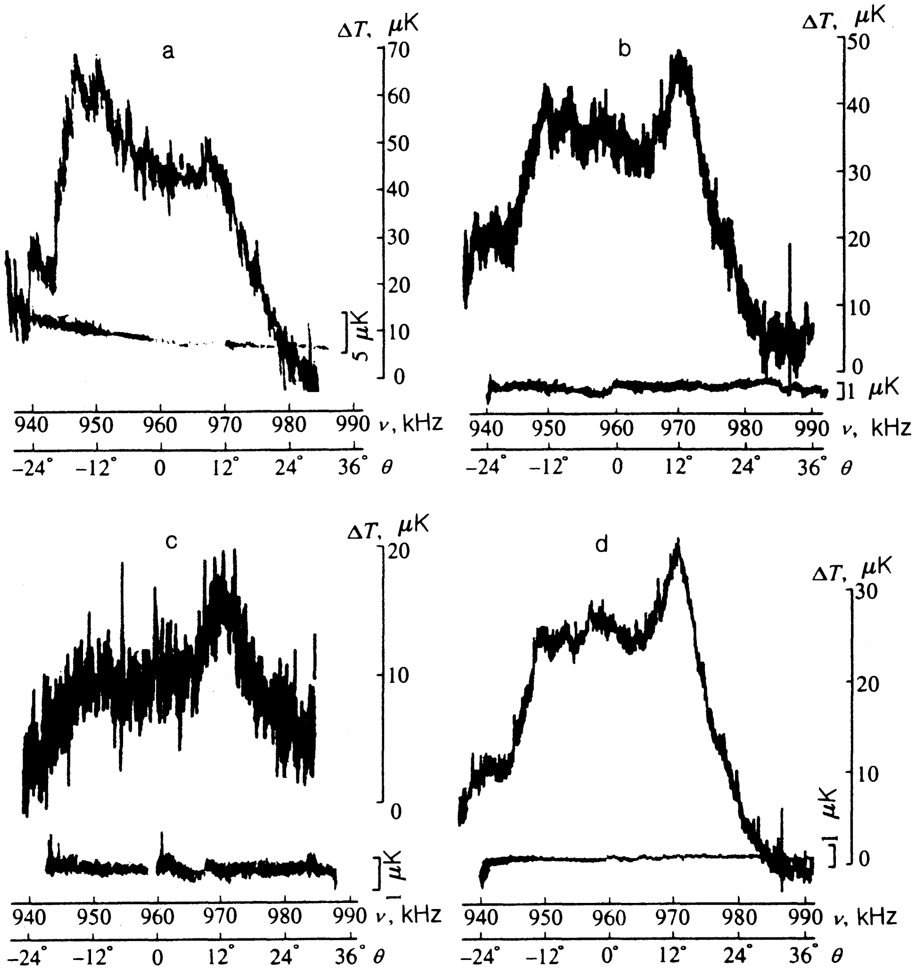

FIG. 2. Experimental spectrum of the absorption of phonons by polycrystalline gold. a—$f = 13$ MHz, $T = 150$ mK; b—39, 150; c—65, 150; d—39 MHz, 200 mK. The curves at the bottom correspond to the temperature of the liquid ^{4}He.

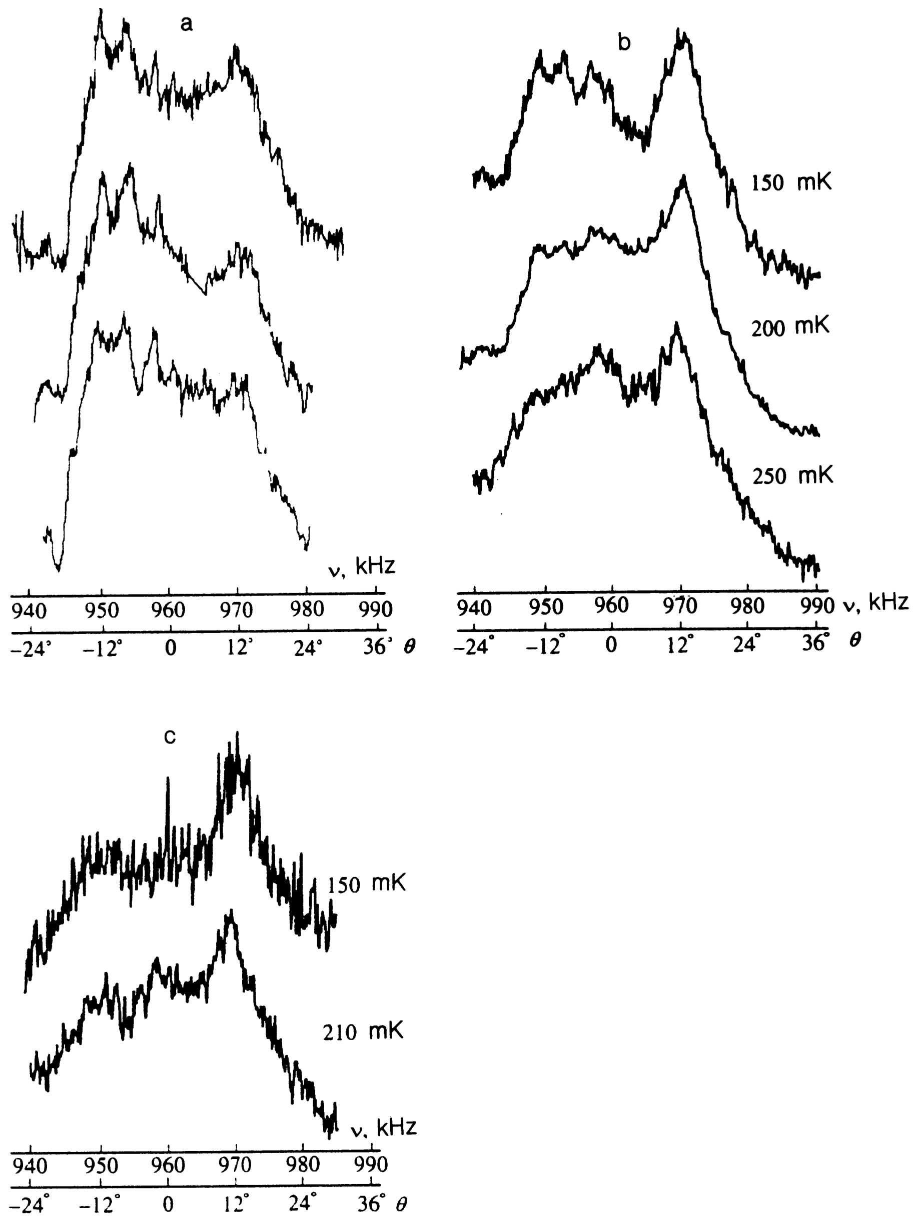

FIG. 3. Experimental spectrum of the absorption of phonons by polycrystalline gold. a—$f=13$ MHz, $T=150$ mK (three different recordings); b—$f=39$ MHz, $T=150$, 200, and 250 mK; c—$f=65$ MHz, $T=150$ and 210 mK.

velocity of a transverse wave in polycrystalline gold, $c_t=1.2\cdot 10^5$ cm/s. The heating (ΔT) of the polycrystalline Au by the gold with respect to the ^{4}He was much less than the heating of W, Cu, and Al single crystals.[4,10–13] This result indicates that the Kapitsa resistance is small in a polycrystalline sample. Rough estimates based on our experiments yield $R_K T^3 \sim 80$ cm$^2\cdot$K^4/W. This figure is close to the value $R_K T^3=46$ cm$^2\cdot$K^4/W found by Folinsbee and Anderson[14] for a mechanically polished and annealed gold foil 0.6 mm thick at $T\sim 40$–300 mK.

The $\alpha(\theta)$ distribution of polycrystalline Au differs greatly in shape from the characteristic angular distributions of single crystals. In this case we do not find the high, sharp peaks of resonant absorption of a Rayleigh wave at the edges of the spectrum; the deep minima separating regions of longitudinal, transverse, and surface waves, or the resolved secondary Rayleigh maxima near the angle of normal incidence. The spectrum is not symmetric. All the structural features are greatly smeared out, and the resonant peaks are wide and short.

Outside the critical angle, the function $\alpha(\theta)$ falls off to zero very slowly. The particular design of the apparatus ruled out a further increase in the angle of incidence and a determination of the heating of the sample at $\theta > 30°$–$40°$.

All these structural features in the $\alpha(\theta)$ distribution point to a high attenuation of sound in the polycrystalline gold sample—an attenuation far higher than that in single crystals. If we use the ultrasound absorption properties for the three principal directions of a gold single crystal as found in the measurements of Ref. 15, and if we average these properties over all directions, then we find $p_l=4.5\cdot 10^{-4}$ and $p_t=3.5\cdot 10^{-4}$ for an electron mechanism for the absorption of phonons in an isotropic polycrystalline gold sample.

On the other hand, p can be determined from experiment if we compare the transmitted-energy integrals

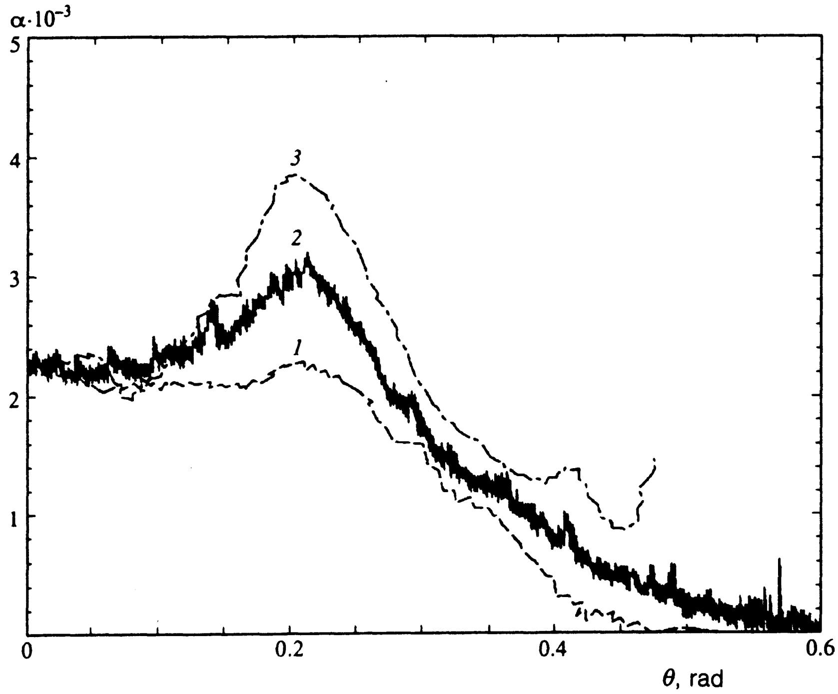

FIG. 4. Normalized experimental curves of $\alpha(\theta)$. 1—$f=13$ MHz, $T=150$ mK; 2—39, 200; 3—65 MHz, 150 mK.

$$J=\int_0^{0.5}\alpha(\theta)d\theta$$

in the cases of the experimental curve (J_{expt}) and the theoretical curve (J_{theor}) of $\alpha(\theta)$ (here we are assuming that the energy integral is conserved). We choose a p value such that $J_{\text{theor}}=J_{\text{expt}}$ holds. The shape of the experimental curve may differ from that of the theoretical curve, primarily because of a broadening of the interval of angles of incidence on a rough surface.

Figure 4 shows experimental curves of $\alpha(\theta)$ for three frequencies: 13, 39, and 65 MHz. The curves are normalized to the same value at $\theta=0$. The values of J_{expt} were calculated over the θ interval from 0 to 0.5 rad. The corresponding values of p were determined from the theoretical $J_{\text{theor}}(p)$ dependence for polycrystalline gold (Fig. 5).

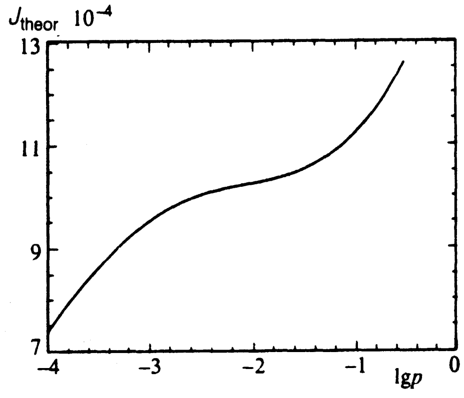

FIG. 5. Theoretical behavior of the acoustic energy transmitted through a polycrystalline gold sample, J_{theor}, as a function of the attenuation parameter p.

It was found that, as the frequency is varied from 13 to 65 MHz, the value of p changes from 10^{-4} to 0.15. In other words, it is highly dependent on the frequency. At high frequencies (39 and 65 MHz), it is one or two orders of magnitude higher than the electron absorption parameter $p_e\sim10^{-4}$ (Table I).

Further evidence for this conclusion comes from the growth of the tails on $\alpha(\theta)$ outside the critical angle as the frequency is raised.

This strong attenuation of the sound in polycrystalline Au and its increase with the frequency stem from scattering of the sound by grain boundaries. As has been shown theoretically by Lifshits and Parkhomovskiĭ,[16] the scattering of ultrasound by inhomogeneities in an elastic medium (by the grains of a polycrystalline sample) becomes the predominant attenuation mechanism at sufficiently high frequencies. It greatly outweighs all other mechanisms. Exact expressions for the attenuation of longitudinal and transverse sound in a polycrystalline material were given in Ref. 16. The absorption of sound depends on the average grain size $\bar{a}$: At low frequencies ($\lambda\gg\bar{a}$) we have $\gamma\propto\omega^4$ (this is Rayleigh scattering), while at high frequencies ($\lambda\ll\bar{a}$) we have $\gamma\propto\omega^2$.

The treatment by Lifshits and Parkhomovskiĭ[16] is limited in several ways (the anisotropy of the grains is slight, the grains are equiaxial, and there is no predominant ori-

TABLE I.

	13 MHz	39 MHz	65 MHz
p	$2\cdot10^{-4}$	$3.5\cdot10^{-3}$	$1.5\cdot10^{-1}$
γ, dB/cm	1.2	62	4430
$\gamma\lambda$, dB	10^{-2}	$2\cdot10^{-1}$	8

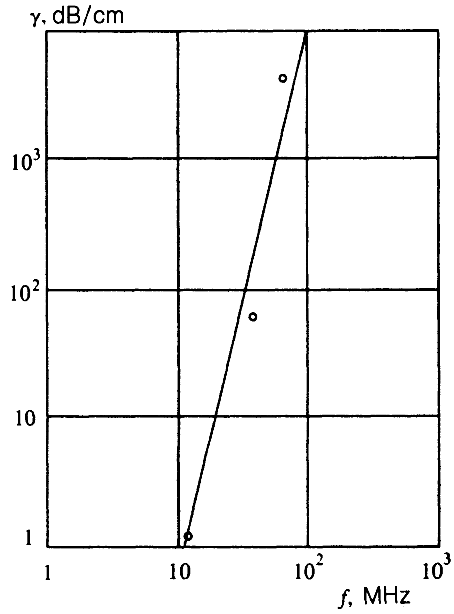

FIG. 6. Experimental points of the attenuation of a Rayleigh wave in polycrystalline gold at frequencies of 13, 39, and 65 MHz, along with a theoretical line[17] $\gamma=\bar{V}f^4S$ for a grain size of 5 μm.

entation). Consequently, the expressions of that paper do not apply to gold. To calculate the attenuation of sound in polycrystalline gold we can use the empirical expressions and tables of scattering coefficients given by Papadakis,[17] which have found good experimental support: $\gamma=\bar{V}f^4S$ at low frequencies and $\gamma=\bar{a}f^2\Sigma$ at high frequencies, where $\bar{V}$ and $\bar{a}$ are the average volume and average diameter of a grain, and S and Σ are constant coefficients which characterize the material and which vary slightly from sample to sample.

The boundary between two regions is determined by the condition $\lambda_b=2\pi\bar{a}$. In our case we have $\bar{a}=5$–10 μm and a boundary frequency $f_b=20$–40 MHz. The frequencies 13, 39, and 65 MHz ($\lambda=90$, 30, and 18 μm) thus correspond primarily to a region intermediate between $\gamma\propto\omega^4$ and $\gamma\propto\omega^2$.

Figure 6 shows the theoretical Papadakis line $\gamma=\bar{V}f^4S$ for a particle size $\bar{a}=5$ μm. The values of S are from Ref. 17. Also shown in this figure are points found from J_{expt} and J_{theor}. The error in the determination of the integrals is at most 5–10%, but the errors in p and γ are on the order of these values themselves ($\sim$100%). The maximum error corresponds to the region of p values between $\sim10^{-3}$ and $\sim10^{-1}$, in which the integral changes by only 10%. It can be seen from this figure that there is a satisfactory agreement between the experimental values of γ and the theoretical line $\gamma\propto\omega^4$. We also note that our data on γ are close to the values of the bulk attenuation in polycrystalline samples as reported for several materials by Papadakis[17] and for stainless steel by Becker and Richardson.[18]

The very strong attenuation of ultrasound at comparatively high frequencies (39, 65 MHz) in polycrystalline materials (10^2–10^3 dB/cm) cannot be measured by direct methods. It can only be estimated, by comparing the experimental attenuation of a Rayleigh wave with the theoretical behavior, as we did above.

The essentially total absorption of the sound at the wavelength corresponding to 65 MHz explains the substantial increase in the overall contribution of phonons to the heat flux across the interface of the polycrystalline sample. It increases because of the tail which stretches out from the critical angle to $\pi/2$, in accordance with the dissipative acoustic theory. The small value of the Kapitsa resistance for polycrystalline gold is completely understandable from this point of view.

At this point we would like to discuss certain features of the experimental curves. First, the $\alpha(\theta)$ curve does not have complete angular symmetry. The probable reason for this result is an anisotropy of the polycrystalline sample (an anisotropy which arises from the rolling, because of the predominant orientation). When the sample and the quartz are not parallel, this circumstance leads to a small difference between the azimuthal angles during scanning to the right and left of $\theta=0$.

The transmitted-energy integrals for positive and negative angles θ are typically equal to within the experimental errors. The isolated peaks on the $\alpha(\theta)$ curves probably correspond to absorption of the Rayleigh wave by groups of grains with principal axes in approximately the same orientation. The positions of the peaks are determined by the propagation velocity of a transverse wave along the principal directions.

A second feature of these curves is that the Rayleigh peak in the absorption [which lies above the level of the continuous spectrum, $\alpha(0)=2.13\cdot10^{-3}$] in the Au sample increases with the frequency from $\sim$10% at 13 MHz to $\sim$50% at 65 MHz (Fig. 4). This effect has been seen previously.[4,17,18] It has been linked with a dependence of the amplitude of the Rayleigh peak on the absorption in the solid: Maximum transmission ($\alpha\sim1$) corresponds to a certain attenuation in each material, in which p depends on the frequency. For tungsten,[4] for example, the transmission peak corresponds to a frequency of 10 MHz; for the interface between stainless steel and water[18] it corresponds to 15 MHz; and for the interface between nickel with a particle size of 25 μm and water it corresponds to more than[18] 400 MHz. For the interface between polycrystalline gold and ^{4}He, the maximum transmission should apparently be observed at $f>65$ MHz. However, we should point out that polycrystalline gold and lead have scattering coefficients S and Σ higher than any other elements, so it would be a problem to observe the $\alpha(\theta)$ dependence at frequencies $f>100$ MHz.

A third feature of the curves is that the Rayleigh peaks are low and have a large angular width, even at the low frequency (13 MHz), where the attenuation is low ($p=10^{-4}$, $\gamma=1$ dB/cm)—comparable to the attenuation by conduction electrons. For attenuation of this sort in single crystals, one observes[4,10–13] high, sharp Rayleigh absorption peaks (the quartz radiator was not changed).

The reason for the broadening of the peak is that the surface of a polycrystalline material consists of distinct grains which are oriented at small angles with respect to the plane. As a result, a narrow incident beam with a divergence of a few arc minutes spreads out to several degrees.

CONCLUSION

In summary, it has been shown experimentally that the spectra of the absorption of sound by the surface of a polycrystalline gold sample differ sharply from those for a single crystal.

The primary difference is that, instead of the high, sharp peaks in the resonant absorption of a Rayleigh wave at a supercritical angle, we find low, wide peaks. There are no minima separating the longitudinal, transverse, and surface excitations. Secondary Rayleigh peaks are not resolved. Outside the critical angle the function $\alpha(\theta)$ falls off to zero very slowly.

Analysis of the data shows that the spectrum corresponds to a high attenuation of sound in the polycrystalline material, which is due to a scattering of the sound by irregularities of the elastic medium: the grain boundaries. The attenuation is proportional to ω^4, i.e., corresponds to the case of Rayleigh scattering with a wavelength much larger than the average grain size.

The absorption properties reach their highest values ($p \sim 1$) at frequencies ~ 100 MHz. Here the transmitted-energy integral increases significantly because of contributions from the $\alpha(\theta)$ tails, which stretch from the critical angle to $\pi/2$. This contribution explains the small Kapitsa resistance of the polycrystalline samples.

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