

INVESTIGATION OF SOLID He⁴ BY STIMULATED BRILLOUIN SCATTERING

S. HUNKLINGER, P. LEIDERER AND P. BERBERICH

Physik Department E 10 der Technischen Universität, München Germany.

INTRODUCTION

Solid helium is generally referred to as a quantum crystal because its zero point energy is comparable to the binding energy. Consequently one expects its behaviour to be quite different from ordinary classical solids. The amplitude of the zero point motion, which is nearly 30 per cent of the interatomic distance, should cause anharmonicities in the lattice potential to play an important role¹. The strength of anharmonicity is expressed by the Grüneisen parameter, which can be determined experimentally from the pressure dependence of specific heat or velocity of sound. The values obtained for solid helium, however, are not too different from those of ordinary substances^{2,3}.

It is well known that direct evidence about the anharmonic behaviour of crystals can be obtained by ultrasonic measurements. In particular, the acoustic attenuation in ideal crystals is determined by phonon-phonon interactions. Until now such measurements have been carried out only in liquid helium⁴. For solid helium there exist rather rough estimations at 10 MHz which only give an upper limit for the sound absorption^{5,6}. This lack of experimental data is not surprising, since the application of the conventional ultrasonic methods to solid helium gives rise to serious technical difficulties: In particular, ultrasonic measurements are hampered by the large acoustical mismatch between the transducer and the sample. In addition they require a high degree of crystal perfection over a large volume. Finally, because of the large acoustic anisotropy of solid helium the energy of a sound wave generally does not travel in the direction of the wave vector, i.e. perpendicular to the transducer, which further complicates the measurements⁷.

All these difficulties can be avoided or become unimportant if one uses stimulated Brillouin scattering (SBS)⁸ for the determination of the sound absorption. The crystal has to be of a high quality only in a small volume, where the laser light is focused. Crystal irregularities influence the propagation of light only slightly as the index of refraction is close to unity and nearly isotropic⁹. Because of the short distance through which the phonons travel during measurement ($< 50 \mu\text{m}$) «walk-off» effects are negligible.

EXPERIMENTAL

The principle of our method is similar to the «Two Pulse Method» used by Heinicke and Winterling¹⁰. Our set-up is shown schematically in fig. 1. A giant pulse from the

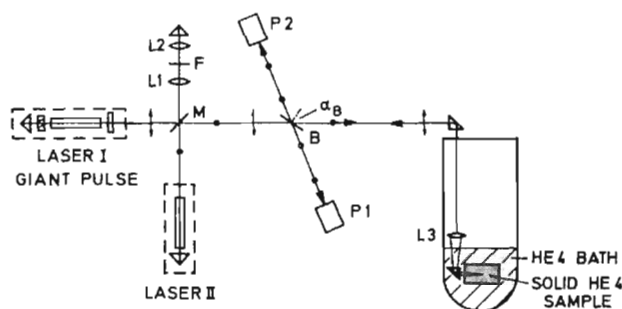


FIG. 1. — Experimental set-up for measuring phonon lifetimes in solid helium. Polarization of laser I (indicated by arrows) is in the plane of the paper; polarization of laser II is perpendicular to it (indicated by circles). Inside the resonator of laser II a semitransparent mirror *M*, a metal film *F* and two lenses *L*₁, *L*₂ are placed. The laser light is focused into the sample chamber with lens *L*₃ ($f = 20 \text{ cm}$). Photo cells *P*₁ and *P*₂ record incident and back-scattered light, respectively. The beam splitter *B* is mounted at the Brewster angle α_B .

ruby laser I is focused into the solid helium sample where it stimulates Brillouin scattering and creates a large number of coherent phonons in the focal volume, which is very small compared to the volume of the sample chamber.

Light from a second ruby laser (II), which is too weak to cause SBS, is subsequently directed into the light path of the first laser and enters the sample at a well defined time. This delayed «test light» is backscattered by those SBS phonons which are still present when the second pulse arrives. The intensity of the backscattered light is proportional to the number of phonons present in the focal volume at that time. Thus from the time dependent ratio of backscattered light to the incident light the phonon life time can be obtained.

The synchronisation of the two lasers is achieved as follows: Within the resonator of laser II two lenses are mounted with a common focal point. A thin metal film in the focus blocks laser II. By means of the semitransparent mirror *M* a certain amount of the giant pulse intensity of laser I is guided into this resonator. This light evaporates the metal film and triggers laser II. Thus a second spiked light pulse develops which is long compared with a giant pulse. Fig. 2*a* shows a typical oscilloscope trace of the incident light registered by photo cell *P*₁. The giant pulse is strongly suppressed because its plane of polarization is perpendicular to the beam splitter *B* (see fig. 1). Fig. 2*b* represents the backscattered light intensity. Stimulated backscattering sets in at time t_b when the incident light reaches the threshold intensity for SBS.

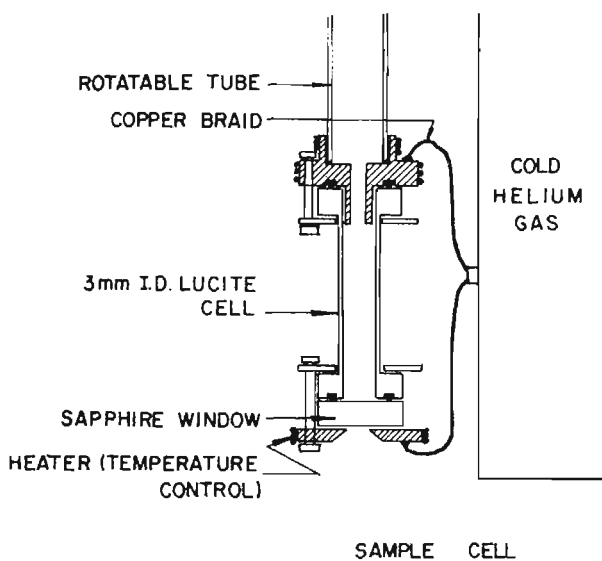


FIG. 4. — Diagram of sample cell for growing neon single crystals.

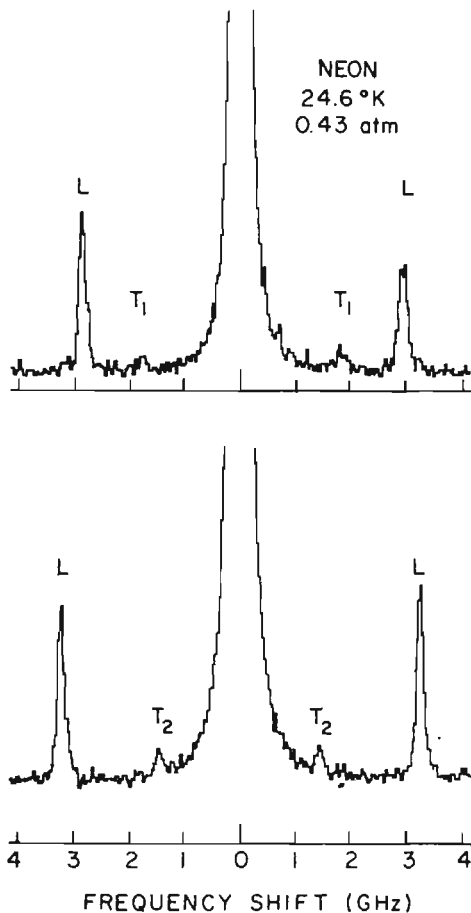


FIG. 5. — Brillouin spectra of two neon single crystals of different orientations.

Fig. 4. The cell is made of carefully machined and annealed Lucite. Lucite is virtually transparent to X-rays, has good optical properties and a very low thermal conductivity. This latter characteristic is necessary in order to eliminate stray nucleation on the walls of the cell. A polished sapphire window is sealed to the bottom of the cell with an indium O-ring. This window allows the laser beam to enter the cell with a minimum of stray scatter. In addition the high thermal conductivity of sapphire makes the inside surface of the window a suitable nucleation site. The cell is cooled by flexible copper braids which are connected to a cold-finger cooled by a flow of helium gas. Heaters control the temperature at the top and bottom of the cell to better than 50 mK.

Prior to crystal growth, the cell is filled with liquid neon and a temperature difference of $\sim 1/2^\circ\text{K}$ is established across the cell. The temperature is then slowly lowered maintaining the same gradient until a seed crystal forms on the window. The seed is annealed for ~ 12 hours. After the annealing the temperature is very slowly lowered another $1/2^\circ$ causing the crystal to grow up the cell. Typical growth is $1/2$ mm/hr.

Brillouin spectra of two neon single crystals of different orientations are shown in Fig. 5. In each spectrum the longitudinal and one transverse component appears. These components are produced by scattering from thermally excited elastic waves in the crystal. In general one expects 2 transverse components in the spectrum, however an estimate of the photoelastic constants for neon allows one to predict that only in certain directions is the Brillouin scattering from both transverse elastic waves sufficiently intense to be seen. If the crystal can be oriented in a direction where both transverse components appear, an accurate determination of the elastic constants is quite simple, as for xenon.

Our present results are based on the Brillouin shifts of the longitudinal and one transverse component for 2 orientations of the same single crystal. From these data we have obtained the following preliminary values for the elastic constants of solid neon, in units of 10^{10} dyne/cm²

$$C_{11} = 1.18 \pm .06 \quad C_{12} = 0.77 \pm 0.06 \quad , \\ C_{44} = 0.56 \pm .06.$$

It should be emphasized that these are preliminary values. We are presently improving both the sample cell and interferometer and hope to have more accurate values soon.

The present experiment has shown that Brillouin scattering is probably the best technique for studying the elastic properties of rare gas single crystals. However, the measurements on xenon and neon crystals described here have covered only a limited temperature range, just below the melting point. Further experimental measurements are necessary and desirable particularly at lower temperatures for comparison with theory.

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In one experiment we also measured the temperature dependence of the phonon lifetime (fig. 4). It was conducted at a constant pressure (35 atm) with an hcp-crystal in the temperature range from 1.2 to 1.9 K. The relatively large error bars presumably come from the lack of sufficient speed in the electronic part of the present apparatus. An influence of anisotropy of the acoustic attenuation can be ignored since all data were collected from the same volume of a distinct crystal.

So far we have not determined the orientation of the helium crystals. Although this should be done in a detailed investigation, our results do not differ too much from one crystal to another.

The observed attenuation of longitudinal phonons at 1 GHz is surprisingly high for these low temperatures. However, the following observation indicates that these large values are not of instrumental origin. With an identical experimental set-up our measurements of the hypersonic attenuation in liquid helium show the expected magnitude and temperature dependence⁴. The short life-times could also be attributed to defective helium crystals. If the sample consists of small crystallites scattering from grain boundaries is to be expected. This, however, seems improbable because of the well known fact that it is very difficult to grow a sample with small crystallites¹³. As the generated phonons travel only 50 μm during observation less than 2 per cent of the phonons can be scattered, if we assume a size of crystallites larger than 1 mm. Another source of error might be scattering from impurities and lattice defects. However, if we use earlier calculations for these processes^{14,15} and estimate the density of defects necessary to account for the observed lifetime, we get concentrations much higher than plausible. From these arguments we believe that we have measured the intrinsic absorption of longitudinal phonons in solid helium.

DISCUSSION

In general scattering of the ultrasonic phonons by thermal phonons is the dominant process for attenuation. For this process calculations are available¹⁶ in which high and low temperature limits are considered. The critical parameter is $\Omega\tau_1$, where Ω is the angular frequency of the ultrasonic phonons and τ_1 is the mean lifetime of thermal longitudinal phonons. At moderately low temperatures, where $\Omega\tau_1 > 1$, but $(T/\theta)^2\Omega\tau_1 < 1$ (θ = Debye temperature) one expects¹⁶ for the lifetime τ of the ultrasonic phonons :

$$\frac{1}{\tau} \propto \Omega T^4 \quad (2)$$

whereas at high temperatures ($\Omega\tau_1 \ll 1$) the absorption of sound should be nearly independent of temperature.

The low temperature limit applies to crystalline quartz for a reduced temperature $T/\theta \sim 1/15$ and a reduced frequency $\omega/\omega_D \sim 1/600$ (ω_D = Debye frequency). These values correspond to $T \sim 1.5$ K and $\omega \sim 1$ GHz for solid helium. Our measurement for longitudinal phonons plotted in fig. 4 suggests a relation

$$\frac{1}{\tau_{\text{long}}} = 3 \cdot 10^7 T \{ \text{sec}^{-1} \} \quad (3)$$

The dependence on frequency has not yet been investigated. Apparently our result does not agree with the prediction of equation (2). A similar discrepancy was observed in experiments on second sound and Poiseuille flow, where it is possible to get an average value for the lifetime of thermal phonons due to normal processes. In these cases the main contribution comes from *transverse* phonons. The mean value $\langle \tau_{\text{trans}} \rangle$ should obey

$$\left\langle \frac{1}{\tau_{\text{trans}}} \right\rangle \propto T^5 \quad (4)$$

This proportionality has proved true in NaF¹⁷. For solid helium, however, at temperatures between 0.3 K and 0.8 K the following relation was found to hold^{18,19}.

$$\left\langle \frac{1}{\tau_{\text{trans}}} \right\rangle = 5 \cdot 10^{11} \left(\frac{T}{\theta} \right)^3 \{ \text{sec}^{-1} \} \quad (5)$$

From both of these experimental results (3) and (5) it is obvious that near 1 K the low temperature limit is not adequate. The same seems to be true for the high temperature limit because of the $1/T$ temperature dependence. It appears that we have made our measurements in an intermediate temperature region and this presumably is also true for the thermal measurements^{18,19}.

Finally, the absolute magnitude of absorption predicted by available theories¹⁶ is more than two orders of magnitude too low compared with the experiment. This discrepancy is presumably caused by using constants of proportionality which are not applicable to helium. On the other hand there may be additional processes which shorten the phonon lifetime. For example, one could imagine that spontaneous decay of phonons occurs¹⁶. In classical crystals this contribution is negligible. However, if lifetime corrections to the spontaneous decay are considered in theoretical calculations²⁰, this effect should play an important role in such an anharmonic crystal like solid helium.

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