Picosecond ultrasonic study of phonon reflection from solid-liquid interfaces

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We have studied the reflection of longitudinal-acoustic phonons with frequencies between 100 and 300 GHz from solid-liquid interfaces using a picosecond ultrasonic technique. Acoustic phonons were generated by absorption of a picosecond light pulse in a metal film. After propagating through a dielectric layer, the phonons were reflected at the solid-liquid interface. We have measured the reflection coefficient at a series of interfaces between dielectric solids and organic liquids for temperatures from 265 to 300 K. The reflection of 205 GHz phonons was also measured between aluminum and liquid argon and nitrogen. The experimental results were compared with the predictions of the classical acoustic-mismatch model. [S0163-1829(97)02804-X]

I. INTRODUCTION

Kapitza[1] was the first to discover that when heat flows across an interface between a solid and liquid helium a temperature jump occurs at the boundary. It was proposed by Khalatnikov[2] that this thermal-boundary resistance (or Kapitza resistance) comes about because phonons incident on the interface may be reflected because of the different acoustic properties of the solid and the liquid helium. Since this early work there have been a large number of studies of the reflection of phonons at interfaces, both at boundaries between solids and liquid helium[3] and at interfaces between two solids.[4] In many of these experiments a pulse of thermal phonons is sent through a crystal towards the interface and the intensity of the reflected phonon pulse is measured. The theoretical studies of the transmission and reflection of phonons at interfaces start with Khalatnikov's acoustic mismatch model[2] (AMM) in which the materials on both sides of the interface are treated as isotropic continua and the reflection coefficient at the interface is given in terms of the densities and acoustic velocities of the materials. At an interface between a solid and liquid helium the reflection coefficients at high frequencies (typically above around 100 GHz), have often been found to be considerably smaller than the predictions from the acoustic mismatch model, yet the microscopic processes involved are still unclear.

The reflection of low-frequency ultrasonic waves from solid-liquid interfaces (less than 1 GHz) at temperatures near room temperature has also been studied in many experiments. In early work by Mason *et al.*[5] the reflection coefficient for shear waves incident on an interface between fused quartz and various polymeric liquids was measured. The frequency range extended up to 60 MHz. The measured value of the complex reflection coefficient was used to find the high-frequency stiffness and viscosity of the liquid.[6] Lamb and co-workers[7] have extended the frequency range of this technique up to a few GHz. The same method has been used to determine the acoustic impedance of liquid helium from the measurement of the reflection of longitudinal-acoustic waves at the crystalline quartz-helium interfaces at 1 GHz.[8]

In this paper we describe a method, picosecond ultrasonics, for the study of reflection of longitudinal-acoustic phonons between solids and liquids at frequencies up to 300

GHz. In the first version of these experiments a high-frequency sound pulse is generated in a metal film, travels through a layer of dielectric (silicon dioxide or silicon nitride), and is then reflected at the interface to a liquid. In an alternate form for the experiment a thin metal film on a substrate is set into vibration at high frequency. The increase in the damping rate of the vibration of the metal film that occurs when a liquid is placed in contact with the metal film is measured. In this form of the experiment the acoustic reflection coefficient at the solid-liquid interface can be calculated from the additional damping of these vibrations caused by sound transmission into the liquid.

II. EXPERIMENTAL TECHNIQUE

In this type of experiment there are several factors that have to be considered in the choice and preparation of samples. (i) Many materials react chemically with liquids, so the solid surface in contact with the liquid during the experiment has to be inert or at least to have a very slow chemical reaction. (ii) Since most solids have an acoustic impedance that is considerably larger than the impedance of liquids, the expected magnitude of the acoustic-reflection coefficient tends to be close to unity. Thus, there is an advantage to a method that can detect small changes in the reflection coefficient, for example, by having an acoustic wave undergo repeated reflections at an interface. (iii) The acoustic wavelength at high frequencies becomes extremely short. In SiO_2 at 100 GHz, for example, the wavelength is 600 Å, and in liquids the wavelength at this same frequency is considerably less. Consequently, it is important to use very smooth surfaces for this type of experiment.

In a first set of experiments we used a sample with the geometry shown in Fig. 1. A 20-Å-thick layer of CF_x, a soft polymer, was first deposited onto a very smooth Si substrate. An aluminum film of thickness in the range 100–300 Å was then thermally evaporated onto the CF_x in a vacuum better than 10^{-6} torr. Finally, a dielectric film of amorphous silicon nitride (a-Si_3N_4) or silicon dioxide (a-SiO_2) of thickness from 500 to 3000 Å was then grown on top of the transducer films by using plasma-enhanced chemical-vapor deposition (PECVD). The thickness of the dielectric film was measured by ellipsometry. It was found that the thickness variation
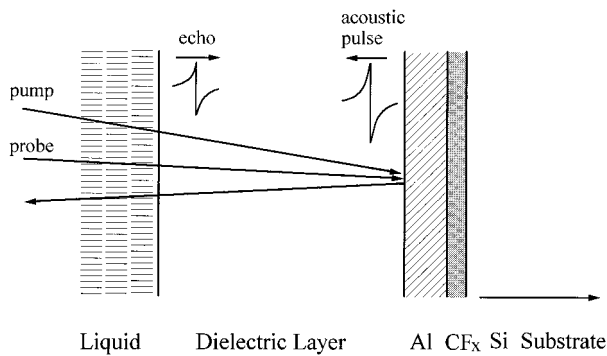
FIG. 1. Schematic diagram of the experiment. The acoustic pulse returning from the dielectric-liquid interface is decreased in magnitude due to partial transmission into the liquid.

FIG. 2. Ultrasonic echoes in a 2500 Å Si_3N_4 film (a) in vacuum and (b) in contact with ethylene glycol at 300 K.

over a surface area of ~ 0.5 cm^2 was less than 1%. The refractive indices of these films at 633 nm were determined to be 1.46 for SiO_2 and 2.10 for Si_3N_4. These values agree well with the bulk values.[9] The Si_3N_4 and SiO_2 layers used in our experiments were deposited by the PECVD technique which is known to produce high-quality, atomically smooth films,[10,11] so that a large amount of diffuse scattering is not expected to occur at the sample surface.

The Al film was used as a transducer to generate and detect ultrasonic waves. To generate sound a light pulse of duration 200 fs was absorbed in the Al film. The light was focused through the dielectric film onto a spot of approximately 20 μm diameter. The light pulses used in the experiment had a wavelength of 730 nm and an energy of 0.5 nJ, and were produced by a self-mode-locked Ti:sapphire femtosecond laser running at a repetition rate of 76 MHz. Absorption of this pulse by the Al film causes a sudden increase in film temperature of a few degrees K. This temperature rise produces a thermal stress distribution in the transducer film that launches a strain pulse into the dielectric film and another pulse through the CF_x towards the silicon substrate. At the high acoustic frequencies studied here the attenuation in the CF_x is very large,[12] so only a very small part of the strain pulse entering the CF_x reaches the silicon and is reflected back to the transducer. The strain pulse that propagates into the dielectric is reflected at the free surface. When it returns to the Al, it changes the optical properties (real and imaginary parts n and κ of the refractive index) of this film, and consequently there is a small change $\Delta R(t)$ in the optical reflectivity. This change is measured by means of a probe light pulse that is time-delayed relative to the pump pulse. The probe pulses has the same duration as the pump pulses, but had approximately ten times less energy and a polarization rotated by 90° with respect to the pump. The magnitude of $\Delta R(t)$ was of the order of 10^{-5}, and so it was necessary to use modulation techniques to obtain the required signal to noise ratio.

Results for $\Delta R(t)$ measured in this way are shown in trace (a) of Fig. 2. These data were obtained from a sample with an Al thickness of 150 Å and with a 2500-Å Si_3N_4 dielectric layer. The oscillations at a frequency of 217 GHz immediately after time zero are the "ringing" of the Al film after it is excited by the light pulse. The feature beginning at around 50 ps is the acoustic echo, i.e., the strain pulse that
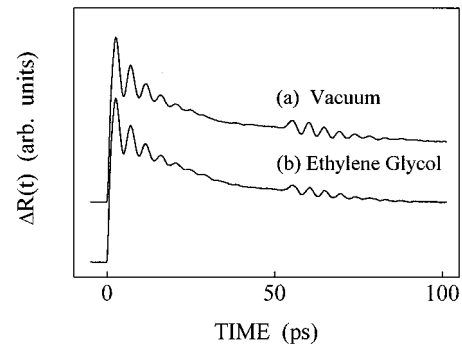
has propagated through the dielectric film and has then been reflected back to the Al film. There is also a smoothly varying background contribution in $\Delta R(t)$ that arises in the following way. When the Al film is heated by the laser pulse the change in its temperature results in a sudden change in the optical reflectivity (thermoreflectance). As the Al film cools via conduction of heat into the materials on either side of it, the optical reflectivity slowly returns to its original value. This gives the background contribution to $\Delta R(t)$ that could be used to determine the cooling curve of the Al film.[13]

The free surface of the dielectric film is then covered with liquid, and $\Delta R(t)$ is measured again giving the results shown in Fig. 2(b). Because part of the acoustic pulse is transmitted into the liquid, the amplitude of the echo is reduced. From the change in the echo size the acoustic-reflection coefficient at the solid-liquid interface can be determined. It is well known that polar-organic liquids absorb well on silica surfaces. In our experiment SiO_2 and Si_3N_4 were used as dielectric layers to be in contact with the liquid. In order to assure strong bonding between the dielectric layer and the liquid we chose to study ethylene glycol as a first application to our method. The data in Fig. 2(b) were obtained with ethylene glycol in contact with the Si_3N_4.

Although the basic idea of this measurement is straightforward, one has to take care to analyze the data correctly in order to determine the acoustic-reflection coefficient. After the introduction of the liquid layer the pump and probe beams have to be refocused and the area of the sample onto which the light is focused may therefore change. In addition, even if the reflection coefficient of the acoustic wave at the solid-liquid interface is unity, the amplitude of the echo feature in $\Delta R(t)$ will change when the liquid is added. This is because the introduction of the liquid changes both the static optical reflectivity of the sample and also the magnitude of any reflectivity change resulting from a modification of the optical properties (e.g., refractive index) of one of the component layers. To allow for this we first make a fit to the thermal background component of the response and subtract this from the data. We then scale the two data sets so that the amplitude of the initial ringing of the transducer (oscillations near to $t=0$) are the same both with and without the liquid present. We next compare the acoustic echoes $\Delta R_{liq}(t)$ and $\Delta R_{vac}(t)$ as they appear in these scaled data. The Fourier transforms of these echoes are taken and the acoustic-reflection coefficient $r(\omega)$ is calculated according to
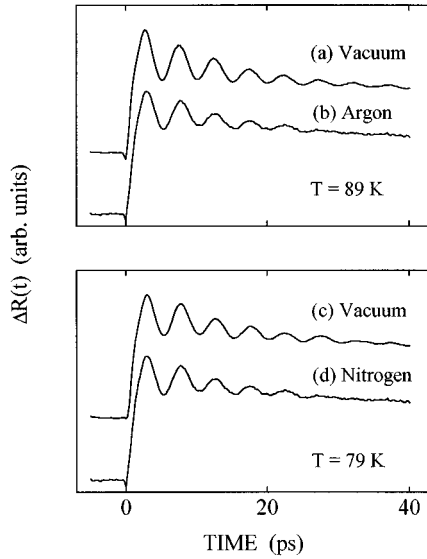
FIG. 3. Measurements of the change $\Delta R(t)$ in reflectivity of the probe pulse for a 150 Å Al film on a Si substrate with a 20 Å CF$_x$ intermediate layer (a) in vacuum at $T=89$ K, (b) in liquid argon at $T=89$ K, (c) in vacuum at $T=79$ K, and (d) in liquid nitrogen at $T=79$ K. The additional damping in (b) and (d) is caused by sound transmission into argon and nitrogen, respectively.

$$r(\omega) = \Delta R(\omega)_{\text{liq}} / \Delta R(\omega)_{\text{vac}}. \qquad (1)$$

The peak response frequency of the transducer ω_0 is related to the thickness d_{Al} and the sound velocity v_{Al} of the Al film by $\omega_0 = \pi v_{\text{Al}} / d_{\text{Al}}$. Thus, by making this type of measurement with a series of samples with different thicknesses of the Al transducer film, we can measure the reflection coefficient r over a range of frequencies.

In this experiment the dielectric layer has two functions. First, the acoustic transit time through this layer makes it possible to see a time-resolved echo coming from the solid-liquid interface. Second, the liquid that is studied does not have to be in direct contact with the particular material that is used as a transducer. Thus, it is possible to perform experiments with liquids that react chemically with the transducer, but do not react with the material of the dielectric layer. In the event that chemical reaction with the transducer material is unlikely, there is no need to use a dielectric layer. In this case when no liquid is present the response of the Al transducer consists of an oscillation damped by acoustic radiation through the CF$_x$ into the silicon substrate. Examples of such responses for a 150 Å Al film are shown in traces (a) and (c) of Fig. 3. The addition of liquid increases the damping rate of the oscillations, and from this increase the acoustic-reflection coefficient at the solid-liquid interface can be calculated. Data taken on the same sample with the Al in contact with liquid argon and nitrogen are shown in traces (b) and (d) of Fig. 3.

To analyze this type of data, let the frequency of oscillation of the Al film when the liquid is present be ω and let the amplitude damping rate be Γ. Then it is straightforward to show that

$$1 - r_{\text{Al-CF}_x} r_{\text{Al-liq}} \exp[2i(\omega - i\Gamma)d_{\text{Al}} v_{\text{al}}] = 0, \qquad (2)$$
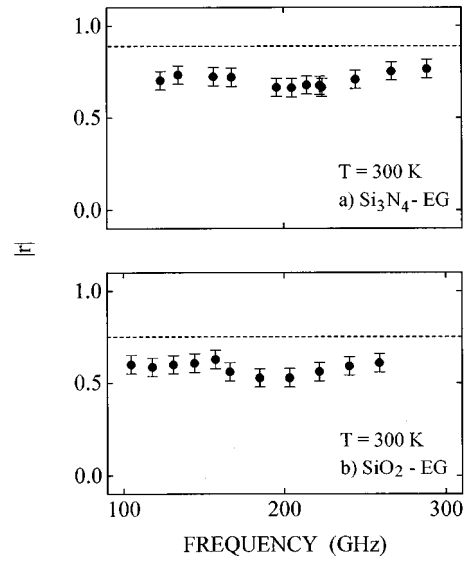

FIG. 4. Magnitude of the acoustic-reflection coefficient measured for (a) Si$_3$N$_4$– and (b) SiO$_2$–ethylene glycol interfaces as a function of frequency at 300 K. The dashed lines are predictions from the acoustic mismatch model.

where $r_{\text{Al-CF}_x}$ and $r_{\text{Al-liq}}$ are the reflection coefficients for Al-CF$_x$ and Al-liquid interfaces. If follows that the magnitude of the reflection coefficient at the Al to liquid interface is given by the expression

$$|r_{\text{Al-liq}}| = \exp\left[\frac{-2(\Gamma - \Gamma_0)d_{\text{Al}}}{v_{\text{Al}}}\right], \qquad (3)$$

where Γ_0 is the damping rate of the Al film in vacuum. To obtain the damping rate of each data set of the type shown in Fig. 3 we first subtracted the thermal background from the raw data leaving the acoustic part of the response. We then fitted the acoustic response to a function of the form

$$\cos(\omega t)\exp(-\Gamma t), \qquad (4)$$

by a nonlinear least-squares-fitting routine.

III. RESULTS AND DISCUSSION

A. Organic liquids

Results for $|r|$ as a function of frequency at the interface between Si$_3$N$_4$ and SiO$_2$ and ethylene glycol at a temperature of 300 K are shown in Fig. 4. The dashed lines at $|r|=0.89$ for Si$_3$N$_4$ and 0.75 for SiO$_2$ were calculated from the AMM,

$$|r| = \left|\frac{Z_{\text{EG}} - Z}{Z_{\text{EG}} + Z}\right|, \qquad (5)$$

where Z and Z_{EG} are the acoustic impedances (product of density and sound velocity) for the dielectric film and ethylene glycol, respectively. We used the values[14–16] 3.19 and 2.2 g cm^{-3} for the densities, and 10.1×10^5 and 5.9×10^5 cm s^{-1} for the sound velocities for Si$_3$N$_4$ and SiO$_2$, respectively. The acoustic impedance of ethylene glycol was determined from its density ($\rho_{\text{EG}} = 1.112$ g cm^{-3}) and sound velocity ($v_{\text{EG}} = 1.67 \times 10^5$ cm s^{-1}) at MHz frequencies.[17]
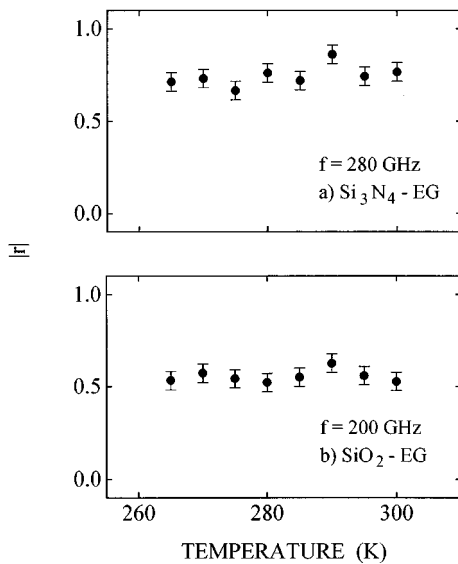
FIG. 5. Magnitude of the acoustic-reflection coefficient at (a) Si_3N_4- and (b) SiO_2-ethylene glycol interface versus temperature at 280 and 200 GHz, respectively.

Results of measurements of $|r|$ versus temperature from 265 to 300 K (ethylene glycol freezes at 260 K) are shown in Fig. 5 for both Si_3N_4 and SiO_2 at frequencies of 280 and 200 GHz, respectively. The results in Figs. 4 and 5 show no frequency or temperature dependence for $|r|$ to within experimental error for both solids.

The theoretical value of the reflection coefficient just calculated from the AMM is based on the use of the sound velocity in the liquid as measured at low frequency. The observation that the measured $|r|$ is smaller than the calculated value could be interpreted to suggest that the sound velocity, and hence also the acoustic impedance, is higher at the very high frequencies studied in our experiment. Our results give an average value of $|r|$ over the frequency range studied of 0.71 and 0.58, for Si_3N_4 and SiO_2, respectively. For the AMM to agree with these values the sound velocity in ethylene glycol would have to be 4.58 and 3.10×10^5 cm s^{-1} for the Si_3N_4 and SiO_2 data, respectively. This would amount to an increase relative to the low-frequency velocity by a factor of 2.7 or 1.9. Because there is no clear frequency dependence indicated in our measurements of $|r|$, the change in velocity from the low-frequency value would have to be essentially complete by the time a frequency of 100 GHz was reached. Brillouin scattering measurements can be used to give the sound velocity in a frequency range around a few GHz. For ethylene glycol such measurements have been made by Wang, Lin, and Jones.[18,19] The velocity that they have obtained is within a few percent of the low-frequency velocity. Thus, if velocity dispersion is the explanation of our results, it would have to occur in the frequency range between a few GHz and 100 GHz. For highly viscous associated liquids like ethylene glycol, analysis of low-frequency ultrasonic attenuation and velocity measurements[20] has led to the conclusion that there may be a 15–60 % dispersion in the sound velocity due to structural relaxation, and this dispersion could possibly occur in the

required frequency range. However, even a change of this magnitude would be insufficient to give the values of $|r|$ that we have measured.

A second possible explanation is that the properties of the liquid near to the solid surface differ from those of the bulk. The molecules of the liquid near the surface can form a more highly ordered layer that is stiffer than the bulk, especially when the liquid wets the surface.[21–23] Interferometric measurements have shown that silica has a low adhesion tension against organic liquids,[24] thus it adsorbs organic liquids quite well. This suggests that ethylene glycol may have a stiff surface layer on Si_3N_4 and on SiO_2, which could serve to acoustically match the two materials, resulting in a greater transmission of phonons across the interface. This approach has been used in the past to try to explain the anomalously large transmission of phonons from solids into liquid helium.[25] In the helium context this approach has not been successful. The essential difficulty has been that anomalous phonon transmission has been found to occur even for phonons of frequency as low as 30 GHz. For these phonons the wavelength in the liquid is ~ 80 Å, i.e., considerably larger than the thickness that one can reasonably assume for an adsorbed layer of high-density helium on the surface of the solid. The same sort of objection can be made to the present measurements; the wave length of sound in ethylene glycol at 200 GHz is 84 Å, assuming the low-frequency value of the sound velocity, and the wave length would be even larger if there is velocity dispersion.

There is an important distinction to be made between the type of measurement reported here and most of the studies on the interface between solids and liquid helium. The helium studies have primarily been carried out using pulses of incoherent phonons radiated by heated thin films or produced by superconducting tunnel junctions.[4] The detectors that were used were either bolometers or tunnel junctions. Because these detectors did not require the reflected phonons to be coherent, it was possible to measure separately the phonons that were reflected specularly and diffusely from the interface, and to see how the intensity of these two components was affected when helium was added to the surface of the solid. The AMM is a theory for the reflection coefficient of the specularly reflected phonons. For interfaces between solids and helium the AMM value for the magnitude of the energy reflection coefficient is typically greater than 0.98. For very smooth surfaces or for phonons of very low frequency the reflection is mainly specular. It is found experimentally that the addition of helium to the surface then causes only a very small reduction of the reflected phonon intensity. Thus, this is in agreement with the predictions of the AMM.[26–29] For less perfect surfaces the helium has a far larger effect. In particular, the diffusely scattered component of the phonon intensity may be reduced to as little as 20% of its original value. It is this reduction in reflection, and the related anomalously large transmission of energy across the interface, which gives rise to the so-called anomalous Kapitza conductance, and which has been the subject of intense study. In our experiment the measured acoustic echo is proportional to the strain due to the returning echo integrated across the surface of the transducer. Any component of the reflected strain wave that is diffusely scattered at the free surface of the sample will have a phase that varies rapidly
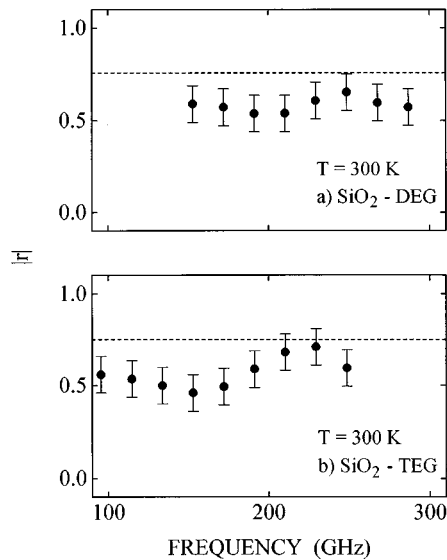
FIG. 6. Magnitude of the acoustic reflection coefficient measured for (a) SiO_2-diethylene glycol and (b) SiO_2-triethylene glycol interfaces as a function of frequency at 300 K. The dashed lines are predictions from the acoustic mismatch model.

across this surface and so will not be detected. Thus, our experiment looks only at the specularly reflected component of the strain. For helium we are unaware of any quantitative study of the reflection coefficient for the *specularly reflected* component of the phonon flux.

The measured reflection coefficients between SiO_2 and diethylene glycol (DEG), and triethylene glycol (TEG) as a function of frequency at room temperature are shown in Fig. 6. The solid lines at $|r| = 0.76$ for diethylene glycol and 0.75 for triethylene glycol are the acoustic mismatch predictions. We have used the values[17] 1.116 and 1.123 g cm^{-3} for the densities and 1.59×10^5 and 1.65×10^5 cm s^{-1} for the longitudinal sound velocities at MHz frequencies for diethylene glycol and triethylene glycol, respectively. Although all three ethylene glycols (EG, DEG, TEG) vary significantly in molecular weight and viscosity,[30] we have not observed a significant difference in the reflection coefficient, although it has to be admitted that there is considerable scatter in these data.

The random error in the measured value $|r|$ arises from the uncertainties in the Fourier transforms of the echoes $\Delta R(w)_{liq}$ and $\Delta R(w)_{vac}$. For a given sample the uncertainty in $|r|$ was about 10% for both SiO_2 and Si_3N_4. For inter-

faces between ethylene glycol and SiO_2 and Si_3N_4 we investigated the variation between different samples and found it to be $\pm 15\%$ for both interfaces.

B. Liquid argon and nitrogen

Measurements of the reflection at interfaces to liquid argon and nitrogen were also made. In these experiments no dielectric buffer layer was used, and the acoustic-reflection coefficient between Al and the liquid was determined from an analysis of the ringing of the transducer as already described. The measurements were made at 89 and 79 K for liquid argon and nitrogen, respectively. We used the values[30–32] 2.70, 1.40, and 0.81 g cm^{-3} for the densities, and 6.6×10^5, 8.5×10^4, and 9.6×10^4 cm s^{-1} for the sound velocities of Al, Ar, and N_2, respectively. These values give theoretical AMM reflection coefficients of 0.875 and 0.916 for Ar and N_2, respectively. The corresponding measured values were 0.87 and 0.86, both with an estimated uncertainty of 5%. Thus for these liquids the AMM is in reasonable agreement with the experimental results. These measurements were made at a single frequency of 205 GHz.

For both Ar and N_2 the velocity measured at the Brillouin frequency is in good agreement with the low-frequency ultrasonic results.[33]

IV. SUMMARY

In this paper we have described a method for the study of phonon reflection from solid-liquid interfaces at frequencies between 100 and 300 GHz, and have presented the data obtained with this technique. The results for the phonon reflection coefficient are compared with the predictions of the acoustic mismatch model. For the ethylene glycol and di- and triethylene glycol the measured reflection is significantly less than the AMM prediction, whereas for argon and nitrogen theory and experiment are in reasonable agreement. In the near future we plan to extend these measurements to cover a broader frequency range. Measurements at lower frequencies for ethylene glycol would be of interest because clearly the AMM must give the correct answer at sufficiently low frequency. In addition, it should be possible to study interfaces to helium using this technique and to relate the results to other studies of phonon reflection in this system and the Kapitza conductance.

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