Thermal-phonon resonance

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(Received 6 September 1994; revised manuscript received 14 June 1995)

We have observed the resonance of thermal phonons confined in a small cavity by Brillouin scattering detected by an optical beating technique. Longitudinal resonance modes of sub-GHz phonons were found at every 760 kHz in the resonance cavity composed of two flat walls set face to face with 880 μm spacing. The optical beating technique offered a frequency resolution as high as ~kHz to the Brillouin scattering measurement, which was thus able to detect the spectrum of phonon resonance. The spectrum observed is well described by a theory which takes the effect of finite correlation time of the phonon phase into consideration.

I. INTRODUCTION

Thermally excited phonons can be observed as the Brillouin components of a light scattering spectrum. The peak frequency gives the phonon energy and the width represents the inverse phonon lifetime.[1] This kind of experiment is usually made in a space without any boundaries, i.e., the scattering volume is sufficiently large in comparison with the decay length of the relevant phonon. If phonons are confined within a cavity smaller than their spatial coherence length, a resonance effect occurs. Sandercock demonstrated the resonance spectrum of phonons in thin solid films up to μm thickness.[2] Recently, we succeeded in observing this effect with a hyper-resolution Brillouin scattering system especially prepared for this purpose. We experimentally demonstrated that thermal phonons of sufficient coherence length can interfere with each other.

Most studies of Brillouin scattering are aimed at observing GHz phonons and the experiments have been conducted almost exclusively with Fabry-Pérot etalons as spectrometers for the scattered light.[3] Experimental instances of thermal-phonon resonances have actually been reported with this conventional method. Sandercock performed experiments in the GHz region and observed the resonance of thermal phonons in thin solid films.[2] Alvarenga *et al.* also showed an analogous result for a thin water sample.[4] In this study, we investigate the phonon-resonance effect in a liquid sample confined within a resonance cavity of macroscopic size. Phonon damping is generally much larger in liquids than in solids. We observed, therefore, sub-GHz phonons whose coherence length is typically as large as ~mm. In addition, the interval of the resonance peaks is expected to be less than MHz when the cavity length is of the order of a mm. Very high resolution frequency analysis is thus required for the present measurement to resolve the fine structure of the phonon-resonance spectrum. High contrast is also needed to reduce the stray central component which is especially harmful in measurements at low scattering angles. We therefore used a system with an optical beating spectroscopy technique, which was capable of analyzing the power spectrum with a resolution of kHz.[5,6] The quantitative examination of the shape of resonance curves was thus possible using known values of phonon velocity and damping constant. The experimental results are in good agreement with the theoretical prediction.

II. EXPERIMENT

The phonon resonator is composed of two optical flat glasses of 150 μm thickness set parallel to each other. An optical flat spacer is sandwiched in between and the parallelism of the cavity is kept better than 10″. Figure 1 shows the schematic view of the phonon resonator and the light scattering configuration. In the experiment, the thickness of the resonator plates was chosen so that they work as ideal reflectors with respect to the phonon wave-

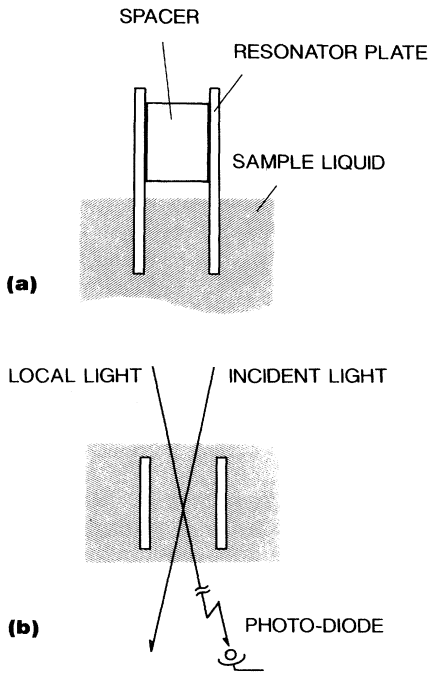

FIG. 1. Schematic view of the phonon resonator and light scattering configuration: (a) side view and (b) top view.

length. The reflection coefficient is theoretically calculated with the plate thickness d, densities and sound velocities of the reflector, and the sample liquid. The reflection coefficient is safely approximated to $R > 0.98$ at the condition $2df = (m + 1/2)v_g$, where v_g is the sound velocity in the resonator plate, f is the frequency, and m is an integer. The sound velocity obtained by the ultrasonic technique is $v_g = 5\,640$ m/s. This approximation holds sufficiently well over the frequency range ± 5 MHz around 30 and 66 MHz, where the experiments were carried out.

The configuration of the light scattering system is very simple.[5] An incident beam and a local beam intersect in the sample, and a high-speed photodetector is placed to receive the local light. The power of the incident and local lights are 200 and 0.3 mW, respectively. Phonon creation through a nonlinear effect, such as stimulated Brillouin scattering, does not occur at these light intensities. We can safely regard the phonons observed as "thermal phonons." The incident light is scattered into the direction of Bragg reflection with respect to the phonon of interest and the scattering angle gives the phonon wave number through the Bragg condition. The local and the scattered light take the same path and are mixed at the photodetector which generates the beating signal, whose power spectrum is analyzed by an electrical apparatus.

Experiments of light scattering in a sample contained in a small cell have been extremely difficult since strong stray scattering inevitably occurs at the optical windows of the sample cell. This stray light gives an overwhelmingly strong central component whose skirt hides the Brillouin doublet if observed with a limited frequency resolution. This problem is particularly serious in the present experiment made at low scattering angles for two reasons: the stray light is usually stronger in the forward direction and the Brillouin peaks are close to the center. Though this problem would be solved with a very narrow pass-band spectrometer, even the most sophisticated system of optical Fabry-Pérot interferometer could hardly have such a high resolution. On the other hand, the frequency resolution of the optical beating Brillouin spectroscopy technique used in this study is as high as 10^4–10^5, sufficient for this experiment.

In addition to this resolution, we developed a polarization modulation of the laser for lock-in detection, which is essentially important for this experiment. In our previous system, we used a mechanical chopper for on-off modulation of the incident light.[5] This conventional lock-in detection with amplitude modulation was shown to be useless in this light scattering. The stray light is so strong that it is readily visible and, in some occasions, glaring at the photodetector. Since its main source is the incident laser, the stray light is chopped and therefore passes through the lock-in system. Of course its dc (and hence the central) component is effectively filtered out with the high resolution of the receiver, but the wide-band shot noise component passes and causes a very serious rise of the base line of the spectrum. This shot noise bias is much larger than the Brillouin signal and the latter is completely submerged under the base line. In our system, we keep the laser intensity constant and, instead, rotate the polarization plane of the incident light so that the optical mixing with the local light occurs periodically at the lock-in frequency. Major part of the shot noise component is removed with the lock-in detection and only the Brillouin signal is clearly observed. Detail of this polarization modulation will be given elsewhere.[7]

Liquid toluene was chosen as the sample because of its large scattering efficiency, as well as the small sound damping. The phonon decay length is about 14 mm at 30 MHz and inversely proportional to the square of the phonon frequency. The laser beam with an initial diameter of 1 mm is focused by a lens with a focal length of 500 mm. The angular divergence induced by the lens ($\sim 0.1°$) gives a wide detection band of phonons, which corresponds to 6 MHz in the frequency domain.

III. RESULTS AND DISCUSSION

Figure 2(a) shows a power spectrum of the thermal phonons in the resonator observed at the scattering angle of 0.5°. The cavity length is 880 μm. The resonance peaks are spaced by 760 kHz, which agrees with the interval of the resonance frequencies theoretically given by $\Delta f = v/2L$, where L is the cavity length and v is the pho-

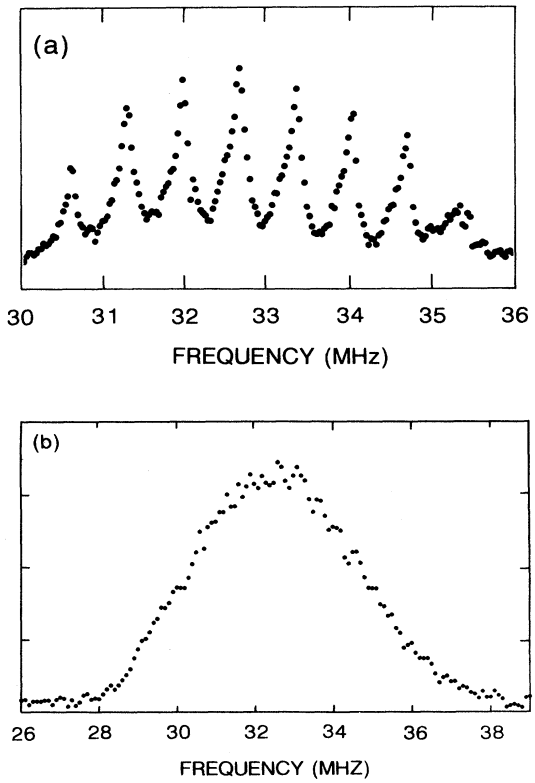

FIG. 2. Power spectra of thermal phonons in liquid toluene observed (a) in the cavity and (b) outside the cavity. The scattering angle is 0.5° in the sample liquid for both experiments. The lower spectrum approximately gives the envelope of the resonance peaks of the upper spectrum.

non velocity in liquid toluene. Figure 2(b) shows the spectrum observed with the same condition as 2(a) but at a scattering point just outside the resonator: in that case, no fine resonance structure is observed. The width of the observed broad Brillouin component is determined essentially by the ambiguity of the scattering angle introduced by the focusing lens. This spectrum therefore represents the instrumental band of this light scattering system, whose width ($\sim$6 MHz) is in agreement with the value estimated from the angular divergence.

Here arises one simple question: why can we observe the resonance of thermal phonon? Phonons are thermally excited fluctuations of local density and the thermal energy is to be equally distributed all over the phonon branches. It means the energy spectrum of the thermal phonon should be "white." This request of thermal equilibrium condition should hold even in the cavity. From the viewpoint of classical statistics, an ensemble of phonons is regarded as thermally excited acoustic waves, which propagate at the sound velocity. Acoustic waves with any wave number can be excited thermally in the resonator: wave numbers off the resonance condition are not prohibited *a priori*. Nevertheless, the observed power spectrum has the resonance structure as shown in Fig. 2. We explain this phenomenon in terms of wave propagation.

This phenomenon is understood by considering a finite correlation time of phase of thermally excited acoustic wave. We have to pay attention to three time constants which may affect the apparent power spectrum. One is the time duration of interaction between phonons and laser light, which is determined as the traveling time of acoustic wave to cross the finite diameter of the laser beam D. The interaction time is then given by D/v. The broadening of the Brillouin peak induced by this effect is usually attributed to the divergence of the laser beam with finite diameter and agrees with the instrumental width described above. Secondly, a phonon has an intrinsic lifetime due to absorption. The broadening of the Brillouin peaks by this effect, Γ, represents the temporal damping constant of phonons, and $\Gamma=100$ kHz in the present case of liquid toluene at the phonon frequency of 30 MHz. Observed Brillouin spectra are usually explained by the convolution integral of these two functions representing the instrumental and intrinsic contribution. In the present case, however, we have to take another correlation time into account. This is the correlation of the phonon phase which is introduced by the resonance cavity. A thermally excited acoustic wave propagates and once intersects the laser beam in the cavity, and then crosses it again after a round trip between the two reflectors. Note that the phonon which propagates forward generates the anti-Stokes component of Brillouin triplet, while that which goes backward contributes to the Stokes component. Scattered lights corresponding to these two components has different frequencies and can be resolved with a spectrometer. When the temporal oscillation of the thermally excited acoustic wave is still in phase after one round trip, the apparent temporal correlation length is infinitely long and the line broadening due to this effect does not occur. The condition of this "resonant" mode is represented as

$$kL=n\pi\ , \tag{1}$$

where n is an integer and k the wave number of the relevant acoustic wave. If Eq. (1) does not hold, on the other hand, the correlation time of the phase is restricted by the period of a round trip of the thermal acoustic wave in the resonator. The observed phase shifts by $2kL$ after each round trip. Taking this into consideration, we calculate the power spectrum of thermal phonons with wave number k as

$$S(\omega,k)\propto\left|\frac{1}{i(\omega-kv)+\Gamma}\times\frac{e^{\{2i(kv-\omega)L/v\}-2L\Gamma/v}-1}{e^{2ikL}e^{\{2i(kv-\omega)L/v\}-2L\Gamma/v}-1}\right|^2 . \tag{2}$$

Here, the cavity is assumed to be composed of two perfect reflectors as discussed previously. When the intrinsic phonon damping is much smaller than the phonon frequency and $\Gamma\ll\omega$ holds, a simple description of the power spectrum $S(\omega)$ is obtained by approximating $2(kv-\omega)L/v\ll1$:

$$S(\omega,k)\propto\frac{1}{(\omega-kv)^2+\Gamma^2}\times\frac{1}{1-2e^{-2L\Gamma/v}\cos(2kL)+e^{-4L\Gamma/v}} . \tag{3}$$

The apparent spectrum is then obtained as

$$P(\omega)\propto\int dk\, I(\omega)S(\omega,k) . \tag{4}$$

The function $I(\omega)$ represents the instrumental band of the detection system. Sandercock derived a similar equation of resonance spectrum for the back scattering configuration assuming $kL=n\pi$ as the possible boundary

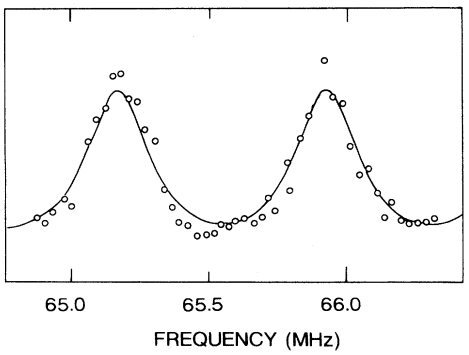

FIG. 3. Thermal phonon resonance observed around 65 MHz. The scattering angle is $\sim$1.0°, and the order of the resonance for the two peaks is $n=84$ and 85. The solid line shows the theoretical curve calculated with Eqs. (3) and (4). The bandwidth of the electrical filter used to analyze the spectrum is 30 kHz, and the line broadening due to the phonon leakage through the resonator wall is expected to be less than 20 kHz. These instrumental effects give 10–15 % additional width to the observed spectrum, which is within the experimental error.

condition for an unsupported thin solid film.[2] In our discussion, the effect of intrinsic phonon damping was taken into consideration for the quantitative examination of the experimentally obtained resonance spectrum.

Figure 3 shows the resonance peaks of phonon observed at the scattering angle of 1.0° in the same resonator. The solid line represents the theoretical curve of resonance spectrum calculated by Eq. (4) using the cavity length, and literature values of sound velocity and sound damping.[8] The observed spectrum is well fitted by the theoretical curve as shown in the figure. This system would be capable of accurate and noncontact measurement of phonon velocity and damping constant in a small cavity.

The phenomenon of thermal phonon resonance and its observation by high resolution Brillouin scattering spectroscopy could be applied to the study of dynamic properties of materials confined in a microscopic space. It is known that various interesting structures are formed in the vicinity of interfaces, such as a wetting layer, an adsorbed layer, and the layer structures seen in liquid crystals. The dynamics of these structures at the molecular level could be investigated through the fluctuation in density, local orientation, and other properties which are observed by the present experiment of thermal phonon resonance.

ACKNOWLEDGMENTS

This work was partly supported by a Grant-in-Aid for Scientific Research from the Ministry of Education, Culture and Science, Japan. The authors thank Mr. Kokubo of the University of Tokyo for his technical help to the experiment.

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