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Low-angle Brillouin scattering under a temperature gradient

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Abstract. The asymmetry appearing in the dynamic structure factor of a medium under a non-equilibrium state of temperature gradient was experimentally verified. Brillouin scattering was carried out in water with the optical beating spectroscopy technique at frequencies lower than 60 MHz where a large asymmetry was expected. The finite-cell-size effect to which the disagreement between the theory and experiment had been attributed was excluded by a specially designed tall cell which suffers from the least effect of the finite size. The observed Stokes line was shown to have intensity 2% to 8% higher than the anti-Stokes line depending on the phonon frequency for a temperature gradient applied into the same direction as the scattering vector. Based on a concept of anisotropic phonon propagation under a temperature gradient, a simple theory was proposed which succeeded in estimating the asymmetry for the temperature distribution of actual experimental condition including the effect of cell boundaries. The observed asymmetry was quantitatively in good agreement with the theoretical prediction.

1. Introduction

A medium kept out of the thermodynamic equilibrium has attracted a great interest for its peculiar phenomena which would provide us with an insight into the profound physics of condensed matters. Inspired by those theoretical works [1–3], several groups have performed experimental studies to detect the behaviour unique to non-equilibrium. The most successful experiments were the ones made on the dynamic structure factor of thermal fluctuation in a medium under a temperature gradient [4, 5]. The authors used a sophisticated system of Brillouin scattering to observe the asymmetry between the Stokes and anti-Stokes components, which should have the same intensity in the equilibrium state. The asymmetry is theoretically predicted to have an order of magnitude sufficiently detectable for the phonon peaks at frequencies around 50 MHz. The very careful experiments have succeeded in detecting the asymmetry, but the magnitude seems consistently smaller than the predicted value.

The most probable cause of this disagreement is the boundaries of the light scattering cell restricting the range of temperature gradient in the medium, which is assumed to have an infinite and uniform extension in the theoretical approach. The quantitative estimation of the boundary effect was reported by some authors [6, 7]. However, it is too difficult to modify the theory to fit the actual experimental conditions, for example inhomogeneous temperature gradient. There have also been some technical difficulties in the quantitative analysis. The experiments have been performed with a Fabry-Pérot interferometer specially designed for a very high finesse. Nevertheless, the frequency resolution cannot exceed the limit of optical

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spectrometers, and is still insufficient to eliminate completely the harm done by the huge central component generated by the stray scattered light. The central peak which is 10^4 times as high as the phonon peaks superimposes its skirt onto the Brillouin components as is usual in experiments of low scattering angles, changing their shape and height. The small asymmetry due between these components would seriously be affected. Further, one spectrum of a Brillouin triplet is an accumulation of numerous data which requires a long experiment time, typically 24 hours [4]. The instability of the overall system is likely to cause an additional ambiguity to the results. The quantitative comparison between the theory and the experiment has not been made in a satisfactory manner.

In this paper, we first present a new theoretical description of the asymmetry in the Brillouin triplet, which is based on a concept of anisotropic phonon propagation under temperature gradient. The advantage of this model is that we can readily take the effect of cell boundaries into account and predict the asymmetry for the real experimental conditions including non-uniform temperature gradient. Focusing the purpose of study onto an experimental verification of the asymmetry, we decided to conduct Brillouin scattering using the technique of optical beating spectroscopy [8] instead of the Fabry-Pérot etalon. This technique has a remarkable frequency resolution of 1 kHz bandwidth, which filters out the troublesome influence of the central peak on phonon spectra even in the range down below 20 MHz [9]. One sweep of the spectrum takes only 15 min and a lack of long-time stability causes no serious problem to the result. We prepared two different types of liquid container for applying a temperature distribution in the medium, a conventional Rayleigh–Benard type and a tall cylindrical cell. The results were successfully correlated with the theory of phonon propagation proposed in this paper. The asymmetry in Brillouin spectra under thermal non-equilibrium was experimentally confirmed.

2. Asymmetric phonon propagation under temperature gradient

There have been various theoretical approaches [7, 10] to statistical physics under non-equilibrium conditions. Though they give the same result in their final form, the most preferred and common theory of these would be the one which solves the hydrodynamic equation [11]. The fluctuation–dissipation theory gives the dynamic structure factor which is associated with the light scattering spectrum. According to the theory, the Stokes component has relatively stronger intensity than the anti-Stokes one when the scattering vector is set in the same direction as the temperature gradient. The asymmetry is characterized by a parameter $\varepsilon = (I_S - I_{AS})/(I_S + I_{AS})$, where I_S and I_{AS} are the intensity of the Stokes and anti-Stokes components, respectively. The theory predicts that ε depends on ∇T, the temperature gradient, and Γk^2, the temporal decay rate of the relevant phonon, as

$$\varepsilon_{th} = \frac{c}{2\Gamma k^2}\frac{\nabla T}{T_0}. \tag{1}$$

Here, c is the phonon velocity and T_0 is the temperature at the scattering point. Note that an infinite range of ∇T is assumed.

From this equation, one can expect a substantial asymmetry of $\varepsilon \sim 15\%$ for phonon at 30 MHz ($k = 1.2 \times 10^3$ cm^{-1}) in water with $\nabla T = 20$ K cm^{-1}, a feasible temperature gradient in a Rayleigh–Benard cell [12] sandwiched between a heater and a cooler. Equation (1) suggests two difficulties in the experimental conditions. First, the dependence of ε on the inverse of k^2 demands that the measurement be made at very low-angle scattering in which the central component gives the most serious problem. This problem was settled in the present study by the hyper-resolution spectroscopy of the optical beating technique.

Secondly, the size of the Rayleigh–Benard cell is to be chosen very carefully. To obtain a sufficient value of ∇T with a practical temperature difference between the two faces, the distance in between must be shorter than a certain length. On the other hand, the limited length may be against the theoretical requirement that ∇T should extend over an infinite range. In fact, the asymmetry would be hardly observable when the distance is shorter than the phonon decay length. To cope with this second difficulty, we propose here the theoretical approach which is applicable to non-uniform temperature distribution in the tall cell designed for the present purpose.

This theory is based on a simple idea that the thermal phonons are excited with a probability proportional to the local temperature, and propagate into the scattering region with the spatial decay. The density–density correlation function in the equilibrium state is represented by

$$S(k,\omega) = 2(k_B T/\omega)\,\mathrm{Im}[\chi(k,\omega)]. \tag{2}$$

The Brillouin triplet is generally included in $\chi(k,\omega)$, the mechanical response function of the system. The probability of phonon excitation in the equilibrium state proportionally depends on $k_B T$. Consider a small volume in the medium with temperature gradient: the total number of phonons generated in the volume per second would be proportional to the local temperature at the point. This idea can be extended to the non-equilibrium state. For simplicity, we take a temperature gradient vertically upward and the scattering vector in the same direction after the theory of equation (1). The experiment in this study was actually performed in this configuration and detected those phonons propagating upward and downward corresponding to the anti-Stokes and Stokes components, respectively. The phonons generated in the hotter region and going downward have larger population than the opposite phonons and contribute to the Stokes peak enhancing its intensity. The asymmetric parameter is determined from I_S and I_{AS} which are given by counting the numbers of phonons generated in both regions and coming into the scattering volume. Note that the phonon number decreases as $\exp(-2a|z|)$ with distance $|z|$ to the scattering volume, suffering from the amplitude attenuation α. We can thus calculate I_S and I_{AS} as

$$\begin{aligned} I_S &\propto \int_0^\infty k_B T(z)\ \exp(-2\alpha|z|)\,\mathrm{d}z \\ I_{AS} &\propto \int_{-\infty}^0 k_B T(z)\ \exp(-2\alpha|z|)\,\mathrm{d}z. \end{aligned} \tag{3}$$

Here, $T(z)$ is a function representing the temperature distribution and the origin of the z-axis is set at the scattering point. When the temperature gradient is uniform, i.e. $T(z) = \nabla T z + T_0$, and the sample size is much larger than the phonon decay length $1/\alpha$, equation (3) gives the asymmetric parameter

$$\varepsilon = \frac{1}{2\alpha}\frac{\nabla T}{T_0}. \tag{4}$$

This equation exactly agrees with equation (1) of the previous theory with the well known relation $\Gamma k^2 = c\alpha$.

The advantage of equation (3) is that one can also treat the case of inhomogeneous temperature gradient by substituting the actual temperature distribution into $T(z)$. In addition, this model enables us to take the boundary effect into account. If the phonon propagation length is longer than the cell size along z, the phonons reflect at the bottom and top boundaries. The phonons from the hot region pass once through the scattering volume downward contributing to the Stokes component and then, after reflection at the bottom, come back up again as the anti-Stokes phonons. The apparent asymmetry would seriously be suppressed. If the measurement is to be made in water at 50 MHz, for example, the propagation length is estimated from $1/\alpha$ to be 2 cm. We cannot expect an observable asymmetry with the cell shorter than 1 cm [13]. This model also suggests that a homogeneous temperature gradient is not necessary but a large temperature difference is important. From these viewpoints, we constructed a new experimental system more appropriate to the present purpose.

3. Experiment

The optical beating technique has an excellent frequency resolution and is especially useful for the measurement of low-frequency phonon. The details of the system have already been described elsewhere [8], and we give here a brief account for the modifications made in the present study. Figure 1 shows a schematic view of the measurement system. Light from an Ar ion laser is divided into two beams and the frequency of one is shifted up with an acousto-optic modulator by 80 MHz. This beam is used as a local oscillator light of the optical heterodyne and the other, as the incident light. The two beams are focused by a lens with 500 mm focal length and intersect each other at their beam waist in the sample. The scattering angle is determined as the crossing angle between them. A high-speed photodiode detects the scattered light together with the local oscillator light. Analysis of the photo beat current gives the Brillouin spectrum, in which the anti-Stokes and Stokes components appear at (80 MHz $-$ f_p) and at (80 MHz $+$ f_p), respectively, where f_p is the phonon frequency.

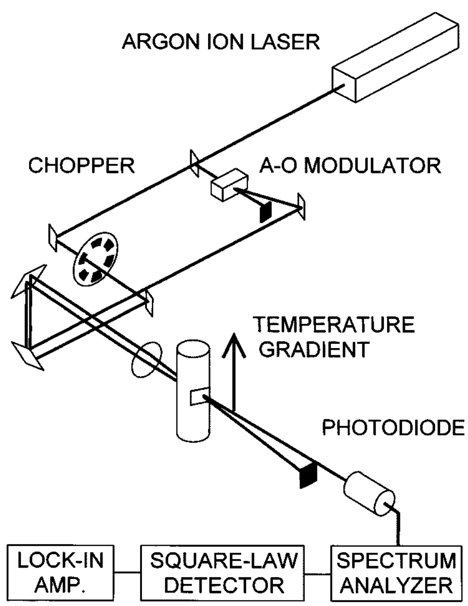

Figure 1. Schematic view of the experimental system. The scattering vector is set vertical along the temperature gradient.

One of the most serious problems common to the light scattering under a temperature gradient is the effect of beam deflection in the index gradient [5]. Non-negligible error is given to the scattering angle, hence to the phonon wave number. In the present system of cross beam geometry, however, the incident and the local beams suffer from the deflection effect to almost the same degree, and error in the scattering angle can be ignored. The temperature gradient is applied upward to prevent the thermal convection of water. The incident and local lights cross in the vertical plane.

In order to obtain a large temperature difference, yet to reduce the boundary effect to a negligible level, we newly designed a tall cylindrical cell 24 cm high and 3.9 cm in diameter with two optical windows at half height. The upper half of the cell is heated up to 90 °C by a heater while the lower part was cooled at 0 °C. The temperature distribution in water was measured by a platinum resistance thermometer and is shown in figure 2. We can expect an asymmetry parameter as large as $\varepsilon \sim 6\%$ at 30 MHz in this cell. We have also prepared a Rayleigh–Benard cell 1.5 cm thick with 4 cm side to check the results of previous studies. The temperature gradient in the cell was measured and shown to be homogeneous with $\nabla T = 20$ K cm^{-1}.

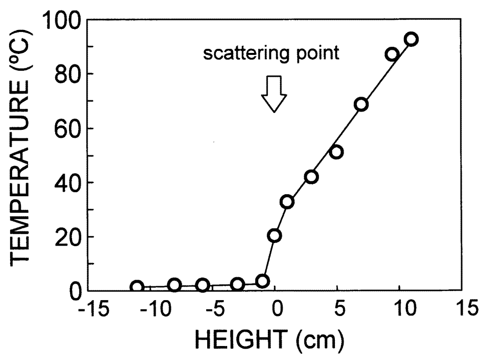

Figure 2. Temperature distribution in the tall cylindrical cell. The arrow indicates the position of the scattering volume.

4. Results and discussion

Figure 3 shows the Brillouin spectra obtained (a) in the equilibrium state and (b) under the temperature gradient in the tall cylindrical cell. While the Brillouin doublet in (a) has a symmetric intensity as is expected, the spectrum of (b) apparently shows asymmetry. The Stokes component at −25 MHz is 8% higher than the anti-Stokes component. Note that the Brillouin peaks are completely isolated from the central component which is too narrow and absent in this spectrum scanned 10 MHz off the centre. Broadening of the Brillouin peaks, as large as several MHz, is mainly the instrumental width due to the angular divergence of the laser beam and is larger by far than the intrinsic broadening expected from the phonon decay. In such a situation, we can safely consider that the observed peak height is proportional to the intensity of the Brillouin component, from which the asymmetry parameter was determined.

The asymmetric parameter obtained in the Rayleigh–Benard cell is shown in figure 4. In this study, we express ε as a function of $1/f^2$ instead of $\nabla T/T_0$ for the reason given later. The corresponding phonon frequencies are indicated in the upper scale. As shown in the figure, the asymmetry is not observable within the experimental error. The dashed and the solid lines represent the theoretical prediction of equations (1) and (3), respectively.

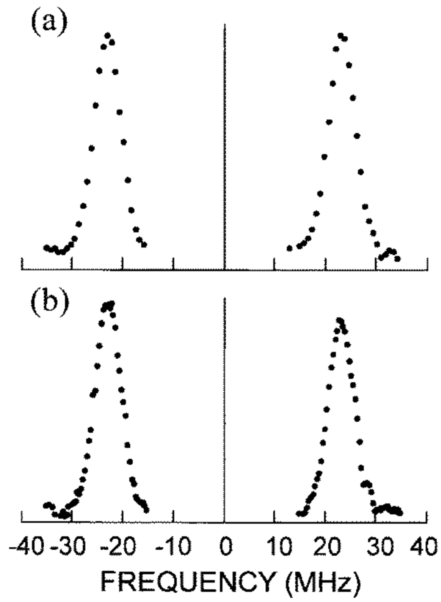

Figure 3. Brillouin spectra obtained with the tall cylindrical cell (a) in the equilibrium state, and (b) under the temperature gradient.

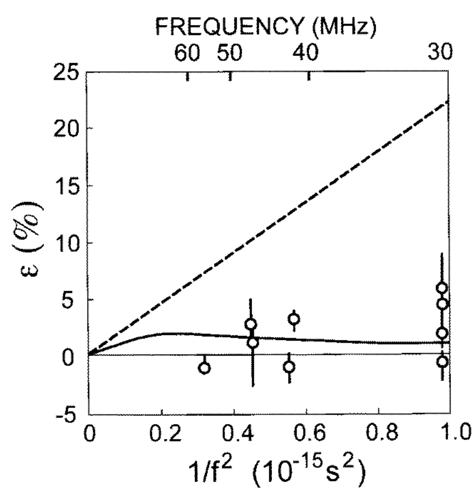

Figure 4. The asymmetric parameter observed in the Rayleigh–Benard cell. The dashed and solid lines are the theoretical curves of equations (1) and (3), respectively.

The experimental points are much lower than the dashed line showing no proportional dependence on $1/f^2$ (hence on $1/k^2$). The theoretical curve of equation (3), which includes the boundary effect, is consistent with the experimental points, though the values are smaller than the experimental error. In the calculation of equation (3), we assumed that top and bottom of the cell work as an ideal phonon reflector.

Figure 5 shows the ε parameter obtained in the tall cell. The temperature gradient is not uniform in this cell and ε cannot be properly described in terms of $\nabla T/T_0$. Therefore, we chose $1/k^2$ as a more suitable variable of the experiment. We changed the scattering

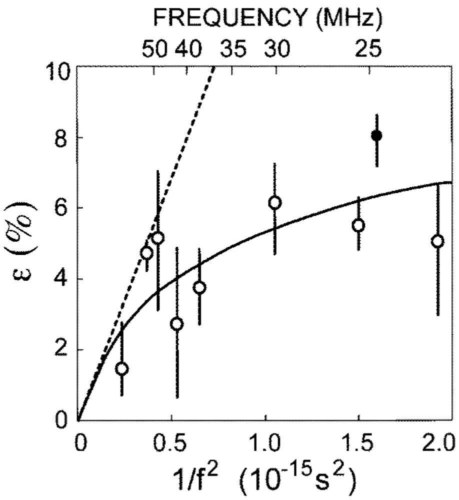

Figure 5. The asymmetric parameter in the tall cylindrical cell. The dashed line shows the prediction of equation (1) as ∇T with the temperature gradient at the scattering point. The solid line represents the prediction of equation (3) as $T(z)$ with the real distribution shown in figure 2. The closed circle is the result obtained with the beat frequency exchanged between Stokes and anti-Stokes components.

angle from 0.4° to 1.3°, corresponding to $k = 1.70 \times 10^3$ cm^{-1} to 2.77×10^3 cm^{-1}. The more practical value would be the phonon frequency $f(= ck/2\pi)$, however, and figures 4 and 5 are shown as a function of $1/f^2$. The phonon frequency was changed over the range 20 MHz to 70 MHz as given in the upper abscissa. The open circles represent the result obtained with the local oscillator whose frequency is shifted up by 80 MHz as written in the preceding section. The detection sensitivity of the photodiode slightly depends on the beat frequency, and might give a systematic error to the intensity ratio of the observed Brillouin peaks. For this reason, we performed the same experiment but with a local oscillator whose frequency is shifted down by 80 MHz, exchanging the beat frequencies of the anti-Stokes and the Stokes lines. The closed circle represents the result of the second experiment, showing the photodiode characteristics give no detectable error. The dashed line is the theoretical curve of equation (1) calculated under the assumption that the temperature gradient at the scattering point extends infinitely. The theory overestimates the asymmetry by neglecting the fact that the actual temperature gradient decreases above and below the scattering region. The solid line shows the theoretical calculation of equation (3) with the measured temperature distribution shown in figure 2. The experimental points are in good agreement with the prediction of equation (3) derived from the present theory.

In conclusion, we succeeded in verifying experimentally the intensity difference between the Stokes and anti-Stokes components of Brillouin spectrum under thermal non-equilibrium. The asymmetry observed in the tall cylindrical cell with a large temperature difference was in quantitative agreement with the prediction of practical theory based on a concept of anisotropic phonon propagation.

Acknowledgments

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