Forced Brillouin Spectroscopy Using Frequency-Tunable Continuous Wave Lasers

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We have developed a new method of phonon spectroscopy using *forced Brillouin scattering*: A scanning interference pattern produced by intersecting two cw lasers generates the density variation (acoustic phonons) through the thermal expansion for a light-absorbing liquid. When the dispersion relation of phonons is satisfied, phonons are generated resonantly in the liquid. Continuous tuning of the frequency difference between the two lasers enables us to measure a resonance spectrum of light-excited phonons using light scattering phenomena. We demonstrate that this resonance spectrum is equivalent to the Brillouin spectrum of thermal phonons both experimentally and theoretically.

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Acoustic phonons that are thermally excited in condensed matter such as liquids and solids scatter light, which is known as spontaneous Brillouin scattering. Brillouin spectroscopy based on this phenomenon has been widely applied for studying acoustic phonons in the frequency range over several hundred megahertz where mechanical excitation of phonons are difficult [1]. Although this method provides us with information on acoustic phonons without any perturbation to the samples, the scattering intensity is extremely weak because of a very weak intensity of thermally excited phonons. Stimulated Brillouin scattering induced by intense laser beams, on the other hand, amplifies the intensities of thermal phonons with a nonlinear process and, thus, the resulting Brillouin scattering intensity is enhanced [2]. However, this method has a crucial disadvantage since the phenomenon is intrinsically nonlinear and distorts the shape of a Brillouin spectrum significantly. Recently, continuous wave stimulated Brillouin gain spectroscopy (SBGS) [3] has been developed as the extension of coherent Raman spectroscopy [4]. This method provides us with a high-frequency resolution phonon spectroscopy on the assumption that the pump laser power is weak enough for the linear approximation to hold. It should also be noted that Nelson *et al.* [5] have succeeded in measuring phonons generated by intersecting pulse laser beams in the time domain (impulsive stimulated Brillouin scattering).

In this Letter, we demonstrate *a new light scattering method of Brillouin spectroscopy using very coherent acoustic phonons optically generated by two frequency-tunable continuous wave lasers.* This method is based on the optical generation of phonons and the simultaneous optical heterodyne detection of the phonons using the phonon-induced Bragg diffraction. Thus, our method is essentially different from SBGS, which is based on the phenomena of the energy transfer from the high-frequency laser beam to the low-frequency one through Brillouin-induced four-wave mixing. In other words, our method is a light scattering method, while SBGS is a method based on nonlinear optical four-wave mixing.

First we briefly explain the principle of the optical generation of acoustic phonons. Since Bell [6] first successfully demonstrated generation of audio-frequency sound waves by using light, some methods to excite radio-frequency sound waves using lasers have been proposed and tried up to now [3,5,7–12]. A common principle of phonon generation using lasers is shown in Fig. 1. There two intersecting laser beams L1 and L2 can be regarded as monochromatic plane waves. Here we define the intensity, angular frequency, and wave vector of Li($i = 1, 2$) as I_i, ω_i, and $\vec{k}_i$, respectively. The light intensity distribution in the region where the two beams intersect, which we call the *intersecting volume*, is given by

$$I_{\text{total}}(\vec{r}, t) = I_1 + I_2 + \Re[2\sqrt{I_1 I_2}\exp i(\vec{k}\cdot\vec{r} - \omega t)], \tag{1}$$

where $\vec{k} = \vec{k}_2 - \vec{k}_1$ and $\omega = \omega_2 - \omega_1$. Equation (1) implies that the scanning interference pattern is produced. Thus, if there exist any mechanisms that induce variations of the density corresponding to the light intensity distribution in the medium, sound waves of frequency ω and of wave vector $\vec{k}$ are likely excited resonantly under the condition that the phase velocity of the scanning interference pattern is nearly equal to the sound velocity c_s, i.e., $\omega \simeq c_s k$, where $k = |\vec{k}|$. The density variation can be produced *in phase* with the light-intensity modulation by two different mechanisms: an electrostriction

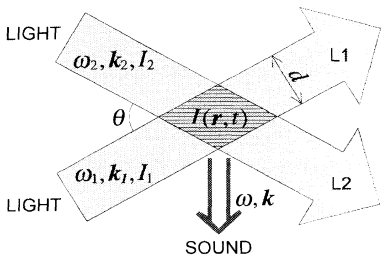

FIG. 1. Schematic representation of sound generation by laser lights.

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effect [7,8] and a thermal-expansion effect due to light absorption [9,13–15].

Here we describe the theoretical background of our new Brillouin spectroscopy using light-excited phonons. We calculate the scattering function of a light-absorbing material under two intersecting light beams, provided that the thermal-expansion effect is much more significant than the electrostriction effect and the latter is negligible. In such a case, the changes in the density $\rho(\vec{r},t)$ and the temperature $T(\vec{r},t)$ induced by the light electric field whose intensity distributions is $I(\vec{r},t)$ are given by the following equations [9]:

$$\rho_0 C_p \frac{\partial T}{\partial t} - \Lambda \nabla^2 T = \alpha_l I, \tag{2}$$

$$\frac{\partial^2 \rho}{\partial t^2} - c_s^2 \nabla^2 \rho - D_s \frac{\partial}{\partial t} \nabla^2 \rho = c_s^2 \beta \rho_0 \nabla^2 T, \tag{3}$$

where ρ_0 is the equilibrium value of ρ, C_p is the specific heat at constant pressure, Λ is the thermal conductivity, α_l is the light absorption coefficient, and β is the coefficient of volume expansion. D_s is the sound damping constant given by $D_s = (c_s^3/2\pi^2)\alpha_s/f^2$, where α_s is the sound damping coefficient and f is the sound frequency $\omega/2\pi$. The steady state solution of Eqs. (2) and (3) for the dynamic perturbation in Eq. (1) is given by

$$\rho(\vec{r},t) = \Re[\rho_1(\vec{k},\omega)\exp i(\vec{k}\cdot\vec{r} - \omega t)], \tag{4}$$

with

$$\rho_1(\vec{k},\omega) = \frac{2c_s^2\beta\rho_0 k^2 \alpha_l \sqrt{I_1 I_2}}{(\omega^2 + iD_s k^2\omega - c_s^2 k^2)(i\rho_0 C_p \omega - \Lambda k^2)}. \tag{5}$$

Equation (4) shows the generation of a traveling sound wave, i.e., acoustic phonons whose angular frequency and wave vector are ω and $\vec{k}$, respectively. Thus, the scattering function is given by

$$S(\vec{k},\omega) = \frac{\rho_0 c_s^3}{4}\left|\frac{\rho_1(\vec{k},\omega)}{\rho_0}\right|^2 = \frac{c_s^7\beta^2 k^4 \alpha_l^2 I_1 I_2}{\rho_0 C_p^2[(\omega^2 - c_s^2k^2)^2 + (D_s k^2\omega)^2][\omega^2 + (\Lambda k^2/\rho_0 C_p)^2]}. \tag{6}$$

The right-hand side of Eq. (6) is equivalent to the Rayleigh-Brillouin spectra of thermal density fluctuations for the case of $c_s k \gg (\Lambda/\rho_0 C_p)k^2$, which is usually satisfied for the range of k of light scattering. Equation (6) clearly indicates that the measurement of the intensity of light-excited sound as a function of k and ω with sweeping ω in the vicinity of $c_s k$ provides us with information equivalent to spontaneous Brillouin spectroscopy.

Next we describe the instrumentation and the experimental results supporting the above theoretical prediction. Figure 2 indicates the block diagram of our experimental system. We used two lasers (model 140-0532-100 and -10, Lightwave Electronics) that are single axial mode, continuous wave output, frequency doubled (green) Nd-YAG (neodymium yttrium aluminum garnet) lasers. The frequency is continuously tunable over 10 GHz by changing the voltage exposed to an external connector of the laser. The jitter that determines the frequency resolution of the system is ~150 kHz/s. The output powers of the two lasers used for this experiment were 20 and 10 mW. The absolute center frequencies of the tunable range are 18 788.4 ± 0.1 cm (roughly 532 nm) for both lasers. Thus, their tunable ranges have an overlap, and the beat frequency between the two lasers can be controlled to be any value between 0 and 10 GHz [16]. Thus, we can produce the coherent phonons having a desired frequency in an intersecting volume, according to the principle described above.

The most serious problem in our method as spectroscopy is how to detect the generated phonons. Although the intensity of the light-induced phonons well exceeds that of thermal phonons inside an intersecting volume, it decays very rapidly outside of the volume, especially at high frequencies. Therefore, it is impossible to detect the phonons mechanically with a transducer. We have to use the Bragg diffraction of light by phonons, which is used in the usual spontaneous Brillouin spectroscopy. A second laser whose wavelength is fairly different from that of the laser for phonon excitation is sometimes introduced as a probe light to detect the phonons by Bragg diffraction [5,9]. In this method, however, we must readjust the incidence angle of the probe light and also the angle for detection of the diffracted light, whenever we change the intersecting angle of exciting lights. This readjustment process complicates the measurement. To avoid this complication, we introduce a probe light made of the exciting light collinearly to it. In this configuration, the Bragg condition for the generated phonons is automat-

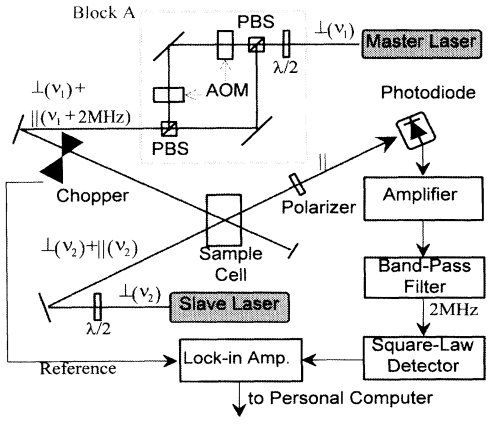

FIG. 2. Block diagram of our experimental system. PBS stands for polarizing beam splitters, AOM for acousto-optic modulators, and $\lambda/2$ for half-wave plates. || shows light with horizontal polarization, while ⊥ light with vertical polarization.

ically satisfied for the probe light, and the high diffraction efficiency can be achieved because of the perfect coincidence between the scattering volume and the intersecting volume. In Fig. 1, L1 and L2 satisfy the Bragg condition to generate sound, and L1 and L2 are diffracted to the path of L2 and L1, respectively.

The only remaining problem is to distinguish the diffracted light from the exciting light. This problem has been successfully solved by combining the selection of the diffraction light by polarization with that in the frequency domain (see Fig. 2). The light output of the block A in Fig. 2 consists of two collinear orthogonally (vertically and horizontally) linearly polarized beams with a frequency difference of 2 MHz $[\perp(\nu_1) + \|(\nu_1 + 2\ \text{MHz})]$. The light output of the slave laser from the half-wave plate, on the other hand, consists of two collinear orthogonally (vertically and horizontally) linearly polarized beams with the same frequency $[\perp(\nu_2) + \|(\nu_2)]$. These two beams intersected in a sample cell, and the light components of $\perp(\nu_1)$ and $\perp(\nu_2)$ produced a scanning interference pattern, which generates sound waves in the sample. The light component of $\|(\nu_2)$ was used as a local beam for the heterodyne detection, and that of $\perp(\nu_2)$ was removed by a polarizer. Thus, the light of $\|(\nu_1 + 2\ \text{MHz})$ was diffracted by excited phonons of a frequency of $\nu_2 - \nu_1$ to produce the diffraction light of $\|(\nu_2 + 2\ \text{MHz})$. This diffraction light was mixed with the above local light on a *p-i-n* photodiode (PD) with a low-noise, transimpedance amplifier (Hamamatsu, S2858). The output signal of the photodiode that includes the beat signal of frequency 2 MHz was fed into a bandpass filter of center frequency 2 MHz and of band width 100 kHz. Then, its output was square-law detected. In order to suppress the influence of the shot noise from the local light, the output of the square-law detector was lock-in detected by light chopping at a frequency of ~100 Hz.

Figure 3 shows phonon spectra of liquid toluene colored with a red dye (Mitsubishi Chemical, HSR-2179), whose light absorption constant α_1 at the wavelength of 532 nm was set to be 1.0 cm^{-1}. It should be noted that the concentration of the dye is a few ppm, so that it never affects the acoustic properties. Open circles show a spectrum under the configuration where the light of $\perp(\nu_2)$ is off and only thermal phonons exist in the sample, which corresponds to spontaneous Brillouin scattering [16]. Closed circles, on the other hand, show a resonance spectrum of phonon under the configuration where the light of $\perp(\nu_2)$ is on and coherent phonons are generated by laser beams, which corresponds to the spectrum predicted by Eq. (6). The half linewidth $\Gamma_B/2$ is about 7.5 MHz for both spectra, and their peak positions are exactly the same. This result clearly shows that the resonance spectrum of optically generated phonons is identical with the Brillouin spectrum of thermal phonons (fluctuation-dissipation theorem). Figure 4 shows the resonant spectra measured at three different k's. The peak positions exactly coincide with the values calculated from the dispersion relation.

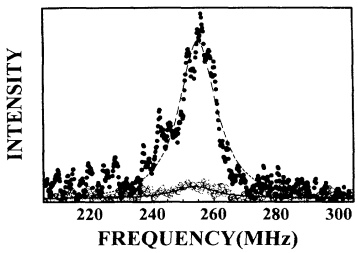

FIG. 3. Phonon spectra of toluene at $T = 300$ K at $k/2\pi = 0.197\ \mu\text{m}^{-1}$. Open circles show a spectrum obtained from spontaneous Brillouin scattering by thermal phonons, while closed circles show a resonance spectrum of phonons excited by continuous wave lasers. Solid and dashed lines are fitted curves of Lorentzians for open and closed circles, respectively.

Here we estimate the ratio of the intensity between the optically generated phonons and the spontaneous thermal phonons theoretically. The intensity of thermal phonons contributing to the scattering is given by

$$S_0 = (k_B T_0 c_s/V)\delta f/(D_s k^2/2\pi)\,, \tag{7}$$

where k_B is the Boltzmann constant, T_0 is the equilibrium value of the temperature T, V is the scattering volume, δf is the bandwidth of the signal detection and 100 kHz in our case, and $D_s k^2/2\pi$ is about 2 MHz. Under the geometrical configuration shown in Fig. 1, we can regard L1 to be an excitation light and L2 to be a light diffracted by thermal phonons. Then, the scattering volume V coincides with the intersecting volume, which is given by $V \sim d^3/\sin\theta$ (θ is the intersecting angle between the two laser beams and d is the diameter of the light beam). On the other hand, the intensity of optically generated phonons can be estimated from Eq. (6) with the resonant condition $\omega = c_s k$ as

$$S_{\max} = S(\vec{k}, c_s k) = c_s^3 \beta^2 \alpha_l^2 I_1 I_2/\rho_0 C_p^2 \Gamma_B^2\,. \tag{8}$$

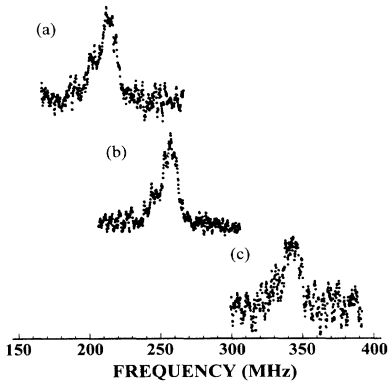

FIG. 4. Phonon spectra of toluene at $T = 300$ K obtained by forced Brillouin spectroscopy. (a) $k/2\pi = 0.164\ \mu\text{m}^{-1}$, (b) $k/2\pi = 0.197\ \mu\text{m}^{-1}$, and (c) $k/2\pi = 0.262\ \mu\text{m}^{-1}$.

For our experiment, the above phonon intensities can be estimated numerically. The physical constants of liquid toluene at $T_0 = 293$ K are $\rho_0 = 0.866$ g/cm^3, $c_s = 1.33$ km/s, $\beta = 1.07 \times 10^{-3}$ K^{-1}, $\Gamma_B/2\pi = 15$ MHz, and $C_p = 1.76$ J/K g. Other parameters are $\lambda_0 = 532$ nm, $I_1 = I_2 = 10$ W/cm^2, $\theta = 4°$, $d = 0.35$ mm, and $\alpha_1 = 1$/cm. With these parameters and Eqs. (7) and (8), we obtain $S_{\max} = 1.1 \times 10^{-12}$ W/cm^2 and $S_0 = 4.4 \times 10^{-14}$ W/cm^2. Thus, we estimate the ratio of the intensity between the optically generated phonons and thermal phonons to be about 25, which reasonably agrees with the ratio of 13 experimentally obtained (see Fig. 3).

Finally, we point a remarkable feature of our method as a measurement method of wave propagation. In our method, both the $\vec{k}$ and ω are fixed at each measurement, and the resonant behavior is observed as a spectrum. Such a method to excite *phonons with both* $\vec{k}$ *and* ω *fixed* has not existed in conventional acoustic phonon spectroscopy so far. In a mechanical method for exciting sound wave, only ω is fixed; in a Bragg reflection method where phonons are mechanically excited, for example, the dispersion of k is measured for a fixed ω. In usual Brillouin scattering measurements, on the other hand, we fix k and measure the dispersion of ω. Thus, all the conventional methods measure a cross section along a line (k = const or ω = const) of the dispersion ridge in the ω-k space. Our method, on the other hand, enables us to measure its height at any point in the ω-k space.

In conclusion, we have developed a new light scattering method of phonon spectroscopy using *forced oscillation of acoustic phonons by coherent light-beating fields.* We call this new method "forced Brillouin spectroscopy" (FOBS), in connection with "forced Rayleigh scattering" where a static interference pattern is produced in a sample colored with a dye by intersecting two laser beams with the same frequency to analyze a self-diffusion mode. FOBS and SBGS [3] are now the only frequency-domain spectroscopic methods using light-induced phonons that can provide us with a very high-frequency resolution. Thus, it is meaningful to point out some advantages of FOBS over SBGS, although they are based on the very different physical processes, as described before: (1) SBGS measures the total signal that is a product of the amplitude gain $g(\omega) \propto \omega S(\vec{k}, \omega)$ and the probe laser power. Thus, the stability of the probe laser power is a prerequisite for obtaining $g(\omega)$ accurately [3]. Our method directly measures $S(\vec{k}, \omega)$ and, thus, is free from this problem. (2) Further, SBGS has a problem of the intrinsic nonlinearity since it is based on the assumption that $\exp[g(\omega)l] \sim 1 + g(\omega)l$ (l is the interaction length) [3]. This nonlinearity problem becomes serious when the pump laser power is increased to improve the signal-to-noise ratio. Our method is free from this dilemma.

Our new method provides a noncontact phonon spectroscopy for liquids, as in spontaneous Brillouin scattering measurements. It should be noted that we must use an electrostriction effect for solids instead of thermal expansion because of the very small thermal-expansion coefficients in solids. If we increase the powers of exciting light for phonon generation (*only 10 mW in our experiment*) up to 100 mW, we can easily generate by an electrostriction effect in both liquid and solid the phonons whose intensity well exceeds that of thermal phonons, according to our estimation. We hope that *forced Brillouin spectroscopy* will be applied in a wide field of condensed matter physics.

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Note added.—After the submission of our paper, the paper by E. P. N. Damen, A. F. M. Arts, and H. W. de Wijn was published in Phys. Ref. Lett. **74**, 4249 (1995). It should be noted that our method is essentially different from theirs in both the mechanism of phonon generation and the phonon detection method, although both use frequency-tunable lasers to generate acoustic phonons.

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