A New Phase-Coherent Light Scattering Method: First Observation of Complex Brillouin Spectra

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We demonstrate here a new *phase-coherent light scattering method*. Spontaneous Brillouin scattering is caused by thermally excited phonons whose phases have no physical meaning because of their random, incoherent nature. However, coherent phonons having *phase* information can be generated resonantly by an optically induced scanning interference pattern, when the dispersion relation of phonons is satisfied. By a phase-sensitive detection of the light scattered by optically generated coherent phonons, we have succeeded in measuring *complex resonance spectra of acoustic phonons*, which we call complex Brillouin spectra, with an ultimate signal-to-noise ratio. [S0031-9007(97)03706-X]

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Dynamic light scattering is one of the most powerful experimental means to study the dynamics of condensed matter, including simple liquids, complex fluids, and solids [1]. This method measures the spectrum of light scattered by thermally excited modes such as diffusion modes and propagating sound modes. Since the phase of any thermally excited mode is inevitably random in principle, the conventional light scattering method can measure only the power spectrum of these modes.

In general, we can measure a response function of the material by two methods: (i) measurement of the power spectrum of thermal fluctuations and (ii) measurement of the response function to an external field. Here we consider the applicability of method (ii) to the light scattering measurement of acoustic phonons, using the simplest equation of a damped oscillator as an example:

$$\frac{\partial^2 \rho}{\partial t^2} + \Gamma \frac{\partial}{\partial t} \rho + \Omega^2 \rho = F(t), \tag{1}$$

where ρ is the density, Γ is the damping constant, and Ω is the angular eigenfrequency. This equation becomes a Langevin equation if $F(t)$ is the random stochastic noise satisfying the fluctuation-dissipation theorem. The resulting density fluctuations have the following power spectrum as a function of the angular frequency ω:

$$\langle \rho_\omega^2 \rangle = V k_B T \rho^2 \chi_T \, \mathrm{Re}\left[\frac{1}{-\omega^2 + \Omega^2 + i\Gamma\omega} \right], \tag{2}$$

where χ_T is the isothermal susceptibility. On the other hand, it becomes the equation of a forced oscillator for $F(t) = F_0 e^{-i\omega t}$. In this case, the density of the ω component, ρ_ω, is directly given by

$$\rho_\omega = \frac{F_0}{-\omega^2 + \Omega^2 + i\Gamma\omega}. \tag{3}$$

Thus, in principle, we can measure the complex spectra of phonon resonance by inducing the forced oscillation via a coherent external field.

The measurement of such complex spectra has two important meanings: (i) observation of complex resonance spectra itself and (ii) realization of the ultimate signal-to-noise ratio (SNR) and frequency resolution. Since we already explained point (i) above, we briefly explain point (ii) below. The scattering intensity of thermal phonons is determined solely by the temperature and frequency resolution of the measurement [see Eq. (2)]. The Fabry-Perot interferometer [2] provides us with the relatively high SNR by sacrificing the frequency resolution (>100 MHz, typically). A superheterodyne Brillouin scattering method expands the frequency range of the usual optical heterodyne method [3]. However, its high frequency resolution (~1 MHz) makes the measurement over a few GHz extremely difficult. The only way to increase SNR is to use optically generated phonons instead of thermal phonons. Along this line, we have recently developed a forced Brillouin scattering method using optical generation of phonons [4]. However, SNR is still not good enough to increase the upper frequency limit over several GHz. To realize both high SNR and frequency resolution, which are fundamental requirements for high-precision spectroscopy, we need another breakthrough. In this Letter, we describe how we can measure complex resonance spectra of acoustic phonons experimentally on the basis of the above principle, and how we can realize both ultimately high SNR and frequency resolution.

The principle of our method is to directly measure *complex resonance spectra of acoustic phonons* under forced oscillation by a light field. This is essentially different from the conventional Brillouin scattering methods, which measure the power spectrum of light scattered by acoustic phonons with a finite lifetime (spatially decaying propagating sound waves). We describe here the theoretical background of our coherent Brillouin scattering method (COBS). It is well known [5–7] that coherent acoustic phonons can be optically generated by intersecting two coherent lasers ($L1$ and $L2$) in a sample. The light intensity distribution in the region where the two beams intersect is given by $I(\vec{r}, t) = I_1 + I_2 + \delta I(\vec{r}, t)$ with $\delta I(\vec{r}, t) = \mathrm{Re}[2E_1 E_2 e^{i(\vec{k}\cdot\vec{r} - \omega t)}]$. Here the intensity, angular frequency, and wave vector of Li $(i = 1, 2)$ are

expressed by $I_i = E_i^2$ (E_i: electric field strength), ω_i, and $\vec{k}_i$, respectively. $\vec{k} = \vec{k}_2 - \vec{k}_1$, and $\omega = \omega_2 - \omega_1$. This relation implies that a scanning interference pattern with a phase velocity of $\vec{k}/\omega$ is produced. Thus, if there exist any mechanisms that induce variations of the density corresponding to $\delta I(\vec{r}, t)$, the sound waves of frequency ω and wave vector $\vec{k}$ are excited resonantly under the condition that the phase velocity of the scanning interference pattern is nearly equal to the phonon velocity c_s, i.e., $\omega \simeq c_s k$, where $k = |\vec{k}|$.

The changes in the density $\rho(\vec{r}, t)$ and the temperature $T(\vec{r}, t)$ induced by the light intensity modulation $\delta I(\vec{r}, t)$ are given by the following equations, including both electrostriction and thermal-expansion effects [8]:

$$\frac{\partial^2 \rho}{\partial t^2} - \frac{c_s^2}{\gamma}\nabla^2\rho - \frac{\eta}{\rho_0}\frac{\partial}{\partial t}\nabla^2\rho - \frac{c_s^2\beta\rho_0}{\gamma}\nabla^2 T = -\frac{1}{4\pi}\left(\frac{n^2-1}{2}\right)\left(\frac{n^2+2}{3}\right)\nabla^2\delta I, \tag{4}$$

$$\rho_0 C_v \frac{\partial T}{\partial t} - \lambda\nabla^2 T - \frac{C_v(\gamma-1)}{\beta}\frac{\partial \rho}{\partial t} = \frac{1}{4\pi} nc\alpha\delta I, \tag{5}$$

where ρ_0 is the equilibrium value of ρ, n is the refractive index, C_v is the specific heat at constant volume, λ is the thermal conductivity, α is the light absorption coefficient, c is the speed of light, β is the coefficient of volume expansion, η is the damping constant, and $\gamma = C_p/C_v$ (C_p: the specific heat at constant pressure). The phonon lifetime τ is given by $\tau \sim \rho_0/\eta k^2$. The steady-state solution of Eqs. (4) and (5) for the dynamic perturbation $\delta I(\vec{r}, t)$ is given by $\rho(\vec{r}, t) = \mathrm{Re}\,[\rho_1(\vec{k}, \omega)e^{i(\vec{k}\cdot\vec{r}-\omega t)}]$, where

$$\rho_1(\vec{k}, \omega) = \frac{E_1 E_2}{4\pi}\frac{\frac{nc\alpha\beta\rho_0 c_s^2 k^2}{\gamma} + (\frac{n^2-1}{2})(\frac{n^2+2}{3})k^2(i\rho_0 C_v\omega - \lambda k^2)}{(-\omega^2 + \frac{c_s^2}{\gamma}k^2 - \frac{\eta}{\rho_0}k^2 i\omega)(i\rho_0 C_v\omega - \lambda k^2) + i\frac{(\gamma-1)}{\gamma}\rho_0 C_v c_s^2 k^2\omega}. \tag{6}$$

This expression gives us the theoretical basis for complex Brillouin spectra. In the linear-response regime, the above complex resonance spectra of phonons under forced oscillations are equivalent to Brillouin spectra of thermal phonons. Equation (6) tells us that the phase of density oscillations is different between electrostriction and thermal expansion effects [see the numerator on the right-hand side of Eq. (6)]. This fact can be used to obtain physical information on the mechanisms of phonon generation, in addition to information on phonon dynamics.

Figure 1(a) indicates a block diagram of our system. The details of the optical configuration are schematically shown in Fig. 1(b). We used two lasers (Model 140-0532-100 and 140-0532-10, Lightwave Electronics) that are single axial mode, continuous wave output, frequency doubled (green) Nd-YAG (neodymium yttrium aluminum garnet) lasers. As previously described [3,4], the tunable ranges of the two lasers have an overlap. The beat frequency between the two lasers can be controlled to be any value between 0 and 10 GHz, and, further, be extended over 60 GHz with several mode hoppings. Thus, we can produce coherent phonons having a desired frequency in a sample, according to the principle described above.

Next we describe how to measure the real and imaginary parts of $\rho_1(\vec{k}, \omega)$ separately, instead of the power spectrum $|\rho(\vec{k}, \omega)|^2$. We develop a new *phase-sensitive optical heterodyne method* to measure both the real and imaginary components of coherent density oscillations described by Eq. (6). The probe light is scattered by coherent phonons produced by the laser beams $L1$ and $L2$. A component of the scattered electrical field with polarization $\vec{n}_l$ at the detector position $\vec{R}$, $E_s(\vec{R}, t)$, is given by [1]

$$E_s(\vec{R}, t) = -\frac{k^2 E_p \vec{n}_p \cdot \vec{n}_l}{4\pi\epsilon_0 R} e^{i[\vec{k}\cdot\vec{R}-(\omega_1+2\pi f)t]}\frac{\partial\epsilon}{\partial\rho} \times \rho_1(k, \omega)e^{-i\omega t}, \tag{7}$$

where $\vec{n}_p$ and $\vec{n}_l$ are the polarization unit vectors of the probe and local light, respectively. We define E_p and E_l, respectively, as the electric field strength of the probe and local light. Since the total electrical field on a photodiode (PD2) is given by the summation of the local and scattered light as $\vec{E}(\vec{R}, t) = \vec{n}_l[E_l e^{-i\omega_2 t} + E_s(\vec{R}, t)]$, we obtain the output current of the photodiode i_{out} as

$$i_{\mathrm{out}}(t) = QEE^* = \mathrm{dc} + 2Q\left[\frac{k^2}{8\pi\epsilon_0 R}\frac{\partial\epsilon}{\partial\rho}(\vec{n}_p\cdot\vec{n}_l)E_l E_p\{\mathrm{Re}[\rho_1(k,\omega)]\cos(2\pi f t) - \mathrm{Im}[\rho_1(k,\omega)]\sin(2\pi f t)\}\right], \tag{8}$$

where ϵ_0 is the dielectric constant of vacuum and Q is the quantum efficiency. Thus, the lock-in detection of i_{out} with a reference signal of $\cos(2\pi f t)$ gives us $\mathrm{Re}[\rho_1(k, \omega)]$ and $\mathrm{Im}[\rho_1(k, \omega)]$, respectively, as the in-phase and out-of-phase components of the signal. In conventional phase-insensitive heterodyne methods [3,9–11], on the other hand, only the power spectrum $|\rho_1(\vec{k}, \omega)|^2$ has a physical meaning. There the square-law detected signal through the bandpass filter is measured as a function of frequency; a spectrum analyzer has often been used for this purpose.

Here we explain how the above principle is realized experimentally. The light output of the master laser of frequency $\nu_1 (= \omega_1/2\pi)$ (100 mW) is transformed into two collinear orthogonally (vertically and horizontally)

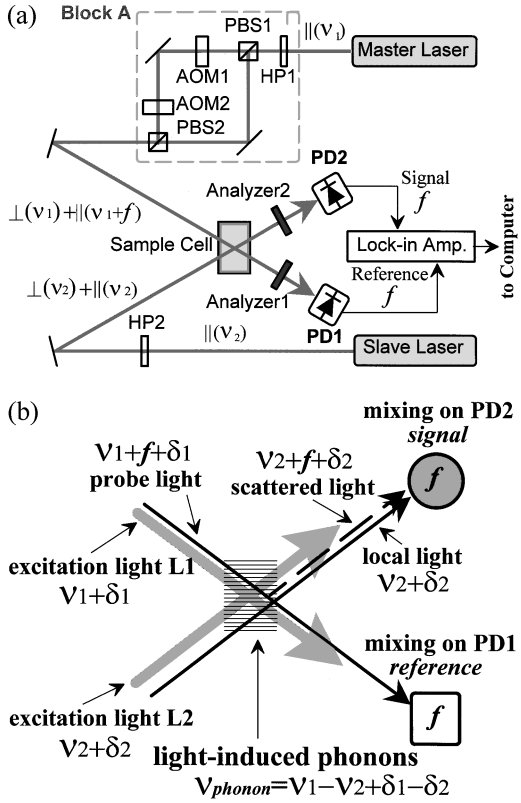

FIG. 1. (a) A block diagram of our experimental system. PBS stands for polarizing beam splitters, AOM for acousto-optic modulators, and HP for half-wave plates. $\|$ shows light with horizontal polarization, while $\perp$ shows light with vertical polarization. A unit for frequency control of the two lasers is not indicated in this figure (see Ref. [3] for details). (b) A schematic figure explaining the extremely high stability of both frequencies and phases of the beat signals for lock-in detection.

linearly polarized beams with a frequency difference of f [$\perp(\nu_1)+\|(\nu_1+f)$] (see Fig. 1). We set f to 2 MHz by using two acousto-optic modulators, whose modulation frequencies are 80 and 82 MHz, to satisfy the following two competing conditions: f needs to be high enough to avoid the harmful $1/f$-like amplitude noises below 1 MHz of the laser light itself and low enough for a direct lock-in detection. On the other hand, the light output of the slave laser of frequency $\nu_2 = (\omega_2/2\pi)$ (10 mW) is transformed by the half-wave plate into two collinear orthogonally (vertically and horizontally) linearly polarized beams with the same frequency [$\perp(\nu_2)+\|(\nu_2)$] (see Fig. 1). The two beams $L1$ [$\perp(\nu_1)$] and $L2$ [$\perp(\nu_2)$], intersected in a sample cell, produce a scanning interference pattern, which generates sound waves in the sample. The light component of $\|(\nu_2)$ is used as a local beam for the heterodyne detection, and that of $\perp(\nu_2)$ is cut by a polarizer before a photodetector (PD2). The probe light of $\|(\nu_1+f)$ is diffracted by excited phonons of a frequency of $(\nu_2-\nu_1)$. This diffracted light [$\|(\nu_2+f)$] is mixed with the above local light [$\|(\nu_2)$] on a p-i-n photodiode (PD2) with a low-noise, transimpedance amplifier (Hamamatsu, S2858). The output signal of PD2 that includes the beat signal of frequency f is fed directly into a two-phase lock-in amplifier. As a reference signal for the lock-in detection we make a sinusoidal wave of frequency f by mixing the excitation light $L1$ [$\perp(\nu_1)$] and the probe light [$\|(\nu_1+f)$] on another photodiode (PD1) after changing their polarization directions by a polarizer to produce the beat signal of frequency f. Thus, the in-phase and out-of-phase components of the lock-in detected signal give us, respectively, the real and imaginary parts of complex Brillouin spectra given by Eq. (8).

Next, we describe a technical, but crucially important point of our method. In the above discussion, we neglect the possible phase and frequency noises of lasers. These noises usually lower the SNR because they lead to the increase in the detection bandwidth. In reality, however, *our method is free from the effects of these noises of lasers* on SNR; namely, the linewidth of the lasers ($\sim$150 kHz in our lasers) does not lower SNR, although it limits the frequency resolution of the measurement ($\sim$300 kHz). This can be explained as follows [see Fig. 1(b)]: Replacement of ν_i by $\nu_i + \delta_i$ [δ_i: laser noises of laser i ($i = 1, 2$)] does not affect the final beat frequencies of both signal and reference for lock-in detection, which are exactly equal to f without noise effects. *This can be realized because we use the same laser sources for both phonon generation and phonon detection.* The signal is almost a δ function centered at f, and it is detected, including the phase, by a narrow-band detector that has a window of δ function centered at f. This fact makes it possible to almost realize the theoretical limit of SNR, as described below.

The SNR of the heterodyne detection of light using a bandpass filter, including the improvement by time average, is generally given by

$$\mathrm{SNR} = 2QP_s/(\hbar\omega\sqrt{B/\tau}), \tag{9}$$

where P_s is the power of the detected light signal, $\hbar\omega$ is the energy of one photon, B is the bandwidth of heterodyne detection, and τ is a period of time average. It is apparent from Eq. (9) that the smaller the B, the greater the SNR. For a signal light that is not monochromatic, as in the case of the light diffracted by *incoherent* thermal phonons, however, P_s becomes smaller in proportion to B with a decrease in B when B is smaller than the spectral width of the light. Since the denominator of the right-hand side of Eq. (9) becomes smaller in proportion to only $\sqrt{B}$, the smaller B leads to the smaller SNR under such conditions. For this reason, B is usually set to be more than 1 MHz in conventional heterodyne methods [3,10,11] to satisfy the two opposing requirements, high frequency resolution and high SNR. In our new method, on the other hand, the signal intensity does not decrease by decreasing B since the optically generated phonons are always coherent to the laser light waves, and all the optical noises can be canceled out in our optical configuration as described before. Thus, an ultimately high SNR can be realized by the direct phase-sensitive lock-in detection of the heterodyne signal in our method.

We demonstrate in Figs. 2(a) and 2(b), respectively, the complex Brillouin spectra of poly(methyl methacrylate) (PMMA) in a glassy solid state and those of carbon disulfide (CS_2) in a liquid state. The typical behavior of complex resonance spectra of forced oscillators can be clearly seen. It should be noted that the spectra having good SNR, such as in these figures, can be obtained within a few minutes. Judging from the phase obtained by fitting Eq. (8) to the spectra, coherent phonons are produced dominantly by an electrostriction effect. The above result clearly indicates that we can measure *complex Brillouin spectra* with very high frequency resolution ($\leq$1 MHz) that is now limited only by the wave-number uncertainty coming from a finite beam-size effect (beam divergence).

The complex spectra do not provide us with physically new information on phonon dynamics compared to the power spectrum in usual cases, except for information on the phonon generation mechanisms, namely, whether phonons are generated by electrostriction effects or by thermal expansion due to light absorption. As a result of the phase-coherent measurement, however, both ultimate SNR and frequency resolution are realized. Further, the fact that we have both real and imaginary parts of spectra increases the accuracy of the determination of the peak phonon frequency, linewidth, and spectra shape. The most important advantage over other methods is that our measurement is not affected by incoherent noises such as slowly fluctuating concentration modes, since our method can pick up only the coherent signal to the excitation: For example, we can make the phonon measurement free from the intense Rayleigh scattering originating from incoherent fluctuations of order parameter near a critical point, which usually makes the accurate phonon measurement difficult near the critical point.

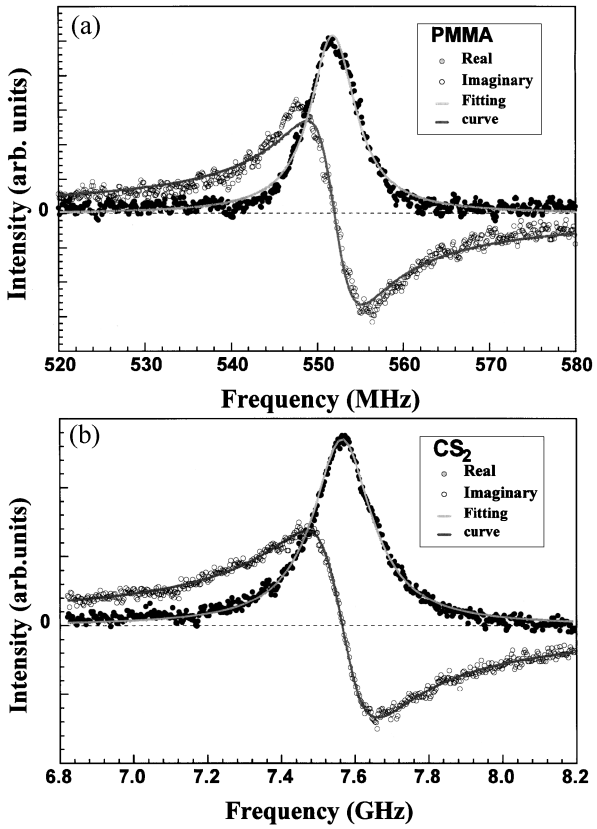

FIG. 2. Complex (anti-Stokes) Brillouin spectra of PMMA at $T = 300$ K and $k/2\pi = 0.197\ \mu\mathrm{m}^{-1}$ (a), and CS_2 at $T = 300$ K at $k/2\pi = 0.197\ \mu\mathrm{m}^{-1}$ (b). Lines are theoretical curves [see Eq. (8)] fitted to the spectra.

Finally, we note the following points: (i) Since the signal intensity is proportional to $E_1E_2E_lE_p$ [see Eqs. (6) and (8)] in our method, the increase in the laser power straightforwardly leads to a significant improvement of SNR. The use of two lasers with a power of 200 mW that are now commercially available, for example, increases SNR by a factor of $\sim$100 compared to our present system. (ii) The speed of photodetectors and electric circuits never limits the upper frequency bound of our method. Our method covers the frequency range from at least 20 MHz to 10 GHz with a frequency resolution of 1 MHz.

In conclusion, we have succeeded in observing *complex Brillouin spectra* by the phase-sensitive detection of coherent phonons using light scattering. A light scattering method can detect the phase of phonons, in addition to the amplitude, simply because the scattered light keeps the phase information of phonons in the scattering process. Our method has both an ultimate SNR and an extremely high frequency resolution, which is a factor of $\sim$100 better than that of a Fabry-Perot interferometer. We hope that COBS will be widely applied for studying the dynamics of acoustic phonons in various condensed matter, superseding a Fabry-Perot interferometer.

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