New Method of Superheterodyne Light Beating Spectroscopy for Brillouin Scattering Using Frequency-Tunable Lasers

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(Received 12 October 1994)

We have developed a new optical superheterodyne method for light scattering using frequency-tunable lasers and succeeded in measuring Brillouin spectra of simple liquids in a wide frequency range, 10 MHz to 3 GHz. Its frequency resolution (~300 kHz) is determined by the short-time coherency between the two lasers. A great advantage of our method over conventional optical beating methods is that the speed of the photodetector and electric circuits never limits the upper frequency bound. This is realized by the continuous tunability of the laser frequency. The method has a high potential for studying the dynamics in a wide class of condensed matters.

PACS numbers: 62.60.+v, 07.60.–j, 33.20.Fb, 78.35.+c

Dynamic light scattering is one of the most powerful experimental means to study the dynamics of condensed matter, including liquids, solids, polymers, and mesophases, without causing any perturbation to the sample [1]. The frequency spectrum of scattered light from a simple liquid typically consists of one Lorentzian unshifted line around the excitation frequency ω_i (Rayleigh line) and of two nearly Lorentzian wings centered around the two frequencies $\omega_i \pm cq$ (Brillouin doublet), where q is the wave number of the phonon satisfying the momentum conservation and c is the phonon velocity. The former comes from entropy fluctuations associated with the translational and rotational degrees of freedom, while the latter comes from the propagation of sound waves (pressure fluctuations) in the scattering medium.

Experimental methods to obtain a spectrum of scattered light could be classified into the following two types: (i) optical filter methods [2] and (ii) optical mixing or beating methods [3,4]. Filter methods are used to study relatively rapid dynamic processes ($\tau_c < 1$ ns, τ_c: the characteristic time). A typical filter method for Brillouin scattering is the spectral decomposition of scattered light by a Fabry-Pérot interferometer [2]. An optical mixing method [3,4] which is an optical analog of the beating techniques developed in radio-frequency spectroscopy, on the other hand, has been used for slower processes ($\tau_c <$ 1 ns) [5–8]. There have so far been many efforts to improve the frequency resolution of Brillouin scattering measurements based on both of the above methods, as discussed below.

A Fabry-Pérot interferometer has been used most commonly for studying Brillouin scattering since the first observations of Brillouin doublets [2] by this technique. This method is suitable to study a thermal phonon whose frequency is more than several GHz. For studying the phonon of a lower frequency, however, the resolution is sometimes not enough. This is an intrinsic difficulty of the optical filter method: The optical signal has a carrier frequency of $\sim 10^{14}$. To get a resolution of 1 MHz, for example, a relative resolution of 10^{-8} is required. This is usually beyond the ability of the filter: Such a high Q value is difficult to reach without greatly sacrificing sensitivity using any techniques including multipath interferometers [9]. The scanning linearity and the determination of the absolute frequency are also serious problems of the Fabry-Pérot interferometer [10].

The optical heterodyne method generally has a much higher frequency resolution than the optical filter method. The reason is as follows: The beating signal produced by optical mixing has a carrier frequency equal to the difference between the excitation and local light. Thus much less relative accuracy is required for the optical beating technique to obtain a certain frequency resolution than for the optical filter method. The application of the optical beating method to Brillouin scattering was first proposed and tried by Lastovka and Benedek [5]. Eden and Swinney [7] are the first to have succeeded in observing the Brillouin doublet as a spectrum by this method, although the frequency range was limited below 100 MHz. Very recently the frequency range has successfully been expanded up to 1 GHz by Matsuoka, Sakai, and Takagi [8] using a fast avalanche-type photodiode as a detector. In all these conventional heterodyne techniques only one common laser source has been used as both the excitation and local lights: This is due to the requirement for the coherency between the excitation and local light. In this case, we need to detect a beating signal around a phonon frequency to obtain a Brillouin spectrum. To study a phonon of 1 GHz, for example, we need a high-speed detector with bandwidth more than 1 GHz. Generally such detectors have lower sensitivity: this fact limits the frequency range of the conventional optical beating method. Namely, the upper frequency bound is limited by the speed of the photodetector and electric circuitry.

In this Letter, we demonstrate a new method of light beating spectroscopy using frequency-tunable lasers, and apply it to Brillouin scattering measurements of simple liquids. Recent advances in the laser technology enable us to apply two independent lasers to the light-beating

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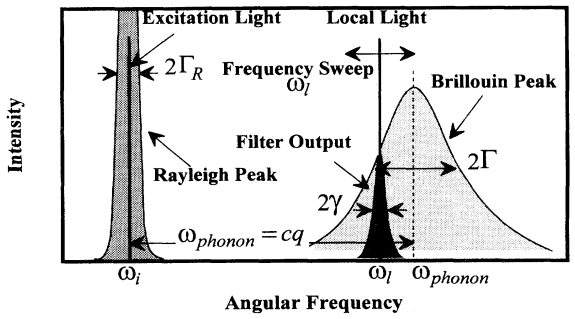

FIG. 1. Schematic figure explaining the basic principle of a new superheterodyne beating spectroscopy. Γ_R is the Rayleigh linewidth. For simplicity, ω_0 is here set to 0.

spectroscopy because of the high coherency between their phases.

Figure 1 schematically shows the principle of the method. Scattered light is optically mixed with a local light on a photodiode and the output current is fed into an electrical bandpass filter whose center frequency and bandwidth are ω_0 and γ, respectively. The output of the filter is square-law detected. This process can be expressed mathematically as follows: The electric field on the photodiode $\epsilon(t)$ is given by

$$\epsilon(t) = \epsilon_i f(t) e^{-i[\omega_i t + \phi_s(t)]} + \epsilon_l e^{i[\omega_l t + \phi_l(t)]},$$

where ϵ_i and ϵ_l are the electric fields of the incident and local lights, respectively, and ω_i and ω_l are the frequencies of the incident and local lights, respectively, and ϕ_i and ϕ_l are the phases of the incident and local light, respectively. $f(t)$ is the modulation produced by a scattering process. The output current from the detector $I(t)$ is thus given by

$$I(t) = \epsilon_l^2 + \epsilon_i^2 f(t)^2 + 2\epsilon_i \epsilon_l f(t) \cos[\Delta\omega t + \Delta\phi(t)],$$

where $\Delta\omega = \omega_i - \omega_l$ and $\Delta\phi = \phi_i - \phi_l$. Then the output of the bandpass filter can be expressed as

$$I_f(t) = \gamma e^{i\omega_0 t} \epsilon_i \epsilon_l \int_{t-1/\gamma}^{t} f(u) \times \cos[\Delta\omega u + \Delta\phi(u)] e^{-i\omega_0 u}\, du.$$

The power spectrum $F(\omega)$ of $f(t)$, which is proportional to the dynamic structure factor $S(q, \omega)$, is

$$F(\omega) = \lim_{\gamma \to 0} \gamma \left| \int_{t-1/\gamma}^{t} f(t) e^{-i\omega u} du \right|^2.$$

For a small γ, thus,

$$\int_{t-1/\gamma}^{t} f(t) e^{-i\omega u}\, du \sim \frac{1}{\sqrt{\gamma}} \sqrt{F(\omega)}\, e^{i\phi_\omega t},$$

where $\phi_\omega(t)$ is the phase of the ω component of $f(t)$. Using these relations and assuming that $\Delta\phi(t)$ is almost constant during a time of $1/\gamma$, the square-law detected signal $|I_f|^2$ is

$$|I_f|^2 = \frac{\gamma \epsilon_i^2 \epsilon_l^2}{4} \{F(\Delta\omega - \omega_0) + F(\Delta\omega + \omega_0) + \cos[\Delta\Phi(t)] \sqrt{F(\Delta\omega - \omega_0) F(\Delta\omega + \omega_0)}\},$$

where $\Delta\Phi(t) = 2\Delta\phi(t) + \phi_{\Delta\omega+\omega_0}(t) - \phi_{\Delta\omega-\omega_0}(t)$. If we average this output for a time interval much longer than $1/\gamma$, the last term including the phase difference disappears and thus we get the final relation

$$|I_f|^2 = \frac{\gamma \epsilon_i^2 \epsilon_l^2}{4} [F(\Delta\omega - \omega_0) + F(\Delta\omega + \omega_0)]. \quad (1)$$

The details of the experimental configuration and the signal processing are shown in the block diagram of Fig. 2. We used two lasers (Model 140-0532-100 & -10, Lightwave Electronics) which are single axial mode, continuous wave output, frequency doubled (green) Nd:YAG (neodymium yttrium aluminum garnet) lasers. They have monolithic traveling ring resonators, which ensure stable, single frequency operation over a range of operating conditions. The wavelength is 532 nm, the linewidth is ~10 kHz/ms, and the frequency jitter is less than 150 kHz/s. The frequency is electrically tunable over 10 GHz without any mode hopping. The two lasers have been specially designed to have a frequency overlap; thus we can observe the beat signal between them with a spectrum analyzer.

First, we describe how to control the frequency difference between the two lasers (see Block A in Fig. 2). Light from the master laser with an output power of 100 mW was used as the incident light inducing the scattering of a sample, while light from the slave laser with an output power of 10 mW was used as the reference (local) light for the optical heterodyne detection. A portion of the former was mixed collinearly with a portion of the latter on a wide-band photodetector (Hamamatsu S4753), and the frequency difference between them ($\Delta\omega$) was then monitored with a spectrum analyzer (Advantest R3361B) whose frequency range was from 9 kHz to 3.6 GHz. The $\Delta\omega$ was controlled by a feedback loop using a computer within ± 150 kHz, and scanned linearly.

The optical configuration used is very convenient for measuring the $\vec{q}$ dependence of the dynamic structure factor $S(\vec{q}, \omega)$. In the conventional optical beating techniques, we had to readjust the beam configuration every

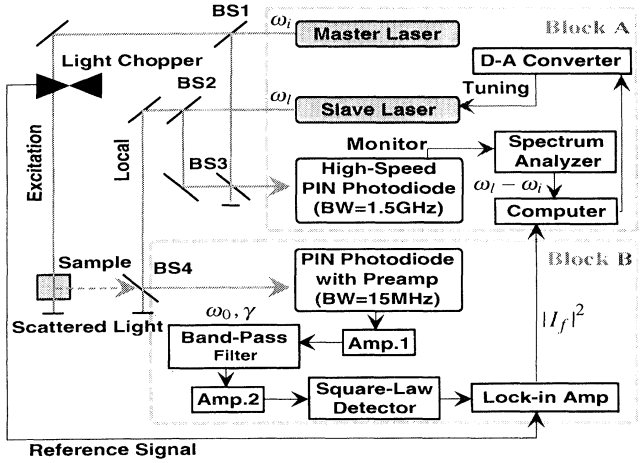

FIG. 2. Block diagram of the system. BS and BW stand for beam splitter and bandwidth, respectively.

time after changing the scattering vector to get the maximum efficiency of the optical beating. This readjustment process usually takes a long time. It further leads to the loss of the information on the scattering intensity, which is extremely sensitive to the geometrical configuration of the beams for the optical heterodyne. We solve this problem by the following method. The light of the master laser was introduced exactly on the rotation axis (z axis) of the scattering cell, namely, of the rotating table. Then the beam direction was changed from the z axis to the radial direction by the mirrors fixed in the rotating table, so that the incident beam always intersects the local beam which is fixed in the laboratory system on the horizontal plane. This special configuration frees us from the readjustment of the beam configuration after rotating the table, or changing the scattering vector $\vec{q}$. However, the change in the scattering angle leads to the change in the polarization direction of the incident light upon reflection at the first mirror fixed in the rotating table; this affects the beat efficiency. To avoid this effect, the polarization of the incident light was transformed from the linear polarization fixed in the laboratory system into the linear one fixed in the rotating coordinate system by using two quarter-wave plates before the light reached the first mirror fixed on the rotating table. This provides us with a constant beat efficiency, irrespective of the setting of the scattering angle.

The wave vector $\vec{q}$ of the scattered light is selected by the local light whose path is fixed in the laboratory system; this selection is due to the requirement for the matching of the wave vectors of the two lights to produce the beat signal. The selected range of $\vec{q}$ was typically $1/20°$. The resolution of $\vec{q}$ is also limited by the diffraction effect, which causes a serious error in $\vec{q}$ at low frequencies below 10 MHz for our beam sizes. The scattering angle was determined by a computer-controlled rotating table with an angular resolution of 0.12" (Phys. Instrum. M-038). The intensity of the local light was controlled to attain the condition that the shot noise well exceeds the thermal noise in the detector output. The scattered light and the local light were mixed on the photodiode (Hamamatsu S2858) with a low-noise amplifier whose cutoff frequency was 15 MHz.

Next, we describe the signal processing (see Block B in Fig. 2). The output signal of the detector was amplified by 40 dB, and then fed into the bandpass filter whose center frequency ω_0 was located at ~5 MHz and whose bandwidth γ can be changed from 150 kHz to 5 MHz. The center frequency was chosen to avoid the $1/f$ noise of the lasers below 1 MHz and also to satisfy $\omega_0 > \gamma$. The output of the filter was further amplified by 34 dB and then square-law detected by a diode bridge. Since the beating signal was overwhelmed by the large shot noise produced by the local light (ϵ_l^2), we made lock-in detection of the signal [5,8] by modulating the incident light (ϵ_l^2) with a light chopper. The modulation frequency was typically set to ~100 Hz. By this superheterodyne method, we can measure Rayleigh scattering, the Stokes and anti-Stokes components of Brillouin scattering, and any other components such as a Mountain component [11], shear modes, and second-sound modes, with a frequency resolution of ~300 kHz. This resolution comes from the effective short-term linewidth of the lasers. The accuracy of the frequency axis in our method is determined by that of the spectrum analyzer which is very reliable. The frequency range is now limited below 3.6 GHz solely by the upper limit of the spectrum analyzer detecting the frequency difference between the two lasers.

The most striking feature of our method is that the signal detection can always be performed at a fixed frequency ω_0 (~5 MHz), irrespective of how fast the dynamics contributing to the scattering are (phonon frequency). Thus we can use a rather slow detector a Si p-i-n photodiode with a low-noise current amplifier even when measuring phonons with a frequency more than a few GHz. Such a detection provides us with the ideal situation where the noise is dominated by the shot noise, and further with a high quantum efficiency; thus it is the best photodetector for optical heterodyne detection. This is a great advantage of our new method over the conventional methods, which require a very high speed detector. In our case, there is, in principle, no limit for the upper frequency bound coming from photodetectors and electrical circuits. Only the S/N ratio limits the frequency range of our method. To improve the S/N ratio, we can use a large value of γ at the expense of the resolution [see Eq. (1)]; the frequency resolution is usually not required for a very high frequency range, since the halfwidth of a Brillouin peak (the decay rate of phonon Γ) is proportional to q^2. The smaller spectral density at higher frequencies, which is believed to be a difficulty of the optical beating spectroscopy at high frequencies [6], can thus be supplemented by the increase in γ in our method.

Figure 3 shows typical Brillouin spectra observed for liquid carbon disulfide (CS_2). The dotted curve is a fitted theoretical line, which is nearly Lorentzian but includes the asymmetric components [see Eq. (39) in Ref. [12]]. Because of the narrow instrumental linewidth, the spectrum

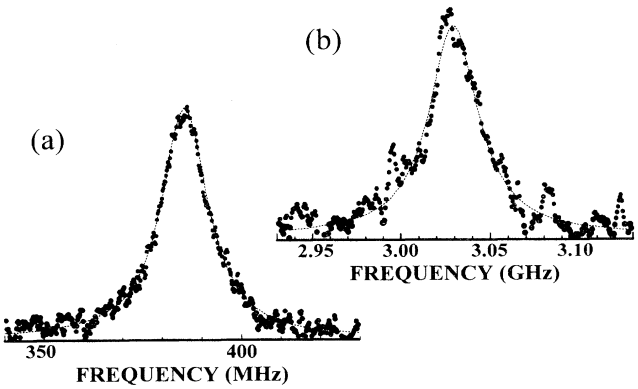

FIG. 3. Typical Brillouin spectra of CS_2 at 20 °C around 400 MHz (a) and 3 GHz (b).

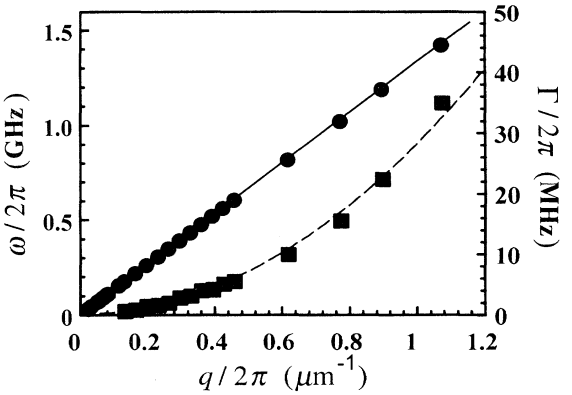

FIG. 4. The q dependence of the phonon frequency ω (filled circle) and the decay rate Γ (filled square) for toluene at 20 °C. Solid line: $\omega = cq$ with $c = 1325$ m; dashed line: $\Gamma = (c/2\pi)^3(\alpha/f^2)q^2$ with $\alpha/f^2 = 80 \times 10^{-17}$ s^2/cm.

almost has an intrinsic shape and thus looks quite different from that measured by the Fabry-Pérot interferometer, which is usually very broad and further overlapped with the tail of the central component due to poor resolution (see, e.g., Ref. [13]). Figure 4 demonstrates the dispersion relation of phonons in toluene, which is the typical behavior of simple liquids. Figure 5 indicates the frequency dependence of the sound velocity c and absorption α of CS_2, which are obtained by curve fitting the Brillouin spectra. To obtain reliable data, α was estimated only when Γ is more than 4 times larger than the instrumental linewidth due to the beam divergence (~1 MHz). The accuracy of the velocity measurement is about ±0.5%. We can clearly see the relaxation phenomena due to the coupling between the translational and internal vibrational degrees of freedom of CS_2 [14]. These results clearly demonstrate the high accuracy of our method and the wide range of the applicability.

In summary, we have developed a new method of superheterodyne light beating spectroscopy of Rayleigh-Brillouin scattering, which at present covers the frequency

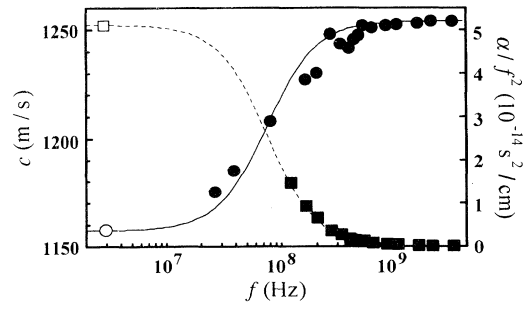

FIG. 5. Frequency (f) dependence of c (filled circle) and α/f^2 (filled square) obtained by Brillouin scattering measurements of CS_2 at 20 °C. Lines indicate theoretical curves of single relaxation with a relaxational frequency of 80 MHz. Open symbols are data measured by the pulse-echo method [14].

range below 3.6 GHz. We call this new optical-frequency-sweep light scattering spectroscopy "superheterodyne Brillouin spectroscopy (SUBS)." This method is intrinsically simple, and no assumptions are necessary for data analysis. Thus, there could be no spurious effects as in the filter methods. This is another advantage over the conventional methods, especially in studying the Mountain mode [11] coming from relaxational effects of fluids and any new mode of a system whose line shape is unknown. The frequency resolution could further be dramatically increased by phase locking the two lasers with a fast feedback loop. The simplest way to improve the S/N ratio further and go to a higher frequency, on the other hand, is to increase the laser power and/or the bandwidth of the filter. A narrow line, single-mode laser (532 nm) with a power of 400 mW is now commercially available. Thus we expect that the frequency could be easily increased up to 10 GHz, at least for samples having large Brillouin scattering cross sections. The technique itself can be applied for any condensed matters including solid samples.

We are grateful to K. Takagi, K. Sakai, and K. Hattori for valuable information through daily conversations and their kind support. We also thank A. J. Lovinger for helpful comments on the manuscript.

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