$1/f$ Phonon-Number Fluctuations in Quartz Observed by Laser Light Scattering

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The energy of each phonon mode is fluctuating around its equilibrium value, and its power spectral density was derived with a quartz specimen which was illuminated by a laser light. Fluctuations of the Brillouin scattered light intensity were measured by means of the photon-counting technique. The power spectral density has been derived to be $8\times10^{-5}/f$ per phonon mode provided that phonon-number fluctuations are independent from mode to mode.

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It is likely that the $1/f$ conductivity fluctuation is an equilibrium phenomenon. This conjecture was directly proved by Clarke and Voss[1] through their observation that fluctuations of the thermal noise power of the electric conductor had a $1/f$ spectrum. One of the origins of conductivity fluctuations is attributed to phonon scattering of charge particles as suggested by Vaes and Kleinpenning[2,3] who investigated Hall voltage fluctuations with and without magnetic induction. Furthermore, Hooge and Vandamme[4] showed the role of lattice scattering in generating $1/f$ mobility fluctuations by using semiconductor specimens with various impurity doping levels. The contribution of phonons was also suggested by Mihaila[5,6] and by Planat and Gagnepain.[7] Musha, Nakajima, and Akabane found that the imaginary part of the dielectric constant of triglycine sulfate is subject to $1/f$ fluctuations near its Curie temperature; this observation also suggests a phonon contribution to $1/f$ fluctuations.[8] In the present Letter we will show direct evidence by means of a laser-light-scattering experiment that fluctuations of the phonon numbers in thermal equilibrium have a $1/f$ power spectral density (PSD).

When laser light is passed through a transparent specimen, a small part of it is scattered in all directions by thermal fluctuations in the dielectric constant of the material. Fluctuations of the dielectric constant are caused by lattice vibrations, namely, "phonons" belonging to various modes; hence the following momentum conservation is satisfied as $\mathbf{k}_i=\mathbf{k}_s\pm\mathbf{q}$, where $\mathbf{k}_i$ and $\mathbf{k}_s$ are wave vectors of the incident and scattered light, $\mathbf{q}$ is the phonon wave vector involved in the scattering, and the double sign refers to emission and absorption of a phonon. The energy conservation requires the relation $f_p=2f_i(u/c)v\sin(\theta/2)$, where f_p is the phonon frequency, f_i is the frequency of the incident laser light which is very close to the frequency of the scattered light, v is the refractive index of the specimen, u is the acoustic velocity, c is the speed of light in free space, and θ is the scattering angle which is nearly equal to 90° in the present experiment. The light-scattering cross section is proportional to the phonon number of that particular mode because the photon flux is proportional to the electric field squared, the electric field generated by lattice vibrations is proportional to the polarization, and because the square of the polarization is proportional to the phonon flux. Therefore, the scattered laser power is proportional to the phonon number, and fluctuations of the phonon number give rise to fluctuations of the scattered light intensity. As a transparent specimen, a synthetic crystalline quartz was used.

The Brillouin scattering spectrum of crystalline quartz was first observed by Gross[9] in 1930. Theoretically the spectrum should have three doublets corresponding to emission and absorption of a single longitudinal and two transverse acoustic-phonon modes. The three doublets were first observed with quartz by Shapiro, Gammon, and Cummins[10] using a 50-mW He-Ne laser at a wavelength of 632 nm with overall finesse of 30. In addition, nonpropagating fluctuations produce scattering whose central frequency is equal to that of the incident one; this is called the Rayleigh component, which was very weak as compared with the Brillouin components.[10]

The light source in our experiment was an Ar-ion laser (Lexel model 95-4) which has a wavelength of 514.5 nm, a longitudinal-mode separation of 150 MHz, a frequency spread of a single longitudinal mode of 3 MHz, and a fractional power fluctuation in a single mode of less than 0.2%/h. The experimental setup is illustrated in Fig. 1. A laser beam with 100 mW and vertical polarization was passed through an ND filter (attenuator) which was placed at an angle to the laser beam to avoid reflection back into the laser, and was incident on the quartz specimen of $12\times10\times10$ mm^3 with the z axis vertical. A slit of 0.1 mm in width and 2 mm in height was placed parallel to the side face of the specimen and 1.5 mm apart from the laser beam. Two photomultipliers (Hamamatsu Photonics C716-02) collected two scattered light beams making angles $90°\pm8°$ with the incident laser beam. A lens of focal length 10 cm made an image of the slit on the active surface of the photomultiplier. The dark count rate was about 100 counts/s, which was more than 3 orders of magnitude smaller than the count rate of the received light. Moreover, it was confirmed that dark counts obeyed Poissonian statistics

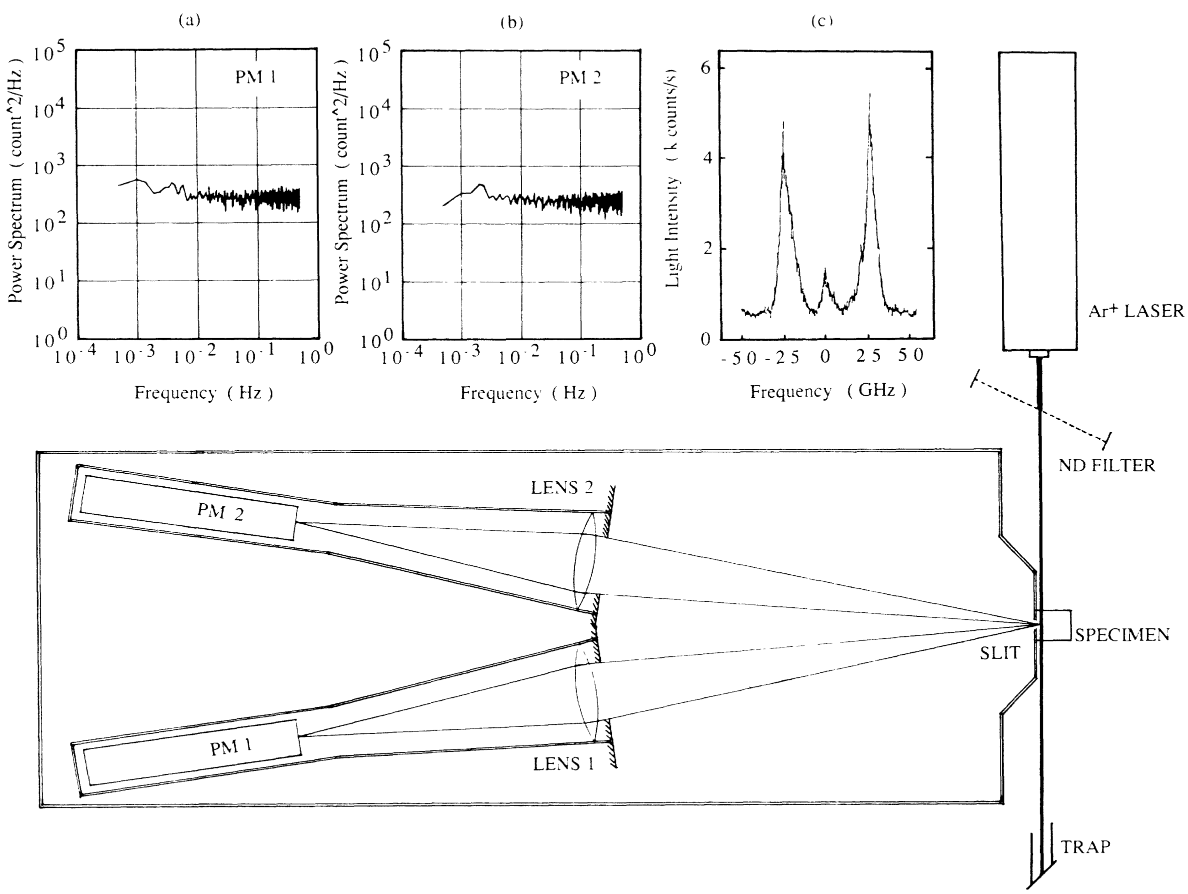

FIG. 1. Experimental setup. Insets (a),(b): PSD of dark counts of photomultipliers 1 and 2. Inset (c): An optical PSD of scattered light.

and that the PSD was white, as shown in insets (a) and (b) of Fig. 1. The PSD of the scattered light was measured with an etalon, and two Brillouin components were observed about 25 GHz above and below the Rayleigh component; this spectrum is also illustrated in inset (c) of Fig. 1. As the finesse of our etalon was 10, three peaks corresponding to a single longitudinal and two transverse-acoustic modes were not separated in each of the Brillouin components. To estimate the PSD of light-intensity fluctuations down to low enough frequencies, a continuous observation as long as 20 to 40 h was required. However, it was difficult to keep a constant resonance frequency of the etalon during such a long period, hence the etalon was not used. Therefore, the Rayleigh component was also collected together with the Brillouin components. From inset (c) of Fig. 1 for the Brillouin spectrum, the intensity of the Rayleigh component is $\frac{1}{12}$ times that of the Brillouin components, and its contribution was neglected. Since the two photomultipliers collected scattered light beams with different scattering angles, outputs of the two photomultipliers refer to different phonon modes. If phonon-number fluctuations are not correlated from mode to mode, these outputs would carry uncorrelated fluctuations. However, since the laser beam was not very stable in intensity, these outputs were modulated by common fluctuations of the incident light intensity. Fluctuations of the incident laser intensity contained in these outputs were removed by taking a ratio between the two output counts.

The incident light intensities to the two photomultipliers I_1 and I_2 are given by

$$I_1 = Af(t)[1+g_1(t)]\,, \tag{1}$$

$$I_2 = Bf(t)[1+g_2(t)]\,, \tag{2}$$

where A and B are numerical factors representing geometrical asymmetry of the optical system, $f(t)$ is the incident-laser-light intensity which is slightly fluctuating, and g_1 and g_2 are fractional fluctuations of the phonon numbers related to the scattering. The photon count is stochastic and the ratio of phonon counts obtained by the two photomultipliers is given by

$$\frac{A(f+df_1)(1+g_1)}{B(f+df_2)(1+g_2)} \approx \frac{(1+df_1/f)(1+g_1)}{(1+df_2/f)(1+g_2)} \approx 1+df_1/f-df_2/f+g_1-g_2\,, \tag{3}$$

where df_1 and df_2 are photon shot noise, f is the mean photon count rate, and we assume $A \approx B$ and $g_{1,2} \ll 1$. As terms $df_{1,2}/f$ refer to fractional photon shot noise, their spectral level is equal to a reciprocal mean photon count rate. There are two such terms in Eq. (3), and the observed PSD of I_1/I_2 contains twice the fractional photon shot noise. For the same reason the last two terms give twice the PSD of fractional phonon-number fluctuations. The laser-beam position as well as intensity was slightly fluctuating and the detected light intensity made a complicated variation in time; after taking the ratio this variation was completely eliminated.

Figure 2 shows an example of the PSD of Eq. (3) with quartz specimen 1. The total observation time is 20 h and the sampling time was 1 and 3 s; as the fast Fourier transform covered 2048 sample data, the PSD was estimated after averaging squared Fourier transforms over 35 and 10 segments, respectively. The PSD at low frequencies is well approximated by a $1/f$ curve, and it merges into photon shot noise at high frequencies; the whole PSD becomes a superposition of a shot-noise PSD and a $1/f$ spectrum. The mean photon count rates of the two photomultipliers were 8.5×10^5 and 8.9×10^5 counts/s, hence the expected fractional-photon shot-noise level is $1/8.5\times10^5+1/8.9\times10^5=2.3\times10^{-6}$, which is in good agreement with the observed result.

The specimen was replaced by a dummy scatterer which was a piece of white chalk with a cavity illuminated by laser light, and the two photomultipliers collected diffused light in the cavity through a slit. The PSD of the output ratio is also shown in Fig. 2; the horizontal part denotes photon shot noise which is determined only by the mean photon count rate, and the low-frequency part, which is the background, is more than 1 order of magnitude smaller than the observed PSD of the Brillouin components. It was confirmed from this comparison that the observed PSD should be attributed to scattering inside the PSD, and the possibility of environmental (temperature, etc.) fluctuations is excluded. However, further evidence is needed to show that the PSD refers to phonon-number fluctuations. If the observed PSD of the ratio refers to phonon-number fluctuations and if phonon-number fluctuations are not correlated from mode to mode, the $1/f$ spectral part must be inversely proportional to the number of phonon modes involved in the scattering. Therefore, evidence can only be obtained through dependence of the $1/f$ spectral level on it.

The number of phonon modes N involved in the Brillouin scattering is given by

$$N=[2V/(2\pi)^3]\,\Omega q^2 dq\,, \tag{4}$$

where Ω is the solid angle spun by the scattered light cone and V is the scattering volume. The uncertainty of q, dq, is attributed to a finite linewidth either of the laser (3 MHz) or the phonon, whichever is larger. Currently, we are not successful in experimentally estimating the linewidth of the Brillouin line, and the result of Pine[11] was adopted. He measured a linewidth of 38 MHz for 28-GHz longitudinal-acoustic phonons in quartz at room temperature. For quartz, $v=1.54$ and $u=5860\ \mathrm{ms}^{-1}$, and we have $f_p=24.8$ GHz; since the decay constant is approximately proportional to frequency within a narrow frequency band,[12] we estimate the linewidth for 24.8-GHz phonons to be 33 MHz, from which $dq=3.54\times10^4\ \mathrm{m}^{-1}$. The slit width and the aperture diameter of the lens were varied to change values of V and Ω. In this way the number of phonon modes involved in the scattering was varied (the count ratio was insensitive to the laser intensity). As the phonon frequency equals 24.8 GHz, the phonon wavelength equals $(5860\ \mathrm{m\,s}^{-1})/(2.48\times10^{10}\ \mathrm{s}^{-1})=2.36\times10^{-7}$ m and $q=2.66\times10^7\ \mathrm{m}^{-1}$. Since we have $V=2.01\times10^{-10}\ \mathrm{m}^3$ and $\Omega=1.73\times10^{-5}$ sr in the case of the data in Fig. 2, we obtain from Eq.

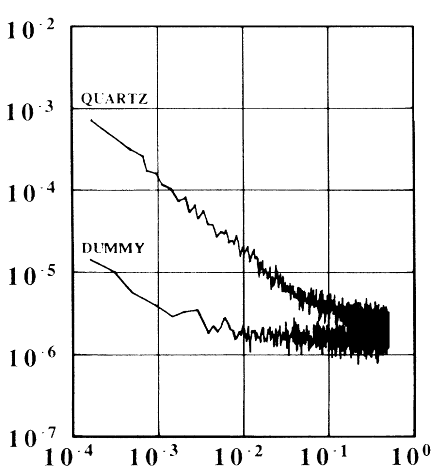

FIG. 2. Estimated PSD of Eq. (3) and the background PSD.

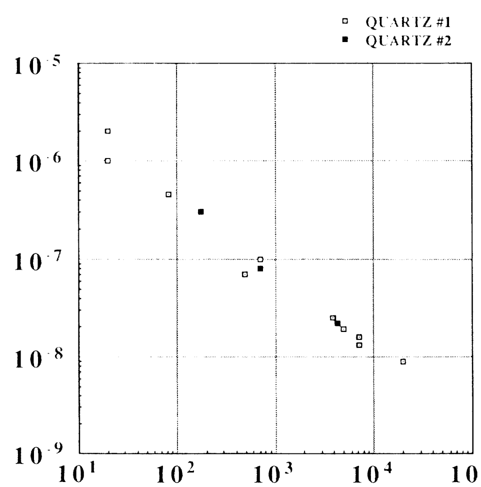

FIG. 3. Dependence of the $1/f$ spectrum at 1 Hz on the number of phonon modes involved in the scattering.

(4) $N=701$.

The $1/f$ spectral curve in Fig. 2 is extended to 1 Hz and the spectral level at this frequency was plotted in Fig. 3 as a function of the N value for two specimens (1 and 2) which have Q values (at 5 MHz) of 3×10^6 and 1×10^6; the Q value is defined as a reciprocal acoustic loss rate in a period of vibration. Measurements with specimen 1 were made over 1 year and the data were quite reproducible; the data referring to the two specimens are on the same line, hence the different Q values made no meaningful difference. It has been found from this plot that the $1/f$ spectral level is inversely proportional to the number of phonon modes N. The present result is evidence for the two facts that the phonon-number fluctuations have a $1/f$ spectrum and that they are not correlated from mode to mode. It should be noted that independence of phonon-number fluctuations from mode to mode does not mean that the $1/f$ lattice vibrations have no spatial correlation. Conclusively, an empirical formula of fractional phonon-number fluctuations per mode is

$$S(f)=8\times10^{-5}/f. \tag{5}$$

Observation of the $1/f$ spectrum means that the original fluctuations are not (even weakly) stationary. If the term "thermal equilibrium" means a stationary state, we should say that the variance of lattice vibrations does not reach thermal equilibrium for a very long period of time. The present observation suggests that the general $1/f$ conductivity fluctuations are, at least partly, related to fluctuations of phonon excitation around its mean value.

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