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Direct optical observations of surface thermal motions at sub-shot noise levels

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We measure surface thermal fluctuation spectra of liquids, solids, complex fluids, and biological matter using reflected light measurements down to $\sim 10^{-17}$ rad^2/Hz in inclinations or $\sim 10^{-29}$ m^2/Hz in vertical displacement. The random thermal fluctuations are delineated from random noise at sub-shot noise levels. The principle of the measurement is not limited to surface measurements. The dynamical evolution of the surface properties is also investigated. The measurement time is short, passive, and applicable to a wide variety of surfaces.

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Thermal fluctuations exist for everything we see and it would be quite interesting to see them directly. However, the *atomic* fluctuations themselves are small and have been directly seen only on few types of surfaces, such as "ripplons" on liquid surfaces [1,2] and high power interferometry measurements on mirrors [3]. When the surface tension is exceptionally low, giant fluctuations have been observed [4]. Our objective is to measure the spectra of common place surface thermal fluctuations at atomic scales. We have directly observed the power spectra (1 kHz ~ 10 MHz) of thermal fluctuations on various kinds of surfaces, presenting some of their interesting cases in this work. The measurements utilize a method that determines the local inclinations of the surface with an optical lever [5] down to orders of magnitude below shot noise levels, separating the random signal from random noise. The optical intensity used in our experiments is relatively low, 1 ~ 10 μW at the photodetectors, with 0.1 ~ 1 mW power light applied to the samples. The latter is higher since the reflectivity of the surfaces we observed are not high. The obtained power spectra are analyzed and are related to the physical properties of materials.

The main difficulty in directly measuring the properties of thermal fluctuations is that the fluctuations are small and random. In any measurement, some random noise exists, from which the signal needs to be extracted. Simple averaging will not suffice to separate out the *random* signal, unless it is large. A detector measurement $D_1 = S + N_1$ consists of the desired signal S and some noise N_1 independent of S. Denoting the Fourier transforms with tildes, the power spectrum is $\langle |\tilde{D}_1|^2\rangle = \langle |\tilde{S}|^2\rangle + \langle |\tilde{N}_1|^2\rangle$ if we simply average. Cross terms of Fourier transforms of decorrelated random observables vanish under averaging. Unless $\langle |\tilde{S}|^2\rangle \gg \langle |\tilde{N}_1|^2\rangle$, the signal cannot be extracted from the noise. However, even a small random signal can be recovered by making a statistically independent measurement, $D_2 = S + N_2$, of the same signal. Then, $\langle \tilde{D}_1 \overline{\tilde{D}_2}\rangle \to \langle |\tilde{S}|^2\rangle (\mathcal{N} \to \infty)$, where $\mathcal{N}$ is the number of averagings. As in $\langle |\tilde{D}_1|^2\rangle$, other terms in the correlation average to zero. The relative error in this method is statistical and its size is $\sim 1/\sqrt{\mathcal{N}}$. While correlation measurements have been used previously in surface light-scattering experiments [6], our approach differs from those in that we use the cross correlation of independent measurements of the same sample at the same instant to reduce the noise. Clearly, this theory of noise reduction is not limited to surface or optical measurements. The crucial requirement for the method is that the random noise in D_1, D_2 are decorrelated.

The experimental setup in Fig. 1 includes an optical lever [Fig. 1(a)] that measures the average surface inclination within the beam spot through the difference between the reflection signal currents. The beam is focused down to μm order, smaller size than the standard light scattering methods. At this scale, even for a nonsmooth surface like that of rubber, the scale of roughness is not large compared to the wavelength. Therefore the light is not scattered in all directions but is reflected back and all such light can essentially be collected by the objective lens. Consequently, we obtain relatively large signals for a given beam power when compared to the standard light scattering, which picks up only a fraction of the scattered light. This is important in low light power measurements.

Since we need two independent measurements for the signal, we use two sets of dual-element photodiodes (DEPDs), DEPD1,2 [S4204 Hamamatsu Photonics, Japan, Fig. 1(b)]. DEPD signals are amplified and fed into a personal computer via an analog to digital converter (ADC) (14 bit, ADXII14–80M, Saya, Japan) to perform fast Fourier transform (FFT) and averaging. The laser beam power at the sample is 2 mW each, at most. When the sample is an organic matter, such as rubber, thermal effects are important so that a neutral density

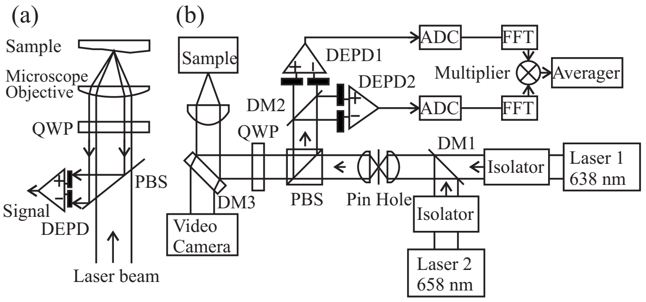

FIG. 1. (a) Schematic of an optical lever: laser beam focused by an objective lens is applied to the sample. The reflected light is picked by the same lens and is detected by DEPD. QWP: quarter-wave plate; PBS: polarizing beam splitter. (b) Experimental setup: by combining the laser beams with wavelengths 638, 658 nm at a dichroic mirror (DM1) and separating them at DM2, we obtain two independent measurements of the same location. DM3 is for observing the sample via a video camera. FFT: fast Fourier transformation.

filter is used to further reduce the beam power. The most significant experimental limitations to the sensitivity are various sources of cross-talk. Cross-talk can arise from such diverse sources as ADC, nonlinear elements in the optical elements, and coupling through the electromagnetic fields. In our experiments, we measured the size of the cross-talk prior to the measurements and corrected for it.

The major causes of noise in the experiment are the shot noise and the thermal noise in the detectors (Nyquist noise), which exist even in ideal situations. Our method enables the measurements to be made at orders of magnitude below the noise level, as seen below. In our measurements, the thermal noise does not dominate for signal light intensities larger than 1 μW, which applies to all the measurements below, and mostly, there is far less thermal noise than the shot noise. We obtain relatively large signals since we collect essentially all the reflected light. Thermal noise can also be reduced, at least in principle, by cooling while the shot noise cannot. To overcome the error due to the shot noise, one can increase the beam power, but this is not always possible since raising the power can affect the sample itself, making it impossible to obtain meaningful results. Furthermore, large power is prohibitive *in situ* where noninvasiveness is required, such as medical applications [7]. Given the same power, our method can extract far weaker signals than those obtainable by other methods. The main error in our work is statistical; in theory, as well as in practice, there is a limitation that if a frequency resolution Δf is required, time $\mathcal{N}/\Delta f$ is necessary to make the measurement. In the experiments, the measurements took 30 s to 30 min, longer times being used for weaker signals.

Thermal fluctuations of simple liquid surfaces appear as capillary waves called “ripplons” and light scattering from them have been studied for some time [1,2]. The dispersion relation of ripplons has been derived theoretically, $P_{\mathrm{R}}(q,\omega) = k_{\mathrm{B}}Tq\tau_0^2/(\pi\omega\rho)\mathrm{Im}[(1+s)^2+y-\sqrt{1+2s}]^{-1}$, where $s \equiv -i\omega\tau_0$, $\tau_0 \equiv \rho/(2\eta q^2)$, $y \equiv \sigma\rho/(4\eta^2 q)$ [8,9], ρ,σ,η are the density, the surface tension, and the viscosity of the liquid, respectively, q,ω are the wave number and the frequency of the capillary waves, respectively, k_{B} is the Boltzmann constant, and T is the temperature. Our method differs from previous measurements in that we directly measure the spectrum of surface inclination fluctuations, corresponding to the spectrum $S_{\mathrm{R}}(\omega) = \int_{q_{\min}}^{q_{\max}} d^2q/(2\pi)^2 q^2 e^{-b^2q^2/8} P_{\mathrm{R}}(q,\omega)$. Here, b is the Gaussian beam diameter and the q^2 factor arises from observing inclinations. $q_{\min}$ is the cutoff due to the system size ($\sim$few mm), which also limits the waves to be only capillary. In the frequency range we study, the corresponding wavelength is much smaller than mm scale so the integral is insensitive to the exact value of $q_{\min}$. The cutoff $q_{\max}$ is at inverse atomic scales, but the integral is damped by a Gaussian and as long as $q_{\max} \gg 1/b$, the difference is negligible. As can be seen here, the beam size cuts off the surface fluctuations with wavelengths or coherence lengths smaller than b. The vertical displacement spectrum is $b^2 S_{\mathrm{R}}(\omega)$ and its spectral density goes down to $\sim 10^{-29}$ m^2/Hz. When integrated over all frequencies, the total vertical displacement is a few angstroms.

The theoretical results $S_{\mathrm{R}}(2\pi f)$ agree well with the experimental observations, as in Fig. 2. The properties of the liquids are (ρ[kg/m^3], σ[kg/s^2], η[kg/(m s)]) $=(1.0\times 10^3,\ 7.3\times 10^{-2},\ 1.0\times 10^{-3})$, $(0.79\times 10^3,\ 2.2\times 10^{-2},\ 1.1\times 10^{-3})$, $(0.92\times 10^3,\ 3.0\times 10^{-2},\ 0.124)$ for water, ethanol, and immersion oil. $b=2.5$ μm for oil, ethanol, and 3.2 μm for water. Due to the relatively high viscosity of oil, there is a qualitative difference for its spectrum which decays as f^{-2} at higher frequencies, while those of water and ethanol decay as f^{-4}. This is well reproduced in the measurements.

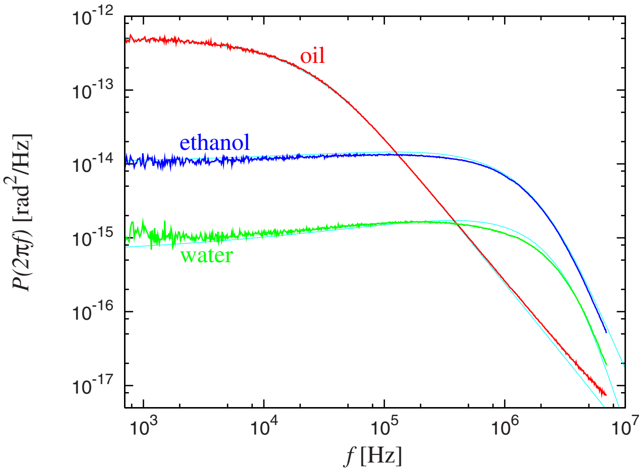

FIG. 2. (Color online) Spectra of capillary waves on water, ethanol, and oil surfaces (labeled in the plot). The theoretical predictions for them are also indicated (thin line, cyan).

The frequency dependence is accurate and quite reproducible, but the overall magnitude is harder to calibrate and a piezoelectrically driven mirror with a known oscillation size was used to determine this magnitude to within a factor of 2. In Fig. 2, the overall magnitude of the signal was adjusted within this range to fit the theoretical formula. The major limitation in this calibration is the precision with which we can adjust the sample surface to the focal plane of the objective lens. If one wants to use the spectrum to determine the physical parameters of the sample, parameters that change the spectrum shape can be obtained with some accuracy, while, at this point, those that change only the overall magnitude are more difficult to ascertain. For example, the spectra in Fig. 2 can be used to determine the surface tension or the viscosity of the liquid to 10%.

The shot noise level is f independent and can be estimated as $N_{\mathrm{SN}}(\omega) = NA^2 e/(2I)$, where I is the photocurrent and NA is the numerical aperture of the objective lens. We used a lens with $NA=0.5$ throughout and in all our measurements $I=0.1$ μA to few μA. $T=25$ °C for all our measurements. $N_{\mathrm{SN}} \simeq 2\times 10^{-15}$ [rad^2/Hz] in the above ripplon measurements so that the results in Fig. 2 go down to a couple of orders of magnitude below the shot-noise level.

Using the same method, the surface fluctuations of rubber with varying strain are observed in Fig. 3. An estimate for the fluctuation spectrum as an elastic medium [10–12] is $S_{\mathrm{S}}(\omega) = 16k_{\mathrm{B}}T(1-\sigma_{\mathrm{P}}^2)\phi/(\sqrt{\pi}\omega b^3 Y)$, where $Y,\sigma_{\mathrm{P}},\phi$ are the Young’s modulus, Poisson’s ratio, and the loss angle of the material, respectively. In the plot, we used natural rubber stretched to various lengths. As a guide, we indicated

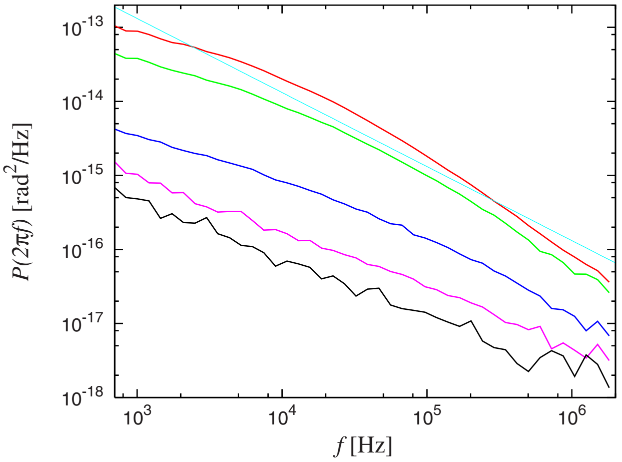

FIG. 3. (Color online) Power spectrum of surface fluctuations of rubber in the elongated direction when the extension ratio is 1 (red), 2 (green), 3 (blue), 4 (magenta), and 6 (black) in the decreasing order of fluctuation amplitude. For comparison, the theoretical formula corresponding to surface fluctuations of an elastic material is also shown (thin line, cyan). $N_{\mathrm{SN}} \simeq 1\times10^{-14}$ [rad^2/Hz] in this measurement.

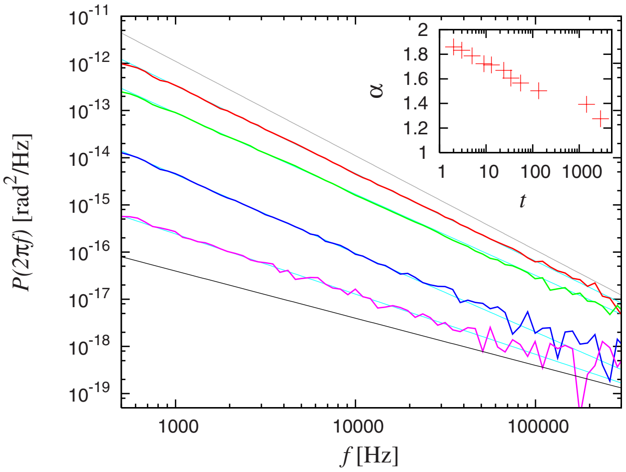

FIG. 4. (Color online) Epoxy adhesive surface fluctuations in decreasing order of its size, 2 (red), 9 (green), 24 (blue) minutes, and 2 days (magenta) after application, together with fits to them to functions of the form const. $\times f^{-\alpha}$ (thin lines, cyan), which work quite well. As a guide, theoretical results for a simple liquid (top straight line, gray) and an elastic material (bottom straight line, black) are shown also (see text). The fluctuations decrease with time. The change in f dependence of the surface fluctuation power spectrum with respect to time t[min] shows a slow decrease between $\alpha=2$ and $\alpha=1$ (inset). $N_{\mathrm{SN}} \simeq 4\times10^{-15}$ [rad^2/Hz] in this measurement.

the spectrum $S_{\mathrm{S}}(\omega)$ with frequency independent $Y=3.5\times10^{6}$ [kg/(m s^2)], $\sigma_{\mathrm{P}}=0.475$, $\phi=0.1$, which are typical values for rubber. The dependence on f does not agree completely with $S_{\mathrm{S}}(\omega)$, which is natural since Y,ϕ depend on f [13] and rubber is a complex material that changes its state as it is elongated [14]. Obviously, the dependence can be perfectly reproduced if we assume a particular f dependence for ϕ/Y. The signal decreases as the Young's modulus increases when the rubber is stretched [14]. We have also measured the fluctuations in the inclinations transverse to the direction of extension and find them to be larger than those parallel to it, as expected, indicating the existence of more flexibility in the transverse direction. Beam had $b=1$ μm with a power 150 μW. Lowering this power did not change the spectrum but substantially raising the power affects the spectrum, presumably by heating up and perhaps melting the material. Consequently, this result is difficult to obtain without separating out the random noise using the logic explained above.

Standard light-scattering methods rely on obtaining measurements for a particular q, and in dissipative materials, such as rubber and the materials we subsequently investigate, surface waves have small coherence lengths so that it would be difficult if not impossible to obtain meaningful surface fluctuation measurements of them. In our method, we do not rely on a measurement for a particular q but instead measure the whole spectrum from the time dependence of the surface deformations. This ability to obtain the surface fluctuation spectra of various materials is what motivated us to pursue this work.

We now consider a more complex material, an epoxy adhesive, whose properties change over time, as the glue "hardens." Our method allows us to obtain spectral properties quickly without any contact, and we can see how the spectrum changes with time. The measurements are shown in Fig. 4 where we used a laser beam with $b=1$ μm. The thermal fluctuation spectrum at each instant can be well described by a simple power, $P(f)\sim f^{-\alpha}$. This power α decreases with time with values between 2 and 1 [Fig. 4 (inset)]. We recall that for highly viscous fluids, $\alpha=2$ in this frequency range, as can be seen in Fig. 2, and $\alpha=1$ for elastic materials, as in $S_{\mathrm{S}}(\omega)$. The results are quite consistent with an evolution of the epoxy adhesive between these two states, which seem quite natural. For comparison, the ripplon spectrum $S_{\mathrm{R}}(\omega)$ with typical values for an epoxy adhesive, $(\rho[\mathrm{kg/m^3}],\ \sigma[\mathrm{kg/s^2}],\ \eta[\mathrm{kg/(m\ s)}])=(1.5\times10^{3},\ 4.0\times10^{-2},\ 5000)$ and the spectrum for an elastic material with $(1-\sigma_{\mathrm{P}}^{2})Y/\phi=1.5\times10^{11}$ [kg/(m s^2)] are also indicated. Phenomenologically, the time dependence of α can be well described by a logarithmic one in the relevant region (Fig. 4 inset) and the reason for this is worth further study.

Our method is well suited to biological materials since low power laser beams can be used. Figure 5 shows the measurements of the surface fluctuation spectrum of an ayu (sweetfish) eye as it dehydrates. The laser beam, focused on the cornea, has a power of 200 μW and $b=1$ μm. Measurements were performed when the eye was wet and then after certain time had elapsed. Not only quantitative but clear qualitative differences in the fluctuation spectrum can be seen with time. At the beginning, the eye surface is wet and at lower frequencies, $f\lesssim40$ kHz, the fluctuations decay as $\sim f^{-1.6}$ as can be confirmed in Fig. 5. This property is similar to that of a highly viscous liquidlike oil in Fig. 2 and viscous complex fluidlike epoxy in Fig. 4. For higher frequencies, $f\gtrsim40$ kHz, the spectrum has almost no frequency dependence and behaves similarly to the water spectrum in Fig. 2, including its magnitude. This suggests that the material contains water substantially and is a gel-like material. With time,

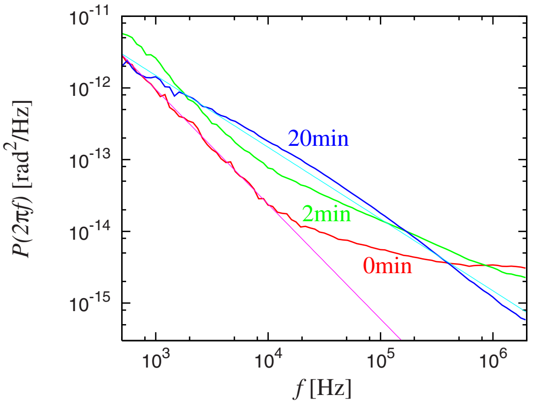

FIG. 5. (Color online) Surface fluctuations of an ayu eye when it is wet, 2 and 20 min thereafter (labeled in the plot). $6.0 \times 10^{-8} f^{-1.6}$ is shown (magenta, bottom straight line). Theoretical formula for an elastic material is also indicated (top straight line, cyan). $N_{\mathrm{SN}} \simeq 2 \times 10^{-14}$ [rad^2/Hz] in this measurement.

water evaporates and the thermal fluctuation spectrum changes from that of a fluid to that of a solid. We show the surface fluctuations of an elastic material in $S_{\mathrm{S}}(\omega)$ with $(1-\sigma_{\mathrm{P}}^2)Y/\phi = 4.0 \times 10^6$ [kg/(m s^2)] in Fig. 5, which describes the spectrum after 20 min quite well. This spectrum is similar to the rubber surface fluctuation spectrum in Fig. 3.

In this work, we used an optical lever [15] to measure power spectra of thermal fluctuations of a wide variety of surfaces, from simple liquids to biological matter. By analyzing the fluctuation spectra of various types of matter and relating their spectra to their physical properties, the spectra for complex materials could be qualitatively explained from the understanding of the spectra of simpler matter. The reason it is possible to make these sensitive measurements at our low optical intensity is because the measurements were performed down to orders of magnitude below the shot-noise level. There are situations, such as gravitational wave measurements, which are usually believed to be shot-noise limited [16,17] and our method can perhaps significantly improve the capabilities of those. The measurements can be noninvasive and is applicable to all kinds of surfaces, including biological matter and may be effective in studying biological phenomena, such as the dry eye syndrome. By making the optical system confocal, depth resolution can also be achieved. Furthermore, sensitive measurements of the properties of liquid/liquid interfaces and the eyeball interior are possible in our approach. Our measurement requires relatively a short time, allowing us to take spectral snapshots of surfaces, observing the time evolution of physical properties, as exemplified above.

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