Milli-Hertz Surface Spectroscopy

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A technique that has been repeatedly employed in high resolution light scattering experiments is that of light beating, or *heterodyne*, spectroscopy.[1] By detecting the beating signal between the scattered light and a 'local oscillator' derived from the same laser source, one can obtain ultrahigh spectral resolution, independent of the random fluctuations of the light source. We reported earlier of a novel Fourier transform heterodyne spectroscopy (FTHS) technique with high resolution[2] which is simpler and more direct than the conventional heterodyne technique; we have since improved our resolution ten-thousand fold to the 20-μHz range. We applied this technique first to study nonequilibrium phenomena at liquid-vapor interfaces. The ultrahigh resolution also enables one to observe the very small Doppler shift of a light beam reflected from a growing silicon crystal.

1. Fourier Transform Heterodyne Spectroscopy

In heterodyne spectroscopy, one detects the beating of the scattered light—with itself or with part of the incident light—and then analyzes the spectrum of the detector signal with a spectrum analyzer or autocorrelator. The main advantage of this scheme is that fluctuations in the phase of the incident laser field, which limit the resolution of spectroscopy, cancel out as long as the two fields reaching the detector are coherent.

One of the major drawbacks of conventional heterodyning, however, is that at near zero frequency the spectrum is usually distorted by high-pass filters used to block the DC component of the signal. The present detection scheme circumvents this problem by shifting the origin of the spectrum from zero to some optimal frequency. In this way, the spectrum at frequencies close to that of the incident light is still reliable, making the detection of very small Doppler shifts possible. As the beating of two signals is sensitive only to the difference frequency, the use of such frequency-shifted local oscillators has the additional advantage of spectrally separating the up-shifted and down-shifted beams—a feature which is indispensable in the study of nonequilibrium fluid interfaces. Figures 1a and 1b show two experimental setups that can be used for doing FTHS. In Fig. 1a the main beam and the local oscillator are mixed after the experiment, while in Fig. 1b they are mixed before incidence on the sample.

2. General Considerations

In each of these setups, the detector signal is sampled for a period T_s (the *sampling time*), and the digitized signal is then Fourier transformed to obtain the spectrum. According to sampling theory, the time interval between adjacent sampling points, δt_s (the inverse of the sampling frequency f_s), and the spectral range F are related by

$$F = \frac{1}{2\delta t_s} = \frac{1}{2} f_s. \tag{1}$$

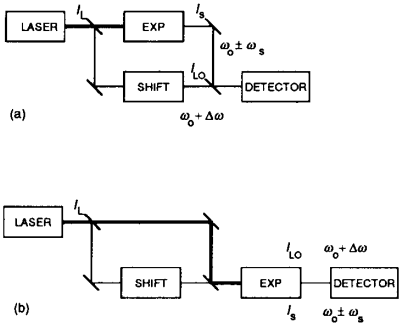

Fig. 1. Experimental schemes for FTHS: (a) post-sample mixing; (b) pre-sample mixing.

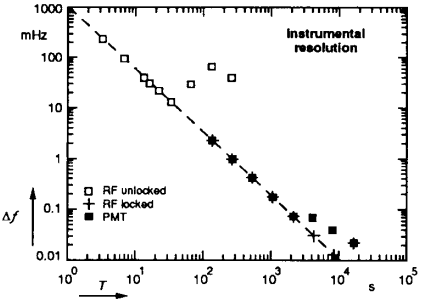

Fig. 2. Instrumental resolution measurements: electronic mixing with independent (□) and locked (+) RF drivers, and optical mixing with locked drivers (■).

We have used a variety of sampling frequencies, between 1 Hz and 20 kHz, resulting in a spectral range from 0.5 Hz to 10 kHz. In general, the upper limit of the range is determined by either the detector response time, or the speed of the signal digitizer; in practice with a 1-ns response time and a fast transient digitizer, the spectral range can be extended to the GHz regime. With a streak camera this could even be further extended by at least two orders of magnitude. Because of this broad range, the technique is applicable to a wide variety of fields of research.

Similarly the sampling time, T_S, and the frequency interval between adjacent spectral points, δf_S, are related by

$$\delta f_S = \frac{1}{T_S} . \qquad (2)$$

Although this shows that the resolution can be made infinitely small for an infinitely long sampling time, in reality the resolution is limited by the electronic and mechanical stability of the setup.

To determine the instrumental resolution, the local oscillator was mixed directly with the main laser beam. The local oscillator was frequency-shifted by a few kHz using a combination of two acousto-optic modulators. The carrier frequency of the modulators was in the 20–40 MHz range, with a frequency difference equal to the desired frequency shift. When the two modulators are driven with two independent radio-frequency (RF) drivers, the resolution decreases with increasing sampling time as expected from Eq. (2). For sampling times in excess of 60 s, a loss of resolution starts to occur (see open symbols in Fig. 2). By comparing the optical beat spectrum with the one obtained by directly beating the two RF signals, it was found that the resolution limit was caused by instabilities in the MHz modulator frequencies. With two Hewlett Packard frequency synthesizers, locked to the same time base, the situation could be improved by a factor of one thousand. Figure 2 shows the resolution of both optical and RF beat spectra. As can be seen, the RF resolution follows Eq. (2) down to 10 μHz. The optical resolution starts to show deviations at about 70 μHz, most likely because of mechanical instabilities of the optical setup during the half-hour sampling time. The highest instrumental resolution of the present setup, 20 μHz, was obtained with a four-hour sampling time.

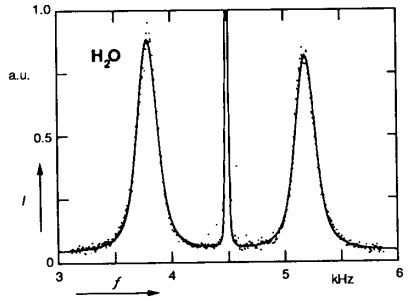

Fig. 3. Spectrum of light scattered from a nonequilibrium liquid surface fitted to three Lorentzian lines.

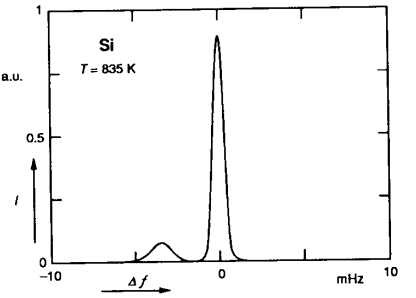

Fig. 4. Spectrum showing the Doppler shift of light reflected from a growing silicon crystal.

3. Application to Liquid Surface Studies

The above technique was first used to study nonequilibrium phenomena at a water-air interface. Thermal fluctuations on a fluid interface give rise to a Brillouin doublet in the spectrum of light scattered from the fluid interface.[3,4] In equilibrium, the Stokes and anti-Stokes Brillouin peaks have the same intensity since the populations, n_S and n_a, of capillary waves travelling in opposite directions are equal. Out of equilibrium, however, n_S and n_a are no longer equal and the spectrum becomes asymmetric. The FTHS, with its directional separation capability, renders the study of such asymmetric Brillouin doublet possible. A schematic diagram of the surface light scattering apparatus is shown in Fig 1a. A more detailed description of the apparatus can be found in Ref. 5. After scattering off a water-air interface subject to a temperature gradient, the scattered beam is combined with the shifted local oscillator. The detector signal is sampled for a certain period of time, stored in a microcomputer and then a fast Hartley transform[6] is applied to the sampled waveform. The resulting spectrum, an example of which is shown in Fig. 3, corresponds to the spectrum of the interfacial fluctuations.[7] These measurements confirm the existence of the predicted asymmetry in the Brillouin spectrum of light scattered from a fluid interface subject to a temperature gradient.[5] A more detailed discussion will appear in a forthcoming paper.

4. Application to Crystal-Growth Measurements

Because of the high instrumental resolution, one can use the FTHS technique to observe very small frequency shifts. A possible application is to measure extremely small Doppler shifts. To illustrate this we have applied this novel technique to study crystal growth at the crystalline-amorphous silicon interface in small silicon samples composed of 300-nm amorphous silicon on a substrate of crystalline silicon. When such a sample is placed on a heating unit, the amorphous material slowly crystallizes and the crystalline-amorphous interface moves up toward the air-amorphous silicon interface. The growth rate can be varied by adjusting the temperature; at 835 K it is approximately 0.2 nm/s. Light reflected from the moving interface will be Doppler shifted by a few mHz.

High temperature air turbulence around the sample and reduced mechanical stability of the heater holding the sample greatly lower the overall stability of the setup. It was therefore necessary to mix the main laser beam with the local oscillator *before* the sample, to ensure constant overlapping of the two (see Fig. 1b). To suppress reflection from the front surface

(air-amorphous silicon), the incidence angle of the beam was kept near the Brewster angle. The main beam was *p*-polarized, the shifted local oscillator *s*-polarized. Most of the local oscillator beam is reflected at the air-amorphous silicon interface, while most of the main beam penetrates the amorphous layer and is reflected at the crystalline-amorphous interface. A beating signal between the two reflected beams was obtained with a 45° polarizer and recorded with a photomultiplier. Figure 4 shows a spectrum obtained this way. The large peak comes from the beating of the main beam and the local oscillator reflected at the air-amorphous silicon interface. The smaller peak is Doppler shifted by the motion of the crystalline-amorphous interface. The Doppler shift in this case is 3 mHz, corresponding to a speed of about 0.2 nm/s. This is in good agreement with interferometric measurements.[8]

In principle, one can obtain the *distribution* of speeds over the sampled area from the broadening of the Doppler shifted peak (the shift reflects only the *average* speed). The resolution of these measurements, however, is limited by the maximum sampling time, which in turn is limited by the ratio of the amorphous layer thickness to the growth rate. For a fixed growth rate this means one needs thicker samples to increase the resolution. Measurements on thicker samples are currently in progress.

5. Conclusion

This paper presents a simple heterodyne technique with ultrahigh resolution. Besides spectrally separating the up-shifted and down-shifted scattered light, the technique also allows one to obtain undistorted data at the low frequency end of the spectrum. This is essential both in low-frequency work as well as in cases where directional separation is called for, such as the study of nonequilibrium liquid interfaces. The ultrahigh resolution makes the technique also suitable to measure extremely small Doppler shifts. In air, a shift of 20 μHz corresponds to a speed of 7×10^{-12} m/s. Since a spectral range of up to 1 GHz can be covered, Fourier transform heterodyne spectroscopy can be applied to study a broad range of physical phenomena.

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