Fourier Transform Heterodyne Spectroscopy: A Simple Novel Technique with Ultrahigh (150 mHz) Resolution

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Light beating spectroscopy has been used from the early days of the laser to study light scattering.[1] By detecting the beating signal between the scattered light and a 'local oscillator' field derived from the same laser, resolving powers of 10^{14} have been achieved. The Fourier transform heterodyne spectroscopy presented here is simpler and more direct than the conventional heterodyne techniques using autocorrelators or spectrum analyzers.

The techniques to measure spectra of scattered light fall into two categories as illustrated in Fig. 1: frequency-domain and time-domain spectroscopy. In the frequency domain (Fig. 1a) one first spectrally filters the incoming light and then detects the transmitted signal. The spectral resolution of this technique is limited by either the resolution of the filter (monochromator, interferometer, etc.), or by the bandwidth of the laser. In the time domain (Fig. 1b) 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 frequency-domain spectroscopy, cancel—provided the two fields reaching the detector are coherent. Also, the *total* signal I_s is detected, rather than the filtered intensity I_ω, yielding a higher signal-to-noise ratio.

In our present detection scheme an acousto-optically shifted local oscillator (Fig. 1c) allows us to study spectra of scattered light near the incident laser frequency. The detector signal is sampled for a certain length of time, stored in a microcomputer, and then a fast Fourier transform is applied to the sampled waveform. It can readily be shown that the resulting datapoints correspond precisely to the (shifted) spectrum of the scattered light $S(\omega)$. This scheme bypasses the need to filter or process the detector signal with an analog spectrum analyzer or autocorrelator.

The local oscillator can also be frequency shifted by reflecting it from a moving mirror (Fig. 1d): a speed of 1 mm/s results in a shift of 3 kHz on a He-Ne beam. Stability in mirror motion then limits the resolution. With a servo mechanism, a stability better than 0.02% can be achieved, so that a resoluion of 1Hz is possible with a shift of 5 kHz. The data shown below were all obtained by acousto-optic frequency shifting.

We are currently applying this technique to the study of hydrodynamic fluctuations in liquid-vapor interfaces.[2] Fig. 2 shows the spectrum of light scattered from the liquid-vapor interface of water at room temperature. The full Rayleigh-Brillouin triplet, centered around the 4.9 kHz frequency shift of the local oscillator, is visible. The central quasi-elastic Rayleigh scattering is due to nonpropagating fluctuations in the interface, whereas the Brillouin peaks at a shift of 1.2 kHz result from propagating fluctuations (capillary waves). Without frequency shifting the two Brillouin peaks merge into one single peak at 1.2 kHz (see position marked

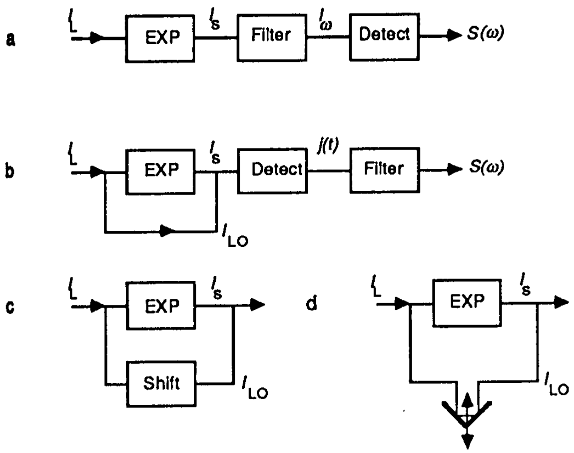

Fig. 1. Spectroscopy configurations: (a) frequency and (b-d) time domain detection techniques.

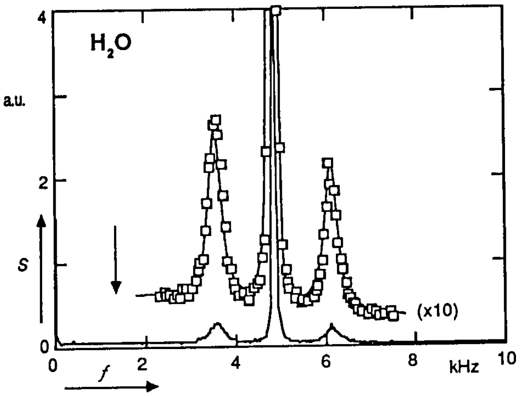

Fig. 2. Fully resolved Rayleigh-Brillouin triplet of light scattered from the liquid-vapor interface of water at room temperature.

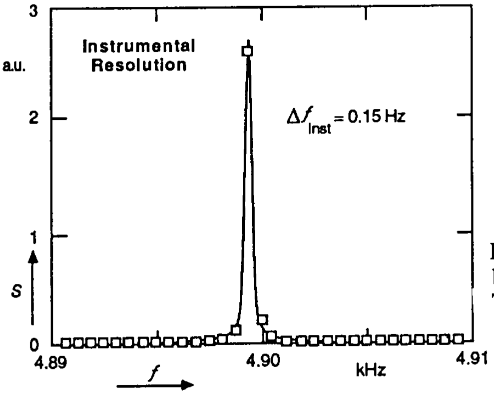

Fig. 3. Instrumental resolution obtained by replacing the sample with a mirror. The halfwidth of the Lorentzian fit is 150 mHz.

with arrow). A full discussion of these results will appear elsewhere.[3] Fig. 3 shows the instrumental resolution, obtained by replacing the interface with a fixed mirror. The resolution is roughly inversely proportional to the sampling time of the signal. For a single 1.5 s sampling the resolution is 150 mHz as shown. For multiple samplings some degradation in resolution was observed due to frequency drifts of the acousto-optic driver. For the same reason the resolution does not increase if the sampling time is increased beyond 1.5 s.

Summarizing, we present here a simple heterodyne technique with ultrahigh resolution. Because a spectral range up to 1 GHz can be covered, Fourier transform heterodyne spectroscopy is applicable to a wide variety of fields of research. The ultrahigh resolution makes the technique also suitable to measure extremely small Doppler shifts: the resolution of 150 mHz corresponds to a speed $v \approx 50$ nm/s. It is limited, however, only to *coherent* scattering processes, and the reported resolution is *relative*, not absolute.

References

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2. See for instance: V.G. Levich, *Physiochemical Hydrodynamics* (Prentice-Hall, New Jersey, 1962); R. Loudon, in *Surface Excitations*, Ed. V.M. Agranovich and R. Loudon (Elsevier, Amsterdam, 1984) 589
3. Eric Mazur and Doo Soo Chung, Physica A *to be published* (December, 1987)