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. 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 $I_{so}$ 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 data points correspond precisely to the (shifted) spectrum of the scattered light $S(o)$. 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 resolution of 1 Hz 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. 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...
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used from the early days of the laser to study light signal between the scattered light and a "local oscillator" driving powers of $10^{14}$ have been achieved. The Fourier presented here is simpler and more direct than the existing autocorrelators or spectrum analyzers.

Two types of scattered light fall into two categories as illustrated for domain spectroscopy. In the frequency domain (Fig. a), the transmitted signal. The is limited by either the resolution of the filter (monochromatic bandwidth of the laser). In the time domain (Fig. b), the incident light—with itself or with part of the incident light—and the signal with a spectrum analyzer or autocorrelator. In the latter case fluctuations in the phase of the incident laser field, in domain spectroscopy, cancel—provided the two fields are equal in phase. The total signal $I_t$ is detected, rather than the filtered signal $I_f$.

We use an acousto-optically shifted local oscillator (Fig. 1c) to frequency shift the scattered light near the incident laser frequency. The detector is a time (of stored in a microcomputer, and then a fast output to an analog spectrum analyzer). The spectral bandwidth of the incident laser, $\Delta f_{\text{LO}}$, is shifted by reflecting it from a moving mirror or a shift of $3\text{kHz}$ on a He-Ne beam. Stability in mirror frequency shifting, $\Delta f_{\text{in}}$, on account of a servo mechanism, a stability better than $0.02\%$ can be achieved with a shift of $5\text{kHz}$. The data shown below illustrate this technique and the results of simulations performed.

The full Rayleigh-Brillouin triplet, centered around the incident frequency, is visible. The central quasi-elastic Rayleigh scattering of the incident light, whereas the Brillouin peaks at higher frequencies (capillary waves). Without frequency shifting, a single peak at $1.2\text{kHz}$ (see position marked on the graph). In the time domain, $S(t)$ is obtained as the output of the amplifier. The instrument resolution, $\Delta f_{\text{in}}$, is $0.15\text{Hz}$. The halfwidth of the Lorentzian fit is $150\text{mHz}$. The instrumental resolution obtained by replacing the sample with a mirror. The halfwidth of the Lorentzian fit is $150\text{mHz}$.

![Diagram](image)

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

![Graph](image)

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

![Graph](image)

Fig. 3. Instrumental resolution obtained by replacing the sample with a mirror. The halfwidth of the Lorentzian fit is $150\text{mHz}$.
with arrow). A full discussion of these results will appear elsewhere. 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 ultra-high 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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High Frequency Modulation
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Frequency modulation (FM) spectroscopy is a sensitive differential absorption technique.
If a laser is frequency modulated, the sidebands are spectrally distinct from the laser
beam and impinge on a square law detector, each at a different frequency. The two beat
notes are exactly out of phase if the modulation frequency is exactly twice the
absorption frequency. The fact that there is no laser noise at the modulation frequency
appears to make this a very sensitive technique.

As a sensitive probe of absorption, FM spectroscopy is ideal for measurements of trace gases in
the atmosphere and in non-radiating systems. Examples of these include the detection of
molecules and collisionally quenched impurities. In some cases the absorption coefficient is
several GHz, due to Doppler and pressure broadening, making it effective, the more

A quick survey of commercially available FM systems shows that lasers of inherently larger bandwidth
are used. In this work, useful electro-optic materials with a radio frequency and optical waves were
investigated, and the resulting phase velocity mismatch of modulator crystals long enough to
overcome the above problem we have used a waveguide near cutoff to increase the frequency
wave to match that of the laser. As a result, efficient resonant cavity modulators at high
frequencies have been developed. [2]

Using these modulators, we have demonstrated that FM spectroscopy is an ideal tool for
the detection of trace gases in the atmosphere and other non-radiating systems. With a dye laser of
short cavity, it has been possible to obtain line widths of 20 MHz or less. [3] A component equally as critical in
the fast photodetector, which was made by Packard. [4]