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Heterodyne Spectroscopy Using Spectral Spread of Short Laser Pulse

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A heterodyne absorption spectroscopy method which utilizes the spectral spread of a short laser pulse was demonstrated with laser pulses generated by an electrooptic modulator from a frequency-stabilized He–Ne laser. The spectrum of a Michelson interferometer was observed by the method in a spectral bandwidth of approximately 100 MHz.

KEYWORDS: heterodyne detection, heterodyne spectroscopy, absorption spectroscopy, short-pulse laser, laser long-path absorption method

In this letter, we report on an experimental study of a heterodyne laser absorption spectroscopy method which utilizes the spectral spread of a short-pulse laser. We described the concept of the method in our previous paper.[1)] In this method, the waveform of the heterodyne beat signals of the laser pulse before and after passing through the sample is recorded with a high-speed transient digitizer, and the spectra of the pulses are obtained from the power spectra of the signals. The absorption spectrum of the sample which lies within the spectral width of the laser is obtained from the ratio of the power spectrum. This method is potentially useful for laser long-path absorption measurements of atmospheric trace species,[2,3)] as discussed in our previous paper,[1)] because the absorption spectrum can be measured, in principle, with a single laser pulse.

To demonstrate the principle of the measurement method, we carried out an experiment using laser pulses generated by an electrooptic (EO) modulator from a frequency-stabilized He–Ne laser at 633 nm. The spectrum of a Michelson interferometer was measured with a bandwidth of approximately 100 MHz. The experimental setup is shown in Fig. 1. The output of a frequency-stabilized He–Ne laser (Newport Co., NL-1) is directed to an acoustooptic (AO) modulator. The output power of the laser was approximately 1 mW. The first-order beam from the AO modulator was used as the local oscillator for heterodyne detection. The frequency shift was 80 MHz. The zeroth-order beam was directed to an EO modulator, and the pulses with a width of 10–100 ns were generated. A Michelson interferometer with an optical path difference of 8 m was fabricated to demonstrate the spectrum measurement. A spherical mirror was used for the longer arm of the interferometer to match the beams from both arms with the local oscillator beam. A piezoelectric translator (PZT) was used for fine adjustment of the path difference of the interferometer. A silicon PIN photodiode with a 2 GHz response was used as a detector. The diameter of the PIN photodiode was 0.3 mm. The signal from the detector was recorded by a computer-controlled transient digitizer with a sampling rate of 500 MHz. A signal band-pass filter with a band pass of 40–140 MHz was used before the digitizer.

Figure 2 shows the heterodyne beat signals of the laser pulses at three different pulse widths. These signals were taken without the interferometer by blocking the beam in the longer arm. The waveform was recorded with a gate time of 2 ns and a record length of 512 words.

Figure 3 shows the spectra of the three beat signals shown in Fig. 2. The pulse shape and the corresponding spectrum seen in Figs. 2 and 3 have complicated structure when the pulse width is short due to the characteristics of the driver circuit for the EO modulator. We did not attempt to improve the pulse shape, however, because it does not pose a problem for measuring the spectrum of the sample or the interferometer in this experiment.

In the measurement of the spectrum of the interferometer, we used the laser pulse shown in Fig. 2(c), which exhibits the spectrum in Fig. 3(c). The measured spectrum of the interferometer is shown in Fig. 4. One hundred signals were recorded in the measurement of the spectrum of the interferometer. The spectrum of the interferometer was calculated for each pulse by dividing the measured spectrum by the averaged laser spectrum. In Fig. 4, the spectrum of the interferometer averaged over 100 pulses and the standard deviation are shown. The periodic spectral structure which has an interval of 37.5 MHz is seen in Fig. 4. This interval is due to the 8 m optical path difference of the interferometer. We observed the frequency shift of the periodic spectral structure when the path difference was changed by the PZT. The error in the spectrum in-

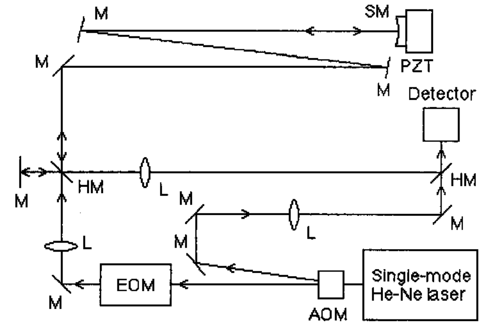

Fig. 1. Experimental setup. M: Mirror, SM: spherical mirror, HM: half-mirror, L: lens, EOM: electrooptic modulator, AOM: acoustooptic modulator, PZT: piezoelectric translator.

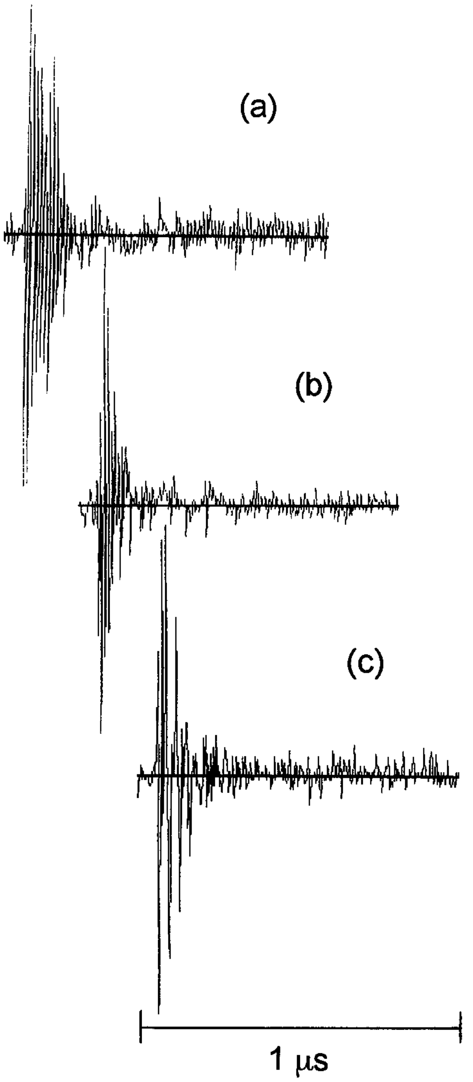

Fig. 2. Laser pulses with different temporal widths generated by the electrooptic modulator.

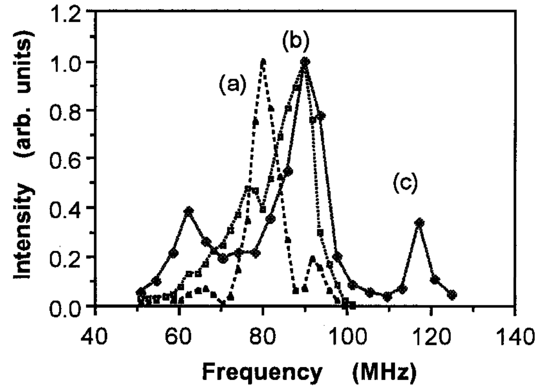

Fig. 3. The spectra of the laser pulses shown in Figs. 2(a)–2(c).

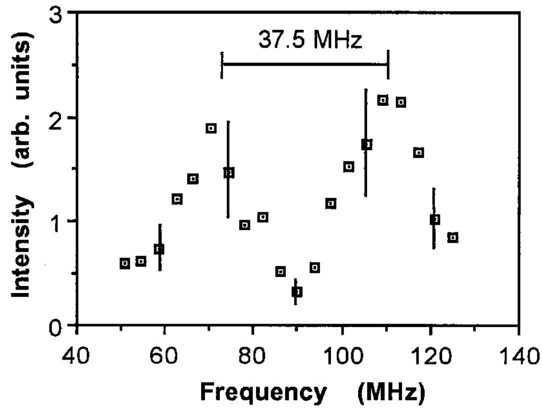

Fig. 4. The spectrum of the interferometer. The periodic structure is due to the 8 m optical path difference of the interferometer.

dicated in Fig. 4 is mainly caused by the small change in the path length due to the vibration of the optics. The change in the frequency of the spectral structure appeared to be random in this experiment because the repetition rate of data aquisition was limited at approximately 1 Hz. However, if we use a signal processor with a high repetition rate, we can analyze the vibration using the shift of the spectrum. In principle, we can measure a spectrum approximately every 1 μs at maximum with this method.

The spectroscopic method described in this paper is potentially useful, for example, for airborne lidar long-path absorption measurements using ground reflection, and for earth-to-satellite or satellite-to-satellite laser long-path absorption measurements.[1] In such applications, however, a much wider spectral bandwidth is required. A bandwidth of several hundred MHz is required for measurements in the stratosphere, and a bandwidth of a few GHz is required for measurements in the troposphere, depending on atmospheric pressure. Aiming at the absorption measurement of low-pressure molecules, we are preparing for an experiment using an infrared single-mode optical parametric oscillator which has a spectral bandwidth of approximately 300 MHz.

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