Inverse Lamb dip spectroscopy using microwave modulation sidebands of CO_2 laser lines

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We report the observation of infrared Lamb dips using a widely tunable laser source. The microwave sidebands generated on CO_2 laser lines provide sufficient power (~ 0.5 mW) and spectral purity ($\Delta\nu \lesssim 100$ kHz) for saturation spectroscopy. Application of this infrared source to the observation of infrared Lamb dip spectra of CH_3F and SiF_4 is reported.

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Until recently, frequency tuned saturation spectroscopy in the mid-infrared region has been limited to cases of accidental coincidences between the center of molecular absorption and the narrow tuning range of the atomic[1] or the molecular[2] lasers, although the use of waveguide lasers increased the tuning range considerably.[3] To overcome this limitation infrared-microwave two-photon spectroscopy[4] and Stark[5] or Zeeman[6] tuning have been exploited in the past. The low power and/or low spectral purity of the available tunable infrared sources, in general, made it difficult to use them for saturation spectroscopy. Only the isolated cases by Patel using a spin-flip laser[7] and by Jennings using the diode laser[8] have been reported. Very recently, color center lasers were used to observe infrared saturation dips.[9] In this letter we report our observation of inverse Lamb dips using microwave modulation sidebands on CO_2 laser lines.

The microwave sidebands on CO_2 laser lines[10] were generated by nonlinear mixing in a CdTe crystal. Infrared sideband radiation with a power of ~ 0.5 mW and spectral purity of $\lesssim 100$ kHz was obtained at the frequencies $\nu_{SB} = \nu_{CO_2} \pm \nu_{Mw}$ and was tunable from 12 to 18 GHz on either side of the CO_2 laser carrier frequency ν_{CO_2}. The use of such a source for Doppler limited spectroscopy has already been reported.[11] Using a 6-mm beam diameter at half-maximum we obtained an average power density of ~ 1.8 mW/cm^2 and a Rabi frequency of $\mu E/h \sim 100$ kHz for a typical vibrational transition dipole moment of 0.15 D. Therefore, we should be able to saturate a gas with a pressure broadening parameter of 20 MHz/Torr up to several mTorr.

Our experimental setup is shown schematically in Fig. 1. The continuous flow CO_2 laser was constructed according to Ref. 12 and its cavity was tuned to the center of the 4.3-μm fluorescence Lamb dip. The fluorescence cell was placed outside the laser cavity and was heated to 150 °C. During the experiment, the laser frequency was not modulated to avoid additional line broadening. The electrooptic modulator[13] consisted of a CdTe crystal ($3.5 \times 3.5 \times 50$ mm) enclosed in a brass housing which acts as the microwave waveguide. We operated the modulator in the traveling-wave mode and obtained 0.5 mW of sideband power for 3 W of incident laser power and 20 W of microwave power. The high power, high spectral purity microwave radiation was provided by a Hewlett–Packard 8672A synthesizer sweeper followed by a Hughes model 1277H traveling-wave amplifier.

The carrier and its sidebands emerge collinearly from the CdTe crystal but they are perpendicularly polarized. They traversed an absorption cell of 1.8-m length and were reflected back slightly tilted with respect to the incoming beam. They were then directed into a 0.5-m Jarrell-Ash grating monochromator which was tuned to the desired sideband frequency. At the exit of the monochromator a rotary grid polarizer was placed in order to further discriminate the residual laser carrier from the desired orthogonally polarized sideband radiation. The signal is then detected by a HgCdTe detector followed by a battery operated preamplifier with a voltage gain of ~ 800. In order to increase the sensitivity of detection, Stark modulation at a few kHz with phase sensitive detection was employed. A pair of Stark electrodes separated by 1 cm was placed in the absorption cell such that the sideband electric field and the Stark field were perpendicular to each other.

We first studied the well established ν_3 band of CH_3F.[14] An example of the $\nu_3\ {}^qR\,(4,3)$ transition is shown in Fig. 2. Figure 2(a) show the Doppler broadened line profile with the inverse Lamb dip at the center. In this "low" resolution spectrum the microwave synthesizer sweeper was stepped at 250-kHz increments and 1f phase sensitive detection was used with a 13-V dc biased 26-Vpp Stark modulation. Figure 2(b) shows a higher resolution, 20-MHz scan with steps of 25

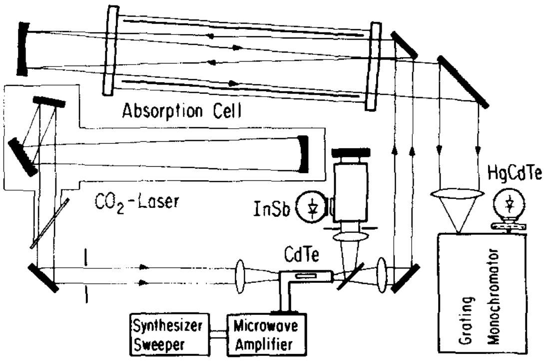

FIG. 1. Experimental setup: typical CO_2 laser output power was 3 W providing about 0.5 mW of sideband power at a microwave drive power level of 20 W.

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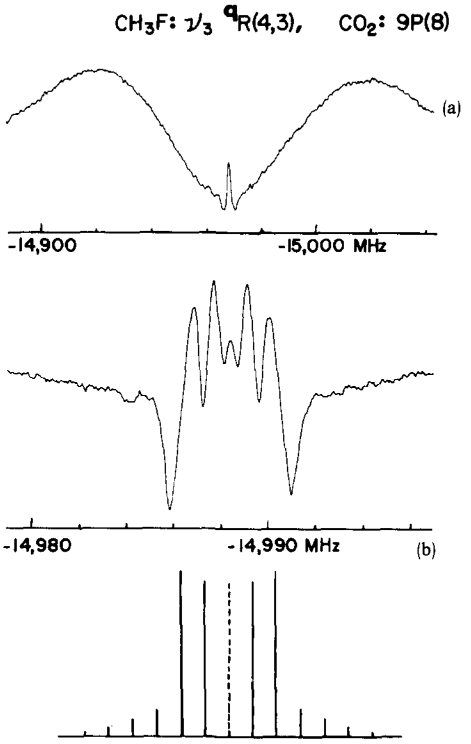

FIG. 2. Stark-modulated spectrum of the ν_3 ^qR (4, 3) transition of CH_3F. The CO_2 laser oscillates at line 9P(8). The microwave-modulation frequency is indicated on the abscissae. (a) Doppler-broadened profile and Lamb dip. (b) Stark splitting (E_{dc} = 22 V/cm) and theoretical line pattern.

kHz. For this result the dc Stark voltage was increased to 22 V and the ac voltage was reduced to 8 Vpp. The calculated Stark pattern is shown under the trace. Clearly resolved are the four strongest components each of which consists of two overlapping Lamb dips and are separated by ~1 MHz. The central component, composed of two overlapping transitions ($M = \pm 3 \leftarrow \pm 2$), is not Stark modulated and is missing. We believe the weak central feature is due to a collision-induced center dip associated with the collisional reorientation of CH_3F.[14–16]

An example of wider frequency tuning is the SiF_4 absorption spectrum shown in Fig. 3. Here the CO_2 laser was tuned to the 9P(38) line and the microwave was tuned over about 600 MHz in the vicinity of the P(11) transition in the ν_3 fundamental of SiF_4.[17] A dc Stark bias of 60 V, an ac Stark modulation of 40 Vpp, and a sample pressure of 10 mTorr were used. Although SiF_4 is a tetrahedral molecule and has no permanent dipole moment in the ground state, the vibrationally induced dipole moment in the triply degenerate upper state[18] makes this transition sensitive to Stark modulation. More than 20 Lamb dips are seen on Fig. 3 which correspond to either the double parity E component or the closely spaced pairs of $A_1 - A_2$ or $F_1 - F_2$ components which have the first order or near first order Stark effects. The small area of crowded lines is expanded in the lower trace of Fig. 3. The half-width at half-height of the strongest feature measures ~200 kHz. The high resolution spectrum will be useful for more detailed assignment of this band and other hot bands and for measuring the vibrationally induced dipole moment.

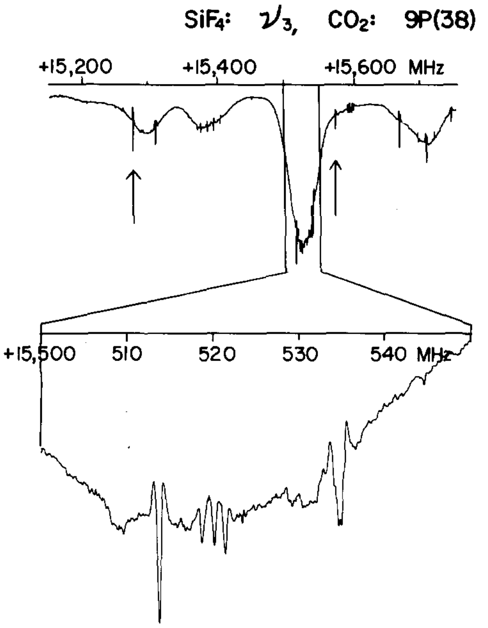

FIG. 3. Part of the ν_3 P branch of SiF_4: The P(11) E lines are indicated by arrows; the usefulness of laser sideband radiation for resolving crowded spectra is clearly demonstrated.

In conclusion, we have observed saturation Lamb dips in the infrared region using a widely tunable source. This opens up the possibility to transfer the high resolution and absolute frequency accuracy of microwave spectroscopy to a substantial part of the mid-infrared region. Although at the moment the resolution is of the order of 100 kHz because of the power broadening, pressure broadening, and laser instability, we anticipate improving this resolution by an order of magnitude in the near future.

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