

Lamb Dip Spectroscopy Using Tunable Laser Sidebands

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So far nonlinear spectroscopy in the IR has been limited to accidental coincidences or near coincidences between laser lines and molecular absorption [1,2], although the use of CO₂ waveguide lasers increased the tuning range considerably [3]. The low power and/or low spectral purity of the available tunable sources, in general, made it difficult to use them for saturation spectroscopy. We report on a widely frequency-tunable infrared source which has sufficient spectral purity and output power for saturation spectroscopy.

The tunable infrared radiation is generated by electrooptically mixing CO₂ laser radiation with 12-18GHz microwave radiation in a CdTe crystal [4]. Using 3W of CO₂ laser power and 20W of microwave power, we have generated sidebands at frequencies $\nu_{\text{CO}_2} \pm \nu_{\text{MW}}$ with $\sim 0.5\text{mW}$ power [5]. With a beam diameter of 6mm, the intensity was sufficient to saturate CH₃F, NH₃, SiH₄, and SiF₄ at a few mTorr pressure. In the spectroscopic experiments described here, we employed Stark modulation with phase sensitive detection to increase sensitivity. The Stark field was oriented perpendicular to the electric field of the sideband radiation.

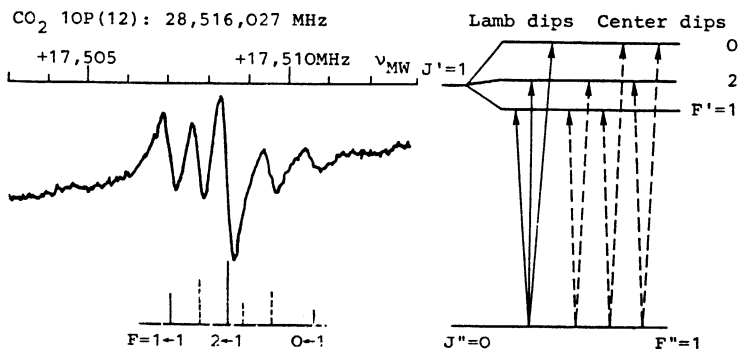


Fig.1: ¹⁴N hyperfine splitting of ν_2 saR(0,0) transition of NH₃

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As an example, the observed hyperfine structure in the ν_2 asR(0,0) transition of NH_3 is shown in Fig.1, together with the energy level diagram. The hyperfine splitting is produced only in the excited vibrational state. A corresponding observation of the ground level hyperfine splitting in the ν_2 asP(1,0) transition enables us to determine the variation of eqQ due to vibrational excitation.

Although tetrahedral molecules like SiH_4 do not have a permanent dipole moment, excitation of a triply degenerate vibration (ν_3, ν_4) induces a small dipole moment on the order of a few hundredths of a Debye. This causes a first order Stark effect on double-parity E levels which can be observed by Lamb-dip spectroscopy. The first observation of this effect was made by LUNTZ and BREWER [6] using the coincidence between the P(7) line of CH_4 and the $3.39\mu\text{m}$ line of the HeNe laser. We used our laser sideband method for the observation of such minute effects in the ν_4 band of SiH_4 and in the ν_3 band of SiF_4 . The observed first order Stark patterns are shown in Fig.2 together with the calculated line positions. The scanings of the infrared frequency are 20MHz wide. From the measurement of the splitting we are able to determine the vibration-induced dipole moment.

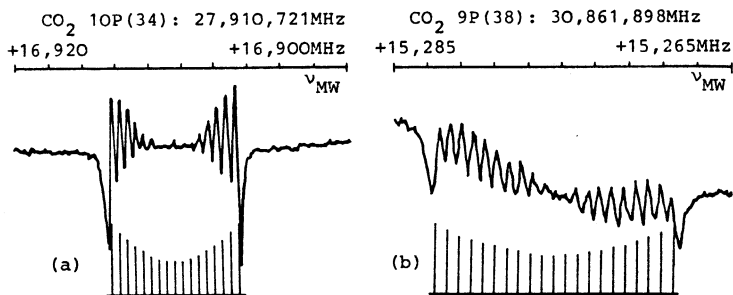


Fig.2: Stark splitting of (a) ν_4 R(7)E transition of SiH_4 and (b) ν_3 P(11)E₁ transition of SiF_4 . DC Stark fields were (a) 2kV/cm and (b) 700V/cm

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