HIGH-PRECISION MILLIMETER-WAVE SPECTROSCOPY WITH THE LAMB DIP*

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Use of the Lamb dip to circumvent Doppler broadening has allowed microwave measurement of rotational lines in the 1-3-mm wave region to accuracies of 1 part in 10^8. Rotational constants of OCS and CH_3F are improved in accuracy by two orders of magnitude.

In a theoretical treatment of the properties of the optical laser Lamb [1] showed that power saturation of a Doppler-broadened line could produce a sharp dip at the exact center of the line if other broadening factors were small in comparison with Doppler broadening and if the radiation passed linearly in opposite directions through the gas.

With a high-Q, Fabry-Perot cavity we have observed the Lamb dip in Doppler-broadened absorption lines occuring in the 1-3-mm wave region. We found that the secondary broadening effects can easily be made small as compared with Doppler broadening and that Lamb dips of only a few kilocycles can be achieved in Doppler widths of more than a megacycle. Thus a considerable improvement in accuracy of alignment of a frequency marker to the line center can be achieved by use of the Lamb dip in the millimeter and submillimeter regions where Doppler widths, which increase directly as the line frequency, become large.

An illustration of the Lamb dip effect, fig. 1, shows the $J = 15 \rightarrow 16$ rotational line of carbonyl sulfide (OCS) centered in the cavity mode and the presence of the small, sharp dip at the center of the Doppler contour. Examples of measurements of high-J rotational transitions of OCS and CH_3F with the Lamb dip are given in tables 1 and 2. From these measurements the rotational constants, also given in these tables, are improved in accuracy by about two orders of magnitude over previously known values.

The Lamb dip method also provides a considerable improvement in resolution capabil-

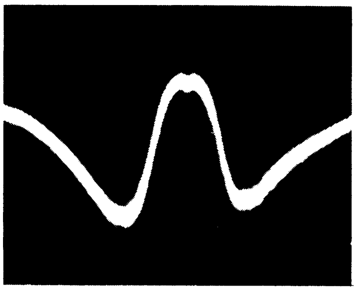

Fig. 1. Lamb dip in the $J = 15 \rightarrow 16$ rotational line of OCS at 194 586 MHz. The broad downward curve is the cavity resonance; the broad upward curve is the Doppler broadened line (Doppler width about 310 kHz); the small sharp depression at the center of the Doppler line is the Lamb dip (width about 10 kHz).

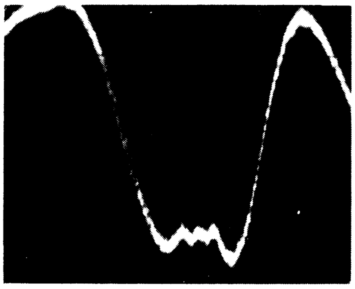

Fig. 2. The $J = 15 \rightarrow 16$ transition of $^{35}ClCN$ showing the closely spaced Lamb dips of hyperfine components with separation of 50 kHz. The Doppler width of the line is approximately 320 kHz.

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Table 1
Frequencies and molecular constants of $^{16}O^{12}C^{32}S$

J (a)	Frequencies in MHz Measured	Calculated	Difference
8	109 463.063	109 463.064	- 0.001
9	121 624.638	121 624.638	0.000
10	133 785.900	133 785.900	0.000
11	145 946.821	145 946.818	+ 0.003
12	158 107.360	158 107.361	- 0.001
13	170 267.494	170 267.498	- 0.004
14	182 427.198	182 427.198	0.000
15	194 586.433	194 586.433	0.000
16	206 745.161	206 745.161	0.000
19	243 218.040	243 218.042	- 0.002
20	255 374.461	255 374.460	+ 0.001

Molecular constants in MHz

$B_0 = 6081.49205 \pm 0.0002$; $D_J = (1.3008 \pm 0.0006) \times 10^{-3}$;
$H_J = (-0.85 \pm 0.8) \times 10^{-9}$.

(a) Rotational quantum number for lower level.

Table 2
Frequencies and molecular constants of $^{12}CH_3{}^{19}F$.

J(a)	K	Frequencies in MHz Measured	Calculated	Difference
1	0	102 142.666	102 142.668	- 0.002
	1	102 140.911	102 140.908	+ 0.003
2	0	153 210.399	153 210.398	+ 0.001
	1	153 207.758	153 207.759	- 0.001
	2	153 199.843	153 199.843	0.000
3	0	204 273.779	204 273.780	- 0.001
	1	204 270.263	204 270.263	0.000
	2	204 259.716	204 259.716	0.000
	3	204 242.148	204 242.148	0.000

Derived constants in MHz

$B_0 = 25\,536.1466 \pm 0.0015$; $H_J = (-0.0107 \pm 0.004) \times 10^{-3}$;
$D_J = 0.05987 \pm 0.0002$: $H_{JK} = (0.020 \pm 0.010) \times 10^{-3}$;
$D_{JK} = 0.44027 \pm 0.0004$: $H_{KJ} = (0.032 \pm 0.020) \times 10^{-3}$.

(a) Rotational quantum number for lower level.

ity of millimeter wave spectroscopy. In fig. 2 are shown three resolved dips of hyperfine components of the $J = 15 \rightarrow 16$ transition of $^{35}ClCN$ separated by only 50 kHz. With G. Winnewisser we have measured the previously unresolved splitting in the $^{R}Q_0$ transitions of the ground vibrational state of H_2S_2 (to be published). The observed splitting of 120 kHz due to relative rotations of the SH group via barrier-tunneling indicates a splitting of only 60 kHz in the rotational levels.

Although the Lamb dip has been widely observed in laser emission lines, its application in absorption spectroscopy has only recently been considered. Brewer, et al. [2] have used it with a laser source to measure Stark splitting of NH_3 lines in the infrared region, and Costain [3] has observed it in the lower frequency microwave region. As far as we know, our work represents the first use of this effect in the 1-mm wave region. The greatest advantage from the effect should be in the shorter millimeter and submillimeter wave regions, where Doppler broadening is the primary limiting factor in resolution in resolution and measurement of gaseous absorption lines. We have demonstrated that with a high-Q Fabry-Perot cavity the power from our harmonic generators is adequate to produce the required saturation even for the CO molecule [4] with the very small dipole moment of 0.11 D and hence a relatively small transition coefficient.

References

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[2] R. Brewer, M. Kelly and A. Javan, Phys. Rev. Letters 23 (1969) 559.
[3] C. C. Costain. Can. J. Phys. 47 (1969) 2431.
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