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NOTE

Sub-Doppler Frequency Measurements of $^{15}NH_3$ Laser Transitions[1]

The ammonia laser is important in the 11–13 μm region of the mid-infrared. For instance, it can be used as a radiation source for a laser Stark spectrometer, a laser magnetic resonance (LMR) spectrometer, and a tunable far-infrared (TuFIR) spectrometer (*1–4*). It is also a candidate for a secondary frequency standard when stabilized to cooled ions such as Ba^+ (*5*). The rich absorption spectrum of the ammonia molecule is a useful calibration standard for Fourier transform infrared and lead-salt diode laser spectrometers (*6*).

We have recently constructed a TuFIR spectrometer in which far-infrared (FIR) radiation is generated by mixing CO_2 laser radiation and $^{15}NH_3$ laser radiation (*7*). In this note, we measure the frequencies of the $aP(4,0)$, $aP(4,3)$, $aP(5,3)$, $aP(6,0)$, and $aP(6,3)$ transitions of $^{15}NH_3$ to determine the frequency of the generated FIR radiation. We also compared these frequencies with the previous measurements by Siemsen and Reid (*8*).

The experimental system for frequency measurement is shown in Fig. 1. The $^{15}NH_3$ laser is longitudinally pumped by the $R(42)_I$ line of a CO_2 laser. The pump power is about 18 W. The $^{15}NH_3$ laser is line tunable to the $aP(4,0)$, $aP(4,3)$, $aP(5,3)$, $aP(6,0)$, and $aP(6,3)$ lines by means of a grating. Typical power is 300–600 mW. More details of this laser can be found elsewhere (*9*). After passing through a CH_3OH cell to block the residual CO_2 pump radiation, the ammonia laser output power is split in half. The first half is used to stabilize the ammonia laser; the second half is used to measure the frequency.

The first half is double passed through a 17-cm-long absorption cell filled with $^{15}NH_3$ gas at a pressure of about 1.3 Pa (10 mTorr), and a saturation-dip signal is observed with a HgCdTe detector. Typical incident power is 150–300 mW and the average beam diameter is ~5 mm. Figure 2 shows both the laser output power profile and the laser power after double passing through the $^{15}NH_3$ cell as a function of the laser frequency for the $aP(4,0)$ line. For frequency stabilization, we frequency-modulated the ammonia laser by applying a 450-Hz ac voltage to the PZT (piezoelectric transducer) on the end mirror and demodulated the absorption signal in a lock-in amplifier using the third harmonic of the modulation frequency as the reference frequency. The resulting signal approximates the third derivative of the saturation dip. The ammonia laser is then locked to the zero crossing of this $3f$ signal using a servo system (*10, 11*).

The rest of the ammonia laser radiation (frequency f) is mixed in a W–Ni metal–insulator–metal (MIM) diode with the radiation from two reference CO_2 lasers (f_1 and f_2) and a microwave sweeper (f_3). The reference CO_2 lasers are locked to the saturation dip of 4.3-μm fluorescence signals in 6.0-Pa (45-mTorr) CO_2 cells using conventional $1f$ servo techniques (*12*). Fifth-order beat notes of $2f_1 - f_2 - f_3 - f$ were observed on a spectrum analyzer, and their frequencies were measured with a calibrated signal generator. The beat frequency was measured 10 times, and the average of these measurements is reported in Table 1 with the 1σ statistical error. The $^{15}NH_3$ laser frequencies (f) are then calculated using the CO_2 laser frequencies in Ref. (*13*). The frequency uncertainties of these CO_2 lasers are ~10 kHz (*14*). The total uncertainty of the $^{15}NH_3$ laser frequency is calculated from the quadratic sum of the uncertainties of the beat frequency and the CO_2 laser frequencies. For comparison, we also measured the frequency with the $^{15}NH_3$ laser locked using the conventional $1f$ servo technique, and results are within 300 kHz.

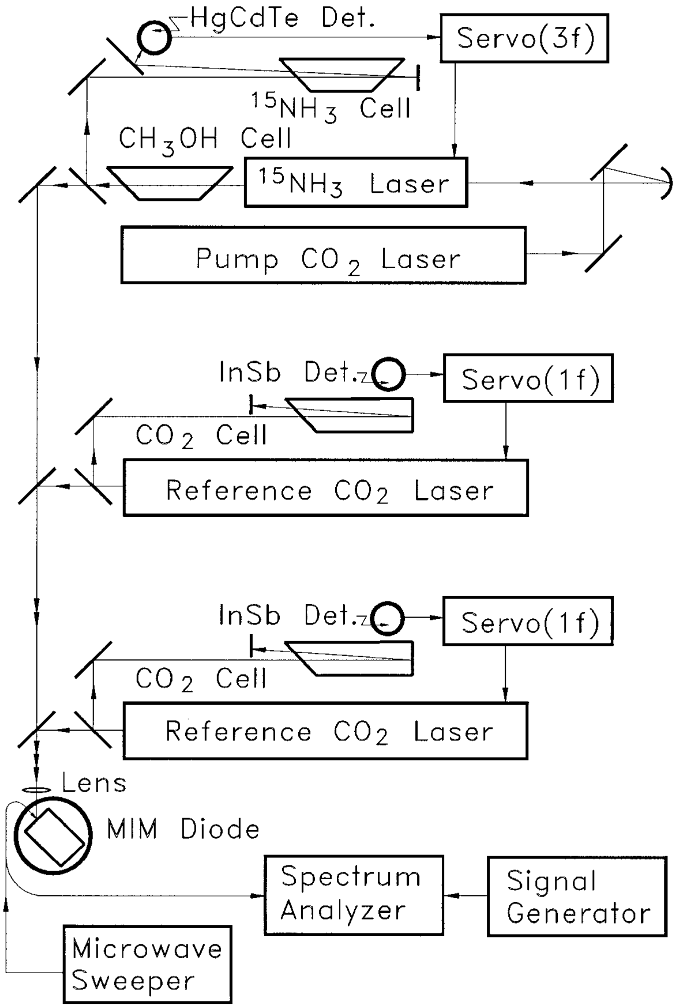

FIG. 1. Experimental setup for frequency measurement.

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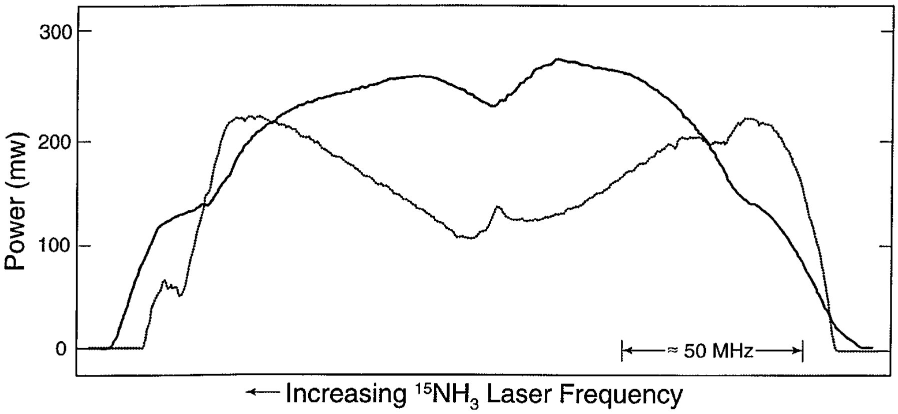

FIG. 2. Typical gain profile of the aP(4,0) line and the absorption spectrum of $^{15}NH_3$. The $^{15}NH_3$ laser power is about 500 mW and the pressure of $^{15}NH_3$ is about 1.3 Pa (10 mTorr). The output power profile is the solid line and the laser power after double passing through the $^{15}NH_3$ cell is the dotted line.

The *aR*(2,0) absorption transition frequency of $^{15}NH_3$ is 53 MHz higher than the center of the $R(42)_I$ CO_2 laser pump line (*8*). Although the lasing frequency of the pump CO_2 laser is upshifted by tuning the PZT on the end mirror, an offset of ≈30 MHz still remains. Since the $^{15}NH_3$ molecules with a nonzero velocity are selectively pumped to the upper level of the lasing transition, two peaks corresponding to this off-center pumping appear in the ammonia laser output power profile from the Doppler effect. These peaks are asymmetric, as shown in Fig. 2. For the *aP*(4,0) line, an additional Raman effect partly explains this asymmetry, because the pumped *aR*(2,0) transition and the lasing *aP*(4,0) transition form a three-level system (*15, 16*). However, we found this asymmetry in every laser line, including collisionally induced laser lines without Raman type transitions; hence, this asymmetry is probably caused by effects such as cavity misalignment.

The slope superposed on the saturation dip in Fig. 2 shifts the center of the saturation-dip signal toward lower frequencies. We numerically calculated the deviation between the true center of the dip and the zero crossing of its third-derivative signal under various conditions of the asymmetric output power profile, the depth of the saturation dip, and the modulation amplitude for stabilization. From these calculations, we estimated the residual systematic error in the 3*f* servo-lock to be less than 100 kHz, and within our measurement reproducibility of 100–150 kHz. The 3*f* servo-lock effectively eliminates the systematic frequency shift due to the asymmetric output power profile of our ammonia laser.

Our measured frequencies differ by a few megahertz from the measurements by Siemsen and Reid (*8*) beyond their estimated 1σ uncertainty of 0.5 MHz. This difference is close to the full linewidth of the saturation dip. Siemsen and Reid pumped their $^{15}NH_3$ laser at the center of the *aR*(2,0) transition of $^{15}NH_3$ by shifting the pump CO_2 laser frequency with an AOM (acoustooptic modulator), so that they obtained a more symmetric laser output power profile with a single peak. However, they made frequency measurements by monitoring the first-derivative signal of the saturation dip in the absorption spectrum and manually tuning the ammonia laser to the dip's center. On the other hand, we lock the ammonia laser with the servo system. Although we off-center pump our $^{15}NH_3$ laser without an AOM to provide more ammonia laser power for a TuFIR spectrometer where typical power of ~150 mW is needed for various lines, we believe that the systematic error due to the asymmetric output power profile with two peaks shown in Fig. 2 has been effectively reduced, using the third-derivative signal of the saturation dip as an error signal for the servo-lock. Further examination of this discrepancy between two measurements is needed.

TABLE 1
Measured Frequencies of $^{15}NH_3$ Laser Transitions

Laser line				Frequency (MHz)		
$^{15}NH_3$	ref. CO_2 (f_1)	ref. CO_2 (f_2)	MW (f_3)	beat($2f_1-f_2-f_3-f$)	$^{15}NH_3$ laser (f)	$f_s{}^a-f$
aP (4,0)	$R(6)_I$	$R(26)_{II}$	2620	310.29 (12)[b]	25 485 558.90 (13)[c]	0.90
aP (4,3)	$P(24)_I$	$P(34)_{II}$	0	215.59 (10)	25 410 438.87 (11)	−0.17
aP (5,3)	$P(24)_I$	$P(14)_{II}$	−2080	−313.07 (8)	24 852 209.40 (9)	−1.40
aP (6,0)	$P(36)_I$	$P(22)_{II}$	−2380	304.40 (13)	24 375 598.22 (14)	−3.02
aP (6,3)	$P(40)_I$	$P(28)_{II}$	−2230	−311.44 (13)	24 303 078.12 (14)	−1.92

[a] f_s denotes the previous frequency by Siemsen and Reid.
[b] The numbers in parentheses are the estimated 1σ uncertainties in units of the last quoted digits.
[c] These uncertainties include those of CO_2 laser frequencies.

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