NASA

Technical Memorandum 79550

Diode Laser Saturation Spectroscopy of NH_3

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MAY 1978

National Aeronautics and
Space Administration

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DIODE LASER SATURATION

SPECTROSCOPY OF NH_3

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ABSTRACT

Saturation of molecular transitions using a tuneable diode laser has been demonstrated for the first time using a standing-wave cavity configuration with an f/8 beam focussed at the sample. Observed saturation effects in NH_3 transitions near 888 cm^{-1} include sub-Doppler (Lamb-dip) resonances at line center.

This *Letter* reports the first demonstration of molecular line saturation using a tuneable diode laser (TDL). Saturation effects on gas samples subjected to intense laser fields are familiar as sources of information concerning, for example, precise line positions, homogeneous lineshapes, and rotational relaxation rates[1], but heretofore these studies have required the use of gas lasers to achieve the necessary power densities. The molecular transitions which could be subjected to saturation were therefore limited to those which occur in coincidence with the discrete frequencies of gas lasers, or which could be shifted into coincidence using the Stark effect.

The aim of the work described here was to show that commercially produced TDL's are available which have sufficient output power to saturate strong molecular transitions. Since Pb-salt TDL's can be fabricated to operate anywhere in the 3-30μm wavelength region the important implication of these results is that saturation spectroscopy should now be possible throughout a large portion of the infrared which includes the fundamental vibrational bands of most molecules. At this laboratory we are particularly interested in utilizing sub-Doppler saturation resonances (Lamb-dips[2]) to lock the output frequency[3] of a tuneable diode laser in heterodyne applications.[4]

Figure 1 is a schematic of the optical arrangement used here. The TDL is mounted on a closed-cycle refrigerator and is shock-isolated as described elsewhere.[5] Off-axis parabolas P_1 and P_2 are f/2 and f/8 respectively. Sphere S_1, in combination with P_1,P_2, and the cleaved face of the diode laser, form a standing-wave cavity. The cavity is tuned with the aid of a pinhole placed at the sample focus, with the cell offset from the focus. The power and beam waist in the focal region can be varied by adjusting the aperture diameter. Minimum beam waist with this optical arrangement is on the order of 10^{-2} cm diameter. A ZnSe window directs a small portion (~17%) of the returned beam to sphere S_2 to be focused at the detector. The detector is a HgCdTe photodiode with an element area of 10^{-4} cm^2, so that only the central region of the focussed beam is sampled. For part of this work a grating monochromator was placed between S_2 and the detector, permitting laser mode isolation at the expense of image aberration.

The TDL was obtained from Laser Analytics, Inc. and has a measured power output in the f/2 beam of 1.5 mW. Approximately, one-half of this power is in the mode used at 888 cm^{-1}, producing an estimated intensity at the sample focus of about 5 W cm^{-2}. With the cavity tuned the circulating power can reach many times this value.

The three ν_2 [sP(4,K)] transitions of NH_3 near 888 cm^{-1} were used to demonstrate saturation. Preliminary attempts with the cavity tuned showed that the entire pressure-broadened profiles of the lines at several Torr could be reduced in absorption, a clear indication of saturation over the entire line profile. Figure 2 shows this effect with 1 Torr NH_3 in a 10 cm cell. With the cavity detuned and the beam power reduced (small aperture diameter) the lines were all greater than 94% absorbing at center. With full power and the cavity tuned the absorption of the sP[4,2] line, for instance, was reduced to approximately 50% at line center.

To observed a sub-Doppler resonance the power density at the sample focus was reduced by narrowing the aperture to 1.2 cm diameter and adjusting S_1 to slightly detune the standing-wave cavity. The intensity in the sample focal region was estimated to be approximately 1 W cm^{-2}. A monochromator was placed between S_2 and the detector to isolate the single TDL output mode at 888 cm^{-1}. The NH_3 was placed in a 3 cm length cell at 1 Torr pressure, The resonance as observed in the sP(4,3) absorption line is shown in **Figure** 3. Apparent frequency and intensity instabilities of the TDL caused some distortion of the line and resonance profiles. These instabilities were presumably due to optical feedback coupling at the laser. At 25°C the Doppler width FWHM of the line is 80 MHz and the pressure-broadened homogeneous width is 46 MHz[6], thus the Voigt width should be 92 MHz. Allowing for distortion of the lineshape, these are in rough agreement with the observed widths.

Extremely narrow resonances at low sample pressure (0.1 Torr) could not be observed consistently, again because of laser instabilities. For the purpose of locking the frequency of a TDL to the center of a saturation resonance a refined optical system will be necessary, including an optical feedback isolator. Improved stabilization of the TDL current and temperature may also be required.

I would like to acknowledge the collaboration of Dr. J.J. Hillman in developing the TDL instrumentation, and to thank both he and Dr. M.J. Mumma for stimulating discussions and encouragement. Thanks also to J.L. Faris for technical assistance.

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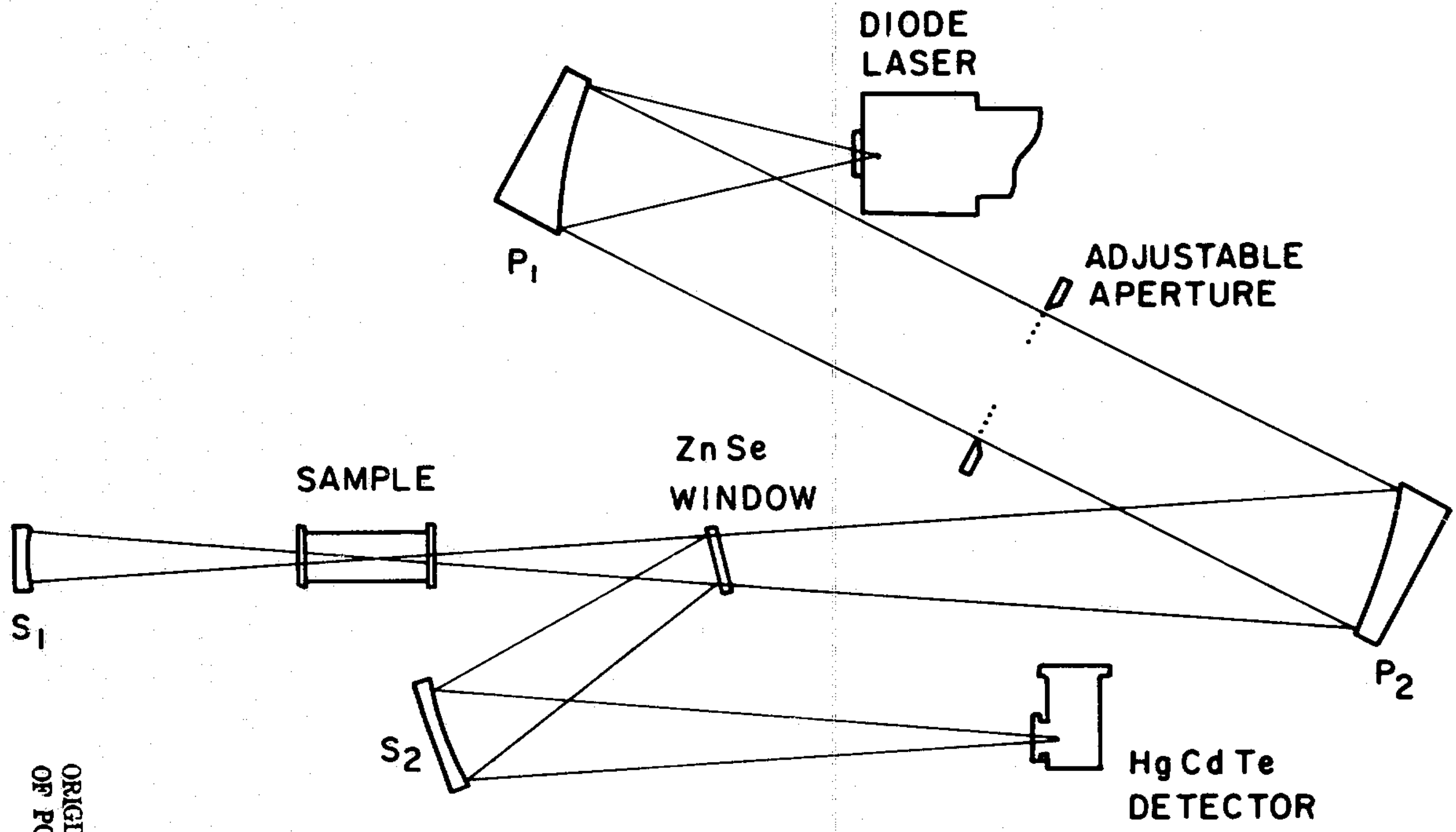
Figure Captions

FIG. 1. Optical arrangement for observing saturation with a TDL. Mirrors P_1 and P_2 are off-axis parabolas. S_1 and S_2 are spheres. The combination S_1, P_2, P_1, and the cleaved face of the TDL form a standing-wave cavity.

FIG. 2. Saturation of all three $\nu_2[sP(4,K)]$ transitions in NH_3. From left to right the lines are K = 1,2, and 3, respectively. The two traces show the effect of subjecting the sample to full beam power with a tuned cavity. The monochromator was in place for these scans.

FIG. 3. Sub-doppler saturation resonance as observed at the center of the $\nu_2[sP(4,3)]$ line of NH_3 in absorption at 887.878 cm^{-1}. The displayed signal is the DC amplified, direct output of the detector, during a single sweep of the TDL current. Pressure 1 Torr. Path 3 cm. Sweep scale 2 ms/div. Zero transmission is at the bottom of the screen.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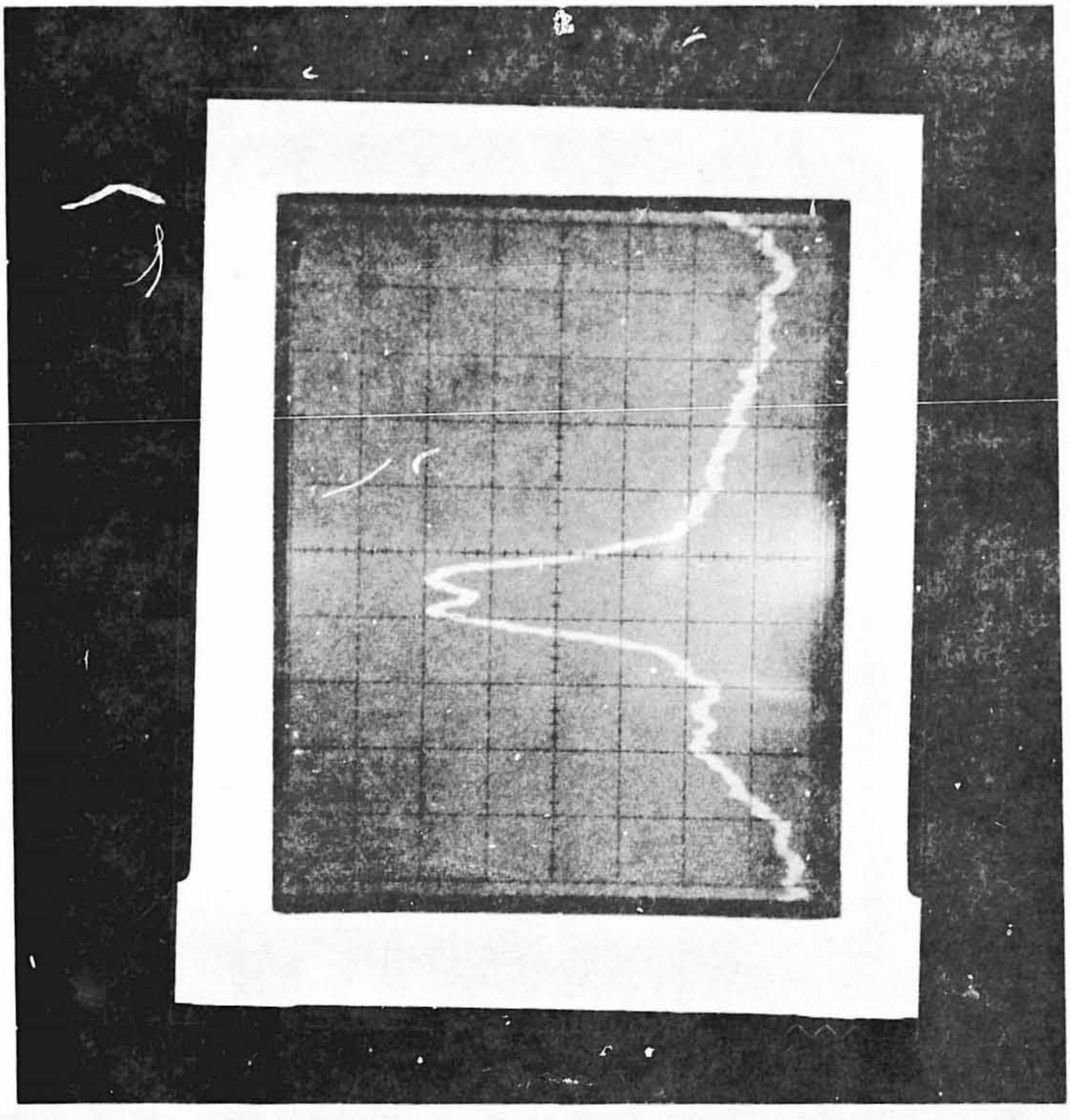

BIBLIOGRAPHIC DATA SHEET

1. Report No.	2. Government Accession No.	3. Recipient's Catalog No.

4. Title and Subtitle	5. Report Date
Diode Laser Saturation Spectroscopy of NH_3	6. Performing Organization Code
7. Author(s) Donald E. Jennings	8. Performing Organization Report No.
9. Performing Organization Name and Address Infrared and Radio Astronomy Branch Laboratory for Extraterrestrial Physics NASA/Goddard Space Flight Center Greenbelt, MD 20771	10. Work Unit No. 11. Contract or Grant No. 13. Type of Report and Period Covered
12. Sponsoring Agency Name and Address	14. Sponsoring Agency Code

15. Supplementary Notes

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17. Key Words (Selected by Author(s))	18. Distribution Statement

19. Security Classif. (of this report)	20. Security Classif. (of this page)	21. No. of Pages	22. Price*

*For sale by the National Technical Information Service, Springfield, Virginia 22151.

GSFC 25-44 (10/77)