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Saturation Processes in Doppler-Broadened HF Vibrational Transitions

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The frequency dependent shapes of Doppler-broadened HF vibrational transitions in the presence of a frequency stabilized, single-mode, saturating radiation field were experimentally determined. It was found that the transitions saturate homogeneously with no effective hole burning observable.

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FIGURES

I. INTRODUCTION

In a previous investigation,[1] we determined the absorption line profiles and the anomalous dispersion of HF gas at pressures for which the vibrational transitions were Doppler broadened. We were able to show that both quantities corresponded to values predicted by standard Doppler-line theory. The present investigation was originally an attempt to extend these measurements to the gain lines of an active, vibrationally inverted medium of a supersonic HF laser. In the study, an additional attempt was also made to measure gain and anomalous dispersion while the transitions were saturated.

To permit a clear interpretation of the experimental results, it was decided to measure the gain-line profiles when they were saturated by an external, single-mode radiation field that passed the medium in one direction only, i.e., the active medium was used as a single-pass amplifier. The pressure of the active medium was maintained at 6 Torr, a value at which standard theory predicts that the lines would be Doppler broadened and pressure broadening would be an order of magnitude smaller.[2]

The investigation concentrated on the $P_2(8)$ line for technical reasons: negligible atmospheric absorption, relatively high gain both in the supersonic laser medium and in the saturating laser oscillator, and no absorption by expended ground-state HF molecules that are always present in the amplifier medium.[3]

As a result, the magnitude of the anomalous dispersion was much smaller than expected, and in fact, it proved impossible to extract meaningful dispersion profiles as a function of frequency from the relatively high noise produced by the amplifier medium. Because we suspected that the reason for this result was connected with the saturation behavior of the transition, we concentrated our investigation on a study of saturated gain-line profiles.

The surprising result was that the investigated HF transition saturated homogeneously under the operational conditions of our supersonic diffusion laser. The amplifier gain line was found to be broadened, which we attribute to source flow effects.[4] Because of this large line broadening we were not able to investigate the line wings and answer the question as to whether the line profile was of Doppler or Lorentzian shape.

II. EXPERIMENTAL APPARATUS

A schematic of the apparatus used in our experiments is indicated in Fig. 1. The investigated active medium was produced by the Aerospace supersonic HF cw laser, which was driven by an arc heater and equipped with a standard 36-slit nozzle.[3] The nozzle produced an active medium of 18 cm width and 2.5 cm height normal to the flow direction. The laser, its flow field, and operating parameters have been exhaustively described elsewhere.[3] We used the laser medium exclusively as an amplifier, so that it was possible to replace the Brewster windows of its oscillator configuration with flat, uncoated calcium fluoride window plates. The windows were not flushed with an inert gas, which permitted operation of the test section at the reduced pressure of 6 Torr.

The amplifier was illuminated by two cw HF lasers that probed the active medium from opposite sides, 4 mm downstream of the slit nozzle exit. The beams from the two probe lasers were arranged concentrically in the amplifying medium, so that both interacted with the same molecules of the medium. In one of the two probe lasers a power density sufficient for saturation was produced; the other laser, of much lower power, which was swept in frequency, served to determine the frequency dependent gain-profile of the investigated transition. The amplification experienced by the low-power probe beam was measured by two uncooled, high-frequency InAs detectors, one before and one after the beam passed the test section.

In order to distinguish the low-power gain signal from the colinear high-power saturating beam, the two beams were polarized orthogonally to each other, which enabled us to separate them with the aid of Glan-Thompson prisms before and after the test section. The prisms were made to our specifications by Karl Lambrecht[5] from single crystals of Rutile (TiO_2). Their rejection ratio, discriminating between S and P polarization, was at least 1/1000 at 2.8 μm.

The laser used as the source of the saturating power was of the type described in Ref. 6. It operated on SF_6, He, O_2 mixtures dissociated by

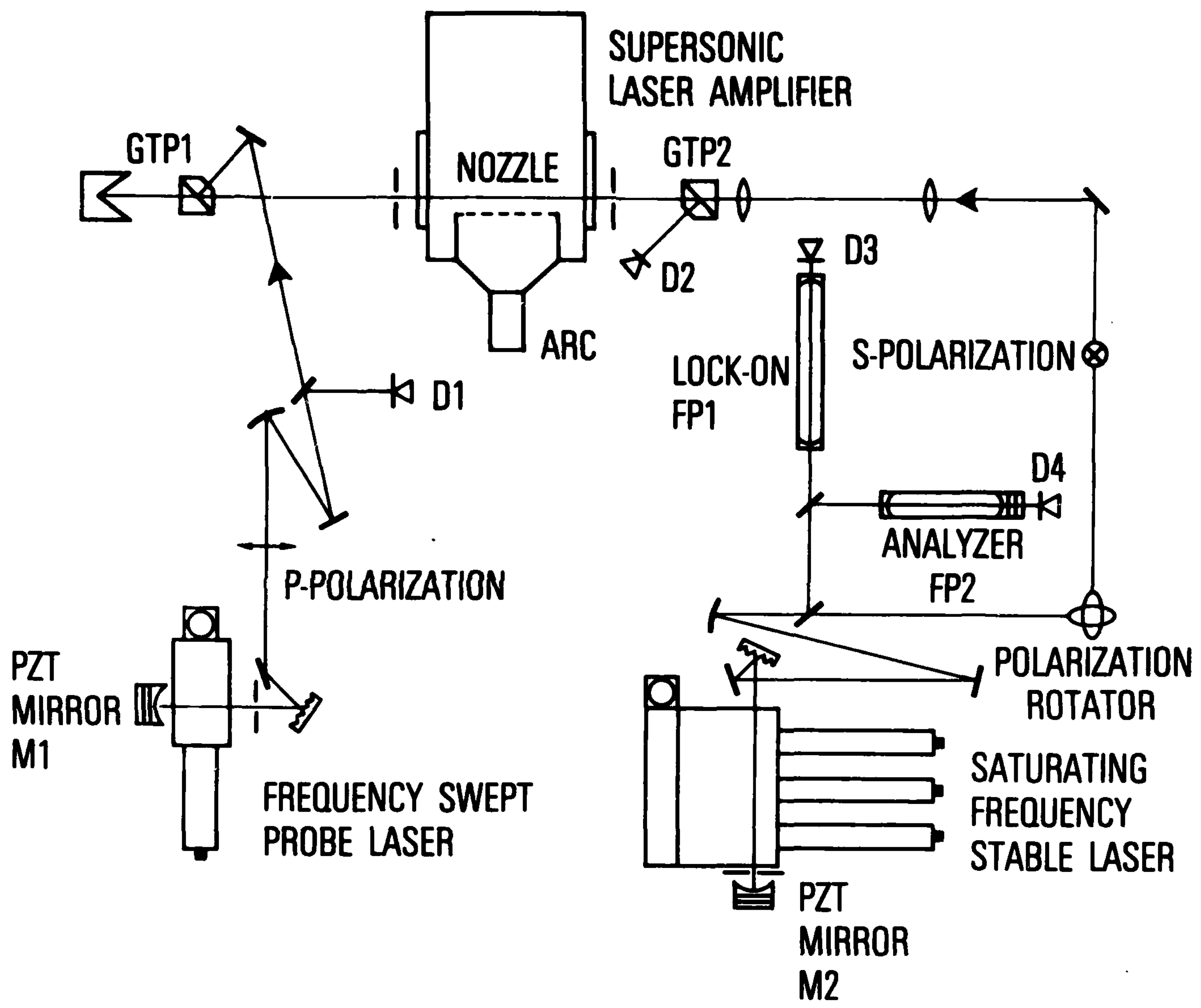

Fig. 1. Optical Layout of Apparatus

a dc discharge in three parallel discharge tubes. H_2 was injected through wall holes into a nearly sonic throat. Equipped with a grating and a 2-m radius gold-coated mirror, this laser produced up to 35 W of continuous, multimode power on the $P_2(8)$ line of HF. In our experiments, the cavity length was reduced to 40 cm. The resultant longitudinal mode separation, after correcting for mode-pulling, was about 320 MHz. This mode separation is of the order of the gain-line width of this laser, and makes its operation on a single longitudinal mode possible.

The laser cavity was frequency stabilized by locking it to a fixed, stable Fabry-Perot interferometer 25 cm long, corresponding to a free spectral range of 300 MHz. As depicted in Fig. 1, this reference Fabry-Perot interferometer was arranged externally to the laser cavity.

The total reflector of the laser cavity was mounted on a piezoelectric transducer, to which a sinusoidal signal of 600 Hz and a few tenths of a volt amplitude were applied. This signal provided a small dither signal, which was sensed by a detector mounted behind the Fabry-Perot interferometer. The error signal, produced by a frequency drift of the laser mode with respect to the Fabry-Perot, was amplified and rectified by a phase-lock amplifier and then used to correct the position of the cavity mirror, closing the control loop. A second Fabry-Perot interferometer of 750 MHz free spectral range was used to monitor the mode structure and stability of the laser cavity. In this manner it was possible to produce up to 30 W in a single, stable mode on the HF $P_2(8)$ line with the freedom of locating this mode at any frequency of the gain-width of this line. Figure 2 indicates typical oscilloscope records of the power output and the mode of this oscillator. The upper trace was obtained by analyzing the laser output with a confocal Fabry-Perot interferometer of 750 MHz free spectral range while oscillating the total reflector of the laser cavity with a large voltage at 600 Hz. The lower trace is a record of the stabilized single-line output integrated over an extended time. Note that the width of the mode increased dramatically in single-line operation; its FWHM is about 17 MHz. After passing several relay mirrors, a simple beam collimator, a polarization rotator, an inverted telescope of magnification 2, the Glan-Thompson prism, and the window of the test section, a Gaussian beam with an

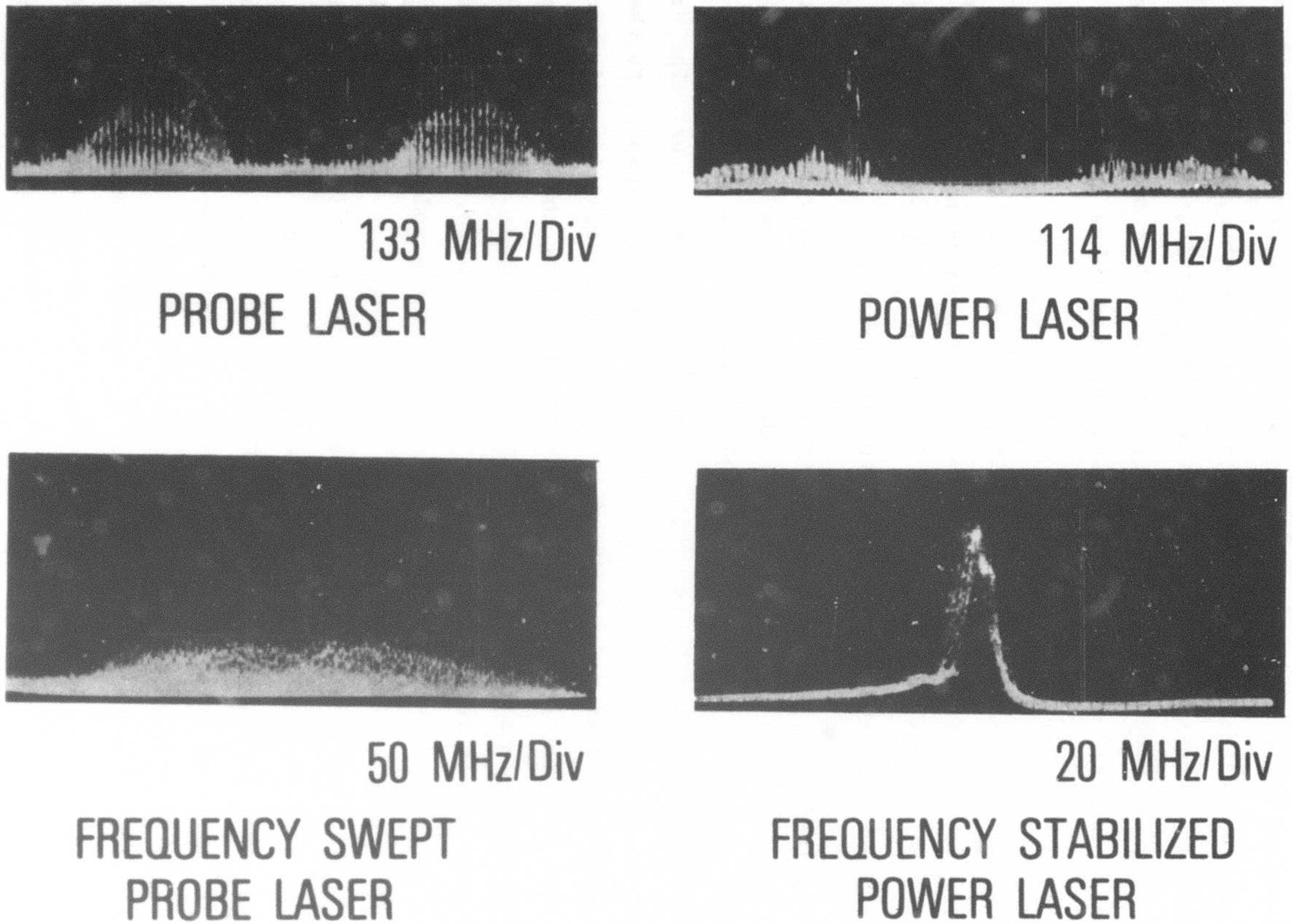

Fig. 2. Frequency Analysis of Laser Oscillators Used in Experiments. Free spectral range of Fabry-Perot: 750 MHz

average maximum power of 8 W across a diameter of 3.5 mm could be produced in the amplifier medium. By attenuating the beam power, densities between 1 and 85 W/cm^2 were available for saturating the medium.

The low-power probe laser, used for the gain measurements, was of the same construction as the saturating laser[6], except that it was equipped with only one discharge tube. The laser cavity consisted of a 300-ℓ/mm grating blazed at 3 μm and a 2-m radius gold-coated mirror. Power was extracted on the zeroth order beam. The cavity was mechanically stabilized inside four invar rods. The cavity was 30 cm long to assure single mode operation when the total reflector was swept across the line width of the gain medium by a piezoelectric translator, which was driven by the sawtooth voltage of an oscilloscope. The duration of a scan could be varied between 20 and 100 msec. The same oscilloscope was used to display the signals from the two detectors monitoring the probe laser beam. The free spectral range of the probe laser, corrected for mode pulling, was 420 MHz. Figure 2 indicates the laser output spectrally dispersed by a 750 MHz Fabry-Perot interferometer when the laser was scanned at 600 Hz. The lower trace shows the power line shape expanded to its full width. Note that the mode of this oscillator was at least an order of magnitude narrower than that of the saturating laser during scanned operation. Also, the width in frequency of the power output was about 400 MHz. This record provided the frequency calibration for the gain scans of the amplifier medium. The pronounced Lamb dip defined the center frequency of the line under investigation.

Under these conditions, this laser produced 0.3 W of single-mode output power on the $P_2(8)$ line of HF. The laser beam passed a beam splitter, diverting about half of its power to the InAs detector D1. The detector D1 was used to measure the input power I_0 to the test section. After passing several relay mirrors, the probe beam was superimposed on the saturating beam in Glan-Thompson prism GTP1. As depicted in Fig. 1, the probe beam entered the test section in a direction opposite to that of the saturating beam. The diameter of the probe beam was controlled by an inverted telescope of power 2 and an aperture of 2.5 mm, resulting in a power density of less than 1 W/cm^2 in the amplifier. After the test section, the probe signal was separated from the power beam in Glan-Thompson prism GTP2. The amplified probe laser signal I_1 was finally determined by InAs detector D2.

The signals from the two detectors were amplified and then recorded digitally by an on-line PDP-11/10 computer. The computer was programmed to average 32 individual frequency scans of the two signals to reduce noise and data scatter. The program digitally recorded the two averaged intensities into 2 × 100 channels for each scan along the time/frequency coordinate. From the averaged data, the ratio of I_1/I_0 and, subsequently, the gain-length product $GL = Ln(I_1/I_0)$ was calculated as a function of frequency point by point.

III. EXPERIMENTAL RESULTS AND DISCUSSION

In Fig. 3, typical averaged intensity records of 32 gain scans as recorded by the two detectors D1 and D2 as a function of frequency are depicted. The saturated intensity signal was measured when a single-mode beam of 60 W/cm^2 illuminated the amplifier medium, 4 mm from the nozzle exit. The saturating laser mode was positioned at the left maximum of the power output curve of the saturating oscillator, about 70 MHz off line center, see Fig. 2. Line center is given by the Lamb dip of the probe laser. The ordinate is calibrated by the free spectral range and the power line width of the probe laser shown in Fig. 2. The Lamb dips also demonstrate that the frequency resolution of our experiment was sufficient to distinguish any significant hole burning in the amplifier medium, if such were present. The amplification produced by the medium is clearly seen in these records as is the decrease of amplification when the saturating laser field is present. No hole burning or unilateral depression of the gain profiles is discernible near the position of the saturating laser mode; the depression is uniform across the entire gain profile.

Fully homogeneous saturation is also shown by the amplifier gain profiles obtained from these records shown in Fig. 4. The intensity depressions of Fig. 3 produced by the Lamb dip in the probe laser power profile have disappeared in calculating the gain length $GL = Ln(I_1/I_0)$, and the gain profiles at various saturating power densities are smooth and self-similar to the unsaturated gain profile.

Mirels,[7] using the theory of Ref. 2, calculated the gain profile to be expected in this medium under saturation by a single-line laser mode of varying power density. The resulting line shapes are shown in Fig. 5, normalized with the parameters of the theory[2]. At low power densities the depth of the hole burned into the Doppler line profile is small. It is, therefore, possible that at the power densities of our experiments, hole burning could not be observed, particularly since the theory of Ref. 2 may underestimate the cross-relaxation between the various Doppler-shifted

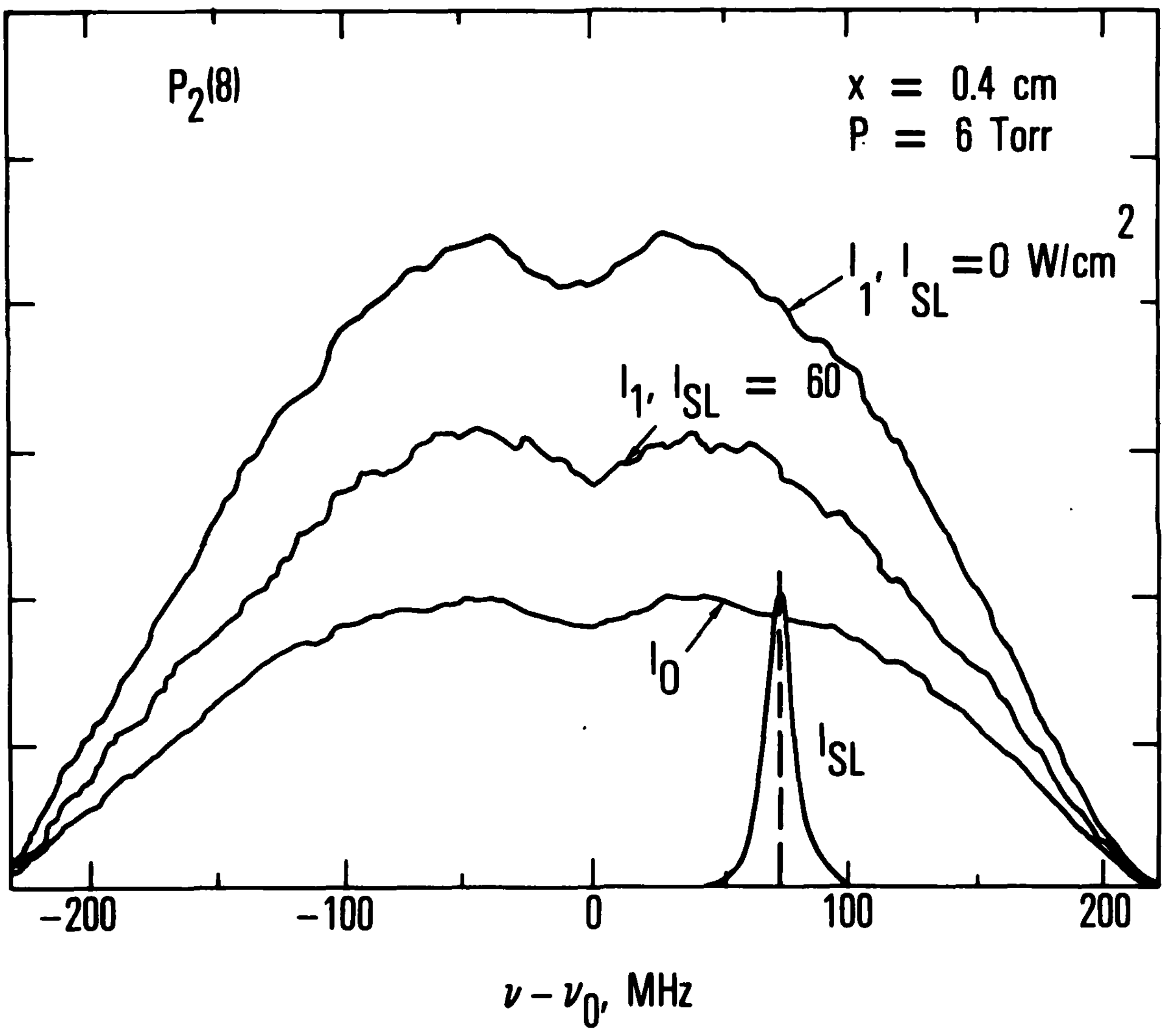

Fig. 3. Intensity of Probe Laser Signal Before and After Amplifier Section With and Without Saturating Laser Present

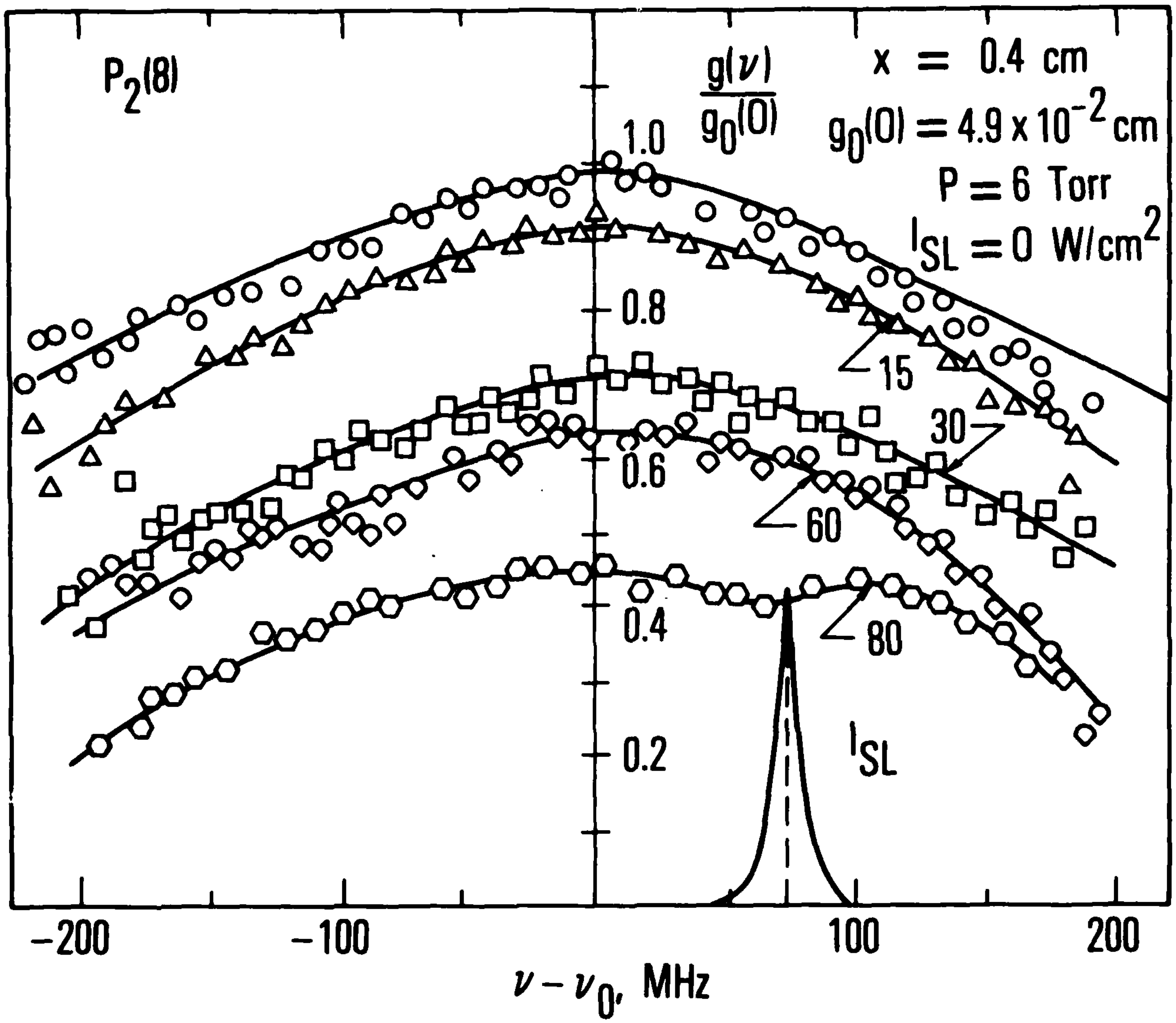

Fig. 4. Measured Line Shapes of Unsaturated and Saturated $P_2(8)$ Transition in The Aerospace Corporation Supersonic HF Diffusion Laser

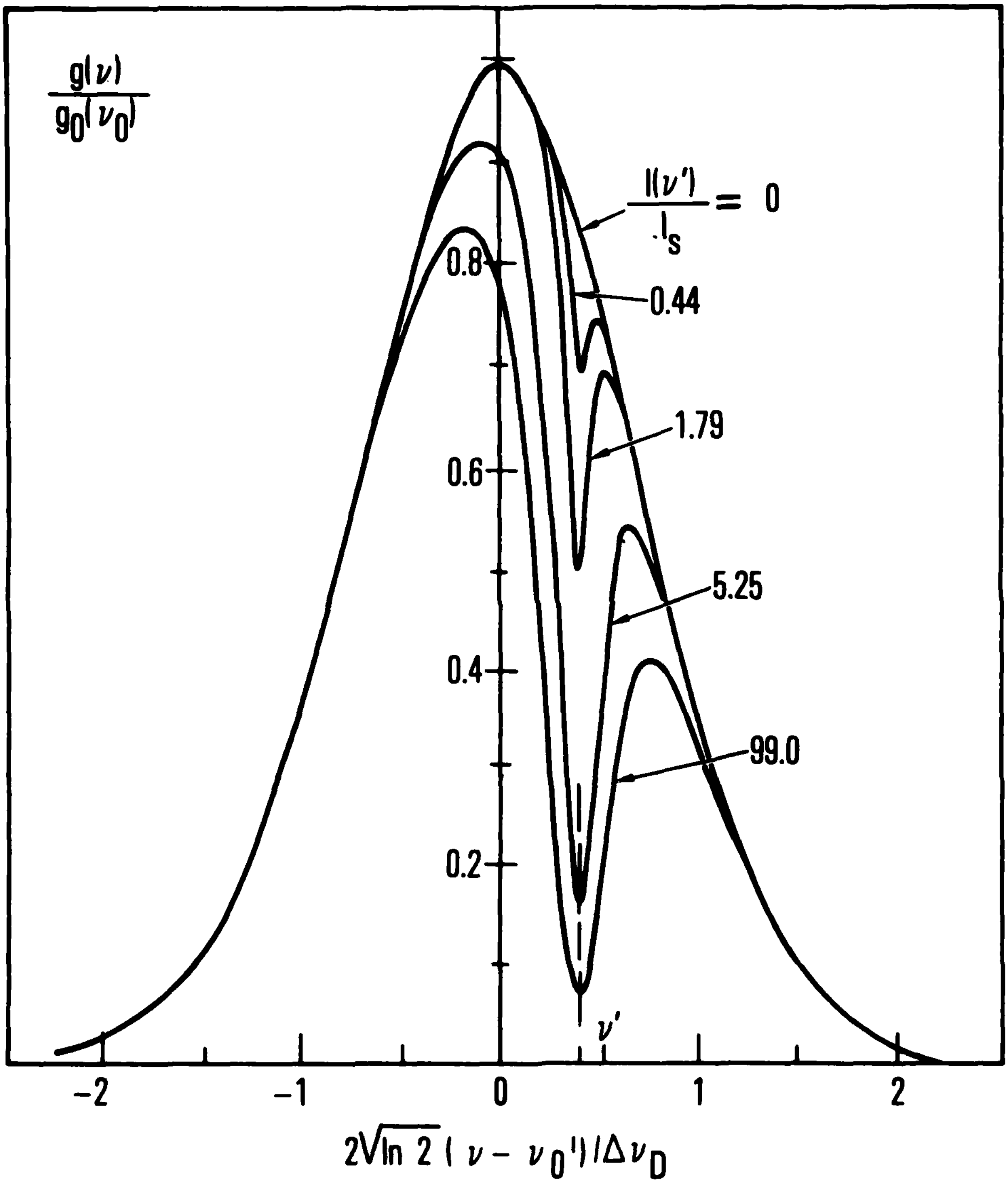

Fig. 5. Hole Burning Effects Predicted by Ref. 2 for a Supersonic Diffusion Laser Amplifier at P=3 Torr

molecules. Nonetheless, this theory does not predict any substantial uniform, "homogeneous" depression of the gain profile as is observed in our experiments.

A similar observation, although under substantially different experimental conditions, was reported in Ref. 8 for a CO_2 laser. The authors found a uniform, "homogeneous" depression of the line profile of their medium when gain is present. This observation was explained by the increased homogeneous relaxation of the investigated transition by higher lying, excited vibrational levels of the CO_2 molecule. It is interesting that they observe inverse hole burning in absorption under otherwise identical conditions in their medium.

With these experimental results we feel justified to calculate a "homogeneous saturation" at line center, as shown in Fig. 6, a process that would not be meaningful if the lines were saturated truly inhomogeneously. We also plot a curve through the experimental points given by the homogeneous saturation relationship $(g/g_0) = 1/(1 + I/I_s)$ that describes our results well for power densities between 1 and 85 W/cm^2. From this curve we calculate a homogeneous saturation density at line center of I_s = 80 W/cm^2.

The gain profiles of the arc-laser medium indicated in Fig. 4 are also seen to be considerably broader than those of the probe laser. By extrapolating the measured unsaturated gain profile to higher and lower frequencies we can estimate a FWHM of over 500 MHz. If one assumes that the line is Doppler broadened, this line width would correspond to a gas temperature in the laser medium of more than 1050 K, a value that appears to be very high, even when the hot gas in the boundary layers of the nozzle is taken into consideration. A much more comfortable explanation is offered by assuming that the nozzles are overexpanded so that the flow field is similar to the source flows investigated by Mirels[4]. The deformation of Doppler profile and the increase in its FWHM caused by such source flow effects is depicted in Fig. 7. The line broadening is substantial, and the profile loses its characteristic Doppler shape. In order to explain our experimental result of a FWHM of 500 MHz, we see that we have to invoke only a relatively moderate flow velocity component along the laser beam direction of Mach number 1.3. In a

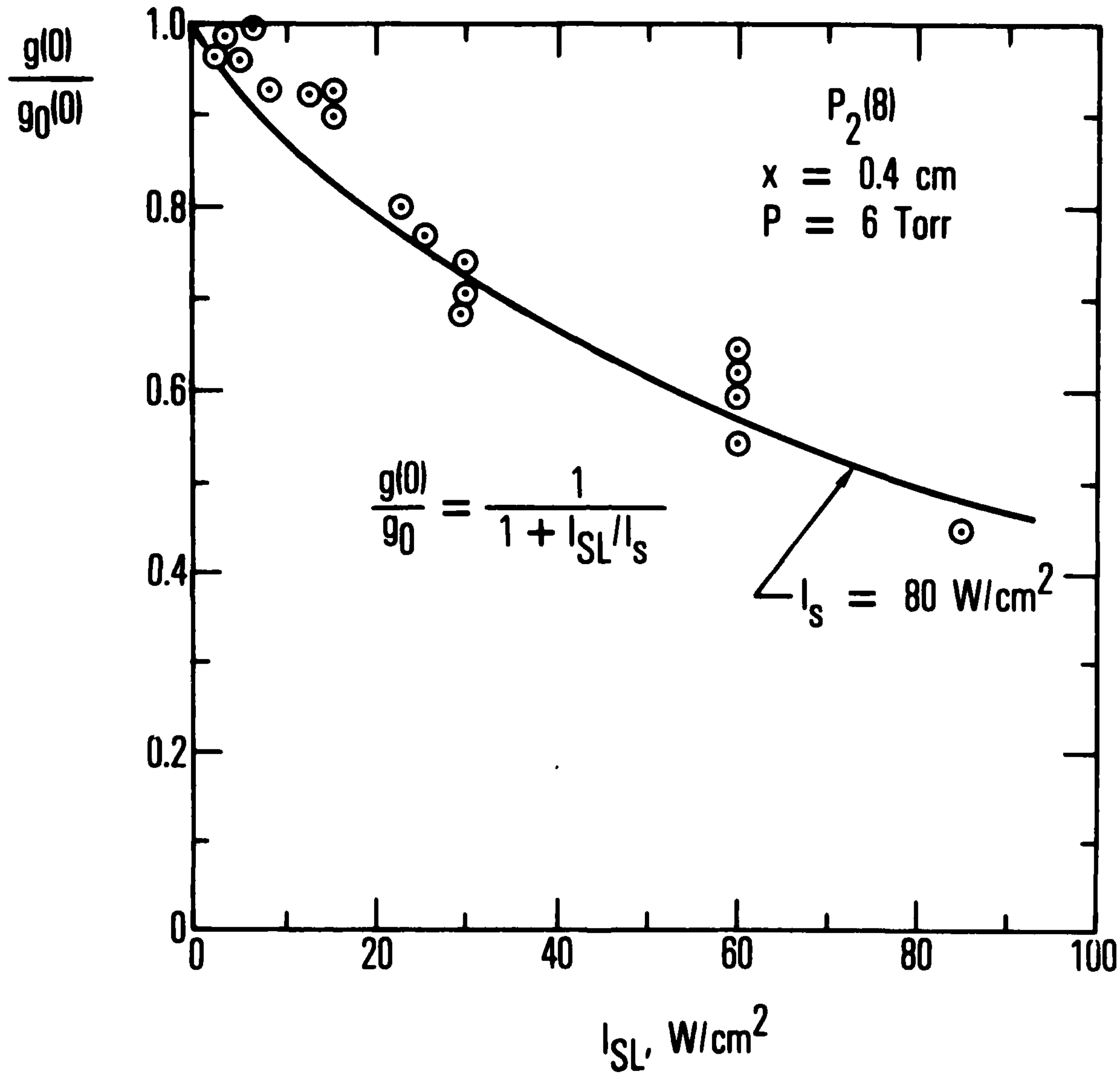

Fig. 6. Measured Gain at Line Center of $P_2(8)$ Transition as a Function of Saturating Laser Power Density I_{SL}

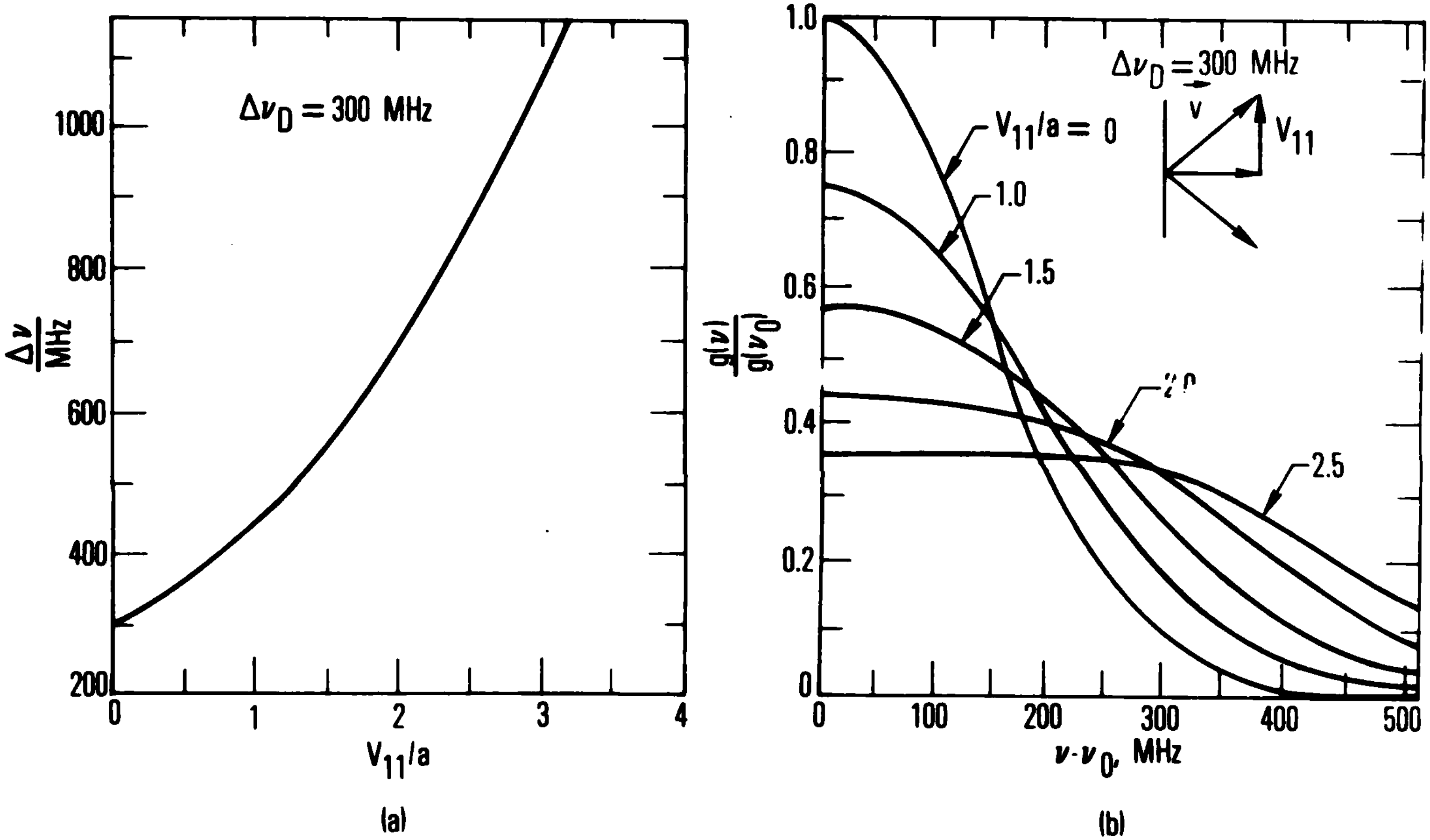

Fig. 7. Source-Flow Effects on Doppler Line Shape[4]. (a) Distortion of Doppler line shape caused by source flow, V_{11} = flow velocity parallel to laser beam, a = speed of sound. (b) Full width at half maximum of distorted Doppler line

flow field of Mach number 4, such as would be expected near the exit of our nozzle, this corresponds to an expansion half-angle of less than 20 degrees, which appears entirely plausible.

In conclusion, we have shown that at a pressure of 6 Torr, the $P_2(8)$ transition in the gain medium of a supersonic mixing laser is saturated homogeneously even if irradiated by a single-mode, single-line laser of quite moderate power density. We determined a homogeneous saturation intensity at line center of 80 W/cm^2 for this line at a position 4 mm downstream of the multi-slit nozzle of our laser. This observation is not explained by present theory[2] assuming a Doppler broadened line in the medium of this laser under our conditions. However, a similar observation has been reported for a CO_2 laser[8], although under sufficiently different experimental conditions to warrant a re-examination of present line shape theory for the HF laser. We also observe a considerable broadening of the gain line that could be explained only by assuming an unreasonably high Doppler temperature of over 1000 K. We propose to explain this observation by a laterally expanding flow similar to the source flows investigated theoretically by Mirels[4].

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