PFC/JA-82-15

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Abstract-First linewidth measurements over a 60 dB dynamic range of a pulsed, high-power, optically pumped far infrared laser are presented. These measurements were made possible by using a 385 μ mD₂O laser with an N₂O absorption filter and a sensitive heterodyne receiver. Studying a 385 μ mD₂O laser oscillator we find that the low-level linewidth (<-20 dB) can be explained by the ac Stark effect due to the high-power pump field. Also, we have not observed any frequency pulling of the main Raman emission frequency due to the strong pump and FIR laser fields.

INTRODUCTION

Since the first published account in 1974 [1], D_2 O has become one of the best developed of the megawatt, optically pumped, far infrared (FIR) molecular lasers [2]. These lasers are generally pumped by the CO₂ laser and operate by near resonant, stimulated Raman emission of which the 385 μ mD₂ O laser is a prime example. A theory for two laser fields of arbitrary strength interacting with a three level system has been developed [3], [4] and experimentally verified in the limit of small FIR signals [5]. However, the performance of high-power FIR laser cavities is difficult to predict.

In this paper we present the first linewidth measurements of a high-power 385 μ mD₂ O laser down to intensities 10⁻⁶ below peak. It is shown that the dominant Raman FIR emission frequency is what would be expected in the limit of small FIR and pump (no ac Stark effect) laser fields. On the other hand, the low-level (< -20 dB) linewidth can be understood in terms of the ac Stark effect due to the high-power pump field.

Our motivation for these measurements results from the application of this laser to collective Thomson scattering in fusion plasmas to measure ion temperature [6]. In this application the scattered signal is down approximately 10^{-14} from the incident laser intensity. In order for this measurement to succeed in a constrained access plasma, one must have very effective FIR beam and viewing dumps and/or a narrow linewidth rejection filter (the desired signal is Doppler broadened by the ion motions) with a corresponding narrow laser linewidth over many orders of magnitude of intensity. The present linewidth measurements were undertaken to investigate the feasibility of the latter approach.

Manuscript received August 2, 1982. This work was supported by the U.S. Department of Energy under Contract DE-AC02-78ET-51013.

N₂O gas has been discovered to be an excellent narrow band rejection filter for the 385 μ mD₂O laser [7]. This filter has also made possible the present linewidth measurements. N₂O has a $J = 30 \rightarrow 31$ transition in the vibrational ground state at 778.2 GHz, approximately - 700 MHz from the line center 385 μ mD₂O transition. The peak absorption is 10 dB per meter with a pressure broadened linewidth of 8.4 MHz.torr⁻¹. The D₂O laser is easily tunable to the N₂O absorption frequency with a tunable CO₂ pump laser. In fact, it has been shown that the power and efficiency of the 385 μ mD₂O laser is optimized by the required detuning [8].

EXPERIMENTAL DETAILS

The D₂O laser used for these measurements was a 4 m long, 11 cm effective aperture oscillator with a Fox-Smith mode selector [9]. Main cavity optics were a 30 lines cm⁻¹ grating and a 49 wires cm⁻¹ copper mesh (reflectivity ~ 30 percent at 385 μ m). The grating tuned the cavity to 385 μ m and polarized the output to 95 percent perpendicular to the pump. Fox-Smith cavity optics were the grating, a 79 wire cm⁻¹ copper mesh at 45° to the main cavity, and a flat gold coated mirror opposite the mesh. The Fox-Smith reflectivity finesse was estimated to be 10 and the free spectral range about 350 MHz. A crystal quartz plate was used to couple the CO₂ laser beam into the main cavity for a single pass pump. The CO₂ pump laser was a single mode, grating and etalon tuned, TEA oscillator-amplifier system [8], [9]. Over 2 GHz of continuous tunability on the CO₂ 9 R(22) transition was possible.

The 385 $\mu m D_2 O$ laser linewidth was studied by first downshifting the D_2O laser frequency with a heterodyne receiver to the microwave x band. The heterodyne receiver used a Schottky diode mixer and a 30 mW, 381 µm DCOOD laser local oscillator [10]. The downshifted signal was frequency resolved using a 32 channel multiplexer covering the band 8.2-10.8 GHz with 80 MHz channels. Provision was also made to use ten percent of the downshifted signal with a second mixer and a 100 MHz bandwidth SAW filter for single shot resolution of the D_2O laser cavity modes [11]. The signal in each multiplexer channel was acquired with a Lecroy CAMAC system 2280 current integrating data acquisition system. A Digital Minc-11 computer then displayed the spectrum. The fraction of the laser pulse over which the signal was integrated could be varied, but we found no significant effect on the detected linewidth by excluding either the start or end of the

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pulse. The D_2O laser pulses were between 120 and 180 ns long at half maximum with ~ 120 ns CO_2 laser pump pulses.

FIR laser energy was measured with a Lumonics model 50D, 12 cm aperture, pyroelectric detector calibrated for 385 μ m (responsivity ~25 percent of 10 μ m calibration).

The laser power available was many orders of magnitude above the receiver sensitivity. Care was taken to attenuate the laser beam to avoid damaging the Schottky diode mixer and saturating the electronics. The linear dynamic range of the receiver was limited. Therefore, the linewidth measurements over a 60 dB dynamic range were accomplished by using a combination of calibrated attenuation and a 7 m path length N₂O cell between the laser and receiver. For attenuation we calibrated sheets of paper using the CW DCOOD laser. The orientation of the papers during calibration was varied to verify the absence of interference effects.

MEASUREMENTS

The linewidth measurements proceeded as follows. First, with the D_2O laser tuned to the N_2O absorption frequency and the N₂O cell evacuated, the FIR beam was attenuated with a paper stack to produce a nonsaturated spectrum on the computer display. This spectrum was recorded. Next, the attenuation was reduced by removing one or more calibrated groups of paper. This brought up additional channels with measurable signal off frequency center, while causing the center channels to saturate. N₂O gas was next let into the absorption cell as needed to selectively filter out the central frequency components and keep the power on the Schottky diode in the same range. The process of reducing the attenuation and increasing the N₂O pressure was repeated several times. The spectrum was recorded at each level of attenuation. Later, all the data were normalized using the known attentuation and N₂O absorption.

The results are shown in Figs. 1 and 2. Each plotted point is the average of several laser shots, the error bar corresponding to the standard deviation. For Figs. 1(a), 1(b), and 2(a), the CO₂ laser pump energy was 20 J in a spacially nonuniform beam area of 56 cm². This would correspond to a time and space averaged pump intensity of 3 MW \cdot cm⁻² at the D₂O laser input. In Fig. 2(b) the pump energy was reduced to 0.46 J for an average pump intensity of 70 kW \cdot cm⁻².

At the lower D_2O laser pressures of 1 and 2 torr [Figs. 1(a), 2(a), and 2(b)], the linewidth spectrum shows two main peaks. The dominant peak at 778.2 GHz corresponds to the stimulated, near resonance Raman emission, while the lower peak at approximately +0.7 GHz is due to the on resonance 385 μ mD₂O transition. For 20 J of pump energy [Figs. 1(a) and 2(a)] these peaks are significantly broadened asymmetrically away from each other. The structure, as explained below, may be due to the nonuniform nature of the CO₂ laser pump beam. At 6 torr and 20 J of pump energy [Fig. 1(b)] only the Raman peak is evident, appearing symmetrically broadened.

Investigation of the D_2 O laser mode structure with the SAW filter revealed distinct longitudinal modes spaced at 35 MHz and less than 10 MHz wide. At the peak Raman emission frequency these modes could be suppressed by up to an order of magnitude by the Fox-Smith mode selector. On the other



Fig. 1. 385 μmD₂O laser linewidth spectrums with 20 J CO₂ laser pump energy (3 MW · cm⁻² time and space average pump intensity);
(a) 1.0 torr D₂O pressure, 7.2 mJ FIR output energy;
(b) 6.0 torr D₂O pressure, 41 mJ FIR output energy.



Fig. 2. 385 μmD₂O laser linewidth spectrums at 2.0 torr D₂O pressure; (a) 20 J CO₂ laser pump energy (3 MW · cm⁻² time and space average pump intensity), 20 mJ FIR output energy; (b) 0.46 J CO₂ laser pump energy (70 kW · cm⁻² time and space average pump intensity), 0.45 mJ (estimate) FIR output energy.

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hand, at low levels (< -20 dB) detuned up to 300 MHz from the peak frequency, we observed that the emission was either distinct longitudinal modes or random frequency components never significantly suppressed as the Fox-Smith was tuned.

The FIR energy measurements are given in the figure captions. In Fig. 2(b) the FIR energy was too low to be detected by our Lumonics Joulemeter. This energy is an estimate based on linearly scaling with pump energy our measurement for Fig. 2(a). Profile measurements of the FIR beam showed an approximate half power diameter of 7 cm.

DISCUSSION

These linewidth measurements can be interpreted in terms of the theory of two laser fields of arbitrary strength interacting with a three level system [3], [4]. Assume the three levels are labeled 1, 2, and 3 in order of increasing energy. There are four dimensionless parameters of importance $\delta_{p}\tau$, $\delta_{s}\tau$, $\beta_{13}\tau$, and $\beta_{32}\tau$ where δ_p is the pump offset frequency, δ_s is the FIR emission offset frequency, $\tau = \Delta \nu^{-1}$ is the relaxation time with $\Delta \nu = 40 \text{ MHz} \cdot \text{torr}^{-1}$ for D₂O, $\beta_{13} = \mu_{13}E_p/2h$ is half the pump Rabi frequency, and $\beta_{32} = \mu_{32} E_s/2h$ is half the emission Rabi frequency. In the Rabi frequency expressions, μ_{ij} is the space averaged dipole moment for transition $i \rightarrow j$, E_p and E_s are the pump and emission electric fields, and h is Planck's constant. For the 385 μ mD₂O laser we use $\mu_{13} = 0.0323$ Debye and $\mu_{32} = 0.68$ Debye. These dipole moments are approximate space averages that do not take into account the details of the ΔM selection rules [4], but are accurate enough for our purposes.

The theory predicts the FIR gain spectrum. In the limit of small FIR intensities $(\beta_{32}\tau \ll 1)$ the line center and Raman components have equal gain and are separated in frequency by $(\delta_p^2 + 4\beta_{13}^2)^{1/2}$ where $2\beta_{13}$ is the ac Stark shift contribution. Increasing the FIR intensity to a few W \cdot cm⁻² causes the line center component to saturate first [5], as evident in Figs. 1 and 2 where this component is weak. Continued increase in the FIR intensity to large values $(\beta_{32}\tau > \beta_{13}\tau)$ causes the Raman gain to go through an optimum and then the Raman gain spectrum broadens, decreases, and is pulled toward line center (see Fig. 5 in [3]).

The observed $D_2 O$ laser emission spectrums do not all follow from the predictions for the gain spectrum. In Fig. 1(b) where $\delta_p \tau = \delta_s \tau = 2.9$ and $\beta_{13} \tau \approx \beta_{32} \tau > 1$ the gain spectrum should be asymmetrically broadened toward line center and δ_p should not necessarily be equal to δ_s . A symmetrically broadened gain spectrum in high power FIR operation when $\delta_p \tau > 1$ is only predicted when $\beta_{32} \tau > \beta_{13} \tau$ and the Raman component is pulled to line center ($\delta_s \tau = 0$).

The lack of seeing a difference between δ_p and δ_s for the peak emission frequency has been true for all data we have taken at D₂O pressures of 1 torr or greater. If $\beta_{32}\tau$ is varied over a large enough range relative to $\beta_{13}\tau$ there would be some frequency shifting of the main gain peak. In Fig. 1 comparing 1 and 6 torr emission for the same $\delta_p = 700$ MHz and pump energy of 20 J, the relative change of $\beta_{32}\tau$ to $\beta_{13}\tau$ is a factor of 2.5 ($\beta_{13}\tau = 9.7 \rightarrow 1.6$; $\beta_{32}\tau = 5.5 \rightarrow 2.3$ for $1.0 \rightarrow 6.0$ torr), yet there is no evidence of any shift in emission frequency

greater than our 80 MHz resolution. However, the lack of observing a shift in frequency may be because $\beta_{32}\tau$ and $\beta_{13}\tau$ have not been varied over a large enough range. More likely, the observation $\delta_p = \delta_s$ at high power may be due to an oscillator cavity effect because the FIR emission builds up from small signals when $\delta_p = \delta_s$ is predicted. Linear tuning of high-power FIR oscillator emission frequency with pump frequency, without any frequency pulling, has been observed previously [11].

The low-level broadening of the D_2O laser linewidth can be understood in terms of the ac Stark effect due to the pump field. In Figs. 1(a) and 2(a) the line center and Raman components are broadened away from each other, the direction of frequency shift expected for the ac Stark effect. High FIR intensities would pull the frequencies in the opposite direction toward line center. Higher pressure rather than higher FIR intensity probably contributed to filling in the spectrum between line center and Raman in Fig. 1(b), but the extreme width of the spectrum is probably due to the pump field ac Stark effect. At the low levels (< -20 dB) the small FIR signal condition $\beta_{32}\tau \ll 1$ is true, but the gain is still large enough for single pass emission as evidenced by the Fox-Smith tuning results.

The pump field intensity necessary to produce the observed broadening can be calculated. The two outer peaks in the spectrum structure of Fig. 1(a) (at +1.2 GHz and -0.6 GHz) are equally displaced from $\delta_p/2$ (a condition for the ac Stark effect) and separated by 1720 MHz. This splitting would correspond to a pump field intensity of 12 MW \cdot cm⁻² or four times the space and time average of the pump beam. It is very likely a hot spot of that intensity exists. The lack of low-level broadening in Fig. 2(b) is explained because for four times the average pump power a splitting of only 740 MHz is expected. At 6 torr [Fig. 1(b)] the pump field is more strongly absorbed down the length of the cavity, reducing, on the average, the pump intensity. This would explain the slight decrease in the total low-level spectrum width relative to 1 torr for the same pump energy.

CONCLUSIONS

Several conclusions can be made based on our measurements and interpretation. 1) The pump beam intensity must be kept low (<1 MW \cdot cm⁻²) without hot spots to prevent ac Stark broadening of the low-level linewidth. This implies large aperture systems for high-power, narrow linewidth operation. Also, we have recently operated our D₂O laser with longer pulses (~ 1 μ s) and achieved a significant reduction in the low-level linewidth for the same FIR energy, though at lower peak power. 2) The pump beam intensity cannot be made too low or the conversion efficiency of the Raman transition will be reduced, as evidenced by the relative increase in the line center component in Fig. 2(b). The presence of a strong line center component along with the Raman component would also complicate linewidth filtering. 3) A narrow linewidth filter in the laser such as a Fox-Smith mode selector is effective in the peak 10 dB of FIR intensity, but not in the low-level (< -20 dB) extremes of the linewidth. A

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distributed feedback laser [12] or filtering at the laser output may be more effective. 4) The dominant frequency of emission from a high-power D_2O laser oscillator is at the Raman frequency expected for small signal pump and FIR emission fields. No ac Stark shifting or strong FIR field pulling has been observed.

ACKNOWLEDGMENT

The authors gratefully acknowledge many useful discussions with R. J. Temkin, H. R. Fetterman, and D. R. Cohn, especially R. J. Temkin for originally suggesting N_2O .

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