Gain Saturation of CW Laser Pumped FIR Laser Gases

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Abstract—The saturation behavior of the 70 μm and the 119 μm transitions in CH_3OH has been investigated both experimentally and theoretically. This knowledge is essential for constructing a complete model of the CW laser pumped FIR laser. Saturated gain measurements made in a single pass amplifier cell were found to be in good agreement with the theory of a quantum-mechanical three-level system in resonant interaction with two coherent fields. The calculation of the gain using the density matrix formalism yields a pump intensity dependent saturation, a result which is not predicted by the rate equation models.

I. Introduction

OPTIMIZATION of laser pumped FIR lasers necessitates a complete understanding of the FIR laser cycle for arbitrary pump and FIR field strengths.

It has been demonstrated that a quantum-mechanical treatment including coherent (dynamic Stark effect) and two-photon effects (Raman transitions, for off-resonant pumping only) is adequate for describing the gain process of both pulsed [1] and CW [2] laser pumped FIR lasers.

In [2], explicit expressions have been derived for the intensities emitted or absorbed in a three-level system by solving the density matrix equations of [3]. The results are valid for arbitrary broadenings and field strengths of the transitions involved. They also account for the removed M-degeneracy of the molecular levels due to the linearly polarized laser fields. In [2] these results were compared to experimental small-signal gain data of some short wavelength CW FIR laser transitions. In this spectral region (40 μm $< \lambda <$ 200 μm) some of the strongest CW FIR laser transitions can be found.

The present paper discusses the saturation behavior of CW FIR laser transitions in the short wavelength FIR region and reports direct gain saturation measurements on the 70 μm and the 119 μm CH_3OH lines using a single pass amplifier cell.

The good agreement found between theory and measurement makes the former a basis for a reliable model of the CW laser pumped FIR laser.

II. Theoretical Saturation Behavior

In the wavelength range $\lambda < 200$ μm, the transitions of a CW laser pumped FIR laser are mixed (Doppler/pressure) broadened. The level scheme of such a laser together with the line shapes induced by the two fields $\vec{E}_1(\Omega_1, t) = E_1 \cdot \cos(\Omega_1 t - k_1 z)$ and $\vec{E}_2(\Omega_2, t) = E_2 \cdot \cos(\Omega_2 t - k_2 z)$ are shown in Fig. 1.

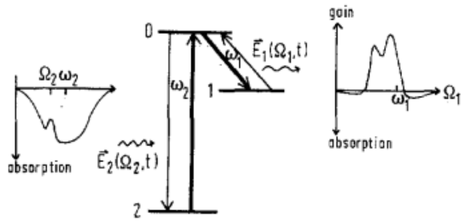

Fig. 1. Level scheme and associated line shapes for a CW laser pumped FIR laser. ($\vec{E}_2(\Omega_2, t)$: pump field at frequency Ω_2. $\vec{E}_1(\Omega_2, t)$: FIR field at frequency Ω_1. ω_2, ω_1: center frequencies of the molecular transitions.

$\vec{E}_2$ is the strong pump field saturating the absorption of the transition 2-0 within the Doppler absorption line at the frequency Ω_2 which may be equal to or different from ω_2, the center frequency of the transition 2-0. The population inversion between levels 0 and 1 produced by $\vec{E}_2$ gives rise to a gain line with the center frequency Doppler shifted from ω_1 by $(\Omega_2 - \omega_2)\omega_1/\omega_2$. This line is probed by the field $\vec{E}_1$. Due to strong coherent pumping at Ω_2, the levels 2 and 0 suffer dynamic Stark splitting by an amount equal to the Rabi frequency. This splitting of the molecular levels 0 and 2 manifests itself in a splitting of the gain line as long as E_1 is small. (For Doppler-broadened transitions, the gain line splitting appears only for copropagating fields, which is the case in the present experiments.) For strong E_1, the gain line shape is altered by gain saturation. The dynamic Stark maxima are expected to broaden and decrease in amplitude in this case (details to follow).

If only a rough estimate of the saturated gain is required, then the rate equation model [1] can be used to give

$$E_{1\,\mathrm{sat}} \cdot |\mu_{10}|/h = \gamma_{10}. \tag{1}$$

$|\mu_{10}|$ is the dipole moment matrix element of the transition 0-1, $\gamma_{10} = \frac{1}{2}(\gamma_1 + \gamma_0)$, and γ_i is the decay rate of level i. The saturation field strength $E_{1\,\mathrm{sat}}$ is defined by

$$g/g_0 = (1 + E_1^2/E_{1\,\mathrm{sat}}^2)^{-1} \tag{2}$$

which is standard saturation formula for a homogeneously broadened transition. g and g_0 are the saturated and the small-signal gain coefficients, respectively.

For several reasons, however, the saturation behavior of the CW laser pumped FIR laser cannot be properly described by the usual saturation formula. First, the strong coherent pump field $\vec{E}_2$ splits the gain line due to the dynamic Stark effect and the saturating FIR field $\vec{E}_1$ broadens the Stark maxima reducing their amplitudes. Due to the overlap of the Stark maxima, this should result in a frequency dependent satura-

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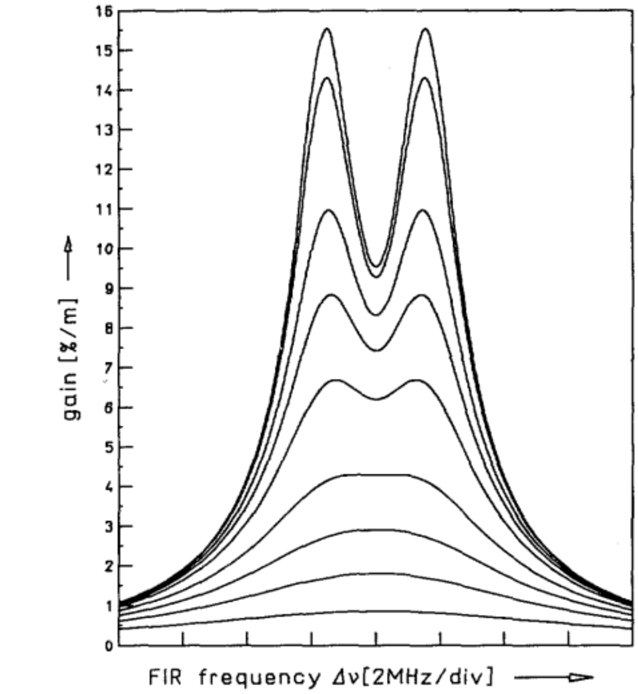

Fig. 2. Theoretical gain line profiles of the 119 μm CH_3OH transition. Parameters: pressure = 5 Pa, pump intensity = 15 W/cm^2, and in ascending order the curves correspond to the FIR intensity values of 0.1, 1, 5, 10, 20, 50, 100, 200, and 500 mW/cm^2.

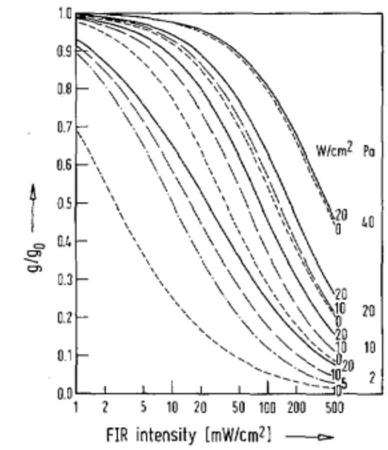

Fig. 3. Theoretical saturation on the 119 μm CH_3OH line center as a function of FIR intensity. The gain g is referred to the small-signal gain g_0. Curve parameters are the pump intensity (W/cm^2) and the pressure (Pa).

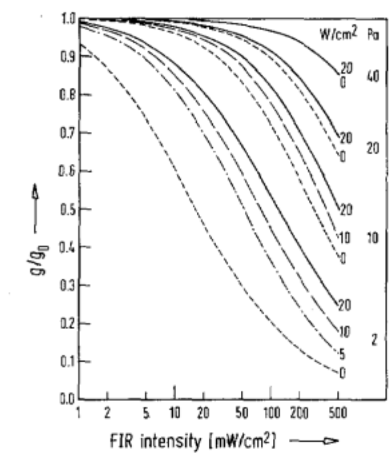

Fig. 4. Theoretical saturation on the 70 μm CH_3OH line center as a function of FIR intensity. Remarks: same as for the 119 μm CH_3OH line.

tion, different on and between the maxima. Second, due to pumping and probing with linearly polarized fields, the M-degeneracy (space orientation of the molecules) of the molecular levels is removed [4]. As $|\mu_{10}|$ depends on M, so does the saturating field strength $E_{1\,\mathrm{sat}}$, introducing thereby an additional inhomogenity into the saturation. An additional frequency dependence of the gain saturation is therefore to be expected: the different M-components of the gain line, being at different frequencies, corresponding to the type ($\Delta J = 0, \pm 1$) of the pump transition, will saturate differently, again depending on the type of transition, in this case the FIR transition. Finally, the separation of the Stark maxima is proportional to the pump field strength E_2. The overlap of the Stark maxima, however, determines the saturation behavior at different frequencies. The saturation, therefore, should also depend on the pump field strength if the dynamic Stark maxima are resolved.

The saturated gain for the 70 μm and the 119 μm CH_3OH transitions was calculated using the density matrix formalism of [2] the Appendix. Fig. 2 shows theoretical gain line profiles for the 119 μm transition at fixed pressure and pump intensity, but different FIR intensities, the pump frequency being the transition center frequency. At low FIR intensity the dynamic Stark splitting is clearly evident, while at higher FIR intensities it disappears due to the FIR power broadening (as discussed before). As expected, the gain saturates less at the line center than at the two Stark maxima.

Figs. 3 and 4 show the gain saturation at line center as a function of the FIR intensity I_1 for different pressures and pump intensities I_2. At low pressure, where the dynamic Stark splitting is already present at medium pump intensities, the saturation at the line center depends strongly on the pump intensity. Even at typical FIR laser operating pressures (between 10 and 20 Pa) the pump intensity dependence of the gain saturation is still apparent but diminishes with increasing pressure as expected. At high pressures, the saturation of the gain at $I_2 = 20$ W/cm^2 equals the saturation of the absorption ($I_2 = 0$ W/cm^2), in conformity with rate equation models. It should be noted that when pumping at line center the effect of coherence shows up as an increase in saturation intensity with pump intensity. This effect, which cannot be described by any rate equation model, is naturally most pronounced at low pressures, where the Rabi frequencies of both transitions are high compared to the collision frequencies. The case of incoherent pumping is included in Figs. 3 and 4 since the saturation behavior is then almost identical to that at $I_2 = 0$ W/cm^2. In that case, (1) and (2) may be used to give the saturating field strengths or intensities for the FIR transitions (see Table I).

It can be seen from the saturation behavior of the two lines considered that the 70 μm transition saturates less compared to the 119 μm transition. This is due to the different dipole moment matrix elements $|\mu_{10}|$ and the collision rates γ_{01} of these lines (see Table I). The 119 μm transition is a pure rotational transition and has a higher dipole moment matrix element compared to the 70 μm line, being a torsional-rotational transition [5]. Also the collision rate γ_{01} is somewhat higher for the 70 μm line than for the 119 μm line, resulting in a stronger saturation for the 119 μm transition.

TABLE I
MOLECULAR DATA FOR GAIN SATURATION IN CH_3OH

FIR transition	119 μm	70,5 μm
γ_{01} $(s \cdot Pa)^{-1}$	$0.69 \cdot 10^6$	$1.07 \cdot 10^6$
$\lvert\mu_{10}\rvert$ (C·m) space average	$1.4 \cdot 10^{-30}$	$0.94 \cdot 10^{-30}$
$E_{1\ sat}$ (V/mPa)	51	99
$I_{1\ sat}$ (W/m^2Pa^2)	3,5	13

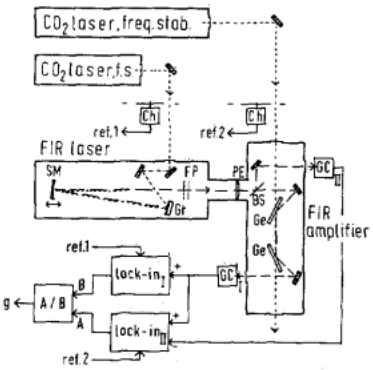

Fig. 5. Experimental setup for the saturated gain measurements. (CH: chopper, SM: spherical mirror, Gr: grating, FP: Fabry–Perot, PE: polyethylene lense, BS: beam splitter, Ge: Germanium plate, GC: Golay cell, g: FIR gain).

It should be mentioned here that the calculated gain saturations apply only for plane traveling waves. For comparison with the experiment, the calculated gain saturations had to be integrated over the Gaussian intensity profile (in this case) of the FIR laser beam. Saturations of the Gaussian beams together with the experiment results are shown in the following section.

III. EXPERIMENTAL GAIN SATURATION

The experimental setup is shown schematically in Fig. 5. Two different CO_2 lasers, frequency stabilized in various ways, pumped the FIR laser and the FIR amplifier. Both CO_2 laser beams were chopped at different frequencies. One of these beams was superimposed coaxially on the FIR beam in the FIR gain cell, Ge plates being used for beam combination and separation. Both beams were collimated inside the gain cell with the CO_2 laser beam larger in diameter than that of the FIR beam to ensure maximum possible homogeneous pumping conditions. Due to two different chopping frequencies, the FIR intensity signal I_1 and the FIR gain signal $(I_1 \cdot g)$ could both be obtained simultaneously from one Golay cell detector for synchronous detection. A second Golay cell was used to detect the FIR laser amplitude noise at the "gain" frequency and subtract it from the gain signal, increasing the S/N ratio by a factor of five.

The FIR laser was specially suitable for the purpose of saturated gain measurements. It was a folded resonator [6] having an 80 1/mm grating and a copper mesh Fabry–Perot output coupler as the laser and mirrors. The pump radiation was coupled to the resonator via the grating in first-order diffraction. This resonator without big coupling holes oscillated preferentially on TEM_{00} modes and the Gaussian output beam made the comparison of measurement and theory easier. For saturated gain measurement, the FIR laser power could be changed rapidly by tuning the Fabry–Perot coupler, all other parameters remaining same. Compared to other configurations the FIR resonator had a relatively high

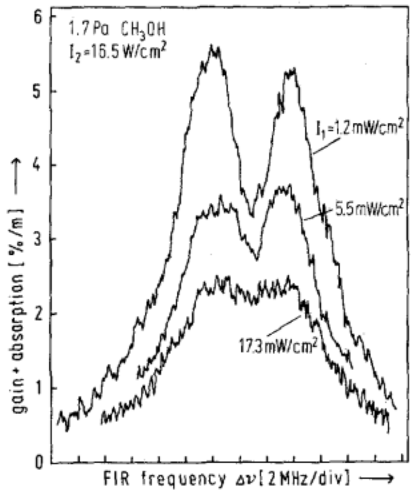

Fig. 6. Measured gain line profiles of the 119 μm CH_3OH transition for three FIR intensities I_1 at a pressure of 1.7 Pa.

mirror loss which thereby limited the available FIR intensity to some extent. With this setup, FIR gain profiles were obtained as functions of the FIR frequency by scanning the FIR resonator mechanically. The parameters varied were pump intensity and frequency, FIR intensity, and pressure.

Fig. 6 shows gain profiles of the 119 μm CH_3OH transition for a particular pressure and pump intensity, but for different FIR intensities I_1. As expected theoretically, the gain saturates by the broadening of the dynamic Stark maxima and the reduction of their amplitude.

A quantitative comparison of measurement and theory is given in Fig. 7(a) and (b) and Fig. 8(a) and (b) for the saturation behavior of the 70 μm and the 119 μm lines for different pressures and FIR frequencies, the pump frequency Ω_2 being the transition frequency ω_2. Part (a) of these figures shows the saturation at the gain line center (ω_1 in this case) while part (b) corresponds to the frequency of one of the dynamic Stark maxima of the small-signal gain line at the pump intensity used. In Figs. 7 and 8 the saturations of the change signal (gain + absorption, the quantity measured) are plotted for a Gaussian intensity distribution of the FIR beam as functions of the FIR intensity. As expected, the 119 μm line has stronger saturation compared to the 70 μm line at the same FIR intensity, and that saturation depends strongly on the FIR frequency. The gain at the dynamic Stark maxima saturates faster than that at the line center due to the FIR power broadening and the inhomogeneous saturation behavior. It may be concluded that the FIR gain saturation can not be described by a simple saturation formula in terms of only one saturation parameter.

However, agreement between saturated gain measurements and theory over the limited available range of FIR intensities indicates that the saturation behavior can be predicted accurately by using the density matrix formalism to evaluate the gain numerically.

In [2], the input data used for gain evaluation were thought to be unreliable mainly in the population densities but not in the dipole moment matrix elements $\lvert\mu_{10}\rvert$ or the collision rates. The results in Figs. 7 and 8 strongly support this assumption as the absolute population densities are not involved in the calculation of the gain saturations in contrast to the dipole moment matrix elements and the collision rates. It

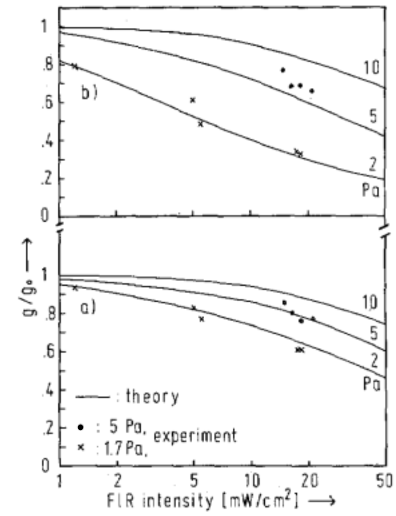

Fig. 7. Comparison of theory and measurement for the saturation behavior of the 119 μm CH_3OH line at various pressures. Case a) and b) differ by the FIR frequency deviation $\Delta\nu$ from the center frequency, i.e., 0 MHz, respectively, 1.5 MHz. The pump intensity was 15 W/cm^2 in both cases.

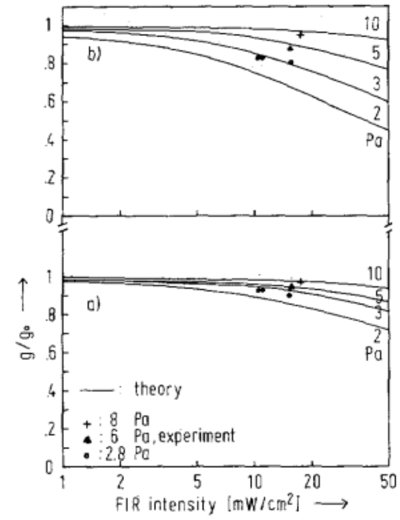

Fig. 8. Comparison of theory and measurement for the saturation behavior of the 70 μm CH_3OH line at various pressures. Case a) and b) differ by the FIR frequency deviation $\Delta\nu$ from the center frequency, i.e., 0 MHz, respectively, 2 MHz. The pump intensity was 15 W/cm^2 in both cases.

should thus be possible to obtain reliable information about the saturation behavior of these FIR laser transitions at higher FIR intensities on the basis of the results of the theoretical calculations presented here.

The encouraging gain saturation results described here stimulated a careful check on the reliability of the absolute population densities used in [2]. It turned out that the partition function of CH_3OH calculated from thermodynamic functions was a factor of three too low due to neglection of the symmetry number 3 of the CH_3 group. Using a value of the partition function of 14280 (at 300 K), the population densities as well as the theoretical gain values of [2] reduce to one third of their initial values. Finally, there is no longer any significant discrepancy between the experimental and the theoretical gain values of CH_3OH, neither in the small-signal nor in the saturated gain values. Therefore, a complete theoretical analysis of the CW laser pumped FIR laser, especially the CH_3OH laser, should be possible. A density matrix analysis of a traveling wave CW laser pumped FIR laser is currently in progress. An experimental verification will be undertaken in the future.

IV. Conclusions

The measured gain saturations for the 70 μm and the 119 μm CH_3OH lines have been found to be in good agreement with calculations of the quantum-mechanical three-level theory.

It has been shown that, for a coherently excited system as a CW laser pumped FIR laser with mixed broadening, the saturation behavior of the FIR transition cannot be described by one single saturation parameter, but only by a quantum-mechanical treatment. Based on the theory presented, a quantitative analysis of CW laser pumped three-level lasers should be possible. For the short wavelength FIR lasers this is presently in progress.

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References

[1] Z. Drozdowicz, R. J. Temkin, and B. Lax, "Laser-pumped molecular lasers. Part I. Theory," *IEEE J. Quantum Electron.*, vol. QE-15, pp. 170–175, Mar. 1979; —, "Laser-pumped molecular lasers–Part II: Submillimeter Laser Experiments," *IEEE J. Quantum Electron.*, vol. QE-15, pp. 865–869, Sept. 1979.

[2] J. Heppner, C. O. Weiss, U. Hübner, and G. Schinn, "Gain in CW laser pumped FIR laser gases," *IEEE J. Quantum Electron.*, vol. QE-16, pp. 392–402, Apr. 1980.

[3] M. S. Feld and A. Javan, "Laser induced line-narrowing effects in coupled Doppler-broadened transitions," *Phys. Rev.*, vol. 177, pp. 540–562, 1969.

[4] N. Skribanowitz, M. J. Kelly, and M. S. Feld, "New laser technique for the identification of molecular transitions," *Phys. Rev. A*, vol. 6, pp. 2302–2311, 1972.

[5] J. O. Hennigsen, "Assignment of laser lines in optically pumped CH_3OH," *IEEE J. Quantum Electron.*, vol. QE-13, pp. 435–441, June 1977.

[6] J. Heppner, "Far infrared laser resonator of TEM_{00} mode operation," *Appl. Opt.*, vol. 17, pp. 1488–1489, 1978.

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