Far-infrared CH_3F Stark laser

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A rectangular metal–dielectric far-infrared waveguide laser has been operated on the individual Stark components of CH_3F by the application of an electric field across the laser medium. The results agree well with theoretical predictions.

This Letter reports oscillation of a far-infrared (FIR) waveguide laser on individual Stark components of the 496-μm transition in CH_3F by the application of an electric field across the laser medium. Stark shifting of vibrational–rotational absorption lines into coincidence with CO_2-laser pump lines giving rise to FIR oscillation in the rotational transitions was first observed in ammonia.[1] Recently much attention has been given to the effect of electric fields on the strong 496-μm FIR transition in CH_3F; a frequency shift was observed,[2,3] and frequency modulation was achieved by use of the Stark effect.[3] The use of rectangular metal–dielectric waveguides for optically pumped FIR lasers provides a simple and effective way of applying an electric field to the laser medium and has been previously used to enhance the power output of the 119-μm CH_3OH laser line.[4]

A metal–dielectric waveguide laser was constructed of a pair of copper plates 8 mm thick, 64 mm wide, and 2.03 m long, which was optically polished on the wide-waveguide faces. These plates were separated by 6-mm-square pieces of common window glass epoxied to the copper pieces with the smooth window surfaces inside the guide. The glass pieces were separated by 35 mm. A fixed flat copper mirror was placed about 1 mm from the end of the waveguide. A 1-mm input coupling hole was centered in this mirror and was covered with a 2-mm-thick slab of NaCl. A 7-mm-diameter output coupling hole was spaced 14 mm from the center of the mirror and was covered with a 0.5-mm-thick piece of crystal quartz. A movable flat copper mirror was positioned with a micrometer on the far end of the laser. A 25-W, grating controlled, piezoelectrically tuned, cw CO_2 laser was used to drive the FIR laser.

The FIR laser oscillated very well at wavelengths shorter than 600 μm (the output power on the 496-μm line was 3 mW) but refused to oscillate on the strong 700-μm line of methyl alcohol. The calculated losses at 1-mm wavelength are less than 2%/m,[5] and the failure of the 700-μm line to oscillate indicates that the waveguide might be somewhat more lossy than was expected. The polarization of the FIR laser was parallel to the copper surface of the waveguide, as expected. The electric Stark field is, of course, perpendicular to the copper plates; hence the dc Stark field is always perpendicular to the FIR-laser field. The electric field of the pump radiation from the CO_2 laser can be oriented either parallel or perpendicular to the Stark field. Thus there are two cases to consider: (A) pumping transitions with $\Delta M = \pm 1$ and FIR transitions with $\Delta M = \pm 1$ and (B) pumping transitions with $\Delta M = 0$ and FIR transitions with $\Delta M = \pm 1$.

The energy levels of the 496-μm transition in CH_3F are given in Fig. 1. The tabulated matrix elements for the dipole moment of the symmetric top molecule[6] give the relative intensity of the single Stark components of a transition $(J,M) \rightarrow (J-1, M \mp 1)$ as shown by the equations:

$$I \propto \frac{1}{2}\cdot(J \pm M)(J \pm M - 1)[J(J+1) - M^2] \tag{1}$$

for case (A) and

$$I \propto M^2(J \pm M)(J \pm M - 1) \tag{2}$$

for case (B).

The Stark frequency shift of the components (M,

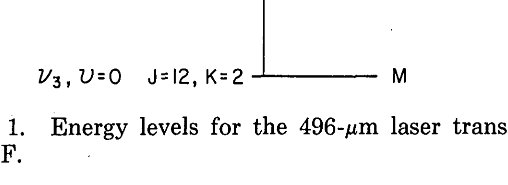

Fig. 1. Energy levels for the 496-μm laser transition in CH_3F.

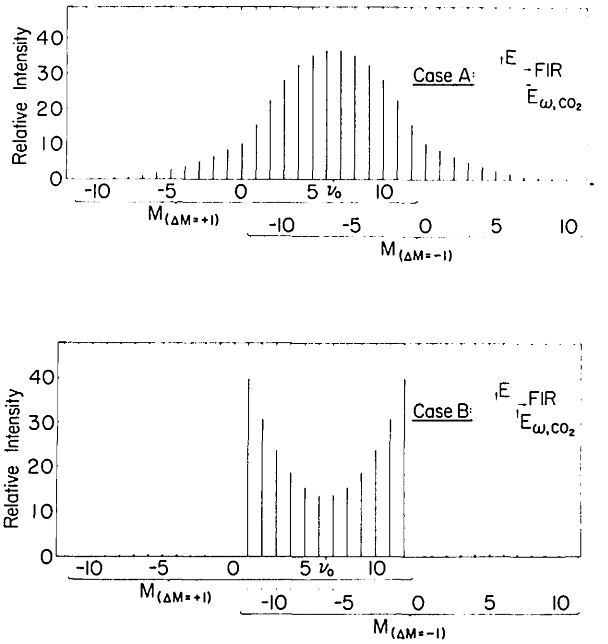

Fig. 2. (a) Intensity pattern of the Stark multiplet for E_{Stark} perpendicular to both E_{CO_2} and E_{FIR} (case A), (b) intensity pattern of the Stark multiplet for E_{CO_2} parallel to E_{Stark} and E_{FIR} perpendicular to E_{Stark} (case B).

ΔM) of the rotational FIR transition in first order is given by

$$\Delta\nu_R = \frac{\mu_e E K}{J(J^2-1)h}\,[2M - (J+1)\Delta M]$$
$$= (2.239\,M - 14.552\,\Delta M)E \text{ (MHz/kV/cm)}, \quad (3)$$

where $J = 12$, $K = 2$, and μ_e (the dipole moment) = 1.9077 D.[7] It follows from Eq. (3) that the 12 lines with $\Delta M = -1$ (from $M = -12$ to $M = -1$) have, respectively, the same frequency shift as the 12 lines with $\Delta M = +1$ (from $M = +1$ to $M = +12$). Higher-order terms in the electric field do not contribute significantly for a field strength of a few kV/cm.

Equations (1) and (2) give the intensity patterns of the Stark multiplets that are shown in Fig. 2. Both patterns are symmetrical with respect to the center frequency, ν_0 (zero electric-field line).

For case (B) only, the 12 degenerate lines have a significant intensity, the strongest being the ($M = 12 \leftarrow 11$) + ($M = 1 \leftarrow 0$) and ($M = -12 \leftarrow -11$) + ($M = 1 \leftarrow 0$). The frequency shifts for these two lines are, respectively,

$$\Delta\nu_R = \pm 12.3 \text{ (MHz/kV/cm)}.$$

Since these two lines have both maximum intensity and maximum frequency shift, they are the most suitable for Stark frequency tuning.

The experimental data are shown in Figs. 3 and 4 for cases (A) and (B), respectively, with methyl fluoride pressures of 30 and 45 mTorr, respectively, and for electric fields of zero and 1.33×10^3 V/cm. The resonant frequency of the laser cavity was scanned by changing the mirror separation by nearly $\lambda/2$. At lower pressures, the gain was insufficient for oscillation, and at higher pressures, the lines were not well resolved. At $V = 0$, four modes were oscillating in the waveguide and complicated the spectrum somewhat. In both cases the frequency separation between the Stark components is in agreement with that predicted by Eq. (3). An asymmetry in the experimental intensity pattern was also observed. This feature may be a direct consequence of the frequency offset between the pumping radiation and the center of the absorption line of CH_3F. The $Q(12,2)$ absorption peak is at +42.5 MHz from the CO_2 9P(20) line center.[8] For the 3.09-m-long CO_2-laser cavity used, the mode spacing is 48.5 MHz. Thus, with the maximum detuning of the CO_2 laser (24 MHz), the CH_3F offset is still 18.2 MHz. (The Doppler FWHM of the CH_3F absorption line is 67 MHz.[8]) The Stark components with different M and ΔM present different offsets from the pumping radiation, as given by the equations[9]:

$$\Delta\nu_v = -0.302\,M \text{ (MHz/kV/cm) for } \Delta M = 0,$$
$$\Delta\nu_v = -0.302(M-1) - 12.3 \text{ (MHz/kV/cm)} \quad \text{for } \Delta M = +1,$$
$$\Delta\nu_v = -0.302(M+1) + 12.3 \text{ (MHz/kV/cm)} \quad \text{for } \Delta M = -1. \quad (4)$$

Since the CO_2 frequency is lower than the $Q(12)$ absorption line of CH_3F, Eqs. (4) indicate that the components with higher M are in closer resonance; therefore the Stark lines at higher frequency are stronger than those at lower frequency, in agreement with the experimentally observed asymmetry. It is worth noting that the remarkable difference between cases (A) and (B) is direct experimental evidence that the collisional randomization between states with different M values in excited levels of the CH_3F is negligible compared with the relaxation between different rotational states. In the presence of a complete collisional randomization between different M states, the Stark intensity pattern would be in both cases like case (B), as shown in Ref. 9.

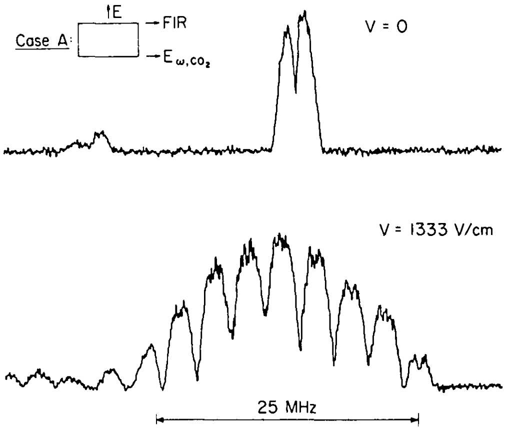

Fig. 3. Experimental Stark effect on the CH_3F laser for E_{Stark} perpendicular to both E_{CO_2} and E_{FIR} (case A); CH_3F pressure 30 μm. The frequency was scanned by increasing the mirror separation (i.e., the high frequency is at the left side of the figure).

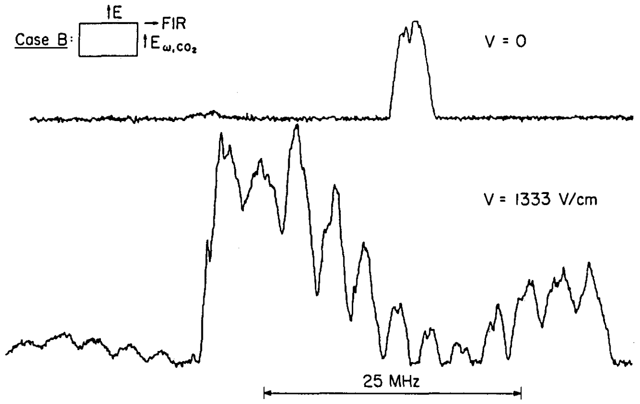

Fig. 4. Experimental Stark effect on the CH_3F laser for E_{CO_2} parallel to E_{Stark} and E_{FIR} perpendicular to E_{Stark} (case B); CH_3F pressure 45 μm. The frequency was scanned by increasing the mirror separation (i.e., the high frequency is at the left side of the figure).

The uniform Stark field in this laser probably made it possible to obtain laser action on individual Stark components while previous experiments[2,3,4,10] failed. This laser still has enough gain for oscillation at electric fields above 1 kV/cm, where the splitting can be easily seen; in previous reports a few hundred V/cm was sufficient to suppress laser action.

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