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CHARACTERISTICS OF THE 615 cm^{-1} SUPERFLUORESCENT EMISSION FROM CO_2 LASER-PUMPED CF_4

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ABSTRACT

A study of superfluorescent emission from CO_2 laser-pumped CF_4 has revealed effects of anomalous dispersion giving rise to large reductions in the group velocity of the superfluorescence. Strong gain modulation of this superfluorescence is reported under conditions where other effects due to saturation of the pump transition are not evident.

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1. INTRODUCTION

We wish to report some preliminary results of an experimental study of superfluorescent emission from an optically pumped molecular laser (OPML) using a single transverse mode CO_2 TEA oscillator-amplifier. In the experiment carbon tetrafluoride (CF_4) was optically pumped with a CO_2 laser operating on its 9 μm R(12) line. Under similar conditions superfluorescent emission at 615 cm^{-1} from CF_4 has already been reported$^{(1,2)}$, but in the present experiment we have compared in detail the temporal structure of the superfluorescent and CO_2 laser pulses under well defined conditions. Superfluorescent pulses travelling both in the same and in the opposite direction to the CO_2 laser pulse have been observed; both pulses were delayed (by different amounts) with respect to the leading edge of the pumping pulse, and both pulses showed modulation with the same periodicity as the mode structure of the pumping pulse. In this paper we examine the fine structure of the superfluorescent pulses and show that the delay of these pulses is consistent with strong effects of anomalous dispersion.

2. EXPERIMENTAL ARRANGEMENT

Figure 1 shows the experimental arrangement. The output from a grating-tuned TEA CO_2 oscillator, suitably apertured for single transverse mode operation, made three passes through a large aperture amplifier (1000 x 100 x 50mm). With the oscillator running on a $1CO_2:1N_2:6He$ mixture, an energy of ∿ 3J was achieved on the 9 μm R(12) line. The beam then made two passes of a cryogenic cell containing CF_4 at a pressure of 4 torr and a temperature of 140 ± 5K along its 2.5 m length. The CO_2 beam diameter within the cell was only 10 mm ($1/e^2$ intensity points) and this allowed the ingoing and return CO_2 pulses to be spatially separated within the 38 mm bore of the cell.

The CO_2 laser and co-propagating superfluorescent pulses emerging from the cell were spatially separated using suitable beam splitters and filters (a LiF_2 reststrahlen reflector and InSb filter to isolate the 615 cm^{-1} component, and a CaF_2 filter to isolate the CO_2 laser components) as shown in Figure 1. The two components were monitored separately on photon drag detectors. Similarly, a KBr beamsplitter isolated a fraction of the counter-propagating superfluorescent component, which was directed onto a photon drag detector through an

InSb filter. The photon drag signals were compared on a dual beam oscilloscope, with the two channels synchronised to within 1 ns. The combined detector plus oscilloscope rise time was calculated to be 1.8 ns in all cases.

3. INITIAL OBSERVATIONS

Figure 2 compares the time resolved CO_2 pumping pulse and the co-progagating and counter-propagating components of the superfluorescent emission from the cell under optimum conditions (4 torr CF_4 at 140K). The detection threshold intensity for the counter-propagating component emerging from the cell is reached approximately 50ns after the peak of the CO_2 pumping pulse has completed a double pass of the cell. The co-propagating pulse is delayed a further 50ns. Both superfluorescent pulses display a temporal structure with a periodicity matching that of the pump laser.

The intensity of both co- and counter-propagating superfluorescent pulses was erratic, with emission being detected in only one shot in five of the pump laser. The shot-to-shot variations in energy of the superfluorescent components were correlated, though occasionally only one component would be (weakly) observed, and very rarely (about one shot in fifty) the co- progagating pulse energy would rise to ∿ 5mJ (compared to an average value of ∿ 1mJ for both components).

4. DELAY OF THE SUPERFLUORESCENCE

The observed delay between the onset of the pumping pulse and the observation of superfluorescent emission remained approximately constant, and independent of both pump energy and superfluorescent output energy.

To achieve superfluorescent output from a 5m gain length, a length-averaged gain of ∿ 8% cm^{-1} is required. With such a high gain in a gas at low pressure and temperature, anomalous dispersion effects may be expected to be large, giving rise to a group velocity much less than the velocity of light. However, for CF_4 gas optically pumped under similar conditions, but in a resonator configuration, similar delays have been reported$^{(1,3)}$ which could in those cases be attributed simply to a build-up time and possibly a lower gain coefficient (a value ∿3% cm^{-1} has been estimated$^{(4)}$).

The possibility of resonant feedback in the present experiment was examined carefully. The inner wall of the cryogenic cell was corrugated to minimise stray reflections, a BaF_2 filter was inserted between the CO_2 laser and the cell to prevent feedback of 615 cm^{-1} radiation, and the surfaces of all windows, filters and detectors were tilted away from normal incidence. Finally, the superfluorescent emission was monitored as various optical components were covered and in no cases were the characteristics of the superfluorescence changed by this procedure. Also, the different shapes of the co- and counter-propagating pulses would be difficult to explain in terms of a resonant feedback mechanism.

5. A PLASMA CUT-OFF EXPERIMENT

In an auxiliary experiment, a plasma cut-off cell was installed between the CO_2 pump laser and the cryogenic cell. The cut-off cell comprised a tube of length 50 cm terminated at both ends with 25 cm focal length converging lenses. By suitable choice of gas pressure in the cell, the intensity at which optical breakdown occurred could be varied. With $\sim$ 70 torr helium in the cell, delays of $\lesssim$ 150 ns could be achieved, and so only on a few occasions was co-propagating CF_4 superfluorescence observed with this experimental arrangement. The detector response is shown in Fig 3 where the vertical arrows indicate the point in time at which plasma cut-off of the CO_2 laser pulse occurred.

Unfortunately, the relationship between the superfluorescent intensity and CO_2 laser-pumping intensity is in general difficult to quantify because a change in pump intensity can produce a change both in the magnitude and in the spectral width (and shape) of the OPML gain. Fast termination of the pump intensity under strongly saturating conditions could lead to a rapid reduction in the gain bandwidth, which may more than offset the effect of the fall in upper laser level population, provided the relaxation of the upper level is sufficiently slow. The results presented in Figure 3 indicate that the conditions required for gain enhancement with fast pump termination are not fulfilled in the present case.

The speed of termination of the superfluorescent emission following cut-off of the pumping pulse gives an indication of the gain lifetime, i.e. the lifetime of the upper laser level. A measured room temperature pressure broadening coefficient of 6MHz $torr^{-1}$ (5) implies a rotational lifetime of 9 ns under the present operating conditions, and this value is consistent with the results in Figure 3. In particular, in all cases the superfluorescence extends no more than 10 ns beyond the point of pump termination. Clearly, this observation alone rules out the unlikely possibility that the upper rotational laser level achieves thermal equilibrium with its vibrational manifold. It is also interesting to note that the decay rate of superfluorescent intensity increases with the value of the superfluorescent intensity at the moment of pump termination. In curve 3(d) the decay rate is consistent with the 9 ns collisional relaxation time of the upper laser level, while in curves (a) to (c) the relaxation is faster, because stimulated emission (at these higher superfluorescent intensities) plays an important role in the relaxation of the upper laser level.

6. ANOMALOUS DISPERSION

There have been several reports of optical pulses travelling through gain media with a group velocity less than the free space velocity(6,7) For the simple case of a medium with a peak gain g_o (at frequency ν_o) and a homogeneously broadened linewidth $\Delta\nu_h$, through which is propagating a pulse of frequency ν_o with a width much less than $\Delta\nu_h$, the delay Δt of the pulse with respect to a pulse travelling with the free space velocity is given simply by(7)

$$\Delta t = \ell g_o / 2\pi\Delta\nu_h \qquad (1)$$

where ℓ is the gain length.

In OPML's generally, the optimum operating pressure corresponds to the condition that the Doppler and pressure broadened contributions to the width of the laser transition are approximately equal(8). Fortunately, in the case of a gain medium for which Doppler broadening is dominant, the equation for Δt is very similar to equation (1), but with the value of Δt reduced by a factor of 0.94(6). Consequently, it is reasonably accurate to use equation (1) for the general case in which the gain lineshape is best approximated by a Voigt profile. It therefore follows that since the observation of a 1mJ pulse requires a 'ℓg_o' product of ∿ 40 (assuming the process to be started by a single 615 cm^{-1} photon), the expected delay of the superfluorescent pulse will be

$$\Delta t \simeq 6.4/\Delta\nu \qquad (2)$$

with $\Delta\nu$ the total gain bandwidth (FWHM).

In evaluating the delay of the co- and counter-propagating superfluorescent pulses it is important to include the change in apparent Doppler width of the gain in the two direction, which occurs only because the processes of absorption and superfluorescent emission in the CF_4 gas take place under collisionless conditions(9). The equivalent Doppler broadening contributions $\Delta\nu_f$ and $\Delta\nu_b$ for the co- and counter-propagating components are, respectively

$$\Delta\nu_f = \Delta\nu_D \left(\frac{\nu_p - \nu_s}{\nu_s}\right) \qquad (3)$$

$$\Delta\nu_b = \Delta\nu_D \left(\frac{\nu_p + \nu_s}{\nu_s}\right) \qquad (4)$$

where $\Delta\nu_D$ is the Doppler width of the laser transition and ν_p and ν_s are the frequencies of the pump and superfluorescent transitions respectively.

Taking a value of 17 MHz for the Doppler width ($\Delta\nu_D$) and computing the Voigt profile width for a 42 MHz pressure broadening contribution(5), values of $\Delta\nu$ for the co- and counter-propagating superfluorescent components are 38 MHz and 68 MHz respectively. Using these values, equation (2) gives values of 168 ns and 94 ns for the delays for the co- and counter-propagating components respectively, corresponding favourably with the observed values of 150 ± 20 ns and 100 ± 20 ns (measured from the leading edges of the pulses, and allowing for the transit time in the cell in the case of the counter-propagating component). It is reasonable to assume that the CO_2 pumping pulse will travel with the free space group velocity in the CF_4 gas since the absorption coefficient is so low. The delay of the co-propagating superfluorescent component corresponds to an average velocity of only ∿ 0.1c, smaller even than the value of 0.4c measured by Casperson and Yariv in a high gain xenon laser(6).

7. GAIN MODULATION EFFECTS

In the simplified analysis of anomalous dispersion effects presented above, no account was taken of power broadening of the gain spectrum; and the good agreement between experimental results and theoretical predictions indicates that this assumption was justified. Other justifications for neglecting power broadening follow from the observations that the probability of observing superfluorescent emission decreased monotonically as the CO_2 laser-energy delivered to the cell was decreased, and that the behaviour of the superfluorescent emission was erratic. This erratic behaviour, a 1 in 5 probability of superfluorescent emission being observed (see section 3) may be associated with a requirement for a coincidence between the centre of the CF_4 absorption feature and an axial mode of the CO_2 laser of only ± 8 MHz (i.e. ± 1/10th of the free spectral range of the CO_2 laser oscillator). Both of these observations imply that the pump transition is not saturated by the CO_2 laser, and it might therefore reasonably be expected that the superfluorescent output should have a temporal structure reflecting only the relatively smooth variation in intensity of any one of the individual axial modes of the CO_2 laser (i.e. a $\sim$ 60 ns FWHM 'spike' followed by a low intensity 'tail' of $\sim$ 2μs duration). Nevertheless, Figure 2 clearly shows a mode structure on the superfluorescent output.

Details of the mode structure in the CF_4 superfluorescence are shown in Figure 4 where, on an expanded time-scale, various CO_2 laser and superfluorescent pulse structures are compared. The mode structure on the CO_2 laser pulse is shown to vary shot-to-shot, as is characteristic of a multi-axial mode laser; yet the periodicity in the structure of both the CO_2 and superfluorescent pulses remains the same, equal to the round-trip time in the CO_2 oscillator.

It is clear form Figure 4 that while there is a similarity between the temporal structure of the CO_2 laser and superfluorescent signals, the peaks in the two pulses are not always coincident, nor is the modulation of the superfluorescent signal as great as that in the CO_2 laser pulse. This latter observation, confirmed by the results presented in section 5, implies that the bandwidth of the superfluorescent pulse is less than that of the CO_2 laser pulse (i.e. it comprises fewer modes) while the two observations taken together give a valuable insight into the temporal behaviour of the OPML gain.

It should be expected that once a strong superfluorescent field is established, the interaction of the combined pump plus superfluorescent fields with the molecule will change. For example, dynamic Stark splitting of the upper laser level by the superfluorescent field may become large compared to the spacing of modes in the pump laser, or 4-wave mixing may occur between two pumping modes (one being the mode in resonance) and the superfluorescent field. Either possibility could give rise to a spectrally broad superfluorescent output, but in each case a necessary requirement is the establishment of a strong radiation field at the line centre of the 615 cm^{-1} transition. However, it should be borne in mind that the depth of modulation due to mode beating can be considerable even when the dominant mode is orders of magnitude more intense than the adjacent modes. For example, if only two modes were beating to produce the modulation in Figure (4a) their intensity ratio would be 35:1.

It can be argued intuitively that once gain saturation has been achieved then the group velocity of the superfluorescence at line centre will approach the free space value i.e. it will travel at approximately the same velocity as the superfluorescence generated away from line-centre.

8. CONCLUSIONS

Temporally resolved measurements of the superfluorescent emission from CF_4 gas pumped by a single transverse mode CO_2 pump laser under non-saturating conditions have revealed a delay in the superfluorescent emission which has been shown to be consistent with strong anomalous dispersion effects.

The importance of dispersion in OPML's has previously been reported; both in producing self-focusing or defocusing of the pump laser field$^{(10)}$ and in giving rise to cavity detuning effects$^{(11)}$. The effect on the group velocity of a pulse being amplified in an OPML gain medium has hitherto not been reported, although it is immediately apparent from equation (1) that under the high gain/narrow bandwidth conditions in a weakly pumped OPML at low pressure and temperature, the effect should be strong. The measured delay of ∿ 150ns in the present case would correspond to an average group velocity much less than the smallest value previously reported for a gaseous gain medium$^{(6)}$. However, it should be noted that the conditions in the present experiment are exceptional. In general the gain spectrum of an OPML will be complicated by strong power broadening of the pump transition. Under these conditions the value of $\Delta\nu$ in equation (2) may be much greater than in the present case, and/or the gain spectrum may not be symmetrical.

Then not only will the effects of anomalous dispersion be much reduced, but power broadening may allow several modes of the pump laser to interact strongly with the molecule without the assistance of the generated OPML field. Under such conditions the observation of gain modulation would be of little significance(12). The results described in section (7) indicate that even for weak pumping of a narrow absorption line the OPML bandwidth can be relatively large (i.e. several times the spacing of adjacent modes of the pump laser).

From the above discussion it is apparent that the advantages of travelling wave excitation (especially that of short pulse generation) can only be realised when the gain bandwidth is sufficiently large (either by virtue of the intrinsic properties of the molecule, collisional broadening, or strong saturation of the pump transition) or when strong off-resonance pumping leads to Raman-like lasing. It is also possible that the generally long ($\gtrsim$ 100ns) build-up time of an OPML may be due to anomalous dispersion effects rather than low gain coefficient, and conversely that a short (<<100ns) build-up time may correspond to the OPML pump transition being strongly saturated.

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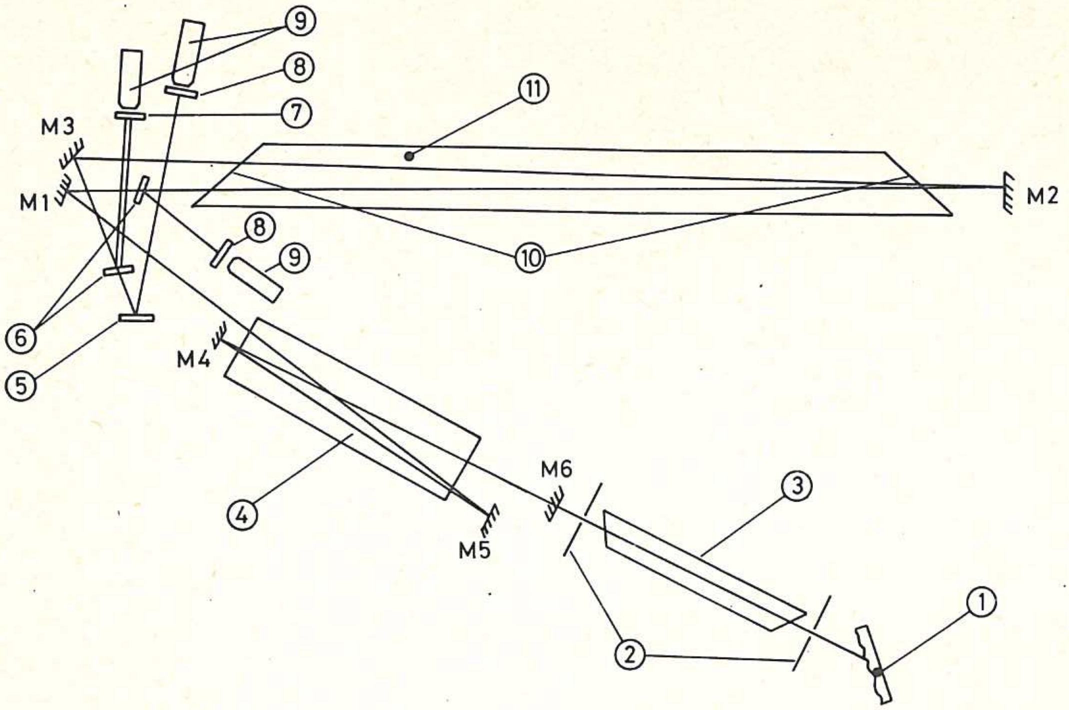

Fig.1 Experimental arrangement. M1 to M5 are plane copper mirrors, M6 is a zero power germanium lens, 4m radius of curvature anti-reflection coated on the rear (convex) surface. The numbers refer to:

(1) 150 lines/mm diffraction grating
(2) iris diaphragms set at 10mm
(3) TEA CO_2 laser oscillator
(4) TEA CO_2 amplifier
(5) LiF_2 restrahlen reflector
(6) KBr beamsplitters
(7) 2mm thick CaF_2 filter
(8) InSb filters
(9) photon drag detectors
(10) KBr windows at Brewster's angle
(11) cryogenic cell

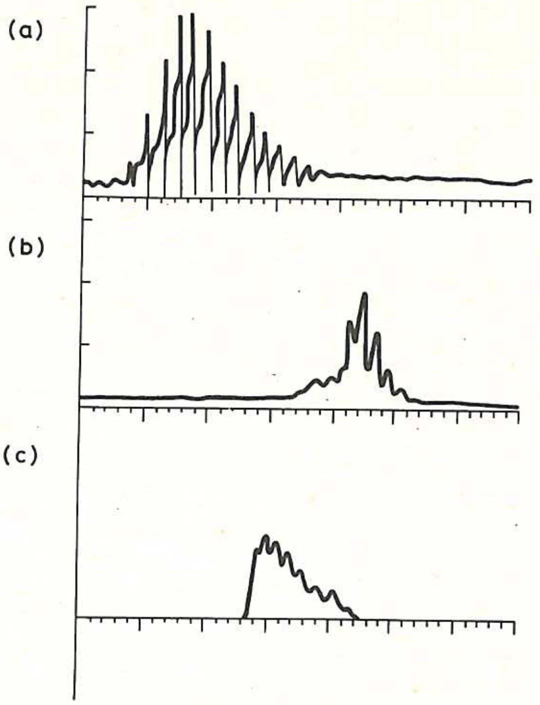

Fig.2 Output of photon drag detectors monitoring
(a) transmitted CO_2 laser intensity (positive going signal) e
(b) co-propagating CF_4 superfluorescent signal and
(c) counter-propagating CF_4 superfluorescent signal.
1 large division = 50ns.

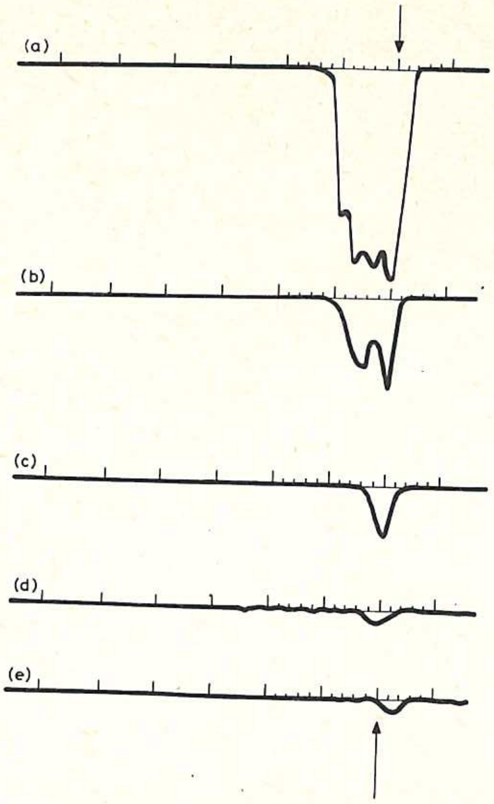

Fig.3 Five examples showing the effect of plasma cut-off of CO_2 laser pumping pulse on co-propagating CF_4 superfluorescent signal. Vertical arrows indicate cut-off point. One large horizontal division = 20ns.

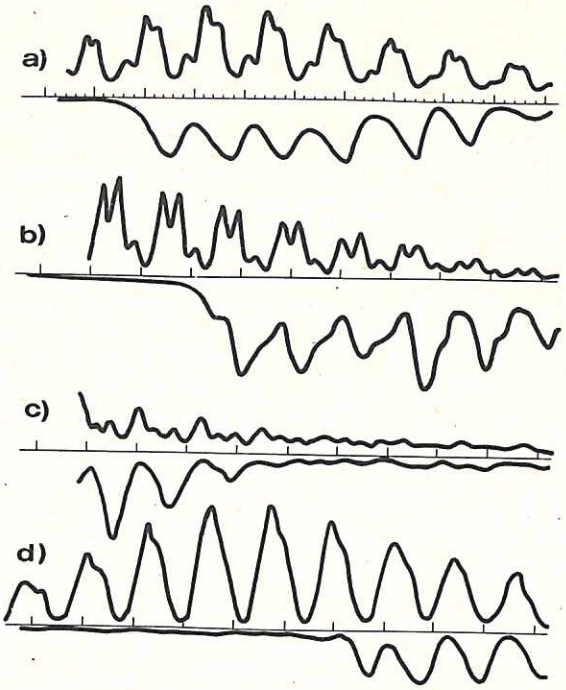

Fig.4 Four sets of photon drag signals showing details of mode beating in the CO_2 laser pulse (positive going signal) and co-propagating CF_4 superfluorescent signal. One large horizontal division = 10ns.