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OPTICAL PUMPING OF CARBON TETRAFLUORIDE BY A FINE-TUNABLE SINGLE MODE CO_2 TEA OSCILLATOR-AMPLIFIER

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ABSTRACT

Superfluorescent emission from carbon tetrafluoride was generated by a single mode CO_2 TEA laser whose frequency was swept over ± 30MHz of the 9R(12) laser line centre. The results confirm the spectroscopic identification of the CF_4 pump transition while the dependence of threshold pump energy on the exact frequency of the CO_2 laser is found to agree with theoretical estimates.

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1. Introduction

Of the many CO_2 laser lines that can generate radiation in the 16μm region by optically pumping the carbon tetrafluoride (CF_4) molecule, the 9R(12) line produces the strongest 16μm emission. High resolution diode spectroscopy has identified an absorption line in CF_4 to lie between 20 and 30MHz to the high frequency side of the 9R(12) line centre[1], and this would explain the striking improvements in the CF_4 output that have been made by line narrowing the output of a TEA CO_2 laser to a single transverse, single-axial mode by incorporating a low pressure CO_2 discharge section into its cavity[2,3]. Using such a hybrid CO_2 TEA laser operating on the 9R(12) line we have generated powerful 615 cm^{-1} superfluorescent emission from CF_4, but have also stabilised and piezoelectrically controlled the cavity length. In this way, the dependence of CF_4 superfluorescence on pump laser frequency over an 80MHz region (corresponding to the free spectral range of the CO_2 laser cavity) has been observed.

2. Experimental Arrangement

The experimental arrangement comprised a tunable single mode TEA oscillator, a large aperture TEA amplifier through which the oscillator output made three passes, and a 2.5m long x 38mm i.d. precision cryogenic cell containing CF_4 gas at 4 torr pressure and 140K temperature through which the CO_2 laser beam made two passes. Details of the experimental layout have been described in an earlier publication[4].

The TEA oscillator cavity was defined by a plane diffraction grating and uncoated Germanium output mirror separated by 1.8m. At either end of the cavity were two 10mm diameter apertures for single transverse mode control, and between the apertures was a 1m long TEA discharge section of 20 x 20mm cross section and a 25cm long x 12mm diameter bore low pressure longitudinal discharge section through which a 6:1:1 ($He:N_2:CO_2$) gas mix flowed at a mean pressure of 15 torr. The energy for the low pressure discharge was stored in a 6.3nF capacitor charged to 5kV, which was discharged some 90μs before the TEA oscillator discharge was initiated. The output mirror of the TEA oscillator was mounted from a precision translation stage onto a heavy iron optical rail. In this way, the mechanical stability of the cavity was high while at the same time the cavity length was held constant by inserting an invar bar held by spring pressure between the grating and output mirror. Variation in cavity length over 6μm was achieved by holding the mirror in a commercial piezoelectric translator which was attached to the mirror mount.

3. CO_2 Laser Performance

A voltage of 760V on the piezoelectric translator was sufficient to change the length of the oscillator cavity by a free spectral range (a distance of 4.659μm for the 9R(12) line), corresponding to a frequency shift of 82MHz. Over approximately half of this range the output laser pulse was single mode with a pulse shape as shown in Figure (1a), while over the remainder of the range the laser output comprised two modes which would beat together to produce a pulse shape such as shown in Figure (1b). However, the CO_2 laser energy remained approximately constant at 1.5J over the full tuning range.

Figure 2 shows a plot of calculated frequency shift from the CO_2 9R(12) line centre as a function of the voltage applied to the piezoelectric translator, as inferred from interferometric measurements of the translation of the output mirror. The voltages at which two adjacent modes of the oscillator cavity were equally spaced either side of the centre of the laser line were assumed to correspond to 100% mode-beating modulation on the output laser pulse. This method of inferring the position of the laser line centre was found to be more accurate than monitoring the optoacoustic signal in CO_2-laser excited CO_2 gas.

4. CF_4 Superfluorescence

As previously described, the 12mm diameter pulses from the CO_2 laser made two passes of the cryogenic cell containing CF_4, with the input and output pulses being spatially separate within most of the cell. Strong CF_4 superfluorescence was generated in the form of two pulses of approximately equal energies, one counter propagating and the other co-propagating with respect to the CO_2 pulses. Figure 3 shows the variation in superfluorescent pulse energy vs. voltage applied to the piezoelectric translator. Three sets of curves are shown, corresponding to different

CO_2 pulse energies.

The shot-to-shot variation of superfluorescent pulse energy is reflected in the size of error bars in Figure 3. This variation is much less than was observed when a multimode CO_2 TEA laser of comparable energy was used to pump the CF_4 gas[4], and can be attributed to fluctuation in the CO_2 laser pulse energy. From Figures 2 and 3, the optimum pump frequency for CF_4 is estimated to be 23 ± 5MHz to the high frequency side of the 9R(12) line centre, in agreement with earlier spectroscopic measurements[1]. However, by measuring superfluorescent emission instead of absorption in CF_4, the identification of the pump transition as being the closest absorption feature to the 9R(12) line is positively confirmed. Indirectly this result implies that optimum pumping does indeed occur at the line centre of the pump transition.

Although the shape of the curves in Figure 3 are difficult to interpret quantitatively, they nevertheless reveal immediately the stringent frequency matching requirements for the CO_2 laser to the pump transition in CF_4. But the variation with CO_2 laser energy of the frequency mismatch at which the CF_4 superfluorescent energy reaches detector threshold can be estimated in a semi-quantitative manner using the full quantum mechanical model of optically pumped laser operation.

The theoretical expression for the maximum gain for a given pump intensity can be greatly simplified by making the reasonable assumptions that, for conditions under which the CF_4 superfluorescent intensity only reaches detector threshold, both the pump intensity and the superfluorescent intensity are insufficient to saturate the pump and laser transitions respectively. Under these conditions, a gain maximum will always be at the line centre of the laser transition in the molecule (although another maximum of equal magnitude will be located at the Raman frequency in the case of off-resonance pumping). Under conditions where the pump and laser transitions in CF_4 are homogeneously broadened, with a relaxation time τ, the maximum gain (g_o) is then given by[5]:

$$g_o = \frac{\alpha \tau I}{\delta^2} \{1 + (1 + \delta^2\tau^2)^{-1} - 2(1 + \delta^2\tau^2)^{-2}\} \qquad (1)$$

Where I is the pump laser intensity, δ is the amount by which the pump laser is off-resonance with the pump transition, and α is a constant determined by the properties of the CF_4 molecule including the equilibrium populations in the molecular levels involved.

Unfortunately, under conditions of optimum gain, the pump and laser transitions are Doppler broadened and pressure broadened to approximately equal extents. In the present experiment the total linewidth (FWHM) of the pump transition and its homogeneous contribution is estimated to be 52.6MHz and 34.7MHz respectively. It might be expected that close to line centre (small δ) a value of τ corresponding to a 52.6MHz homogeneous linewidth would be the most appropriate value to substitute into equation (1) (since this would in some way account for the effect of Doppler broadening) while for larger values of δ the true relaxation time should be used. This is indeed found to be the case in Figure 4 where the threshold pump energy is plotted against frequency offset, δ. The points are the experimentally measured values while the two solid curves were derived from equation (1) (assuming the threshold energy to be proportional to threshold intensity) using the two different values for τ and normalising both curves to agree with the threshold energy value at zero frequency shift. The experimental points are seen to fall close first to one curve and then to the other as δ increases from zero, in the manner predicted above.

It is interesting to note that the dependence of threshold energy (or intensity) on frequency offset δ is much stronger than might be anticipated from the width of the pump transition in CF_4. This is illustrated in Figure 4 by the dashed curve in which the threshold intensity was estimated on the assumption that the maximum gain (g_o) is simply proportional to the product of the pump intensity and the absorption coefficient of the pump transition at the pump frequency (i.e. shifted an amount δ from line centre) assuming a Voigt profile with the estimated width of 52.6MHz. This dashed curve is seen to greatly underestimate the threshold energy dependence on frequency offset.

5. Conclusions

The construction and operation of a fine-tunable single mode CO_2 TEA oscillator/amplifier system has been described. The close frequency coincidence of an absorption line in CF_4 with the 9R(12) laser line in CO_2 has allowed measurement to be made of CF_4 superfluorescent emission as a function of frequency offset from 0 to ± 30MHz. The results confirmed a previous identification of the CF_4 pump transition, while measurements of the dependence of threshold pump energy with frequency offset have been found to be in semi-quantitative agreement with theoretical predictions.

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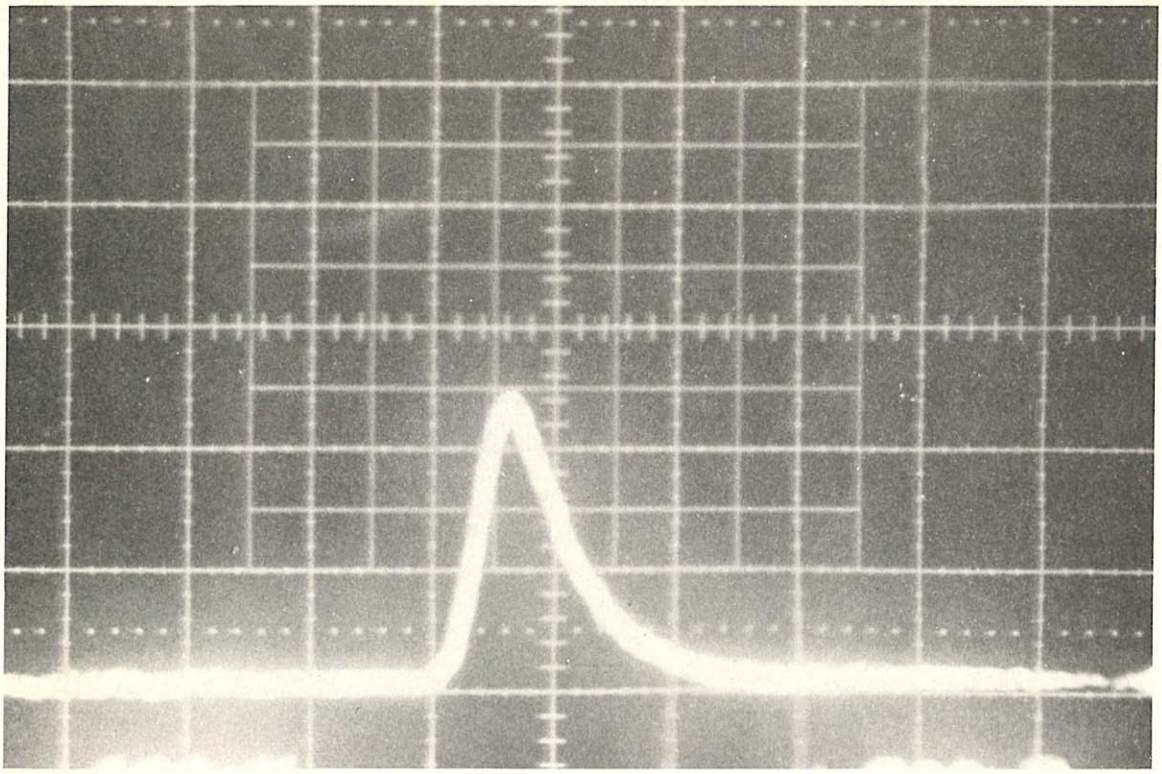
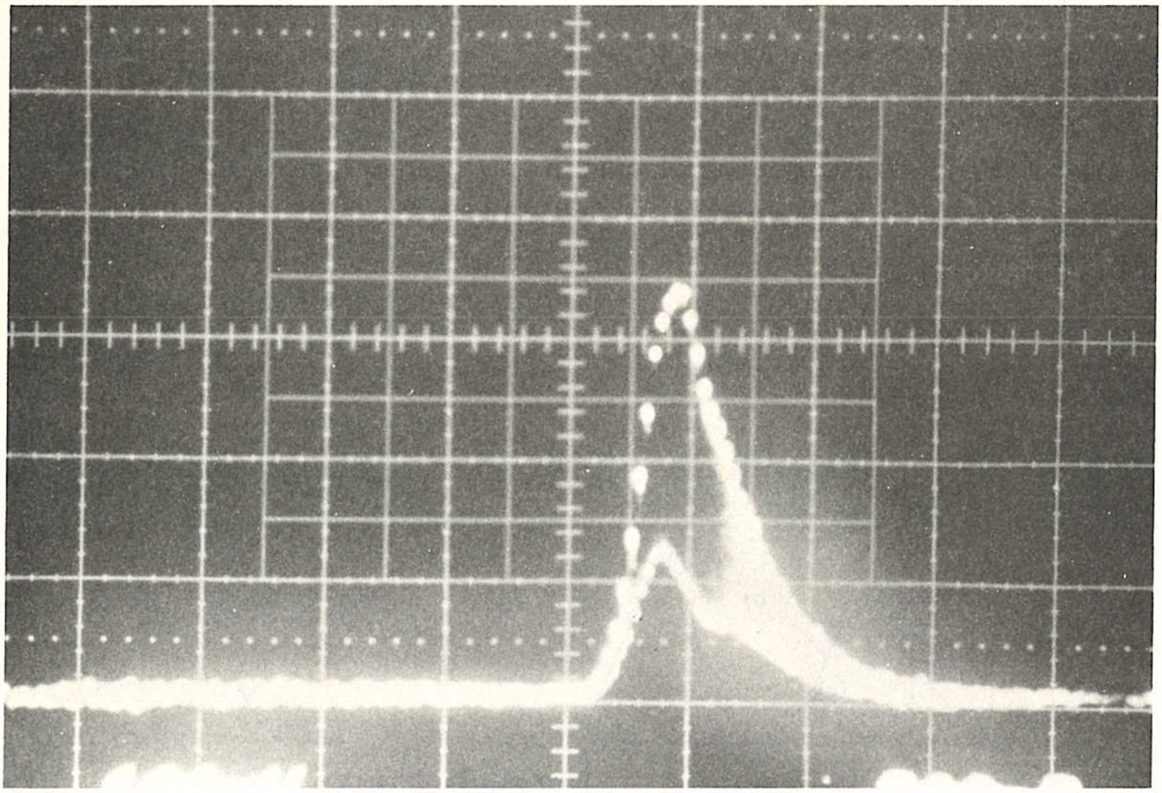
References

1. L.J. Radziemski, Jr., R.F. Begley, H. Flicker, N.G. Nereson and M.J. Riesfeld. 'Spectroscopy of $^{12}CF_4$ Features Near the 9μm $^{12}C^{16}O_2$ R(10), R(12) and R(18) Lines by CO_2-Laser and Diode Spectroscopy" Opt. Lett. 3, 241-243 (1978).
2. A. Stein, P. Rabinowitz and A. Kaldor. 'Radiance Enhancement of the 16μm CF_4 Laser' Opt. Lett. 3, 97-99 (1978).
3. J.J. Tiee, T.A. Fischer and C. Wittig. 'High-energy Optically Pumped Molecular Lasers in the 13 and 16μm Regions' Rev. Sci. Instrum. 50 958-961 (1979).
4. J.M. Green, T. Stamatakis and D.A. Aldcroft. 'Characteristics of the 615cm^{-1} Superfluorescent Emission from CO_2 Laser-pumped CF_4' Opt. Commun. 39 99-104 (1981).
5. R.J. Temkin. 'Theory of Optically Pumped Submillimeter Lasers' IEEE J. Quantum Electron. QE-13 450-454 (1977).



(a)



(b)

Fig.1 Pulse shape of output from CO_2 TEA oscillator-amplifier system as monitored by a photon drag detector, under single mode operation (a) and two-mode operation (b). Time scale 200 ns/division.

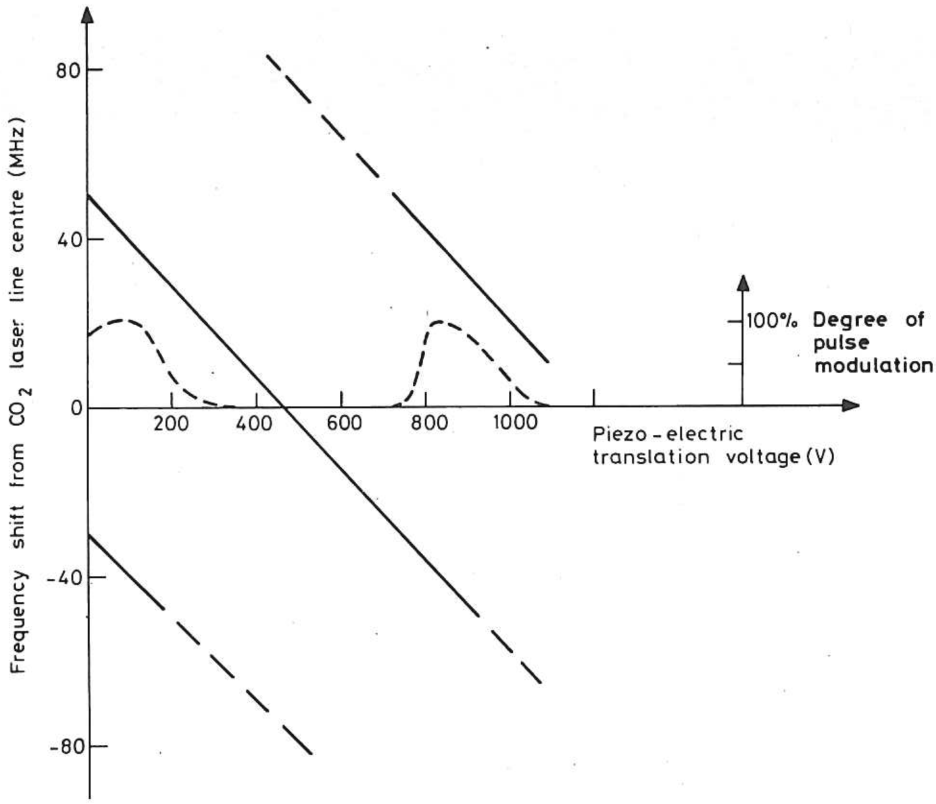


Fig.2 Calculated frequency shift of laser output from line centre as a function of the voltage applied to the piezoelectric translator on the CO_2 oscillator. Lines show calculated positions of adjacent axial modes, and are solid over regions where that mode is lasing. Dashed curves show the measured degree of modulation (mode heating) in the laser pulse and refer to the right hand side ordinate scale.

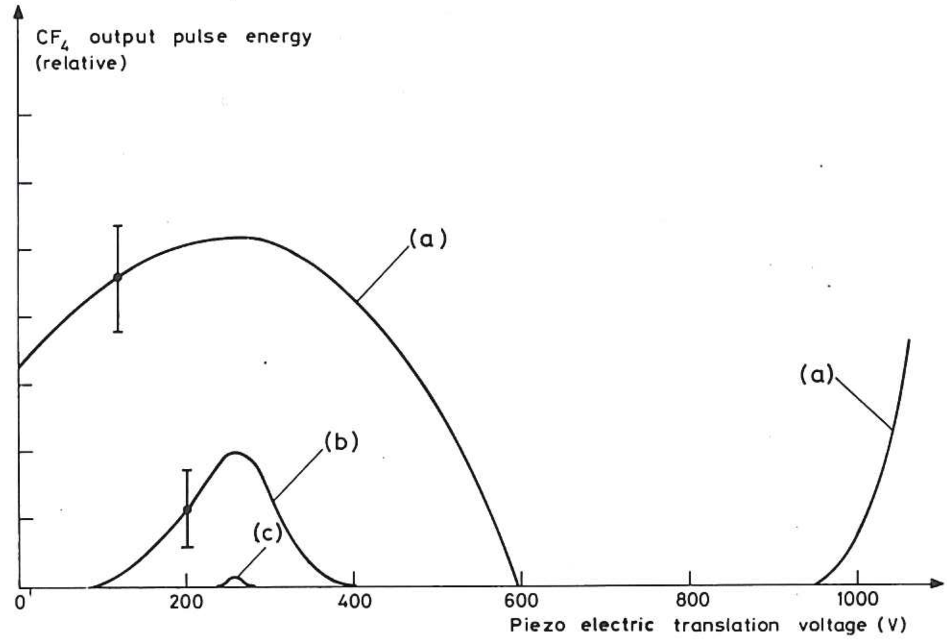


Fig.3 Variation of CF_4 laser output with voltage applied to the piezoelectric translator on the CO_2 oscillator. Curves refer to varying attenuation of the CO_2 pumping pulse: (a) full transmission, (b) 41% transmission, (c) 14% transmission.

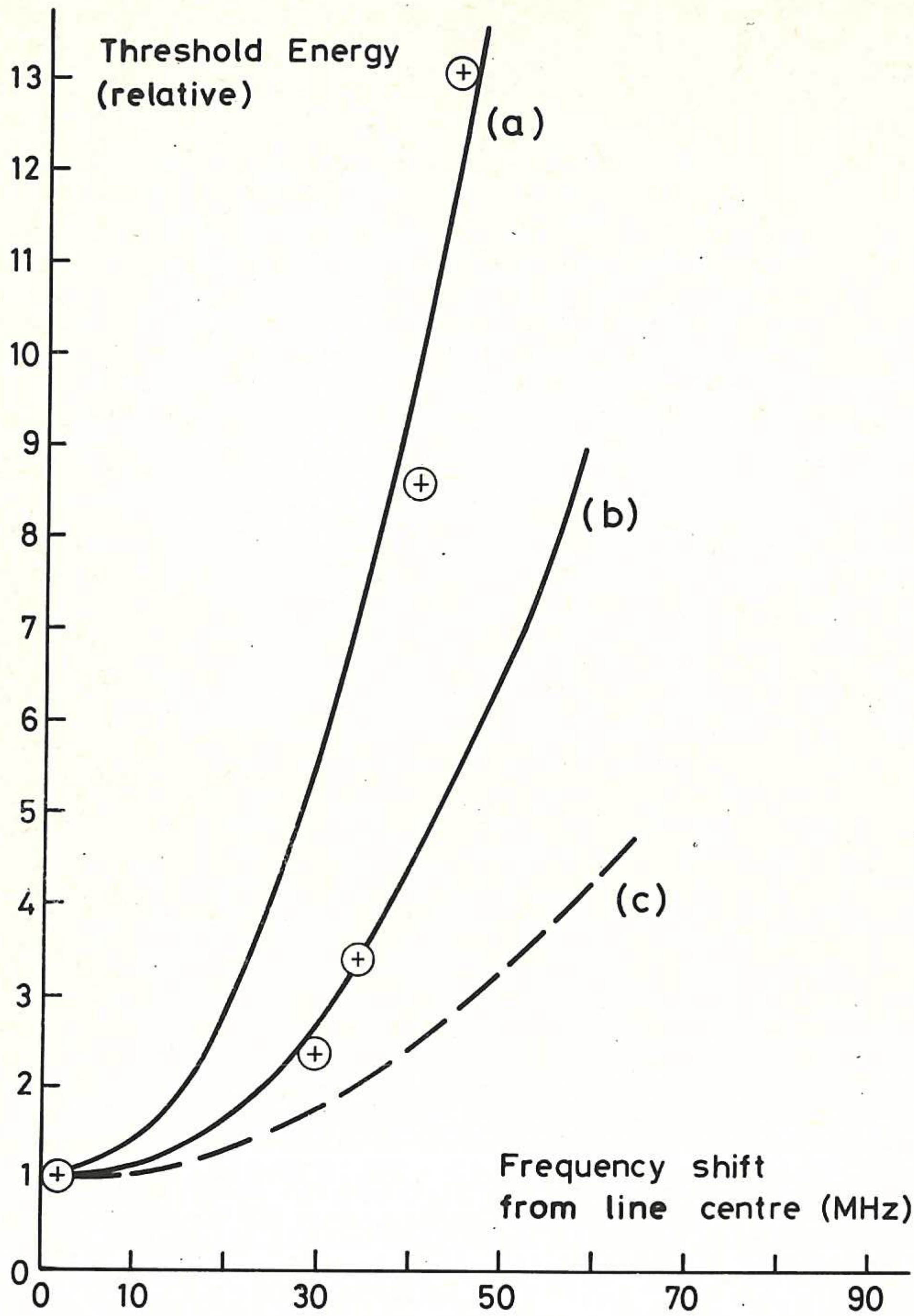


Fig.4 Variation of threshold pump energy for CF_4 laser operation with frequency shift of the CO_2 pump laser from line centre. Points show experimental results. Curves (a) and (b) are theoretical curves generated using Equation (1) with values for the rotational relaxation time corresponding to homogeneous linewidths for the pump transition of 34·7 MHz respectively. Curve (c) shows the predicted gain assuming it to be proportional to the absorption coefficient of the pump transition at the frequency of the pump laser.