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SOME APPLICATIONS OF POLARISATION PHENOMENA IN OPTICALLY PUMPED MOLECULAR LASERS

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Abstract

When an optically pumped molecular laser is pumped with linearly polarised light, an orientational anisotropy of excited molecules is produced. We present ways in which this phenomena may be exploited in the estimation of excited state densities in optically pumped molecular lasers and in the design of optically pumped molecular amplifiers.

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1. Introduction

When molecules absorb energy from linearly polarised light by resonant absorption on an electric dipole transition, the energy absorption rate by any molecule depends on the relative orientation of its dipole moment to the plane of polarisation of the light, and so an anisotropic distribution of orientations of excited molecules is produced. This anisotropy is reflected in a partial polarisation of any spontaneous emission from the excited molecules under collision-free conditions. However, since rotation of molecules takes place at a time scale typically many orders of magnitude faster than spontaneous emission, the degree of polarisation of the spontaneous emission depends on the types of transitions (P,Q or R) involved in the optical pumping and emission processes (Zare, 1966).

A manifestation of this polarisation phenomena is the general observation that when an optically pumped molecular laser (OPML), either pure rotational or roto-vibrational, is pumped with linearly polarised light, the OPML output is also linearly polarised parallel or perpendicular to the plane of polarisation of the pump radiation. In this case the orientational anisotropy of excited molecules produced by linearly polarised pump radiation effectively leads to an anisotropy in stimulated emission cross section for the OPML transition. Since OPML's are invariably operated under collisional conditions, the polarisation phenomena is only observed because orientation changing collisions generally also change the rotational quantum number (J) of the molecule, so that molecules undergoing such collisions no longer participate in the laser process. However, the situation can be complicated by optical saturation of the pump and/or laser transition, which has the effect of reducing the anisotropy (Drozdowicz et. al., 1979). Nevertheless, for purposes of discussion the theoretical results for the case of high J transitions and non-saturating fields are often sufficient, and the results can then be summarised quite simply in terms of the effective transition probability (A) measured by a probe laser beam at the OPML frequency orientated first parallel and then perpendicular to the plane of polarisation of the pump radiation. Using subscripts '11' and 'h' to refer to these two orientations respectively the ratio A_{11}/A_{h} can take on three values, as follows:

$\frac{1}{2}$ for either pumping on a Q transition and lasing on a P or R transition, or pumping on a P or R transition and lasing on a Q transition; $\frac{3}{4}$ for pumping and lasing on R or P transitions, and 3 for pumping and lasing on Q transitions (Chang 1977).

One way in which the above polarisation phenomenon is routinely used is to assign (as P,Q or R) the pump and laser transitions involved in an OPML. Here we present other ways in which this phenomenon might be exploited; in the estimation of excited state densities in OPML's and in the design of optically pumped molecule amplifiers (OPMA's). In the case of excited state density measurements, some of the results are illustrated for a 16μm CF_4 (Freon - 14) OPML pumped by the 9μm R(12) and R(22) lines of a CO_2 TEA laser (Tiee and Wittig 1977). For both of these CO_2 laser lines, optical pumping is believed to proceed on a P branch transition of CF_4, but laser action occurs on a P branch line for R(12) pumping and on a Q branch line for R(22) pumping.

2. Excited State Density Measurements

If we make the normally valid assumption that the build up of stimulated emission inside on OPML cavity is slow compared to both the time for a round trip of the cavity and also the rate of change of gain in the laser medium, then we can equate the laser build up time τ to the gain coefficient g(t), the length of the laser cavity ℓ and the reflectivities of the cavity mirrors R_1, R_2, by the simple formulae

$$\tau c \left\{\frac{1}{2\ell}\,\ell n(R_1R_2) + f\,\frac{\int_0^{\tau} g(t)dt}{\tau}\right\} = \gamma \qquad (1)$$

where c is the speed of light, f is the fraction of the cavity length occupied by the gain medium and γ is the exponential factor required to increase spontaneous noise to some (arbitrary) fixed power level of stimulated emission as detected at the output of the OPML. A value $\gamma = 32$ is typical, and varies little for different laser build up conditions.

Equation (1) can be used to estimate the average gain coefficient in an OPML and from this value the time average population inversion density may be deduced if the stimulated emission cross section for the OPML transition is known. Time averaged upper laser level population densities cannot readily be deduced in this way, not only because it is generally difficult to make adequate allowance for the lower laser level population, but also because the interpretation of build up time measurements is complicated by the need to allow for all losses in the laser cavity; for example, in the case of cooled OPML cavities an important loss can be the presence of hot gas at the ends of the OPML cell. These problems can be overcome by introducing a polarisation dependent loss into the laser cavity

If an inclined beam splitter is introduced into the laser cavity, then the reflective losses of its faces will be dependent on the polarisation of the incident light, and by choosing an inclination which produces the highest reflective losses for the polarisation for which the OPML gain is maximised, OPML operation orthogonal to this plane of polarisation may be achieved. At some intermediate inclination of the beam splitter, however, the build-up times for both (orthogonal) polarisations will be the same and the OPML output polarisation will vary from shot to shot. Under these conditions we may write

$$\frac{f\int_0^{\tau} (g_{11} - g_h)\,dt}{\tau} = \frac{1}{\ell} \ln \left(\frac{T_h}{T_{11}}\right) \qquad (2)$$

where we have equated the build up times for both orthogonal polarisations assuming that equation (1) (modified to include the transmission (T) of the beam splitter) holds independently for each polarisation prior to the intracavity OPML power reaching saturation.

The importance of equation (2) lies in the fact that the only losses experienced differently by the two orthogonal polarisations within the OPML cavity are due to the beam splitter. Other losses will not appear in equation (2). Further, since molecules in the lower laser level prior to laser action will exhibit no orientation anisotropy (because molecules in this state are produced largely as a result of thermalising collisions) the difference in gains $(g_{11} - g_h)$ will reflect only the upper laser level population (N_u) i.e.

$$(g_{11} - g_h) = N_u\,(\sigma_{11} - \sigma_h) \qquad (3)$$

where σ is the stimulated emission section for the laser transition, with the ratio σ_{11}/σ_h being proportional to A_{11}/A_h for which values were given previously. Thus equations (2) and (3) can be used either to evaluate an '$N_u\sigma$' product or conversely (if adequate allowance can be made for the lower laser level population prior to laser action and for all losses inside the laser cavity) for the effects of collisions and optical saturation on the σ_{11}/σ_h ratio.

In an experimental check of equation (2), gain measurements were made in a low temperature CF_4 OPML. Fig. 1 shows the experimental arrangement of the OPML cavity. The cavity included a beam splitter on a rotating table in a part of the cavity free from CO_2 pump radiation, and a three-mirror polarisation rotator (Johnston 1977) was situated between the pump laser and OPML cavity. The liquid nitrogen cooled OPML cell has been fully described elsewhere (Green 1979), but in this case the cell window was held at near-normal incidence. A linear polariser and pyroelectric detector were arranged behind the output mirror.

With the experimental arrangement described above, we were able to 'flip' the plane of polarisation of the OPML output through 90° simply by rotating the intracavity beam splitter. Potassium Bromide (KBr) and Cadmium Telluride (CdTe) beam splitters were tried, covering between them a wide range of '$\ell n(\frac{T_{11}}{T_h})$' values. For the R(12) pumped line in CF_4 'flipping' could only be achieved using CdTe as the beam-splitter material, while for the R(22) pumped line a KBr beam-splitter was sufficient. Using the CO_2 laser polarisation rotator to rotate the polarisation of the pump radiation through 90° we were able to verify that any polarisation dependent effect induced on the OPML output by the multiple bounce arrangement of the pump radiation within the OPML cell was negligible. Further, we observed that the wavelength of the OPML output for both CO_2 pump lines remained unchanged when its plane of polarisation was 'flipped'. This result is perhaps surprising in view of the multiplicity of possible transitions from a single upper level in this highly degenerate molecule.

Table (1) compares the results of '$N_u\sigma$' and gain measurements using equations (1) - (3). For '$N_u\sigma$' measurements the table lists the material of the beam splitter and its inclination to the optic axis of the OPML cavity at which 'flipping' occurred. For gain measurements the build-up times given were measured with a HgCdTe detector. Errors are due prinicipally to shot-to-shot variations in the CO_2 pump laser. The laser gas conditions given in the table correspond to those for optimum energy output from the OPML.

Comparing '$N_u\sigma$' and gain measurements we see that the '$N_u\sigma$' measurements are consistently low. This we attribute to the effect of pump saturation, which would have the effect of reducing the ratio σ_h/σ_{11} from the values given in Table (1). Strong pump saturation is believed to occur during the 100 ns duration of the gain switched spike which is estimated to give rise to peak powers of ~ 40 $MWcm^{-2}$ inside the OPML cell. For the R(22) pumped line the effect is not so apparent. This may be attributed to the lower gain which reduces the effect of pump saturation by increasing the time over which the gain is averaged in the build-up time measurement. Further, Q-band absorption of R(22) pumped OPML emission (Eckhardt et.al.1979) is believed to occur but could not quantitatively be allowed for in the calculation of gain from build-up time measurements.

3. Optically Pumped Molecule Amplifiers

The characteristics of the gain medium created by laser pumping of a molecular gas generally do not satisfy the requirements for high energy storage amplifiers, principally because at the pressures of lasant gas necessary for efficient absorption of the pump laser radiation, collisions destroy the population

TABLE 1

Measurement of Gain in the CF_4 OPML

CO_2 Pump line (9 μm band)	R(12)	R(22)
CF_4 Gas pressure (torr)	1.9	0.5
CF_4 Gas temperature (^oK)	127	118
Beam splitter material	CdTe	KBr
Inclination to optic axis	55 ± 2^o	47 ± 2^o
$N_u(\sigma_{max} - \sigma_{min})cm^{-1}$ using equation (2)	$(3.0 \pm 0.4)10^{-3}$	$(2.4 \pm 0.3)\ 10^{-3}$
$N_u\sigma_{max}$ using theoretical $\frac{\sigma_{max}}{\sigma_{min}}$	$(12 \pm 2)10^{-3}$	$(4.8 \pm 0.6)10^{-3}$
OPML build-up time (ns) with beam-splitter	210 ± 30	550 ± 100
Gain $\dot{g}_{max}cm^{-1}$ using equation (1)	$(17 {}^{+2}_{-1})10^{-3}$	$(6.8 {}^{+0.9}_{-0.6})10^{-3}$

inversion in a time typically much shorter than the duration of the pumping pulse. However, not only are there applications in plasma diagnostics for OPMA's where high powers and narrow linewidths are required simultaneously, but there may be good efficiency arguments in favour of such devices. For example, in low gain OPML's the laser build-up time commonly exceeds the duration (~100ns) of the leading gain-switched 'spike' characteristic of CO_2 TEA laser outputs and a good fraction of the available pump energy is therefore not utilised. Further, where the low optimum pressures in OPML's results in only a small fraction of the available pump energy being absorbed in laboratory-size devices (Armandillo and Green 1979), an OPMA operating at a higher pressure can often be used. Previous work has demonstrated that (at least under some conditions) the optimum operating pressure in an OPML for maximum energy extraction corresponds to the maximum gain, rather than to the maximum inversion density (which is reached at a higher pressure, Green 1979). This conclusion has been borne out in the present work, where the pressure dependence of the R(22) pumped CF_4 OPML was examined for both parallel and perpendicular output polarisations. The results for output energy vs. pressure are shown in Fig (2). Since the same upper laser level population is common to both curves, the optimum pressure is observed to be the same in both cases. At this optimum pressure the ratio of OPML output energies for the two polarisations is observed to be approximately the same as the 2:1 ratio of gains. We conclude that an OPMA can be operated at significantly higher pressures than those which optimise OPML operation, greatly increasing the efficiency of utilisation of the energy available from high power CO_2 pump laser by improving the specific energy storage in the optically pumped gas.

Given the possible advantages of OPMA's, the polarisation phenomena displayed by OPML's may be used to great advantage in isolating the elements of a multi-stage OPMA. This isolation is necessary to prevent parasitic oscillations which otherwise limit the size of the amplifier. Here we propose the use of a polarisation filter (e.g. a stack of plates at Brewster's angle) to isolate amplifier stages against the plane of polarisation for which the OPMA gain is a maximum. With such an arrangement the orientations of the planes of polarisation of the (colinear) high power laser pumping pulse and the pulse to be amplified are chosen to mimimise the gain of the amplified pulse in the OPMA. Illustrations of such an arrangement are shown in Fig (3) for the plane of polarisation of maximum OPMA gain parallel and perpendicular to the plane of polarisation of the pumping pulse. For P or R band pump and laser transitions this technique offers no particular advantage. For other cases, factors up to three in total amplifier length may be gained, by choosing the length of each stage just small enough to prevent parasitic oscillation. When compared to the possibility of using circularly polarised light for pumping and amplifying, the improvement in amplifier length is a factor up to 1.7. The improvement achieved experimentally may be somewhat less, because of the complications of saturation by both the pump and amplified signal (Drozdowicz et. al. 1979). Because of the M_J selection rules, molecules in certain (quantised) orientations will not participate in the absorption/emission process, but this effect is only important in low J transitions. The gain in length of an OPMA which can be achieved in this way must in general be estimated by considering the behaviour of each J, K, M_J transition in the detail undertaken for the D_2O and CH_3F FIR lasers by Drozdowicz et. al. 1979.

4. Conclusions

One of the unique features of OPML's is that the gain anisotropy predicted to occur under collisionless conditions for linearly polarised pump radiation is also found to hold (at least approximately) under collisional conditions.

This gain anisotropy has been shown to provide a useful indication of upper laser level population density, but could conversely be used to study the effects of collisions in OPML's at higher pressures. An application of gain anisotropy to the design of OPMA's has also been proposed. For this we require that laser action on a different transition should not occur when oscillation is constrained to occur in the plane of polarisation perpendicular to that of maximum gain. Examples were provided by the two CF_4 OPML transitions studied here.

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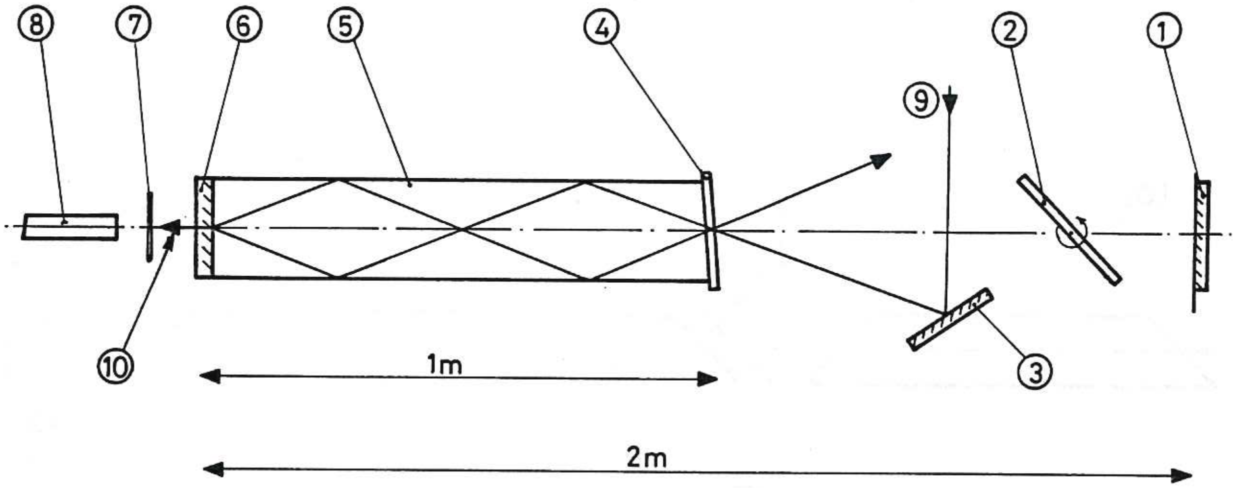

Fig.1 Schematic arrangement of OPML cavity. 1 – total reflector R = 5 m; 2 – beam splitter in rotatable holder; 3 – external reflector; 4 – KBr entrance window inclined at 10° to normal; 5 – low temperature OPML cell; 6 – total reflector with 2 mm diameter central pinhole, R = 5 m; 7 – infrared polariser; 8 – pyroelectric detector; 9 – CO_2 pump radiation; 10 – OPML output radiation.

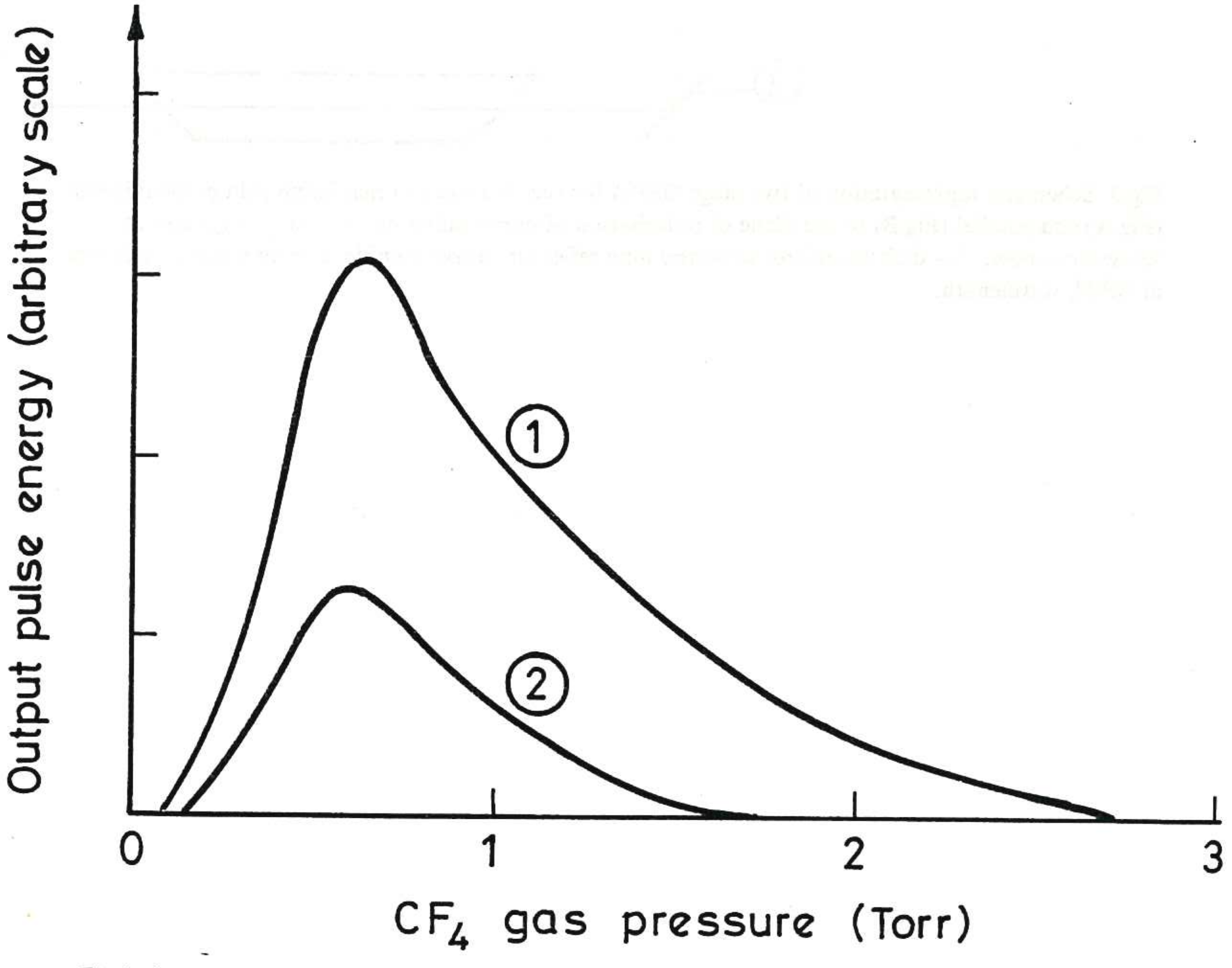

Fig.2 Output energy vs gas pressure for R(22) pumped CF_4 OPML at 118 K. (1) output polarised perpendicular to pump polarisation (i.e. normal operation). (2) output polarised parallel to pump polarisation (i.e. beam splitter orientated in appropriate plane at Brewster's angle.).

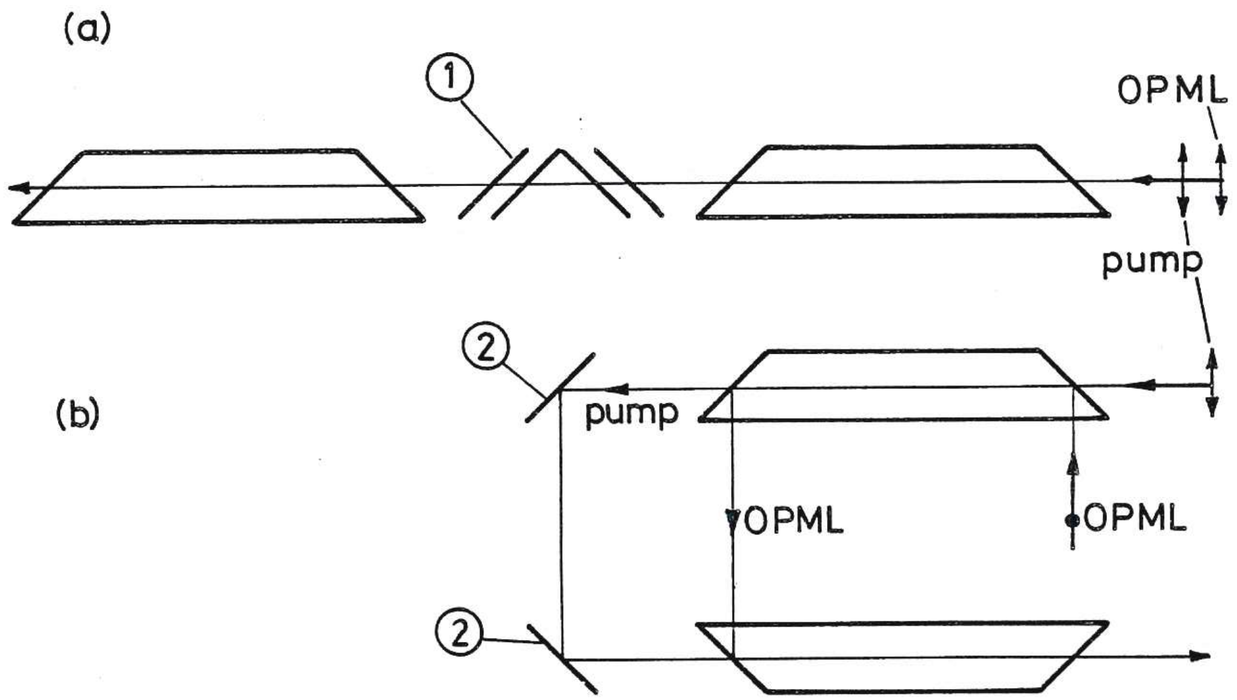

Fig.3 Schematic representation of two stage OPMA for the two cases of maximum gain perpendicular (Fig.A) and parallel (Fig.B) to the plane of polarisation of pump radiation. 1 – stack of plates at Brewster's angle; 2 – dichroic mirror or restrahlung reflector/mirror combination to remove radiation at OPML wavelength.