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OBSERVATION OF OPTICAL NUTATION IN AN ACTIVE MEDIUM*

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An intense optical pulse at 10.6 μm, of rise time that is short compared with the inverse bandwidth of an active CO_2 medium in which it is amplified, has been observed experimentally to attain a sharpened leading edge and attenuated lagging edge. This is an effect attendant upon optical nutation in the CO_2 medium. Computer solutions are presented for comparison.

The response of an amplifying or attenuating medium to intense pulses of length comparable with, or shorter than, T_2 (the phase-coherence time) can exhibit nonlinear inertial effects such as nutation and self-induced transparency. Although nutation effects are known at low frequencies in nuclear magnetic resonance, they have only been reported in the optical region for the attenuating case, using SF_6 as the medium.[1,2] In SF_6, the exact absorbing transitions are not known and so the results cannot be fitted to theory.

In this paper, we report the first observation of the optical nutation effect in an amplifying medium. The transitions are known and experimental results agree well with theory. The experiments were done in a CO_2-N_2 laser amplifier fed with a pulsed signal from a CO_2-N_2-He oscillator at 10.6-μm wavelength. A combination of high gain, high power, and large T_2 makes this medium uniquely suitable for demonstrating the effect. On the other hand, a number of factors tend to smooth out the nutation.

(a) Each of the multiply degenerate vibrational-rotational transitions of the CO_2 molecule has its own dipole moment. In the case of the $P(20)$ CO_2 transition, the pulse interacts with 20 different dipole moments in the amplifier. The rate of population reversal is proportional to the product of the dipole moment and the electric field.

(b) Rotational relaxation also affects the interaction, particularly at high operating pressures.

(c) The nonuniform intensity of the beam introduces further complexity. These factors have not proved severe enough in practice to prevent the observation of nutation. The smoothing effects prevent repetitive ringing, yet still allow population reversal leading to attenuation of the lagging edge of the pulse. A realistic theory is obtained by taking account of degeneracy. Experimentally, the population-reversal effect is achieved by operating the amplifier at low pressure to reduce rotational relaxation.

Observation of the nonlinear effect in an amplifier of reasonably short length requires an intense pulse with a very sharp leading edge. Such a pulse with a rise time of 5 nsec and a peak power on the order of 1 kW was generated by a laser oscillator with electro-optic Q switching and cavity dumping.[3] As a result of optical inhomogeneities in the GaAs, the output was multimode and the intensity profile was relatively uniform over the beam cross section.

The pulse was fed into a five-pass amplifier[4] which repeatedly refocused the beam through the same discharge tube without spatial overlap. Refocusing ensures high intensities over the full length of the amplifier, while absence of overlap avoids interaction between separate beams. The refocusing mirrors of 1-m radius were spaced by 1.5 m of which 1 m was occupied by active amplifier discharge giving a total amplifying path of 5 m. At each reflection, the beam was apertured to 8.5 mm diam. Since the pulses were not perfectly repetitive in shape, both the input and output to the amplifier were monitored with Ge:Cu:Sb photoconductor detectors having measured rise times of better than 1 nsec, and were observed simultaneously on an oscilloscope with a 7-nsec rise time. The recorded pulse shapes were corrected by means of measured power characteristics of the detectors.

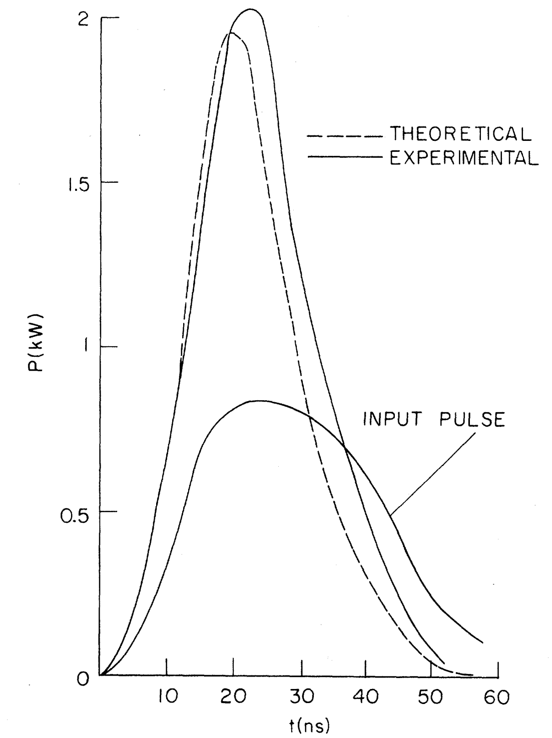

FIG. 1. Experimentally observed input and output pulses and computed output pulse.

The desired effects were only seen at amplifier pressures <1 Torr. At higher pressures, rotational relaxation apparently smoothed out the nutation. Gain coefficients below 1 Torr were comparatively small, being typically ≈1.4 dB/m. Figure 1 shows examples of corrected traces for a single pulse of the $P(20)$ line. The gas mixture in the amplifier contained CO_2 and N_2 with partial pressures of 0.25 and 0.55 Torr, respectively. Sharpening of the leading edge and attenuation of the trailing edge are apparent. Figure 1 shows that the trailing edge of the output pulse is below that of the input pulse, illustrating the population-reversal effect. At pressures we have used, the CO_2 transitions are inhomogeneously broadened and the appropriate equations used to describe the pulse amplification are those of Hopf and Scully,[5] generalized to take into account the orientational degeneracy of the upper and lower levels characterized by the quantum number M, $-J<M<+J$. Except where specifically noted, the notation is that of Ref. 5. The excitation is assumed to be at line center. Then, the action of the material on the field (ignoring losses) is fully described by [compare Eq. (2.1a) of Ref. 5]

$$\frac{\partial \mathcal{E}(t,z)}{\partial z}+\frac{1}{c}\frac{\partial \mathcal{E}(t,z)}{\partial t}=\frac{-\nu}{2c\epsilon}\sum_{M=-J}^{+J} S_M(t,z), \tag{1}$$

where the in-phase component of the polarization $S(t,z)$ is given by [compare Eq. (3.6) of Ref. 5]

$$S_M(t,z)=\frac{\rho_M{}^2N}{\hbar}\int_{-\infty}^{\infty}d\omega\,\sigma(\omega)\int_{-\infty}^{t}dt'\,\mathcal{E}(t',z)\exp\frac{-(t-t')}{T_2}\cos(\omega-\nu)(t-t')[\rho_{aa}{}^{(M)}(\omega,t',z)-\rho_{bb}{}^{(M)}(\omega,t',z)], \tag{2}$$

and the difference of the diagonal matrix elements of the density matrix is related to the electric field amplitude by [compare Eq. (3.7) of Ref. 5]

$$-\partial[\rho_{aa}{}^{(M)}(\omega,t,z)-\rho_{bb}{}^{(M)}(\omega,t,z)]/\partial t=(\mathcal{P}_M{}^2/\hbar^2)\mathcal{E}(t,z)\int_{-\infty}^{t}dt'\mathcal{E}(t',z)\exp[-(t-t')/T_2]$$
$$\times\cos(\omega-\nu)(t-t')[\rho_{aa}{}^{(M)}(\omega,t,z)-\rho_{bb}{}^{(M)}(\omega,t,z)]. \tag{3}$$

Here, the summations are over the orientational quantum number M. The matrix elements ρ_M are related to the matrix elements $\mathcal{P}_0$ for $M=0$ by

$$\mathcal{P}_M{}^2/\mathcal{P}_{M=0}{}^2 = 1-M^2/J^2. \tag{4}$$

These equations have been programmed for a digital computer by Rhodes and Hopf[6] using numerical integration of the density-matrix equations. Computer runs were made for our case, taking into account the degeneracy of the CO_2 transitions. A uniform-intensity plane wave was assumed with a diameter of 8.5 mm (the amplifier aperture size). Diffraction effects were ignored, the average value of intensity along the beam being used. The parameters used in the computation, corresponding to the experimental conditions, were $T_2 = 85$ nsec (at a pressure of 0.8 Torr), exponential gain coefficient 0.32 m^{-1}, Doppler width 60 MHz, and the matrix elements appropriate to the CO_2 transition. The curve labeled "theoretical" in Fig. 1 was obtained. The agreement is reasonably good, in spite of the fact that variations of intensity along the length of the amplifier due to diffraction and refocusing were ignored in the computation.

Apart from their theoretical interest, the effects described here have possible practical application to the production of very short and intense radiation pulses at 10-μm wavelength.

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OBSERVATION OF SIMULTANEITY IN PARAMETRIC PRODUCTION OF OPTICAL PHOTON PAIRS

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The quantum mechanical description of parametric fluorescence is the splitting of a single photon into two photons. This description has been verified by observing coincidences between photons emitted by an ammonium dihydrogen phosphate crystal pumped by a 325-nm He-Cd laser. The coincidence rate R_C decreases to the calculated accidental rate [$<0.03R_C(\max)$], unless the two detectors are arranged to satisfy energy and momentum conservation and have equal time delays.

In the elementary quantum process of decay of a photon (ω_p) into two new photons (ω_1, ω_2), emission of the products should be simultaneous[1]:

$$t_1 = t_2. \tag{1}$$

The decay is allowed in a medium which lacks inversion symmetry. If the medium is invariant to translations in space and time, momentum and energy must be conserved:

$$\vec{k}_p = \vec{k}_1 + \vec{k}_2, \tag{2}$$

$$\omega_p = \omega_1 + \omega_2. \tag{3}$$

This process is called[2] parametric fluorescence, parametric scattering, or parametric noise, and (2) and (3) are already well known as the phase-matching conditions. We have verified that photon coincidence occurs unless any of the conditions of (1)-(3) is violated.

The optical arrangement is illustrated in Fig. 1. Phase matching was satisfied by using the birefringence of an ADP crystal, $L = 25$ mm long, whose optic axis made an angle of 52.4° with the normal to the faces. The pump was the 325-nm beam of a He-Cd laser (Spectra-Physics model No. 185) with single-isotope cadmium, power $P_p = 9$ mW, and about 2-mm beam diameter. Phase matching requires that the two new beams, to be at visible frequencies, be of ordinary polarization. It follows that each new frequency is emitted in a cone at angle $\varphi_{1,2}$ around the pump beam.