Coherent amplification of intense nanosecond pulses in a multilevel system

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Coherent amplification in a multilevel system is analyzed for the particular example of the iodine lasing transition $^2P_{1/2} \rightarrow {}^2P_{3/2}$. The evolution of the pulses leads to a situation with the highest possible energy output. An experiment could be carried out on the chemical pumping of iodine.

1. Study of the coherent amplification of light pulses is an important part of the problem of the coherent interaction of electromagnetic radiation with matter. Most of the theoretical work on coherent amplification has been restricted to a two-level model of the amplifying medium.[1–4] The actual systems which are of physical and practical importance, however, are multilevel and degenerate systems. Nevertheless, in only a few papers can we find attempts to determine the effect of degeneracy on coherent amplification.[5,6] In the present letter we use the particular example of the $^2P_{1/2} \rightarrow {}^2P_{3/2}$ lasing transition of atomic iodine to study the distinctive features of coherent amplification in the case with a hyperfine splitting and a degeneracy. In particular, we study how the pulsed emission of energy from the active medium differs from that in the case of a two-level system. We show that among all the stable solutions given by the "area theorem"[7,8] the solution corresponding to the greatest output of energy from the active medium holds in the limit of long pulse propagation distances. We also show that for a sufficiently short pulse a degenerate multilevel medium "works" in the same way as a two-level system without degeneracy.

2. The upper level of the lasing transition $^2P_{1/2}\to{}^2P_{3/2}$ of the iodine atom is split into two sublevels, while the lower level is split into four.[9] These sublevels have a total angular momentum F. Three transitions go from each of the two upper sublevels to lower levels, so that there are always six hyperfine components of the transition $^2P_{1/2}\to{}^2P_{3/2}$. Working from the frequency intervals between the components,[9] we conclude that a pulse of length $\tau_p \gtrsim 10^{-10}$ s interacts resonantly with only one of the components, removing the population inversion from only one of the two upper sublevels. For definiteness, we consider the interaction of a pulse with a transition from the $F=3$ sublevel to the $F'=4$ lower sublevel. This transition is degenerate with respect to the projection (m) of the total angular momentum F, and in the case of linearly polarized light the transitions occur with essentially no change in m.

We begin our analysis of the pulse amplification with the "area theorem." This theorem was derived in Ref. 8 for the case of an absorbing medium and an inhomogeneously broadened line; an approximate derivation was offered in Ref. 9 for the case of homogeneous broadening. In the case of the amplification of a light pulse considered by us, the area theorem can be written

$$\frac{d\theta}{dz} = \frac{\alpha}{\sum_m p_m^2} \sum_{m=-3}^{3} p_m \sin p_m \theta . \qquad (1)$$

Here

$$\theta(z) = \frac{\hat{\mu}}{\hbar} \int_{-\infty}^{+\infty} dt\, E(z,t)$$

is the pulse "area," $E(z,t)$ is the amplitude of the wave which is propagating along the z axis, α is the linear gain, $p_m = \mu_m/\hat{\mu}$, μ_m is the dipole moment of the transition between sublevels (F,m) and (F',m), and $\hat{\mu}$ is the dipole moment of any of these transitions. In our case we assume $\hat{\mu}$ to be the dipole moment of the transition $(F,3)\to(F',3)$: $\hat{\mu}=\dot{\mu}_3 = 0.27\times10^{-20}$ abs. unit. We thus have $p_0 = 1.51$, $p_{\pm1} = 1.46$, and $p_{\pm2} = 1.31$. During the passage of a pulse of area θ, the difference (n_m) between the populations of levels (F,m) and (F',m) changes from 1 to $\cos p_m\theta$. According to (1), "stable" values $\theta=\theta_i$ lead to a minimum in the average population difference $\bar{n} = 1/(2F+1)\Sigma_m n_m$ after the passage of the pulse and thus maxima in the efficiency of the pulsed emission of energy from the active medium, $\eta = 1/2[1-\bar{n}]$. Among the "stable" values θ_i, there is the value $\theta_1 = 0.75\,\pi$, which leads to the greatest value of the coefficient η: $\eta_1 = 0.95$. In the interval $0<\theta<4\pi$, on the other hand, the stable solutions are $(\eta_1\approx0.95)$, $\theta_2 = 2.2\pi(\eta_2=0.67)$, and $\theta_3 = 3.4\pi(\eta_3=0.72)$. A numerical simulation of the pulse amplification, however, shows that, regardless of the initial conditions, the same definite situation is ultimately established: the situation with the maximum value of η. In other words, the system tends to emit a maximum

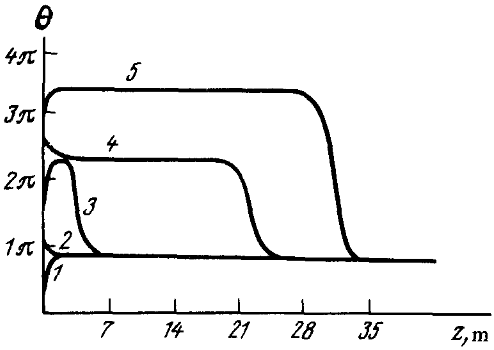

FIG. 1. Change in the "area" θ during the propagation of a pulse for various values of the initial area θ_{in}: $1 - 0.2\pi$; $2 - \pi$; $3 - 1.5\pi$; $4 - 2.5\pi$; $5 - 3\pi$.

value of the energy stored in the active medium during coherent amplification. We get the impression that this remarkable fact has a profound thermodynamic meaning.

A simulation of the amplification was carried out through a numerical solution of the Maxwell-Bloch equations. The initial pulse length is chosen to be $\sim 10^{-9}$ s, so we assume T_1, $T_2 = \infty$ and ignore the inhomogeneous broadening. The curves shown below refer to the case (a typical case experimentally) in which the density of atoms in the upper sublevel is 10^{16} cm^{-3}. Figure 1 shows the behavior of θ during the propagation of the pulse for five different initial values (θ_{in}) of our area. In the first two cases, the behavior of θ is in accordance with (1), and there is a tendency toward the value 0.75π. In the other cases, θ initially behaves in accordance with (1), goes to the nearest value θ_i, spends a certain time there, and then acquires the value $\theta_1 = 0.75\pi$, at which we have the maximum value $\eta \cong 0.95$.

Figure 2 shows the evolution of the pulse amplitude $E(z,t)$ and the change in the population $\bar{n}$. We see that in the course of the amplification the pulse becomes modulated and narrower. Comparison of the evolution of the pulses with the various initial areas shows that the shape of the pulses at the ouput may differ substantially for an essentially identical amount of energy emitted.

The modulation and narrowing (to $\tau_p \sim 10^{-10}$ s) of the pulse in the course of the amplification have the consequence that the pulse becomes resonant with three transitions at the same time: $F = 3 \rightarrow F' = 4, 3, 2$. It can be shown that transitions from $(F = 3; m)$ to $(F' = 4; m)$, $(F' = 3; m)$, and $(F' = 2; m)$ are described in the same way as a transition in a two-level system with $\mu_{\text{eff}}^m = (\mu_{2,m}^2 + \mu_{3,m}^2 + \mu_{4,m}^2)^{1/2}$ [$\mu_{i,m}$ is the dipole moment of the transition $(F = 3; m) \rightarrow (F = i; m)$]. The value of μ_{eff}^m is independent of m at $\mu_{\text{eff}}^m = 0.43 \times 10^{-20}$ abs. unit. Consequently, as a result of the narrowing, the pulse interacts with the medium of atomic iodine as it would with a two-level nondegenerate medium with $\eta = 1$.

The maximum possible value of η for the transition $F = 2 \rightarrow F' = 3$ (as for the transition $F = 3 \rightarrow F' = 4$) is ≈ 0.95. Consequently, two pulses, at resonance with the transitions $F = 3 \rightarrow F' = 4$ and $F = 2 \rightarrow F' = 3$, can remove essentially all the energy stored in the two upper sublevels, and the shape of the pulse at the output can be adjusted by changing the parameters of the input pulse.

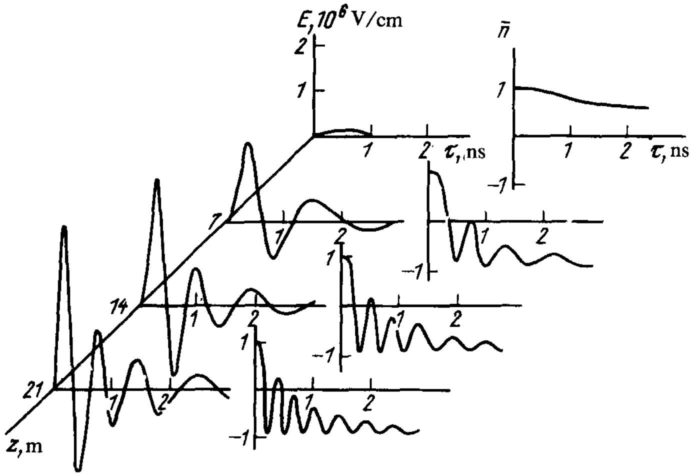

FIG. 2. Evolution of the pulse amplitude $E(z,t)$ during propagation; change in the average population difference $\bar{n}(z,t)$. Here $\tau = t - z/c$ is the comoving time, and c is the velocity of light.

3. Coherent-amplification effects have not yet found application in high-power lasers. We believe that the reason is that the intensity of the light at which the coherent amplification occurs lies above the thresholds for self-effects in the active media (self-focusing, breakdown, and many-photon absorption). In high-power lasers the saturation energy E_{sat} is high, while the polarization relaxation time T_2 is quite short. For this reason, the intensity (E_{sat}/T_2) at which coherent-amplification effects become manifested are above 10^9 W/cm^2. Such intensities are close to the thresholds for self-effects in the condensed matter and compressed gases which are ordinarily used in high-power lasers. This is apparently the basic reason why coherent-amplification conditions could only be approached in the experiments of Refs. 10 and 11. The ideal medium for the practical realization of coherent amplification in a high-power laser would be a medium with a high stored-energy density but without self-effects.

The medium of an iodine laser with chemical pumping meets these requirements. A population inversion is produced in this laser[12] through a rapid, near-resonance transfer of energy from the $O_2(a'\Delta g)$ metastable state of the oxygen molecule to the $I(5^2P_{1/2})$ metastable state of iodine. The necessary concentration of $O_2(a'\Delta g)$ (singlet oxygen) is produced in the chlorination of an alkali solution of hydrogen peroxide.[13] Analysis[14] of the pulsed operation of this laser shows that at a singlet-oxygen pressure ~ 10 torr and an iodine pressure ~ 0.3 torr the stored energy is about 15 J/liter, and the intensity of the pulse which is being amplified can reach several gigawatts through a square centimeter. At such pressures, the saturation energy is $E_{sat} < 0.01$ J, and we have $T_2 > 10$ ns; i.e., the pulse intensity exceeds that required for the coherent amplification of nanosecond pulses, and there is no reason to expect any restrictions because of self-effects. The features of coherent amplification which have been derived can thus be observed experimentally in the pulsed operation of a laser of this type.

[1]E. M. Belenov, A. N. Oraevskiĭ, and V. A. Shcheglov, Zh. Eksp. Teor. Fiz. **56**, 2143 (1969) [Sov. Phys. JETP **29**, 1153 (1969)].
[2]G. L. Lamb, Phys. Rev. A **12**, 2052 (1975).
[3]V. E. Zakharov, Pis'ma Zh. Eksp. Teor. Fiz. **32**, 603 (1980) [JETP Lett. **32**, 589 (1980)].
[4]S. V. Manakov, Zh. Eksp. Teor. Fiz. **83**, 68 (1982) [Sov. Phys. JETP **56**, 37 (1982)].
[5]A. O. Markano and V. T. Platonenko, Kvantovaya Elektron. (Moscow) **7**, 759 (1980) [Sov. J. Quantum Electron. **10**, 433 (1980)].
[6]I. R. Gabitov and S. V. Manakov, Phys. Rev. Lett. **50**, 495 (1983).
[7]S. L. Mc Call and E. L. Hahn, Phys. Rev. **183**, 457 (1969).
[8]C. K. Rhodes, A. Stökes, and A. Javan, Phys. Rev. Lett. **21**, 1151 (1968).
[9]V. S. Zuev, V. A. Katulin, V. Yu. Nosach, and O. Yu. Nosach, Zh. Eksp. Teor. Fiz. **62**, 1673 (1972) [Sov. Phys. JETP **35**, 870 (1972)].
[10]V. Yu. Baranov, V. L. Borzenko, D. D. Mamota, Yu. V. Perushevich, Yu. A. Satov, A. Yu. Sebrant, Yu. B. Smakovskiĭ, A. N. Starostin, and A. P. Strel'tsov, Pis'ma Zh. Eksp. Teor. Fiz. **30**, 593 (1979) [JETP Lett. **30**, 558 (1979)].
[11]O. P. Varnavskiĭ, A. N. Kirkin, A. M. Leontovich, R. F. Malikov, A. M. Mozharovskiĭ, and E. D. Trifonov, Zh. Eksp. Teor. Fiz. **86**, 1227 (1984) [Sov. Phys. JETP **59**, 716 (1984)].
[12]D. J. Bernard, W. E. Mc Dermott, D. J. Bernard, and R. R. Bousek, Appl. Phys. Lett. **34**, 40 (1979).
[13]W. E. Mc Dermott, N. R. Pchelkin, D. J. Bernard, and R. R.Bousek, Appl. Phys. Lett. **32**, 469 (1978).
[14]G. N. Hays and G. A. Fisk, IEEE J. Quantum Electron **QE-17**, 1823 (1981).

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