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BLEACHING WAVES IN TWO-LEVEL SYSTEMS

V. M. OVCHINNIKOV and V. E. KHARTSIEV

A. F. Ioffe Physico-technical Institute

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Propagation of a monochromatic pulse of resonance radiation in a medium containing centers with two energy levels is considered. It is shown that the propagation of a pulse in such a medium is accompanied by a bleaching wave with a narrow front whose velocity may be smaller than that of light by several orders of magnitude. The bleaching wave effect can arise in many systems with narrow intense absorption lines and especially in the presence of metastable levels.

SUBSTANCES of the type of Mg-phthalocyanine and cryptocyanine, which have narrow bands of rather intense absorption and whose absorption coefficient can decrease sharply and abruptly under the influence of a pulse of resonant radiation passing through them have been investigated in a number of experimental papers.[1-3] Recently such substances have come into use as passive optical shutters for optical quantum generators of giant pulses.[3] In connection with the properties of these substances, we consider a model problem involving the passage of pulses of monochromatic resonant radiation through an infinite plane-parallel layer containing uniformly distributed active centers with two levels.

Let n_1 and n_2 be the populations of the lower and upper levels, respectively, N—the density of the active centers, σ—the capture cross section, and l—the thickness of the layer. When $t \ll \tau_2$, where τ_2 is the lifetime of the excited state, neglecting nonresonant losses the coefficient of absorption is determined by the relation $k(x, t) = \sigma n(x, t)$, with $n = n_1 - n_2$ being obtained from the system of equations

$$\frac{\partial n}{\partial t} = -2\sigma c u n, \quad \frac{\partial u}{\partial t} + c\frac{\partial u}{\partial x} = -\sigma c n u. \tag{1}$$

Here u is the density of the photons of the resonant radiation, c is the velocity of light in the layer, and the flux is incident on the layer from the left.

In the case of an originally unexcited layer, initial and boundary conditions $n(x, t = 0) = N$ and $u(x = 0, t) = u_0(t)$ correspond to the nonlinear system (1). Substituting the values for n from the integrated first equation of the nonlinear system (1) in the second equation, and making the change of variable

$$\int_0^t u\,dt = w(x, t),$$

we obtain an equation analogous to that for the amplification of radiation in an excited layer[4]:

$$\frac{\partial w}{\partial t} + c\frac{\partial w}{\partial x} = \frac{1}{2}(e^{-2\sigma c w} - 1). \tag{2}$$

Integrating Eq. (2) by the method of characteristics and determining the value of n, we obtain an expression for the absorption coefficient:

$$k(x, t) = \sigma N\left\{\exp\left[\sigma\left(2\int_0^{t-x/c} u_0(s)\,ds - Nx\right)\right] + 1 - e^{-\sigma N x}\right\}^{-1}. \tag{3}$$

In the case of a rectangular pulse with $\exp(-\sigma N x) \ll 1$, we have

$$k(x, t) = k(vt - x),$$

where

$$v = 2u_0 c(N + 2u_0)^{-1}. \tag{4}$$

Thus, the propagation of a monochromatic pulse in a resonantly absorbing medium corresponds to a bleaching wave moving with velocity v. The distance δ at which the wave is formed is determined from the condition $\exp(-\sigma N\delta) = 5 \times 10^{-2}$. The width of the wave front, determined from the condition that the absorption coefficient decrease from the maximum value $k_m = \sigma N$ to a value $10^{-p}k_m/2$ is found from the relation

$$\Delta = \ln 10(10^p - 2) / \sigma(N + 2u_0)$$

for $p > 1$. The time for the bleaching of the layer by the wave is

$$\tau = (N + 2u_0)l / 2u_0 c. \tag{5}$$

Here $\tau'/\tau = \ln 20/\sigma N l$, where τ' is the time of formation of the wave.

In the general case, where the amplitude of the pulse incident on the layer depends on the time, the rate of displacement of the constant absorption coefficient, determined by the equation $dk/dt = 0$, is

$$v = 2cu_0(t - x/c)[N + 2u_0(t - x/c) - NA]^{-1},$$

$$A = \exp\left(-2\sigma c \int_0^{t-x/c} u_0(s)\,ds\right). \tag{6}$$

From (3) and (6) it follows that at first the bleaching moves with a velocity c, and then, after the condition $A \ll 1$ is satisfied, the velocity decreases and a bleaching wave is established. Under ordinary conditions $N \gg u_0$, and therefore in (6) we have $t \gg x/c$, and then $v(t) = 2u_0(t)c/N$. It is typical that the velocity of the wave does not depend on σ but the bleaching wave arises at large photon absorption cross sections $\sigma \gg 1/Nl$. The latter condition corresponds to large optical thicknesses.

Let us estimate the parameters of the bleaching wave produced by a giant pulse from a ruby laser in a medium containing cryptocyanine, assuming that the pulse is rectangular. In the spectral region of the laser emission line R_1, the cryptocyanine has an absorption line with a photon capture cross section $\sigma = 8 \times 10^{-16}$ cm^2. Let us assume that $N = 10^{17}$ cm^{-3}, $l = 0.5$ cm, $J = 0.5$ MW/cm^2, $c = 2 \times 10^{10}$ cm/sec. The velocity of the bleaching wave is then $v = 2.4 \times 10^7$ cm/sec, $k_m = 80$ cm^{-1}, $\delta = 0.38$ mm, the width of the wave front corresponding to a decrease in the absorption coefficient by a factor of 10^2 is $\Delta = 0.75$ mm, and the bleaching time is $\tau = 42$ nsec.

In systems of the type of Mg-phthalocyanine, below the level 2, corresponding to the two-level system under consideration, there is a metastable level with a lifetime $\tau_m \cong 10^{-3}$ sec and the lifetime of the level 2 is determined by the transitions to the metastable level, $\tau_2 = \tau_{2m} = 10^{-7}$—$10^{-8}$ sec, that is, the system is analogous to a ruby-type system.[1,2] The bleaching of systems of this type with intermediate levels also has a wavelike character. Because of the metastable state, bleaching in this case can be realized at low intensities and correspondingly low wave velocities. At these values of N, l, and c we obtain $\tau = 10^{-3}$ sec, $v = 5$ m/sec at $J = 2 \times 10^{-2}$ kW/cm^2.

We note that the condition of large optical thicknesses $\sigma Nl \gg 1$ can be satisfied also for media used in solid state lasers.

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