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RESONANCE INTERACTION OF SHORT LIGHT PULSES WITH MATTER

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ONE of the most important accomplishments of laser physics in recent times has been the generation of ultrashort giant pulses of light. Lasers have already yielded pulses with durations on the order 10^{-12} sec[1]. A method has already been proposed of forming powerful pulses with characteristic times 10^{-13}–10^{-14} sec[2].

The interest in such short pulses is due to a considerable degree to the possibility of observing them with the aid of new nonlinear effects in strong fields. One such effect was predicted and observed by mcCall and Hahn[3,4], and consists in an increase of the transparency of an absorbing medium to short intense pulses of resonant radiation (self-induced transparency). This phenomenon, like photon echo[7], is due to the features of coherent interaction of powerful radiation with a two-level system.

Let us recall first some results of the theory of interaction of two-level systems with an electromagnetic field[8,9]*. Assume that such a system is acted upon by a pulse of resonant coherent radiation, having a duration

$$\tau_p \ll T_1, T_2, \tag{1}$$

where T_1 and T_2 are respectively the times of longitudinal and transverse relaxation of this system. Condition (1) denotes that irreversible relaxation processes do not have time to violate greatly the coherence of the interaction within the time τ_p. Then, if the pulse satisfies the condition

$$\Theta = \frac{2d}{\hbar} \int_{-\infty}^{\infty} \mathscr{E}(t)\, dt = (2n-1)\pi,$$

the system, which was previously in the equilibrium state, goes over under the influence of the field into a fully invariant state. Here d is the electric dipole moment of the quantum transition, $\mathscr{E}(t)$ is the slowly varying field amplitude, and n = 1, 2, ... Such 180° pulses are used to produce inversion in certain types of quantum amplifiers.

On the other hand, if

$$\Theta = \frac{2d}{\hbar} \int_{-\infty}^{\infty} \mathscr{E}(t)\, dt = 2n\pi, \tag{2}$$

then the system returns to the initial state after the end of the interaction with the pulse. Consequently, the energy of the pulse satisfying Eqs. (1) and (2) will be conserved as the pulse propagates in the resonant two-level medium. The medium becomes transparent to this pulse. As seen from the foregoing, this transparency differs radically from the well known bleaching of absorbing media as a result of the equalization of the populations of the ground and excited states.

Let us examine now, following[4], the propagation of pulses satisfying (1) in a resonant absorbing medium in greater detail*. Assume that there are N two-level optical ions (per cm^3) dissolved in the crystal. Owing to the statistical scatter of the local crystal fields, the absorption line will be inhomogeneously broadened with a reciprocal width $T_2^* = g(0)$. The distribution function of the deviations of the resonant frequencies ω_0 of the ions from the frequency ω of the incident pulse will be denoted by $g(\Delta\omega)$, where $\Delta\omega = \omega_0 - \omega$, with

$$\int_{-\infty}^{\infty} g(\Delta\omega)\, d\Delta\omega = 1.$$

We also assume that the following condition is satisfied

$$\omega^{-1} \ll T_2^* \ll \tau_p \ll T_2.$$

The polarization vector of the two-level medium, represented in the language of magnetic resonance in the form $P = \hat{u}_0 u + \hat{v}_0 v + \hat{w}_0 (2d/\mathscr{P}) W$, obeys the Bloch equation[11], given for a time $t \ll T_1, T_2$ by

$$\frac{\partial \mathscr{P}}{\partial t} = \left[\mathscr{P}\left(\hat{u}_0 \frac{2d}{\hbar}\mathscr{E} + w_0 \Delta\omega\right)\right]; \tag{3}$$

here $\hat{u}_0$, $\hat{v}_0$, and $\hat{w}_0$ are unit vectors of a fictitious orthogonal coordinate system, rotating with frequency ω aroung $\hat{w}_0$; v and u are combinations of off-diagonal elements of the density matrix of the two-level system; these elements are responsible for the electric dipole absorption (v) and dispersion (u); W is the spectral energy density of the absorbing particle per cm^3, proportional to the population difference between the upper and lower levels.

The light pulse propagating in the resonant medium along the z axis is represented in the form of a plane wave, say circularly polarized:

$$E = \mathscr{E}(z, t)[i \cos(\omega t - kz) - j \sin(\omega t - kz)], \tag{4}$$

where $k = \omega\eta/c$ is the wave number, η is the refractive index, and the slowly varying amplitude $\mathscr{E}(z, t)$ satisfies the relation $\partial\mathscr{E}/\partial z \ll k\mathscr{E}$ and $\partial\mathscr{E}/\partial t \ll \omega\mathscr{E}$. Then the propagation of the pulse can be described by the equation

$$\frac{\partial \mathscr{E}(z, t)}{\partial z} + \frac{\eta}{c}\frac{\partial \mathscr{E}(z, t)}{\partial t} = -\frac{2\pi\omega}{\eta c}\int_{-\infty}^{\infty} g(\Delta\omega)\, v(z, t, \Delta\omega)\, d\Delta\omega. \tag{5}$$

*The investigation of the behavior of a two-level quantum system in a radiation field consisting of one and two quasiresonant lines is also the subject of [10] (where a bibliography is given).

*Certain results of an investigation of the propagation of pulses in amplifying media can be found in [12], which contains also the basic bibliography on this subject.

From (3) and (5) we get the relation

$$\operatorname{tg} \frac{1}{2} \Theta(z) = \operatorname{tg}\left(\frac{1}{2} \Theta_0\right) e^{-1/2\,\alpha z}; \qquad (6)$$

Here Θ_0 is the value of Θ for ions with $\Delta\omega = 0$ at $z = 0$,

$$\alpha = \frac{8\pi^2 d^2 \omega g(0) N}{\eta \hbar c}.$$

Figure 1 shows the dependence of Θ on z, corresponding to Eq. (6). We see that as $z \to \infty$ we have $\Theta \to 2(n-1)\pi$ if $2(n-1)\pi < \Theta_0 < (2n-1)\pi$ and $\Theta \to 2n\pi$ if $(2n-1)\pi < \Theta_0 < 2n\pi$. When $\Theta_0 < \pi$, the pulse attenuates as it propagates, and its area $\Theta(z)$ tends to zero. In the limit of very small Θ, the damping is described by the usual exponential law $\mathscr{E} = \mathscr{E}_0 \exp(-\alpha z/2)$. On the other hand, if $\Theta_0 > \pi$, then at a distance of several times α^{-1} the pulse acquires a form satisfying (2), losing at the same time a certain fraction of its energy. After taking shape, it propagates further without losses, just as in a transparent medium. These conclusions are demonstrated in Fig. 1b, which shows the solutions $\mathscr{E}(z, t)$ for $\Theta_0 = 0.9\pi$ and $\Theta_0 = 1.1\pi$.

It follows from (5) that the formed pulse is stationary and is described by the formula

$$\mathscr{E}(z, t) = \frac{\hbar}{d\tau} \operatorname{sech}\left[\frac{1}{\tau}\left(t - \frac{z}{V}\right)\right]; \qquad (7)$$

here V is the propagation velocity of the stationary pulse, and τ is its duration, which is inversely proportional to the value of $\mathscr{E}$ at the maximum.

Calculations show that an arbitrary input pulse with $\Theta_0 \approx 2n\pi$ propagating in a resonant absorbing medium breaks up into n stationary pulses described by formula (7).

As the stationary pulse propagates, it loses energy on its leading front to coherent excitation of the atoms, and it reacquires the same energy on the trailing edge. Consequently, its propagation velocity V should be smaller than the velocity of light. Calculations lead to a formula confirming this conclusion:

$$\frac{1}{V} = \frac{\eta}{c} + \frac{\alpha\tau}{2} \qquad (8)$$

We see that more intense pulses travel more rapidly than weak ones. If the light beam at the entrance to the medium has a transverse intensity distribution with a maximum at the center, then its transverse dimensions on the leading edge will decrease as it propagates further, owing to the lag of the peripheral parts of the beam relative to the central ones. The narrowing may be so appreciable, that diffraction effects can no longer be neglected.

We note that Eqs. (3) and (5) for a stationary pulse (7) were solved in[4] without assuming that $T_2^* \ll \tau_p$.

Experiment[4,5] has qualitatively confirmed the main theoretical conclusions. In[4] the employed two-level absorbing system was a ruby rod cooled with liquid helium to decrease the rate of relaxation processes (to increase T_2). The source of the intense pulses of light was a Q-switched ruby laser cooled with liquid nitrogen. It produced plane-polarized radiation due to generation on the transition

$$\bar{E}(2E) \longleftrightarrow 4A_2\left(\pm\frac{3}{2}\right). \qquad \text{A}$$

The radiation frequency was resonant for the transition

$$4A_2\left(\pm\frac{1}{2}\right) \longleftrightarrow \bar{E}(2E) \qquad \text{B}$$

in the liquid-helium-cooled sample. The pulse duration was 10—20 nsec.

Pulses with low intensity, passing through the ruby absorbing rod, were attenuated by almost 10^5 times, whereas pulses with energy in excess of threshold passed with practically no attenuation. An appreciable delay of the output pulses was observed and reached a value corresponding to an increase of the optical path by 100 sample lengths.

The indicated anomalous transmission can be treated as a result of the ordinary saturation effect, but then it becomes impossible to explain such long pulse delays. Furthermore, the rate equations for the populations are not applicable here, since the relaxation times in ruby at liquid-He temperatures exceed the durations of the employed pulses.

With increasing temperature of the absorbing ruby rod, its anomalous transmission decreases and vanishes completely at 40°K[4]. This fact is further evidence that the observed transparency of the absorbing sample is due to coherent interaction.

Patel and Slusher[5] performed similar experiments, using radiation from a Q-switched CO_2 laser. The absorbing medium was SF_6 gas filling a double-pass cell with total length 4.7×2 m. This gas system is convenient because it is easy to control in it the relaxation times of the absorbing levels, the Doppler line width, and the absorption coefficient, by varying the pressure, temperature, and the added buffer gas, making it possible to carry out a detailed investigation of the dependence of the characteristics of the output pulses on the parameters of the medium.

As shown in Fig. 2, for pulses of duration 200—300 nsec, a sharp increase of the transparency of the SF_6 was observed, starting with a certain threshold energy. This threshold value remained practically constant in the pressure range 0.01—0.041 Torr.

Certain results of the investigation of the shape of the output pulses and their delays are demonstrated in Fig. 3. We see that the output pulses are much more symmetrical than the input pulses.

An investigation of the pulse delays as a function of their intensity[5] has revealed qualitative agreement with the theoretical conclusions, namely, that when the intensity of the input pulses increases from the thres-

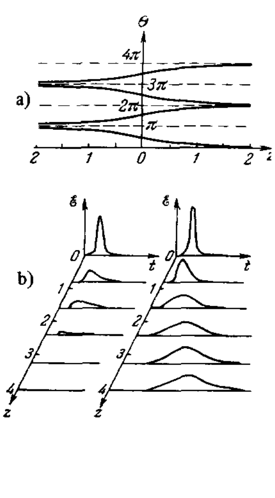

FIG. 1. a) Dependence of Θ on z. For the excited sample, Θ develops in the direction of −z (z is measured in units if $\pi\alpha^{-1}$). b) Dependence of $\mathscr{E}$ on z and t for $\Theta_l = 0.9\pi$ and 1.1π. The initial form is Gaussian (the time is measured in units of the pulse width τ_p).

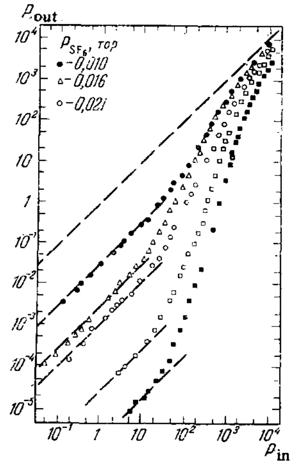

FIG. 2.

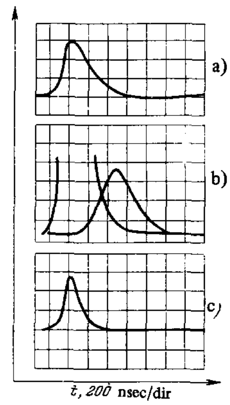

FIG. 3.

FIG. 2. Dependence of the output energy P_{out} on the energy of the input pulses P_{in} at different pressures (□ – 0.033, ■ – 0.041). The energy units are arbitrary.

FIG. 3. Oscillograms of input and output pulses in the case of a slight excess above threshold. a) Typical output pulse in the absence of SF_6 in the cell. b) Double exposure of output pulses. The non-delayed pulse is the same as in Fig. a, magnified in the vertical direction by a factor of 4. The delayed output pulse was obtained at an SF_6 pressure 0.04 Torr. To decrease the transverse-relaxation time, He at a pressure of 2 Torr was added. The narrowing of the output pulse and the almost complete absence of delay are noted.

hold value to the maximum attainable, the delay changes from 0.7 μsec to 50 nsec.

A picture obtained by double exposure and shown in Fig. 3b, indicates that the delayed pulse greatly exceeds in its intensity the "tail" of the input pulse, to which it corresponds in accordance with the delay time. This is evidence against the interpretation of the observed transparency of the medium as bleaching resulting from saturation of the quantum transition.

When the threshold was greatly exceeded, the output pulse contained several maxima, in agreement with the theory of[4].

If the duration of the intense pulse is $\tau_p > T_2$, then the pulse is expected to narrow down on propagation, owing to the absorption on the trailing edge. Such a narrowing was observed in[5] and is demonstrated in Fig. 3c. The authors of[5] believe that it is well possible to determine the relaxation time T_2 from the duration of the shaped 2π pulses at slight excess over threshold. They also assume that low threshold intensities may make it possible to investigate the relaxation times with the aid of photon echo.

Curious experimental results were obtained by the authors of[6]. Experimenting with neodymium-glass and ruby lasers Q-switched by rotating mirrors, they observed in the radiation pulses of picosecond duration. More accurately, these pulses, which appeared spontaneously, had a duration of the order of 1 psec for the neodymium laser and 10 psec for the ruby laser. They were observed by a procedure of two-photon pulse collision and by a superhigh speed procedure.

The appearance of these pulses can be explained on the basis of coherent interaction between a two-level inverted system and a resonant field. When the laser generation is initiated by the noise, certain parts of the generated wave can turn out to be shorter and more intense than others. Let the duration of such a spike be less than T_2. Then, the longer the pulse whose coherent interaction with the medium is interrupted by the relaxation processes, the larger the number of active particles whose coherent emission is stimulated. It is clear that in this case a short pulse will build up in the resonator more rapidly than a long one. In a sufficiently long amplifying rod, the pulse would grow until it would become a 180° pulse. In a resonator, its growth is limited by the losses in the mirrors and by the decrease of the population inversion.

Favoring this explanation are energy estimates presented in[6], as well as the fact that the duration of the observed ultrashort pulses is in poor agreement with the ruby and neodymium-glass line widths.

Picosecond pulses were previously observed in the emission of lasers Q-switched with rotating filters. Their appearance was attributed to mode synchronization due to nonlinearity of the filter. The results of[6] show that such pulses are contained also in the emission of lasers Q-switched by rotating mirrors. This gives grounds for assuming that all sufficiently powerful Q-switched lasers produce picosecond pulses and that many experiments performed using giant pulses should be reviewed with allowance for the presence of powerful picosecond pulses in the radiation.

In conclusion we note that the results of[3-6], the content of which has been briefly reviewed in this note, far from exhaust the problem. It is necessary to explain in greater detail the influence of the relaxation of the diffraction effects, and the influence of the shape of the input pulses. It is important to ascertain the role of the phase relations, the multimode character of the radiation, and of various types of line broadening. As shown in[6], it is also of interest to investigate the coherent interaction of ultrashort pulses with an active medium placed in a resonator.

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