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REDUCTION OF PULSE DURATION IN A RESONANTLY-ABSORBING MEDIUM OF HIGH OPTICAL DENSITY

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We investigate the absorption saturation induced by powerful pulses of resonant monochromatic radiation of frequency ν_{21} in media of high optical density $D(\nu_{21}) = k(\nu_{21})$, where $k(\nu_{21})$ is the absorption coefficient and l is the thickness of the medium. In this case a bleaching wave appears in a medium that is opaque to low-intensity resonance beams. The front of the wave corresponds to a continually moving region of absorption saturation. Reduction of the pulse duration by a factor 1.5–3 was observed experimentally, and the slopes of the fronts of giant ruby-laser pulses passing through a vanadium phthalocyanine solution of optical density D = 6—13 were found to increase. Optically dense media can be used to regulate the duration, slope, amplitude, and spatial structure of monochromatic pulses.

WE know presently of several media (solutions of organometallic and organic compounds such as metal-phthalocyanine, cryptocyanine, and polymethine dyes) that have narrow intense-absorption bands in the region of the ruby laser emission lines [1-3] and neodymium-glass laser lines [4,5]. The absorbing centers in such media, which are used as passive shutters for giant-pulse lasers, have large cross sections for the capture of the resonant-emission photons. It is therefore possible to produce, at a relatively low absorbing-center concentration, a medium with a large absorption coefficient $k(\nu_{21})$ at the frequency ν_{21} of the resonant transition. Armstrong [6] and Dediogio and Potenza [7] investigated experimentally the absorption, at the resonant-transition frequency, of laser radiation by media of low optical density, $D(\nu_{21}) = k(\nu_{21})l$, where l is the thickness of the medium. The initial transmission at the lasing frequency exceeded 15%, and no changes in the shape of the pulse after passing through the absorbing medium were observed by Armstrong [6]. It is clear from qualitative considerations, on the other hand, that a medium which is optically dense at the frequency ν_{21} of the incident monochromatic radiation can alter the pulse parameters, its duration and its spectrum.

In this paper we describe the results of a theoretical and experimental investigation of the saturation of absorption in the optical region of the spectrum under the influence of powerful pulses at high optical density ($D(\nu_{21}) \gg 1$), when the spatial inhomogeneity of the photon flux density of the monochromatic radiation in the absorbing medium (with respect to the coordinate x directed inward from its surface) is appreciable. We observed experimentally not only a decrease in the amplitude but an appreciable change

in the shape and duration of the giant ruby-laser pulses passing through a resonantly absorbing optically dense medium, namely a solution of vanadium phthalocyanine. Many peculiarities of the kinetics of the phenomena under consideration were discussed by us earlier [8,9].

The passage of a pulse of monochromatic radiation through a one-dimensional medium containing two-level resonantly absorbing centers is described by a system of equations for the photon density u and the level populations $n_{1,2}$:

$$\frac{\partial u}{\partial t}+c\frac{\partial u}{\partial x}=-\sigma c(n_1-n_2)u,$$
$$\frac{\partial}{\partial t}(n_1-n_2)=2\sigma c(n_1-n_2)u+\frac{n_2}{\tau}, \quad (1)$$

where σ is the cross section for photon capture by the absorbing center, c the speed of light in the medium, and τ the lifetime of the excited state. The system (1), which is based on the rate-equation approximation, corresponds to initial and boundary conditions $u(x=0, t)=u_0(t)$, $n_1(x, t=0)=N$, and $n_2(x, t=0)=0$. In the case of resonant bleaching of the medium, two extreme cases are possible. In the first, the pulse duration T_p is much shorter than the lifetime of the excited state τ and the second term of the second equation of (1) can be neglected.

It follows from the solution of the system (1) that the photon flux density deep inside the medium is determined, for a rectangular pulse, by the expression

$$\Phi(x,t)=u_0c\{1+\exp[\sigma N(1+2u_0/N)(vt-x)]\}^{-1}, \quad (2)$$

that is, the passage of the pulse corresponds to a bleaching wave of velocity $v=2u_0c(N+2u_0)^{-1}$. The front of the wave is a bleached region (absorption-saturation region) that moves continuously inside the medium. The energy of the front part of the pulse should be consumed in bleaching the medium, after which the remaining part of the pulse passes unobstructed if other loss mechanisms are neglected.

The rise time of the leading front of the pulse passing through the optically dense medium is determined by the time during which the bleaching wave covers a distance l_0 equal to the width of the wave front: $T_r \approx l_0/v$. The value of l_0 can be determined with the aid of (2) from the condition that the initial flux decreases from the level 0.9 to the level 0.1. Then $l_0 \approx 5[\sigma(N+2u_0)]^{-1}$, and the rise time of the output pulse is given by

$$T_r \approx 2.5h\nu_{21}/\sigma P_{in}, \quad (3)$$

where P_{in} is the density of the power flux of the input pulse. At values $\sigma \approx 3\times10^{-16}$ cm^2, which are typical for metal-phthalocyanines [3], we get $T_r \approx 0.5$ nsec in the case of giant pulses with $P_{in} = 5$ MW/cm^2 and $c = 2\times10^{10}$ cm/sec. This is smaller by a factor 10—30 than the rise time of the giant laser pulses. Thus, the use of optically dense media with sufficiently large values of σ and τ makes it possible to reduce the duration of the giant pulses (without decreasing the power flux density) and greatly increase their slope.

In the second extreme case we have $T_r \gg \tau$, and allowance for relaxation is essential. The pulse energy is consumed not only in bleaching the medium, but also in maintaining the bleached state. Let us consider first the passage of the front part of the pulse. During a time interval $\Delta t \approx \tau/2$ there will propagate in the medium a bleaching wave which is weakly distorted by the relaxation. Its velocity v is determined by the photon flux density $\Phi_0 = u_0c$ at the boundary of the medium. Therefore in the case of a rectangular pulse with $v\tau/2 \gtrsim l$, that is, when the condition

$$P_{in} \gtrsim Dh\nu_{21}/\sigma\tau \quad (4)$$

is satisfied for the power flux density of the input pulse, the pulse emerging from the medium during the time $\tau/2 \lesssim t \lesssim l/v$ is also distorted little by the relaxation, and its rise time is determined by the relation (3). When $P_{in} \ll Dh\nu_{21}/\sigma\tau$, the stationary front of the bleaching wave in the medium does not have time to become established within a time $\tau/2$, and relaxation leads to a slowing down of the wave and to a decrease in the amplitude of the output pulse. In the case of a pulse of arbitrary shape, condition (4) must be generalized. The relaxation does not distort the leading front of the pulse emerging from the absorbing medium, provided the following relation holds true for the leading front of the incident pulse

$$\int_0^{\tau/2} P_{in}(t)\,dt \gg G_{cr},$$

where the critical energy flux density is $G_{cr} = Dh\nu_{21}/\sigma$. In the derivation of the last inequality and of the condition (4), we took into account the relation $N \gg u_0$, which is satisfied when $N \gtrsim 5\times10^{16}$ cm^{-3} and $P_{in} \lesssim 50$ MW/cm^2.

The passage of the remaining part of the pulse can be analyzed with the aid of the quasistationary approximation, when the photon flux density at the boundary of the medium is a sufficiently slow function of the time, and when the condition $u_0^{-1}\,du_0/dt \ll \tau^{-1}$, c/l is satisfied. Then the derivative with respect to t in (1) can be neglected, and the solution of the system takes the form

$$\ln\frac{u(x)}{u_0} = 2\sigma u_0 c\tau\left(1+\frac{u(x)}{u_0}\right) - \sigma N x. \qquad (5)$$

It follows from (5) that noticeable transmission of an optically dense medium opaque to low fluxes will take place in the case of powerful pulses when $d/D \gtrsim 1$, where $d = 2u_0c$.[1)] Thus, the medium will transmit predominantly part of the pulse, for which $P_{in} \gtrsim Dh\nu_{21}/2\sigma\tau$. This will result not only in a reduction of the rise time and a decrease of the pulse amplitude after passage through the optically dense medium with small lifetime, but also a reduction of the pulse duration as well as a decrease of the fall-off time of the trailing edge. When the pulse power is increased, and the density D and the pulse duration T_p are kept constant, the duration of the pulse transmitted through the medium will increase. With increasing D and with the parameters of the incident pulse fixed, a decrease will be observed in the amplitude and duration of the transmitted pulse. When $D \gg 1$, we can obtain from relation (5) two approximate expressions for the quasistationary transmission of an optically dense medium, for two regions of variation of the main dimensionless parameters of the problem, $d + 3 \lesssim D$ and $d \gg D$, respectively:

$$\frac{u(x)}{u_0} = e^{-\sigma N x + d}, \quad \frac{u(x)}{u_0} = e^{-\sigma N x/d}$$

We present below the results of experiments on bleaching of a solution of vanadium phthalocyanine in nitrobenzene, placed in a cell of variable thickness. To determine the features of the bleaching of the medium, we used giant ruby-laser pulses of different duration. The resonator was 60 cm long, and the crystal length was 90 mm and its diameter 6.5 mm. The output mirror had a transmission of 50%. Q switching was by means of a total internal reflection prism rotating at 24,000 rpm, and also by means of a combination of a rotating prism with a saturating KS-19 filter, the initial transmission of which was 53%. In the former case the power of the single pulses was ~1 MW and the duration at half-width 40–60 nsec. When the KS-19 plate was introduced into the resonator, the pulse power increased to 5 MW and the duration decreased to 20–25 nsec. A mirror with reflection coefficient 99% was used in many experiments in lieu of the rotating prism, and a cell with a solution of vanadium phthalocyanine in nitrobenzene, with initial transmission 25%, was placed next to it for Q switching. To limit the beam cross section, a diaphragm of 2.5 mm diameter was placed inside the resonator near the output mirror, thus ensuring lasing in the central region of the crystal, where the pump energy was most uniform. In this case the pulse power was 0.5 MW at a duration ~ 20 nsec. The diameter of the beam incident on the cell was 3 mm.

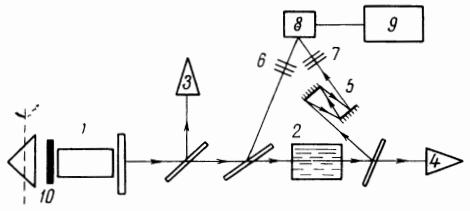

FIG. 1. Block diagram of the experiment: 1 – ruby laser with rotating prism, 2 – cell with resonantly-absorbing medium, 3, 4 – calorimeters, 5 – optical delay line, 6, 7 – neutral calibration filters, 8 – high-speed photodiode, 9 – oscilloscope, 10 – KS-19 plate.

The measurement setup (see Fig. 1) made it possible to register on one oscillogram the waveforms of the pulses before and after passage through the cell with the optically-dense medium, and also to measure their energy. Both pulses were registered by the same photoreceiver. To this end, the pulse passing through the cell was delayed by an optical delay line for ~ 100 nsec. The photoreceiver was a low-inertia surface-barrier germanium photodiode with a time constant shorter than 1 nsec, developed at the Physico-technical Institute of the USSR Academy of Sciences. Operation of the photodiode on the linear section of the lux-ampere characteristic was effected by introducing sets of calibrated neutral filters. The photodiode signals were fed to the input of the amplifier of an S1-11 oscilloscope. The pulse energy was measured with calorimeters accurate to 15%.

The optical density of the absorbing medium at the ruby laser wavelength could be varied continuously by changing the length of the cell, keeping the solution concentration constant in the range D = 0.1—25 corresponding to a change in transmission in the interval 0.9—10^{-11} at low radiation fluxes. Control experiments have shown that the solvent has no noticeable influence on the parameters of the pulse passing through the cell. At low optical densities of the absorbing medium (D = 1—2), the amplitude and the energy of the pulse decreased. With further increase of the optical density, regardless of the Q-switching method employed, the decrease in the energy of the pulse passing through the cell was also accompanied by an appreciable change in the shape of the pulse.

[1)] When $d = D = 1 - 50$, the transmission u/u_0 changes in the interval 0.57 – 0.057; when $2d + 0.7 \gg D$ we have $u/u_0 \geq 0.5$.

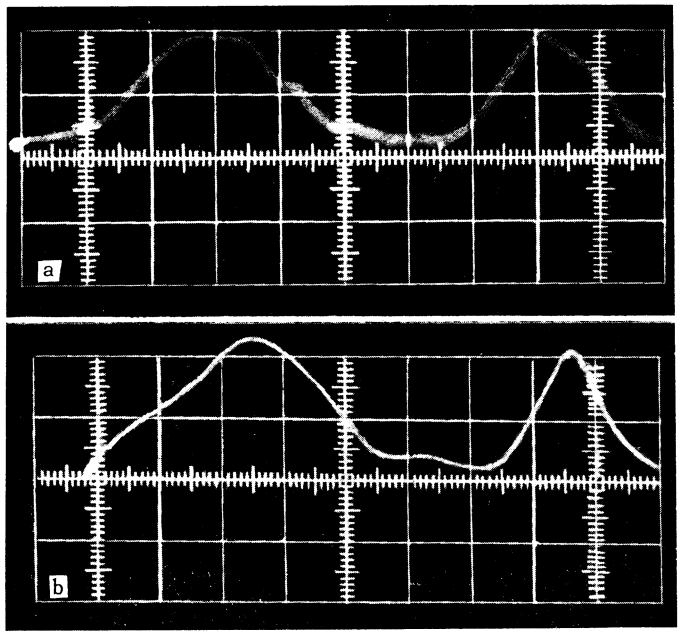

FIG. 2. Oscillograms of pulses before and after passage through a cell with a resonantly-absorbing optically-dense medium (solution of vanadium phthalacyanine in nitro benzene): a – optical density D = 6.5; b – D = 13. The direction of the sweep is from left to right, the rate is 20 nsec/cm. The Q-switching was by means of a rotating prism.

As seen from the oscillograms of Fig. 2, which were obtained with Q-switching by means of a rotating prism, the duration of the pulse passing through a cell was decreased by a factor of 1.5 at D = 6.5, and by a factor of 3 at D = 13. The durations at half-level of the input pulse and of the pulse passing through the cell were in the latter case 60 and 20 nsec, and the rise times from the half-power level to maximum were 30 and 10 nsec. The output-pulse energy in cases a and b was 0.15 and 0.01 of initial (~50 mJ), respectively. With further increase of the optical density, the pulse became strongly attenuated; it could no longer be registered at D = 25.

Dediogio and Potenza [1] noted that in a solution of vanadium phthalocyanine, unlike in phthalocyanines of other metals, the transitions to the metastable triplet level from the excited singlet level, the lifetime of which for transitions to the ground level was 3 nsec, play an insignificant role. Therefore, under the experimental conditions, the absorbing centers can be regarded at two-center systems with $T_p \gg \tau \gg l/c$, and the transmission of the giant pulse through the absorbing medium is determined by relation (5). According to our measurements, at the absorbing-center density $N = 6 \times 10^{16}$ cm^{-3} used in our experiments, the cross section for the capture of a resonant-radiation photon was 2.6×10^{-16} cm^2.[2)] The presence of a spatial structure in the giant pulse [10-12] causes the bleaching of the medium to be uneven over the cross section, and introduces an uncertainty in the parameters characterizing the input and output pulses.[3)]

The value of P_{in} estimated from the calorimetric measurements, amounted to ~3 MW/cm^2 for the oscillograms shown in Fig. 2, so that at the indicated values of σ and τ the maximum of the parameter d was ~ 18. With the aid of relation (5) the input-pulse parameters taken from the oscillograms, and the values of the optical densities D, we estimated the durations of the output pulses at half-width. These values agreed, within 20%, with the data obtained from the oscillograms of the output pulses. The critical flux densities G_{cr} amount to 0.37 D mJ/cm^2 for a vanadium phthalocyanine solution. Under the experimental conditions, the value of $\int_0^{\tau/2} P_{in}(t)\,dt$ for the leading front of the input pulse was smaller than the critical value, owing to the long growth time, and therefore the bleaching of the medium corresponded to the case where the stationary front of the bleaching front did not have time to become established.

The experimentally observed reduction of the duration of the output pulse, and the increase of slope of its front, agree with the above-considered features of the propagation of a monochromatic pulse through a resonantly absorbing optically-dense medium with strong relaxation. Optically dense media can be used to control the duration, slope, amplitude, and spatial structure of monochromatic radiation pulses. It is therefore of interest to investigate in detail, theoretically and experimentally, the influence of the transformation properties of a resonantly-absorbing optically-dense medium when the input pulse has a complicated space-time structure. In this case it is advantageous to investigate simultaneously the space-time structure of the pulse before and after passing through the cell, using the methods developed by Ambartsumyan et al.[11] and Korobkin et al.[12]

[2)] An increase in σ was observed when the concentration of the solution was decreased.

[3)] In several of the experiments we limited the beam cross section by using an additional diaphragm made up of crossed spectrograph slits and placed outside the resonator ahead of the cell with the optically-dense medium. A reduction in the pulse duration was observed at different dimensions of this diaphragm, down to the smallest size used, 0.2 × 0.2 mm.

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