

Water and ethanol as bleachable radiation absorbers in an yttrium-erbium-aluminum garnet laser ($\lambda = 2.94 \mu\text{m}$)

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A passive shutter was developed for 2.94- μm radiation and was used to generate giant pulses with a laser based on an yttrium-erbium-aluminum garnet. The passive-shutter materials were associated liquids containing the hydroxyl group, namely water and ethanol. An energy of 20 mJ was obtained in a monopulse of 120 nsec duration with a TEM₀₀-mode diameter 1.7 mm.

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The burgeoning progress in the research into the 3- μm band in the recent year has led to the development of a laser using an yttrium-erbium-aluminum garnet crystal and emitting at a wavelength $\lambda = 2.94 \mu\text{m}$ (Ref. 1) at room temperature. Giant pulses were generated by this laser both with the aid of a rotating mirror as a Q switch² and with the aid of a lithium-niobate crystal as an electro-optical shutter.^{3,4}

We report here the development of a passive shutter for $\lambda = 2.94 \mu\text{m}$ and its use to generate giant pulses in an yttrium-erbium-aluminum garnet laser. The material of the passive shutter was ordinary water or ethyl alcohol (ethanol). It appears that other substances, containing a hydroxyl group, with an absorption band near the frequency 3400 cm^{-1} ($\lambda = 2.94 \mu\text{m}$) of the laser transition, will also be suitable for this purpose. We recall that in IR spectra the valence vibrations of the non-associated hydroxyl groups lie in the range 3670-3580 cm^{-1} . The participation of these groups in the formation of the intermolecular hydrogen bonds manifests itself in intense bands at lower frequencies (3550-3450 cm^{-1}).⁵ Thus, the condition of finding an absorption band of a hydroxyl group near the frequency 3400 cm^{-1} can be satisfied by associated liquids, such as water and ethanol.

Water as a substance for a passive shutter has attracted interest because of its tremendous damping coefficient κ at the wavelength 2.94 μm , which equals 0.3080 according to the data of Ref. 6. We note that the maximum value of the damping coefficient $\kappa_m = 0.3100$ is reached at $\lambda = 2.93 \mu\text{m}$, i.e., near the wavelength of interest to us.

Knowing the density of the water and the damping coefficient κ , we can easily obtain the transition cross section $\sigma_{\text{H}_2\text{O}}$ for water at the wavelength 2.94 μm :

$$\sigma_{\text{H}_2\text{O}} = 4\pi\kappa/\lambda N = 3.9 \cdot 10^{-19} \text{ cm}^2,$$

where N is the number of molecules of water per cm^3 . The intensity of the light with $\lambda = 2.94 \mu\text{m}$ decreases on passing through a layer of water in accord with the Bouguer law with an absorption coefficient $\alpha = 4\pi\kappa/\lambda = 1.32 \cdot 10^4 \text{ cm}^{-1}$.

The cross section for the laser transition between the Stark components $x_2(^4I_{11/2})$ and $\gamma_7(^4I_{13/2})$ at the wavelength 2.94 μm is, according to the data of Ref. 7, $\sigma_L = 2.6 \times 10^{-20} \text{ cm}^2$.

As a rule, in passive shutters based on saturable dyes the transition cross section reaches very high values ($\sim 10^{-16} \text{ cm}^2$) and exceeds by several orders of magnitude the laser-transition cross section (e.g., $\sigma_L = 2.5 \times 10^{-20}$ for ruby). The mechanism of bleaching these passive shutters is connected with the saturation of the absorption. In our case the cross sections differ by 15 times, and in principle the mechanism of bleaching via absorption saturation could operate here, too. What remains unknown, however, is the relaxation time τ of water from vibrational-rotational levels located near 3400 cm^{-1} , so that the question of the saturation intensity $I_s = \hbar\omega/2\sigma_{\text{H}_2\text{O}}\tau$ at the wavelength 2.94 μm remains open. Knowledge of the relaxation time τ would help understand the mechanism whereby a passive shutter with water becomes bleached, but for the time being we can only discuss a number of mechanisms for bleaching a passive shutter.

1. Saturation of a two-level system, such as water. It is meaningful to discuss this mechanism only in the case of not too short a relaxation time $\tau \geq 10^{-9}$ sec. In this case the saturation power density is $I_s \leq 85 \text{ MW/cm}^2$. Another open question is the possible stepwise absorption on higher-lying vibrational-rotational levels.

2. The "liquid-superheated vapor" phase transition, when the water heated by the radiation at the wavelength 2.94 μm goes over rapidly into the critical or even transcritical state with temperature $T \geq T_{\text{cr}}$ and pressure $p \geq p_{\text{cr}}$. In this case a two- or threefold increase of the initial volume occupied by the irradiated layer of water is sufficient to cause its density to drop by a factor 2-3 (we recall that $\rho_{\text{cr}} = 0.329 \text{ g/cm}^3$), and consequently to increase in suitable fashion the transmission of the passive shutter. This mechanism can operate only in the case of rapid vibrational-translational relaxation in water. As will be shown later, to produce a giant pulse it suffices to produce bleaching corresponding to a much lower increase of the initial volume.

3. It is possible also that the superheating of the water shifts the absorption spectrum, particularly because of the breaking of the hydrogen bonds between the water (ethanol) molecules, and the result is bleaching of the passive shutter. The energy of the hydrogen bond is 0.7-1.9 kJ/cm^3 (Refs. 5, 8). In the presence of rapid vibrational-translational relaxation in water, such an energy input to break the hydrogen bond can be easily

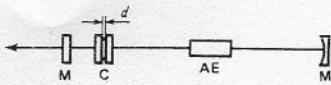


FIG. 1. Experimental setup with cell inside the laser cavity.

provided even at a power density 1.0–2.5 MW/cm².

4. Even slight heating that broadens the absorption band of the water can decrease the absorption at the laser-transition frequency enough to produce a giant pulse.

Thus, additional research is necessary to determine the operating mechanism of a water or ethanol passive shutter.

Leaving aside the question of the mechanism whereby the passive shutter becomes bleached in our case, let us dwell on the giant-pulse generation in an yttrium-erbium-aluminum garnet laser with the aid of this shutter.

The experiment was performed in the following manner. A cell consisting of two plane-parallel quartz plates of 1 mm thick and containing water and ethanol was placed in the cavity of a laser consisting of two dielectric mirrors with reflectances 60 and 100% and an active laser element between them (Fig. 1). The active element was a Y₃Al₅O₁₂:Er³⁺ crystal with Er³⁺ ion density 50 wt.%, in the form of a rod of 5 mm diameter and 140 mm length with plane-parallel end faces. The active element was pumped in a quartz mono-block by an IFP-1200 flash lamp into which a capacitor with C = 100 μF was discharged. The cell with water or ethanol was placed either normal to the radiation direction or at the Brewster angle. The resonator length was 125 cm. The thickness *d* of the liquid layer between the cell windows was chosen such that the transmission of the cell placed outside the resonator was approximately 50% in the case of weak free generation of the erbium laser. It was found that *d* ≈ 0.5–1 μm for water, and *d* ≈ 10–20 μm for ethanol, where the absorption is weaker. Both for water and for ethanol, the threshold pump energy needed to produce a giant pulse was 135 J. The duration of the flash of the IFP-1200 lamp was in this case 100 μsec. The threshold pump energy required for free lasing without a passive shutter in the resonator was 75 J. At a pump energy 150 J and at a repetition frequency 2 Hz, a stable single monopulse was observed for the cell with water in the laser cavity (Fig. 3). The output lasing energy was 15–20 mJ at a relative stability 20%. The temporal characteristics of the radiation of the laser were investigated with a thin-film pyroreceiver⁹ with time resolution ~10 nsec, the signal from which was fed to an oscilloscope. The duration of the monopulse at half maximum (Fig. 3b) was 100–130 nsec, depending on the pump energy. The spatial distribution of the



FIG. 2. Experimental setup with total internal reflection prism inside the laser cavity.

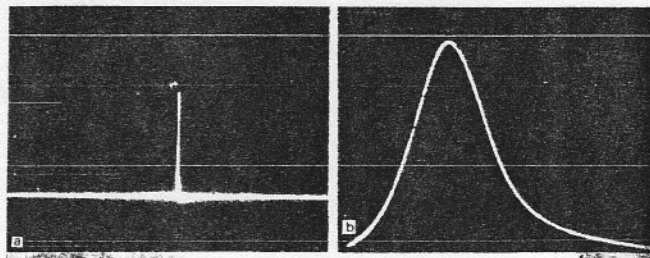


FIG. 3. Oscillograms of giant pulse. Sweep rates 20 μsec (a) and 50 nsec (b) per division.

radiation was monitored against the exposed part of the photographic emulsion and corresponded to the TEM₀₀ mode. No irreversible changes in the liquid layer were observed. When the threshold was decreased by 40–50%, a second monopulse was produced, lagging the first by 20–25 μsec, thus indicating a rather rapid restoration of the absorptivity of the shutter. Further increase of the pump increased the number of lasing pulses and decreased the interval between them, as is typical of lasers with saturable filters. In this case irreversible changes, in the form of reddish spots with dimension close to the diameter of the TEM₀₀ mode, were produced inside the cell.

The second experimental setup differed from the first in the cavity configuration, which contained a total internal reflection prism (Fig. 2) made of a material with large refractive index (LiNbO₃, *n* = 2.2). A drop of water or ethanol was spread on the reflecting face of the prism, thereby disrupting in part, owing to the absorption, the total internal reflection. The absorption of the intra-cavity radiation was due to its partial penetration into the liquid, to a depth of the order of the wavelength. The cavity had mirrors with 100% reflection, so that the glare from the end face of the prism was used as the output radiation. For a dry reflecting prism face the threshold pump energy for free lasing was 108 J. When water was coated on the reflecting face of the prism, at a pump energy 240 J, a giant radiation pulse was produced and damaged the reflecting face of the prism. The situation was similar with ethanol. The optical damage can apparently be attributed to the small initial transmission of the passive shutter, and accordingly to the large initial population inversion in the active element, which led to a very powerful giant radiation pulse.

For the first experimental setup (Fig. 1) we can estimate the degree of bleaching of the absorbing liquid. Knowing the duration of the giant pulse (~120 nsec), and the lifetime of the photons in the resonator (5–7 nsec) we obtain, following Ref. 10, that the ratio of the initial inverted population of the active element for the bleached shutter to the threshold inverted population is small, ~1.2, assuming instantaneous Q switching. This means that

$$\frac{\ln(1/T)}{\ln(1/T')} = \frac{2\alpha d + \ln(1/T_0)}{2\alpha' d + \ln(1/T_0)} = 1.2,$$

where *T* denotes the cavity transmission for a round trip with allowance for all the losses, *T*₀ is the trans-

mission without allowances for the losses in the passive shutter, d is the thickness of the liquid layer, the prime refers to the bleached state. At $T_0 = 0.35$ and $\exp(-2\alpha d) = 0.25$ we obtain $\alpha' = 0.6\alpha$, i.e., the absorptance decreases by 40%, and the shutter transmission per pass increases from 50 to 65%. The shape and duration of the calculated pulse following this change of transmission of the passive shutter agree well with the shape and duration of the experimentally recorded laser pulse.

Explanation of the mechanism whereby a passive shutter with water and other liquids containing hydroxyl groups becomes bleached will undoubtedly call for further research. It is clear, however, that to obtain a giant pulse in an yttrium-erbium-aluminum garnet crystal the required degree of bleaching of the passive shutter is relatively low.

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