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Measuring nonlinear reflectance of laser radiation with a wavelength of 2940 nm from silica glass–water and silica glass–ethanol interfaces

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Abstract. **We have measured the reflectance of an Er^{3+} : YAG laser pulse from silica glass–water and silica glass–ethanol interfaces at high (0.9 J cm^{-2}) and low (5 mJ cm^{-2}) energy densities of incident radiation. The nonlinearity of reflectance dynamics is found for high-power radiation during the laser pulse action.**

Keywords: *nonlinear reflectance, interface, Er^{3+} : YAG laser.*

1. Introduction

It is known that water has a high absorption coefficient ($\sim$1.3 $\times$ 10^4 cm^{-1}) for Er^{3+} : YAG laser light (λ = 2940 nm) [1]. This ensures a high energy contribution per unit volume of water (about 10^4 J cm^{-3}) in irradiation of its surface by Er^{3+} : YAG laser pulses (the input energy density of about 1 J cm^{-2}). If a *Q*-switched laser is used, water is heated during 100–200 ns, and in the interaction area the existence of a strongly non-equilibrium state of water, with a temperature of 1500–2000 °C [2] at a virtually constant density, becomes possible. The emergence, existence and decay of this state is accompanied by various effects, including acoustical [3–5] and optical [2, 6, 7]. For example, a significant reduction in the absorption coefficient of high-power Er^{3+} : YAG laser radiation in a thin water layer was found by Vodop'yanov [2]. Vodop'yanov et al. [7] also observed a strong decrease in the water refractive index in the case of radiation in the visible spectrum range. Therefore, studying the interaction of high-power IR radiation with water is of interest from the viewpoint of physics of nonequilibrium states of water.

Interest in studying the processes occurring in the interaction of high-power laser IR radiation with water is also stipulated by the practical needs of medicine, because biological tissues contain water and therefore readily absorb radiation at certain wavelengths in this spectral range. As an example, we may point out review [8], which highlights different aspects of the interaction of IR radiation with biological tissues, and paper [9], which presents the results of studying the ablation of biological tissues irradiated by high-power Er^{3+} : YAG laser pulses.

Since radiation with a wavelength of 2940 nm is absorbed in a thin layer of water, a change in its state should have the strongest effect on the reflectance from the water–substrate interface. Another liquid with a high absorption coefficient at this wavelength is ethanol. Water and ethanol at λ = 2940 nm have different refractive indices, and their absorption coefficients differ by several times. Furthermore, thermodynamic characteristics of these compounds are also different. In this regard, it is interesting to compare the reflectance behaviour of high-power laser pulses from the interfaces of fused silica with water and ethanol. In the present work, we have measured reflectances of high-power and attenuated IR radiation from fused silica–water and fused silica–ethanol interfaces, and also the reflectance dependence on temperature.

It should be noted that, in almost all previous studies on the interaction of high-power IR radiation with water [2, 3, 6, 7], measured were the characteristics of radiation passed through a cell with water. An exception is work [4] in which the reflectance of high-power radiation with λ = 2940 nm from the cubic zirconia (phianite)–water interface is measured in the total internal reflection regime. As far as we know, the reflectance measurements of high-power IR radiation from the fused silica–strongly absorbing liquid interface at the incidence angle close to normal have not yet been reported.

2. Scheme of the experiment and its results

The optical scheme of the setup is shown in Fig. 1. Radiation from a 2940-nm, actively *Q*-switched Er^{3+} : YAG laser falls on the interface between silica glass and liquid or air. The laser pulse energy is equal to 12 mJ, the pulse duration is about 220 ns and the transverse distribution is given by the TEM_{00} mode. The energy density at the interface is 0.9 J cm^{-2} for high-power radiation (strong field) and 5 mJ cm^{-2} in the case of attenuated radiation (weak field). Radiation reflected from the interface is recorded with photodiode (*4*), and incident radiation is measured with photodiode (*2*). In measuring the reflectance in a strong field, filters (*5*) are located in front of photodiode (*4*), while, in measuring the reflectance in a weak field, these filters are placed in front of lens (*3*) (shown in Fig. 1 by a dotted line). Temperature sensor (*11*) and heater (*10*) are installed inside cell (*8*). Diffuser (*12*) is mounted in the focal plane of lens (*9*) in front of photodiode (*4*). This design should eliminate the effect of divergence of radiation reflected from the interface on the signal from the photodiode.

The reflectance was measured in the following way. Constant components u_{01} and u_{02} were deducted from signals u_1 and u_2 recorded by photodiodes (*2*) and (*4*), respectively. To remove the high-frequency noise components from the signals $U_1 = u_1 - u_{01}$ and $U_2 = u_2 - u_{02}$, we performed their Fourier transform and removed all the Fourier components

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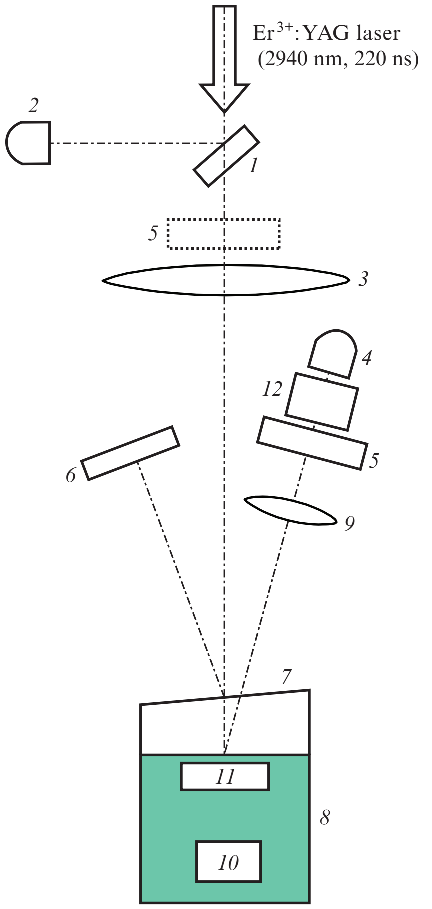

Figure 1. Scheme of the setup: (*1*) substrate outcoupling radiation to photodiode (*2*); (*2*) PD29 photodiode to record radiation incident on the water–silica glass interface (the angle of incidence on the interface is about 10°); (*3*) BaF_2 focusing lens (F = 300 mm); (*4*) measuring PD29 photodiode to record radiation reflected from the liquid–silica glass interface; (*5*) attenuating filters; (*6*) screen to attenuate radiation reflected from the front face of the silica glass substrate; (*7*) wedge-shaped silica glass substrate closing the liquid; (*8*) cell with water or ethanol; (*9*) CaF_2 lens (diameter 20 mm, F = 60 mm); (*10*) heater; (*11*) temperature sensor; (*12*) scatterer consisting of 14 GGG plates (thickness 350 μm, diameter 5 mm) installed in a polished copper pipe.

with a frequency higher than the intermodal beat frequencies. After that, the inverse Fourier transform is fulfilled. As a result, the signals U_{f1} and U_{f2} were obtained. At each time moment, the ratio $r = U_{f1}/U_{f2}$ of these signals were taken. The measurements were started at the moment when radiation in the measurement channel was reflected from the silica glass–air interface (let $r = r_0$ in this case). Then water (or ethanol) was poured into the cell, and the signals of the incident and reflected radiation were once again measured. If the ratio of the signals in this case was r_1, the reflectance for each time moment was calculated as $R = R_0 r_1/r_0$, where $R_0 = 3.0\%$ is the calculated reflectance from the silica glass–air interface at normal incidence (the refractive index n = 1.42 at the wavelength of 2900 nm [10]).

Figure 2 shows the measured values of reflectance from the silica glass–air interface in the strong and weak fields versus time, and also the incident radiation intensity normalised to its maximum. It is seen that the reflectance values for the weak and strong fields during the radiation pulse action are virtually constant and close to each other.

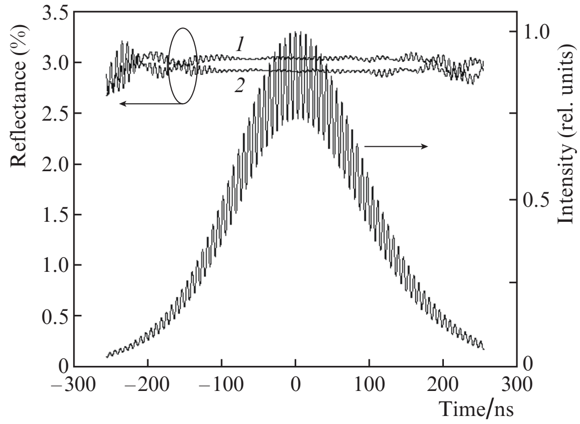

Figure 2. Reflectance from the silica glass–air interface during the laser pulse action in the case of [0.9 mJ cm^{-2}, (*1*)] strong and [5 mJ cm^{-2}, (*2*)] weak fields, as well as the incident light intensity normalised to the maximum.

Figure 3 presents the same reflectance dependences for the strong and weak fields, but from the silica glass–water interface. It can be seen that, in the case of a strong field, the reflectance decreases from 1.4% to its minimum value of 0.4% and then increases up to 0.7% at the pulse end. In the case of a weak field, the reflectance during the pulse is virtually constant and constitutes approximately 1.4%, which is close to the calculated value of 1.5% at a temperature of 0 °C, if the optical constants of water from [1] are used. The same Figure presents the pulses of incident and reflected radiation normalised to the relevant maxima. It is evident that the shapes of these pulses are different, which affects the time dependence of the reflectance.

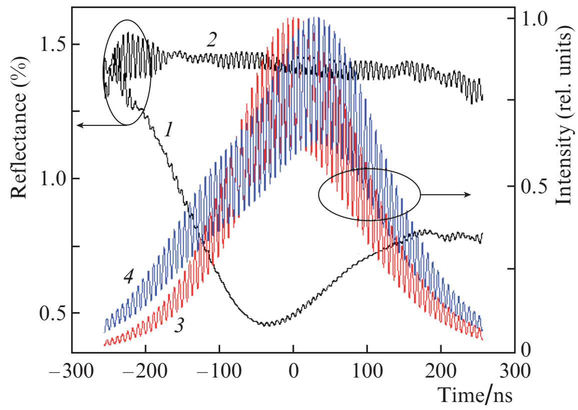

Figure 3. Reflectance from the silica glass–water interface during the laser pulse action in the case of [0.9 J cm^{-2}, (*1*)] strong and [5 mJ cm^{-2}, (*2*)] weak fields, as well as the incident (*3*) and reflected (*4*) signals in a strong radiation field normalised to the maximum.

Figure 4 shows the results of the same measurements for the silica glass–ethanol interface. It is seen that, at the pulse onset, the reflectance remains virtually constant and then increases from 0.28% to 0.5%.

The changes in reflectance both for water and ethanol after the onset of the laser pulse can be explained by heating of a thin liquid layer adjacent to the substrate. To test the possibility of this explanation, we have measured the depen-

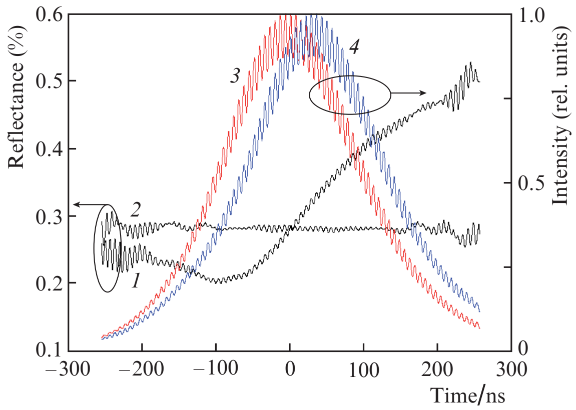

Figure 4. Reflectance from the silica glass–ethanol interface during the laser pulse action in the case of [0.9 J cm^{-2}, (*1*)] strong and [5 mJ cm^{-2}, (*2*)] weak fields, and also the incident (*3*) and reflected (*4*) signals in a strong radiation field normalised to the maximum.

dences of the reflectance on the temperature in a weak field (Fig. 5). One can see from this Figure that the reflectance of water decreases from 1.4% at 20 °C to 0.75% at 90 °C, which is in good agreement with the calculated value, if use is made of the data for the optical constants of water at a temperature of 0 °C and 50 °C given in [1]. In the case of ethanol, the reflectance does not depend on the temperature when it changes in the range of 20–70 °C. The measured absorption coefficient for ethanol at a wavelength of 2940 nm turned out equal to 3100 cm^{-1}.

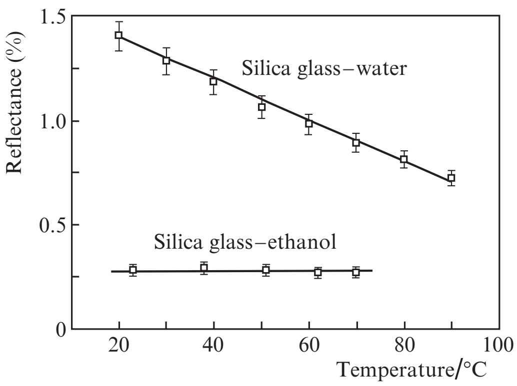

Figure 5. Reflectance from the silica glass–water and silica glass–ethanol interfaces in a weak field vs. temperature.

Another possible mechanism for reducing the reflectance is bleaching of water and ethanol under the action of high-power radiation [2], when the imaginary part of the refractive index of a medium is decreased, which should result in a decrease in reflectance.

The increase in reflectance by the end of the radiation pulse action for both water and ethanol can be explained by a decrease in the material density contacting the substrate, and by a possible phase transition.

3. Conclusions

We have measured the reflectance from the fused silica–water and fused silica–ethanol interfaces exposed to the action of a 2940-nm pulse. The reflectance of high-power radiation in the case of water decreases from 1.4% (at the onset of the pulse) to 0.4%, and then increases up to 0.7% by the end of the laser pulse. A decrease in reflectance can be explained by the water heating and bleaching under the action of high-power IR radiation, while an increase in reflectance – by a decrease in the density of the water layer being in contact with the substrate, which should lead to a decrease in the refractive index of water.

The ethanol reflectance in a strong field remains virtually constant at the beginning of the pulse action and then rises from 0.28% to 0.5%.

We have investigated the temperature dependence of reflectance from the interface between fused silica and water and fused silica and ethanol in a weak field. We have found that the reflectance of water decreases in the temperature range of 20–90 °C, while the reflectance of ethanol remains constant in the temperature range of 20–70 °C.

The experimental results suggest that, during the action of a high-power radiation pulse, the substrate is contacted with a heated material whose density is close to the density of the liquid both for water and ethanol, i.e. the substrate remains 'wet' in the course of the action of the laser pulse with an energy density of about 1 J cm^{-2}.

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