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Limitation of laser intensity using binary stratifying solutions

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Abstract. **The possibility of limiting laser intensity using light-induced critical scattering under optothermodynamic transfer of stratifying triethylamine–water and 2-butoxyethanol–water solutions to the unstable equilibrium state has been investigated. A decrease in the intensity of neodymium laser radiation (λ = 1.06 μm) by a factor of ~10 has been obtained for the triethylamine–water solution. The time dependences of the intensities of the incident light and the radiation transmitted through solution layers are compared.**

Keywords: *intensity limitation, laser radiation, stratifying solution, critical state.*

1. Introduction

Intensive development of laser technique and its wide implementation in the different fields of human activity requires new tools for protecting organs of sight and light sensors from light damage. Conventional devices for attenuating solar radiation and welder-flash light use glass or plastic color light filters. Light intensity can be reduced relatively slowly, taking into account the characteristic inertia time of human sight (~0.1 s) [1].

As for high-power lasers, the increase in their radiation intensity stimulated development of a new field of optical technique: limitation of the laser energy and power. The problem of limiting pulsed laser intensity, beginning with the pioneer studies [2, 3], is of great interest for researchers, as is evidenced by the papers [4–7]. A specific feature of the dynamic reduction of pulsed laser intensity is the light-induced change (for several tens of nanoseconds) in the absorbing, scattering, or spatial properties of the nonlinear optical material used in the laser radiation intensity limiter (LRIL).

Despite the existence of extensive information about the design and nonlinear optical media used in LRILs based on different mechanisms of limiting light intensity, the problem of reducing laser intensity to a level safe for practical applications remains unsolved. In addition, the necessity of excluding colour perception disturbance for operators of optical devices or narrowing significantly the sensitivity range of sensors (which is related to the LRIL selectivity) causes large difficulties. In our opinion, these spectral problems can successfully be solved using practically nonselective mechanisms of limiting radiation intensity; such processes include light-induced thermal scattering and focusing/defocusing laser radiation accompanied by its orificing.

Since the effect of thermal scattering, which is generally implemented in one-component liquids, is related to local boiling and formation of strongly scattering vapour bubbles, the response time of the corresponding LRILs is relatively long; thus, the case in point is the limitation of intensity of not very short laser pulses. In addition, thermal scattering can only be implemented in practice by introducing a rather large amount of energy into a liquid. The practical use of laser focusing/defocusing is limited by the possibility of initiating an optical breakdown in the LRIL scheme, which is a hazard for the protected organ of sight or optical device.

The problem of reducing the relatively low-power cw laser radiation formed by laser pointers is of particular importance and calls for new tools to reduce intermediate laser intensities [8]. For this reason, an urgent problem is to search for new nonselective mechanisms of limiting laser intensity, to which light-induced scattering of radiation in stratifying solutions (SSs) can be assigned [9–11].

The purpose of this study was to reveal the specific features of applying light-induced scattering in SSs near the critical stratification point (CSP) in order to limit laser intensity. One can satisfy the stringent requirements imposed on the experimental implementation of this mechanism (design of compact devices capable of operating under various conditions) using the optothermodynamic method of transforming a SS into the labile state through the CSP under laser irradiation. The essence of this method is the change in the thermodynamic state of liquid under its nonresonant interaction with high-power optical radiation. An advantage of this technique is that the pulse intensity and width can be varied in wide limits. In this case, absorption of even relatively low-power laser radiation by the working medium of limiter may significantly change the thermodynamic state of the medium [12].

2. Optothermodynamic transformation of stratifying solution into labile state

The early stage of light-induced scattering in the binary liquid methanol–cyclohexane SS was observed for the first time in [13] by studying the character of scattered He–Ne laser radiation. Reverse light scattering from the temperature anomaly in a SS was investigated in [14–16], where a significant influence of anisotropy fluctuations on the occurring processes was noted. The optothermodynamic method of measuring the spinodal position in a SS with a lower CSP was described

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in [17]. The kinetics of the spatial structure of liquid SSs at delayed convection in them was investigated in [18]. A comprehensive analysis of the potential of laser methods in the study of light-induced scattering was performed in [19–21], where the efficiency of light-induced pressure diffusion in the processes under consideration was also indicated.

In a binary SS, where the binodal and spinodal have a single common CSP, premature separation of the system into two phases as a result of heterogeneous nucleation hinders the solution existence in the phase state in the supercritical labile region between the binodal and spinodal. To achieve the vicinity of spinodal, one can use only nonequilibrium optothermodynamic processes, which, in particular, can be initiated by absorption of short laser pulses in a SS [22, 23].

Among binary SSs with a lower CSP, aqueous solutions of triethylamine (TEA) with a lower CSP and 2-butoxyethanol (BOE) (Butyl Cellosolve), which possesses both lower and upper CSPs, are of practical interest for us. For the TEA–water SS, the critical stratification temperature is T_{cr} = 291.32 K and the critical concentration C_{cr} = 32.12 wt %. For the BOE–water SS, with a lower CSP, we have T_{cr} = 321.44 K and C_{cr} = 30.14 wt % [24–27].

Under optothermodynamic effect on the phase state of a SS with a lower CSP with light absorption, the initially homogeneous solution passes to the metastable region. Laser radiation absorption in SSs causes formation and growth of a new phase in the labile region, which is accompanied by the formation of refractive index inhomogeneities and intense radiation scattering (light-induced critical opalescence [28]). These phenomena accompany the light-induced spinodal decay in liquids, which was observed in [10] for a BOE–water solution with a critical concentration exposed to a pulsed xenon lamp.

When studying experimentally the laser intensity limitation in critical SSs using light-induced critical opalescence, one must take into account that an increase in the temperature deviation from critical reduces the degree of fluctuations development in the system. As a result, the influence of the light-induced spinodal decay of the 'seed' structure (an ensemble of nonequilibrium heterogeneous centres) on the process decreases, thus making the laser intensity limitation less efficient. In turn, an increase in the penetration depth into the instability region, which is determined by the final temperature of the system (this temperature depends on the energy density and the incident laser pulse duration), makes it possible to obtain a state with a high degree of lability, i.e., to increase the efficiency of laser intensity limitation.

3. Experimental

The TEA and BOE compounds under study, with a purity of no less than 97%, were dried by alkali, treated using activated coal, and then subjected to vacuum distillation. As a result, the degree of their purity (determined by chromatography) was no worse than 99.5%. This careful purification makes it possible to avoid a temperature shift for phase stratification due to the influence of chemical impurities. The mass concentration of the organic components of solutions with distilled water was measured with an error of ~1 wt %.

The limitation of high-power laser intensity using SSs implies transfer of the latter from the initial thermodynamic equilibrium to the lability region at a temperature as close to CSP as possible; therefore, careful thermal stabilisation of solutions must be performed. The thickness of the solution layer interacting with radiation was 3 mm.

A cell was placed in a bulky stainless-steel chamber filled with a coolant (circulating distilled deionised water) and connected to a thermostat through hoses. The coolant was thermally stabilised with an error of ±0.1 K.

The system loses its initial thermodynamic equilibrium under laser irradiation; therefore, to repeat analysis, one must intensively stir the liquid to return it to the initial state. To enhance this process, a small air bubble was introduced into the solution. This procedure, along with damping pressure in the liquid (which arises during solution irradiation) facilitated its subsequent, more intense mixing. Mixing of the liquid under additional mechanical agitation of the cell was performed after each experiment for 10–15 min to eliminate temperature and concentration gradients in the solution. This technique makes it possible to recover thermodynamic equilibrium in the system for two to three hours after placing the cell in the chamber [12].

The specific features of applying light-induced scattering in SSs near the CSP were studied on an experimental bench; one of its elements was a Nd:glass laser with pump pulse energy and duration ~8 kJ and ~1.5 ms, respectively. The laser operated in the single-pulse regime with a pulse repetition period of ~3 min.

The scheme for measuring the characteristics of SSs absorbing radiation of the Nd:glass laser (*1*) is shown in Fig. 1. To make the distribution of laser intensity incident on the cell from the solution more uniform, the laser beam with an initial diameter of ~20 mm was orificed to a diameter of ~7 mm, due to which one could cut the central (most uniform) part of the beam. The time distributions of the lasing power at the input and output of the cell (*3*) with a 3-mm-thick solution layer were measured using photodiodes (*6*), the signal of which was applied to a double-beam storage oscilloscope. The radiation energy at the input and output of the cell was measured by solid-state calorimeters (*7*). The laser beam transverse cross section was monitored by irradiating photo paper (*8*). The lasing energy density could be varied using a set of neutral light filters (*9*).

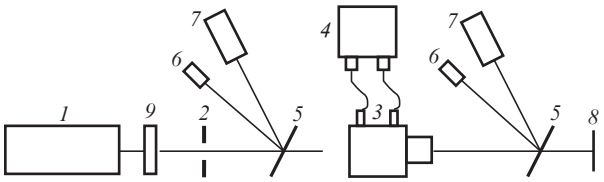

Figure 1. Schematic of the setup for studying the optothermodynamic characteristics of stratifying solutions: (*1*) laser, (*2*) diaphragm, (*3*) cell, (*4*) thermostat, (*5*) deflection plates, (*6*) photodiodes, (*7*) energy meters, (*8*) photo paper, and (*9*) a set of neutral light filters.

The response of the system to the optothermodynamic effect was aligned and studied using probe single-mode He–Ne laser radiation (λ = 633 nm, power ~0.5 mW, beam diameter ~1 mm) directed along the beam axis of the neodymium laser.

The approach of the SS to the equilibrium state in the early stage of light-induced spinodal decay excited by radiation of the laser was monitored by measuring the intensity of probe He–Ne laser radiation transmitted through the SS layer. This radiation was recorded using a photoelectron multiplier, located in the plane of a real enlarged image of the

exposed SS volume; orificing of the photoelectron multiplier reduced the diameter of the probed region of the solution to ~0.5 mm at the point of intersection of neodymium laser beam with the solution layer. An interference light filter was placed at the photoelectron multiplier input to select the probe laser radiation transmitted through the SS.

4. Results

The measured transmission spectra of the SSs were used to determine the absorption coefficient at the neodymium laser wavelength 1.06 μm. For the TEA–water and BOE–water solutions it was, respectively, 0.16 ± 0.01 cm^{-1} and 0.17 ± 0.01 cm^{-1}. To increase the solution penetration rate and depth into the stratification region under laser irradiation, water-soluble salt $CuCl_2$ was introduced into the SS; this additive increased the SS absorption at λ = 1.06 μm by a factor of about 10.

The measured dependences of the transmittance of the solutions under study on the intensity I of incident laser radiation (with working LRIL characteristics), obtained at different initial deviations ΔT_i of the SS temperature from the critical value, are shown in Fig. 2. It can be seen that the attenuation coefficient of the incident radiation intensity, $k = T_0/T$ (T_0 is the initial transmittance), increases with a decrease in ΔT_i. The k values are maximum at ΔT_i = 0.01 K and $I \sim$ 55 kW cm^{-2} and amount to ~10 and ~4 for TEA–water and BOE–water SSs, respectively.

It is noteworthy that the laser radiation is significantly reduced in the SSs at an average laser intensity several orders of magnitude lower than in the case of typical LRILs (10–100 MW cm^{-2}) [4–7].

An analysis of the oscillograms of laser pulses before and after their transmission through the cell with a nonlinear medium (Fig. 3) shows that the leading edge of a pulse transmitted through the SS approximately coincides with that of the incident laser pulse. In contrast, the trailing edge of the transmitted pulse is shifted to the pulse beginning, which is caused by the attenuation of the incident laser radiation in the nonlinear optical medium (SS). For the TEA–water system this effect is less pronounced because of lower laser intensity.

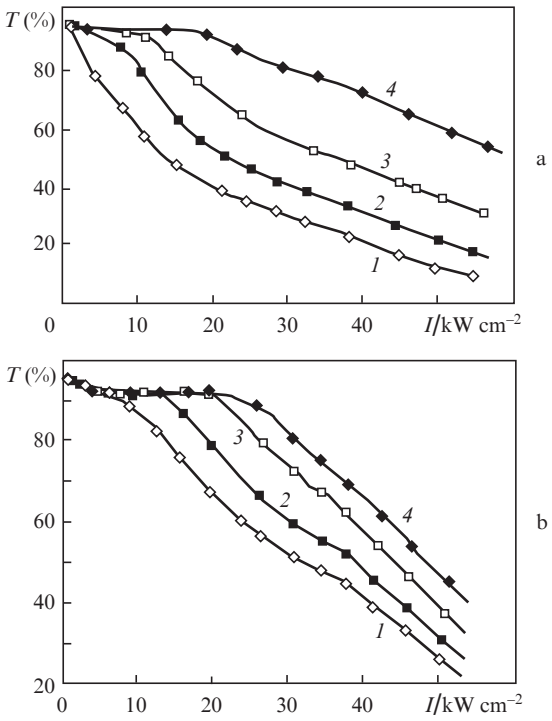

Figure 2. Dependences of the transmittance T on the laser radiation intensity I (λ = 1.06 μm) incident on a cell with (a) TEA–water and (b) BOE–water SSs. The initial deviations from critical temperature, ΔT_i, are (a) 0.01 (*1*), 0.05 (*2*), 0.20 (*3*), and 0.55 (4) K and (b) 0.01 (*1*), 0.05 (*2*), 0.20 (*3*), and 0.90 (*4*) K.

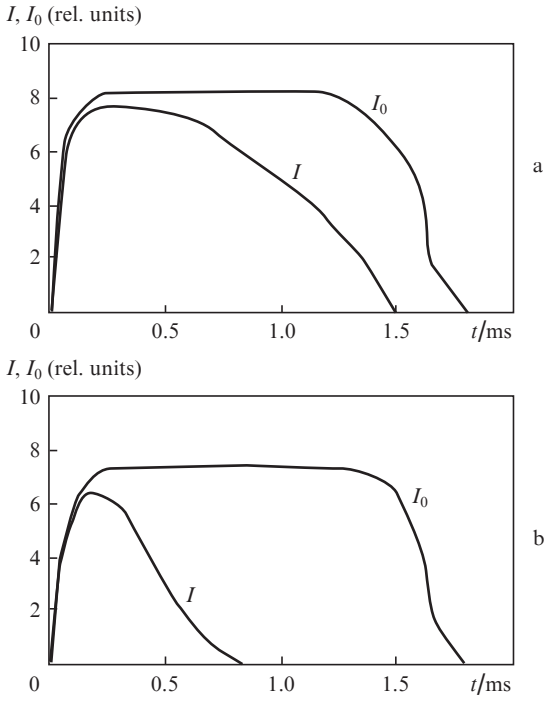

Figure 3. Oscillograms of the intensities of the incident radiation pulses (I_0) and the pulses transmitted through solution layers (I) for (a) TEA–water and (b) BOE–water SSs at ΔT_i = 0.01 K; I_0 = (a) 8 and (b) 35 kW cm^{-2}.

5. Conclusions

The study of the laser radiation attenuation in critical TEA–water and BOE–water SSs showed that a labile state (phase instability) may arise, which relaxes into the stable state with the formation of microheterophase concentration inhomogeneities and their subsequent stratification, which enhances radiation scattering near the critical temperature. The IR laser intensity was limited using light-induced critical scattering of light. The characteristics of LRILs based on the investigated solutions were measured at different incident laser intensities and different initial deviations ΔT_i of the temperature of liquids from critical. The attenuation of the incident radiation increases with a decrease in ΔT_i and reaches a maximum value (~10) for the TEA–water SS. In this case, the measured laser intensities are relatively low: several orders of magnitude smaller than in the case of typical LRILs.

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