DIRECT OBSERVATION OF REFRACTIVE-INDEX GRADIENTS PRODUCED IN A LIQUID BY AN ULTRASHORT LASER PULSE

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As is well known [1, 2], self-focusing and self-modulation of ultrashort laser pulses (USP) lead to a natural limitation of the pulse power, and therefore a study of the changes produced by USP in the refractive index n and leading to the indicated phenomena is of considerable interest. We have observed directly, for the first time, gradients of n produced when single USP are focused in a liquid.

The experiment was performed with the setup shown in Fig. 1. A single USP of duration not longer than 20 psec (resolution limit of the high-speed camera) with energy $\sim 10^{-2}$ J was focused with a lens of focal length 5 cm into the center of a cell 5 cm long filled with the investigated liquid. The gradients of n were photographed with a Schlieren system from the side of the cell, the illumination being produced by second harmonic ($\lambda = 0.53$ μ) of the same USP but delayed in time. The delay time could be varied in a wide range accurate to 3 psec, so that it was possible to trace the dynamics of the development of the gradients of n. The time resolution of the method (the photograph exposure time) did not exceed the duration of the USP. The single USP used in the experiment was first passed through a stable two-component medium, which made it possible, in addition to amplification, to improve the parameters (contrast and duration) of the USP [3]. The knife-edge of the Schlieren system was placed in such a way that it was possible to photograph gradients dn/dx satisfying the condition $\ell(dn/dx) \geq 8 \times 10^{-3}$, where ℓ is the dimension of the inhomogeneity in the direction of the illuminating beam.

The experiments have demonstrated the following:

1. Gradients of n are produced in liquids by single USP, and the effect has a threshold. Accurate to within the order of magnitude, the threshold is the same for ethanol and nitrobenzene, and equals $\sim 10^{-2}$ J.

2. The gradients of n occur in both liquids within a time ≤ 5 psec.

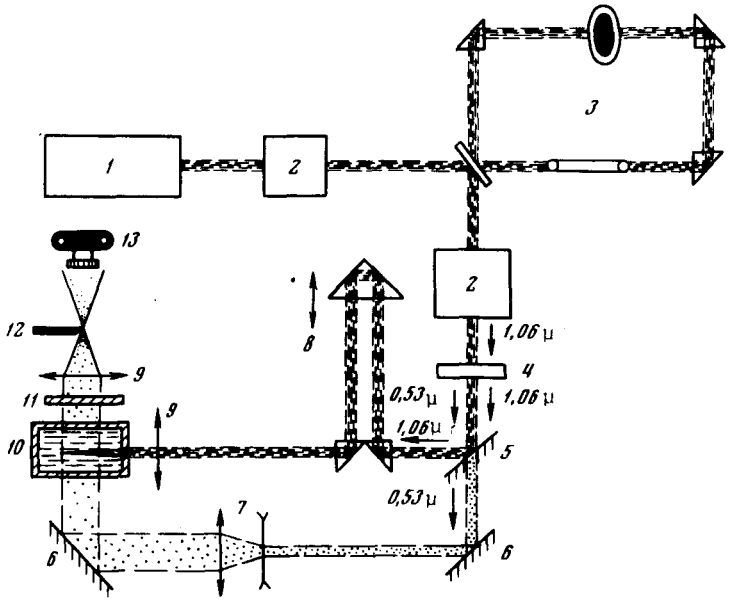

Fig. 1. Experimental setup: 1 - USP generator, 2 - system for separating a single USP from a train, 3 - stable two-component medium, 4 - second-harmonic converter, 5 - mirror reflecting the radiation with $\lambda = 1.06$ μ; 6 - mirror reflecting radiation with $\lambda = 0.53$ μ; 7 - telescope, 8 - adjustable optical delay, 9 - focusing lenses, 10 - cell with investigated liquid, 11 - SZS-22 light filter, 12 - knife edge of Schlieren system, 13 - photographic camera.

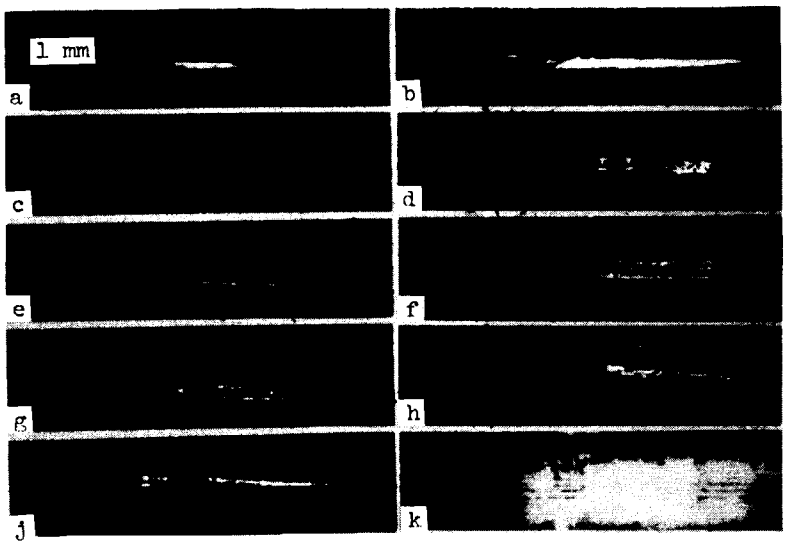

Fig. 2. Schlieren photographs of gradients of n: a - nitrobenzene, c - ethanol, initial instant; b - nitrobenzene, delay 1 nsec; d - nitrobenzene and e - ethanol, delay 75 nsec; f - nitrobenzene and g - ethanol, delay 120 nsec; h, j - nitrobenzene, case of two succeeding pulses separated by 10 nsec; k - nitrobenzene, case of a train of USP. The laser beam direction is from left to right. The gradient of n is directed upward (the knife edge is horizontal).

3. The gradients produced in nitrobenzene have the form of strongly inhomogeneous conical (sometimes cylindrical) formations with length from 3 to 5 mm and minimum transverse dimension ~100 μ. Inside these cones there are thin (not thicker than 10 μ) filaments, whose length fluctuates from several tens of microns to 5 mm; they may be even longer, but then they exceed the field of view of the recording system. The thin filaments are usually located where the field gradient is maximal - on the edges and along the axes of the cones (Figs. 2a and 2b). Such a picture agrees with the large-scale filaments observed in [4]. No large-scale filaments were observed in ethanol, where only one thin filament not longer than 3 mm, rather inhomogeneous in length (Fig. 2c), appears at the initial instant.

4. The large-scale filaments in nitrobenzene last approximately 1 - 2 nsec, after which they vanish.

5. In both liquids, cylindrical or spherical shock waves propagate respectively from the thin filaments or from the individual points where the jump of n originates. Their initial velocities exceed slightly the sound velocity (Figs. 2d - 2g). After approximately 30 nsec, the shock-wave velocities become equal to the sound velocity. The waves have very sharp leading fronts and more gently sloping and "ragged" trailing edges.

6. The gradients that appear at the very outset in the form of points or filaments are observed at the center of the shock wave for at least 120 nsec following the instant of their appearance, and their dimensions remain unchanged in time. The gradients become noticeable inside the wave only at sufficiently large delays (75 and 120 nsec). This is connected in all probability with the fact that by that time a sufficiently large rarefaction is produced inside the shock wave.

7. From photographs obtained with large delays it is seen that gradients are produced in nitrobenzene both in the form of filaments and in the form of individual points, whereas in ethanol either individual points or clusters of points are observed (Figs. 2d - 2g).

8. When several USP act in succession in nitrobenzene, the succeeding pulses are "captured" by the gradients of n produced by the preceding ones. Such a "capture" occurs either at the filaments themselves (Fig. 2j), or in the shock waves that propagate from them (Fig. 2h), if these waves have not yet left the focal volume. No formation of "new" gradients by the action of the

succeeding pulses is observed, that is to say, only the first pulse of the sequence is self-focused in pure form. The same occurs in ethanol, but no "capture" of the succeeding pulses is observed in this case.

Thus, the dynamics of the phenomenon reduces to the following. Gradients of n, leading to self-focusing of the radiation, are produced in the liquid by the USP within a very short time. Self-focusing causes a rapid local heating of the liquid, as a result of which shock waves propagate from the heating regions (filaments or points). The local-heating regions spread out slowly at the thermal-diffusion rate. The next USP entering the liquid encounters in the focal region gradients of n in the shock wave and in the regions of local heating at the center of the shock wave; these gradients can "capture" part of the USP energy, leading to a further increase of n in the gradient-localization regions. A rarefaction region is produced inside the shock wave, where self-focusing is thus hindered. Figure 2k shows a Schlieren photograph obtained with a USP train in nitrobenzene. One can see in the center of the photograph bright converging filaments and shock waves propagating from them, in the form of a superposition of many instantaneous pictures of each pulse of the train. Unlike the picture for one pulse, the filaments seem brighter here than the waves, since they exist during the entire USP train and produce the same picture for each pulse. Photographs of the same type, taken through crossed polaroids, were obtained in [5], but the authors erroneously identified the shock waves with the self-focusing filaments.

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SPIN-LATTICE RELAXATION TIME IN SF_6 IN THE CRITICAL TEMPERATURE REGION

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Investigation of transport processes in the vicinity of the liquid-vapor critical point by the method of nuclear magnetic resonance are very important for the study of the liquid state. It is therefore of interest to investigate in this region the behavior of the spin-lattice NMR relaxation time T_1, which is determined in a liquid by the character of the thermal motion of the individual molecules and of their nearest neighbors.

Reports have already been published on the temperature dependence of the spin-lattice relaxation time along the liquid-vapor coexistence curve in different substances [1 - 3], but no detailed investigations have been made as yet of the behavior of T_1 at constant density, when the distances between the molecules remain constant on the average, and as the temperature changes T_1 depends only on the character and velocity of the molecule thermal motion.