## ABSORPTION OF LASER RADIATION IN TRANSPARENT SUBSTANCES UNDER CONDITIONS OF APPEARANCE OF OPACITY

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The effects of absorption of laser radiation (free generation regime) in transparent organic dielectrics are studied. The absorbed part of the energy comprises a large fraction of the incident energy, about 0.5 to 0.8. The absorbed energy referred to the area of the fissures is  $1.1 \pm 0.22$  J/cm<sup>2</sup>. A dependence of the absorptivity of liquids on light intensity is observed. The absence of ultrasound generation is proved, and the role of other mechanisms in producing opacity followed by destruction of the solid is discussed.

I N the study of the destruction produced in transparent solid materials by laser light and in the elucidation of the nature of the appearance of opacity in these substances, the question of the effect of absorbed energy naturally arises. Experiments<sup>[11]</sup> have shown that in the presence of destruction the laser beam is greatly attenuated by passing through the sample; however, it follows from these same experiments that a fraction of the light is reflected or scattered from the fissures that are produced, so that, generally speaking, it is impossible to attribute all of the attenuation of the beam to absorption.

This paper presents for the first time, as far as we know, the results of a direct measurement of the absorbed energy of laser light. The experimental arrangement is shown in the figure. Light from a ruby laser 1, operating in the free generation regime, was focused by lens 3 (f = 70 mm) inside the sample 5. The samples were of polymethylmethacrylate (PMA) and polystyrene (PS) of dimensions  $43 \times 9 \times 9$  mm. The point of focus was usually located at a distance of 27-29 mm from the entrance face of the sample. Specially designed meters for measuring the light energy were used, made up of fine insulated wire wound in the form sketched in the figure. The light energy incident on the inside surface of the device heated it up, and this heating was determined by the change in its total electrical resistance. The meter A served to determine the initial energy of the beam  $E_0$ , a known fraction of which was deflected toward it by the transparent plane-parallel plate 2. The sample under investigation 5 was placed inside the meter B, which registered the energy Et transmitted through the sample, including that scattered and reflected from fissures. Finally, meter C should register the energy  $E_r$  reflected or scattered by the sample in the reverse direction and hence not acquired by B. The energy absorbed inside the sample is then, obviously,  $E_a = E_0 - E_t$  $- E_r$ . The lower limit of the sensitivity of the meter C was approximately 0.001 J.

The principal series of measurements was made at energies  $E_0 \approx 0.5-1.1$  J. At lower initial energies the measurement errors were large; at higher energies, expanding fissures would break the surface and the gas evolved from them<sup>[2]</sup> could strongly affect the indications of meter B. It was found that in PMA the fraction



of absorbed energy was  $E_a/E_0\approx 0.46\pm 0.04$ , and in PS.  $0.84\pm 0.03$ . The accuracy of relative measurements of energy by our meters for PMA was not worse than 9%, for PS, not worse than 4%. Significantly, device C did not give indications, i.e., the light energy reflected or scattered backwards was less than 0.01 J, taking into account the sensitivity of 0.001 J and the reflection coefficient of plate 2 of the order of 0.1. This, in our opinion, is direct experimental proof of the absence of ultrasound generation, so that in spite of many assertions in the literature, the ultrasound mechanism cannot play a role in the destruction of a material by a non-giant laser pulse.

Besides measuring the absorbed energy, we computed the total surface of the fissures formed in the sample. The error in this was not greater than 20%. It was found that the absorbed energy, referred to the area of the fissures, averaged over all investigated samples of PMA and PS, was  $1.1 \pm 0.22 \text{ J/cm}^2$ . This quantity, of course, should not be considered as the energy expended in creating cracks in the material, since a significant portion of the energy goes to the formation and heating of gas.<sup>[2]</sup> as well as to the heating and deformation of the layer of solid around a crack.

As has been established by experiment,<sup>[2]</sup> the principal reason for the expansion of fissures in PMA and PS is the loosening up of material by the pressure of the gas formed as a consequence of absorption of light energy and heating of the substance at places that have lost transparency. However, the reasons for the appearance of this opacity remain unclear.

It must be assumed that any kind of process in this respect ought to show up in investigating liquids. In fact, if the sites of absorption in solids are formed at places where there are inhomogeneities, defects, or impurities, then in very pure transparent liquids absorption should not arise. On the other hand, if the opacity appears in the host medium as a consequence of non-linear, e.g., two-photon, absorption, then a liquid should behave in general just like a solid (remember that for  $\lambda \approx 3500$  Å, corresponding to  $2h\nu$  for ruby, PMA, PS, and many other transparent substances already have low transparency).

We investigated the absorption of light in distilled water and the monomer of PMA, methylmethacrylate (MA) (the degree of freedom from inhibitor was not very high). The laser beam was passed through a 40-mm liquid layer. We measured the incident and transmitted beam energy. In order to calculate the contribution of reflections from the bottom of the cuvette and the surface of the liquid, measurements were made with empty and filled cuvettes. The measurements showed that by focusing the beam in a region outside the liquid, i.e., illuminating the liquid with a nonfocused beam of low intensity, the water absorbed about 4%, which agrees with a coefficient of ordinary linear absorption  $\kappa \approx 10^{-2} \text{ cm}^{-1}$ , but by focusing the beam inside the water, the absorption rose to 17%. In the case of MA the absorption amounts to respectively 2.4 ( $\kappa \approx 0.6 \times 10^{-2} \text{ cm}^{-1}$ ) and 16%. Thus, although we do observe an increase in absorption in the liquid upon a sharp increase in light intensity, the absorption in liquids under the same conditions of irradiation is much less than in solids. This obviously points to the important role of inhomogeneities as centers of absorption in solids. One should think that high-speed photography, as in<sup>[2]</sup>, of phenomena in liquids will permit settling the question of whether or not sites of absorption are localized in them as in solid polymers (unfortunately, the indicators of absorptionfissures, by which one can judge where absorption occurs, do not stay put in liquids). We are preparing to carry out such an investigation.

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In conclusion, we shall present some considerations that contradict the hypothesis of two-photon absorption of ruby quanta as a mechanism of nonlinearity.

It is known that light of the neodymium laser acts on polymeric materials in about the same way as does ruby light. At the same time, it is not  $2h\nu_{Nd}$  but only  $3h\nu_{Nd}$  that falls in the region of low transparency, and the probability of three-quantum absorption, in contrast to two-quantum absorption, is very low, based on estimates for experimental intensities.

In addition, the opacity boundary in MA, which special measurements show to lie in the region  $\lambda \approx 3500$  Å, is somewhat shifted toward longer wavelengths compared to PMA, i.e., two-photon absorption in MA ought to occur more easily than in PMA. The measured absorption in MA, however, is much less than in PMA.

We intend to publish more detailed experimental results and an analysis of possible absorption mechanisms.

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