Selective laser heating and nonlinear light scattering in a homogeneous medium

M. V. Belyaev, A. P. Maĭorov, V. A. Smirnov, and V. P. Chebotaev
Institute of Thermal Physics, Siberian Branch, Academy of Sciences of the USSR

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A scattering of light by temperature inhomogeneities which are produced by laser radiation and whose centers are associated with individual absorbing molecules has been observed. This is the first report of the observation of such a scattering. The scattering intensity observed experimentally agrees well with a theoretical prediction.

1. Let us consider a transparent liquid containing a few molecules that absorb light at a wavelength λ. Since essentially all the absorbed energy is converted into heat if quenching collisions occur, each absorbing molecule is a point source of heat. In the steady state, the deviation of the temperature from the average temperature around this point heat source is $\Delta T(r) = P/4\pi\kappa r$, where P is the amount of heat evolved per unit time, and κ is the thermal conductivity. To derive some simple estimates, we assume that the smallest value of r for which this expression is valid is $r_1 = 2.5 n_l^{-1/3}$, where n_l is the density of molecules of the liquid. For water we have $r_1 \approx 7.8 \times 10^{-8}$ cm. For an absorption cross section $\sigma = 1.5 \times 10^{-15}$ cm^2 and a laser intensity $I = 1$ GW/cm^2 we would have $P = \sigma I = 15$ erg/s and $\Delta T(r_1) \approx 260$ K. A significant heating of the medium in the immediate vicinity of an absorbing molecule can have a wide variety of consequences, in particular, a distinctive nonlinear scattering of light, which may be taken as evidence of the effect. Since the refractive index depends on the temperature, each absorbing molecule is a center of an optical inhomogeneity, whose magnitude is proportional to the intensity of the incident light. The scattering of light by such inhomogeneities differs qualitatively in its basic physics from both nonlinear scattering by small particles[1] and scattering by thermal gratings created by an interference of two light beams.[2] In the present letter we report an observation of this new type of nonlinear scattering in an optically homogeneous medium.

2. We assume that the light scattered by different inhomogeneities is summed in an incoherent way. We can then use the well-known expression for the Rayleigh scattering of linearly polarized light:

$$I_s = I \frac{\pi^2 V}{\lambda^4 r^2} A \sin^2 \Phi [1 + BI^2]. \qquad (1)$$

Here I and I_s are the intensities of the incident light and the scattered light, V is the observed volume, Φ is the angle between the field vector of the incident light and the observation direction, and the constant A is a measure of the molecular scattering of light in the given liquid. The 1 inside the brackets corresponds to ordinary molecular scattering, while the second term reflects the temperature inhomogeneities:

$$BI^2 = \frac{n_{eff}}{A}\left(\frac{d\epsilon}{dT}\right)^2 (\int \Delta T dV)^2 = \frac{n_{eff}}{3A}\left(\frac{d\epsilon}{dT}\right)^2 \left(\frac{\tau \sigma I}{c\rho}\right)^2, \text{ at } \tau \ll \tau_{st} \quad (2)$$

$$= \frac{n_{eff}}{A}\left(\frac{d\epsilon}{dT}\right)^2 \left(\frac{l^2 \sigma I}{2\kappa}\right)^2, \text{ at } \tau \gg \tau_{st}. \quad (3)$$

Here c is the specific heat, ρ is the density of the liquid, τ is the length of the incident light pulse, σ is the cross section for absorption of light at the given wavelength, $\tau_{st} = l^2 c\rho/\kappa$, $d\epsilon/dT$ is the total derivative of the dielectric constant with respect to the temperature, $l = n_{eff}^{-1/3}$ is the average distance between inhomogeneities, and n_{eff} is the density of molecules at resonance with the incident light.

3. Figure 1 shows the apparatus used in the present experiments. The light source is an LTIPCh-8 YAG laser operating at a repetition frequency of 12.5 Hz at a wavelength of 0.53 μm in one transversed mode. The length of the output pulse is 20 ns, and the maximum energy in the pulse is 1.2 mJ. The laser light is focused into a cell holding an aqueous solution of iodine by a spherical or cylindrical lens. The light intensity is varied over a dynamic range of 10^3 by a set of filters and is monitored by a photodiode. The maximum light intensity at the lens focus is 250 MW/cm^2. The intensity of the scattered light is measured with an FÉU-118 photomultiplier in the direction perpendicular to the propagation of the laser beam. We measure the intensity of the scattered light as a function of the intensity of the incident laser beam, $I_s(I)$, for distilled water and for a dilute iodine solution ($n = 2.8 \times 10^{17}$ cm^{-3}); the results are shown in Fig. 2. The linear behavior for the distilled water corresponds to ordinary Rayleigh scattering. The solid line is a least-squares fit on the basis of expression (1). The value of BI^2 found in this manner is $BI^2_{max} = 15$ at the maximum intensity. In the course of the experiments, we also measured the absorption coefficient of the solution for various intensities of the incident light. We observed no saturation. When a cylindrical lens is used, the scattering intensity is independent of the angle between the axis of the cylinder and the observation direction; we conclude that the observed scattering is thus not a stimulated scattering, in agreement with our model. This circumstance is a fundamental distinction between the effect observed here and type II stimulated thermal scattering.[4]

4. We estimate n_{eff} from the known absorption coefficient ($\alpha = 0.21$ cm^{-1}) and the dipole moment of the electronic transition,[5] $d = 1$ D. Assuming a homogeneous line half-width $\Gamma = 10^{12}$ s^{-1}, we find $n_{eff} = 1.4 \times 10^{14}$ cm^{-3}. Correspondingly, we

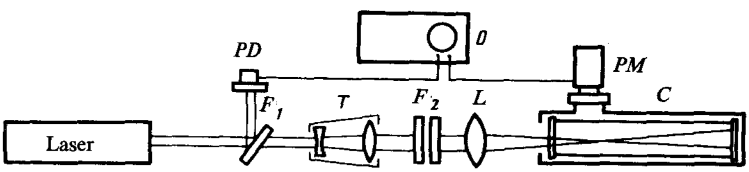

FIG. 1. The experimental arrangement. *PD*—LFD-2 photodiode: *O*—S1-75 oscilloscope: F_1, F_2—filters which absorb light at $\lambda = 1.06$ and $\lambda = 0.53$ μm, respectively; *T*—telescope; *L*—lens with $F = 100$ mm; *C*—cell holding the solution.

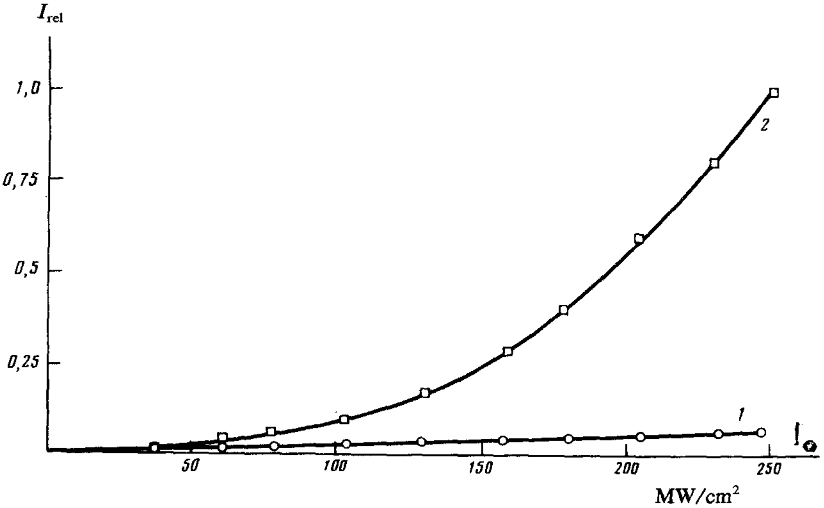

FIG. 2. Experimental intensity of the scattered light versus the intensity of the laser beam in the cell holding the solution. 1—Distilled water; 2—dilute aqueous solution of iodine ($n = 2.8 \times 10^{17}$ cm^{-3}).

have $l = 2 \times 10^{-5}$ cm, $\tau_{st} = 65$ ns, and $\sigma = 1.5 \times 10^{-15}$ cm^2. Since $\tau < \tau_{st}$, we can use (2) for an estimate. Setting $A = 10^{-24}$ cm^{-1} (p. 121 in Ref. 3) and $(d\epsilon/dT)^2 = 5.23 \times 10^{-8}$ K^{-2}, we find $BI^2_{max} = 7.8$, in comparison with the value of 15 found experimentally. We judge the agreement to be good in view of the approximate nature of the formulas used here. According to the estimate in Section 1, the value of ΔT at the center of the inhomogeneity at an intensity $I = 250$ mW/cm^2 is 65 K, while the average heating of the water at the focus is 0.4 K.

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