NONLINEAR SCATTERING OF LIGHT BY SMALL INHOMOGENEITIES IN CORUNDUM CRYSTALS

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Light scattering by small optical inhomogeneities in corundum crystals is investigated for various radiant energy densities. The angular and frequency dependences of the scattering intensity are studied. Nonlinear effects are discovered in the scattering of high density light (from a Q-switched ruby laser). The effects are revealed in the appearance of oscillations and in the nonlinear dependence of the scattered light intensity on the incident radiant flux. The effects observed are interpreted on the basis of a thermal mechanism of nonlinearity of the index of refraction of the scattering centers with allowance for heat removal to the ambient medium.

1. INTRODUCTION

In the propagation of powerful light radiation in media with absorbing inhomogeneities, the effects of strong light scattering can appear on the so-called refracting halos[1] (bubbles in a liquid,[2,4] defects in solids, "diffusion or gas-kinetic propagation of evaporated material from inhomogeneities"[1]).

In addition to such effects, nonlinear scattering of light can take place in inhomogeneous media, due to changes in the index of refraction of the scattering inhomogeneities and of the medium surrounding them in a field of an intense light wave.[4] Such changes in the index of refraction can be produced by different mechanisms (Kerr effect, electrostriction, thermal effects).

The purpose of the present research was the discovery and investigation of the nonlinear light scattering from optical inhomogeneities in transparent media. The experiments were carried out on corundum crystals containing scattering centers. For the clarification of the nature of these centers and the mechanism of nonlinear scattering of the light, we also studied the ordinary (linear) scattering at low intensities of the incident light. The following light sources were used in the experiments: in the region of low intensities, an incandescent light and a He-Ne laser; in the region of high intensities, a ruby laser operating in the regime of gigantic pulses.

2. LIGHT SCATTERING AT LOW INTENSITIES OF THE INCIDENT RADIATION

To explain the nature and properties of the scattering centers, we undertook the study of the frequency and angular dependences of the scattering intensity. The first was studied in the range of 600–900 nm with the use of an incandescent lamp as a light source, with a radiation spectrum close to the spectrum of a black body with a temperature T = 3000° K. The block diagram of the arrangement for detection of the scattering is shown in Fig. 1. Since the intensity of the scattered

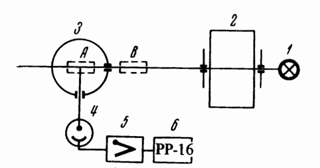

FIG. 1. Block diagram of the experimental setup for the investigation of the frequency dependence of light scattering: 1—incandescent lamp, 2—monochromator, 3—integrating sphere, 4—photomultiplier, 5—broadband amplifier, 6—scaling apparatus.

light was very low, the detector (of type FÉU-79) operated in the regime of photon counting with 100-second accumulation time. The background of the photomultiplier amounted to no more than 30 pulses per second.

The corundum samples had the shape of cylinders of diameter 14 mm and length 80 mm with polished lateral surfaces. The optic axis of the crystal was perpendicular to the geometric axis of the sample, which was placed inside an integrating sphere (point A in Fig. 1).

The observed dependence of the intensity of scattered light on the wavelength of the incident radiation is described by the formula

$$I = I_0 + I_1(\lambda),$$

where the term I_0 does not depend on λ and $I_1(\lambda)$ is proportional to λ^{-4}. The relative weight of I_0 and I_1 varies strongly from sample to sample and the typical value of I_0/I_1 was ~0.1 for λ = 600 nm. The frequency dependence of the term $I_1(\lambda)$ shows that it is connected with scattering from small inhomogeneities, the dimensions of which $a \ll \lambda$.

Investigation of the scattering diagrams (we used an He-Ne laser in these experiments) in a plane containing the optic axis of the crystal and perpendicular to the direction of propagation of the incident light showed that they are also described by the scattering formula for small particles (Rayleigh scattering):[5]

$$I_R = I_0 \frac{9\pi^2 n_0^4}{\lambda^4 r^2} |\alpha_0|^2 \sin^2 \varphi, \qquad (1)$$

where φ is the angle between the direction of observation of the scattered light and the electric vector **E** of

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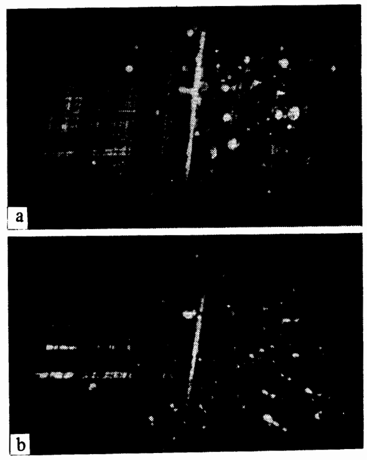


FIG. 2. Microphotographs of scattering particles: a—before annealing of the crystal, b—after annealing in an oxygen atmosphere.

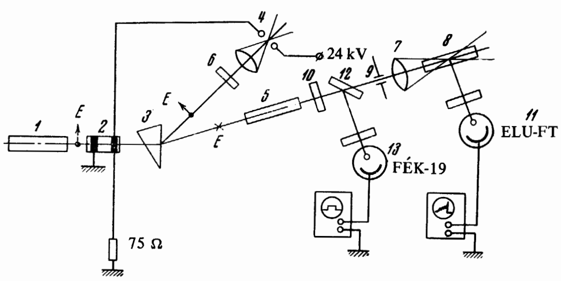


FIG. 3. Block diagram of the experimental apparatus for observation of nonlinear light scattering: 1—master oscillator, 2—electro-optical crystal, 3—prism of Iceland spar, 4—discharger, 5—amplifier, 6 and 10—light filters, 7—focusing lens, 8—sample, 9—disphragm, 11 and 13—light detectors, 12—delay plate.

the incident light wave, r is the distance from the point of scattering to the point of observation, α_0 is the polarizability of the scattering center ($\alpha_0 = a^3(m^2 - 1)/(m^2 + 2)$, $m = n_1/n_0$ is the index of refraction of the inhomogeneity relative to the surrounding medium).

We note that the intensity of the scattered light for fixed φ changes upon rotation of the optic axis of the crystal relative to the vector **E**, which indicates the anisotropy of the polarizability of the scattering centers. In scattering along the optic axis, it is approximately 12% greater than in the direction perpendicular to it. A similar anisotropy of α_0 was also observed in [6].

Ultramicroscopic investigations carried out by us with use of the He-Ne laser as the light source showed that the mean distance between the scattering centers, was $\bar{l} \simeq (5-6)\times10^{-4}$ cm. They were distributed preferentially along the boundaries of the blocks of the lines of intersection of them with the slip lines (Fig. 2).

An estimate of the absolute intensity of the scattered light showed that the coefficient of the integrated scattering $\gamma_{sc} \approx 10^{-5}$ cm^{-1}, and the total losses on passage along the sample, measured on the apparatus of Fig. 1 for location of the sample outside the integrating sphere (point B), have the value $\gamma_{tot} \approx 6 \times 10^{-3}$ cm^{-1} for a wavelength of 850 nm.

Thus, in the corundum samples we studied, the losses by scattering (at large angles) was much less than the absorption loss.

Investigation of the conditions of formation of scattering centers shows that they appear in annealing of the crystals in a CO atmosphere in the presence of atomized carbon (Fig. 2a) and disappear in vacuum annealing and annealing in an oxygen atmosphere (Fig. 2b). The nature of these particles remains unclear. Probably they appear as the result of diffusion of foreign impurities (carbon) along the defects of the crystalline lattice in the time of annealing and are connected with the chemical reactions of carbon with aluminum.

3. LIGHT SCATTERING FOR HIGH INCIDENT RADIATION DENSITIES

For the study of scattering in the region of high intensities of the incident light, we used a ruby laser system (Fig. 3), consisting of a master oscillator which operated in a Q-switched regime with a saturable filter and a power amplifier. A solution of cryptocyanin in nitrobenzene was used as the saturable filter. By variations of the concentration of the solution and the length of the resonator, the duration of the emitted radiation pulses of the master oscillator was changed within the range 8–50 nsec. Here the pulses had a smooth bell shape. In a series of experiments for the study of the kinetics of the scattering, we also used radiation pulses of rectangular shape, which were obtained by cutting off the leading and trailing edges of the bell-shaped pulses of the master oscillator with the aid of a Pockels electro-optical shutter. The latter consisted of a KDP crystal and a prism of Iceland spar which assured the spatial splitting of the rays with different polarizations. The electrical circuit supplying the shutter consisted of cable lines and a nitrogen high-pressure discharger (12 atm), controlled by the light radiation of the laser.

The shutter thus described made it possible to separate a rectangular pulse of duration 18 nsec, with fronts no worse than 3 nsec, from an output bell-shaped pulse. of 50-nsec duration. The location of the cut-off part on the pulse of the master oscillator was regulated by the cable delay lines and by the change in the light intensity incident on the discharger, by means of light filters. We note that, for most complete suppression of the leading and trailing edges of the radiation pulse of the master oscillator, it is necessary that its crystal have the minimum value of the anomalous double refraction.

After amplification, the radiation of the laser was focused by a lens (F = 30 cm) inside the sample studied. A diaphragm located in front of the sample served to limit the cross section of the beam. Change in the level of the radiation density in the sample was produced by means of calibrated light filters, and its maximum value was $P_{max} \approx 10^9$ W/cm^2.

The scattered light was recorded at an angle of 90° by means of a cathode-ray amplifier of the type ÉLU-FT with resolution no worse than 2×10^{-9} sec and the oscillograph I2-7. For control of the pulse envelope, a coupler and receiver of the type FÉK-19 was used.

The results of the investigation of the scattering of powerful light radiation by the setup described above are shown in Fig. 4. Here, oscillograms are given of the scattered radiation for different shapes and lengths

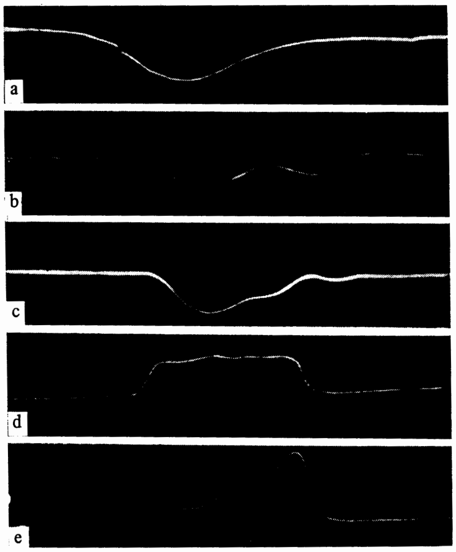


FIG. 4. Oscillograms of pulses of incident and scattered radiation: a—pulse of incident radiation with length 50 nanosec, b—pulse of scattered radiation at an angle $\theta \approx 65°$, c—$\theta \approx 115°$, d—pulse of incident radiation with length 18 nanosec, e—pulse of scattered radiation for $\theta = 90°$ and power of incident radiation $P_0 \approx 0.9P_d$.

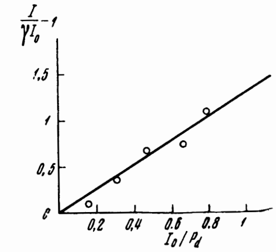
of the pulses of incident radiation for several scattering angles θ. It is seen from these oscillograms that the shape of the pulse of scattered radiation is materially different from the shape of the incident light pulse. Oscillations of intensity are clearly seen in the scattered light, the period of which depends on the scattering angle (see Figs. 4b and c). Moreover, it is seen on the oscillogram of Fig. 4e, which was obtained for a rectangular pulse, that the average intensity of the scattering increases from beginning to end of the pulse.

The investigations showed that marked peculiarities in the scattering are noted only for sufficiently high densities of the incident radiation $P_0 \approx 10^8-10^9$ W/cm^2. Upon lowering of the intensity of the incident light, the extent of the oscillations and the degree of rise of the mean intensity of scattering fall off rather rapidly and even at $P_0 \lesssim \frac{1}{3} P_d$, where P_d is the threshold power for destruction of the samples, the shape of the pulse of the scattered light is practically identical with the shape of the pulse of the incident radiation. The value of P_d in our experiments was not measured precisely; one could only note that it lay in the range 10^8-10^9 W/cm^2.

We note that the effects observed in the scattering for $P_0 < P_d$ are not connected with any irreversible processes in the sample (of the type of microfractures). This is confirmed by the fact of the reproducibility, from scintillation to scintillation, of the shape and amplitude of the scattering signals for $P_0 < P_d$, which was always violated for $P_0 > P_d$, when irreversible structural changes (defects) appeared in the samples.

The data obtained by us indicate the nonlinear character of the light scattering in corundum crystals for high densities of the incident radiation. For the purpose of making clear the nature of the nonlinearity, in addition to the effects of change in shape of the pulses of scattered light that we noted above, we also investigated the spectrum of scattered radiation and the dependence of the peak intensity of the scattered light on the intensity of the incident radiation I_0, the pulse of which had a smooth bell shape with length 8 nsec. The pulse of scattered light in this case also had a smooth shape without oscillations and its peak intensity followed the law (see Fig. 5) $I \approx \gamma I_0(1 + \beta I_0)$, where γ and β are the constants of linear and nonlinear scattering. Approximate measurements of these quantities gave $\gamma \approx 10^{-5}$, $\beta \approx 1.3/P_d$.

FIG. 5. Dependence of the peak intensity of scattered light on the power of the incident radiation (τ = 8 nanosec).



Investigation of the spectrum of the scattered light showed that it was identical with the spectrum of the incident radiation of the laser, with accuracy up to the resolution of the Fabry–Perot interferometer used, $\Delta\nu \approx 0.1$ cm^{-1}.

4. DISCUSSION OF THE MECHANISM OF NONLINEAR SCATTERING

The nonlinear effects in light scattering can be due to changes in the index of refraction both in the scattering inhomogeneities and in the surrounding medium (the crystalline matrix) in the strong field of the light wave.[4]

Here we shall attempt to explain the experimental data given in the preceding sections by assuming that the observed nonlinear effects of scattering are connected with heating of the scattering particles by the laser radiation. Such heating can lead both to a change in the index of refraction of the inhomogeneities and to a nonstationary change in the index of refraction of the crystalline matrix in the vicinity (as a consequence of the heat transfer).

Thus the absorbing centers themselves and the thermal waves propagating from them are scattering centers for high densities of the incident light radiation. For analysis of the scattering in this case, we can use the methods of the scattering theory of Rayleigh–Gans for particles with an inhomogeneous distribution of the index of refraction.[7] The intensity of the scattered light from such complicated objects (particle + thermal wave in the surrounding medium) can be written in the form

$$I = \frac{I_R}{|\alpha_0|^2}|R_1 + R_2|^2, \tag{2}$$

where I_R is the intensity of the ordinary Rayleigh light scattering, determined from Eq. (1), R_1 and R_2 are corrective coefficients which take into account the nonlinearity of the polarizability of the scattering center and surrounding medium, respectively, due to their heating from the light radiation. Following [7], we can write down the expressions

$$R_1 = \frac{1}{v_0}\int_0^a 4\pi r\alpha_1 \frac{\sin br}{b}\,dr, \tag{3}$$

$$R_2 = \frac{1}{v_0}\int_a^{r_m} 4\pi r \alpha_2(r)\frac{\sin br}{b}\,dr, \quad (4)$$

for the corrective coefficients, where v_0 is the volume of the scattering center, α_1 and α_2 are volume polarizabilities of the center and surrounding medium, respectively, which are assumed to have spherical symmetry, $b = 2k_0 \sin(\theta/2)$, $k_0 = 2\pi n_0/\lambda$, r_m is the radius of the thermal wave in the medium at the instant of time t. By assuming the heating of the center to be homogeneous, and of the surrounding medium to fall off as $r^{-1}[1 - \Phi(r/2r_m)]$ (a diverging spherical thermal wave), we can write for α_1 and α_2

$$\alpha_1 = \alpha_0 + \Delta\alpha_1,$$
$$\alpha_2(r) = \Delta\alpha_2 \frac{a}{r}\left[1 - \Phi\left(\frac{r}{2r_m}\right)\right], \quad (5)$$

where $\Delta\alpha_1$ and $\Delta\alpha_2$ are the corrections to the polarizabilities which depend on the intensity of the incident radiation, $\Phi(r/2r_m)$ is the probability integral. We then get for R_1 and R_2

$$R_1 = \alpha_0 + \Delta\alpha_1, \quad (6)$$

$$R_2 \approx \frac{3\Delta\alpha_2}{a\cdot b^2}\left\{1 - \left[1 - \Phi\left(\frac{1}{2}\right)\right]\cos br\right\}. \quad (7)$$

For the thermal mechanism assumed by us, the nonlinearities of the polarizability $\Delta\alpha_1$ and $\Delta\alpha_2$ can be written as

$$\Delta\alpha_1 \approx \left(\frac{dn_1}{dT}\right)\frac{\varkappa P a^2}{6\pi k_1}, \quad (8)$$

$$\Delta\alpha_2 \approx \left(\frac{dn_0}{dT}\right)\frac{\varkappa P a^2}{6\pi k_2}, \quad (9)$$

where P is the flux power density of the incident radiation, κ the absorption coefficient of the material of the inhomogeneity, k_1 and k_2 the coefficients of thermal conductivity of the material of the inhomogeneity and the surrounding medium, respectively. We note that Eqs. (3), (4) are valid under the condition of weakness of the inhomogeneities, which in our case reduces to the following:

$$2k_0\int_0^{r_m} |m(r) - 1|\,dr < 2\pi k_0 a \Delta\alpha_2 \ln\frac{t}{t_{\rm char}} \ll 1. \quad (10)$$

A comparison of Eq. (2), in which R_1 and R_2 are given by Eqs. (6) and (7), with the experimental data (Sec. 3) shows that the thermal mechanism of nonlinearity of the polarizability of the scattering centers gives a satisfactory account of the observed oscillations of the scattered light and the dependence of the peak intensity of the scattering on the incident power. From (7), we have for the time of the first oscillation

$$\tau_{\rm osc} = c\rho\lambda^2/4\pi k_2 n_0^2 \sin^2\frac{\theta}{2}. \quad (11)$$

This formula explains qualitatively the observed increase in the time of the first oscillation for decrease in the scattering angle (see Figs. 4b, c). For $\theta = \pi/2$, this time is seen to be equal to $\sim 10^{-8}$ sec, which is close to the observed value $(0.7\text{–}0.9)\times 10^{-8}$ sec. The estimate of the value of the coefficient of modulation of the intensity of scattered light $\eta = (I_{max} - I_{min})/I_{max}$, which depends on the nonlinearity parameters $\Delta\alpha_1$ and $\Delta\alpha_2$, shows that it agrees with the experimentally observed values $\eta = 0.1\text{–}0.5$, if we assume that the absorption coefficient of the scattering particles is $\kappa \approx 10^4$ cm^{-1}. This value of κ does not contradict the measured loss coefficient $\gamma_m = 6 \times 10^{-3}$ cm^{-1} in the sample (Sec. 2). We note that for carbon, the presence of which in the samples studied by us is quite probable (see Sec. 2), $\kappa \approx 10^5$ cm^{-1}.[7] For the estimates of η, we have assumed $(dn/dT) = 10^{-5}$ deg^{-1} (a value that is typical for transparent solids).

The observed dependence of the peak intensity of the scattered light on the power of the incident radiation is also well explained by formula (2) if the parameters of nonlinearity $\Delta\alpha_1$ and $\Delta\alpha_2$ are sufficiently small in comparison with α_0. The latter is in agreement with the estimate of the ratio $\Delta\alpha_2/(\alpha_0 + \Delta\alpha_1) \simeq 4\times 10^{-3}$, which is obtained from the modulation coefficient η.

We note that all the foregoing analysis is based on the single particle scattering model, which is valid for distances between the scattering centers $l \gg \lambda$. In the crystals investigated by us, this condition was satisfied in the mean (see Sec. 2), however, clustering of the scattering centers could take place with $l \gg \lambda$. For such clusterings, the effect of overlap of the thermal fields from the individual centers can have a significant effect on the time dependence of the scattering intensity. It is seen from the given analysis that this effect leads to a growth in time of the mean scattering intensity for $t \lesssim c\rho L^2/k_2$, where L is a characteristic dimension of the clustering of the scattering centers. Estimates have shown that this effect can be responsible for the observed temporal growth of the scattering intensity.

One of the possible mechanisms leading to an increase of the scattering intensity with time can also be the nonlinear absorption of the light by the scattering centers.

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