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Abstract

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PHYSICS

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NONLINEAR SCATTERING OF LIGHT BY SMALL PARTICLES

(Presented by Academician A. M. Prokhorov on 7 VII 1969)

In connection with the appearance of powerful light sources—lasers—numerous different nonlinear optical effects have been predicted, discovered, and investigated (self-focusing, various types of stimulated light scattering, etc.). In the phenomena of stimulated scattering studied up to the present (combination scattering, Mandelstam–Brillouin scattering), the “seed” sources of scattering are **dynamic** optical inhomogeneities of the medium (molecular vibrations, fluctuations of density, concentration, etc.).

In this note we wish to draw attention to the possibility of the existence of a new nonlinear effect—nonlinear scattering of light by **static** optical inhomogeneities, such as foreign impurities and inclusions in optically transparent media. This possibility follows from the fact that the refractive indices of the material of the scattering particles n_p and of the medium n_m in which they are located may depend on the intensity of the incident light (as occurs in other nonlinear effects, for example self-focusing), and not in the same way, so that the relative refractive index of the particles $n = n_p/n_m$ may vary with the field strength of the light.

Following the methods developed in ordinary (linear) optics of turbid media (see, for example, ⁽¹⁾), let us analyze the nonlinear scattering effects caused by the dependence of the refractive index of the scattering particles on the intensity of the incident light. The scattering problem, as is known, is a problem of diffraction of light and reduces to the solution of Maxwell’s equations under definite boundary conditions. In the general case of particles of arbitrary size and shape this problem proves to be very complicated even in linear optics, and it is greatly complicated if the refractive index of the particles depends on the intensity of the incident light. Therefore we shall confine ourselves here to the practically frequently encountered case of scattering by small particles, whose dimensions $a \ll \lambda$ (λ is the wavelength), and whose mean separation $l \gg \lambda$ (Rayleigh scattering). In addition, we shall consider only single scattering, for which the parameter $\tau = a^2 R/l^3 \ll 1$, where R is the size of the scattering region. In the approximations indicated, the light field inside the particles is homogeneous, all particles of the medium scatter independently, and the

total scattering intensity, being the sum of the scattering intensities I_s of the individual particles, is much smaller than the intensity of the incident light I_0 . In this case the solution of the problem of light scattering by particles whose refractive index depends on the strength of the electric field of the light wave is considerably simplified, and the result essentially reduces to the fact that in the final formulas of the ordinary theory of light scattering one must put the refractive index equal to a specified function of the field. Thus, for scattering of linearly polarized light by spherical particles we have ⁽¹⁾:

$$I_s = \frac{16\pi^4 a^6}{\lambda^4 r^2} \left| \frac{m^2 - 1}{m^2 + 2} \right|^2 (1 - \sin^2 \theta \cos^2 \varphi) I_0. \quad (1)$$

Here λ is the wavelength of the incident radiation in the medium in which the scattering particle is immersed; r , θ , and φ are polar coordinates ($\beta = \pi - \theta$ is the scattering angle, measured from the direction of the incident ray); m is the relative complex refractive index of the particle material. For absorbing and conducting particles we have:

$$m = n - i\chi, \quad m^2 = \varepsilon' - i(\varepsilon'' + 4\pi\sigma/\omega), \quad (2)$$

where n and χ are, respectively, the refractive index and absorption coefficient of the particles; $\varepsilon = \varepsilon' - i\varepsilon''$ is the dielectric constant; σ is the conductivity; and ω is the frequency of the incident radiation. The nonlinear dependence of the scattering intensity on the intensity of the incident radiation may be associated with the dependence of either the real or the imaginary part of the refractive index on the field; this dependence may be caused by various mechanisms, such as the Kerr effect, electrostriction, and direct or multiphoton absorption. For example, for nonabsorbing dielectric particles one may assume

$$m^2 = n^2 = \varepsilon_0 + \varepsilon_2 E^2, \quad (3)$$

where ε_0 and ε_2 are constants independent of the electric-field strength E of the light wave. Substituting (3) into (1), we obtain

$$I_s = k \left(\frac{\varepsilon_0 - 1 + \varepsilon_2 E^2}{\varepsilon_0 + 2 + \varepsilon_2 E^2} \right)^2 f(\theta, \varphi) I_0, \quad (4)$$

where

$$k = \frac{16\pi^4 a^6}{\lambda^4 r^2}, \quad f(\theta, \varphi) = 1 - \sin^2 \theta \cos^2 \varphi.$$

It follows from this formula that the character of the dependence of I_s on I_0 is essentially determined by the relation between the quantities $\varepsilon_2 E^2$ and $\varepsilon_0 - 1$. For $\varepsilon_2 E^2 \ll \varepsilon_0 - 1$, this dependence will be quadratic,

$$I_s = k_0 I_0 + k_2 I_0^2 \quad (5)$$

(where

$$k_0 = k \left(\frac{\varepsilon_0 - 1}{\varepsilon_0 + 2} \right)^2 f(\theta, \varphi), \quad k_2 = \frac{8\pi k (\varepsilon_0 - 1) \varepsilon_2}{c n_m (\varepsilon_0 + 2)} f(\theta, \varphi),$$

c is the speed of light, and n_m is the refractive index of the scattering medium), whereas for $\varepsilon_2 E^2 \sim \varepsilon_0 - 1$ it becomes more complicated.

It is interesting to note that, since scattering is determined by the relative refractive index of the particles and the surrounding medium, the nonlinearity parameter for scattering

$$\xi = \varepsilon_0 E^2 / (\varepsilon_0 - 1)$$

may be considerably larger than the analogous parameter

$$\eta = (\varepsilon_2 E^2 / \varepsilon_0)^{1/2},$$

which characterizes the self-focusing properties of nonlinear media (2-4). This circumstance indicates that in some media the effect of scattering nonlinearity may be observed at substantially lower intensities of light beams than the self-focusing effect.

It should be noted that formula (4) is valid, generally speaking, only for the case in which the effect of scattering nonlinearity is due to the dependence $\varepsilon(E)$ for the material of the scattering particles, although, as indicated above, it also occurs when the quantity ε for the surrounding medium depends on the field. The latter case is considerably more difficult to analyze, since here the field in the medium is not homogeneous, as in the case of light scattering by small particles, and, in addition, the self-focusing effect may be superimposed. In the presence of both effects, the scattering problem becomes mathematically much more complicated and requires computer calculations.

The nonlinear effects analyzed may play a significant role in the propagation of light in various gaseous and condensed media containing foreign impurities or structural defects. In particular, in doped laser crystals, owing to the nonlinear growth of the scattering intensity, scattering may considerably exceed nonresonant losses at high radiation powers. For the mechan-

nonlinearities that have short characteristic establishment times, this source of losses may be a significant factor limiting the output power of laser generators and amplifiers of nanosecond and picosecond pulses.

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