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NONLINEAR SCATTERING OF LIGHT IN INHOMOGENEOUS MEDIA

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A theory of the nonlinear scattering of light in inhomogeneous media is developed. Different mechanisms (the Kerr optical effect, electrostriction, and heating) are considered to account for nonlinearity of the refractive index. Expressions are derived for the intensity and spectrum of the scattered light. Particular cases of scattering by small particles and of stationary scattering are analyzed.

INTRODUCTION

WHEN light is scattered by optical inhomogeneities (such as foreign particles in transparent media) one can observe nonlinear effects that result from the dependence of the refractive indices of these inhomogeneities and the surrounding medium on the intensity of the light. Different mechanisms (the Kerr effect, electrostriction, or thermal effects) can account for a variable refractive index.

We have observed experimentally the nonlinear scattering of light by small optical inhomogeneities in corundum crystals.[1] The observed effects were interpreted on the basis of a thermal mechanism for the nonlinearity of the refractive index of the scattering centers with allowance for heat transfer to the surrounding medium.

It is of interest to consider the problem of nonlinear light scattering more generally for the different mechanisms that may account for the nonlinear refractive index of the inhomogeneities, and allowing for dynamic effects that can arise in the field of an intense light wave and alter the scattered light spectrum.

Our present work is a general analysis of nonlinear light scattering in media containing inhomogeneities described by a spherically symmetric variation of the refractive index, which is also assumed to be quadratically dependent on the field strength of the incident radiation. This formulation encompasses a wide range of media and of mechanisms for a nonlinear refractive index. The results are applicable to an analysis of nonlinear light scattering by small static optical inhomogeneities.[2]

1. THEORY OF NONLINEAR LIGHT SCATTERING IN WEAKLY INHOMOGENEOUS MEDIA

We know that in the general case the refractive index, which is a complex quantity, is determined completely by the state of the medium, i.e., by the temperature, electric field, and deformation (T, E_i, u_{ik}) distributions in the medium. A light wave induces variation of the refractive index because the latter is dependent on the field strength; also because of nonuniform heating associated with the imaginary part of the refractive index, and the propagation of elastic deformations.

In a centrally symmetric medium the variation of the refractive index can be described by

$$\Delta n_{ik} = (\partial n_{ik}/\partial T)_{E_i, u_{ik}} \Delta T + \beta_{iklm} E_l \dot{E}_m + p_{iklm} u_{lm}, \tag{1}$$

where β_{iklm} and p_{iklm} are electro-optical and elasto-optical coefficients, u_{lm} is the deformation tensor, $\Delta T = T - T_0$, and T_0 is the initial temperature of the medium. Here only the first nonvanishing terms in the expansion of the refractive index are taken into account.

We consider a spherically symmetric isotropic medium having volume polarizability $\alpha_0(r)\delta_{ik}$. An electromagnetic field induces anisotropy of the medium. We shall confine our analysis to effects that lead to spherical anisotropy:

$$\alpha_{rr} \neq 0, \quad \alpha_{\theta\theta} = \alpha_{\varphi\varphi} \neq 0, \quad \alpha_{r\varphi} = \alpha_{r\theta} = \alpha_{\varphi\theta} = 0.$$

To calculate the scattered field we use the Rayleigh-Gans method for weakly inhomogeneous media.[3] At large distances the scattered wave is a diverging spherical wave with the amplitude

$$E_{\text{div}} = R(\theta, \varphi) k^2 r^{-1} e^{-ikr+ikz} E_0, \tag{2}$$

where $R(\theta, \varphi)$ is the scattering amplitude function, and k is the wave vector of an incident plane wave with amplitude E. It can be shown that when light is scattered by the spherically anisotropic inhomogeneities that we are considering $R(\theta, \varphi)$ will be given by

$$R(\theta, \varphi) = \int_V (\alpha_{rr} \cos^2\theta + \alpha_{\theta\theta} \sin^2\theta) e^{i\delta} dV, \tag{3}$$

where $e^{i\delta}$ is a phase factor that takes into account the interference between waves scattered by different elements of the optical inhomogeneity.

In this way our problem regarding nonlinear light scattering is reduced to the obtaining of the polarizability tensor components α_{rr} and $\alpha_{\theta\theta}$, which in our approximation (a weakly inhomogeneous medium) are expressed in terms of the refractive index variation as follows:

$$\alpha_{rr} = \alpha_0 + \Delta n_{rr} / 2\pi, \quad \alpha_{\theta\theta} = \alpha_0 + \Delta n_{\theta\theta} / 2\pi. \tag{4}$$

We calculate Δn_{rr} and $\Delta n_{\theta\theta}$ by means of (1), where the temperature and deformation fields ΔT and u_{lm} induced by the electromagnetic field are derived from the equations[4]

$$\frac{\partial T}{\partial t} = \chi \nabla^2 T + \frac{\chi}{k_T} P(r, t), \tag{5}$$

$$\partial^2 \mathbf{u} / \partial t^2 = c_l^2 \nabla \operatorname{div} \mathbf{u} + \mathbf{f}(r, t) \tag{6}$$

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subject to the following initial and boundary conditions:

when $t=0$: $T=T_0$, $\mathbf{u}=0$, $\partial\mathbf{u}/\partial t=0$;
when $r\to\infty$: $T\to T_0$, $\partial T/\partial r\to 0$, $\mathbf{u}\to 0$, $\partial\mathbf{u}/\partial r\to 0$;
when $r=0$: T, and $\mathbf{u}$ — and are finite

Here $P(r, t) = \omega_0 n'' E_0^2/4\pi$ is the energy absorbed by a volume unit in unit time; n'', the imaginary part of the refractive index of the medium, is an arbitrary function of the radius and field; k_T and χ are the thermal conductivity and thermal diffusivity, respectively; $\mathbf{u}$ is the displacement vector; c_l is the velocity of longitudinal elastic waves;

$$\mathbf{f}(r,t) = -\nabla[AT + (E_0^2/8\pi\rho)(\varepsilon + a)]$$

is the density of body forces induced by electrostriction and the temperature gradient; ρ is the density; $A = c_l^2(1+\sigma)\alpha/3(1-\sigma)$; σ, α, and a are the Poisson, thermal expansion, and electrostriction coefficients, respectively. In liquids and gases we have $a = -\rho\partial\epsilon/\partial\rho$. We note that in (6) the damping of elastic waves has been neglected.

The deformation tensor components are expressed in terms of the displacement vector $\mathbf{u}(r) = \mathbf{u}r/r$:

$$u_{rr} = \partial u/\partial r, \quad u_{\theta\theta} = u_{\varphi\varphi} = u/r. \tag{7}$$

In deriving the temperature and deformation distributions we assume that the medium is homogeneous and isotropic with respect to its thermal and elastic properties. The solutions of (5) and (6) will be sought by using Fourier transforms with respect to the coordinates. The scattered field will thus be obtained in terms of Fourier transforms of the temperature and deformation fields without an actual calculation of T and $\mathbf{u}$. For T we have

$$T(r,t) = \frac{2}{\pi}\int_0^\infty \frac{\sin sr}{r} F_1(s,t)\,ds, \tag{8}$$

where

$$F_1(s,t) = \frac{\chi}{k_T}\int_0^t dt' \exp[-\chi s^2(t-t')]\int_0^\infty r'\sin sr'\, P(r',t')\,dr'. \tag{9}$$

In solving (6) the displacement vector $\mathbf{u}$ will be sought in the form $\mathbf{u} = \nabla\psi$. For Ψ we then obtain the scalar equation

$$\partial^2\Psi/\partial t^2 = c_l^2\nabla^2\Psi - [AT + (E_0^2/8\pi\rho)(\varepsilon - 1 + a)] \tag{10}$$

and for the components of the deformation tensor we have

$$u_{rr} = \partial^2\Psi/\partial r^2, \quad u_{\theta\theta} = u_{\varphi\varphi} = r^{-1}\partial\Psi/\partial r. \tag{11}$$

Solving (10) by the Fourier transform method, we obtain

$$\Psi(r,t) = -\frac{2}{\pi}\int_0^\infty ds \frac{\sin sr}{r}\int_0^t \frac{\sin sc_l(t-t')}{sc_l}\Phi(s,t')\,dt', \tag{12}$$

where

$$\Phi(s,t') = AF_1(s,t') + \frac{E_0^2}{8\pi\rho}F_2(s,t'),$$
$$F_2(s,t') = \int_0^\infty r'\sin sr'\,(\varepsilon - 1 + a)\,dr'. \tag{13}$$

We can now see that $u_{rr} \neq u_{\theta\theta}$, from which spherical anisotropy results.

For the components of the polarizability tensor we obtain

$$\alpha_{rr} = \alpha_0 + \frac{1}{2\pi}\left\{\left(\frac{\partial n}{\partial T}\right)_{E_i,u_{ik}}\Delta T + n_2E_0^2 - \frac{1}{2}\left[p_{11}\frac{\partial u}{\partial r} + p_{12}\frac{2u}{r}\right]\right\} \tag{14}$$

$$\alpha_{\theta\theta} = \alpha_0 + \frac{1}{2\pi}\left\{\left(\frac{\partial n}{\partial T}\right)_{E_i u_{ik}}\Delta T + n_2E_0^2 - \frac{1}{2}\left[p_{12}\frac{\partial u}{\partial r} + (p_{11}+p_{12})\frac{u}{r}\right]\right\}, \tag{15}$$

where for the photoelastic constants we have used the conventional notation ($p_{11} \equiv p_{1111}$, $p_{12} \equiv p_{1122}$) and $n_2 \equiv \beta_{1111}$.[5]

Substituting (14) and (15) into (3), and utilizing (8) and (12), for the scattering amplitude function we obtain

$$R(\theta,\varphi) = R^L(\theta,\varphi) + R^{NL}(\theta,\varphi), \tag{16}$$

where

$$R^L(\theta,\varphi) = \int_0^\infty 4\pi r^2\alpha_0(r)\frac{\sin qr}{qr}\,dr$$

is the amplitude function of ordinary (linear) scattering, $q = 2k\sin(\theta/2)$, and θ is the scattering angle. The field of the scattered wave includes a nonlinear term that is proportional to the incident radiation intensity, because the optical properties of the medium are changed by intense light:

$$R^{NL}(\theta,\varphi) = \frac{2}{q}\left(\frac{\partial n}{\partial T}\right)_{E_i,u_{ik}}F_1(q,t) + \frac{2}{q}E_0^2\int_0^\infty n_2(r')r'\sin qr'\,dr'$$
$$-\frac{p_{11}+2p_{12}}{3}\int_0^t \sin qc_l(t-t')\,\Phi(q,t')\,dt'. \tag{17}$$

The intensity of the scattered light is calculated from

$$I_{\text{scat}} = \frac{k^4}{r^2}|R^L + R^{NL}|^2 I_0. \tag{18}$$

The first term of the scattered field in (17) allows for variation of the refractive index due to thermal heating; the second term allows for variation due to the Kerr effect; and the third term allows for variation due to the propagation, in the medium, of elastic deformations whose sources are the temperature gradient and ponderomotive forces.[6] In deriving (17) it was assumed that the light pulse varies very slowly throughout its duration. In other words, the characteristic time of light intensity variation was taken to be much longer than the characteristic times for the propagation of heat and elastic deformations, which are $1/\chi q^2 \sim 10^{-9}$ sec and $1/qc_l \sim 10^{-10}$ sec, respectively; monochromatic incident radiation is assumed here. On the other hand, it must be remembered that (17) is valid for times $t < \tau_l$, where τ_l is the damping time of elastic waves having the frequency $\Omega = qc_l$.

In the calculation of (17) it was taken into consideration that

$$\frac{2}{\pi}\int_0^\infty \sin qr\sin sr\,dr = \delta(q-s), \quad q>0, \ s>0.$$

This has a profound physical meaning. In solving (5) and (6) by means of Fourier transforms we actually expanded the temperature and deformation fields in spherical waves with the "wave vector" s. The occurrence of the δ function signifies that the scattered field receives a contribution at a given angle θ only from spherical waves with the "wave vector" $s = q \equiv 2k\sin(\theta/2)$; contributions from the other spherical waves are cancelled by interference.

We shall now consider the spectrum of scattered radiation. The incident radiation of frequency ω_0 will be assumed to have the form of a square pulse; in this case we have

$$F_1(q,t)=f(q)(1-e^{-\chi q^2 t}),$$
$$\Phi(q,t)=Af(q)(1-e^{-\chi q^2 t})+\mathcal{F}(q),\quad \mathcal{F}(q)=\frac{E_0^2}{8\pi\rho}F_2(q) \tag{19}$$

and for the spectrum of the scattered field we obtain

$$E_{\text{scat}}(\omega)=\frac{k^2e^{-ikr}}{r}E_0\Big\{\pi Q_1(q)\delta(\omega_p)+\frac{Q_2(q)}{\chi q^2+i\omega_p} \tag{20}$$
$$+\pi Q_3(q)\delta(qc_l-\omega_p)+\pi Q_4(q)\delta(qc_l+\omega_p)\Big\},$$

where $\omega_p = \omega - \omega_0$, and the Q_i terms are defined by

$$Q_1(q)=R^L(\theta,\varphi)+\frac{2}{q}\int_0^\infty r'n_2(r')\sin qr'\,dr'+\frac{2}{q}\left(\frac{\partial n}{\partial T}\right)_{E_i,u_{ik}}f(q)$$
$$-\frac{p_{11}+2p_{12}}{3qc_l^2}[Af(q)+\mathcal{F}(q)],$$
$$Q_2(q)=\frac{2}{q}f(q)\left[\left(\frac{\partial n}{\partial T}\right)_{E_i,u_{ik}}-\frac{A(p_{11}+2p_{12})}{6c_l^2}\frac{1}{1+(\chi q/c_l)^2}\right],$$
$$Q_3(q)=\frac{p_{11}+2p_{12}}{6qc_l^2}-\frac{\chi q^2[Af(q)+\mathcal{F}(q)]+iqc_l\mathcal{F}(q)}{\chi q^2+iqc_l}$$
$$Q_4(q)=Q_3^*(q)$$

(an asterisk denotes the complex conjugate).

Therefore the spectrum of the scattered light consists of an unshifted component that is broadened by an amount of the order χq^2 because of thermal heating, and shifted components where the shift qc_l depends on the direction of scattering and decreases at smaller scattering angles. The ratio of the shifted and unshifted amplitudes is of the order $(\chi q/c_l)^2$, which in solids equals $\sim 10^{-3}$–10^{-4}.

2. PARTICULAR THEORETICAL CASES OF NONLINEAR LIGHT SCATTERING

A. Light scattering by small particles. We consider the case of a small "Gaussian" particle of diameter $a_0 \ll \lambda$ and volume polarizability $\alpha_0(r) = \alpha_0 \exp(-r^2/a_0^2)$, with $n''(r) = n_0''\exp(-r^2/a_0^2)$ as the imaginary part of its refractive index. This model represents a more adequate description of optical inhomogeneities in real media than the model of a "step-like" particle with a sharp change of the refractive index.

In this case, for a square light pulse we have

$$F_1(q,t)=\frac{\sqrt{\pi}a_0^3}{4k_T q}P_0(1-e^{-\chi q^2 t}), \tag{21}$$

where $P_0 = \omega_0 n_0'' E_0^2/4\pi$ is the absorbed energy, and from (17) we obtain the nonlinear scattering amplitude function

$$R^{NL}(\theta,\varphi)=\sqrt{\pi}a_0^3\frac{E_0^2}{8\pi}\Big\{4\pi n_{20}+\frac{\omega_0 n_0''}{k_T q^2}\Big[\left(\frac{\partial n}{\partial T}\right)_{E_i,u_{ik}}$$
$$-\frac{A(p_{11}+2p_{12})}{6c_l^2}\Big](1-e^{-\chi q^2 t})+\frac{\chi q}{c_l}\frac{\omega_0 n_0''}{k_T q^2}\frac{A(p_{11}+2p_{12})}{6c_l^2}\sin qc_l t$$
$$-\frac{\varepsilon_0}{4q\rho}\frac{p_{11}+2p_{12}}{3c_l^2}(1-\cos qc_l t)\Big\}. \tag{22}$$

Here we have the electrostriction coefficient $\epsilon_0 = (\epsilon - 1 + a)\exp(r^2/a_0^2)$.

B. Stationary Scattering by Nonabsorbing Particles. When the time required for a change in the intensity of the incident radiation exceeds the characteristic damping time τ_l of elastic waves in the medium, light scattering will be stationary (the shape of the scattered light pulse will coincide with that of the incident pulse). Then from (3) and (6), neglecting the second time derivative, for the Kerr and electrostriction mechanisms we obtain the scattering amplitude function

$$R^{NL}(\theta,\varphi)=\frac{2}{q}E_0^2\int_0 n_2(r')r'\sin qr'\,dr'-\frac{p_{11}+2p_{12}}{3qc_l^2}\frac{E_0^2}{8\pi\rho}F_2(q). \tag{23}$$

For small particles this equation becomes the familiar result given in[2].

Our theory can account for nonlinear light scattering in media where multiple scattering may be neglected (media containing a low density of distributed scattering inhomogeneities).

[1] Yu. K. Danileĭko, A. A. Manenkov, V. S. Nechitaĭlo, and V. Ya. Khaimov-Mal'kov, Zh. Eksp. Teor. Fiz. 58, 1083 (1970) [Sov. Phys.-JETP 31, 588 (1970)].

[2] A. A. Manenkov, Dokl. Akad. Nauk SSSR 190, 1315 (1970) [Sov. Phys.-Dokl. 15, 155 (1970)].

[3] H. C. Van der Hulst, Light Scattering by Small Particles, John Wiley and Sons, New York, 1957, (Russ. transl., IIL, 1961).

[4] L. D. Landau and E. M. Lifshitz, Teoriya uprugosti (Theory of Elasticity), Nauka, 1965 (Engl. Transl., Addison-Wesley, Cambridge, 1959).

[5] J. F. Nye, Physical Properties of Crystals, Oxford Univ. Press, New York, 1957 (Russ. transl., IIL, 1960).

[6] L. D. Landau and E. M. Lifshitz, Elektrodinamika sploshnykh sred (Electrodynamics of Continuous Media), Gostekhizdat, 1959 (Engl. transl., Addison-Wesley, Cambridge, 1960).

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