Rayleigh superscattering of electromagnetic waves at an interface between vacuum and a superradiating medium

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An integral-equation method is used to examine the processes of reflection and refraction of a weak probe wave at the interface between two media, one of which contains a subsystem which is in a superradiating state. The electric polarizability is calculated for an atom in the medium which is acted on simultaneously by the field of the probe wave and that of the coherent oscillations, leading to a shift of the resonance levels of the atom. Formulas are found for the reflection and transmission coefficients of the interface between vacuum and a superradiating medium, and also the laws of reflection and refraction, which differ from the Fresnel laws.

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1. The propagation of powerful nanosecond and picosecond resonant laser pulses through a medium cause it to go into a superradiating state,[1] which is characterized by specific features of the kinetics of its deexcitation by emission of electromagnetic radiation and of the intensity of the radiation and its angular distribution, as affected by the shape of the emitting volume and the direction of the exciting pulses. Passage of an optical medium into a superradiating state is also possible as the result of spontaneous emission, under conditions in which the atoms are in an excited state (photon avalanche, see Ref. 2). A medium in which a particular superradiative effect (self-induced transparency, light induction and echo, photo avalanche, optical nutation, stimulated light induction) is produced decidedly alters the character of the Raman scattering (RS) of weak (probe) nonresonance radiation in the medium.[3,4] The probability of Raman scattering in a medium which is in a Dicke pure state[1] have been calculated by Walls[5] and by Makhviladze and Shelepin,[6] and those for a medium in a mixed state have been calculated by Nagibarov and the writer.[7] In recent years the idea of probing the coherent oscillations in a medium by means of the RS of weak radiation has been successfully developed in the method of stationary and nonstationary active spectroscopy of Raman scattering (ASRS).[8]

In the present study the coherent oscillations in the medium were excited by powerful resonance radiation with frequency ω_0, and the necessary conditions were satisfied for the occurrence of the appropriate superradiative processes. These processes were sounded with a weak probe wave of frequency ω which did not coincide with any of the frequencies in the spectrum of the atoms. The intensity of the probe wave was such that it did not excite nonlinear effects, but its scattering could be observed during the time of occurrence of the superradiative process. The frequency of the scattered radiation was the same as the frequency ω of the probe wave, i.e., the scattering process did not change the state of the medium (Rayleigh scattering component). For definiteness we shall consider a situation in which two resonant laser pulses with frequency ω_0 excite in the optical medium a light echo with its maximum intensity at the time 2τ, where τ is the time interval between the activation of the exciting laser sources.

The scattering of the probe wave is observed during the entire time of the formation of the light echo and of its decay. The process of the scattering of the probe wave has the characteristic features of a superradiative process and can be called Rayleigh superscattering. We shall here take into account the boundary effects at the vacuum-superradiating medium interface. We shall also indicate the possibility of using this method to study the behavior of the dielectric constant of a superradiating medium in the region of its absorption bands, which obviously will be different from the dielectric constant in the absence of the exciting pulses.[9]

2. Let us calculate the electronic polarizability of an atom in the optical medium, subjected to the simultaneous action of powerful resonance radiation with frequency ω_0 and of the probe wave with frequency ω. Let φ_1, φ_2 be states of the atom with frequencies ω_1, ω_2 such that $\omega_1-\omega_2=\omega_0$. The resonance radiation leads to mixing of the states φ_1 and φ_2, and so we write the wave function of the atom in the field of the two waves in the following way:

$$\psi(t)=\sum_{m=1}^{2}a_m\left\{\varphi_m+\sum_k(a_{mk}e^{i\omega t}+b_{mk}e^{-i\omega t})\varphi_k\right\}e^{-i\omega t},\qquad(1)$$

where $k\neq m$ are indices of the remaining states of the atom. The coefficients a_m depend on the time and on the parameters of the resonance action on the system (the electric field strength of the exciting radiation, the duration of its action, the coupling of the atom with the dissipative subsystem, etc.). The coefficients a_{mk} and b_{mk} can be found if we substitute Eq. (1) in the Schrödinger wave equation for the atom in the external field and obtain the terms of first order in the amplitude E_0 of the probe field. The mean value of the electric dipole moment of the atom in the state (1) takes the form

$$\langle d_\beta\rangle=\sum_{m,m'=1}^{2}\rho_{mm'}d^{\beta}_{mm'}-\frac{1}{2\hbar}\sum_{m,m'}^{2}\rho_{mm'}E_{0\alpha}(\tilde{\alpha}_{\alpha\beta})_{mm'},\qquad(2)$$

where summation over all repeated indices is under-

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stood; $d_{mm'}$ are the matrix elements of the electric dipole moment of the atom for the wave functions φ_1 and φ_2; $\rho_{mm'}$ is the second-order density matrix which is constructed with the coefficients a_m and describes the behavior of the resonance subsystem of the atom in the field of the powerful wave. The tensor $(\overline{\alpha}_{\alpha\beta})_{mm'}$ is symmetric in the indices α and β and can be put in diagonal form by a suitable choice of the coordinate axes. Far from resonance, if the approximation equation

$$\omega_{m'k}^2-\omega^2\approx\omega_{mk}^2-\omega^2$$

is satisfied, we can separate the time factor in the components of the tensor $(\overline{\alpha}_{\alpha\beta})_{mm'}$. Then we get the following formula for the part of the quantity (2) that depends on the field of the probe wave:

$$\langle d_\beta\rangle=-E_{0\beta}\alpha_{\beta\beta}\cos\omega t,$$

$$\alpha_{\beta\beta}=\frac{1}{\hbar}\sum_{m,m'}\rho_{mm'}\sum_k\frac{d_{mk}^\beta d_{km'}^\beta(\omega_{mk}+\omega_{m'k})}{\omega_{mk}^2-\omega^2}. \tag{3}$$

To calculate the quantity (3) it is necessary to know the density matrix $\rho_{mm'}$. We now consider, as we have proposed to do, the case of the action of two powerful laser pulses of durations Δt_1 and Δt_2 on the system of atoms, with a time interval τ between the times at which they are turned on. It is assumed that

$$\Delta t_1,\ \Delta t_2\ll T_1,\ T_2,\ T_2^*,\quad T_2^*\leqslant\tau\ll T_1,\ T_2, \tag{4}$$

where T_1 is the longitudinal relaxation time of the system of atoms, and T_2, T_2^* are the respective times of irreversible and reversible transverse relaxation.[10]

At a time t after the turning off of the second exciting pulse,

$$\rho(t)=\mathscr{L}(t)\rho_0\mathscr{L}^{-1}(t),\quad \mathscr{L}(t)=\exp[i\hbar^{-1}\mathscr{H}_0t]$$
$$\times\exp[i\hbar^{-1}(\mathscr{H}_1^{(2)}+\mathscr{H}_0)\Delta t_2]\exp[i\hbar^{-1}\mathscr{H}_0\tau]\exp[i\hbar^{-1}(\mathscr{H}_1^{(1)}+\mathscr{H}_0)\Delta t_1], \tag{5}$$

where ρ_0 is the equilibrium density matrix of the atom; $\mathcal{H}_0$ is the Hamiltonian operator of the atom in the absence of alternating external fields; $\mathscr{H}_1^{(1)}, \mathscr{H}_2^{(2)}$ are the Hamiltonians for the interaction of the atom with the fields of the first and second exciting pulses. We consider the exponential operators in Eq. (5) as functions of the matrices, and by using the method for calculating such operators,[10] we get the density matrix $\rho(t)$ in general form. We separate in it the terms corresponding to the light-echo signal with maximum intensity at the time 2τ, which are of the form

$$\overset{\text{echo}}{\rho_{12}}(t)=(\overset{\text{echo}}{\rho_{21}}(t))^*=-{}^1/{}_2 i\sin^2\theta_2\sin 2\theta_1(\rho_{11}^{(0)}$$
$$-\rho_{22}^{(0)})\exp[-i(\mathbf{k}_1-2\mathbf{k}_2)\mathbf{r}]\exp[i\omega_0(t-2\tau)],$$
$$\theta_1=2^{1/2}\hbar^{-1}d_0E_1\Delta t_1,\quad \theta_2=2^{1/2}\hbar^{-1}d_0E_2\Delta t_2,\quad d_0=|d_{12}|, \tag{6}$$

where $\mathbf{k}_1$ and $\mathbf{k}_2$ are the wave vectors of the first and second exciting pulses, $\mathbf{r}$ is the radius vector of the position of the atom in the medium, and E_1 and E_2 are the amplitudes of the electric fields of the exciting pulses. In the case of isotropic atoms we have

$$\overset{\text{echo}}{\alpha_{xx}}=\overset{\text{echo}}{\alpha_{yy}}=\overset{\text{echo}}{\alpha_{zz}}=\alpha_{\text{echo}}=\alpha_{\text{echo}}^{(0)}\exp[-i(\mathbf{k}_1-2\mathbf{k}_2)\mathbf{r}+i\omega_0(t-2\tau)]+\text{c.c.},$$
$$\alpha_{\text{echo}}^{(0)}=-\frac{i}{2\hbar}\sum_k\frac{d_{1k}^x d_{k2}^x(\omega_{1k}+\omega_{2k})}{\omega_{1k}^2-\omega^2}(\rho_{11}^{(0)}-\rho_{22}^{(0)})\sin^2\theta_2\sin 2\theta_1. \tag{7}$$

3. The electric field intensity of the probe wave obeys the integro-differential equation[11]

$$\mathbf{E}(\mathbf{r},t)=\mathbf{E}_i(\mathbf{r},t)+\int\operatorname{rot}\operatorname{rot}\mathbf{D}\left(\mathbf{E}\left(\mathbf{r}',t-\frac{R}{c}\right)\right)\frac{dV'}{R}, \tag{8}$$

where $\mathbf{E}_i$ is the intensity of the external field, $R=|\mathbf{r}-\mathbf{r}'|$, and $\mathbf{D}$ is the electric dipole moment per unit volume as a function of the field $\mathbf{E}$. Since the field of the probe wave is weak, we have

$$\mathbf{D}=\alpha_{\text{echo}}(\mathbf{k}_1,\ \mathbf{k}_2,\ t)\mathbf{E}N, \tag{9}$$

where N is the number of atoms per unit volume.

Noting the expression (7) for the polarizability of the atom, we write the condition for phase synchronism

$$\mathbf{k}_{\text{RSS}}=\mathbf{k}_L-\mathbf{k}_{\text{coh}},\quad \mathbf{k}_{\text{coh}}=2\mathbf{k}_2-\mathbf{k}_1, \tag{10}$$

where $\mathbf{k}_L$ is the wave vector of the probe wave in the medium, and $\mathbf{k}_{\text{RSS}}$ is the wave vector of the scattered wave. As can be seen from Eq. (10), the coherent oscillations with the wave vector $\mathbf{k}_{\text{coh}}$ can greatly alter the direction of the Rayleigh superscattering, and consequently the directions of the reflected and refracted waves at the interface vacuum-superradiating medium. Owing to this we consider the following types of processes at the interface (Fig. 1).

Case a. Suppose a plane wave with wave vector $\mathbf{k}_i$ and frequency ω is incident on the vacuum–superradiating medium interface. The process of reflection and refraction at the interface produces two plane waves with the respective wave vectors $\mathbf{k}_r$ and $\mathbf{k}_t=\mathbf{k}_L-\mathbf{k}_{\text{coh}}$, with the direction of the wave $\mathbf{k}_r$ connected with observed direction of the refracted wave. The wave vector $\mathbf{k}_L$ coincides with the direction of the refracted probe wave in the equilibrium medium in the absence of the exciting pulses. The wave of polarization in the medium is of the form

$$\mathbf{D}=[n^2(\omega)-1](\omega/c)^2\mathbf{Q}(\mathbf{r},t)e^{-i\omega t},\quad \mathbf{Q}=\mathbf{Q}_0\exp(i(\mathbf{k}_L-\mathbf{k}_{\text{coh}})\mathbf{r}), \tag{11}$$

where $n(\omega)$ is the index of refraction of the superradiating medium at the frequency ω.

The solution of the equation (8) reduces to the calculation of the integral

$$\mathbf{J}_t=\int_\Sigma\left(\mathbf{Q}\frac{\partial G}{\partial\nu'}-G\frac{\partial\mathbf{Q}}{\partial\nu'}\right)dS',\quad k=\omega/c, \tag{12}$$

where $\partial/\partial\nu'$ denotes differentiation along the outward normal to the interface Σ, and $G=e^{ikR}/R$ (see Ref. 11). The components of the unit vector $\mathbf{s}_i$ of the incident probe wave are

$$s_{yi}=-\sin\theta_i,\quad s_{xi}=0,\quad s_{zi}=-\cos\theta_i, \tag{13}$$

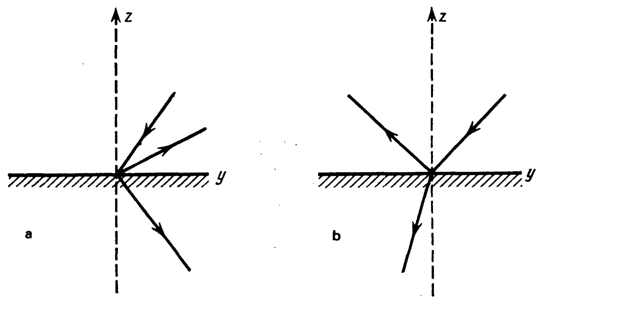

FIG. 1.

where θ_i is the angle of incidence and yz is the plane of incidence.

We shall suppose that the plane of incidence coincides with the direction of propagation of the coherent oscillations. Let the resultant vector $\mathbf{k}_t$ be directed into the medium along $\mathbf{s}_t$. Then we get for the components of $\mathbf{s}_t$ the following equations

$$s_{yt}=\sin\theta_t, \quad s_{xt}=0, \quad s_{zt}=-\cos\theta_t, \tag{14}$$

where θ_t is the angle of refraction of the probe wave in the superradiating medium. Using the principle of stationary phase,[11] which holds for distances r much larger than the wavelength, we get

$$\mathbf{J}_t=-2\pi \mathbf{Q}_0(t)\frac{\sin(\varphi+\theta_t)}{\cos\varphi\sin\theta_t}\exp\left[\frac{ikr(1+\sin^2\varphi)}{(1-\sin^2\varphi)^{1/2}}\right], \tag{15}$$

where the angle φ is determined from the equation $n\sin\theta_t=\sin\varphi$.

From the extinction theorem and Eq. (15) follows the refraction law connecting the angle of incidence θ_i with the angle of refraction θ_t:

$$\cos\theta_i=\frac{1+n^2\sin^2\theta_t}{(1-n^2\sin^2\theta_t)^{1/2}} \quad \text{or} \quad \mathbf{s}_i=\frac{1+\sin^2\varphi}{\cos^2\varphi}\mathbf{s}, \tag{16}$$

where $\mathbf{s}$ is a unit vector with the components $s_y=-\sin\varphi$, $s_x=0$, $s_z=-\cos\varphi$. From Eq. (16) we get

$$\sin\theta_t=iq_t=i\left[\frac{1}{2n^2}(2+\cos^2\theta_i\pm((2+\cos^2\theta_i)^2-4\sin^2\theta_i)^{1/2})\right]^{1/2}. \tag{17}$$

Let $q_t=\sinh A_t$; then $\theta_t=iA_t$, i.e., the angle of refraction is imaginary. The concept of complex angles is sufficiently fully analyzed in the optics of laminar media,[12] and in the general case the wave vector $\mathbf{k}_t$ can put in the following form:

$$\mathbf{k}_t=(\omega/c)(n-i\varkappa)(\mathbf{a}_t+i\mathbf{b}_t)=(\omega/c)(n-i\varkappa)\mathbf{s}_t, \tag{18}$$

where $\mathbf{s}_t$ is a complex vector with unit square ($\mathbf{a}_t^2-\mathbf{b}_t^2=1$, $\mathbf{a}_t\cdot\mathbf{b}_t=0$) which allows for the fact that the direction $\mathbf{N}_t$ of the propagation of the wave (the planes of equal phase) does not always coincide with the direction $\mathbf{K}_t$ in which it is damped (plane of equal amplitude); $\varkappa$ is the index of extinction of the superradiating medium at the frequency ω. The components of the vectors $\mathbf{N}_t$ and $\mathbf{K}_t$ can be found from the system of equations

$$a_{yt}+ib_{yt}=iq_t, \quad a_{yt}b_{yt}+a_{zt}b_{zt}=0, \quad a_{yt}^2+a_{zt}^2-b_{yt}^2-b_{zt}^2=1, \tag{19}$$

from which we get the following values of the components:

$$\begin{aligned} N_{yt}&=na_{yt}+\varkappa b_{yt}=\varkappa q_t, & N_{zt}&=na_{zt}+\varkappa b_{zt}=-n(1+q_t^2)^{1/2},\\ K_{yt}&=\varkappa a_{yt}-nb_{yt}=-nq_t, & K_{zt}&=\varkappa a_{zt}-nb_{zt}=-\varkappa(1+q_t^2)^{1/2}. \end{aligned} \tag{20}$$

Far from resonance we can set $\varkappa=0$ and find that for a definite mutual position of the directions of propagation of the probe and coherent oscillations in the medium, when the vectors $\mathbf{s}_i$ and $\mathbf{s}_t$ lie on both sides of the normal to the interface (Fig. 1a) a situation arises in which the direction of propagation of the refracted wave is perpendicular to the interface of the two media and directed into the medium for arbitrary angles of incidence (isotropic penetration of the interface vacuum-superradiating medium).

Let us find the formula connecting the amplitude of the incident probe wave, $\mathbf{E}_{0i}$, and that of the refracted wave, $\mathbf{T}_0=4\pi k^2\mathbf{Q}_0$, using the extinction theorem:

$$\mathbf{E}_{0i}=-\operatorname{rot}\operatorname{rot}\mathbf{J}_t. \tag{21}$$

Performing the differentiation of the expression (15), we get the following formula:

$$\mathbf{E}_{0i}=\frac{1}{2}\frac{\sin(\varphi+\theta_t)}{\cos\varphi\sin\theta_t}[\mathbf{T}_0-\mathbf{s}_i(\mathbf{s}_i\mathbf{T}_0)]. \tag{22}$$

Let us examine the case when the observations are located outside the medium ($z>0$). The calculations of the amplitude $\mathbf{E}_{0r}$ of the reflected wave are similar, except that in the relevant equations we must replace $z'=-r$ with $z'=r$. Then

$$\mathbf{E}_{0r}=-\frac{1}{2}\frac{\sin(\varphi-\theta_t)}{\cos\varphi\sin\theta_t}[\mathbf{T}_0-\mathbf{s}_r(\mathbf{s}_r\mathbf{T}_0)], \tag{23}$$

where the unit vector $\mathbf{s}_r$ determines the complex direction of the reflected wave with the components (θ_r is the angle of reflection)

$$s_{yr}=\sin\theta_r, \quad s_{xr}=0, \quad s_{zr}=-\cos\theta_r. \tag{24}$$

In an analogous way we find the vector components N_{ry}, N_{rz}, K_{ry}, K_{rz}:

$$N_{ry}=0, \quad N_{rz}=n(q_t^2+1)^{1/2}, \quad K_{ry}=-nq_t, \quad K_{rz}=0, \tag{25}$$

i.e., the real direction of the reflected wave is along the z axis for an arbitrary angle of incidence of the probe wave on the vacuum-superradiating medium interface (isotropic reflection).

Since the electronic polarizability (7) depends on the time, oscillating with the frequency of the resonant subsystem, the amplitude of the wave of electronic polarization in the medium, Eq. (11), must be averaged over the frequencies of the resonating atoms with allowance for the nonuniform broadening of the spectrum line, as is done in the theory of light echoes.[12] Then the amplitude of the passing wave must include a dependence on a time factor $\mathscr{A}(t)$, which takes its maximum value at $t=2\tau$ and falls to zero in a time interval larger than the time for obliteration of the phase memory. Furthermore Eq. (22), which gives the relation of the incident and passing waves, must be normalized to allow for the fact that as the phase memory fades out in the superradiating medium $\varphi\to\theta_i$ and the processes at the interface of the two media come to obey the Fresnel laws.

Using Eqs. (22) and (23), we find the transmitting power $\mathscr{T}$ and the reflecting power $\mathscr{R}$ of the vacuum-superradiating medium interface:

$$\mathscr{T}=\frac{n\cos\theta_t}{\cos\theta_i}\frac{|\mathbf{T}_0|^2}{|\mathbf{E}_{0i}|^2}, \quad \mathscr{R}=\frac{|\mathbf{E}_{0r}|^2}{|\mathbf{E}_{0i}|^2}. \tag{26}$$

Substituting the expressions (22) and (23) in Eq. (26), we find that $\mathscr{T}+\mathscr{R}=1$ only for $\varphi=\theta_i$. For all other angles, even when $\varkappa=0$, this equation is not satisfied; this indicates that there is additional absorption in the medium, owing to the fulfillment of the spatial conditions (10). For the components of $\mathbf{T}_0$ and $\mathbf{E}_0^r$ perpendicular to the plane of incidence we get the absorbing power in the following form:

$$A_\perp=\frac{4n\sin^2\theta_t\cos\theta_t\cos\varphi(\cos\varphi-\cos\theta_i)}{\cos\theta_i\sin^2(\varphi+\theta_t)}. \tag{27}$$

Case b. This case is trivial, since the formulas for the directions of the reflected and refracted waves, and also the corresponding amplitudes, are identical with the well known formulas of the optics of laminar media.[11,12]

DISCUSSIONS OF RESULTS AND CONCLUSIONS

The dielectric properties of a superradiating medium are determined by the direction of propagation of the exciting pulses, and also by the relaxation processes that promote the obliteration of the phase lattice in the medium. As can be seen from Eq. (7), the electronic polarizability of the atom depends on the wave vectors of the exciting pulses, the coordinates, and the time. Consequently, an optical medium composed of such atoms is inhomogeneous and anisotropic and changes in the course of time. Unlike the traditional method for describing wave-propagation processes in an optical medium with dielectric constant having a sinusoidal dependence on the coordinates,[13] we assume in the present work that the dielectric constant does not depend on the coordinates, but that at the interface of the two media the incident wave (of frequency ω and wave vector $\mathbf{k}_i$) is totally extinguished and is replaced by another wave with the frequency ω and the wave vector $\mathbf{k}_t$ given by Eq. (10). Furthermore, besides the usual locations of the wave vectors of the reflected, passing, and incident waves (Fig. 1b) we consider the case *a* (Fig. 1), which is possible if we note that the wave vector of the scattered wave is complex even for $\varkappa=0$ owing to the requirements of the synchronism conditions (10).

This process of Rayleigh superscattering can be used to determine numerical values of the optical constants of a superradiating medium at a resonance frequency ω_0 as functions of the angle of incidence of the probe wave on the interface between vacuum and the superradiating medium. In fact, using the relation (10) and the formulas (20), we find a system of equations for determining the indices of refraction, $n_0(\omega_0)$, and of absorption, $\varkappa_0(\omega_0)$, of the medium brought into the superradiating state by two exciting pulses directed in a definite way:

$$2n_0a_{2z}+2\varkappa_0b_{2z}-n_0a_{1z}-\varkappa_0b_{1z}-\frac{\omega}{\omega_0}n\left(1-\left(\frac{\sin\theta_i}{n}\right)^2\right)^{1/2}=\frac{\omega}{\omega_0}n(1+q_i^2)^{1/2},$$
$$2\varkappa_0a_{2z}-2n_0b_{2z}-\varkappa_0a_{1z}+n_0b_{1z}=0, \qquad (28)$$

where a_{1z}, a_{2z}, b_{1z}, b_{2z} are the components of the real vectors $\mathbf{a}_1$, $\mathbf{a}_2$, $\mathbf{b}_1$, $\mathbf{b}_2$ which determine the complex directions of the exciting pulses in the medium. Their values can be found easily from the boundary conditions that the tangential components of the electric field strengths must be continuous:

$$a_{z1}=-\frac{a_{y1}b_{y1}}{[-{}^1/_2p+({}^1/_4p^2+s)^{1/2}]^{1/2}},\qquad b_{z1}=[-{}^1/_2p+({}^1/_4p^2+s)^{1/2}]^{1/2},$$
$$p=1-\frac{(n_0^2-\varkappa_0^2)\sin^2\theta_{1i}}{(n_0^2+\varkappa_0^2)^2},\qquad s=\frac{n_0^2\varkappa_0^2\sin^4\theta_{1i}}{(n_0^2+\varkappa_0^2)^4} \qquad (29)$$
$$a_{y1}=\frac{n_0\sin\theta_{1i}}{n_0^2+\varkappa_0^2},\qquad b_{y1}=\frac{\varkappa_0\sin\theta_{1i}}{n_0^2+\varkappa_0^2},$$

where θ_{1i} is the angle of incidence of the first exciting pulse on the interface. The values of a_{2z} and b_{2z} are obtained from Eq. (29) by replacing θ_{1i} with θ_{2i}, where θ_{2i} is the angle of incidence of the second exciting pulse.

Starting from Eqs. (10) and (20), we also obtain a condition connecting the angles of incidence of the exciting pulses and of the probe wave:

$$2\sin\theta_{2i}-\sin\theta_{1i}=(\omega/\omega_0)\sin\theta_i. \qquad (30)$$

To calculate the quantity q_i in Eq. (28) one must choose the sign in Eq. (17). The proper choice can be found unambiguously by measuring the reflecting power of the vacuum-superradiating medium interface and comparing with Eq. (26).

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