Nonlinear reflection and refraction of ultrashort light pulses by surfaces of resonant media and phase "memory" effects

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A method of integrodifferential equations associated with the optical Bloch equations is used to consider the problem of nonlinear reflection (refraction) of a laser beam scanning the surface of a resonant medium excited by traveling and standing surface electromagnetic waves of resonance frequency. An allowance is made for the phase memory effects exhibited by surface atoms in response to pulses of fields resolved in space and time. A polarization wave on the surface is regarded as the result of a nonlinear superposition of a reflected wave and a surface wave. A generalized Lorentz-Lorenz formula is derived for a nonlinear complex refractive index of an optical medium in the region of an absorption band when this index depends nonlinearly on the field inside the medium. An allowance is made for possible transient effects in the response of the medium to pulses because of the finite time needed to establish polarizing fields. A theory of quenching is used to obtain generalized reflection and refraction laws for a scanning beam. These laws can be used to determine the direction of propagation of a reflected (refracted) wave. It is shown that, in general, a reflected wave is inhomogeneous. An analysis is made of the case of reversal of a scanning beam by an excited surface, characterized by phase conjugacy of the wavefront. A study is made of the spectrum of transient nonlinear surface polaritons as a function of the area of an exciting pulse and of the depth of penetration of these polaritons in a resonant optical medium.

INTRODUCTION

Transient coherent effects of the optical echo type[1] which are due to the so-called phase "memory" effect are beginning to play an important role in optical studies of resonant media. A new and promising branch of laser spectroscopy, which can be called optical echo spectroscopy,[2] has now been established. This spectroscopy has been used already to study spectral and relaxation characteristics of particles (atoms, molecules, ions) located inside a resonant medium. For practical reasons it is important to extend such studies to particles on the surface of a medium. However, this is not a trivial extension because the solution of the nonlinear optical problem requires an allowance for the boundary conditions and this, in turn, needs a review of a number of important concepts applicable to bulk media. This problem is important also because the discovery of a long-term optical memory in an LaF_3:Pr^{3+} crystal[3] has demonstrated that the theoretically predicted applications of phenomena of the optical echo type in dynamic holography and in construction of optical memory devices[2] have now reached the stage of practical realization and technical importance. Further progress may be facilitated by the recently developed method of scanning with a laser beam.[4–6] The present paper deals with the solutions of all these fundamental problems in echo spectroscopy of surfaces. Reversal of the wavefront has been realized in the optical echo experiments.[2] It has been shown in Refs. 7 and 8 that under certain conditions (for example, with the aid of a standing wave) it is possible to reverse the direction of optical echo signals. In the case of stimulated optical echo signals the reversal effect occurs under conditions when two waves traveling in opposite directions and forming a standing wave are separated in time. We shall consider in detail the case when the first exciting signal is in the form of a laser beam scanning the surface of a medium along a definite closed path. In the two-pulse excitation regime the second pulse is in the form of two counterpropagating pulsed surface light waves of the same resonance frequency as that of the scanning laser beam. The situation is explained by the diagram in Fig. 1. The reversed optical echo signal emitted by each part of the scanning path travels exactly opposite to the scanning beam. A beam splitter BS directs this signal to a fast-response photomultiplier. The most promising is the three-pulse excitation regime because the time interval between the second and third pulses in some crystals can be very considerable (record results have been reported for an LaF_3:Pr^{3+} crystal for which the stimulated optical echo signal due to the $^3H_4$–$^3P_0$ transition at the 4777 Å wavelength appeared after up to 30 min from the action of the first pair of the exciting pulses). In this regime the first exciting signal is once again the scanning light beam; the second exciting signal is a pulse of a surface light wave of the resonance frequency in the direction $\mathbf{k}_2$, and the "readout" or reconstructing third pulse (which, in principle, can be applied even after several minutes) is the reversed pulse of a surface light wave traveling in the $-\mathbf{k}_2$ direction. The signal of the reversed stimulated optical echo may be emitted by

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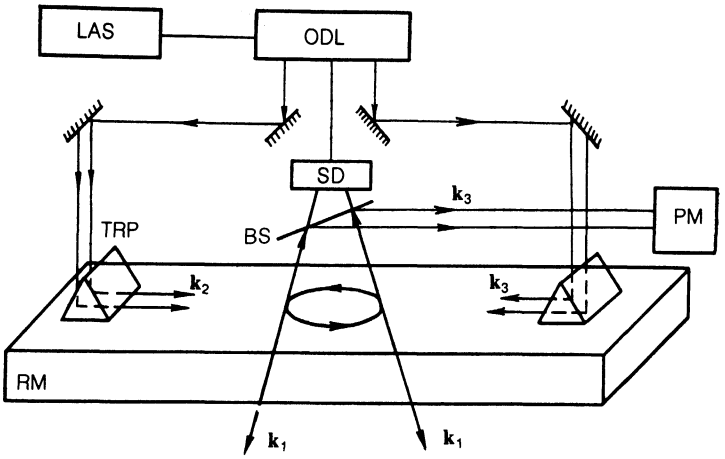

FIG. 1. Schematic diagram of the apparatus which can be used to excite the surface of a resonant optical medium (RM). LAS is a laser, ODL is an optical delay line, SD is a scanning device, BS is a beam-splitting plate, TRP is a total internal reflection prism, and PM is a fast-response photomultiplier.

active particles on the crystal surface in the direction $-\mathbf{k}_1$.

We shall now consider the scanning laser beam. In principle, its function can be performed by a cw laser and the coherent nature of the interaction with a resonant medium is ensured by the fact that at scanning velocities v_{sc} satisfying the inequality $D/v_{sc} < T_1$, T_2' (where D is the laser beam diameter, T_1 and T_2' are the longitudinal and transverse irreversible relaxation times), the duration of action of the beam decreases. Clearly, the beam path can be arbitrary and the storage and retrieval ("readout") by surface waves can be of the line-by-line type.

Similar experiments can be carried out not only on the surfaces of crystals, but also in thin films, which may be of interest in integrated optics.[9–17] However, solution of these problems depends on a detailed theoretical analysis of some of the fundamental topics in nonlinear optics of surfaces, which are dealt with in several sections below. We shall use the method of integral equations in a systematic study of various boundary-value problems in nonlinear optics. We shall consider specifically the reversal of a scanning beam by an interface between two media along which a surface electromagnetic wave pulse is traveling.

§1. SYSTEM OF INTEGRODIFFERENTIAL EQUATIONS DESCRIBING PROPAGATION OF OPTICAL WAVES IN A RESONANT MEDIUM

Interaction of a scanning laser beam with a resonant medium can be investigated using a two-dimensional system of the differential Maxwell and Bloch equations subject to certain initial and boundary conditions.[4–6] An allowance for the boundary conditions in solving such boundary-value problems (particularly in the case of the spatial dispersion effects) complicates greatly a theoretical analysis.[18] Therefore, in contrast to the usual method based on the "matching" of the Maxwell equations for a resonant medium and vacuum, we shall solve the boundary-value problems by integrodifferential equations for the propagation of light in an optical medium. This avoids the need to introduce explicitly the boundary conditions at refracting surfaces. The necessary integrodifferential equations suitable for the investigation of the propagation of light in resonant optical media were obtained in Refs. 19 and 20. We shall write them as follows:

$$\mathbf{E}(\mathbf{r},t)=\mathbf{E}_I(\mathbf{r},t)+\int \operatorname{rot}\operatorname{rot}\mathbf{D}(\mathbf{r}',t-R/c)\,dV'/R, \qquad (1)$$

where $\mathbf{E}(\mathbf{r},t)$ is the intensity of the electric field at an observation point $\mathbf{r}$; $\mathbf{E}_I$ is the intensity of the electric field of an external wave; $R=|\mathbf{r}-\mathbf{r}'|$; $\mathbf{r}'$ is the radius vector of an arbitrary point inside the medium; $\mathbf{D}$ is the induced electric dipole moment per unit volume at the point $\mathbf{r}'$ inside the medium, dependent in some way on the field $\mathbf{E}$. The differentiation in Eq. (1) is with respect to the coordinates of the observation point. Equation (1) is three-dimensional so that it can be used to study various optical resonances under conditions of three-dimensional scanning with a laser beam.

Equations (1) are obtained from general laws of quantum electrodynamics, and the polarizing field created by atoms in the radiation field is regarded as third-order effects involving virtual exchange of photons of all polarizations betweeen atoms and also emission (absorption) of a real photon.[19] Obviously, such effects include spontaneous emission of photons, because in this case the field $\mathbf{E}(\mathbf{r},t)$ should be regarded as a quantized field of spontaneous radiation. Consequently, the range of validity of the system (1) is very wide and these equations can be used for a theoretical study of a wide class of nonlinear optical phenomena, including the optical echo and the Dicke superradiance. Moreover, equations (1) can be used also in the case of conducting media. In this connection we must point out that in their book[21] Born and Wolf limit the range of validity of the system (1) to nonconducting media, which is unjustified. For example, formulas for polarizing fields created by moving charges (not necessarily atomic) at arbitrary observation points are obtained in Ref. 22 using retarded potentials. In this case the electric dipole moments in a medium are formed by charge clusters in elementary volumes which have linear dimensions much smaller than the distance to the observation points. In all other cases we must obviously modify the system (1), which together with the equations for the medium, forms a general system of equations for the study of the propagation of high-intensity light in homogeneous and inhomogeneous optical media in the region of an isolated absorption band. We shall determine the dependence of the dipole moment **d** of an isolated atom on the field **E** inside an insulator, using Bloch's equations. We shall therefore use the representation of an effective spin and express **d** in terms of the Pauli operators σ as follows[1]: $\mathbf{d} = \mathbf{d}_r\sigma_1 - \mathbf{d}_i\sigma_2$, where $\mathbf{d}_r$ and $\mathbf{d}_i$ are the vectors in the coordinate space. The equations of motion for local dipole moments and the differences between the populations of the resonance levels of an atom are derived, for example, in Ref. 1. We shall adopt the standard notation used in that book. In the case of the $\Delta m = 0$ transitions we have $d_i = 0$ (Ref. 1) and we note that $(2d_rE/\hbar) \equiv \varkappa_0$. Then, in a coordinate (reference) system rotating at a frequency ω, the equations of motion become

$$\dot{u}=-\Delta v, \quad \dot{v}=\Delta u+\varkappa_0 E_0 w, \quad \dot{w}=-\varkappa_0 E_0 v, \tag{2}$$

where the frequency detuning is $\Delta = \omega_0 - \omega$ and E_0 is the amplitude of the field **E** in the medium. We shall also introduce a dimensionless quantity

$$\theta(\mathbf{r}',t)=\varkappa_0\int_{-\infty}^{t} E_0(\mathbf{r}',t')\,dt',$$

which represents the effect of a light pulse at a point $\mathbf{r}'$ inside the investigated medium. It can be used to write down the solutions of Eq. (2) in accordance with Ref. 1. The solution can be used to find a polarization wave $\mathbf{D}(\mathbf{r}',t - R/c)$, which depends nonlinearly on the field **E** inside the medium. Introducing $\mathbf{D}_r = \mathbf{d}_r(N/V)$, where N/V is the concentration of atoms, we obtain

$$\mathbf{D}^{\pm}(\mathbf{r}',\ t-R/c,\ \Delta) = {}^1/_2\mathbf{D}_r\delta_{\pm}(\mathbf{r}',\ t,\ \Delta)\,e^{\pm i\mathbf{k}\mathbf{r}'}e^{\pm i\omega(t-R/c)} \equiv \mathbf{D}_0{}^{\pm}e^{\pm i\omega R/c}; \tag{3}$$

here, $\delta_{\pm} = u \pm iv$ and **k** is the wave vector of the polarization wave in the medium. Substituting Eq. (3) into Eq. (1), we obtain the field at an arbitrary observation point **r** at a moment t.

§2. REFRACTIVE INDEX OF A NONLINEAR OPTICAL MEDIUM IN THE REGION OF AN ABSORPTION BAND. QUENCHING THEOREM

We shall consider the processes of reflection of waves from an abrupt interface between two optical media, one of which is nonlinearly resonant. In this case an observation point is located outside the nonlinear medium so that the operation curl curl in Eq. (1) can be taken outside the integral. Then, the field at the observation point becomes

$$\mathbf{E}(\mathbf{r},t)=\mathbf{E}_I(\mathbf{r},t)+\mathrm{rot\ rot}\int\mathbf{D}(\mathbf{r}',t-R/c)\,dV'/R. \tag{4}$$

The second term in Eq. (4) represents a reflected wave $\mathbf{E}_R$.

We shall assume that the function (3) satisfies the Helmholtz wave equation $\nabla^2\mathbf{D}_0^{\pm} + \tilde{n}^2(\omega/c)^2\mathbf{D}_0^{\pm} = 0$, where $\tilde{n}$ is the complex refractive index of the optical medium in the region of an absorption band, and generally depends on the coordinates of the observation point and on time. Since the function $G_0^- = (1/R)e^{i\omega R/c}$ represents a spherical wave in vacuum, integration of the volume integral by the Green theorem gives

$$\int G_0{}^-(R)\mathbf{D}_0{}^+(\mathbf{r}')\,dV'=(c^2/\omega^2)(\tilde{n}^2-1)^{-1}\int_{\Sigma}\{\mathbf{D}_0{}^+(\partial G_0{}^-/\partial\nu') - G_0{}^-(\partial\mathbf{D}_0{}^+/\partial\nu')\}\,dS', \tag{5}$$

where Σ is the interface between the two media and the symbol $\partial/\partial\nu'$ represents differentiation along the outward normal to the interface Σ. Therefore, a calculation of the field above the interface Σ reduces to a calculation of the surface integral (5), where $\mathbf{D}_0^+$ is governed by the field E on the surface Σ. We shall denote the surface integral by $\mathbf{I}_{-+}$, so that instead of Eq. (4) we now obtain a different integral equation depending on the state of the surface Σ.

The integrodifferential equation obtained in this way contains an unknown quantity $\tilde{n}$ which governs the velocity of propagation of the field inside the medium. We can find this quantity by returning to the initial system (1). We shall assume that an observation point is located inside the medium so that the operation curl curl cannot be simply taken outside the integral. Assuming that $\mathbf{D}_0^+$ is an arbitrary function of the coordinates, we find that when the condition $\mathrm{div}\mathbf{D}_0^+ = 0$ is satisfied, instead of Eq. (1) we now have

$$\mathbf{E}(\mathbf{r},\ t)=\mathbf{E}_I(\mathbf{r},\ t)+\mathrm{rot\,rot}[(c^2/\omega^2)(\tilde{n}^2-1)^{-1}]\mathbf{I}_{-+} - {}^8/_3\pi\mathbf{D}_0{}^+(\mathbf{r},\ t)+4\pi\tilde{n}^2\mathbf{D}_0{}^+(\tilde{n}^2-1)^{-1}. \tag{6}$$

Equation (6) can be separated into two equations if we identify separately two groups of terms, each of which represents a wave traveling at its own velocity. The equation

$$\mathbf{E}_I(\mathbf{r},\ t)+(c^2/\omega^2)\,\mathrm{rot\,rot}\,\mathbf{I}_{-+}/(\tilde{n}^2-1)=0 \tag{7}$$

represents the quenching theorem which is satisfied by a nonlinear optical medium in the region of an absorption band. The equation

$$\mathbf{E}(\mathbf{r},\ t)=-{}^8/_3\pi\mathbf{D}_0{}^+(\mathbf{r},\ t)+4\pi\tilde{n}^2\mathbf{D}_0{}^+(\tilde{n}^2-1)^{-1} \tag{8}$$

allows us to calculate the complex refractive index of a nonlinear optical medium for an arbitrary dependence of $\mathbf{D}_0^+$ on **E**. If $\mathbf{E} = E\mathbf{e}$, we find that (**e** is a unit polarization vector of the field)

$$\tilde{n}^2=\varepsilon'+i\varepsilon''=[E_0+{}^8/_3\pi\tilde{\mathbf{D}}_0{}^+\mathbf{e}][E_0-{}^4/_3\pi\tilde{\mathbf{D}}_0{}^+\mathbf{e}]^{-1}, \tag{9}$$

where $\tilde{\mathbf{D}}_0^+$ is found by dropping the phase factor $\exp[-i(\omega t - \mathbf{k}\cdot\mathbf{r})]$ from the quantity $\mathbf{D}_0^+$. Equation (9) is obtained for observation points inside the medium. A modification of the lemma proposed in Ref. 21 can be used to calculate the complex refractive index at observation points located on the surface or close to it. Then, the refractive index depends on the depth of the observation point and, consequently, on the polarization of the medium at this depth. In linear optics the quantity $\mathbf{D}_0^+$ is proportional to the first power of $\mathbf{E}_0$ and Eq. (9) reduces to the Lorentz-Lorenz formula.[21,22] In general, the refractive index of Eq. (9) depends on the amplitude E_0 in a complex manner and this dependence is governed by the equations of motion (2). We shall substitute Eq. (3) into Eq. (9). We shall represent the complex refractive index in the form $n + i\varkappa$, where n is the real refractive index and $\varkappa$ is the extinction coefficient, which gives two equations for the determination of n and $\varkappa$ expressed in terms of u and v.

Equation (9) is a generalization of the Lorentz-Lorenz formula to nonlinear isotropic media in the region of an isolated resonance. In the case of strongly excited quantum dipoles in a medium the polarization $\mathbf{D}_0^+$ of the medium depends in a complex manner on the field amplitude $\mathbf{E}_0$, so that the values of n and $\varkappa$ depend on E_0, which is one of the manifestations of the nonlinearity of the medium. In the approximation of a constant field, the parameters n and $\varkappa$ are independent of the coordinates $\mathbf{r}$ of the observation point. When an allowance is made for the transmission effects, E_0 depends on $\mathbf{r}$ so that n and $\varkappa$ also include a dependence on $\mathbf{r}$, i.e., the optical medium becomes inhomogeneous and this is again a manifestation of its nonlinearity. It should be pointed out that the dependence of the complex refractive index of Eq. (9) on the coordinates of the observation point should also be different, for example, it may be governed by the surface geometry. Therefore, we can allow in Eq. (9) for a transition layer on the surface which may be of field or structure origin.

When ultrashort light pulses interact with a resonant medium, the problem of the origin of the refractive index is no longer trivial. This is due to the fact that the polarizing fields are then induced in a finite time τ_c, which can be longer or shorter than the pulse duration Δt. The time $\tau_c = W_c^{-1}$ is calculated in Ref. 23, where W_c is the probability of emission of a photon by an arbitrary atom in a medium interacting with its environment. Obviously, in the former case $(\tau_c > \Delta t)$ the investigated medium cannot respond in the available time to the propagation of a pulse, there is no reaction by the medium, and the refractive index should not differ from unity. In the latter case $(\Delta t \gg \tau_c)$ the refractive index of the medium can be calculated from Eq. (9). Depending on the method used to excite the resonance medium, we can have different values of the induced polarization and, consequently, different dependences of the refractive index on the field. When a resonant medium is excited by ultrashort light pulses of duration much less than all the relaxation times, the induced polarization of the medium is due to transient processes. After the necessary averaging of the polarization over the frequency detuning, we obtain a factor $\exp(-t/T_2^*)$ in the expression for the polarization of the medium and this factor indicates that the polarization decays rapidly because of inhomogeneous broadening of the atomic levels. Therefore, the optical properties of the medium change significantly during a characteristic phase memory time T_2^* of atoms and this transient behavior of an optical medium is also a manifestation of its nonlinearity. Figure 2 shows the dependences of the permittivity components ε' and ε'' on the field intensity. Clearly, an increase in the field amplitude alters greatly ε' and ε''. An analysis of Eq. (9) shows that pulsed illumination of a resonant medium from a high-intensity light source can be used to vary the optical properties of the medium within a wide range.

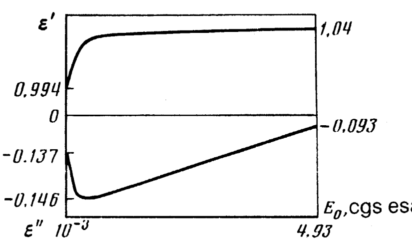

FIG. 2. Dependences of the permittivity of a nonlinear medium on the direction of an electric field of an ultrashort light pulse. The following numerical values of the physical parameters were used to plot these dependences: $d_r = 4.8\times10^{-21}$ cgs esu, $N/V = 1.6\times10^{19}$ cm^{-3}, $\Delta = 10^7$ sec^{-1}, $\tau = 35$ nsec. The characteristic time for inducing polarizing fields in the medium is $\tau_c = 2/T_2'(\varkappa_0 E_0')$, where E_0' is the amplitude of the polarizing field of the surrounding atoms.[20]

§3. REFLECTION AND REFRACTION OF A SCANNING LASER BEAM BY AN INTERFACE BETWEEN TWO MEDIA, ONE OF WHICH IS RESONANT

Let us assume that electromagnetic waves interact with an interface Σ between two media, as shown in Fig. 3:

$$s_I^x = -\sin\theta_I\cos\varphi_I, \quad s_I^y = \sin\theta_I\sin\varphi_I, \quad s_I^z = -\cos\theta_I, \quad (10)$$

where the angles θ_I and φ_I generally depend on time. We shall describe an incident wave by $\mathbf{E}_I(\mathbf{r},t) = \mathbf{E}_0^I\exp\{i[(\omega/c)(\mathbf{r}\mathbf{s}_I) - \omega t]\}$. In the case of three-dimensional scanning with a laser beam incident on an interface between two media, one of which is a nonlinear resonant medium, we must drop the usual concept of the plane of incidence. This is because the interaction of resonant laser radiation with a nonlinear medium may give rise to optical transient effects at the interface due to the phase memory of atoms. For example, it is shown in Ref. 10 that an ultrathin nonlinear layer of resonating atoms at an interface between two linear transparent media may give rise to nonlinear components of reflected and refracted waves. In contrast to the Fresnel components of reflection and refraction, the nonlinear components now result in afterglow of the interface between the two media for a period equal to the phase memory time. Therefore, in the case of three-dimensional scanning each direction of incidence of a scanning beam has its own history of reflection and refraction for the previous directions of incidence. This is the reason for introduction of two angles of incidence: θ_I and φ_I. Obviously, in the case of two-dimensional scanning this complication of the problem is unnecessary. The field at an arbitrary observation point above the surface can be found from Eq. (4) where we need to know the polarization $\mathbf{D}_0^+$ at the interface Σ, which is generally represented by Eq. (3) with a wave vector $\mathbf{k}$, the meaning of which will be defined later. Substituting $\mathbf{D}_0^+$ into the surface integral I_{-+}, we find that differentiation yields

$$\frac{\partial G_0^-}{\partial \nu'} = \frac{\omega i}{c}\frac{\partial R}{\partial \nu'}G_0^-\left(1+\frac{ic}{\omega R}\right),$$

$$\frac{\partial \mathbf{D}_0^+}{\partial \nu'} = \frac{\mathbf{D}_r}{2}\left(\frac{\partial \delta_+}{\partial \mathbf{r}'}\frac{\partial \mathbf{r}'}{\partial \nu'} + i\delta_+\mathbf{k}\frac{\partial \mathbf{r}'}{\partial \nu'}\right)\exp(i\mathbf{k}\mathbf{r}' - i\omega t). \quad (11)$$

Introducing a spectral response function F of Ref. 1 and the area under a pulse θ, we find that

$$\frac{\partial \delta_+}{\partial \mathbf{r}'}\frac{\partial \mathbf{r}'}{\partial \nu'} = \left[\mp\frac{1}{u}F(F-1) + \sin\theta + iF\cos\theta\right]\frac{\partial \theta}{\partial z}. \quad (12)$$

Then, the surface integral becomes

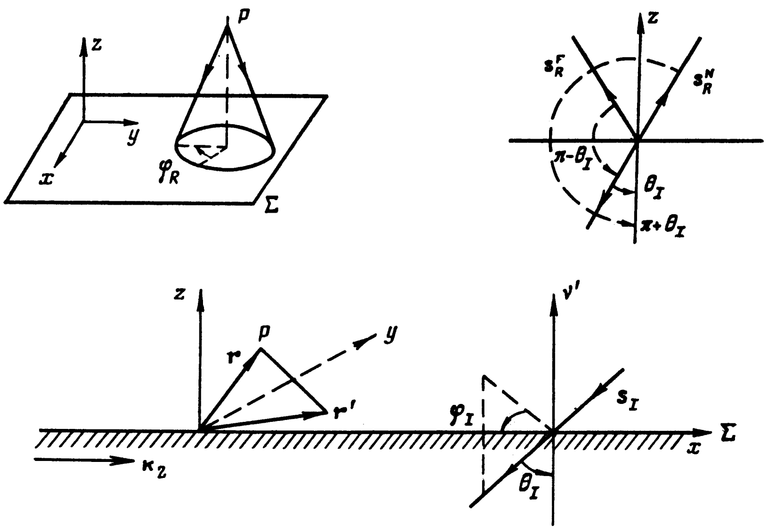

FIG. 3. Schematic representation of the distribution of vectors: $\mathbf{s}_I$ is a unit vector along the direction of incidence of a scanning beam reaching a surface Σ; $\mathbf{s}_R^F$ is a unit vector along the direction of the Fresnel reflection; $\mathbf{s}_R^N$ is a unit vector along the direction of the nonlinear reflection.

$$\mathbf{I}_{-+}=\frac{\mathbf{D}_r}{2}\int\int_{-\infty}^{\infty}\left\{\delta_+\omega\frac{i}{c}\frac{i}{R^2}\left(1+\frac{ic}{\omega R}\right)-\frac{1}{R}\left(\frac{\partial\delta_+}{\partial\mathbf{r}'}\frac{\partial\mathbf{r}'}{\partial\mathbf{v}'}+i\delta_+k_z\right)\right\}\exp\left[i\left(\frac{\omega}{c}R+k_xx'+k_yy'\right)\right]dx'\,dy'. \qquad (13)$$

The exponential factor in Eq. (13) is a rapidly oscillating function of the variables x' and y'. Under these conditions a good approximation to the integral $\mathbf{I}_{-+}$ is obtained by the application of the principle of stationary phase.[21] Then, instead of Eq. (13), we obtain

$$\mathbf{I}_{-+}=\pi i\mathbf{D}_r g_1(x_1', y_1')\exp(irp)/|\alpha_1\beta_1-\gamma_1^2|^{1/2}, \qquad (14)$$

which includes the following quantities:

$$g_1(x_1',y_1')=\delta_+(x_1',y_1')\,\omega\frac{i}{c}\frac{r}{R_1^2}\left(1+\frac{ic}{\omega R_1}\right)-\frac{1}{R_1}\left(\frac{\partial\delta_+}{\partial\mathbf{r}'}\frac{\partial\mathbf{r}'}{\partial\mathbf{v}'}\right)_{x_1',y_1'}-\frac{i}{R_1}k_z\delta_+(x_1',y_1'),$$
$$\alpha_1=\frac{p}{r}\left(1-k_x^2\frac{c^2}{\omega^2}\right),\quad \beta_1=\frac{p}{r}\left(1-k_y^2\frac{c^2}{\omega^2}\right),$$
$$\gamma_1=-k_xk_y\frac{p}{r}\frac{c^2}{\omega^2},\quad R_1=\frac{r}{p}\frac{\omega}{c},\quad p=\left[\frac{\omega^2}{c^2}-k_x^2-k_y^2\right]^{1/2}, \qquad (15)$$

where g_1 is calculated at a point on the surface Σ which has the coordinates $x_1' = -k_x r/p$ and $y_2' = -kr/p$. We shall now substitute Eq. (14) into Eq. (7). The resultant equation applies at arbitrary observation points r provided

$$-(\omega/c)\cos\theta_y=\{(\omega/c)^2-k_x^2-k_y^2\}^{1/2}. \qquad (16)$$

In the case of a fixed plane of incidence ($k_y = 0$), we find from Eq. (16) the following Fresnel reflection law: $\cos\theta_y = \cos\theta_R$, where $\theta_R = \pi - \theta_y$ is the angle of reflection.[21] In general, the quantity $\mathbf{k}$ in Eq. (16) represents the wave vector of a polarization wave excited by high-intensity laser radiation on the surface in the region of an absorption band, i.e., it is defined by $|\mathbf{k}| = (\omega/c)\tilde{n}$, where $\tilde{n}$ is the complex refractive index dependent on the field on the Σ surface of an optical medium. All the quantities in the system (15) are affected by this value of the wave vector, because of the quenching theorem (7). We shall assume that

$$k_x=-(\omega/c)\sin\theta_R\cos\varphi_R+2k_{2x},$$
$$k_y=(\omega/c)\sin\theta_R\sin\varphi_R+2k_{2y}, \qquad (17)$$

where θ_R and φ_R are the reflection angles, and $\mathbf{k}_2$ is the wave vector of a resonance pump wave traveling along the interface Σ. Then, the equality (16) represents the generalized law of reflection of a scanning laser beam. We shall consider a specific physical situation when a scanning beam describes a circle on the interface Σ (Fig. 3). We shall find the conditions under which the reflection applied from different parts of the circle produces an effect at an observation point P. Obviously, in linear optics it follows from the Fresnel reflection law that such a formulation of the problem is meaningless. However, in the situation considered here, there are additional possibilities for reflection which are associated with the phase memory of the interface. We shall substitute Eq. (17) into Eq. (16) and determine all possible values of the angles θ_R for a fixed value of φ_R:

$$\sin\theta_R=(1/2A)\,[B\pm(B^2-4AC)^{1/2}],\quad A=(\omega/c)^2,$$
$$B=4(\omega/c)\,(-k_{2y}\sin\varphi_R+k_{2x}\cos\varphi_R),$$
$$C=-(\omega/c)^2\sin^2\theta_I+4(k_{2x}^2+k_{2y}^2). \qquad (18)$$

We can see that for any given value of the angle of incidence θ_R, we can generally have two complex directions of reflection governed by the wave vector $\mathbf{k}_2$ of a surface resonance pump wave. We shall consider in greater detail the case when $\mathbf{k}_2\|x$ corresponds to complete reversal of the scanning beam, i.e., we shall assume that $\sin\theta_R = \sin\theta_I$ (Fig. 3). We then have

$$-\sin\theta_I=(c/\omega)\,\{2k_{2x}\cos\varphi_R\pm[4k_{2x}^2\cos^2\varphi_R-C]^{1/2}\}\equiv\sin\theta_R, \qquad (19)$$

which is satisfied if $-4k_{2x}^2 \sin^2\varphi_R + (\omega/c)^2\sin^2\theta_I \geqslant 0$. When the condition $C > 4k_{2x}^2 \cos^2\varphi_R$, is obeyed, the angle of reflection is a complex variable. The concept of complex angles is used frequently in the optics of layer absorbing media[24] and the quantity θ_R does not simply represent the angle of reflection. We shall introduce a vector $\mathbf{s}_R$, which describes the complex direction of propagation of a reflected wave. It is convenient to express s_R^z in the form $s_R^z = qe_r^{i\gamma}$, where q and γ are real. We shall then square this quantity. We can obtain equations for the calculations of q and γ by equating the real and imaginary parts. A reflected wave is proportional to the phase factor $\exp(i\mathbf{s}_R \mathbf{r}\omega/c)$ and, therefore, we can use the values of q and γ found in this way to identify the surfaces of constant amplitude of the reflected wave and surfaces of the real phase, which are generally not coincident, i.e., the reflected wave is inhomogeneous. When the above conditions are satisfied, we can use the values of the components of the unit vectors $\mathbf{s}_I$ and $\mathbf{s}_R$ to obtain the following result: the phases of the reflected and incident waves are identical if $\varphi_R = \varphi_I + 2\pi m$, where $m = 0,1,2,\ldots$. It follows that parts of the surface directing a reflected wave to an observation point P do not have to coincide when an allowance is made for the phase memory effects. Therefore, in the case of nonlinear reflection of a scanning beam by an excited surface of a resonant medium, we may expect phase conjugacy of the reflected wave relative to the incident wave.

We have considered above the case when an exciting pulse propagating along a surface (interface) is a traveling surface electromagnetic wave. A wave of polarization of a resonant medium is formed by nonlinear superposition of the reflected wave field and of the field of the traveling surface electromagnetic wave, so that the wave vector $\mathbf{k}$ of the polarization wave can be written in the form of Eq. (17). An equally important case is that of the excitation of a surface by standing surface electromagnetic waves. The method of solution of the optical Bloch equations for a bulk coherent response excited by standing waves can be found in Ref. 2. We can use the results given there to write down the relationships for the wave vectors participating in the process of nonlinear reflection, which can be done by analogy with Eq. (17) but assuming that $k_{2x} = k_{2y} = 0$.

We can similarly study the properties of light transmitted by the surface Σ of an optical medium under selected conditions of illumination of this surface. In the case of a refracted wave, we find that Eq. (17) is replaced with

$$k_x = -(\omega/c)\tilde{n}\sin\theta_T\cos\varphi_T + 2k_{2x},$$
$$k_y = (\omega/c)\tilde{n}\sin\theta_T\sin\varphi_T + 2k_{2y}, \tag{20}$$

where θ_T and φ_T are the refractive angles. In the limiting case of $k_y \to 0$, ignoring nonlinear effects of refraction, we find from Eq. (20) the law of refraction of linear optics, i.e., $\sin\theta_I = \tilde{n}\sin\theta_T$, where $\tilde{n}$ is a field-independent refractive index of the medium.[21]

§4. EXCITATION OF SURFACE ELECTROMAGNETIC WAVES. DISPERSION LAW OF NONLINEAR SURFACE POLARITONS

The relationships obtained above include the wave vector $\mathbf{k}_2$ of a surface electromagnetic wave excited by an external pulsed light source. We shall now consider the properties of a surface electromagnetic wave in the case when the surface is illuminated with an external wave along the direction $s_I^{(2)}$ and the electric field intensity in this wave has the amplitude $\mathbf{E}_{0I}^{(2)}$. We shall select a trial solution for a transmitted wave traveling along the x axis in the form of a plane wave of amplitude $\mathbf{E}_{0T}^{(2)}(x,t)$ and with a wave vector $k_{2x} = (\omega/c)\tilde{n}_2$, where $\tilde{n}_2$ is the complex refractive index dependent on the amplitude $\mathbf{E}_{0T}^{(2)}$ and on the amplitude of the polarization wave $\mathbf{D}_0^+$ in the illuminated part of the surface Σ. The value of D_0^+ is calculated from the equations of motion (2) as a result of successive interactions with various parts of the surface of both the scanning beam and the surface electromagnetic wave.

The quenching theorem (7) yields a formula relating to amplitude $\mathbf{E}_{0I}^{(2)}$ to the polarization of the resonant medium on the surface and it also gives the dispersion law of transient nonlinear surface polaritons. The term "transient nonlinear surface polariton" is introduced to distinguish those nonlinear surface polaritons which are due to transient processes in an optical medium when the quantum dipoles are induced by ultrashort laser pulses of duration much less than any of the relaxation times. In this case the phase memory of resonating atoms is maximal and the relaxation terms can be omitted from the equations of motion (2). In our case a resonant medium has an arbitrary nonlinearity with respect of the field and the nonlinear polarization of this medium can be excited by fields which are separated in space and in time. We shall consider the case of oblique incidence on the interface Σ of a single ultrashort laser pulse when the plane of incidence xz is fixed. The wave vector of the field in the medium is $k_x = -(\omega/c)\tilde{n}\sin\theta_T = -(\omega/c)\sin\varphi$; then, the quantities in Eq. (15) simplify greatly. We shall assume that at a point x_1', y_1' on the surface we have $[(\partial\delta_+/\partial\mathbf{r}')(\partial\mathbf{r}'/\partial\nu')]_1 = 0$; then, we find that

$$g_1 = \delta_1^+ \frac{\omega}{c}\frac{i}{R_1}\left[\frac{r}{R_1}\left(1 + \frac{ic}{\omega R_1}\right) + \tilde{n}\cos\theta_T\right].$$

The second term in the brackets vanishes if the point of observation lies far from the boundary Σ. We shall not drop this term bearing in mind the need to investigate the boundary or surface region of a nonlinear medium. Equations of motion (2) can be used to find δ_1^+, where θ_1 is the area under a laser pulse on the surface Σ at a point x_1', y_1'. For simplicity, we shall consider the case of the s polarization of the external wave. The quenching theorem (7) yields the values of ε' and ε'' and these can then be used to obtain the real refractive index n and the extinction coefficient $\varkappa$, so that the law of dispersion of transient nonlinear surface polaritons can be found.

This dispersion law can be investigated as a function of changes in various parameters (angles of incidence of external radiation $\theta_1 = \varphi$, depth of penetration of nonlinear surface polaritons into a nonlinear medium, duration of pulses, concentration of resonating impurity atoms, etc.). We shall study the dispersion law as a function of changes in the parameter θ_1. The dielectric properties of a nonlinear medium enter in the dispersion law, where they are represented by the

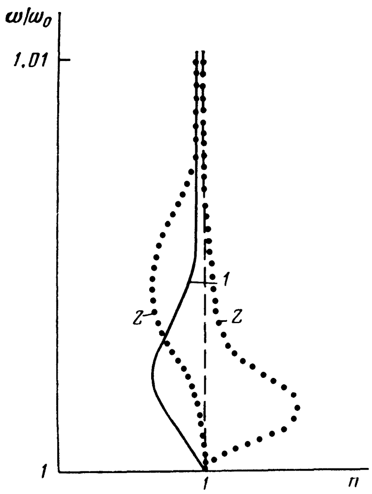

FIG. 4. Dispersion curves of transient nonlinear surface polaritons plotted as a function of the area under a pulse θ_1 incident on a surface: 1) $m = 1.1$; 2) $m = 0.5$; $c/\omega_0 r = 0.1$, $\cos\theta_l = 10^{-2}$, $\theta_1 = \pi m$, $b_1 = -10^{-2}/m$.

nonlinear polarization, which in turn is governed by the area θ_1 under a pulse at the boundary of the medium. If we assume that the field at the point x_1', y_1' is identical with the external field E_{0l}^y and that the external pulse is rectangular, then in the case of weak fields we have $\cos\theta_1 \approx 1$ and the dispersion law ceases to depend on the field, which agrees with the dispersion law of linear surface polaritons. Figure 4 shows the dispersion curves for the real refractive index of a boundary region as a function of the parameter θ_1. Variation of θ_1 in a wide range of values $m = 1.1$–0.1 alters considerably the optical properties of the surface. A numerical analysis of the dispersion law of nonlinear surface polaritons shows that for selected values of θ_1 the extinction coefficient vanishes, indicating that undamped waves (solitons) can then travel along the surface. A second polariton branch appears beginning from $m = 1$, and this branch gradually broadens on increase in the parameter m. Therefore, smooth variation of θ_1 for fixed other parameters of the theory can result in excitation of nonlinear surface polaritons. An additional criterion is then provided by the condition $\cos\theta_l = (1 - \tilde{n}_2^2)^{1/2}$ which follows from the quenching theorem (7).

§5. REFLECTIVITY OF AN INTERFACE BETWEEN TWO MEDIA CALCULATED ALLOWING FOR THE PHASE MEMORY OF THE SURROUNDING SURFACE

The reflection law (16) allows us to determine the conditions under which a scanning beam is reversed to reach a point P (Fig. 3). We shall now calculate the electric field of a reflected wave using Eq. (4). Each part of the surface is illuminated twice for time intervals Δt_1 and Δt_2 first by a scanning beam and then, after a time τ, by a surface electromagnetic wave. If τ, Δt_1, $\Delta t_2 \ll T_1$, T_2', where T_1 is the longitudinal relaxation time and T_2' is the transverse irreversible relaxation time[1,2] of a system of surface atoms, then the condition for coherent interaction of light with the surface is obeyed and at a moment 2τ we can expect afterglow of the surface Σ directed toward the observation point P. The phase memory of the surface atoms is governed by the wave vector (17). We can calculate the field of the reflected wave, representing a coherent response of the surface atoms to the two-pulse interaction with light if we know the polarization wave D_0^+ at moments after the end of the second pulse in a certain part of the surface. The induced polarization wave can be calculated during each of the light pulses by matched solution of the equations of motion for the atoms (2) and of the equation for the field (1). For each pulsed interaction we obtain equations that follow from the quenching theorem (7) and have the relevant interaction parameters g_1, α_1, β_1, and γ_1. We must bear in mind that at the moment of arrival of the second light pulse on a certain part of the surface, the polarization of the medium differs from zero because it has been induced by the first pulse. Therefore, calculation of the polarization wave in a system of surface atoms resembles calculation of the microscopic dipole moment in the optical echo effect.[1,2] An important difference is the procedure of matching of the waves at the boundary of an optical medium. This procedure includes the following stages. 1) The quenching theorem is used to find an equation relating the amplitude of an external wave to the polarization wave on the surface of the medium. The quantities u and v, governing the polarization wave, are found from the optical Bloch equations (2), where the exciting field is a certain field having a pulse area θ_1 at a point x_1', y_1' on the surface Σ. This field represents a nonlinear superposition of the reflected wave field and the field of the surface electromagnetic wave. When the conditions for reversal of the scanning beam are satisfied, we can find the quantities p, R_1, α_1, β_1, γ_1, and g_1 at the point x_1', y_1' on the surface Σ. Using the generalized Lorentz-Lorenz formula on Eq. (9), we can express the complex refractive index occurring in the quenching theorem in terms of the parameter θ_1. After the curl curl operation on the coordinates of the observation point, we obtain the differential equation the solution of which allows us to find the relationship between θ_1 and the amplitude of the external wave. 2) The generalized Lorentz-Lorenz formula (9) is then used to find the refractive index n and the extinction coefficient $\varkappa$ of the surface after the end of the second pulse. Selecting suitably the conditions for such two-pulse illumination of the surface, we must satisfy the law of reflection (19) with relevant values of n and $\varkappa$ on the surface of the investigated optical resonant medium.

It follows from Eq. (4) that the electric field intensity in the reflected wave can be described approximately by

$$E_R = [c^2/\omega^2(\tilde{n}^2-1)]\,(\pi i g_1|\alpha_1\beta_1-\gamma_1^2|^{-1/2})\ \mathrm{rot\,rot}\,(D_2 e^{irp}), \quad (21)$$

where $\tilde{n}$ is the complex refractive index of the surface of a resonant optical medium, which depends nonlinearly on the polarization of the medium after the end of the second exciting pulse. In the linear approximation [when $\mathbf{D}_0^+$ in Eq. (9) is a linear function of $\mathbf{E}_0$] we can ignore the effects of the phase memory of the surface atoms and then Eq. (21), for a fixed plane of incidence, reduces to the Fresnel reflection law for the intensity of the electric field in the reflected wave. Equation (20), together with the formula (19) for the direction of propagation of the reflected wave, describes the properties of a nonlinear mirror operating on the basis of the phase memory of the reflecting surface. We shall calculate the reflectivity of this mirror, selecting the reflecting surface of a ruby crystal activated with Cr^{3+} crystals. Applying the formulas from Eq. (15), we find the reflectivity of the interface between vacuum and a nonlinear resonant medium $R = |E_0^R|^2/|E_{0I}^{(1)}|^2$, where $E_{0I}^{(1)}$ is the amplitude of the electric field in the scanning beam. In the case of the s polarization of this beam, when the coherent response of the crystal surface is formed by a standing surface wave, we obtain the following formula from Eq. (21):

$$R=[(1-w^2)/(E_{0I}^{(1)})^2](D_r v)^2 \cos^4 \theta_1 4\pi^2(\tilde{n}^2-1)^{-2}, \qquad (22)$$

where w is the inversion of the surface atoms which have interacted extensively with two light pulses. We shall assume that the fields on the surface are such that the areas of the pulses are $\theta_1^I = \pi/2$ and $\theta_1^{II} = \pi$. It follows from the results in §4 that we have $n \to 1$ in the limit $\theta_1^{II} \to \pi$ when the extinction coefficient is $\varkappa \to 0$. The quantity $E_{0I}^{(1)}$ can be estimated if we assume that $\varkappa_0 E_{0I}^{(1)} D v_{sc} \approx \pi/2$. When the beam diameter is $D = 0.1$ cm and the scanning velocity is $v_{sc} = 10^6$ cm/sec, we shall assume that $E_{0I}^{(1)} = 1$ cgs esu; then, a nonlinear mirror utilizing the phase memory of a reflecting surface with a phase conjugacy of the wavefront of a scanning beam can have a high reflectivity ($R \to 1$) if the optical properties of the surface of such a resonant medium are modified suitably by a surface electromagnetic wave.

We have thus investigated some laws governing a coherent interaction between ultrashort light pulses and the surfaces of a resonant medium when the phase memory effects are exhibited by surface atoms. The example of a two-pulse excitation of the surface by a scanning beam and a traveling surface electromagnetic wave is used to show how the problem can be solved completely, i.e., how the reflectivity of the surface can be calculated. The proposed procedure for the calculation of the optical properties of surfaces can of course be generalized to more complex cases in which three or more exciting laser pulses are used.

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