Coherent interaction of laser pulses in a resonant optically dense extended medium under the regime of strong field-matter coupling

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The nonstationary pump-probe interaction between short laser pulses propagating in a resonant optically dense coherent medium is considered. Special attention is paid to the case where the density of two-level particles is high enough that a considerable part of the energy of relatively weak external laser fields can be coherently absorbed and reemitted by the medium. Thus, the field of the medium reaction plays a key role in the interaction processes, which leads to collective behavior of an atomic ensemble in the strongly coupled light-matter system. Such behavior results in fast excitation interchanges between the field and a medium in the form of optical ringing, which is analogous to polariton beating in solid-state optics. This collective oscillating response, which can be treated as successive beats between light wave packets of different group velocities, is shown to significantly affect the propagation and amplification of the probe field under its nonlinear interaction with a nearly copropagating pump pulse. Depending on the probe-pump time delay, the probe transmission spectra show the appearance of either a specific doublet or coherent dip. The widths of these features are determined by the density-dependent field-matter coupling coefficient and increase during the propagation. Besides that, the widths of the coherent features, which appear close to resonance in the broadband probe spectrum, exceed the absorption-line width, since under the strong-coupling regime, the frequency of optical ringing exceeds the rate of incoherent relaxation. Contrary to stationary strong-field effects, the density- and coordinate-dependent transmission spectra of the probe manifest the importance of collective oscillations and cannot be obtained in the framework of a single-atom model.

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I. INTRODUCTION

Investigation of the nonlinear medium response to coherent radiation is a powerful tool to analyze the microscopic properties of matter and ultrafast processes taking place therein. Moreover, a detailed study of resonant light-matter interactions enables the development of techniques of active control of the processes under investigation. Optically dense media attract significant attention, since they provide information about phenomena taking place in macroscopic ensembles of resonant particles. On the other hand, a dense medium represents a rather complicated object, because it can strongly modify the characteristics of the probing radiation by coherent absorption and reemission of photons. These processes lead to the appearance of the medium reaction field, which plays an essential role and becomes comparable to the field of an external radiation source. Thus, the coupled electromagnetic field and optically dense matter should be considered self-consistently as a single system with specific characteristics and evolution.

In the limit where the reemission field is negligible in comparison to the strong, externally applied one, the model of a single driven atom is usually used. Atomic variables can be calculated on the basis of Bloch equations without taking into account Maxwell equations. Thus, the spectral features of the process are fully determined by the strong external field, but not by the density of the medium. In this context, phenomena such as Rabi-sideband generation due to the stationary probe-pump Mollow-Boyd effect [1] or to transient Rabi flopping [2] can be mentioned.

The main peculiarity of the coherent interaction between light and optically dense media is related to the collective behavior of resonant particles due to their interaction through the reemission field. This behavior can significantly modify the mutual light-matter dynamics, which cannot be described in the framework of the single-atom model. In the case where a high density of the resonant medium enables the atomic system to coherently absorb and reradiate a considerable part of the energy of the external electromagnetic field, fast excitation interchanges between field and matter appear, which is analogous to polariton beats in solid-state optics. The frequency of such collective oscillations is determined by the density-dependent field-matter coupling coefficient and exceeds the relaxation rates of a medium. Under the cavity interaction, this "strong-coupling regime" results in vacuum Rabi oscillations [3–7]. In the field of cavity QED, the nonclassical properties of the strong-coupling regime (photon antibunching and sub-Poissonian statistics [5], squeezing [6], and application for entanglement [7]) have been discussed. Under free-space pulse propagation, the strong-coupling regime leads to the formation of collective optical ringing [8,9]. The generation of such a signal may appear even under weak-field excitation, without essential

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changes in the population difference of a medium. This fact is different from the case of the resonant pulse breakup into 2π solitons of self-induced transparency [2], where the incident pulse is strong enough to invert a part of a medium (the area of the external field should be at least greater than π). In this context, optical ringing represents a nonsolitonic solution in the form of a 0π pulse.

In this paper, we present a study of the coherent free-space interaction between two (probe and pump) short laser pulses propagating in a two-level extended medium under the conditions of the strong-coupling regime. We center our discussion on the question as to how the collective oscillatory response of an optically dense medium, excited by a short pump pulse, affects the transmission spectrum of the probe field. Since optical ringing originates from the linear response of a dense resonant medium, we consider the case of a weak pump field, which does not invert atoms. Hence, all obtained transformations of the probe pulse, particularly its amplification, take place in a medium without population inversion.

Optical ringing represents an essentially transient phenomenon and can be observed in a time scale shorter than the relaxation times of a medium. Due to its general character, such an oscillating response accompanies a variety of coherent phenomena taking place under short pulse propagation [8] in dense media and should be taken into account in analyses of experimental data. Having a superradiant character, it was studied in the context of the so-called "polarium model" (purely coherent interaction) [9] and oscillatory regime of Dicke superradiance [10]. Its influence on the quantum coherent control and pulse shaping techniques was mentioned in Refs. [11–13]. The works in [14] were devoted to a determination of the relaxation times of homogeneously and inhomogeneously broadened spectral lines of molecular substances in the presence of optical ringing. In Refs. [11,12,15], different aspects of the coherent interaction of ultrashort laser pulses with extended multilevel atomic media, particularly two-photon absorption, were considered. The existence of ultrafast collective oscillations was demonstrated in Ref. [16], where not only the external laser pulse, but also the optical ringing itself belonged to the femtosecond time scale.

Besides the atomic and molecular media, optical ringing was also rediscovered for the case of solid-state exciton polaritons [17]. It was treated as quantum beats between two branches of the polariton dispersion curve corresponding to a single resonance, contrasting to the case of the usual quantum beats between different optical transitions. Moreover, the transition from the strong-field regime of Rabi oscillations to the weak-field optical ringing under the strong-coupling regime was recently observed in Ref. [18]. Femtosecond ringing was obtained for the cases of both bulk crystals [19] and multiple-quantum-well structures [20].

In the presence of optical ringing, the interaction of two intersected probe and pump beams (this configuration is also used in the present work) was considered for the cases of atomic media [21,22] and solids [20,23–25]. Most of the results of these studies are related to the time-integrated and time-resolved properties of the diffracted four-wave mixing signal. In contrast, our discussion is centered on modification of the probe field and, particularly, on its spectral properties. In our previous works [26–29], we studied both theoretically and experimentally the interaction of broadband polychromatic pulses of a multimode dye laser without mode locking in an optically dense resonant medium (metastable neon atoms in gas discharges). Different spectral types of probe amplification were obtained and analyzed. Our present paper is devoted to the interaction of short transform-limited pulses, since the fundamentals of the phenomena considered arise from the coherent interaction between short pulses and a dense medium.

The paper is organized as follows. In Sec. II, the main equations of the theoretical model are obtained. In Sec. III, we outline some specific features of the collective optical ringing originating from the dispersion of a dense resonant medium. The numerical results and discussion of the probe-field propagation in the presence of a pump pulse are presented in Sec. IV. The main results of the paper are summarized in Sec. V.

II. EQUATIONS OF THE MODEL

The theory of transient processes of the interaction between electromagnetic fields and resonant media is based on a joint solution of the semiclassical Maxwell-Bloch equations [2]. We restrict our study to the two-level system approximation assuming that the interaction between the field and neighboring atomic transitions is negligibly small. The electric field $E(t,\mathbf{r})$ and the polarization of the medium $P(t,\mathbf{r})$ can be written in the form

$$E(t,\mathbf{r}) = \frac{1}{2}\frac{\hbar}{d}[\Omega_s(t,\mathbf{r})e^{i\omega_0 t} + \text{c.c.}],$$

$$P(t,\mathbf{r}) = \frac{1}{2}nd[p_s(t,\mathbf{r})e^{i\omega_0 t - i\pi/2} + \text{c.c.}],$$

introducing the complex amplitudes $\Omega_s(t,\mathbf{r})$ and $p_s(t,\mathbf{r})$ slowly varying in time but having arbitrary spatial dependence. Here, d and ω_0 are the electric dipole moment and the resonance frequency of the atomic transition; n is the density of atoms. The amplitude $\Omega_s(t,\mathbf{r})$ is expressed in units of the Rabi frequency of the electric field.

Using the rotating-wave approximation, the system of Bloch equations can be written as

$$\frac{\partial p_s}{\partial t} = \Omega_s D - \gamma_2 p_s, \tag{1a}$$

$$\frac{\partial D}{\partial t} = -\frac{1}{2}(\Omega_s p_s^* + \Omega_s^* p_s) - \gamma_1(D - D^{eq}), \tag{1b}$$

where D is the population difference of an atom, D^{eq} is the value of D in the absence of an external field (the value $D=1$ corresponds to an atom in the ground state), and γ_1 and γ_2 are the relaxation rates.

The problem of the interaction between two intersected plane linearly polarized waves is considered. In the case of nearly copropagating pulses, the amplitude of the field is given by

$$\Omega_s = \Omega_0(t,z)e^{-ik_0 z} + \Omega_1(t,z)e^{-i\mathbf{k}_1\mathbf{r}}, \tag{2}$$

with Ω_0 and Ω_1 slowly varying in space. The field Ω_0 with wave vector $\mathbf{k}_0$, which is parallel to the z axis, corresponds to the strong pump wave, whereas Ω_1 with wave vector $\mathbf{k}_1$ is assumed to be a weak probe field propagating at the small angle φ with respect to the z direction. A nonlinear interaction of the intersected waves leads to the appearance of spatial polarization harmonics with wave vectors $\mathbf{k}_0+m\Delta\mathbf{k}$ ($m=0,\pm1,\pm2,...$, $\Delta\mathbf{k}=\mathbf{k}_1-\mathbf{k}_0$) and harmonics of the population difference with $m\Delta\mathbf{k}$ wave vectors:

$$p_s = \sum_{m=-\infty}^{\infty} p_m(t,z)e^{-i(\mathbf{k}_0\mathbf{r}+m\Delta\mathbf{k}\mathbf{r})}, \tag{3a}$$

$$D = \sum_{m=-\infty}^{\infty} D_m(t,z)e^{-im\Delta\mathbf{k}\mathbf{r}}, \quad D_m = D_{-m}^{*}. \tag{3b}$$

The emission of p_0 and p_1 polarizations corresponds to the pump and probe fields, respectively. The emission of higher harmonics is considered to be suppressed in a thick medium, due to the mismatch in the dispersion relation. Substituting the expansions (2) and (3) into Bloch equations (1), adding Maxwell equations, and using the first-order perturbation theory with respect to the small amplitude of the probe field, one can get the coupled Maxwell-Bloch system, describing propagation of the strong pump field,

$$c\frac{\partial\Omega_0}{\partial z} + \frac{\partial\Omega_0}{\partial t} = -\omega_c^2 p_0, \tag{4a}$$

$$\frac{\partial p_0}{\partial t} = \Omega_0 D_0 - \gamma_2 p_0, \tag{4b}$$

$$\frac{\partial D_0}{\partial t} = -\frac{1}{2}(\Omega_0 p_0^{*} + \Omega_0^{*} p_0) - \gamma_1(D_0 - D^{eq}), \tag{4c}$$

and a weak probe,

$$c\cos\varphi\frac{\partial\Omega_1}{\partial z} + \frac{\partial\Omega_1}{\partial t} = -\omega_c^2 p_1, \tag{5a}$$

$$\frac{\partial p_1}{\partial t} = \Omega_1 D_0 + \Omega_0 D_1 - \gamma_2 p_1, \tag{5b}$$

$$\frac{\partial D_1}{\partial t} = -\frac{1}{2}(\Omega_1 p_0^{*} + \Omega_0^{*} p_1 + \Omega_0 p_{-1}^{*}) - \gamma_1 D_1, \tag{5c}$$

$$\frac{\partial p_{-1}^{*}}{\partial t} = \Omega_0^{*} D_1 - \gamma_2 p_{-1}^{*}, \tag{5d}$$

where

$$\omega_c = \sqrt{\frac{2\pi d^2\omega_0 n}{\hbar}} \tag{6}$$

is the cooperative frequency of the medium, which plays a role of the coupling coefficient between field and matter. We will present the numerical results of our work in the dimensionless form in units of ω_c.

If the functions $\Omega_{0,1}(t,z)$ are real (exactly resonant interaction without phase modulation of input pulses), the areas of the pulses, defined as

$$s_{0,1}(z) = \int_0^{\infty} \Omega_{0,1}(t,z)dt,$$

are very important parameters characterizing coherent propagation of short laser pulses. In the case of complex functions $\Omega_{0,1}(t,z)$, the definition of the area does not have a general form.

III. PECULIARITIES OF OPTICAL RINGING IN A SINGLE LASER BEAM

Contrasting to the case of nonlinear pulse breakup into 2π solitons [2], the origin of collective oscillations can be traced to the linear light-matter interaction. In this section, we outline the main features of this phenomenon, considering the propagation of the single (pump) field described by Eqs. (4). In the linear limit of a weak (small-area) field, where the atoms are assumed to be in the ground state with $D(t,z)=1$, Eqs. (4a) and (4b) can be written as a single second-order differential equation

$$c\frac{\partial^2\Omega}{\partial z\,\partial t} + \frac{\partial^2\Omega}{\partial^2 t} + \gamma_2\left(c\frac{\partial\Omega}{\partial z} + \frac{\partial\Omega}{\partial t}\right) + \omega_c^2\Omega = 0, \tag{7}$$

describing oscillations of the laser field, which propagates in a medium with high cooperative frequency ω_c, Eq. (6). In this section, considering the propagation of a single beam, we have omitted the subscript of the field amplitude. The problem of the free-space propagation of a short laser pulse entering an initially unperturbed medium can be solved taking the time Fourier transform of Eq. (7). The inverse Fourier transform then gives a solution in the form (cf. Ref. [8])

$$\Omega(t,z) = \frac{1}{2\pi}\int_{-\infty}^{\infty} F_{\mathrm{in}}(\omega)e^{-ik(\omega)z}e^{i\omega t}d\omega, \tag{8}$$

where $F_{\mathrm{in}}(\omega)$ is the spectrum of the laser pulse at the input of the medium at $z=0$. The spatial evolution of spectral components is determined by the wave vector $k(\omega)$:

$$ck(\omega) = \omega - \frac{\omega_c^2}{\omega - i\gamma_2}. \tag{9}$$

Here, the frequency ω and wave vector k are defined as detunings from the resonant values ω_0 and ω_0/c, respectively. The real part of $k(\omega)$ determines the dispersive characteristics of the propagating field, whereas its imaginary part determines the absorption.

Depending on the relation between the field-matter cou-

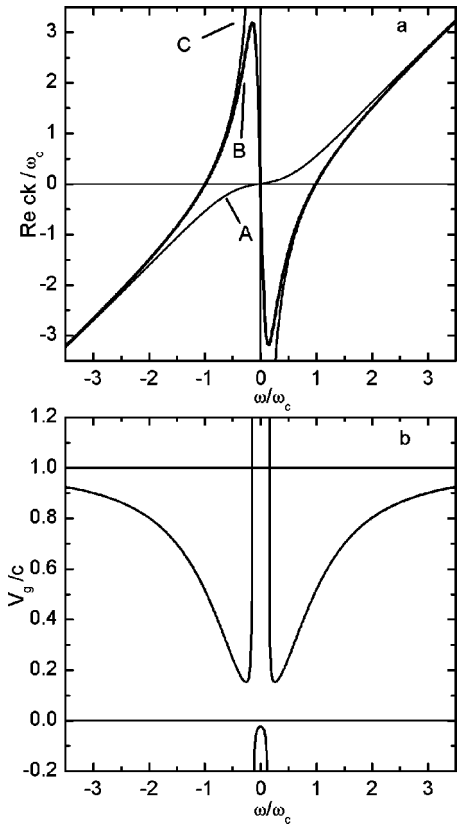

FIG. 1. Dispersion characteristics of dense resonant medium. (a) Linear dispersion Re $k(\omega)$ for $\gamma_2/\omega_c=1.1$ (curve *A*), 0.15 (curve *B*), and 0.0 (curve *C*); (b) group velocity $V_g(\omega)$ for $\gamma_2/\omega_c=0.15$.

pling coefficient ω_c and the rate of incoherent relaxation γ_2, two qualitatively different frequency dependences of the real part of $k(\omega)$ are possible, which is demonstrated in Fig. 1(a). If the relaxation rate exceeds the cooperative frequency, only one value of the frequency ω corresponds to each wave vector k (curve *A*). In the opposite case, where

$$\omega_c > \gamma_2, \tag{10}$$

there may exist three frequencies that correspond to the same value of k (curve *B*). The central frequency falls into the region of the anomalous dispersion with strong absorption of the field, whereas two other solutions appear at the wings of the spectral line, where absorption may be very small.

The existence of photons of equal wave vectors, but of different frequencies, is a characteristic property of the resonant light-matter interactions in optically dense coherent media. This phenomenon can be treated as a density-dependent splitting of normal modes in the system of strongly coupled field-matter oscillators (polaritons). In the limiting case of purely coherent interactions ($\gamma_2=0$), the polariton dispersion is represented by two separated anticrossing branches, displayed by curve *C* in Fig. 1(a). Coherent beating between normal modes of these kind can be observed, if the spatial spectrum of the field is fixed, for example, by initial conditions or by the presence of a cavity. In this context, effects such as cavity collective vacuum Rabi oscillations [3,4] and the oscillatory regime of superradiance under side excitation of a sample [10] can be mentioned. In these cases, fixing of wave vectors leads to the appearance of a characteristic doublet in the field spectrum, which corresponds to the normal modes and manifests the existence of the "strong-coupling regime" determined by Eq. (10).

In the situation under analysis, Eqs. (7) and (8), where a laser pulse propagates in an initially unperturbed medium, the frequency content of the output field is entirely determined by the spectrum of the external source $F_{\text{in}}(\omega)$. Due to the linearity of interaction, the absolute value of the output spectrum does not vary during the propagation, except for the trivial appearance of the absorption line. Thus, in this case, contrary to collective vacuum Rabi oscillations, no coherent density-dependent spectral features can be extracted from spectral measurements. At the same time, coherent oscillations can be observed under analysis of the temporal behavior.

Using the theorem about the Fourier transform of a product, the solution (8) can be rewritten in the form (cf. Ref. [14])

$$\Omega(t,z) = \Omega_{\text{in}}(\tau) - \int_0^\tau \Omega_{\text{in}}(\tau - t')e^{-\gamma_2 t'}\omega_c\sqrt{\frac{z}{ct'}} \times J_1(2\omega_c\sqrt{zt'/c})dt' \tag{11}$$

for $\tau \geqslant 0$ and $\Omega(t,z)=0$ for $\tau<0$. Here $\tau=t-z/c$, $\Omega_{\text{in}}(t)$ is the complex amplitude of the electric field at the input of the medium, and $J_1(x)$ is the first-order Bessel function. The integral kernel represents the oscillatory response of the atomic ensemble to the short δ pulse $[\Omega_{\text{in}}(t)=\delta(t)]$ and, thus, represents the Green function of the problem.

Optical ringing, which is described by the Bessel function in Eq. (11), corresponds to oscillations with frequency varying in time and space. This fact reveals the principal role of propagation effects in the formation of optical ringing. In a cross section of the medium with coordinate z, the inverse duration of the first short pulse, which was reradiated by the medium, is proportional to the quantity

$$\omega_D = \frac{\omega_c^2 z}{c}. \tag{12}$$

The frequency of subsequent oscillations decreases in time and at the tail of the ringing is given by $\omega_c\sqrt{z/(c\tau)}$. Hence, to observe at least the initial stage of the coherent optical ringing, the following criterion should be fulfilled:

$$\omega_D = \frac{\omega_c^2 z}{c} \gg \gamma_2. \tag{13}$$

The frequency of optical ringing in its initial stage increases with propagation distance z and medium density n. Thus, it is proportional to the number of atoms interacting via the reemission field (here, only propagation in the forward direction is considered). Such a dependence directly demonstrates the collective character of the field reemission by the atomic ensemble. The quantity ω_D, Eq. (12), is a well-known parameter in the theory of Dicke superradiance [9,10]. Unlike a single superradiant pulse, which is considered as fast collec-

tive relaxation of atoms to the ground state, optical ringing represents fast collective oscillations that nevertheless remain coherent in a time scale of $1/\gamma_2$, which may be much longer than the duration of a single pulse of superradiance.

The condition of the appearance of optical ringing, Eq. (13), is different from that of the collective vacuum Rabi oscillations, Eq. (10). Nevertheless, the relation between the field-matter coupling coefficient ω_c, Eq. (6), and the rate of incoherent relaxation of the medium is very important for both types of collective oscillations. The relaxation rates included in our model may account for radiative and collisional broadening of a spectral line, while the inhomogeneous Doppler broadening is not taken into account. Nevertheless, if the processes of interest are much faster than the relaxation and the detailed shape of a narrow absorption contour is not of importance, the Doppler broadening may be also approximately included in the relaxation rates considered [8].

The expressions (10) and (13) can be considered as conditions determining a threshold value for the atomic density, which should be exceeded to obtain collective oscillations. It is important to stress that these conditions are essentially weaker than the one necessary for other density-dependent phenomena in resonant media, such as total backward reflection [9] and local field effects [30] leading to a Lorentz-Lorenz frequency shift and intrinsic optical bistability. This fact substantiates our model presented in Sec. II, which does not take into account the latter high-density effects.

The statement—that the initial optical ringing frequency (12) displays an unlimited increase during the propagation—is valid only for the idealized situation of an input pulse having an infinitely broad spectrum. To describe more realistic conditions, we will present a simple analytical expression giving a relation between the temporal behavior of the field and the spectrum of the input pulse.

For this purpose, the solution (8) can be analyzed using the stationary phase method, which gives the following asymptotical value for an integral expression:

$$\int_{-\infty}^{\infty} f(\omega) e^{i\lambda s(\omega)} d\omega \sim \sum_{\omega_s} \sqrt{\frac{2\pi}{\lambda |s''(\omega_s)|}} f(\omega_s) e^{i\lambda s(\omega_s) \pm i\pi/4} \tag{14}$$

for $\lambda \to \infty$ and $s''(\omega_s) \neq 0$. The sum is taken over all points of the stationary phase ω_s such that $s'(\omega_s)=0$; the plus or minus sign is chosen for $s''(\omega_s)>0$ and $s''(\omega_s)<0$, respectively. In the limit of coherent interaction, $\gamma_2=0$, the solution (8) and (9) can be reduced to the integral on the left-hand side of Eq. (14) by the substitutions

$$f(\omega) = \frac{F_{\text{in}}(\omega)}{2\pi}, \quad \lambda = \frac{\omega_c z}{c}, \quad s(\omega) = \frac{\omega}{\omega_c}\beta + \frac{\omega_c}{\omega},$$

where $\beta = c\tau/z$. In this case, there exist two stationary phase points $\omega_{s1,2} = \pm \omega_g$:

$$\omega_g = \omega_c \sqrt{\frac{z}{c\tau}}. \tag{15}$$

Using Eq. (14), the asymptotic solution of the problem for high values of $\omega_c z/c$, finite values of the parameter β, and a smooth function $F_{\text{in}}(\omega)$ can be written as

$$\Omega(t,z) \sim \frac{1}{2\tau\sqrt{2\pi}} (2\omega_c \sqrt{z\tau/c})^{1/2} \left[F_{\text{in}}\left(\omega_c \sqrt{\frac{z}{c\tau}} \right) e^{i2\omega_c \sqrt{z\tau/c} + i\pi/4} + F_{\text{in}}\left(-\omega_c \sqrt{\frac{z}{c\tau}} \right) e^{-i2\omega_c \sqrt{z\tau/c} - i\pi/4} \right]. \tag{16}$$

Expression (16) shows that the field amplitude $\Omega(t,z)$ is determined by the complex amplitudes of the input spectrum at frequencies $\pm\omega_g$, Eq. (15). These frequencies are symmetrically placed around the frequency of the atomic transition and depend on z and t. Under an increase of the propagation distance z, the field amplitude is determined by the spectral components lying farther and farther from the resonance. Thus, the increase of the ringing frequency is limited by the frequency of non-negligible spectral components farthest from the resonance.

On the other hand, in a fixed cross section of the medium, the time evolution of the field is determined by the spectral components, which are placed closer and closer to the resonance frequency. In other words, Eqs. (15) and (16) show that in the cross section z, a spectral component of an arbitrary frequency ω determines the field amplitude at the time moment $\tau(\omega)$ such that

$$\tau(\omega) = \frac{\omega_c^2 z}{\omega^2 c}.$$

One can easily show that $\tau(\omega)$ corresponds to the time delay related to the difference between the speed of light in vacuum and the group velocity of a wave packet of frequency ω. The frequency-dependent group velocity $V_g(\omega)$ is defined by the derivative of the real part of the wave vector $k(\omega)$, Eq. (9),

$$\frac{V_g(\omega)}{c} \equiv \left(c \frac{d\ \text{Re}\ k}{d\omega} \right)^{-1} = 1 - \frac{\omega_c^2(\omega^2 - \gamma_2^2)}{(\omega^2 + \gamma_2^2)^2 + \omega_c^2(\omega^2 - \gamma_2^2)},$$

and is displayed in Fig. 1(b). In the case $\omega_c \gg \gamma_2$, the width of the absorptive (anomalous dispersion) area $2\gamma_2$ is much smaller than the width of the $V_g(\omega)$ contour, which is equal to $2\omega_c$ and hence increases with atomic density. Figure 1(b) shows that in an optically dense coherent medium, the group velocity significantly varies over the spectrum and can be essentially reduced near the resonance.

Thus, long oscillations of optical ringing can be treated as successive beats between light wave packets of frequencies symmetrical with respect to the resonance [17]. Equation (16) gives the quantitative characteristics of this treatment. In contrast to collective vacuum Rabi oscillations, which are explained by the beating between two monochromatic waves of equal wave vectors [see Fig. 1(a)], free-space optical ring-

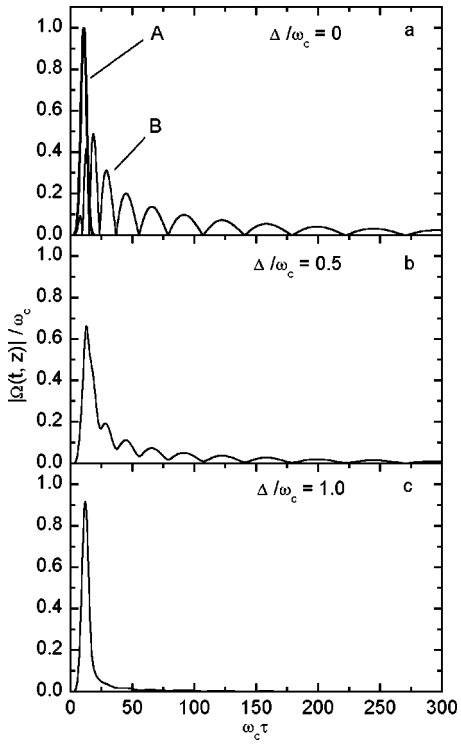

FIG. 2. Optical ringing at different detunings of the pulse spectrum from resonance frequency. (a) Input pulse (curve *A*) and output pulse for $\Delta/\omega_c=0.0$ (curve *B*); (b) output pulse for $\Delta/\omega_c=0.5$; (c) output pulse for $\Delta/\omega_c=1.0$; $\gamma_2/\omega_c=10^{-3}$, FWHM of the spectrum $\gamma_{sp}/\omega_c=1.0$, propagation distance $\omega_c z/c=1.0$.

ing originates from the beats between wave packets of equal group velocities, which should be present in the broad spectrum of a short input pulse [see Fig. 1(b)].

Moreover, Eq. (16) shows that to observe optical ringing in the absolute value of the electric field $|\Omega(t,z)|$, the spectrum of the input pulse $F_{in}(\omega)$ should excite both red and blue wings of the spectral line. This fact confirms the essentially resonant character of the collective optical ringing. In the case where the input spectrum covers only one of the wings, oscillations in the absolute value are absent, though they are still present in the quadrature components of the field, which are determined by the real and imaginary parts of $\Omega(t,z)$.

Figure 2 displays the temporal behavior of optical ringing [the solution of Eq. (7)] excited by a short Gaussian-shaped input pulse

$$\Omega_{in}(t)=\frac{s}{a\sqrt{\pi}}e^{-[(t-t_0)/a]^2+i\Delta t}\vartheta(t) \tag{17}$$

at different detunings Δ between the pulse central frequency and the frequency of the resonance. The full width at half maximum (FWHM) of the spectrum $F_{in}(\omega)$, which corresponds to the input pulse (17), is determined through the parameter a and given by

$$\gamma_{sp}=\frac{4}{a}\sqrt{\ln 2}. \tag{18}$$

In Eq. (17), the time delay $t_0\gg 2\pi/\gamma_{sp}$ and the Heaviside step function $\vartheta(t)$ are included to fulfill the formal requirement of $\Omega_{in}(t)=0$ for $t<0$; quantity s is the area of the input pulse.

Figure 2 shows that for fixed parameters of the medium, the amplitude of the ringing in $|\Omega(t,z)|$ essentially decreases under an increase of the detuning Δ and, hence, under a decrease of spectral amplitudes at one wing of the resonance in comparison with amplitudes at another one. Deformation of the input pulse also decreases, manifesting the transition to the regime of the usual nonresonant pulse propagation with a mean group velocity. At the same time, the frequency of optical ringing, which depends on the characteristics of the medium, does not change, since it is determined by the oscillating exponents in Eq. (16) but not by the amplitudes of the wave packets, $F_{in}(\omega)$. This fact was experimentally demonstrated, particularly in Refs. [16,19].

IV. NONLINEAR PUMP-PROBE INTERACTION

The dynamics of coherent collective phenomena under the regime of strong light-matter coupling should significantly affect the nonlinear interaction of laser fields in resonant media. The collective model of "spectrum condensation" (or "self-frequency locking") [26,31–33] under the generation of a multimode dye laser with an intracavity absorbing cell was proposed in Ref. [26]. Particularly, in the case of short transform-limited pulses, the "spectrum condensation" was investigated in Refs. [31,32]. Giant parametric amplification of polaritons was observed in semiconductor microcavities under a large vacuum Rabi splitting [6,34,35].

In this paper, we present an investigation of influence of collective optical ringing on the transmission of a probe pulse in the presence of a short pump pulse. In the presence of optical ringing, the configuration of two intersected beams was also considered in investigations of the resonant four-wave mixing in atomic [21,22] and solid-state media [20,23–25]. In these works, the collective oscillations of the field were obtained in the time-resolved and time-integrated studies of a new diffracted pulse. In contrast, our present paper deals with spectral properties of the probe beam. In Refs. [20,21,23–25], the probe field was not affected by the pump pulse, since the latter one was considered to be very weak. The theoretical model of Ref. [22] accounted for a modification of the probe field, but the carrier frequency of the pump pulse was chosen to be shifted from the resonance; thus no optical ringing was produced in the absolute value of the pump field and in the population difference of the medium. In the present paper, the case of a resonant pump pulse is analyzed, and optical ringing is able to affect the probe field also through collective oscillations of the population difference $D_0(t,z)$, which couples two beams intersected at a small angle [cf. Eqs. (5a) and (5b)]. In Ref. [36], where the interaction of two short resonant pulses was considered under an exactly collinear configuration, it was impossible to distinguish between the probe and pump fields, when the long oscillating field of the medium response appeared.

In our previous works [26–29], we studied, both experimentally and theoretically, the pump-probe interaction of broadband polychromatic pulses produced by dye lasers

without mode locking. We obtained and analyzed different spectral types of probe amplification in an optically dense resonant medium. Even in the case of quasistochastic pulses of correlation time determined by the width of the laser spectrum, collective optical ringing was shown to accompany the propagation and amplification processes. In the experiments, a neon gas discharge containing a significant amount of neon metastable atoms (up to 10^{13} cm^{-3}) was used as an optically dense medium. Measurements were carried out at several neon transitions with the metastable level $2p^5 3s\ ^3P_2$ as a lower state. Particularly, for the strongest transition of wavelength 640.2 nm, the cooperative frequency $\omega_c/2\pi$ reached a value of 30 GHz, which was enough to fulfill the conditions of the strong-coupling regime. The widths of laser spectra greatly exceeded both homogeneous and inhomogeneous broadening of the spectral line, and took a value of about 200 GHz (FWHM), giving a correlation time (duration of a single spike in quasistochastic signal) of about 5 ps.

In contrast to Refs. [26–29], the present paper deals with smooth transform-limited pulses, since fundamentals of the phenomena have been considered earlier and arise from the coherent interaction between short pulses and a dense medium. Under the mode locking of dye lasers, our experimental parameters described above can be used for a quantitative analysis of the results obtained in this work, which are presented in dimensionless form. Moreover, as mentioned in the Introduction, collective optical ringing was already observed on the femtosecond time scale. Thus the results obtained have a direct relation to the study of the so-called "sharp-line limit" of the resonant light-matter interaction. In the framework of this limit, single-pulse propagation was considered in Refs. [16,37–39].

The numerical model is based on a solution of the nonlinear system of equations (4) and (5) describing the propagation of two intersected waves. In this system, the weak probe field $\Omega_1(t,z)$ is treated in the first order of perturbation theory, while the amplitude of the pump field $\Omega_0(t,z)$ can take arbitrary values. Nevertheless, since the optical ringing originates from the linear response of a medium under small changes in the population difference, we center our discussion on the case where the input area of the pump field takes values less than $\pi/2$. Thus, in the situation considered, the pump field does not invert a medium. Moreover, long ringing can be obtained after both probe and pump pulses, which provides an interaction of two fields much longer than the durations of the short input pulses.

Equation (5b) shows the influence of the pump field on the polarization component p_1 radiating in the direction of the probe Ω_1, whose propagation is described by Eq. (5a). The first term on the right-hand side of Eq. (5b) can be treated as forward scattering of the probe wave Ω_1 on the spatiotemporal oscillations of the population difference component $D_0(t,z)$, which is determined by transient collective ringing in the pump field Ω_0 [cf. Eq. (4c)]. The second term in Eq. (5b) can be described as scattering of the pump field Ω_0 on the nonstationary grating of the population difference $D_1(t,z)$ with wave vector $\Delta\mathbf{k}$, Eq. (3b), which originates from the spatiotemporal beating between pump and probe waves [cf. Eq. (5c)].

In the numerical simulations, the pump pulse at the input of the medium was chosen in the form (17) with pulse area $s_0 < \pi/2$ and spectral FWHM (18) $\gamma_{sp0}/\omega_c = 10.0$. To provide the maximum amplitude of pump-matter energy exchange D_0, which is determined by the amplitude of the ringing in $|\Omega_0|$, the pump central frequency was tuned exactly to the resonance, giving $\Delta_0 = 0$.

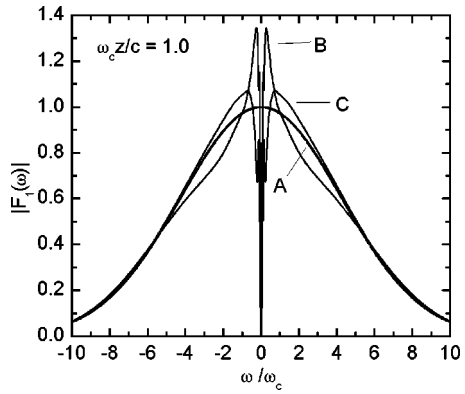

FIG. 3. Spectra of the probe field at the input (curve *A*) and output of the medium at probe-pump time delays $\omega_c\tau_0 = -0.5$ (curve *B*) and $\omega_c\tau_0 = 0.5$ (curve *C*); $\gamma_{1,2}/\omega_c = 10^{-3}$, FWHM of the probe and pump spectra $\gamma_{sp0}/\omega_c = \gamma_{sp1}/\omega_c = 10$, pump area $s_0 = 0.49\pi$, propagation distance $\omega_c z/c = 1.0$.

The input probe pulse was also chosen in the form (17) with introducing an additional time delay τ_0 ($\tau_0 < 0$ corresponds to the probe pulse preceding the pump). The probe has different values of γ_{sp1} and Δ_1, and an area s_1 much less than s_0 (the exact value of s_1 is not of importance due to the linearity of the equations with respect to the probe amplitude). The value of the small angle φ did not significantly affect the results and was set to 1°.

A typical result of numerical simulations for the case of exactly resonant broadband probe pulse (with parameters similar to those of the pump: $\gamma_{sp1}/\omega_c = 10.0, \Delta_1 = 0$) is presented in Fig. 3. It shows the spectra of the probe pulse $|F_1(\omega)|$ at the input of the medium and after the propagation for the cases of positive and negative time delays τ_0 between the probe and pump pulses. The characteristic peculiarity of the output spectra obtained consists in the fact that, in addition to the narrow absorption line, either a broad spectral doublet for $\tau_0 < 0$ or a dip for $\tau_0 > 0$ appears. Both of these spectral features appear close to the resonance. Besides that, at the wings of the spectra, there exist dumping for $\tau_0 < 0$ and amplification for $\tau_0 > 0$. Integration over the spectrum squared shows that such amplification or dumping at the spectral wings gives an increase or decrease, respectively, in the total energy of the pulse under propagation. This fact is a consequence of the existence of energy exchange between probe and pump, which interact by the ringing tails originating from the field of medium reemission.

The details of the probe-field transmission spectra (output to input spectrum ratios) in the vicinity of the resonance are presented in Figs. 4 and 5.

Figure 4 displays the spectra for the dimensionless length of the medium $\omega_c z/c = 1.0$ and different time delays τ_0 between probe and pump pulses. Figure 4(a) illustrates the transmission of the probe in the presence of the pump for the

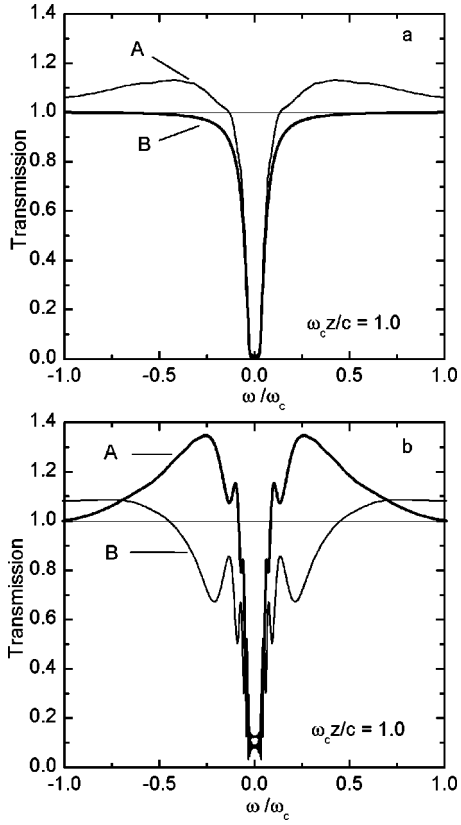

FIG. 4. Transmission spectra of the probe field in the vicinity of resonance for propagation distance $\omega_c z/c=1.0$ and different probe-pump time delays. (a) $\omega_c\tau_0=0$ (curve *A*) and linear transmission contour (curve *B*); (b) $\omega_c\tau_0=-0.5$ (curve *A*) and $\omega_c\tau_0=0.5$ (curve *B*); $\gamma_{1,2}/\omega_c=10^{-3}$, FWHM of the probe and pump spectra $\gamma_{sp0}/\omega_c=\gamma_{sp1}/\omega_c=10$, pump area $s_0=0.49\pi$.

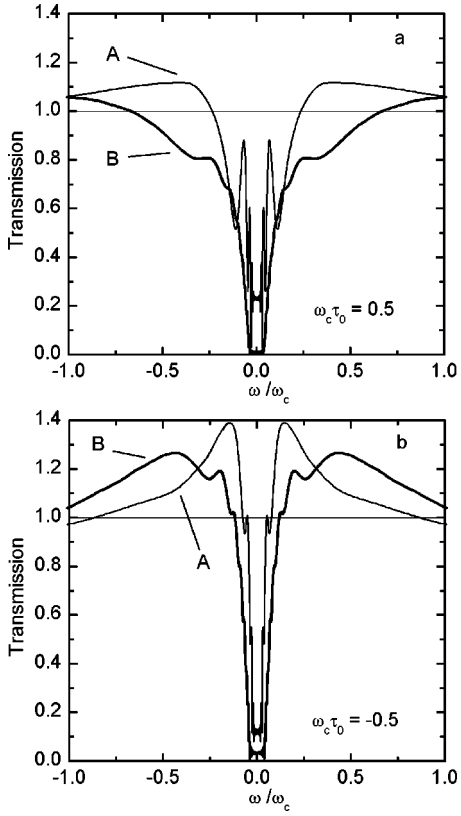

FIG. 5. Transmission spectra of the probe field in the vicinity of resonance for different propagation distances and probe-pump time delays. (a) $\omega_c\tau_0=0.5$ for $\omega_c z/c=0.5$ (curve *A*) and $\omega_c z/c=2.0$ (curve *B*); (b) $\omega_c\tau_0=-0.5$ for $\omega_c z/c=0.5$ (curve *A*) and $\omega_c z/c=2.0$ (curve *B*); $\gamma_{1,2}/\omega_c=10^{-3}$, FWHM of the probe and pump spectra $\gamma_{sp0}/\omega_c=\gamma_{sp1}/\omega_c=10$, pump area $s_0=0.49\pi$.

probe-pump time delay $\tau_0=0$ (curve *A*). It also shows the linear transmission contour in the absence of the pump pulse (curve *B*). In Fig. 4(b), the probe spectra with resonant doublet for $\omega_c\tau_0=-0.5$ and broad dip for $\omega_c\tau_0=0.5$ are shown, presenting the detailed description of Fig. 3. Comparison of Figs. 4(a) and 4(b) shows that the widths of both the spectral doublet and dip are greater than the width of the linear transmission contour, which corresponds to the incoherent absorption. Besides that, the broad features are not significant at $\tau_0=0$. The latter fact demonstrates the importance of the phase shift, which is introduced by the presence of the time delay between probe and pump pulses.

Figure 5 compares the probe transmission spectra for different propagation distances ($\omega_c z/c=0.5$ and 2.0). Figure 5(a) presents this comparison for the time delay $\tau_0>0$, whereas the comparison for $\tau_0<0$ is displayed in Fig. 5(b). These figures together with Fig. 4(b) for $\omega_c z/c=1.0$ show that the widths of the spectral dip and doublet significantly depend on the coordinate z and increase during the propagation. Such behavior of the spectral features in the probe field and the fact that its width exceeds the absorption-line width give evidence of the coherent optical ringing importance in the process of nonlinear interaction.

The temporal behavior of the probe pulse in the presence of the pump field is shown in Fig. 6 for $\tau_0<0$ (curves *A*) and $\tau_0>0$ (curves *B*). Here, the argument τ was shifted to $\tau-\tau_0$ for convenient comparison of these curves. The initial stage of the process $[\omega_c(\tau-\tau_0)<15]$ and the probe pulse at the input of the medium (curve *C*) are presented in Fig. 6(a). The most interesting contribution to the output signal is given by the coherent oscillating tail in the form of collective optical ringing, which is displayed in Fig. 6(b) $[\omega_c(\tau-\tau_0)>15]$. The temporal dynamics corresponds to the spectra displayed in Figs. 3 and 4(b). The dumped oscillations at the tail of the optical ringing (curve *B*) correspond to the broad dip in the probe spectrum, while the amplified oscillations (curve *A*) reflect the appearance of the spectral doublet.

It is important to stress that in the problem considered in Sec. III, due to the linearity of the interaction, no features corresponding to coherent optical ringing can be observed in the transmission spectrum, except for the usual incoherent absorption contour [cf. Fig. 4(a)]. In contrast, the results presented in this section show the existence of such spectral features under the nonlinear pump-probe interaction. The broadening of the coherent doublet and dip during the propagation is a consequence of the growing ringing frequency, which increases with an increase in the number of atoms participating in the collective interaction. In other words, the broadening of the coherent doublet and dip reflects the fact that wave packets of increasing detunings from the resonance

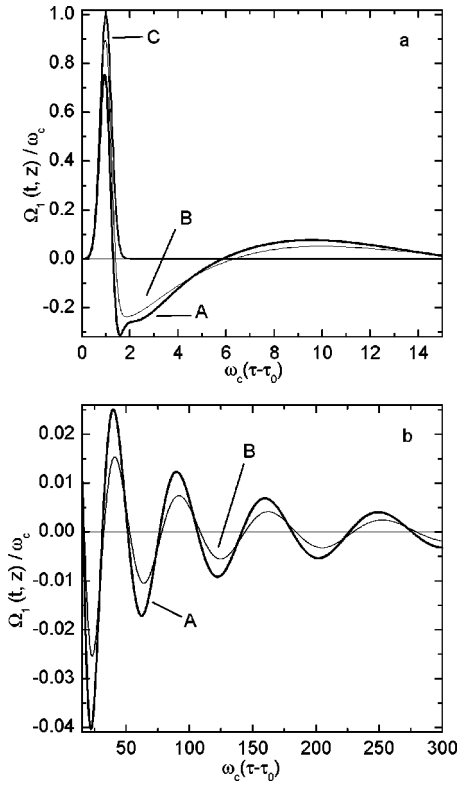

FIG. 6. Temporal behavior of the probe field for propagation distance $\omega_c z/c=1.0$ and probe-pump time delays $\omega_c\tau_0=-0.5$ (curves *A*) and $\omega_c\tau_0=0.5$ (curves *B*). (a) Initial stage of the process and the input pulse (curve *C*); (b) collective oscillating tail of the probe field; $\gamma_{1,2}/\omega_c=10^{-3}$, FWHM of the probe and pump spectra $\gamma_{sp0}/\omega_c=\gamma_{sp1}/\omega_c=10$, pump area $s_0=0.49\pi$. Corresponding spectra are shown in Figs. 3 and 4(b).

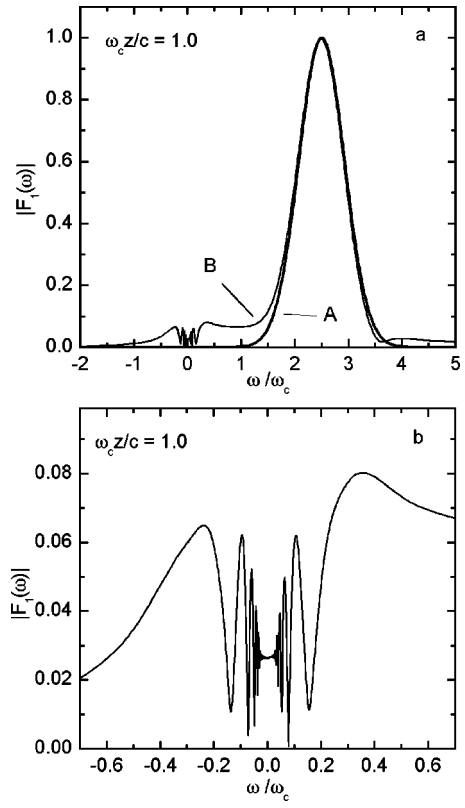

FIG. 7. Spectra of the narrow-band probe field for propagation distance $\omega_c z/c=1.0$. (a) Input (curve *A*) and output (curve *B*) spectra; (b) detailed output spectrum of (a) in the vicinity of resonance; $\gamma_{1,2}/\omega_c=10^{-3}$, FWHM of the probe input spectrum $\gamma_{sp1}/\omega_c=1.0$, FWHM of the pump input spectrum $\gamma_{sp0}/\omega_c=10$, pump area $s_0=0.49\pi$.

are involved in the creation of the ringing [see Eq. (16)] and hence in the dynamical interaction of two fields.

The relaxation rates γ_1 and γ_2 and the "optical density" $\alpha_0 z\equiv\omega_c^2 z/(\gamma_2 c)$ of a medium, which together describe the absorption contour, are not the characteristic parameters of the optical ringing, since they include incoherent parameters of a medium. The characteristic frequencies of the coherent collective oscillations (13) and (15) do not depend on the rates of incoherent relaxation and increase during the propagation. The width of the absorption contour also increases during the propagation. Nevertheless, since it is determined by the relaxation rate, it may be significantly smaller than the width of the coherent features.

The propagation of the probe pulse $\Omega_1(t,z)$ can be treated as a parametric process under the modulation of parameters in Eqs. (5) by the pump field. A detailed analysis shows that it is the first term on the right-hand side of Eq. (5b) with oscillating $D_0(t,z)$ that mainly contributes to the appearance of the near-resonant features presented in Figs. 4 and 5. Thus, the physical origin of these features is mainly related to the forward scattering of the probe wave Ω_1 on the collective spatiotemporal oscillations of the population difference component $D_0(t,z)$. It is the transient ringing in the pump field Ω_0 that is able to modulate $D_0(t,z)$ [cf. Eq. (4c)]. The near-resonant features described are determined by the frequencies of the parametric modulation. Moreover, the importance of a value of the phase shift between probe and pump fields, which is introduced by the time delay τ_0, substantiates the parametric treatment of the phenomenon.

Under the increase of the time delay between the probe and pump pulses $|\tau_0|$, the efficiency of modifications in the probe spectrum decreases. Under the decrease of the pump area s_0, the amplitude of the spectral features also decreases, but its characteristic frequencies do not significantly vary. The latter fact is different from the case of the strong-field stationary Mollow-Boyd contour [1], where characteristic frequencies depend on the amplitude of the pump field, but not on the length and density of a medium.

Parametric peculiarities of the process are also illustrated by the numerical simulations with the narrow-band probe pulse shifted from the resonance. The pump parameters were chosen to be the same as described above. The parameters of the probe pulse had the values $\gamma_{sp1}/\omega_c=1.0$, $\Delta_1/\omega_c=2.5$, and $\tau_0=0$. The results of the simulations are presented in Figs. 7 and 8.

Figure 7 displays the input and output spectra of the narrow-band probe pulse for the case of propagation distance $\omega_c z/c=1.0$. The figure shows that a characteristic doubletlike feature in the vicinity of the resonance appears even in the case where significant near-resonant components are absent in the input spectrum. Therefore, the probe output spectrum

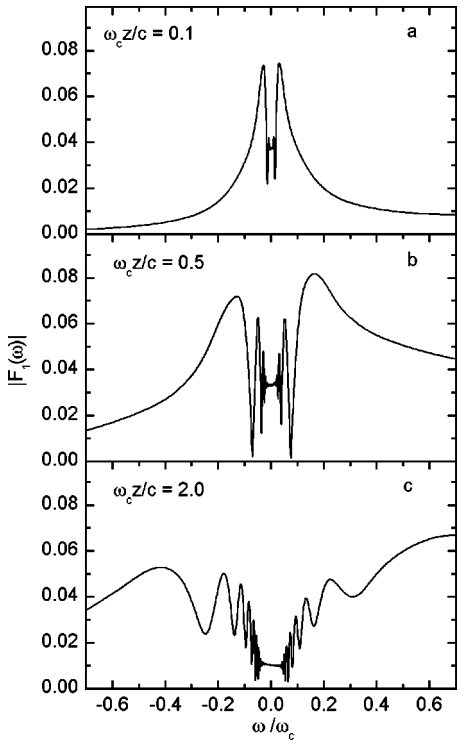

FIG. 8. Output spectra of the narrow-band probe field in the vicinity of resonance for different propagation distances. (a) $\omega_c z/c=0.1$; (b) $\omega_c z/c=0.5$; (c) $\omega_c z/c=2.0$; corresponding input spectrum is shown in Fig. 7(a); $\gamma_{1,2}/\omega_c=10^{-3}$, FWHM of the probe input spectra $\gamma_{sp1}/\omega_c=1.0$, FWHM of the pump input spectra $\gamma_{sp0}/\omega_c=10$, pump area $s_0=0.49\pi$.

does not originate only from amplification of existing components, but also from the generation of new ones via modulation of medium parameters by the pump field. Moreover, one can see the appearance of a specific oscillating structure in the output spectrum.

The specific structure becomes richer and broader under an increase of the propagation distance, which is demonstrated by Fig. 8 for $\omega_c z/c=0.1$, 0.5, and 2.0 and by Fig. 7(b) for $\omega_c z/c=1.0$. The total width of the amplification region also increases. The spectral structure can be also seen in Figs. 4(b) and 5 for the broadband resonant probe field, though being more strongly suppressed by the absorption.

Such oscillating structure, which depends on the density and length of the medium, is characteristic for spectral measurements in the presence of optical ringing. It was also obtained numerically [37,39] and experimentally [38] under an investigation of the spectral properties of the self-induced transparency of single ultrashort pulses. The structure was shown to appear if both nonlinear solution (particularly, solitons) and optical ringing are significant in the output signal. The experiments [38] were carried out in a potassium vapor cell (the interaction length was 2 mm) at the $D1$ spectral line; the pulse duration was equal to 415 fs, and the vapor density reached a value of 10^{14} cm^{-3}, which corresponds to the cooperative frequency $\omega_c/2\pi$ of about 90 GHz. The width of the oscillating structure obtained in these experiments was about 40 GHz.

This kind of spectral feature enables an analysis of the free-space optical ringing by spectral methods. The features can be obtained even in the case of linear interference with participation of the optical ringing in an optically dense resonant medium. Particularly in the case where a short laser pulse, which has passed through a medium, interferes at the output with a similar pulse, which has traveled the same distance in vacuum and having additional arbitrary time delay τ_1. Using the integrand of Eq. (8) and the expression for the wave vector $k(\omega)$, Eq. (9), in the limit of $\gamma_2=0$, the summed spectrum of two fields $F_+(\omega)$ can be written as:

$$F_+(\omega)=F_{\rm in}(\omega)e^{-i\omega z/c+i\omega_c^2 z/(\omega c)}+F_{\rm in}(\omega)e^{-i\omega z/c-i\omega\tau_1}.$$

Then, the absolute value of the spectrum is given by

$$|F_+(\omega)|=2|F_{\rm in}(\omega)|\left|\cos\left[\frac{1}{2}\left(\frac{\omega_c^2 z}{\omega c}+\omega\tau_1\right)\right]\right|. \qquad (19)$$

In the region of the parameters, where the phase brought by the medium (the first term) is greater than the phase brought by the time delay (the second term), Eq. (19) gives spectral oscillations which are qualitatively similar to the ones presented in Figs. 7 and 8. This phase is also responsible for the appearance of optical ringing. The function (19) has an infinite number of extrema, whose frequency increases when the detuning approaches zero. If the relaxation is taken into account, the number of oscillations is limited, and the oscillations are absent in the region of the absorption.

So the characteristic spectral oscillations can be made visible in the spectrum of optical ringing by a change of the phase in the linear solution (8), particularly, due to the presence of another pulse. Moreover, it is the nonlinearity of the medium that can bring a specific phase shift to the interaction process. In Refs. [37–39], the appearance of spectral oscillations can be treated as interference of the quasilinear optical ringing with nonlinear solutions (particularly, solitons), which have specific time delays due to the nonlinearity of the interaction. In the case of the pump-probe interaction presented here, it is the pump pulse that changes the propagation conditions of the probe. Unlike the works in [37–39], in our study, the spectral structure is obtained in the weak amplified probe field intersected with the pump. The input pump pulse may have an area less than $\pi/2$. Moreover, contrary to Refs. [37–39], the amplitude of the input spectrum may be negligibly small in the vicinity of the resonance and hence produces no spectral background for the observation of specific output features.

Thus, the pulsed pump-probe interaction may be significantly affected by the generation of the optical ringing signal, and it is the dynamic peculiarities of the collective reemission of a resonant medium that play a key role in the processes considered. The efficiency of collective dynamics increases with an increase of the propagation distance and density of the medium. On the other hand, the efficiency of the two-beam interaction may be limited by those parameters, since under an increase of the oscillation frequency, the amplitude of the field and population difference modulation decreases. This fact reduces parametric coupling of two fields and leads to their linear propagation.

V. CONCLUSIONS

We have presented a theoretical investigation of the transient pump-probe interaction between short laser pulses in an optically dense resonant medium. A high value of the field-matter coupling coefficient, which is determined by the medium density, and relatively weak intensities of the external pulses give rise to the collective oscillating response of a medium in the form of coherent optical ringing. The long ringing response, which exists in a time scale of the order of relaxation times, provides an interaction of light fields with a medium much longer than the duration of short input pulses. The analytical expression obtained gives the relation between the temporal behavior of the ringing and the spectrum of the input laser pulse. It describes the limitation of oscillation-frequency growth and the reduction of the ringing efficiency under the increasing detuning from the resonance. Moreover, it supports the treatment of optical ringing as successive coherent beats between light wave packets of different group velocities, which in the case of a dense coherent medium can significantly vary over the broad spectrum of the pulse.

The collective dynamics of the medium is shown to essentially affect the propagation and amplification of a probe pulse under its nonlinear interaction with the pump field. In the transmission spectrum, either a characteristic doubletlike structure or a broad dip appears in the vicinity of the resonant frequency, depending on the time delay between the probe and pump pulses. In accordance with the spatiotemporal behavior of the optical ringing, the widths of the coherent spectral features increase with an increase of the medium density. Besides that, broadening of the coherent doublet and dip during the propagation reflects the fact that wave packets of increasing detunings from the resonance are involved in the interaction of two fields. Modification of the spectrum comes from modulation of the probe and medium parameters by the pump pulse, especially from oscillations of population difference due to the existence of optical ringing in the pump field. Because of the modulation, spectral features can appear not only in the spectral region of the existing input components, but also in the region where the spectrum of the input probe pulse is negligibly small.

Thus, contrary to the stationary strong-field effects, the density- and coordinate-dependent transmission spectra of the probe manifest the importance of collective oscillations and cannot be obtained in the framework of the single-atom model. We would like to point out that the doublet spectral features and their sensitivity to the atomic density (particularly, the existence of a threshold) qualitatively resemble the analogous properties of the "spectrum condensation," or "self-frequency-locking," phenomenon [26,33].

ACKNOWLEDGMENT

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