Resonant Raman amplification of ultrashort pulses in a *V*-type medium

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(Received 10 December 1998)

Propagation of a pair of ultrashort coherent pulses in a *V*-type medium is studied. Both pulses are supposed to be resonant with corresponding atomic transitions. It is shown that the resonant character of the atom-field interaction yields ''pulse locking,'' in which two pulses propagate with equal group velocities. Additionally, in the presence of Raman inversion, the system exhibits resonantly enhanced gain for the probe pulse. These two effects add together to decrease the pump laser threshold intensity by orders of magnitude in comparison with conventional (far-off-resonant) Raman laser schemes. [S1050-2947(99)06304-0]

PACS number(s): 42.55.Ye, 42.50.Gy, 84.30.Ng

I. INTRODUCTION

Creation of new sources of coherent UV and x-ray radiation is an important and challenging task of modern optics. Ordinary lasers operating with population inversion have, among other features, one crucial obstacle to high-frequency generation: the spontaneous decay rate grows proportionally to the transition frequency cubed (ω^3); therefore creation of population inversion on a high-frequency transition is problematic, and conventional lasers with inversion require rapid excitation rates in order to operate in the high-frequency domain.

On the other hand, two mechanisms of coherent light amplification are known which do not require population inversion directly on the amplifying transition: Raman gain [1] and *gain without inversion* (commonly referred to as LWI) [2]. Although population inversion is no longer needed in these two cases, practical implementation of both examples in high-frequency generation is not free from technical difficulties.

Both Raman and LWI gain require strong pumping (or driving) of an active medium with external coherent laser fields. Of course, the closer the fields are tuned to the corresponding atomic resonances, the stronger the atom-field interaction is, and, typically, the higher Raman and LWI gain are. On the other hand, approaching atomic resonances leads to resonantly enhanced absorption and dispersion of the pumping fields, and this imposes a limit on the density of active atoms and, therefore, on the gain of the laser transition.

An additional difficulty involves the inhomogeneous broadening (Doppler broadening, for the case of gaseous medium). In the frequency up-conversion regime, the pumping (or driving) field has a lower frequency than that of the laser field. Typically, both LWI and Raman-gain effects involve some specific detuning between the two fields. If the frequency of the laser field is higher than that of the driving field, then relative detuning between the fields is different for different groups of atoms of an inhomogeneously broadened medium, resulting in a reduced net gain.

One may think that both the above-mentioned obstacles can be overcome by pumping the medium with a short pulse. For example, if the duration of the pulse is chosen to be smaller than the relaxation times in the system, then such a pulse is not attenuated while propagating through the medium. Furthermore, if the pulse is so short that its spectral width exceeds the linewidth of the inhomogeneously broadened atomic transition, then the problem of inhomogeneous broadening is mitigated. The only remaining question is how the two mechanisms of inversionless amplification—Raman and LWI gain—will work in such a pulsed regime.

In fact, multilevel systems driven by short optical pulses have been a subject of extensive experimental study. The basis of LWI in a Lambda-type medium—the effect of electromagnetically induced transparency (EIT)—has been demonstrated using short pulses [3]. Since some fixed pump pulse energy has to be spent to convert the medium into a coherent state necessary for EIT, and field strength has to overcome effects of inhomogeneous broadening, pump pulses in those experiments were strong (a power density on the order of 10^7 W/cm^2).

Raman gain has been very well studied in the pulsed regime. The reason for working with short pulses in this case is simple: To avoid strong absorption and dispersion of the pumping laser field, it is conventionally taken to be detuned far from the corresponding atomic resonance frequency. Since, at such a detuning, atom—pumping-field interaction is weak, the threshold intensity of the pump field is so high that it is available only in the form of a short pulse. Typical pump intensities in these kinds of experiments are on the order of 10^8 W/cm^2 and higher [1]. In the present paper we will pump the system resonantly, and therefore will be dealing with much weaker pulses: The pump pulse area will be on the order of several π (meaning that the pumped transition undergoes small number of Rabi cycles), while for the above-mentioned experiments it was thousands of π or more.

There are also several theoretical works on inversionless amplification with short pulses. In Ref. [4], the gain of the weak probe pulse relied on the initial preparation of active atoms in some specific coherent superposition of atomic bare states. In Ref. [5] only part of the pulse in a driven medium was shown to be amplified, but then the rest of it was subjected to absorption. In Ref. [6] the instability of the weak probe pulse in a *V*-type medium resonantly driven by short

2π pulse was studied. Gain for the probe was predicted, and it was attributed to the temporal ''inversion window'' opened on the probed transition by the 2π driving pulse. In a recent paper [7], the far-off-resonant Raman gain was revised, and an anti-Stokes Raman laser without inversion has been proposed. It has been found that a multilevel system may yield gain independently of the initial distribution of population among atomic states, provided that incoherent pumping mechanisms are strong enough to redistribute atomic populations considerably within duration of the coherent pump pulse. This, of course, requires either very strong sources of incoherent pumping, or a very long pulse of the coherent pumping field.

Pursuing practical aspects of efficient frequency upconversion, in the present paper we will study in detail coherent pulse amplification in the *V*-type medium pumped by a short laser pulse. The duration of the pump pulse is supposed to be shorter than all relaxation times in the medium. Both pump and probe pulses are tuned close to corresponding atomic resonance frequencies. Atoms constituting the medium are not supposed to be initially prepared in a coherent superposition of bare atomic states. We will show analytically and numerically that, in the system under consideration, linear *gain without inversion* is impossible. At the same time, the resonant character of the atom-field interaction has two practically important consequences: In the presence of Raman inversion the probe pulse can experience strong resonantly enhanced two-photon gain. Furthermore, the system yields a ''pulse locking,'' i.e., the probe and pump pulses propagate together, with the same group velocity, and without a substantial change of shape while propagating (this effect is similar to the propagation of simultons in a three-level medium [8]). Since the pulses are chosen to have a duration considerably shorter than all relaxation times in the medium, the gain in the system under consideration is not accompanied by devastating absorption of the pump pulse. This is in contrast to the case of long pulses, where a transfer of energy from the resonant pump pulse into spontaneously emitted photons limits the propagation distance and, consequently, the net gain for the probe. Hence the gain for the probe pulse is higher than in conventional, far-off-resonant, Raman laser schemes, and the threshold power of the pump source can be reduced by orders of magnitude compared to those schemes.

In the weak probe pulse case, it is possible to study the system analytically. In this limit we will derive the formula for the probe energy gain; this formula is valid for the arbitrary shape of the pulses. When the amplified probe pulse acquires an appreciable value, analytical study becomes difficult, and we study this regime numerically. We will demonstrate that, in this case, the two-photon gain does not decrease. Conversely, when the intensity of the probe pulse becomes comparable to or even exceeds that of the pump pulse, the amplification rate of the probe increases. Probe pulse amplification ends only with the vanishing of the pump pulse, i.e., with a complete exchange of pump photons into probe photons.

The physics beyond the resonant two-photon amplification and ''pulse locking'' effects will be discussed in detail. It will be shown, that, in spite of the fact that the 2π pulse of the pump opens a temporary ''inversion window'' on the probed transition, this population inversion does not contribute to the probe gain at all, and all gain comes from the resonant two-photon Raman-like process.

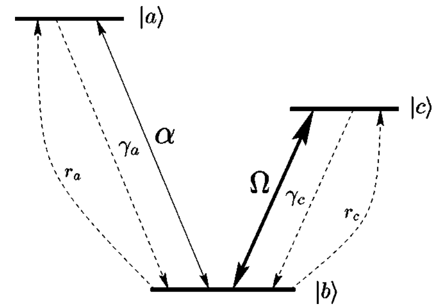

FIG. 1. Level scheme of the *V*-type medium showing coherent pump (Ω) and laser (α) fields, decays γ_a and γ_c, and incoherent pump rates r_a and r_c.

We will usually take the pump pulse in the form of the 2π soliton [9], because such a pulse is distortion free, and yields the most clear pattern of the probe amplification. If the pump pulse is initially prepared in a form different from the 2π soliton, then the pulse dynamics becomes more complicated, showing an interplay between effects of self-induced transparency for the pump pulse and two-photon gain for the probe. This situation will be discussed, and we will consider a specific example of the pump pulse with area equal to 4π.

II. DESCRIPTION OF THE MODEL

Consider a *V*-type three-level system with the ground state $|b\rangle$ and excited states $|a\rangle$ and $|c\rangle$, as shown in Fig. 1. Transition $|b\rangle \leftrightarrow |c\rangle$ is driven by a short pulse with a Rabi frequency $\Omega(z,t)$. A probe (or laser) pulse with Rabi frequency $\alpha(z,t)$ is applied to the transition $|b\rangle \leftrightarrow |a\rangle$. We suppose the carrier frequency of the both pulses to be tuned to exact resonance with the corresponding transitions and the pulses to be free of phase modulation. Then, in the slowly varying envelope approximation, the temporal and spatial evolution of the pulse envelopes is governed by wave equations

$$\left[\frac{\partial}{\partial z}+\frac{n_\Omega}{c}\frac{\partial}{\partial t}\right]\Omega(z,t)=i\kappa_\Omega\rho_{cb}, \tag{1}$$

$$\left[\frac{\partial}{\partial z}+\frac{n_\alpha}{c}\frac{\partial}{\partial t}\right]\alpha(z,t)=i\kappa_\alpha\rho_{ab}, \tag{2}$$

with propagation constants

$$\kappa_\Omega=k_\Omega\wp_{cb}^2N/\epsilon_0\hbar n_\Omega,$$

$$\kappa_\alpha=k_\alpha\wp_{ab}^2N/\epsilon_0\hbar n_\alpha. \tag{3}$$

ρ_{ab} and ρ_{cb} are off-diagonal density matrix elements in the rotating frame, k_Ω and k_α are wave vectors of the pump and probe fields in vacuum, $\wp_{cb}$ and $\wp_{ab}$ are dipole matrix elements of the transitions $|c\rangle \leftrightarrow |b\rangle$ and $|a\rangle \leftrightarrow |b\rangle$ (supposed to be real, for simplicity), and N is the density of resonant atoms. In Eqs. (1) and (2), n_Ω and n_α stand for the host refractive index at frequencies of the pump and probe fields, respectively. Slowly varying amplitudes of the pump and probe electric fields, $\mathcal{E}_\Omega$ and $\mathcal{E}_\alpha$, are related to the Rabi frequencies Ω and α according to

$$\mathcal{E}_\Omega = \frac{\hbar\Omega}{\wp_{cb}}, \quad \mathcal{E}_\alpha = \frac{\hbar\alpha}{\wp_{ab}}. \tag{4}$$

We suppose the medium to be homogeneously broadened. The semiclassical density matrix equations of motion under the rotating wave approximation are

$$\frac{\partial}{\partial t}\rho_{ab} = -\Gamma_{ab}\rho_{ab} - \frac{i}{2}\alpha(\rho_{aa}-\rho_{bb}) - \frac{i}{2}\Omega\rho_{ac}, \tag{5}$$

$$\frac{\partial}{\partial t}\rho_{ac} = -\Gamma_{ac}\rho_{ac} + \frac{i}{2}\alpha\rho_{bc} - \frac{i}{2}\Omega^*\rho_{ab}, \tag{6}$$

$$\frac{\partial}{\partial t}\rho_{cb} = -\Gamma_{cb}\rho_{cb} - \frac{i}{2}\Omega(\rho_{cc}-\rho_{bb}) - \frac{i}{2}\alpha\rho_{ca}, \tag{7}$$

$$\frac{\partial}{\partial t}\rho_{aa} = -\gamma_a\rho_{aa} + r_a\rho_{bb} - \frac{i}{2}(\alpha^*\rho_{ab} - \alpha\rho_{ba}), \tag{8}$$

$$\frac{\partial}{\partial t}\rho_{bb} = \gamma_a\rho_{aa} + \gamma_c\rho_{cc} - (r_a + r_c)\rho_{bb} + \frac{i}{2}(\alpha^*\rho_{ab} - \alpha\rho_{ba}) + \frac{i}{2}(\Omega^*\rho_{cb} - \Omega\rho_{bc}), \tag{9}$$

$$\frac{\partial}{\partial t}\rho_{cc} = -\gamma_c\rho_{cc} + r_c\rho_{bb} - \frac{i}{2}(\Omega^*\rho_{cb} - \Omega\rho_{bc}), \tag{10}$$

where Γ_{ab}, Γ_{ac}, and Γ_{cb} are dephasing rates, and γ_a, γ_c, r_c, and r_a are the decay and pumping rates indicated in Fig. 1. The analysis is referred to the case of the closed V system, so that one of Eqs. (8)–(10) is to be replaced by the normalization condition

$$\rho_{aa} + \rho_{bb} + \rho_{cc} = 1. \tag{11}$$

In the above equations, $\Omega = \Omega^*$ and $\alpha = \alpha^*$. (The fields are resonant and therefore can be chosen real.) We will suppose that durations of the both pulses are much less than all incoherent decay and pumping rates in the system.

Before the arrival of the pulses, the medium is under the effect of continuous incoherent pumping, and thereby is prepared in the following manner:

$$\rho_{bb}^0 \equiv \rho_{bb}(t\to-\infty) = \frac{1}{1 + r_a/\gamma_a + r_c/\gamma_c},$$
$$\rho_{aa}^0 \equiv \rho_{aa}(t\to-\infty) = \frac{r_a/\gamma_a}{1 + r_a/\gamma_a + r_c/\gamma_c}, \tag{12}$$
$$\rho_{cc}^0 \equiv \rho_{cc}(t\to-\infty) = \frac{r_c/\gamma_c}{1 + r_a/\gamma_a + r_c/\gamma_c}.$$

All the coherences (i.e., off-diagonal elements of the density matrix) are initially zero, so that *the system is not initially prepared in a coherent state*. We will consider the case when initially there is no population inversion on the laser transition: $\rho_{aa}^0 < \rho_{bb}^0$. If, in addition to that, there is an inversion on a two-photon transition $|a\rangle \to |b\rangle \to |c\rangle$, i.e., $\rho_{aa}^0 > \rho_{cc}^0$, we say that the system is prepared in a state with Raman inversion. In the opposite case of $\rho_{aa}^0 < \rho_{cc}^0$ there is no inversion of any kind.

III. ANALYSIS IN THE WEAK PROBE FIELD APPROXIMATION

A. Evolution of the pump pulse

With an eye toward a partial analytical analysis, we suppose that the pulses are so short that all incoherent terms in the density-matrix equations (10) can be neglected. Suppose also that the probe pulse is weak, so that it does not excite any appreciable population to the state $|a\rangle$, and *a linear approximation in* α is applicable. In this limit, the probe does not change atomic populations and has a negligible effect on the dynamics of the pump pulse. Consequently, the problem of the pump dynamics is reduced to that of a pulse propagating in a resonant two-level medium [9]. The equations for ρ_{cb}, ρ_{cc}, and ρ_{bb} decouple from the other density matrix equations:

$$\frac{\partial}{\partial t}\rho_{cb} = -i\frac{\Omega}{2}(\rho_{cc}-\rho_{bb}),$$
$$\frac{\partial}{\partial t}(\rho_{cc}-\rho_{bb}) = -2i\Omega\rho_{cb}. \tag{13}$$

Since the pump field is resonant, ρ_{cb} is always purely imaginary. The system of equations (13) has a conservation law—the length of the Bloch vector for the two-level subsystem $|b\rangle$-$|c\rangle$ remains constant:

$$(\rho_{cc}-\rho_{bb})^2 - (2\rho_{cb})^2 = (\rho_{cc}^0 - \rho_{bb}^0)^2. \tag{14}$$

This allows us to solve Eq. (13) in terms of the Bloch angle θ:

$$\rho_{cc} - \rho_{bb} = (\rho_{cc}^0 - \rho_{bb}^0)\cos\theta, \tag{15}$$

$$\rho_{cb} = -\frac{i}{2}(\rho_{cc}^0 - \rho_{bb}^0)\sin\theta, \tag{16}$$

where

$$\theta(t,z) = \int_{-\infty}^{t} dt'\,\Omega(t',z), \tag{17}$$

so that $\theta(t=+\infty,\ z) = \Theta(z)$ is total area of the pump pulse. Substituting Eq. (16) into Eq. (1), we find a closed equation for dynamics of the Bloch angle (and, therefore, of the pump pulse) in the form

$$\left[\frac{\partial^2}{\partial z\,\partial t} + \frac{n_\Omega}{c}\frac{\partial^2}{\partial t^2}\right]\theta = \frac{\kappa_\Omega}{2}(\rho_{cc}^0 - \rho_{bb}^0)\sin\theta. \tag{18}$$

B. Evolution of the probe pulse

As follows from Eq. (2), the probe pulse is driven by the off-diagonal element ρ_{ab} of the density matrix. The latter depends on the evolution of the pump pulse via ρ_{bb} and two-photon polarization ρ_{ac}. Note again that the populations do not depend on the dynamics of the probe pulse (the linear approximation in α). Therefore ρ_{aa} is equal to ρ_{aa}^0 and does not change in time. Upon introducing new variables

$$R=\rho_{ab}+\rho_{ac},$$

$$Q=\rho_{ab}-\rho_{ac},$$

we may rewrite Eqs. (5) and (6) in the following ways:

$$\dot{R}=-i\frac{\alpha}{2}(\rho_{aa}-\rho_{bb})+i\frac{\alpha}{2}\rho_{bc}-i\frac{\Omega}{2}R,$$
$$\dot{Q}=-i\frac{\alpha}{2}(\rho_{aa}-\rho_{bb})-i\frac{\alpha}{2}\rho_{bc}+i\frac{\Omega}{2}Q. \tag{19}$$

Then the formal solutions for R and Q read

$$R=\frac{i}{2}\exp(-i\theta/2)\times\int_{-\infty}^{t}dt'\,\alpha[-(\rho_{aa}-\rho_{bb})+\rho_{bc}]\exp(i\theta/2), \tag{20}$$

$$Q=-\frac{i}{2}\exp(i\theta/2)\times\int_{-\infty}^{t}dt'\,\alpha[(\rho_{aa}-\rho_{bb})+\rho_{bc}]\exp(-i\theta/2),$$

where the Bloch angle θ is defined in Eq. (17). For the off-diagonal density-matrix element ρ_{ab} we find

$$\rho_{ab}=-\frac{i}{2}\mathrm{Re}\left\{\exp(-i\theta/2)\times\int_{-\infty}^{t}dt'\,\alpha[(\rho_{aa}-\rho_{bb})-\rho_{bc}]\exp(i\theta/2)\right\}. \tag{21}$$

Using normalization condition (11) and Eq. (15), one can obtain an expression for population difference $(\rho_{bb}-\rho_{aa})$ in the form

$$\rho_{bb}-\rho_{aa}=(\rho_{cc}^0-\rho_{aa}^0)-(\rho_{cc}^0-\rho_{bb}^0)\cos^2(\theta/2). \tag{22}$$

Substituting this and Eq. (16) into Eq. (21) yields

$$\rho_{ab}=-\frac{i}{2}(\rho_{aa}^0-\rho_{bb}^0)\cos(\theta/2)\int_{-\infty}^{t}dt'\,\alpha\cos(\theta/2)-\frac{i}{2}(\rho_{aa}^0-\rho_{cc}^0)\sin(\theta/2)\int_{-\infty}^{t}dt'\,\alpha\sin(\theta/2). \tag{23}$$

Finally, combining Eqs. (23) and (2), we find an equation governing evolution of the probe pulse:

$$\left[\frac{\partial}{\partial z}+\frac{n_\alpha}{c}\frac{\partial}{\partial t}\right]\alpha=\frac{\kappa_\alpha}{2}\times\left[(\rho_{aa}^0-\rho_{bb}^0)\cos(\theta/2)\int_{-\infty}^{t}dt\,\alpha\cos(\theta/2)+(\rho_{aa}^0-\rho_{cc}^0)\sin(\theta/2)\int_{-\infty}^{t}dt'\,\alpha\sin(\theta/2)\right]. \tag{24}$$

Define the total energy of the probe pulse:

$$J_\alpha(z)\equiv\frac{\varepsilon_0 c n_\alpha^2}{2}\int_{-\infty}^{+\infty}dt|\mathcal{E}_\alpha|^2(z,t)=\frac{\varepsilon_0 c\hbar^2 n_\alpha^2}{2\wp_{ab}^2}\int_{-\infty}^{+\infty}dt|\alpha|^2(z,t); \tag{25}$$

then, integrating Eq. (24) over time, we find

$$\frac{\partial}{\partial z}J_\alpha(z)=\frac{N\hbar\omega_{ab}}{4}\times\left\{(\rho_{aa}^0-\rho_{bb}^0)\left[\int_{-\infty}^{+\infty}dt\,\alpha\cos(\theta/2)\right]^2+(\rho_{aa}^0-\rho_{cc}^0)\left[\int_{-\infty}^{+\infty}dt\,\alpha\sin(\theta/2)\right]^2\right\}. \tag{26}$$

Equation (26) is the main analytical result of the present paper. It describes the evolution of the probe pulse energy for *arbitrary* shapes of the two pulses as long as the probe pulse remains weak. The first term on the right-hand side of Eq. (26) is proportional to the bare population inversion $(\rho_{aa}^0-\rho_{bb}^0)$ and, due to our initial assumption that this inversion is negative, corresponds to the one-photon absorption of the probe on the transition $|a\rangle\leftrightarrow|b\rangle$. This absorption is, however, modified by the presence of the pump pulse, and, under certain pulse shapes [obeying $\int_{-\infty}^{+\infty}dt\,\alpha\cos(\theta/2)=0$], may be canceled. The second term is proportional to the Raman inversion between the two upper states $|a\rangle$ and $|c\rangle$, $(\rho_{aa}^0-\rho_{cc}^0)$, and, if this inversion is positive, corresponds to the two-photon gain of the probe pulse. One can see that, at any point z of the propagation, one of the two kinds of inversion is necessary for gain on the probe transition. This proves that, in the system under consideration, *linear LWI gain with ultrashort pulses is impossible*.

Note that Eq. (26) describes the evolution of the *total* energy of the probe pulse. Therefore, even in the absence of any kind of inversion, some part of the pulse can experience gain, but then the remaining part will experience absorption, so that the total probe pulse energy will typically decrease (as in Ref. [5]). It can, however, remain constant under some specific pulse shapes, as will be shown below. Additionally, Eq. (26) does not tell anything about the evolution of the probe pulse envelope. For instance, if Raman inversion is prepared in the system, and, according to Eq. (26), the probe pulse experiences energy gain, it is not *a priori* clear that the pulse would preserve its shape: it could spread out due to resonantly enhanced dispersion, decompose into weaker and shorter pulses, etc. Of course, an increase of the total pulse energy in such a case is of no practical value.

To investigate the dynamics of the probe envelope, Eq. (24) has to be solved together with Eq. (18). We will show below that, if the atoms are initially prepared in the ground state ($\rho_{bb}=1$, and the Raman inversion is equal to zero), then special matching of shapes of the pump and probe pulses leads to *full absorption and distortion cancellation* for the probe. If launched into the medium in such a matched form, the two pulses propagate without changing shapes. Furthermore, we will show that even if the pulses do not meet the matching condition at the entrance to the medium, they quickly evolve to a matched state after a short transient process. The presence of small Raman inversion in the medium does not disturb the shape-preserving propagation of the two pulses: the time scale of the transient process is considerably faster than that of the two-photon amplification process.

C. Stationary pulse propagation

In order to obtain a clear picture of pulse dynamics in the system under consideration, one can separate the effect of linear absorption cancellation from the two-photon Raman amplification. To do so, first consider the case when the medium is not excited, $\rho_{aa}^0=\rho_{cc}^0=0$, so that Raman amplification vanishes. Further, any pulse with an area greater than π in a resonant two-level medium breaks up into a sequence of the 2π soliton pulses after a short transient process [9]. Then, if relaxation in the medium is negligible, each of the 2π pulses propagates through the medium without any attenuation or reshaping. For a better understanding of the probe dynamics in the pumped medium under consideration, we want the pump pulse to behave in the simplest possible way. Therefore, in our following considerations, we choose the pump pulse to be initially injected into the medium in the form of the 2π soliton of Hahn and McCall. If the pump pulse is prepared in a form different from the 2π soliton, the situation becomes more complex, and we will postpone a discussion of such a case until Sec. V.

The probe pulse is supposed to be weak, and its area to be much less than π. It is known, that, in a resonant two-level medium, such a pulse would take an oscillatory form and rapidly spread out [10]. In the presence of the pump pulse, the situation is completely different. We performed numerical simulations showing that the pump pulse, if launched into the medium in the form of the 2π soliton, causes the probe pulse to attain stationary bell-shaped form, and then both pulses propagate with stationary shapes and equal group velocities (Fig. 2). This effect of ''pulse locking'' takes place independently of the initial shape of the probe pulse.

Under the assumption of stationary pulse shapes, these shapes can be found analytically. The problem reduces to solving Eqs. (18) and (24) for the probe envelope α and the Bloch angle θ, in terms of a single dimensionless variable $u=(t-z/v)/\tau_\Omega$ (v is a group velocity of the both pulses, τ_Ω is duration of the pump pulse)

$$\frac{d^2\theta}{du^2}=\frac{\kappa_\Omega}{2}\left(\frac{1}{v}-\frac{n_\Omega}{c}\right)^{-1}\sin(\theta(u)), \tag{27}$$

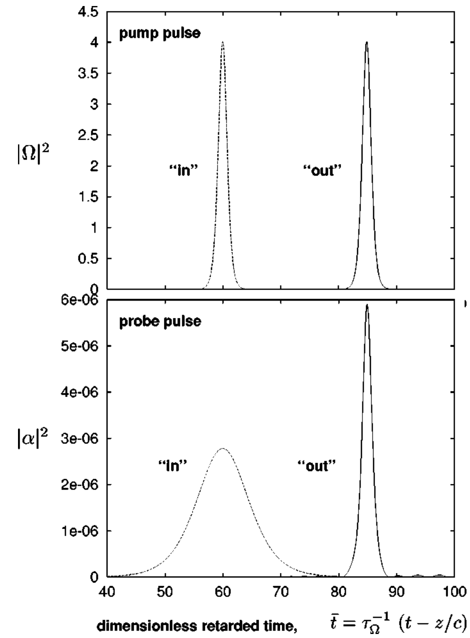

FIG. 2. Input and output intensity profiles of the pump and probe pulses as functions of dimensionless retarded time. The medium is initially prepared in the ground state $|b\rangle$. The total dimensionless propagation distance $\overline{z}\equiv\kappa_\Omega\tau_\Omega z=50$. At the entrance to the medium, the pump pulse has a 2π soliton shape [Eq. (29)]; the input probe pulse is intentionally chosen to be different from the ''locked'' shape [Eq. (36)].

$$\frac{d\alpha}{du}=\frac{\kappa_\alpha}{2}\left(\frac{1}{v}-\frac{n_\alpha}{c}\right)^{-1}\cos(\theta(u)/2)\times\int_{-\infty}^{u}du'\,\alpha(u')\cos(\theta(u')/2). \tag{28}$$

Writing Eqs. (27) and (28), we explicitly used the fact that atoms are initially prepared in the ground state $|b\rangle$, so that $\rho_{bb}^0=1$, and $\rho_{aa}^0=\rho_{cc}^0=0$.

Since the probe pulse is supposed to be weak, and does not influence the pump pulse at all, the latter propagates just like usual 2π soliton in a two-level medium. Accordingly, the only localized solution of Eq. (27) reads

$$\Omega(u)=\frac{2}{\tau_\Omega\cosh(u)}. \tag{29}$$

The group velocity of the soliton is defined by the relation [9]

$$\frac{1}{v}=\frac{n_\Omega}{c}+\frac{\kappa_\Omega\tau_\Omega^2}{2}. \tag{30}$$

Then, from Eq. (17), we find

$$\cos(\theta(u)/2)=\tanh(u). \tag{31}$$

Using Eq. (31) one can rewrite Eq. (28) as follows:

$$\epsilon^2\,\mathrm{cotanh}(u)\frac{d}{du}\left[\mathrm{cotanh}(u)\frac{d}{du}\alpha\right]=\alpha, \tag{32}$$

where

$$\epsilon^2 = \frac{\kappa_\Omega}{\kappa_\alpha} + 2\frac{n_\Omega - n_\alpha}{c\kappa_\alpha \tau_\Omega^2}. \tag{33}$$

Substitution

$$x = \ln(\cosh u) \quad (x \text{ runs from 0 to } \infty) \tag{34}$$

reduces Eq. (32) to

$$\frac{d^2\alpha}{dx^2} - \epsilon^{-2}\alpha = 0. \tag{35}$$

Therefore, for the localized solution of Eq. (31), we find

$$\alpha = \alpha_0 e^{-x/\epsilon} = \frac{\alpha_0}{\cosh^\epsilon(u)}, \tag{36}$$

where α_0 is the amplitude of the probe pulse. Parameter ϵ has to be real, i.e.,

$$\kappa_\Omega > 2\frac{n_\alpha - n_\Omega}{c\tau_\Omega}, \tag{37}$$

otherwise the localized solution does not exist.

It is easy to show that a pair of pulses (29) and (36) makes the linear absorption term in Eq. (26) for the probe energy equal to zero for any z. The simultaneous propagation of the two pulses can be referred to as ''pulse locking:'' the two pulses have equal group velocities, in contrast with the case when they propagate independently. As we already pointed out, if the pump pulse has a 2π soliton shape [Eq. (29)], the initial form of the weak probe pulse is not essential: the probe pulse of arbitrary envelope evolves toward the stationary ''pulse-locked'' form [Eq. (36)] (Fig. 2). Furthermore, we will show numerically in Sec. II D, that the effect of ''pulse locking'' is retained in the nonlinear regime when the probe pulse becomes strong.

D. Linear probe pulse amplification

Now consider the case when nonzero Raman inversion is prepared in the medium:

$$\frac{r_a}{\gamma_a} > \frac{r_c}{\gamma_c} \Rightarrow \rho_{aa}^0 > \rho_{cc}^0. \tag{38}$$

As before, the pulses are short enough not to be devastated by absorption. Therefore, transfer of the coherent pulse energy into spontaneously emitted photons is negligible, and the pulses are not devastated by absorption while propagating. According to Eq. (26), the probe pulse energy will increase during the propagation. If the Raman inversion is small,

$$\rho_{aa} - \rho_{cc} \ll \rho_{bb} - \rho_{aa} \tag{39}$$

(which is the case we will concentrate on), one can separate the process of pulse reshaping toward the matched state from the amplification process. [Roughly, reshaping has $(\rho_{bb}^0 - \rho_{aa}^0)/(\rho_{aa}^0 - \rho_{cc}^0)$ times faster time scale than amplification.] Thus, during the first short stage of the probe propagation, it is reshaped into the matched form (36). After that the probe pulse is amplified adiabatically, while its shape is preserved.

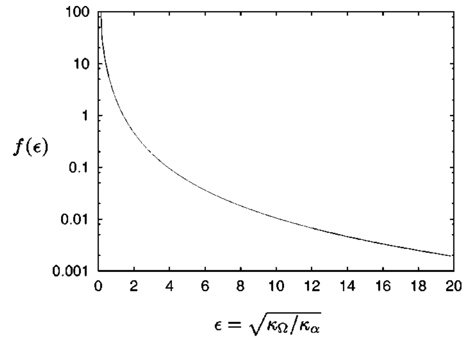

FIG. 3. Plot of the function $f(\epsilon)$ as given by Eq. (41).

In the following we will suppose that the host index of the medium is the same for the pump and probe pulses: $n_\Omega = n_\alpha$. Substituting Eqs. (28), (31), and (36) into Eq. (26), we find the energy gain coefficient for the probe pulse in the matched state:

$$G_\alpha \equiv \frac{1}{J_\alpha}\frac{\partial J_\alpha}{\partial z} = \frac{\kappa_\alpha}{2n_\alpha}\tau_\Omega(\rho_{aa}^0 - \rho_{cc}^0) f(\epsilon), \tag{40}$$

where

$$f(\epsilon) = \epsilon^{-2}\frac{\left[\int_{-\infty}^{+\infty} du \cosh^{-(1+\epsilon)}(u)\right]^2}{\int_{-\infty}^{+\infty} du \cosh^{-2\epsilon}(u)} \tag{41}$$

and ϵ is simply the square root ratio of the propagation constants:

$$\epsilon = \sqrt{\frac{\kappa_\Omega}{\kappa_\alpha}} = \sqrt{\frac{\omega_{cb}\wp_{cb}^2}{\omega_{ab}\wp_{ab}^2}}. \tag{42}$$

The function $f(\epsilon)$ is plotted in Fig. 3. One can see that the larger the probe transition oscillator strength (at fixed oscillator strength of the driven transition), the higher the linear gain for the probe that can be achieved. We will show numerically in Sec. IV that this tendency persists in the nonlinear stage of amplification when the probe pulse becomes strong.

IV. NONLINEAR PROBE PULSE AMPLIFICATION

The analytical results of Sec. III are obtained in the linear approximation in the probe field. As the probe pulse intensity increases, one cannot neglect its effect on dynamics of level populations and on the pump pulse. To study this nonlinear regime of the probe amplification, we solve the system of Maxwell-Schrödinger equations (1), (2), and (5)–(10) numerically. We again suppose that the host refractive index is the same for both the pump and probe fields. This assumption considerably simplifies computations. Then, without loss of generality, we can put the host refractive index for both fields equal to unity.

Our numerical procedure is standard [11], and we describe it here only briefly. First, having chosen the incoherent pump rates r_a and r_b and decay constants γ_a and γ_c, we find the populations in the medium prior to arrival of the pulses. We do this by solving the master equations (8), (9), and (11) in the steady state with $\alpha=\Omega=0$:

$$
\begin{aligned}
&-\gamma_a\rho^0_{aa}+r_a\rho^0_{bb}=0,\\
&\gamma_a\rho^0_{aa}+\gamma_c\rho^0_{cc}-r_a\rho^0_{bb}-r_c\rho^0_{cc}=0,\\
&\rho^0_{aa}+\rho^0_{bb}+\rho^0_{cc}=0.
\end{aligned}
\tag{43}
$$

We introduce the dimensionless coordinate $\bar{z}$ and retarded time $\bar{t}$ as follows:

$$\bar{z}=\kappa_\Omega\tau_\Omega z, \tag{44}$$

$$\bar{t}=\frac{t-z/c}{\tau_\Omega}. \tag{45}$$

We also define dimensionless Rabi frequencies $\bar{\Omega}$ and $\bar{\alpha}$:

$$\bar{\Omega}=\tau_\Omega\Omega, \quad \bar{\alpha}=\tau_\Omega\alpha. \tag{46}$$

All decay and pumping rates are also measured in units of $1/\tau_\Omega$. Then the wave equations (1) and (2) can be rewritten in terms of the above dimensionless quantities in the following ways:

$$
\begin{aligned}
&\frac{\partial\bar{\Omega}}{\partial\bar{z}}=i\rho_{cb},\\
&\frac{\partial\bar{\alpha}}{\partial\bar{z}}=i\epsilon^{-2}\rho_{ab},
\end{aligned}
\tag{47}
$$

with ϵ defined in Eq. (42). The density-matrix equations have exactly the same form as Eqs. (5)–(10) with replacements $t\to\bar{t},\ \Omega,\alpha\to\bar{\Omega},\bar{\alpha}.$ We solve propagation problem for the Rabi frequencies $\bar{\Omega},\bar{\alpha}(\bar{z},\bar{t})$ in the following way.

The fourth-order predictor-corrector method is used for both $\bar{z}$ and $\bar{t}$ integrations. Given the time dependence of the fields at some value of propagation distance $\bar{z}$, we integrate the master equations (5)–(10) in retarded time $\bar{t}$ and find off-diagonal elements of the density matrix ρ_{ab} and ρ_{bc} as functions of $\bar{t}$ at this fixed position $\bar{z}$. According to Eqs. (47), these matrix elements drive the fields, and we propagate the whole distributions $\bar{\Omega},\bar{\alpha}(\bar{t})$ a step further in $\bar{z}$. This integration scheme has been found to be stable for broad range of step sizes in $\bar{z}$ and $\bar{t}$.

We found that, after being launched into the medium with Raman inversion, weak probe pulse quickly attains the matched form (36), and then its amplitude steadily increases.

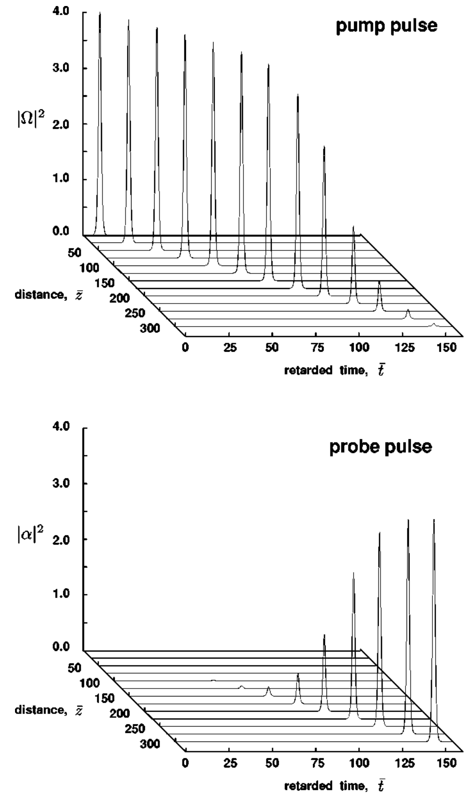

FIG. 4. Three-dimensional (3D) plot of the pump and probe intensity as a function of retarded time and propagation distance. The medium is prepared in a state with 5% Raman inversion: $\rho^0_{aa}=0.05$, $\rho^0_{bb}=0.95$, and $\rho^0_{cc}=0$. Frequencies and dipole matrix elements are the same for the pump and laser transitions. The input pump pulse has 2π soliton shape [Eq. (29)].

Before the probe pulse amplitude becomes comparable to that of the pump, evolution of the probe is accurately described by Eq. (24). When the probe pulse gains substantial energy, the gain coefficient for the probe considerably increases. Amplification of the probe ends with a vanishing of the pump pulse, and, ideally, all photons of the pump eventually become exchanged with the photons of the probe. [By ideal here we mean the case when the pulses are so short that relaxation in the medium can be completely neglected. In addition, the probe pulse at the entrance to the medium has to be so weak compared to the pump pulse, so that the energy lost while reshaping it into the matched form (36) is negligible.] A typical picture of the pulse dynamics is shown in Figs. 4(a) and 4(b).

We studied in detail how efficient resonant Raman scattering is in the system under consideration in the frequency up- and down-conversion regimes. As we pointed out above, ideally, all photons of the pump are exchanged on the photons of the probe, one by one. Therefore, the maximum output energy of the probe is simply equal to the input energy of the pump multiplied by the ratio of the corresponding atomic frequencies:

$$\frac{J^{out}_\alpha}{J^{in}_\Omega}=\frac{\omega_{ab}}{\omega_{bc}}. \tag{48}$$

The question of interest is the following: what length of the medium is needed for the probe pulse to attain the maximum

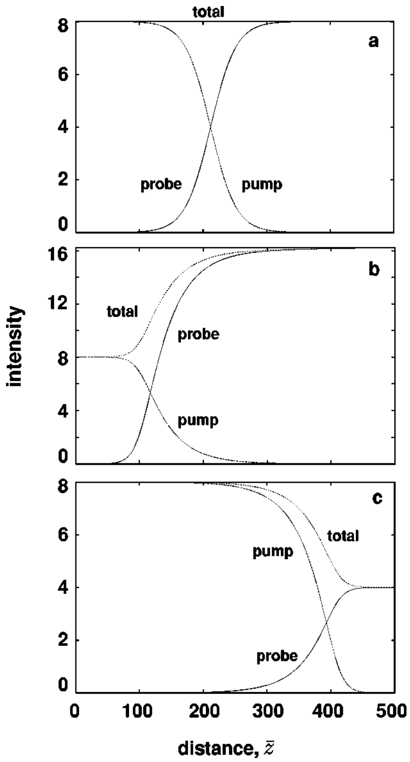

FIG. 5. Pump, probe, and total (pump plus probe) energy vs propagation distance for different values of the probe field frequency: (a) $\omega_{ab}=\omega_{cb}$, (b) $\omega_{ab}=2\omega_{cb}$, and (c) $\omega_{ab}=0.5\omega_{cb}$. For all three cases, the dipole matrix elements of the pump and laser transitions are the same, and the medium is prepared with 5% Raman inversion: $\rho^0_{aa}=0.05$, $\rho^0_{bb}=0.95$, and $\rho^0_{cc}=0$; the input pump pulse has a 2π soliton shape.

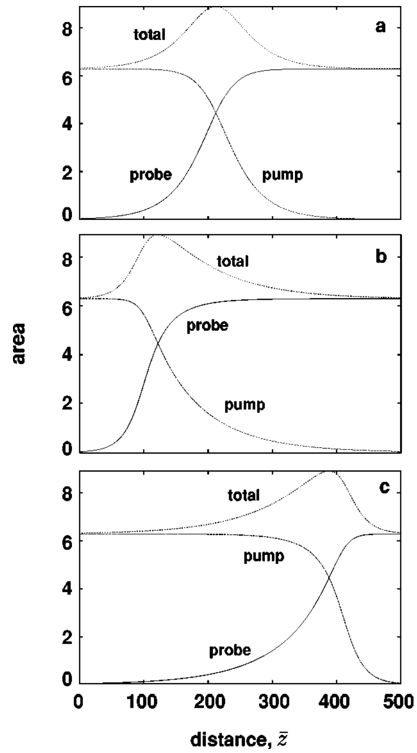

FIG. 6. Pump, probe, and total (pump plus probe) pulse area vs propagation distance for the three different cases of Fig. 5. The probe pulse at the output is always a 2π soliton due to the dominance of the SIT effect in the final stage of the probe amplification.

possible energy given by Eq. (48)? In the following consideration we take *equal dipole matrix elements of the two transitions*: $\wp_{ab}=\wp_{cb}$. In Fig. 5 the total energy of the pulses is plotted as a function of the propagation distance for different frequencies of the probe. We have shown analytically in Sec. III that, in the weak probe field limit, a larger oscillator strength of the probe transition yields a higher gain for the probe pulse. As follows from Fig. 5, this is true for the nonlinear stage of amplification as well: If the pump frequency is kept fixed, then length of conversion of the pump into the probe decreases with increasing frequency of the probe.

In Fig. 6 the probe and pump pulse *area* versus distance is shown for the three different cases of Fig. 5. In all three cases the area of the probe pulse tends to 2π, and the pulse is reshaped to become a typical 2π soliton of self-induced transparency (SIT):

$$\alpha=\frac{2}{\tau_\alpha \cosh(u)},$$

where (49)

$$u=(t-z/v_p)/\tau_\alpha.$$

v_p is a final group velocity of the probe pulse. This is because, at this developed stage of amplification, the probe pulse has enough energy to excite a considerable population into the state $|a\rangle$. Therefore, effects of SIT become comparable to the two-photon amplification. Eventually, when the pump pulse becomes weak and almost does not affect the probe, the SIT effect dominates. As a result, the probe pulse at the output is always a 2π soliton. Recalling that the final number of photons in the probe pulse is equal to the initial number of photons in the pump pulse, one can derive a simple relation between the initial duration of the pump pulse τ_d and the final duration of the probe pulse τ_α:

$$\tau_\alpha=\tau_\Omega/\epsilon^2. \tag{50}$$

Thus, in the up-conversion regime ($\epsilon>1$), the output duration of the probe is smaller than the input duration of the pump, and the medium acts as a pulse compressor.

V. ESSENTIAL PHYSICS

In this section we will discuss the physics beyond the two-photon resonant amplification of the probe pulse in the medium under consideration. Due to the interdependent dynamics of the fields and populations, this question is not as trivial as it may seem. As before, we will not consider the case when $\rho^0_{aa}>\rho^0_{bb}$, i.e., no population inversion is initially created directly on the laser transition. Then, as follows from Eq. (26), gain for the probe takes place only when atoms are prepared in a state with Raman inversion: $\rho^0_{aa}>\rho^0_{cc}$. This suggests that the amplification is due to the stimulated Raman scattering $|a\rangle\rightarrow|b\rangle\rightarrow|c\rangle$. On the other hand, resonantly pumping the 2π pulse temporarily transfers *all* population from ground state $|b\rangle$ to state $|c\rangle$. Under the same condition, $\rho^0_{aa}>\rho^0_{cc}$, this population transfer results in a temporary

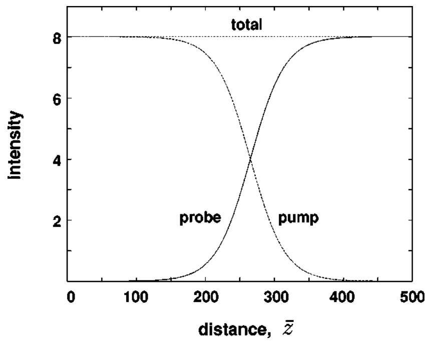

FIG. 7. Energy of the pulses vs propagation distance for parameters of Fig. 5(a), but with the pump and probe fields detuned from the corresponding resonances: $\Delta_\Omega = \Delta_\alpha = 1/\tau_\Omega$. At this detuning, the population of state $|a\rangle$ never exceeds the population of state $|b\rangle$—$\rho_{aa}(z,t) < \rho_{bb}(z,t)$—so that population inversion on the laser transition is absent. Two-photon probe gain, however, is reduced only slightly compared to the resonant case of Fig. 5(a), where this population inversion exists temporarily.

''window'' of inversion on the laser transition. Consequently, one may think that the gain for the probe pulse is due to this temporary inversion window.

To check what actually happens in the system, we performed an additional numerical simulation: We slightly detuned frequencies of both pulses from the corresponding atomic transitions, keeping them in a two-photon resonance. In this case, the 2π pulse of the pump [Eq. (29)] still propagates without absorption and dispersion, but excites the population from $|b\rangle$ to $|c\rangle$ to a lesser extent: if the pump frequency detuning is Δ, then the maximal population temporarily excited to the state $|c\rangle$ is $[1+(\Delta\tau_\Omega)^2]^{-1}$. Obviously, there is some value of Δ at which the inversion window disappears, and $\rho_{aa}(z,t) < \rho_{bb}(z,t)$ for any z and t. If the ''window'' is the reason for the probe gain, then the gain must vanish or considerably decrease at this detuning. Figure 7 demonstrates that this does not happen. The gain in this case persists, although it is slightly weaker than for resonant pulses. This proves that the mechanism responsible for the probe amplification is two-photon Raman scattering, and not the temporary inversion window.

Additionally, we studied the evolution of populations and coherences in the system under the action of the pump and probe pulses. Having fixed some position z where the probe pulse is expected to attain an appreciable amplitude (i.e., the probe experiences the nonlinear gain described in Sec. IV), we plot elements of the density matrix as functions of time (Fig. 8). As follows from Eqs. (1) and (2), the pulses are amplified when the imaginary part of the corresponding matrix elements ρ_{cb} and ρ_{ab} is negative; otherwise the pulses are absorbed. In the usual far-off-resonant Raman amplification, the ground state $|b\rangle$ participates in the process only virtually: By means of the two-photon process $|a\rangle \to |b\rangle \to |c\rangle$, population is transferred from state $|a\rangle$ to state $|c\rangle$, and absorption of the pump photon is simultaneously accompanied by emission of the probe photon. In the resonant case (which is the subject of consideration here), the situation is quite different. The ground state $|b\rangle$ participates in the process of Raman scattering, and the population of this state can be considerably changed by the fields. As a result, imaginary parts of ρ_{cb} and ρ_{ab} oscillate with time synchronously [Fig. 8(b)]; therefore, amplification (and absorption) of the pump and probe pulses happens at the same time: both pulses are first absorbed, then both are amplified. Due to the presence of Raman inversion, curves Im$[\rho_{cb}(t)]$ and Im$[\rho_{ab}(t)]$ are not symmetric: The first one has a larger absorptive part, and the second a larger gaining part. Hence the net results are amplification for the probe pulse and absorption for the pump pulse.

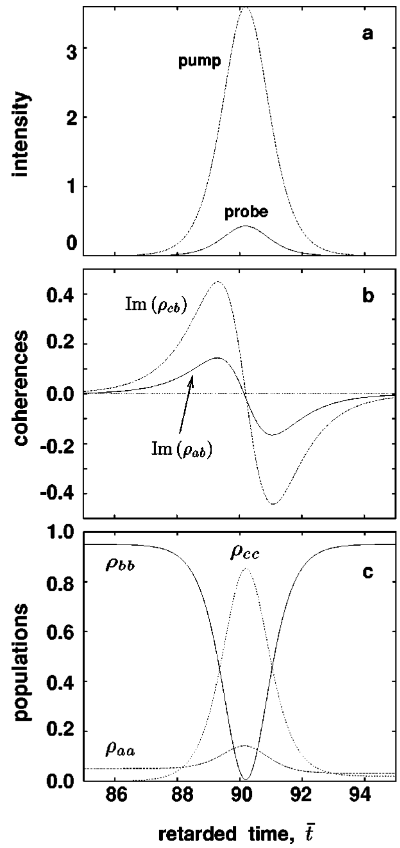

FIG. 8. Time dependence of density-matrix elements under simultaneous action of pump and probe fields: (a) Rabi-frequency envelope of the pump and probe pulses, (b) time dependence of the polarizations driving the fields, and (c) time dependence of the populations. Both fields are on resonance. A comparison of (b) and (c) shows that most of the probe gain takes place when there is no inversion of any kind in the system.

In Fig. 8(c) we plot the populations as functions of time for the case of Fig. 8(a). Comparison of Figs. 8(b) and 8(c) shows that, for the resonant case, the dominant part of the gain takes place when $\rho_{aa} < \rho_{bb}$ *and* $\rho_{aa} < \rho_{cc}$, i.e., *there is no inversion of any kind in the system, neither the usual one-photon inversion nor Raman inversion*. However, we do not refer to this effect as gain without inversion. The reason for this is that, what matters practically is how the population is distributed among atomic states *before* arrival of the pulses. (Although, of course, this is a terminological question.)

To demonstrate the advantage of using resonant pulses in the system under consideration, in Fig. 9 we plot the energy of the probe pulse as a function of propagation distance for the case when the pulses are in two-photon resonance but detuned far from the atomic resonance frequencies. Comparison with Fig. 5(a) shows that, indeed, conversion of the

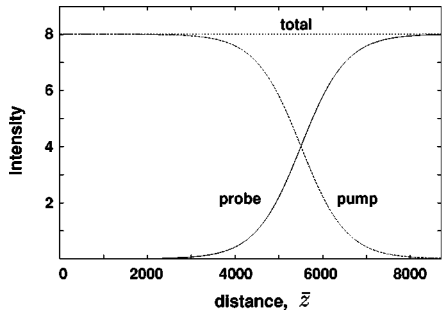

FIG. 9. Pump, probe, and total pulse energy vs propagation distance for the parameters of Fig. 5(a), but with both fields detuned far from the corresponding resonances: $\Delta_\Omega = \Delta_\alpha = 10/\tau_\Omega$. The length of complete conversion of pump into probe is much larger than for resonant case of Fig. 5(a), demonstrating the advantage of using resonant pulses.

pump into a probe is greatly enhanced in the resonant case.

In the previous discussion we dealt mainly with a pump pulse in the form of a 2π soliton. If the medium is pumped by a pulse with an area noticeably exceeding 2π, the situation becomes more complicated.

According to the theory of SIT, a pulse of any area greater than π in a resonant two-level medium breaks up into a sequence of 2π solitons [9]. After that all the solitons propagate through the medium independently of each other. This happens because the 2π soliton in a two-level medium, after exciting population from the ground state, brings it all back, leaving the medium in exactly the same state as it was before the arrival of the soliton. Therefore, subsequent soliton pulses do not ''feel'' the preceding ones.

However, this is not generally true for a multilevel medium in the presence of additional fields. In such a case, each of the 2π pulses may not leave a medium in exactly the same state as it was before arrival of the pulse. In particular, populations in the medium can be redistributed, and the pulse can leave behind a trace of excited polarization. Both of these will affect propagation of the subsequent pulses. Such a situation is illustrated in Fig. 10, where the pump pulse initially has an area equal to 4π. After entering the medium, the pump quickly breaks up into two 2π soliton pulses. These two solitons have different amplitudes; therefore they propagate through the medium with different velocities. The faster propagating 2π pump pulse creates a pulse on the probe transition, and amplification of the latter proceeds in exactly the same way as we discussed before for the case of individual 2π pulse of the pump. So does the second pump pulse, but only until the probe pulse, ''locked'' to the first pump pulse, attains a considerable amplitude. After that both pump and probe pulses of the first pair are strong enough to change the state of the medium substantially, creating a negative Raman inversion for the probe pulse of the second pair ($\rho_{aa} < \rho_{cc}$), and exciting the polarization on forbidden transition $|a\rangle \leftrightarrow |c\rangle (\rho_{ac} \neq 0)$. But negative Raman inversion for the probe means positive Raman inversion for the pump; therefore, at some point, the probe pulse of the second pair starts to attenuate while propagating, and the corresponding pump pulse grows. This ''opposite'' propagation dynamics ends with a vanishing of the pump pulse of the first pair. Eventually, at the output, we have two

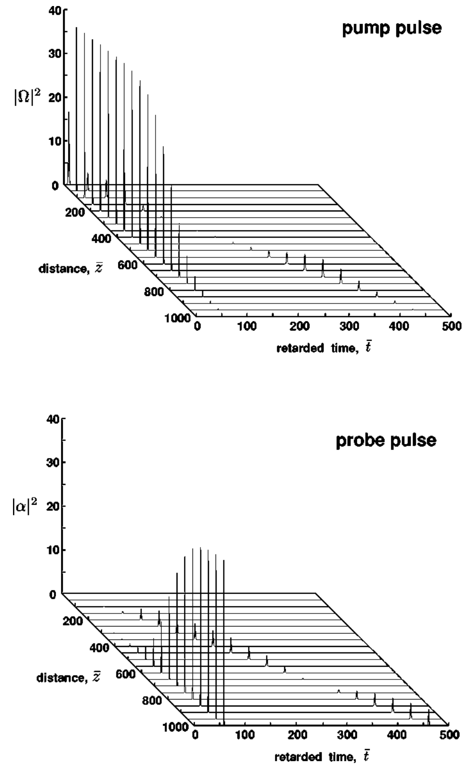

FIG. 10. 3D plot of the pump and probe intensity as a function of retarded time and propagation distance. All parameters are the same as in Fig. 4, but the input pump pulse has an area equal to 4π.

2π pulses on the probe transition, with the pump completely absorbed.

VI. CONCLUSION

We have considered resonant amplification of a short probe pulse in a *V*-type medium driven by a strong pump pulse. It has been shown that the resonant character of the atom-field interaction yields the effect of ''pulse locking,'' i.e., the pump and probe pulses propagate together, with the same group velocity. Further, in the presence of Raman inversion in the medium, the two-photon Raman-like amplification is resonantly enhanced, and, if the pump pulse is prepared in the form of a 2π soliton, this is not accompanied by fast resonant absorption of the pump. Therefore, the length of the complete conversion of pump photons into probe photons is considerably reduced in comparison with conventional (far-off-resonant) Raman laser operation. Analytical formulas describing the pulse dynamics in the linear-in-probe approximation have been derived. Those show that linear gain without inversion in this scheme is impossible. The nonlinear regime of the probe amplification has been studied numerically. Various aspects of frequency up- and down-conversion via resonant two-photon amplification have been discussed.

ACKNOWLEDGMENTS

The authors gratefully acknowledge support from the U.S. Office of Naval Research, the National Science Foundation (Grant No. PHY-9732445), the Welch Foundation, and the U.S. Air Force Research Laboratories.

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