Phase-dependent nonlinear optics with double-Λ atoms

E. A. Korsunsky and D. V. Kosachiov*
Institut für Experimentalphysik, Technische Universität Graz, A-8010 Graz, Austria
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We present a theory of a continuous-wave light propagation in a medium of atoms with a double-Λ configuration of levels. This is a configuration with a closed cycle of radiation-induced transitions. An interference of excitation channels in such closed-loop systems leads to a strong dependence of the atomic state on the relative phase and the relative amplitudes of applied electromagnetic waves. Therefore, the medium response may be controlled by the phases. On the other hand, the phases themselves change during the propagation. Thus the state of the medium and all the field parameters are tightly coupled to each other in the present problem. We consider the propagation of four-frequency laser radiation through the double-Λ medium for two situations. At resonant or near-resonant excitation of atoms, both the medium and the field evolve into a nonabsorbing state. This state implies specific coherent superposition for atoms (''dark state''), and particular relations for the field phases, amplitudes, and frequencies. In this way, the propagation results in the phase, amplitude, and frequency matching of the laser waves. In the second case, one Λ system in double-Λ atoms is excited resonantly, while the second Λ system is far off resonance. Such an excitation scheme ensures the preparation of atoms in the nearly dark state throughout the medium. Therefore, the total light energy is dissipated very weakly, whereas each individual laser wave can vary considerably along the propagation path. We have found that the resonant fields change as much as the far-detuned ones. The intensities oscillate sinusoidally with the optical length, with the energy being transferred back and forth between two waves in each frequency pair, resonant and far detuned. This gives the possibility for an almost lossless amplification of two of the laser waves, or an even generation of one of them. [S1050-2947(99)04412-1]

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I. INTRODUCTION

Nonlinear optics has recently gained a new impetus with the introduction of the effects of quantum interference and atomic coherence into the field. The interference of excitation channels in quantum systems (atoms) composing a medium allows one to modify and control optical properties of this medium. It has been found, for example, that in multilevel schemes interference can be destructive for linear susceptibility (absorption and refractive index). A strong reduction of the resonant absorption has given the name ''electromagnetically induced transparency'' (EIT) to the group of effects using quantum interference in nonlinear optics [1]. Especially promising is the use of EIT in nonlinear optical wave-mixing processes, because the reduction of the linear susceptibility may be accompanied by an enhancement of the nonlinear susceptibility, leading to a dramatic enhancement of the frequency conversion efficiency [2–10]. Moreover, the generated field components can possess almost perfect noise (including quantum noise) correlations, since the resonant absorption and associated noise processes are strongly suppressed [5,11].

A useful interpretation of the EIT phenomenon is given in terms of the so-called ''dark state,'' which is an atomic state superposition not excited by the radiation. The preparation of atoms in this superposition (the process referred to as ''coherent population trapping'' (CPT) [12]) eliminates the effect of the medium on a propagating beam of electromagnetic (e.m.) radiation. In fact, the dark state is created via destructive quantum interference, which requires specific conditions on the interaction parameters such as laser frequencies, phases, and Rabi frequencies of involved transitions. In many situations, however, the dark state may exist for any (mean) field phases and intensities. The only necessary condition is a multiphoton resonance of the atom-radiation interaction. As long as the laser fields do not change or change adiabatically, the dark state can be always found, and atoms can be prepared in this state by optical pumping in the continuous-wave (c.w.) regime or by adiabatic following in the pulsed regime.

The present paper deals with EIT in the medium of four-level atoms in a double-Λ configuration (Fig. 1). This scheme is a closed-loop system in which atomic transitions form a closed cycle. A peculiarity of such systems is the dependence of the atomic state on the relative phase and the relative amplitudes of applied e.m. fields [13,14]. In particular, only at some specific values of the relative phase and

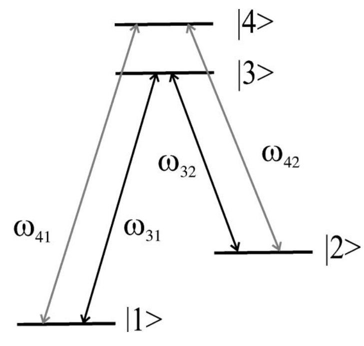

FIG. 1. Double-Λ system.

*Permanent address: Tyumentransgas Co., 627720 Yugorsk, Russia.

amplitudes may the dark superposition exist. Therefore, the response of atoms to the e.m. radiation may be controlled by phases and relative amplitudes of the multifrequency field. It has been suggested, for example, that closed-loop interaction schemes can be used for the phase control of photoionization [15], of nonadiabatic losses in coherent population transfer process [16], of spontaneous emission [17], and of subrecoil laser cooling and localization of atoms [18]. Laser light propagation in an optically dense medium consisting of closed three-level atoms was theoretically investigated in Ref. [19]. Experimentally, phase-sensitive population dynamics was observed in three- [20,21] and four-level [22,23] closed-loop systems. Recently, the phase-dependent EIT was demonstrated with optically dense sodium vapor excited as double-Λ atoms [24].

In this paper we focus our attention on the propagation of a c.w. laser light in the double-Λ medium. We show that both absorptive and dispersive properties of such a medium range from regular ''two-level'' to typical ''EIT'' character depending on the relative phase and amplitudes of the light. The medium is almost transparent to the field when phases, amplitudes, and frequencies of input field components satisfy specific conditions corresponding to the CPT conditions for double-Λ atoms. For other input parameters neither absorption nor refraction vanish, so that the field intensities as well as the phases are changing in the course of propagation. This leads to a change of the optical properties of the medium and, in turn, to a change of the very character of the propagation. Thus we face a problem where laser phases and intensities are entangled with each other through the closed-loop atomic medium. Solution of this problem requires an explicit consideration of the self-consistent set of equations including amplitude and phase Maxwell equations, and the density matrix (Bloch) equations for atoms in the medium.

Here we consider two practically important schemes of the excitation of double-Λ atoms. In the first one, all four frequency components of the laser field are tuned on resonance or near resonance with corresponding atomic transitions. In this case, the total energy of the field is dissipated by the medium if the CPT conditions are not satisfied for the input field. The optical length variation of individual components depends on the frequency relations. The phases do not change, and the intensities decay rapidly to zero when laser detunings in each Λ system differ considerably. However, if the frequency differences are in the narrow range around two-photon resonance (in ''transparency window'' range), then the phases vary with the length, and the amplitudes may be either amplified or attenuated. We find that these variations always proceed toward establishing a nonabsorbing state for both the medium and the field. After a sufficient long propagation length the medium is prepared in the CPT state, and the field frequencies, phases, and amplitudes become matched. This also occurs for the case when one of the frequency components is absent at the entrance to the medium, and it is generated in the medium – the case referred to as resonant four-wave mixing. The propagation of light in a double-Λ medium was studied earlier in Ref. [25] with respect to amplification without inversion. The authors considered the case of complete resonance and the CPT conditions initially fulfilled, and found the possibility of amplification of one of the frequency pairs when there is some population in excited states created by additional pumping mechanism. Here we are interested in a situation where double-Λ atoms are excited only by a four-frequency laser field with arbitrary parameters at the entrance to the medium.

The second case corresponds to the ''maximum coherence medium'' or ''phaseoneum'' [9,26]. Here one pair of frequency components is single photon resonant with transitions in one Λ system of the double-Λ atom, and the other pair is far detuned from the excited state in a second Λ system. The resonant pair creates a dark state and supports atoms in this state everywhere in the medium, while the far-detuned field may only weakly disturb the CPT. Therefore, the total light energy is dissipated very weakly during the propagation even when the CPT conditions are not satisfied. Nevertheless, a small disturbance of the dark state is sufficient to produce a parametric instability in the medium which leads to considerable amplification or attenuation of individual frequency components. It turns out that not only the far-detuned waves, but also the resonant waves, are varying to the same degree and with the same rate as the far-detuned fields. We find that the intensities oscillate sinusoidally with the optical length, with the energy being transferred back and forth between two waves in each frequency pair — resonant and far detuned. This allows us to use such an excitation scheme for very efficient frequency conversion. We calculate the case of a new frequency generation, and analytically find the optimum conditions.

The paper is organized as follows. In Sec. II we derive the coupled set of Maxwell-Bloch equations including amplitude and phase propagation equations and the atomic density matrix equations. In Sec. III an explicit expression for the dark state in a double-Λ system is obtained, as well as necessary conditions for the relative frequencies, phases, and amplitudes of transitions. Section IV is devoted to optical properties of the medium and to the light propagation in the case of resonant excitation of double-Λ atoms. The frequency conversion in phaseoneum is considered in Sec. V. In Sec. VI we discuss assumptions made in the present treatment, and summarize the main results of the paper.

II. BASIC EQUATIONS

We consider a transmission of the beam of four-frequency laser radiation propagating along the z axis:

$$\mathbf{E}(z,t)=\sum_{s=3,4}\sum_{m=1,2}\mathbf{e}_{sm}E_{sm}(z,t)\frac{1}{2}\exp[-i(\omega_{sm}t-k_{sm}z+\varphi_{sm}(z,t))]+\text{c.c.} \tag{1}$$

in a medium of double-Λ atoms (Fig. 1). Here E_{sm}, φ_{sm}, and $\mathbf{e}_{sm}$ are the amplitude, the phase, and the unit polarization vector, respectively, of the laser wave with frequency ω_{sm} and wave number $k_{sm}=\omega_{sm}/c$. Both the amplitudes E_{sm} and the phases φ_{sm} are regarded as slowly varying functions of time and coordinate, i.e., they vary little within an optical period and an optical wavelength. We assume that each field component with frequency ω_{sm} interacts with one atomic transition $|s\rangle$-$|m\rangle$ only, as depicted in Fig. 1.

The response of the medium, neglecting higher harmonics, is given by the polarization, which can also be represented as a sum of four frequency components with slowly varying amplitudes:

$$\mathbf{P}(z,t)=\sum_{s=3,4}\sum_{m=1,2}\mathbf{e}_{sm}P_{sm}(z,t)\exp[-i(\omega_{sm}t-k_{sm}z+\varphi_{sm}(z,t))]+\text{c.c.} \tag{2}$$

The Maxwell equations then yield the following set of equations for the slowly varying amplitudes $E_{sm}(z,t)$ and phases $\varphi_{sm}(z,t)$ of the optical field components [27,19]:

$$\frac{\partial E_{sm}}{\partial z}+\frac{1}{c}\frac{\partial E_{sm}}{\partial t}=-4\pi k_{sm}\ \mathrm{Im}(P_{sm}), \tag{3a}$$

$$\frac{\partial \varphi_{sm}}{\partial z}+\frac{1}{c}\frac{\partial \varphi_{sm}}{\partial t}=-\frac{4\pi k_{sm}}{E_{sm}}\ \mathrm{Re}(P_{sm}). \tag{3b}$$

Usually, the medium polarization components P_{sm} do not depend on the laser phases. Therefore, Eq. (3a) is completely decoupled from Eq. (3b), and it is often sufficient to consider the propagation problem with Eq. (3a) only, which gives the behavior of the laser intensity $I_{sm}=cE_{sm}^2/8\pi$. For the closed-loop light-atom interaction schemes, however, the relative phase of the e.m. field determines the state of atomic system, and hence, of the medium polarization. Thus, the refraction of the medium (change of the light phase) directly determines the absorption in this case, and one has to solve the complete set of Eqs. (3).

The macroscopic polarization of the medium, $\mathbf{P}(z,t)$, is a sum of atomic dipole moments induced by laser light on optical transitions, averaged over an ensemble of double-Λ atoms:

$$\mathbf{P}(z,t)=N_{\text{act}}\mathrm{Tr}(\hat{\mathbf{d}}\hat{\rho})=N_{\text{act}}\int_{-\infty}^{+\infty}dv_z\sum_{s=3,4}\sum_{m=1,2}\mathbf{d}_{ms}\rho_{sm}(v_z)w(v_z)+\text{c.c.}, \tag{4}$$

where N_{act} is the density of active atoms, $\hat{\mathbf{d}}$ is the electric-dipole moment operator, and $\hat{\rho}$ is the atomic density matrix. An ensemble averaging is carried out quantum mechanically over atomic states, and over the atomic velocities with the distribution $w(v_z)$, where v_z is the z projection of the atom velocity. In Eq. (4), the matrix elements $\mathbf{d}_{ms}$ and ρ_{sm} are in the basis of bare atomic states $|n\rangle$, $n=1$, 2, 3, and 4: $\mathbf{d}_{ms}\equiv\langle m|\hat{\mathbf{d}}|s\rangle$, and $\rho_{sm}\equiv\langle s|\hat{\rho}|m\rangle$. From Eqs. (2) and (4) we obtain (with the rotating-wave approximation being used)

$$P_{sm}=N_{\text{act}}|\mathbf{e}_{sm}\mathbf{d}_{ms}|\tilde{\sigma}_{sm}, \tag{5}$$

with

$$\tilde{\sigma}_{sm}=\int_{-\infty}^{+\infty}dv_z\ \ \sigma_{sm}(v_z)w(v_z), \tag{6}$$

where

$$\sigma_{sm}=\rho_{sm}\exp[i(\omega_{sm}t-k_{sm}z+\chi_{sm})], \tag{7}$$

and the phase χ_{sm} is the sum of the laser phase φ_{sm} and the phase η_{sm} of the atomic dipole moment $\mathbf{d}_{ms}=|\mathbf{d}_{ms}|e^{i\eta_{sm}}$: $\chi_{sm}=\varphi_{sm}+\eta_{sm}$.

It is more convenient to consider light propagation problems in terms of dimensionless variables: a dimensionless field amplitude (Rabi frequency) $g_{sm}=|\mathbf{e}_{sm}\mathbf{d}_{ms}|E_{sm}/2\hbar\gamma_{31}$, where γ_{31} is the spontaneous decay rate in a channel $|3\rangle\rightarrow|1\rangle$; a dimensionless optical length $\zeta=\kappa z$; and a dimensionless time $\tau=c\kappa t$ with the (absorption) coefficient $\kappa=2\pi N_{act}\omega_{31}|\mathbf{d}_{13}|^2/c\hbar\gamma_{31}$. The Maxwell equations (3) reduce then to the following set:

$$\frac{\partial g_{sm}}{\partial\zeta}+\frac{\partial g_{sm}}{\partial\tau}=-\frac{|\mathbf{d}_{ms}|^2}{|\mathbf{d}_{13}|^2}\frac{\omega_{sm}}{\omega_{31}}\ \mathrm{Im}(\tilde{\sigma}_{sm}), \tag{8a}$$

$$\frac{\partial \varphi_{sm}}{\partial\zeta}+\frac{\partial \varphi_{sm}}{\partial\tau}=-\frac{|\mathbf{d}_{ms}|^2}{|\mathbf{d}_{13}|^2}\frac{\omega_{sm}}{\omega_{31}}\frac{1}{g_{sm}}\ \mathrm{Re}(\tilde{\sigma}_{sm}). \tag{8b}$$

In the present paper we concentrate on a regime of light propagation in the continuous-wave limit, that is, we shall assume that the characteristic time of a change in the field amplitudes and phases, and the interaction time of atoms with the light, are much longer than the characteristic time of a change in the internal state of an atom. Then the polarization components can be found by using the steady-state values of the density-matrix elements σ_{sm}. The density-matrix equations can be reduced to the following forms [14]:

$$\dot{\rho}_{11}=ig_{31}(\sigma_{31}-\sigma_{13})+ig_{41}(\sigma_{41}-\sigma_{14})+\rho_{33}+(\gamma_{41}/\gamma_{31})\rho_{44},$$

$$\dot{\rho}_{22}=ig_{32}(\sigma_{32}-\sigma_{23})+ig_{42}(\sigma_{42}-\sigma_{24})+(\gamma_{32}/\gamma_{31})\rho_{33}+(\gamma_{42}/\gamma_{31})\rho_{44},$$

$$\dot{\rho}_{33}=-ig_{31}(\sigma_{31}-\sigma_{13})-ig_{32}(\sigma_{32}-\sigma_{23})-(\gamma_{32}/\gamma_{31}+1)\rho_{33},$$

$$\rho_{44}=1-(\rho_{11}+\rho_{22}+\rho_{33}),$$

$$\dot{\sigma}_{31}=-\frac{[\gamma_3-i(\Delta_{31}-k_{31}v_z)]}{\gamma_{31}}\sigma_{31}+ig_{31}(\rho_{11}-\rho_{33})+ig_{32}\sigma_{21}-ig_{41}\sigma_{34},$$

$$\dot{\sigma}_{32}=-\frac{[\gamma_3-i(\Delta_{32}-k_{32}v_z)]}{\gamma_{31}}\sigma_{32}+ig_{32}(\rho_{22}-\rho_{33})+ig_{31}\sigma_{12}-ig_{42}\sigma_{34}e^{-i\Phi}, \tag{9}$$

$$\dot{\sigma}_{41}=-\frac{[\gamma_4-i(\Delta_{41}-k_{41}v_z)]}{\gamma_{31}}\sigma_{41}+ig_{41}(\rho_{11}-\rho_{44})+ig_{42}\sigma_{21}e^{-i\Phi}-ig_{31}\sigma_{43},$$

$$\dot{\sigma}_{42}=-\frac{[\gamma_4-i(\Delta_{42}-k_{42}v_z)]}{\gamma_{31}}\sigma_{42}+ig_{42}(\rho_{22}-\rho_{44})+ig_{41}\sigma_{12}e^{i\Phi}-ig_{32}\sigma_{43}e^{i\Phi},$$

$$\dot{\sigma}_{12}=-\frac{[\Gamma+i(\Delta_{31}-\Delta_{32}-(k_{31}-k_{32})v_z)]}{\gamma_{31}}\sigma_{12}+ig_{31}\sigma_{32}-ig_{32}\sigma_{13}+i(g_{41}\sigma_{42}-g_{42}\sigma_{14})e^{-i\Phi},$$

$$\dot{\sigma}_{34}=-\frac{[\gamma_3+\gamma_4+i(\Delta_{41}-\Delta_{31}-(k_{41}-k_{31})v_z)]}{\gamma_{31}}\sigma_{34}+ig_{31}\sigma_{14}-ig_{41}\sigma_{31}+i(g_{32}\sigma_{24}-g_{42}\sigma_{32})e^{i\Phi},$$

$$\sigma_{ij}=\sigma_{ji}^{*}.$$

Here $\dot{\sigma}_{ij}\equiv(\partial\sigma_{ij}/\gamma_{31}\partial t)$,

$$\sigma_{12}=\rho_{12}\exp[(\omega_{31}-\omega_{32})t-(k_{31}-k_{32})z+\chi_{31}-\chi_{32}],$$

$$\sigma_{34}=\rho_{34}\exp[(\omega_{41}-\omega_{31})t-(k_{41}-k_{31})z+\chi_{41}-\chi_{31}].$$

The following notations are used in Eqs. (9): $\Delta_{sm}=\omega_{sm}-(\mathcal{E}_s-\mathcal{E}_m)/\hbar$ are the laser frequency detunings from transitions $|s\rangle$-$|m\rangle$ ($s=3$ and 4; $m=1$ and 2); $\mathcal{E}_n$ is the eigenenergy of the atomic bare state $|n\rangle$; γ_{sm} are the spontaneous relaxation rates for transitions $|s\rangle$-$|m\rangle$ ($s=3$ and 4; $m=1$ and 2) (see Fig. 1); and $\gamma_s=(\gamma_{s1}+\gamma_{s2})/2(s=3$ and 4). The rate Γ represents the decay rate of the atomic coherence σ_{12} determined by uncorrelated laser fluctuations, atomic collisions and other random phase disturbing processes. The relative phase Φ is given by

$$\Phi=\Delta\omega t-\Delta kz+\Phi_0, \tag{10}$$

with the multiphoton detuning

$$\Delta\omega=(\omega_{31}-\omega_{32})-(\omega_{41}-\omega_{42}), \tag{11}$$

the wave-vector mismatch

$$\Delta k=(k_{31}-k_{32})-(k_{41}-k_{42}), \tag{12}$$

and the constant (initial) phase

$$\Phi_0=(\chi_{31}-\chi_{32})-(\chi_{41}-\chi_{42}). \tag{13}$$

We observe from the density-matrix equations (9) that there is a steady state in the closed-loop atomic system only if the multiphoton resonance condition is satisfied, $\Delta\omega=0$, and that this steady state is determined by the relative phase Φ_0.

In what follows, we will always assume the condition $\Delta\omega=0$ to be satisfied. Moreover, we suppose that all four laser frequencies are close to each other in the magnitude $|\omega_{sm}-\omega_{rn}|\ll(\omega_{sm}+\omega_{rn})$. Such a situation occurs, for example, at the excitation of double Λ system on D lines of alkali-metal atoms, where the states $|1\rangle$ and $|2\rangle$ are the ground hyperfine sublevels [8,24]. We therefore can neglect the dispersion of the medium induced by far-detuned atomic states not belonging to the double-Λ system. Hence we neglect the ''usual'' phase mismatch, and always have $\Delta k=0$.

III. DARK STATES IN DOUBLE-Λ SYSTEM

The double-Λ scheme provides a very rich spectrum of phenomena based on atomic coherence. This is due to the manifold interference of the excitation channels in such a system. Coherent population trapping [12] is the most prominent and famous effect. The basic feature of CPT is the creation, through quantum interference, of a specific superposition $|NC\rangle$ of atomic states which is not excited by the laser field. Mathematically, this requires a condition

$$\hat{V}|NC\rangle=0, \tag{14}$$

where $\hat{V}$ is the interaction operator in the interaction picture:

$$\hat{V}=-\hbar\gamma_{31}\sum_{s=3,4}\sum_{m=1,2}g_{sm}\exp[-i(\Delta_{sm}t-k_{sm}z+\chi_{sm})]|s\rangle\langle m|+\text{H.c.} \tag{15}$$

One can easily prove that under the conditions

$$\frac{g_{31}}{g_{32}}=\frac{g_{41}}{g_{42}}, \tag{16}$$

$$\Phi=2\pi n \quad (n=0,1,2,\ldots), \tag{17}$$

and

$$\Delta_{32}-\Delta_{31}=\Delta_{42}-\Delta_{41}=0, \tag{18}$$

there are two superposition states which are not excited, i.e., which satisfy Eq. (14):

$$|NC\rangle_g=\frac{g_{32}/g_{31}}{\sqrt{1+g_{32}^2/g_{31}^2}}|1\rangle-\frac{1}{\sqrt{1+g_{32}^2/g_{31}^2}}|2\rangle, \tag{19}$$

$$|NC\rangle_e=\frac{g_{42}/g_{32}}{\sqrt{1+g_{42}^2/g_{32}^2}}|3\rangle-\frac{1}{\sqrt{1+g_{42}^2/g_{32}^2}}|4\rangle. \tag{20}$$

This means that, under conditions (16)–(18), the four-level double-Λ system is effectively reduced to the two-level system $|C\rangle_g$-$|C\rangle_e$ (the states $|C\rangle_g$ and $|C\rangle_e$ are orthogonal to $|NC\rangle_g$ and $|NC\rangle_e$), and two states $|NC\rangle_g$ and $|NC\rangle_e$ not coupled by the laser radiation. The state $|NC\rangle_e$ is unstable since it is a superposition of the radiative unstable states $|3\rangle$ and $|4\rangle$. In opposite, the ground-state superposition $|NC\rangle_g$ is stable, and spontaneous emission feeds this superposition, so that most of the atomic population is trapped in $|NC\rangle_g$ after some optical pumping time. The degree of imperfection of CPT is determined by the relaxation rate Γ of the coherence between states $|1\rangle$ and $|2\rangle$. The rate Γ should be sufficiently small in order to allow the population trapping in $|NC\rangle_g$.

The stationarity of the dark state requires, as we discussed in Sec. II the multiphoton resonance condition

$$\Delta\omega=(\omega_{31}-\omega_{32})-(\omega_{41}-\omega_{42})=0. \tag{21}$$

This condition also implies $\Delta k=0$ for close values of the laser frequencies. Thus we have, from Eq. (17),

$$\Phi=\Phi_0=(\chi_{31}-\chi_{32})-(\chi_{41}-\chi_{42})=2\pi n,\quad (n=0,1,2,\ldots). \tag{22}$$

The CPT conditions (16) and (22) are specific for the closed-loop interaction schemes. In other schemes, for example in a three-level Λ system, only the two-photon resonance condition [similar to Eq. (18)] is required. Therefore, the closed-loop systems have a resonant behavior not only with respect to the laser frequency, but also with respect to the laser intensity and the phase. Nonlinear properties of the medium consisting of ''closed-loop atoms'' may, therefore, be modified in a very dramatic way by a change of either phases or intensities of input laser fields.

IV. PHASE-DEPENDENT EIT WITH RESONANT EXCITATION

We start our analysis with the simplest situation of a complete one-photon resonance: $\Delta_{sm}=0$; equal Rabi frequencies of input laser fields: $g_{sm}\equiv g$; and equal spontaneous relaxation rates: $\gamma_{sm}\equiv\gamma$ (for all $s=3$ and 4 and $m=1$ and 2). In this case, an analytical solution of the density-matrix equations (9) can be obtained. The optical off-diagonal density-matrix elements σ_{sm} responsible for the medium polarization have the following form (for zero atomic velocity v_z):

$$v_{31}=v_{32}=v_{41}=v_{42}=\frac{2g^2(1-\cos\Phi)+\bar{\Gamma}}{16g^3(1-\cos\Phi)+8g}, \tag{23}$$

$$u_{31}=-u_{32}=-u_{41}=u_{42}=\frac{2g^2\sin\Phi+\bar{\Gamma}}{16g^3(1-\cos\Phi)+8g}, \tag{24}$$

where $v_{sm}\equiv\mathrm{Im}(\sigma_{sm})$, $u_{sm}\equiv\mathrm{Re}(\sigma_{sm})$, $\bar{\Gamma}=\Gamma/\gamma$, and the CPT conditions $\bar{\Gamma}\ll 1$, $g^2\gg\bar{\Gamma}$ are assumed to be satisfied. The ground-state coherence $u_{12}\equiv\mathrm{Re}(\sigma_{12})$, indicating the CPT state of the medium, is

$$u_{12}=-\frac{1}{2}\,\frac{(1+\cos\Phi)(1-\bar{\Gamma}/2)}{4g^3(1-\cos\Phi)+2}. \tag{25}$$

These simple expressions show that both the absorption and the refraction of laser radiation depend critically on the phase Φ. For $\Phi=2\pi n$ the absorption coefficients v_{sm} are very small, and the steady-state Maxwell equation

$$\frac{dg}{d\zeta}=-\frac{\bar{\Gamma}}{8g} \tag{26}$$

gives the linear attenuation of the laser intensity with a very gentle slope: $I\propto g^2(\zeta)=g^2(\zeta=0)-(\bar{\Gamma}/4)\zeta$. Such a dependence is typical for the light transmission through the medium prepared in the CPT state [28]. If the relaxation of the ground-state coherence is negligibly small, $\Gamma=0$, then the medium does not absorb light at all. We note that the ground-state coherence is maximum at this phase, $|u_{12}|\cong 1/2$, which means that atoms are in the dark state. In contrast, for $\Phi=\pi(2n+1)$ the absorption becomes similar to the case of two-level atom, $v_{sm}=g/(8g^2+2)$, which gives exponential attenuation $g^2(\zeta)=g^2(\zeta=0)\exp(-\zeta)$ at small intensities $g^2\ll 1$ (famous Beer's law), and linear attenuation $g^2(\zeta)=g^2(\zeta=0)-\zeta/4$ for the saturation $g^2\gg 1$ [29]. There is no coherence between the ground states at the phase $\Phi=\pi(2n+1)$: $u_{12}=0$. Note that for initial $\Phi=2\pi n$ and $\Phi=\pi(2n+1)$ the refraction coefficients are very small, so that the change of phases during transmission may be neglected.

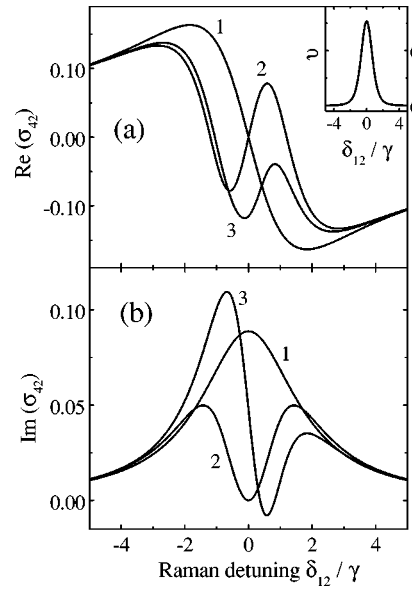

FIG. 2. (a) Real and (b) imaginary parts of the atomic density matrix element σ_{42} as a function of the Raman detuning $\delta_{12}=\Delta_{32}-\Delta_{31}=\Delta_{42}-\Delta_{41}$ for different phases: (1) $\Phi=\pi$, (2) $\Phi=0$, and (3) $\Phi=\pi/4$. Dependencies for other optical coherences σ_{sm} are similar. Parameters are as follows: atom velocity $v_z=0$, Rabi frequencies $g_{sm}\equiv g=0.6$, spontaneous relaxation rates: $\gamma_{sm}\equiv\gamma$ (for all $s=3$ and 4 and $m=1$ and 2) and $\bar{\Gamma}=10^{-3}$, and detunings $\Delta_{31}=\Delta_{41}=0$. The inset in (a) shows the Raman detuning dependence of the ''absorption coefficient'' υ for the phase Φ: $\upsilon=(u_{31}/g_{31})-(u_{32}/g_{32})-(u_{41}/g_{41})+(u_{42}/g_{42})$.

Thus one can control optical properties of the double-Λ medium by the phase Φ. Figure 2 demonstrates the frequency dependence of absorption and refraction coefficients. We see that for the phase $\Phi=\pi(2n+1)$ both absorption and refraction have a ''two-level'' frequency dependence, while for $\Phi=2\pi n$ both curves acquire typical ''CPT shape'': the absorption coefficient has a sharp narrow dip at the two-photon resonance (''black line'' or ''transparency window''), and the refraction coefficient steepens and changes the sign near the resonance as compared to the two-level case [27]. For the phases Φ not equal to $2\pi n$ or $\pi(2n+1)$, the absorption and refraction have some ''intermediate'' frequency dependence, which can be quite unexpected. For example, the absorption coefficient for $\Phi=\pi/4$ in Fig. 2(b) is negative at some frequency range, which means amplification of the wave with frequency ω_{42} at expense of the other waves.

Also important is the fact that at intermediate initial phases the refraction coefficients are not negligibly small even at exact two-photon resonance. Therefore, the phases φ_{sm}, and correspondingly, the phase Φ change during the transmission, thus leading to the change of absorption and refraction. The general trend in the resonance case is the same for different initial conditions: the phase Φ tends to the value $2\pi n$, and the field amplitude g decreases until it

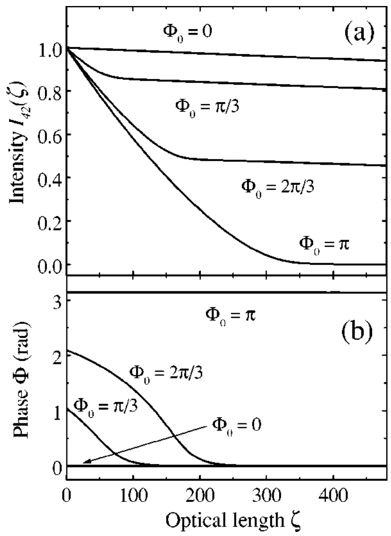

FIG. 3. Transmission of radiation in a double-Λ medium at resonance: $\Delta_{sm}=0$, equal Rabi frequencies of input fields $g_{sm}\equiv g=1$ (for all $s=3$ and 4 and $m=1$ and 2) and $\overline{\Gamma}=10^{-3}$ for different phases Φ. Here and all following transmission calculations, the averaging over atomic velocities, [Eq. (6)] is carried out with the assumption that the medium is a sodium vapor kept at temperature 410 K and interacting with light on the D_1 line. (a) Light intensity I_{42} (ζ) (in arbitrary units). Optical length dependences for laser waves with other frequencies are the same. (b) Optical length dependence of the phase $\Phi(\zeta)$.

reaches some steady-state value (if $\Gamma=0$) corresponding to the ''CPT phase'' $\Phi=2\pi n$. Thereafter, both the phase Φ and the amplitude g do not change anymore — the medium becomes transparent. The numerical calculations of the propagation equations (8), taking into account the Doppler broadening of the medium and nonzero relaxation rate Γ, are shown in Fig. 3. In Fig. 3, a small intensity decrease after the phase has reached zero is due to the presence of the relaxation Γ, in accordance with Eq. (26). Thus the net phase Φ of the four-frequency laser radiation goes to the value $2\pi n$ after a sufficiently long propagation length; that is, the laser waves become phase matched.

Along with the phase matching, a correlation of the laser frequencies occurs in the double-Λ medium. Corresponding to the calculations presented in Fig. 2, we may expect the phase dependence in the light propagation only when the laser frequencies lie in the range of the black line. The inset in Fig. 2 shows the frequency dependence of the ''absorption coefficient'' for the phase Φ: $\upsilon=(u_{31}/g_{31})-(u_{32}/g_{32})-(u_{41}/g_{41})+(u_{42}/g_{42})$. This quantity is different from zero only in the black line range. At the same time, the ''normal'' absorption coefficients v_{sm} are very small in this range only for the phase $\Phi=2\pi n$. Figure 4 demonstrates that the laser frequency components outside the black line range are extinguished, while the resonant component survives: the phase decays to zero in the black line range, and thereafter both the intensities and the phase propagate unaltered. Therefore, the laser frequencies also become matched after the transmission.

It turns out that the amplitudes are also matched by the double-Λ medium in such a way that the condition (16) is satisfied. When Rabi frequencies of input laser fields are not

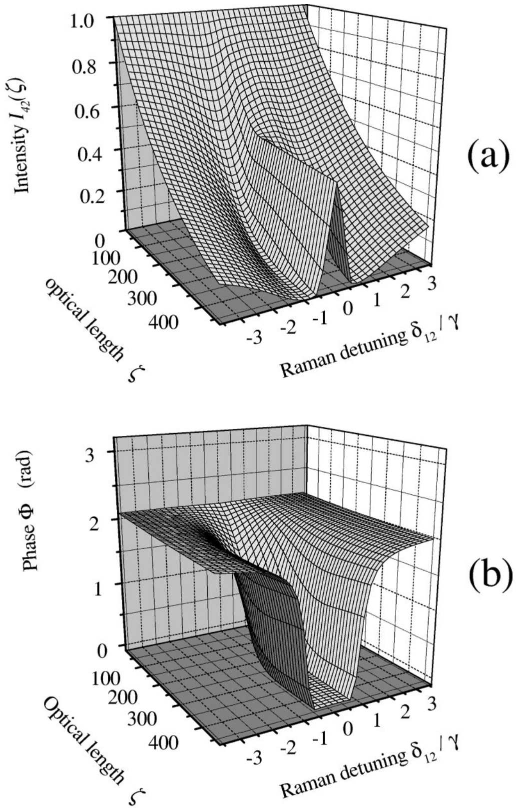

FIG. 4. Transmission of radiation in a double-Λ medium as a function of the Raman detuning δ_{12} for $\Delta_{31}=\Delta_{41}=0$, equal Rabi frequencies of input fields $g_{sm}\equiv g=1$ (for all $s=3$ and 4 and $m=1$ and 2), $\overline{\Gamma}=10^{-3}$, and initial phase Φ ($\zeta=0)=2\pi/3$. (a) Light intensity $I_{42}(\zeta)$ (in arbitrary units). The optical length dependences for laser waves with other frequencies are almost the same. (b) Optical length dependence of the phase $\Phi(\zeta)$.

all equal [so that condition (16) is not fulfilled] and the phase $\Phi=2\pi n$, one pair of the waves (E_{31} and E_{42} in Fig. 5) is exponentially amplified, while the other pair (fields E_{32} and E_{41} in Fig. 5) is exponentially attenuated. The short period of exponential changes is followed by saturation on the level corresponding to condition (16). The total light energy is dissipated at the initial transient stage of this process, and it is almost conserved when condition (16) is satisfied. The

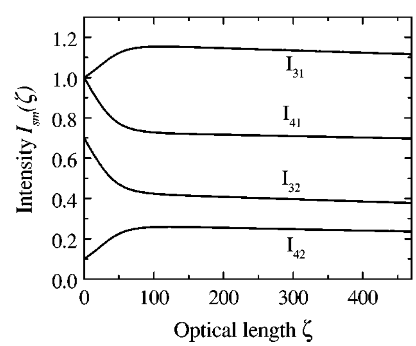

FIG. 5. Optical length dependence of the light intensities $I_{sm}(\zeta)$ in a double-Λ medium (in arbitrary units) at resonance: $\Delta_{sm}=0$; initial phase $\Phi(\zeta=0)=0$; Rabi frequencies of input fields $g_{31}=g_{41}=1$, $g_{32}=0.84$, and $g_{42}=0.32$ [condition (16) is not satisfied], and $\overline{\Gamma}=10^{-3}$.

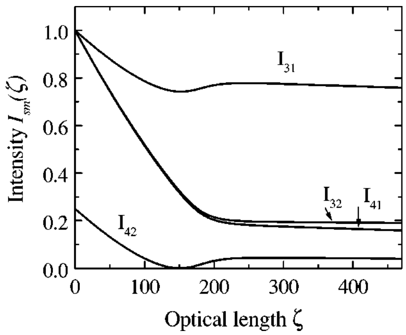

FIG. 6. Optical length dependence of the light intensities in a double-Λ medium (in arbitrary units). Parameters are the same as in Fig. 5, except for the initial phase $\Phi(\zeta=0)=\pi$.

phase $\Phi=2\pi n$ does not change. A similar effect of the amplitude correlation was reported in Ref. [30], but for the pulsed laser radiation propagating in the double-Λ medium.

In the case when the phase is initially not equal to $2\pi n$ and condition (16) is not fulfilled, both laser intensities and the phase Φ first decay until the phase reaches the value $2\pi n$. Afterwards, the waves E_{31} and E_{42} are amplified and the fields E_{32} and E_{41} are attenuated and reaching a steady state corresponding to condition (16), while the phase does not change anymore.

Such an amplitude matching occurs even in the case of initial phase $\Phi=\pi(2n+1)$, although in a quite specific way: one of the intensities (the smallest one, is that of the wave E_{42} in example of Fig. 6) decays at initial stage to zero. The phase Φ stays unaltered, equal to $\pi(2n+1)$, at this stage, and then it becomes undetermined at the point where $I_{42}=0$. From this point on, a four-wave-mixing process starts which generates the wave E_{42} in such a way that finally the laser wave amplitudes satisfy condition (16): $g_{42}=g_{41}g_{32}/g_{31}$ (Fig. 6). The total light energy is dissipated in the resonant four-wave mixing even though the two waves E_{31} and E_{42} are amplified initially. The wave E_{42} is generated with a frequency corresponding to the Raman detuning belonging to the black line range. The phase of the generated wave E_{42} is immediately matched to the phases of other laser waves, so that the phase Φ is equal to zero throughout the whole generation process.

However, the four-wave-mixing generation process becomes much more complicated as soon as at least one of the ''pump'' waves E_{31}, E_{32}, or E_{41} is slightly detuned from the corresponding one-photon transition. Figure 7 shows the generation process for such a case. The main difference as compared to the complete resonance case is that the phase of the generated wave is matched to other phases only at the steady state, when the system reaches a nonabsorbing state. This means, in particular, that in real experiments the phase matching is not granted *a priori*; one has to use media which are optically dense enough. We should note that dependences of the generated wave intensity on different laser detunings are quite complicated. For example, the maximum generation occurs not necessarily at zero detunings. Our numerical calculations reveal several resonances whose position and intensity, and even number, depend on the relative intensities of all the waves at a given propagation length. We believe that this is due to different manifestations of the quantum interference in the double-Λ system. It has been shown, for example, that some other decoupled states, different from those given by Eqs. (19) and (20), may appear at particular relations between the detunings and the Rabi frequencies, different from Eqs. (16) and (18) [31] (these states contain some admixture of one of the excited states $|3\rangle$ or $|4\rangle$, and are therefore not absolutely dark). The appearance of additional peaks in a spectrum of the generated wave due to interferences between dressed states in the double-Λ system, and due to interference of light from different velocity groups of atoms, was found in Ref. [32]. Nevertheless, a more detailed investigation, including the propagation effects, seems to be necessary.

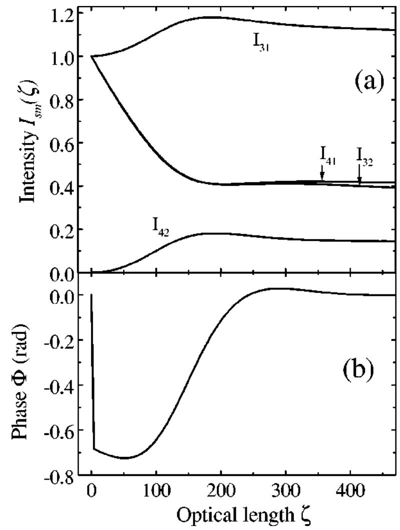

FIG. 7. Resonant four-wave-mixing generation in a double-Λ system. Spontaneous relaxation rates $\gamma_{sm}\equiv\gamma$ (for all $s=3$ and 4 and $m=1$ and 2); Rabi frequencies of input fields $g_{31}=g_{32}=g_{41}=1$, and $g_{42}=0$; and detunings $\Delta_{31}=0$, $\delta_{12}=0$, $\Delta_{41}/\gamma=1$; and $\bar{\Gamma}=10^{-3}$. (a) Light intensities $I_{sm}(\zeta)$ (in arbitrary units). (b) Optical length dependence of the phase $\Phi(\zeta)$.

Several experiments on the four-wave mixing by use of c.w. resonant or near-resonant excitation of the double-Λ atoms have been performed in last few years [4,6–8]. The experiments were motivated by the fact that for continuous frequency conversion at comparatively low pump intensities, a high nonlinear susceptibility is necessary, which is possible by tuning to resonances. The use of Λ or double-Λ atoms as a medium with induced atomic coherence should allow one to enhance the efficiency of the generation due to the cancellation of the resonant absorption and elimination of the refractive index by CPT. The enhancement of the generation efficiency was indeed demonstrated in the experiments. Unfortunately, no investigations have been performed on the propagation effects, which could be of vital importance for the generation, as we have shown above. Our calculations also indicate that the conversion efficiency of the resonant four-wave mixing cannot be very large: the process always includes the irreversible absorption of the pump fields, which leads to the establishment of a steady state. This is in contrast to the ''maximum coherence'' case, where absorption is very small at any stage of the process so that almost all energy can be transferred to the generated fields; see Sec. V.

V. FREQUENCY CONVERSION IN PHASEONEUM

Recently, nonlinear optical effects in the ''medium with maximum coherence'' (or ''phaseoneum'' [26]) have attracted a great deal of attention. It has been shown both theoretically and experimentally that such a medium is capable of high-efficiency nonlinear frequency conversion and generation [9,10]. For example, Jain *et al.* [9] reached a blue to UV conversion efficiency of 40% with atomic Pb vapor. The idea behind is to create and support a CPT medium by a strong pair of laser-radiations, and then to probe such a medium with a relatively weak, far-detuned laser beam. The atoms prepared in the CPT state may be viewed as a strong atomic local oscillator. The spectral components of a probe beam beat with the local oscillator, and are converted to a corresponding spectrum of sum or difference frequencies. At the same time, the problems associated with resonant absorption, phase shifts, and unwanted nonlinearities are avoided for the most part because the atoms are in the nonabsorbing CPT state.

One of the schemes which allows maximum-coherence nonlinear processes is a double-Λ system where one of the laser pairs (fields E_{31} and E_{32}) is resonant with corresponding transitions ($\Delta_{31}=\Delta_{32}=0$), while the other pair is far detuned from the excited state $|4\rangle$:$\Delta_{41}=\Delta_{42} \gg k_{sm}v_z, \gamma_{sm}, g_{sm}\gamma_{sm}, g_{sm}g_{s'm'}\gamma_{sm}$. A very similar scheme was also used in the experiment Ref. [9]. Since the second pair is far detuned, one would intuitively expect that its influence on the internal dynamics of double-Λ atoms is negligibly small, so that the atoms are pumped by the first pair E_{31} and E_{32} into a dark state which may hardly be disturbed by the second pair even if CPT conditions [Eqs. (16) and (17)] are not satisfied. At first glance, therefore, one may expect a quite boring propagation dynamics: the far-detuned fields E_{41} and E_{42} are simply attenuated weakly in a manner similar to the CPT case with nonzero relaxation Γ—see Eq. (26) and the upper curve in Fig. 3(a)—while the fields E_{31} and E_{32} are almost not depleted. It turns out, however, that this first intuitive idea is not absolutely correct. We have found that intensities of the far-detuned fields E_{41} and E_{42} oscillate with the optical length. Moreover, the resonant ''driving'' fields E_{31} and E_{32} change in the same manner and with the same rate as the ''probe'' fields E_{41} and E_{42}. This occurs because the optical susceptibilities for all four waves are of the same order at resonance. Figure 8 demonstrates the optical susceptibilities as a function of the Raman detuning for the phase $\Phi=\pi/2$. At frequencies out of the black line range, both the refraction and absorption coefficients for the fields E_{31} and E_{32} are quite large since they are almost resonant and atoms are not in the dark state, while those for the far-detuned waves E_{41} and E_{42} are almost zero because most of the atomic population circulates in the Λ system $|1\rangle$-$|3\rangle$-$|2\rangle$, so that state $|4\rangle$ is not excited. At the two-photon resonance, however, the absorption of the resonant waves is strongly reduced (atoms are in the CPT state), but not to zero. The imperfection of the absorption cancellation is caused by the action of the far-detuned fields which slightly disturb the CPT if either of conditions (17) and (16) is not satisfied. On the other hand, the (imperfect) reduction of the resonant fields absorption allows state $|4\rangle$ to be excited, thus leading to the absorption (or amplification) of the far-detuned fields. The order of all optical susceptibilities becomes the same at the two-photon resonance, and is determined by the detuning $\Delta_{41}=\Delta_{42}$. Therefore, it is not correct to consider the nonlinear processes in phaseoneum with the

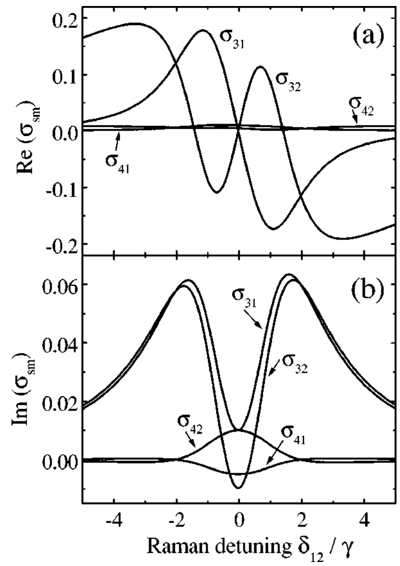

FIG. 8. (a) Real and (b) imaginary parts of the atomic density matrix elements σ_{sm} as a function of the Raman detuning $\delta_{12}=\Delta_{32}-\Delta_{31}=\Delta_{42}-\Delta_{41}$ for the ''maximum coherence'' case. Parameters are as follows: atom velocity $v_z=0$; spontaneous relaxation rates $\gamma_{sm}\equiv\gamma$ (for all $s=3$ and 4 and $m=1$ and 2) and $\Gamma=10^{-4}$; Rabi frequencies $g_{31}=g_{32}=g_{42}=1$, and $g_{41}=2$; detunings $\Delta_{31}=0$ and $\Delta_{41}/\gamma=100$; and phase $\Phi=\pi/2$.

assumption of undepleted resonant fields. One may expect from the above consideration that the maximum attenuation (amplification) occurs at the phase $\Phi=\pi$, the worst for CPT. Surprisingly, this appears not to be true. It is possible to derive an analytical solution for the present case. This shows, in particular, that the most interesting situation occurs for the phases not equal to zero and π.

Solution of the density-matrix equations (9) for $v_z=0$ and $\Gamma=0$, equal spontaneous relaxation rates $\gamma_{sm}\equiv\gamma$ (for all $s=3$ and 4 and $m=1$ and 2), and detunings $\Delta_{31}=\Delta_{32}=0$ and $\Delta_{41}/\gamma=\Delta_{42}/\gamma\equiv\Delta\gg 1, g_{sm}, g_{sm}g_{s'm'}$ gives, to the first order in $(1/\Delta)$,

$$v_{31}=\frac{g_{32}g_{41}g_{42}}{g_0^2\Delta}\sin\Phi,$$

$$v_{32}=-\frac{g_{31}g_{41}g_{42}}{g_0^2\Delta}\sin\Phi,$$

$$v_{41}=-\frac{g_{31}g_{32}g_{42}}{g_0^2\Delta}\sin\Phi, \tag{27}$$

$$v_{42}=\frac{g_{31}g_{32}g_{41}}{g_0^2\Delta}\sin\Phi,$$

$$u_{31}=\frac{g_{32}}{\Delta}\frac{g_{41}g_{42}(g_{32}^2-g_{31}^2)\cos\Phi+g_{31}g_{32}(g_{41}^2-g_{42}^2)}{g_0^4},$$

$$u_{32}=-\frac{g_{31}}{\Delta}\frac{g_{41}g_{42}(g_{32}^2-g_{31}^2)\cos\Phi+g_{31}g_{32}(g_{41}^2-g_{42}^2)}{g_0^4},$$

$$u_{41}=\frac{g_{32}}{\Delta}\frac{g_{31}g_{42}\cos\Phi-g_{41}g_{32}}{g_0^2},$$
$$u_{42}=\frac{g_{31}}{\Delta}\frac{g_{32}g_{41}\cos\Phi-g_{42}g_{31}}{g_0^2}. \tag{28}$$

The populations of the ground states are $\rho_{11}=g_{32}^2/g_0^2+O(1/\Delta)$, and $\rho_{22}=g_{31}^2/g_0^2+O(1/\Delta)$, and the ground-state coherence is $u_{12}=-g_{31}g_{32}/g_0^2+O(1/\Delta)$ and $v_{12}=O(1/\Delta)$, where $g_0^2=g_{31}^2+g_{32}^2$. Thus the atoms are really in the nearly dark (''gray'') state, and the term ''maximum coherence medium'' is indeed valid: $|\sigma_{12}|\approx\sqrt{\rho_{11}\rho_{22}}$. It is wrong, however, to say that the ground-state coherence is fixed for all the medium, and that it should be $u_{12}\approx-0.5$ for phaseoneum. As we will show below, the Rabi frequencies of the resonant fields change along the propagation path, so that even the case $|u_{12}|\ll 1$ is possible at some points. However, the condition $|\sigma_{12}|\approx\sqrt{\rho_{11}\rho_{22}}$ is always satisfied and the atoms are always in a gray state — the internal state of atoms follows the spatial change of the fields. This situation closely resembles the case of adiabatic population transfer, where the internal (gray) state of the atom adiabatically follows the temporal change of the fields [34].

We see from Eq. (27) that absorption vanishes for phases $\Phi=0$ and π. It vanishes to all orders in $(1/\Delta)$ for $\Phi=0$, if $\Gamma=0$ and condition (16) is satisfied, since atoms are dark for all laser waves (see Sec. IV), and there is absorption proportional to the second order in $(1/\Delta)$ for $\Phi=\pi$. For all other phases waves E_{31} and E_{42} are attenuated (amplified), and waves E_{32} and E_{41} are amplified (attenuated) for $0<\Phi<\pi$ $(-\pi<\Phi<0)$. The maximum effect occurs at $\Phi=\pm\pi/2$. In order to understand why the phase $\Phi=\pm\pi/2$, not the intuitive $\Phi=\pm\pi$, is optimum, it is useful to consider the classical model of the phaseoneum as a medium of atomic dipoles all oscillating at frequency $\omega_{31}-\omega_{32}=(\mathcal{E}_2-\mathcal{E}_1)/\hbar$ with the same phase $(\varphi_{31}-\varphi_{32})$ (suppose for simplicity that all the atomic phases $\eta_{sm}=0$). When the Raman field (E_{41},E_{42}), with the phase $(\varphi_{41}-\varphi_{42})$ and frequency $\omega_{41}-\omega_{42}=(\mathcal{E}_2-\mathcal{E}_1)/\hbar$, propagates in phaseoneum, the total-energy flow along the direction of propagation (Poynting's vector) consists of two parts: a contribution from the incident Raman field alone and a contribution from the interference between the incident field and the field radiated by the dipoles (see, e.g., Ref. [35]). For the plane e.m. wave, a model of a dipole sheet is appropriate, radiation from which is known to be shifted in phase by $\pi/2$ with respect to the dipole oscillations (Appendix A in Ref. [35]). Therefore, the interference term is proportional to $\sin((\varphi_{31}-\varphi_{32})-(\varphi_{41}-\varphi_{42})=\Phi)$, and it vanishes at $\Phi=0$ and $\Phi=\pi$. For the relative phases $\Phi\neq 0,\pi$ the Raman field is amplified or attenuated. At the same time, the dipole oscillations are forced by the Raman field (E_{41},E_{42}). Therefore, its amplification (attenuation) leads to an increase (decrease) of the dipole oscillation amplitude, that is, to attenuation (amplification) — remember the $\pi/2$ shift — of the resonant ''driving'' field (E_{31},E_{32}).

The problem of the field propagation can be solved analytically in the present case. The relevant Maxwell equations take the following forms:

$$\frac{dg_{31}}{d\zeta}=-\frac{g_{32}g_{41}g_{42}}{g_0^2\Delta}\sin\Phi,$$
$$\frac{dg_{32}}{d\zeta}=\frac{g_{31}g_{41}g_{42}}{g_0^2\Delta}\sin\Phi,$$
$$\frac{dg_{41}}{d\zeta}=\frac{g_{31}g_{32}g_{42}}{g_0^2\Delta}\sin\Phi, \tag{29}$$
$$\frac{dg_{42}}{d\zeta}=-\frac{g_{31}g_{32}g_{41}}{g_0^2\Delta}\sin\Phi,$$
$$\frac{d\Phi}{d\zeta}=\frac{g_{41}^2g_{42}^2(g_{31}^2-g_{32}^2)+g_{31}^2g_{32}^2(g_{42}^2-g_{41}^2)}{g_{31}g_{32}g_{41}g_{42}g_0^2\Delta}\cos\Phi+\frac{g_{31}^2-g_{32}^2+g_{42}^2-g_{41}^2}{g_0^2\Delta}.$$

The above equations are typical for nonlinear wave-mixing processes in lossless media [36]. The particular example of Eqs. (29) describes the $\chi^{(3)}$ process. This coincidence with well-known problems of nonlinear optics is not accidental, of course. In fact, the energy-level schemes in media used for the nonlinear wave mixing are all the closed-loop schemes. We believe, therefore, that many of the effects described in this paper may be observed under some conditions in ''traditional'' nonlinear materials.

We solve the set of Eqs. (29) by use of the method developed in seminal paper by Armstrong *et al.* [36]. First of all, from the first four of Eqs. (29), one obtains the conserved quantities

$$g_{31}^2+g_{32}^2=g_0^2\equiv C_1,$$
$$g_{31}^2+g_{41}^2=C_2, \tag{30}$$
$$g_{31}^2-g_{42}^2=C_3,$$

which are equivalent not only to the conservation of total power flow in the lossless medium (constant $g_0^2+C_2-C_3$), but also to the conservation of power flows for each Raman field: resonant field (E_{31},E_{32}) (constant g_0^2) and probe field (E_{41},E_{42}) (constant C_2-C_3) separately. These three constants reduce the problem to solution of the set consisting of the first and last of Eqs. (29). The first term in the right-hand side of the last of Eqs. (29) can be transformed to the following form by use of first four of Eqs. (29):

$$\frac{g_{41}^2g_{42}^2(g_{31}^2-g_{32}^2)+g_{31}^2g_{32}^2(g_{42}^2-g_{41}^2)}{g_{31}g_{32}g_{41}g_{42}g_0^2\Delta}\cos\Phi=\frac{\cos\Phi}{\sin\Phi}\frac{1}{g_{31}g_{32}g_{41}g_{42}}\frac{d(g_{31}g_{32}g_{41}g_{42})}{d\zeta}.$$

Therefore, the equation for the phase can be rewritten as

$$\frac{d\Phi}{d\zeta}=\frac{\cos\Phi}{\sin\Phi}\frac{d}{d\zeta}(\ln(q_1q_2q_3q_4))+\frac{q_1^2-q_2^2+q_4^2-q_3^2}{\Delta}, \tag{31}$$

which can immediately be integrated to give the fourth constant of motion,

$$q_1q_2q_3q_4\cos\Phi=\Pi+\tfrac{1}{4}(q_1^4+q_2^4+q_3^4+q_4^4), \tag{32}$$

where we have introduced new variables: $q_1=g_{31}/g_0$, $q_2=g_{32}/g_0$, $q_3=g_{41}/g_0$, and $q_4=g_{42}/g_0$. The value of the constant Π can be determined from the known values of q_i and Φ at the entrance to the medium, $\zeta=0$. Expression (32) is used then to express $\sin\Phi=\sqrt{1-\cos^2\Phi}$ in terms of the conserved quantity Π, and substitute it, together with the constants given in Eqs. (30), into the first of Eqs. (29). This gives the equation for the quantity $y_1=q_1^2=g_{31}^2/g_0^2$:

$$\frac{dy_1}{\sqrt{Ay_1^2+By_1+C}}=d(\zeta/\Delta), \tag{33}$$

with coefficients

$$A=4(a+b(1+a))-(1+a+b)^2-2(1+a^2+b^2)-8\Pi,$$

$$B=-4ab+(1+a+b)(1+a^2+b^2)+4\Pi(1+a+b),$$

$$C=-\frac{1}{4}(1+a^2+b^2)^2-4\Pi^2-2\Pi(1+a^2+b^2), \tag{34}$$

$$a=C_2/g_0^2,$$

$$b=C_3/g_0^2.$$

Equation (33) can now be solved easily. The value of the integral $\int dy_1(Ay_1^2+By_1+C)^{-1/2}$ depends on the sign of a quantity $D=4AC-B^2$. One can show that $D<0$ always. Therefore, the integral is $\int dy_1(Ay_1^2+By_1+C)^{-1/2}=-(1/\sqrt{-A})\arcsin[(B+2Ay_1)/(\sqrt{-D})]$, so that the general solution is

$$y_1=-\frac{1}{2A}\left(B-\sqrt{-D}\sin\left(\phi-\frac{\sqrt{-A}\zeta}{\Delta}\right)\right), \tag{35}$$

and $q_2^2=1-y_1$, $q_3^2=a-y_1$, and $q_4^2=y_1-b$, with the constant ϕ and coefficients (34) determined from the conditions at $\zeta=0$. The solution indicates that as the optical length increases, energy is transferred back and forth between the waves E_{31},E_{42} and E_{32},E_{41} with a period $(2\pi\Delta/\sqrt{-A})$. This behavior is very similar to the case of ‘‘traditional’’ nonlinear wave mixing in a dielectric [36], where solution is, in general, given by periodical Jacobi elliptic functions.

We now consider two specific examples of boundary conditions. The first case of interest is a situation when all four waves are present at the entrance to the medium, and the CPT condition for the phase is not satisfied. Corresponding to the discussion above, we choose the input phase $\Phi_0(\zeta=0)=\pi/2$, and equal input Rabi frequencies of all four waves. For this case we have: $a=1$, $b=0$, and $\Pi=-1/4$. The solution is then

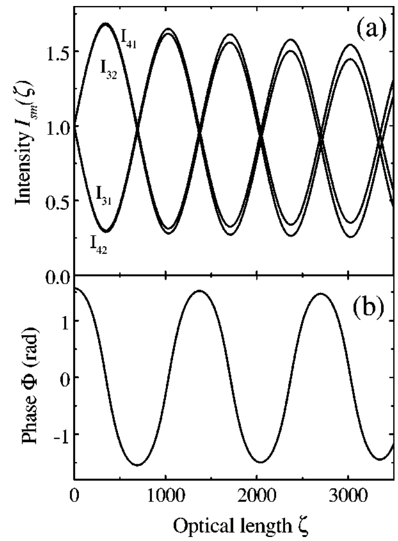

FIG. 9. Transmission of radiation in a double-Λ medium in the ‘‘maximum coherence’’ case. Rabi frequencies of input fields $g_{sm}\equiv g=1$ (for all $s=3$ and 4 and $m=1$ and 2); $\overline{\Gamma}=10^{-4}$; initial phase $\Phi(\zeta=0)=\pi/2$; and detunings $\Delta_{31}=0$, $\delta_{12}=0$, and $\Delta_{41}/\gamma=100$. (a) Light intensities $I_{sm}(\zeta)$ (in arbitrary units). (b) Optical length dependence of the phase $\Phi(\zeta)$.

$$y_1=\tfrac{1}{4}(2-\sqrt{2}\sin(\sqrt{2}\zeta/\Delta))$$

which gives, for the laser intensities,

$$g_{31}^2(\zeta)=g_{42}^2(\zeta)=\frac{g_0^2}{4}(2-\sqrt{2}\sin(\sqrt{2}\zeta/\Delta)),$$

$$g_{32}^2(\zeta)=g_{41}^2(\zeta)=\frac{g_0^2}{4}(2+\sqrt{2}\sin(\sqrt{2}\zeta/\Delta)),$$

and, for the phase,

$$\cos\Phi=\frac{1-\cos^2(\sqrt{2}\zeta/\Delta)}{1+\cos^2(\sqrt{2}\zeta/\Delta)}.$$

Thus the intensities of both far-detuned and resonant pairs of radiation oscillate over the optical length with the same period $\sqrt{2}\pi\Delta$ and amplitude $g_0^2/2\sqrt{2}$. We note that the period is much longer than the characteristic length scale of the intensity changes in the resonant case (since $\Delta\gg1$), and that the intensities oscillate between $g_0^2(2-\sqrt{2})/4$ and $g_0^2(2+\sqrt{2})/4$ so that it is not possible to transfer all the energy of one wave to the other in the present case. The phase Φ is also a periodical function of the optical length—it oscillates between $\pi/2$ and $-\pi/2$.

The situation becomes more complicated if we take into account in Eqs. (27) and (28), the terms to second order in $(1/\Delta)$, nonzero coherence decay rate Γ, and averaging over atomic velocities. Figure 9 shows the results of numerical calculations of the propagation dynamics in this case, when detuning Δ is larger than the Doppler width $k_{sm}v_z/\gamma$ of each single-photon transition. One sees that the intensity oscillations persist but the period of oscillations is not constant anymore—it decreases slightly with the optical length, and the total energy is exponentially dissipated with the rate

$\propto(1/\Delta)^2+\overline{\Gamma}$. Also, the inclusion of the relaxation Γ leads to slightly different behavior for the intensities in each conjugated pair: $g_{31}^2(\zeta)\neq g_{42}^2(\zeta)$ and $g_{32}^2(\zeta)\neq g_{41}^2(\zeta)$.

Nevertheless, we see that a medium with a nearly maximum coherence is indeed capable of an exchange of a considerable amount of energy between two pairs of laser waves even in real situations when all possible relaxation processes are taken into account. We note that this exchange occurs between the pairs (E_{32},E_{41}) and (E_{31},E_{42}), but not between resonant and far-detuned pairs. All this suggests that it is possible to convert a large amount of energy to one initially very weak seed wave (say E_{42}) or even generate it, with (E_{32},E_{41}) being strong pump waves and the wave E_{31} being arbitrarily small.

We now consider such a case of generation of a new e.m. wave by the use of phaseoneum [9]. This corresponds to a zero input intensity of one of the far-detuned probe waves: $g_{42}(\zeta=0)=0$. As one can see from Eq. (32), the solution does not depend on the phases of input fields in this case, which is also clear from a physical point of view: the behavior of the system does not depend on the phases if the transition loop is not closed [14]. However, as soon as the generation of the fourth wave starts, the interaction system becomes closed and the phases start to influence the process.

For simplicity, we suppose equal input intensities of the pump waves E_{32} and E_{41}: $g_{32}^2(\zeta=0)=g_{41}^2(\zeta=0)$ (detailed analysis shows that this is also the situation which gives the maximum conversion efficiency). We then have: $a=1$, $\Pi=-(2-4b+3b^3)/4$, and a solution can be expressed through the constant $b=g_{31}^2(\zeta=0)/g_0^2$:

$$y_1=g_{31}^2(\zeta)/g_0^2=\frac{1}{4-3b}(2-b^2-2(1-b)^2\times\cos(\sqrt{4b-3b^2}\zeta/\Delta)).$$

Intensities of other waves are given as

$$g_{32}^2(\zeta)=g_{41}^2(\zeta)=g_0^2\frac{1-b}{4-3b}(2-b+2(1-b)\times\cos(\sqrt{4b-3b^2}\zeta/\Delta)),$$

$$g_{42}^2(\zeta)=g_0^2\frac{2(1-b)^2}{4-3b}(1-\cos(\sqrt{4b-3b^2}\zeta/\Delta)). \tag{36}$$

The intensity of the generated wave reaches its maximum,

$$g_{42}^2(\max)=g_0^2\frac{4(1-b)^2}{4-3b}, \tag{37}$$

at the optical length $\zeta_{\max}=\pi\Delta/\sqrt{4b-3b^2}$. The efficiency η for conversion of power from the pump waves E_{32} and E_{41} to the wave E_{42} can be defined by

$$\eta=\frac{g_{42}^2(\max)}{g_{32}^2(\zeta=0)+g_{41}^2(\zeta=0)}.$$

We see that both the intensity of the generated wave [Eq. (37)] and the conversion efficiency,

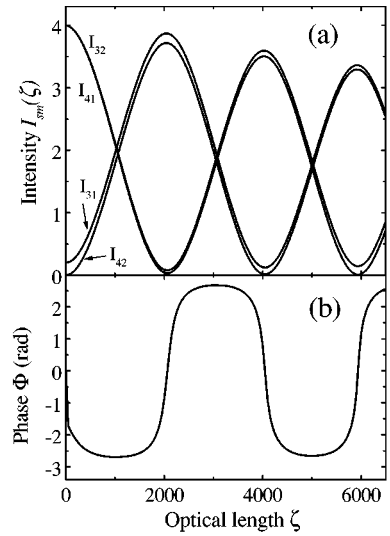

FIG. 10. Frequency conversion in a double-Λ system in the ''maximum coherence'' case. Rabi frequencies of input fields $g_{31}=0.45$, $g_{32}=g_{41}=2.0$, and $g_{42}=0$, $\overline{\Gamma}=10^{-4}$ and detunings $\Delta_{31}=0$, $\delta_{12}=0$, and $\Delta_{41}/\gamma=200$. (a) Light intensities $I_{sm}(\zeta)$ (in arbitrary units). (b) Optical length dependence of the phase $\Phi(\zeta)$.

$$\eta=\frac{2(1-b)}{4-3b}, \tag{38}$$

depend on the input intensity of the E_{31} wave, and are maximized at $b\ll1$: $g_{42}^2(\max)\approx g_0^2(1-5b/4)$ and $\eta\approx0.5(1-b/4)$. Thus we have arrived at the somewhat surprising result that the maximum conversion efficiency in phaseoneum is achieved when one of the resonant waves is taken to be weak at the entrance to the medium. This implies that the coherence $|u_{12}|\approx g_{31}g_{32}/g_0^2\ll1$, which is in contrast to the intuitive requirement $|u_{12}|\approx0.5$ for phaseoneum taking place at $g_{31}^2\approx g_{32}^2$. We recall, nevertheless, that the far-detuned excitation of the second Λ system, considered here, ensures that $|\sigma_{12}|\approx\sqrt{\rho_{11}\rho_{22}}$, and that atoms are in the gray state everywhere in the medium even at $g_{31}^2\ll g_{32}^2$. We also note that this takes place only if the frequency difference of the resonant waves is in the black line range, as follows from calculations in Fig. 8. At the same time, the generated wave may be tuned in a quite wide range: at a fixed optical length $\zeta_{\max}$ it remains within 50% of its maximum [Eq. (37)] over a frequency range of the order of $\Delta\gamma$ (of the order of few GHz, for example, Fig. 10).

The results of numerical simulation of the frequency conversion in phaseoneum are presented in Fig. 10. The influence of the ''real situation'' parameters — finite detuning Δ, coherence relaxation Γ, and the Doppler broadening — is similar to the calculations in Fig. 9. The oscillation period decreases slightly with the length, and the total e.m. energy is exponentially dissipated with the rate $(1/\Delta)^2+\overline{\Gamma}$. Therefore, we have to correct expression (37) for the generated intensity to

$$g_{42}^2(\max)=g_0^2\frac{4(1-b)^2}{4-3b}\exp\left(-\frac{\pi\Delta[(1/\Delta]^2+\overline{\Gamma})}{\sqrt{4b-3b^2}}\right),$$

and expression (38) for the conversion efficiency to

$$\eta=\frac{2(1-b)}{4-3b}\exp\left(-\frac{\pi\Delta((1/\Delta)^2+\overline{\Gamma})}{\sqrt{4b-3b^2}}\right). \qquad (39)$$

These two expressions indicate that decrease of the input intensity of the E_{31} wave leads not only to an increase of the ''ideal'' efficiency [Eq. (38)], but also to an increase of $\zeta_{\max}$ and, therefore, to an increase of losses. Hence there is an optimum for Δ and b which is determined by a particular experimental situation—by relaxation rate Γ, Doppler width, and energy-level spacing in real atoms. In the example of Fig. 10, the double-Λ medium is assumed to be a gas of Na atoms at a temperature 430 K with density $N_{\mathrm{act}}=3\times10^{11}$ cm^{-3}, excited on the D_1 line. The generated wave reaches a maximum at $\zeta=2050$, which corresponds to a density-length product of 2.5×10^{12} cm^{-2} and a gas cell length of 8 cm. At this optical length, the conversion efficiency is $\eta\approx0.465$ from numerical results, and $\eta\approx0.485$ calculated from Eq. (39), which does not take into account the Doppler broadening.

We note finally that the behavior of the phase in Fig. 10(b) is a bit different from that in Fig. 9(b). We observe that, as soon as the generation starts, the phase Φ acquires some value different from zero. Then it rapidly approaches a value close to π, and remains unchanged for quite a long optical length. However, when the generated wave approaches its maximum (and the pump waves their minimum), the phase varies very rapidly to change the sign. This means that the phase of the generated wave will be hardly locked to the phases of other waves, because any small change in the optical length around $\zeta_{\max}$, or even its fluctuation (e.g., due to gas temperature fluctuation) would lead to a considerable change of the phase. Therefore, the correlation of the phases in the present scheme seems problematic.

VI. SUMMARY AND CONCLUSIONS

We should note and comment on some assumptions which have been made in the treatment.

(1) Only the case when all four laser frequencies are close to each other in the magnitude is considered. Significantly different laser frequencies may lead to two important effects not accounted for in this paper. First, dispersion of the medium, induced by the interaction with far-detuned states other than those of the double-Λ system, may give different changes of the wave vectors k_{sm}. Hence the phase mismatch $\Delta kz\neq0$, even if the multiphoton resonance condition (21) is satisfied. However, as has been shown theoretically and experimentally [9], the phase mismatch can be compensated for by a small two-photon detuning δ_{12} which is in the range of the black line. The second effect of different frequencies is a nonzero Doppler width of the Raman transition $|1\rangle$-$|2\rangle$ which adds to decay of the ground-state coherence. Therefore, the Rabi frequencies of the laser waves should exceed this Doppler width in order to establish CPT in the medium. Thus, the processes considered here can be observed in double-Λ media with different laser frequencies, with an appropriate change of input laser intensities and detunings.

(2) Only copropagating waves are considered. For noncollinear propagation, the phase mismatch Δkz as well as the Doppler broadening of the $|1\rangle$-$|2\rangle$ transition will appear, which could be compensated for, as noted above, by two-photon detuning and a higher input intensity, respectively. An interesting regime of the nonlinear generation was found in Ref. [33] for counterpropagating light waves exciting double-Λ atoms — a large parametric gain and mirrorless oscillation within a very short optical length.

(3) The transversal profile of the laser beams is not taken into account. This would not influence the results as long as the profiles of all four frequency components are the same. However, new features in the light propagation through the double-Λ medium may appear for distinct input profiles. For example, we can expect the matching of the profiles at resonant excitation and long optical length. At small lengths, rebuilding of the profiles should occur, which may allow their control by the phases of input fields. Also, the process of self-focusing should be substantially modified as compared to both two- and three-level media, and should allow the phase control. These issues will be addressed in our future investigation.

The following conclusions summarize the theory presented here. We have theoretically investigated the propagation of c.w. laser radiation through the medium of double-Λ atoms. A coupled set of Maxwell-Bloch equations has been derived and solved both numerically and analytically for some situations where the effects of quantum interference play an important role. The interference of closed-loop excitation paths in the double-Λ system may be either constructive or destructive depending on the specific relations between the phases and amplitudes of applied e.m. waves. The latter case, realized under conditions (16) and (17), leads to the appearance of a superposition of atomic ground states, [Eq. (19)], not excited by the laser light, and to pumping of the population into this dark state. This results in a strong reduction of light absorption by atoms. Such an absorption reduction for specific light phases, amplitudes, and frequencies is a basic feature of the process.

We have considered two cases of the laser excitation of a double-Λ medium. In the first one, all four frequencies of the laser field are tuned on resonance or close to resonance with corresponding atomic transitions. Here the medium is transparent and not refractive for the laser light when conditions for the amplitudes [Eq. (16)], phases [Eq. (17)], and frequencies [Eq. (18)] are satisfied at the entrance to the medium. If any of these conditions is not initially fulfilled, then the laser energy is dissipated during transmission through the medium, and field amplitudes, and phases are changed. The amplitudes of individual laser waves may be either amplified or attenuated in the process. The general trend is, however, always the same: the changes proceed in a direction toward establishing CPT conditions [Eqs. (16) and (17)], and lossless propagation afterwards. This process can be understood in terms of the ''dressed fields'' [37,30]: the CPT component of the field which implies conditions (16)–(18) propagates without absorption, while other field components are damped out by the medium. Thus, after some transient regime, both the atomic medium and the laser field arrive at the nonabsorbing state. In the case of conventional EIT, only a particular relation between field frequencies (two-photon resonance) is required for such a nonabsorbing state. In fact, both the pulse matching [37] and fluctuation correlation [38] found for conventional EIT are due to the extinction of Fourier components of the laser field which do not satisfy the two-photon resonance condition. In contrast, specific relations for the field amplitudes, phases, and frequencies are important for EIT in closed-loop media. Therefore, the resonant excitation of double-Λ atoms yields an efficient preparation of the medium in a coherent state, and complete correlation of the laser fields not only in the pulsed regime, but also in the c.w. regime. This effect can be used, for example, for laser phase locking, for amplitude matching, for amplitude and frequency stabilization of c.w. radiation, and for amplitude and phase fluctuation correlation.

Another mode of the light-atom interaction investigated in the present paper is a resonant excitation of one Λ system, and far-off-resonance excitation of the second Λ system in double-Λ atoms. Such an excitation scheme ensures preparation of atoms in a nearly dark state throughout the medium. Therefore, the total energy of laser waves propagating in this coherent medium is dissipated very weakly, even when the CPT conditions for the amplitudes and phases are not fulfilled. Nevertheless, the change of each individual laser wave along the propagation path can be dramatic. We have found that both the far-detuned waves and resonant waves are changing. There are two important consequences of this fact. First, the state of the medium, while always dark, changes along the propagation path since it follows the change of the resonant fields. Second, such changes provide the possibility of an efficient transfer of energy from one pair of laser waves to another, which could be used for almost lossless amplification of two of the laser waves or generation of one of them. Our calculations show that maximum generation (e.g., of the wave E_{42}) takes place when the conjugated wave E_{31} is weak, and intensities of the strong pump waves E_{32} and E_{41} are of the same order in magnitude. Then one may expect an almost complete transfer of energy from the waves E_{32} and E_{41} to E_{42} and E_{31} at some predefined optical length. Advantages of this scheme over resonant four-wave mixing include the much larger efficiency of the generation due to very weak total energy dissipation, and the possibility to tune the frequency of the generated wave. On the other hand, it requires quite large optical densities, and the phase of the generated wave is not stable with respect to the optical density fluctuations.

Recent experiments have demonstrated that quantum-interference-induced nonlinear optical effects with the double-Λ excitation scheme can be realized not only with atomic [9,4,8,24] and molecular [7] gases, but also with solid media [6,21]. These processes provide highly efficient frequency conversion and generation of coherent radiation in a wide frequency range, for very low input pump powers, with nearly ideal quantum noise properties as they combine large nonlinearity with substantially reduced spontaneous emission noise [5,11]. All this leads one to expect a bright future for coherent media in many areas of optics, atomic and laser physics, and quantum control.

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