Resonant nonlinear optics in coherently prepared media: Full analytic solutions

E. A. Korsunsky and M. Fleischhauer
Fachbereich Physik, Universität Kaiserslautern, D-67663 Kaiserslautern, Germany

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We derive an analytic solution for pulsed frequency conversion based on electromagnetically induced transparency or maximum coherence in resonant atomic vapors. In particular, drive-field and coherence depletion are taken into account. The solutions are obtained with the help of a Hamiltonian approach, which in the adiabatic limit allows one to reduce the full set of Maxwell-Bloch equations to simple canonical equations of Hamiltonian mechanics for the field variables. Adiabatic integrals of motion can be obtained and general expressions for the spatiotemporal evolution of field intensities derived. Optimum conditions for maximum conversion efficiency are identified and the physical mechanism of nonlinear conversion in the limit of drive-field and coherence depletion discussed.

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I. INTRODUCTION

Resonant nonlinear optics in atomic gases has received a great impetus in recent years due to new concepts based on the application of specific coherence and interference effects. Two of these concepts attracted particular attention. The first mechanism uses the effect of electromagnetically induced transparency (EIT) [1,2], the second uses maximum coherence between two metastable atomic levels [3]. Both schemes allow for high-efficiency nonlinear conversion with substantially alleviated phase-matching problems in a rather dilute ensemble of atoms if the medium is driven by an undepleted coherent coupling field or if an undepleted atomic coherence is assumed. These assumptions do not take into account the resources needed to maintain the drive field or atomic coherence. Thus in the considered limit, the *overall* efficiency of the resonant nonlinear processes is small. In the present paper we analyze and compare EIT- and maximum-coherence based systems with each other and with the conventional nonresonant schemes of nonlinear optics, taking into account the transparency- or coherence-maintaining fields and their depletion. Effects of inhomogeneous broadening are, however, disregarded. Deriving analytic solutions for the nonlinear pulse propagation problem, we show that in both systems also a large *overall* conversion efficiency can be achieved. The physical nature of the conversion can, however, no longer be associated with EIT or maximum coherence alone.

In EIT a strong resonant electromagnetic (EM) field of frequency ω_2 is applied to the transition between two excited states $|2\rangle$ and $|3\rangle$ [Fig. 1(a)], causing a splitting of both (Autler-Townes effect). When a weak probe field of frequency ω_3 resonant with the transition from the ground state $|1\rangle$ to the bare state $|3\rangle$ is applied, its linear interaction with the medium is almost perfectly canceled. The vanishing of the linear susceptibility, i.e., absorption and refraction, for the probe field is due to a destructive interference between the two excitation paths through the Autler-Townes doublet and persists even for small splittings. If the ground state $|1\rangle$ is coupled to the intermediate state $|2\rangle$, on the other hand (e.g., by a two-photon transition), photons of frequency ω_3 are generated. While the resonant linear absorption of these photons is suppressed by destructive quantum interference (EIT), the nonlinear susceptibility responsible for their generation is only affected by the splitting, which can be rather small. Since there are no resonant contributions to the refractive index, there is, furthermore, perfect phase matching except for contributions from other off-resonant transitions. Owing to the large dispersion near the transparency frequency of EIT, a small detuning δ_2 can be applied to compensate for these contributions and phase matching can easily be obtained without sacrificing the cancellation of absorption. Thus EIT provides a perfect system for large nonlinear conversion with a minimum of atoms. Since the first demonstration of phase matching in EIT-assisted four-wave frequency mixing [4], many other experiments confirmed considerable improvement in the conversion efficiency when EIT has been used (see, e.g., Refs. [5,6]).

Another mechanism proposed recently is referred to as nonlinear optics with maximum coherence [3,7]. The idea here is to prepare and maintain the atoms in a coherent superposition of atomic states $|1\rangle$ and $|2\rangle$ with equal amplitudes. This can be done either by two strong fields exciting the $|1\rangle-|2\rangle$ coherence via a Raman transition [3] or by other means such as rapid adiabatic passage [8]. The coherence, established in such a way, plays the role of a strong

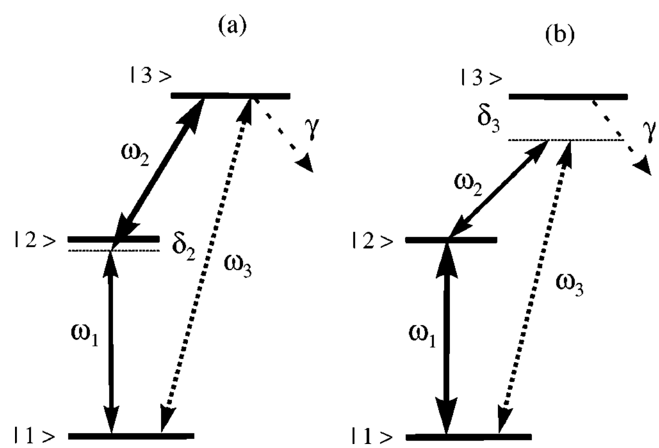

FIG. 1. Resonant sum-frequency generation. (a) Strong drive field (ω_2) between metastable state $|2\rangle$ and excited state $|3\rangle$ creates EIT; (b) strong drive ω_1 (two-photon, Raman, or magnetic coupling) generates maximum coherence between $|1\rangle$ and $|2\rangle$.

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local oscillator. If then a relatively weak field ω_2 detuned by a sufficiently large amount δ_3 from the $|2\rangle-|3\rangle$ transition is applied, Fig. 1(b), it will beat against the local oscillator to generate the sum (or difference) frequency. Here, the refraction due to resonant transitions does not vanish. But since the nonlinear coupling strength is of the same order as the linear susceptibility, complete conversion occurs over a distance smaller than the coherence length [9]. Consequently there is no need to phase match the propagating beams. We note that the relation between the $|1\rangle-|2\rangle$ coherence and the wave-mixing processes has been pointed out in earlier work on nonlinear optics (see, e.g., Refs. [10] and references therein).

In the limit of undepleted drive or constant coherence, the nonlinear conversion process in both the EIT- and maximum-coherence schemes affects the quantum state of the atoms only in the perturbative sense. Consequently the probability of photon conversion per atom is small. For the same reason the overall conversion efficiency including the transparency- and coherence-generating fields is small. We here analyze the properties of the two mechanisms for arbitrary strength of the drive fields taking into account their depletion and compare them to conventional off-resonant nonlinear optics.

The drive depletion turns the propagation problem into a truly nonlinear one. Its solution is particularly challenging for pulses and is, in general, possible only numerically. In order to obtain transparent analytical solutions we apply here the so-called Hamiltonian approach [11–14], which allows for a solution in a wide range of physically relevant situations. This approach is especially useful under adiabatic conditions, i.e., when the atoms are excited by the laser pulses in such a way that they remain in the same instantaneous eigenstate of the interaction Hamiltonian during the entire process. In the present work, we will assume that the interaction is adiabatic. The adiabatic approximation requires a slow rate of evolution as compared to the frequency separation of the adiabatic eigenstates. This results usually in a requirement for the product of the pulse duration and the Rabi frequency of the radiation field to be much larger than unity. Thus, for sufficiently intense fields, the process can be adiabatic even for short pulses. It should be noted, however, that this assumption rules out such important effects as group-velocity reduction, which result from lowest-order nonadiabatic corrections. For sufficiently large intensities or cw fields these effects do not influence the process. An analysis of nonadiabatic corrections to the Hamiltonian approach taking into account group delays will be given in a future publication.

To simplify the calculations and the interpretation of the analytic results, we restrict ourselves to a somewhat idealized three-level atomic excitation scheme (Fig. 1). The nonlinear three-wave mixing in a three-level system is normally forbidden due to symmetry [9]. However, this scheme provides the simplest example where all above-mentioned mechanisms of nonlinear-optical wave mixing may take place. It should be noted that similar results, however with much lengthier expressions and larger number of atomic parameters, can be found for a more realistic three-level scheme, where $|1\rangle-|2\rangle$ transition is a two-photon one [15]. Moreover, the three-wave-mixing processes are possible when a dc electric field is applied to the atomic sample [16].

The paper is organized as follows. In Sec. II we discuss resonant nonlinear-optical processes based on EIT or maximum coherence for undepleted drive fields or undepleted coherence, respectively, and compare them to conventional off-resonance nonlinear optics. In Sec. III we outline the Hamiltonian approach, which allows to eliminate the atomic degrees of freedom assuming adiabatic following and to map the pulse propagation problem to the dynamics of a one-dimensional nonlinear pendulum. Making use of this formalism we derive the full analytic solution for EIT- and maximum-coherence based nonlinear optics in Sec. IV, taking into account drive-field and coherence depletion.

II. RESONANT NONLINEAR OPTICS IN A COHERENTLY PREPARED THREE-LEVEL SYSTEM

We first consider the propagation of pulsed EM fields in a medium of three-level atoms (Fig. 1) in which either a constant drive field mixes the two excited states $|2\rangle$ and $|3\rangle$ or in which a constant coherence between the lower two states $|1\rangle$ and $|2\rangle$ is maintained. The first case corresponds to resonant nonlinear frequency conversion based on EIT [1,2], the second one to nonlinear optics with maximum coherence [3]. The electric field propagating in the z direction is assumed to consist of three components with carrier frequencies ω_1,ω_2, and $\omega_3=\omega_1+\omega_2$:

$$E(z,t)=\sum_j \{\mathcal{E}_j(z,t)\exp[-i(\omega_j t-k_j z)]+\text{c.c.}\}. \tag{1}$$

Here $k_j=n_j\omega_j/c$ with n_j being the refractive index at frequency ω_j due to levels outside the three-level system of Fig. 1. This background refraction gives rise to the ''residual'' phase mismatch determined as

$$\Delta k=k_1+k_2-k_3. \tag{2}$$

In the approximation of slowly varying amplitudes and phases, Maxwell's propagation equations read in a moving frame

$$\frac{\partial\mathcal{E}_j}{\partial z}=i2\pi\frac{\omega_j}{c}\mathcal{P}_j, \tag{3}$$

where $\mathcal{E}_j$ and $\mathcal{P}_j$ are functions of the coordinate z and the retarded time $\tau=t-z/c$. $\mathcal{P}_j$ are the components of the medium polarization:

$$P=\sum_j \{\mathcal{P}_j\exp[-i(\omega_j t-k_j z)]+\text{c.c.}\},$$

which can be expressed in terms of the atomic probability amplitudes c_n in levels $|1\rangle$, $|2\rangle$, and $|3\rangle$:

$$\mathcal{P}_1=N\,d_1c_1^*c_2, \tag{4}$$

$$\mathcal{P}_2=N\,d_2\,c_2^*c_3e^{i\theta-i\Delta kz}, \tag{5}$$

$$\mathcal{P}_3=N\,d_3c_1^*c_3, \tag{6}$$

N being the density of active atoms. d_1, d_2, and d_3 are the real dipole moments of the transitions $|1\rangle \to |2\rangle$, $|2\rangle \to |3\rangle$, and $|1\rangle \to |3\rangle$, respectively. θ is phase of the $|2\rangle \to |3\rangle$ dipole moment, which in general cannot be chosen freely.

Assuming decay only from the topmost state $|3\rangle$ with rate γ the state amplitudes obey in the rotating-wave approximation the equations

$$\dot{c}_1 = i\Omega_1^* c_2 + i\Omega_3^* c_3,$$

$$\dot{c}_2 = i\Omega_1 c_1 + i\Omega_2^* e^{i\theta - i\Delta kz} c_3 + i\delta_2 c_2,$$

$$\dot{c}_3 = i\Omega_2 e^{-i\theta + i\Delta kz} c_2 + i\Omega_3 c_1 + i(\delta_3 + i\gamma) c_3, \tag{7}$$

where δ_2 and δ_3 are the frequency detunings indicated in Fig. 1,

$$\delta_2 = \omega_1 - \omega_{21}, \quad \delta_3 = \omega_3 - \omega_{31}, \tag{8}$$

with ω_{ij} denoting the transition frequencies between the corresponding levels. Ω_1, Ω_2, and Ω_3 are the Rabi frequencies for transitions $|1\rangle - |2\rangle$, $|2\rangle - |3\rangle$, and $|1\rangle - |3\rangle$, respectively:

$$\Omega_j = \frac{d_j \mathcal{E}_j}{2\hbar}. \tag{9}$$

A. EIT with undepleted coupling field

Here, we consider three fields interacting with the three-level system. For simplicity we assume $\delta_3 = 0$. Furthermore, we consider the case of a strong, undepleted drive field with frequency ω_2,

$$|\Omega_2| \gg |\Omega_1|, |\Omega_3|, \gamma, |\delta_2|.$$

Then, the solution of the atomic equations of motion (7) with the initial condition $c_1(t \to -\infty) = 1$ yields

$$|c_1| \approx 1.$$

Assuming quasiadiabatic evolution, i.e., not too fast changing fields, we find

$$c_2 = -\frac{\Omega_3 \Omega_2^* e^{i\theta - \Delta kz} - i\Omega_1 \gamma}{|\Omega_2|^2 - i\delta_2 \gamma},$$

$$c_3 = -\frac{\Omega_1 \Omega_2 e^{-i\theta + i\Delta kz} - \Omega_3 \delta_2}{|\Omega_2|^2 - i\delta_2 \gamma}.$$

Substitution into Maxwell's propagation equations gives

$$\frac{\partial \mathcal{E}_1}{\partial z} = -\frac{\pi N d_1^2}{\hbar} \frac{\omega_1}{c} \frac{\gamma}{|\Omega_2|^2} \mathcal{E}_1 - i \frac{\pi N d_1 d_3}{\hbar |\Omega_2|} \frac{\omega_1}{c} e^{i\theta - i\Delta kz} \mathcal{E}_3, \tag{10}$$

$$\frac{\partial \mathcal{E}_3}{\partial z} = i \frac{\pi N d_3^2}{\hbar} \frac{\omega_3}{c} \frac{\delta_2}{|\Omega_2|^2} \mathcal{E}_3 - i \frac{\pi N d_1 d_3}{\hbar |\Omega_2|} \frac{\omega_3}{c} e^{-i\theta + i\Delta kz} \mathcal{E}_1. \tag{11}$$

Equations (10) and (11) are linear differential equations, which can easily be solved. We now consider the case in which no $\mathcal{E}_3$ field is incident on the medium, $\mathcal{E}_3(z=0)=0$. Introducing the normalized intensity (photon flux)

$$\eta_j = \frac{I_j}{\hbar \omega_j} \equiv \frac{c|\mathcal{E}_j|^2}{8\pi\hbar\omega_j} \tag{12}$$

and the coupling strength

$$\mu_j = \frac{2\pi\omega_j d_j^2}{\hbar c}, \tag{13}$$

the solution of Eqs. (10) and (11) reads

$$\eta_3(z) = \frac{\eta_{10}}{1 + (\Delta k'/2\kappa)^2} e^{-\Gamma \kappa z} \sin^2[\kappa z \sqrt{1 + (\Delta k'/2\kappa)^2}], \tag{14}$$

where $\eta_{10} = \eta_1(z=0)$ is the photon flux at the entrance to the medium and we have introduced the conversion coefficient κ,

$$\kappa = \frac{N}{2} \frac{\sqrt{\mu_1 \mu_3}}{|\Omega_2|}, \tag{15}$$

the loss coefficient Γ,

$$\Gamma = \frac{\gamma}{|\Omega_2|} \sqrt{\frac{\mu_1}{\mu_3}}, \tag{16}$$

and

$$\Delta k' = \Delta k - \frac{N}{2} \frac{\mu_3 \delta_2}{|\Omega_2|^2} \tag{17}$$

is the total phase mismatch, including the background value Δk and the contribution from the resonant transition $|1\rangle \to |3\rangle$. In the EIT case, the resonant contribution to the phase mismatch is always smaller than the conversion coefficient κ by a factor $\sim \delta_2/|\Omega_2|$. Moreover, a small detuning δ_2 can be introduced to compensate the residual phase mismatch. Thus, the EIT scheme represents an ideal situation with complete phase matching, where the optimum conversion occurs for a density-length product

$$Nz|_{\text{opt}} = \pi \frac{|\Omega_2|}{\sqrt{\mu_1 \mu_3}}. \tag{18}$$

In order to minimize the absorption losses, Eq. (16), the Rabi frequency of the coupling field $|\Omega_2|$ has to be sufficiently large:

$$|\Omega_2| \gg \sqrt{\frac{\mu_1}{\mu_3}} \gamma.$$

B. Undepleted coherence ρ_{12}

In the following we discuss the scheme of resonant nonlinear optics with maximum coherence. For this situation, constant state amplitudes c_1 and c_2 are assumed, maintained, e.g., by a constant strong drive field Ω_1. In this case, the wave vector of the atomic coherence $c_1^* c_2$ is equal to k_1, which can be changed by, e.g., the small detuning δ_2 [3]. The back action of the atoms to this field is disregarded and thus the corresponding coupling does not need to be taken into account. Under the condition

$$|\delta_3 + i\gamma| \gg |\Omega_j|$$

the amplitude of the excited state can be adiabatically eliminated, which yields

$$c_3 = -\frac{\Omega_2 e^{-i\theta + i\Delta kz} c_2 + \Omega_3 c_1}{\delta_3 + i\gamma}. \tag{19}$$

Substitution of Eqs. (5), (6), and (19) into the Maxwell propagation equations (3) gives

$$\frac{\partial \mathcal{E}_2}{\partial z} = -i\frac{\pi N d_2^2}{\hbar}\frac{\omega_2}{c}\frac{\delta_3 - i\gamma}{\delta_3^2 + \gamma^2}|c_2|^2 \mathcal{E}_2 - i\frac{\pi N d_2 d_3}{\hbar}\frac{\omega_2}{c}\frac{\delta_3 - i\gamma}{\delta_3^2+\gamma^2}\rho_{12} e^{i\theta - i\Delta kz}\mathcal{E}_3, \tag{20}$$

$$\frac{\partial \mathcal{E}_3}{\partial z} = -i\frac{\pi N d_3^2}{\hbar}\frac{\omega_3}{c}\frac{\delta_3 - i\gamma}{\delta_3^2 + \gamma^2}|c_1|^2 \mathcal{E}_3 - i\frac{\pi N d_2 d_3}{\hbar}\frac{\omega_3}{c}\frac{\delta_3 - i\gamma}{\delta_3^2+\gamma^2}\rho_{12} e^{-i\theta + i\Delta kz}\mathcal{E}_2, \tag{21}$$

where $\rho_{12} = |c_1^* c_2|$.

This is again a set of linear differential equations whose solution has the same form as that for the EIT case, Eq. (14), with the substitution $\eta_{10} \to \eta_{20}$ and corresponding parameters (assuming $\delta_3 \gg \gamma$):

$$\kappa = \frac{N}{2}\frac{\sqrt{\mu_2\mu_3}}{\delta_3}\rho_{12}, \tag{22}$$

$$\Gamma = \frac{\gamma}{\delta_3}\frac{\mu_2|c_2|^2 + \mu_3|c_1|^2}{\rho_{12}\sqrt{\mu_2\mu_3}}, \tag{23}$$

$$\Delta k' = \Delta k + \frac{N}{2}\frac{\mu_3|c_1|^2 - \mu_2|c_2|^2}{\delta_3}. \tag{24}$$

The total phase mismatch $\Delta k'$ includes the background value Δk and the contributions from resonant transitions $|1\rangle \to |3\rangle$ and $|2\rangle \to |3\rangle$.

For atomic media, which we consider here, the off-resonant (background) contributions to the refractive index n_j are of the order of [9]

$$n_j \approx 1 + N\frac{c}{\omega_j}\sum_m \frac{\mu_{jm}}{\delta_{jm}}, \tag{25}$$

where μ_{jm} and δ_{jm} are the coupling constants and detunings, respectively, for the wave with frequency ω_j and transition to the far-detuned state $|m\rangle$ not belonging to the three-level system of Fig. 1. Since $\delta_{jm} \gg \delta_3$, the resonant contributions to the phase mismatch are the dominant ones. As can be seen from Eqs. (22) and (24), they are in general of the same order as the conversion coefficient κ if the atomic coherence ρ_{12} is large (of the order of 1/2). Therefore, efficient energy transfer from the ω_2 field into the ω_3 field, $\eta_3(l) \sim \eta_{20}$, occurs already within a length $l = \kappa^{-1} \sim L_c = 2/\Delta k'$, the coherence length. This feature constitutes the main advantage of the maximum-coherence scheme over conventional nonlinear optics, because the phase matching is no longer important.

If the conversion length l is much smaller than the coherence length L_c, $l/L_c \ll 1$, and the losses are small, $(\Gamma/l)z|_{\rm opt} \ll 1$, maximum conversion occurs for a density-length product

$$Nz|_{\rm opt} = \frac{\pi}{2}Nl = \pi\frac{\delta_3}{\rho_{12}\sqrt{\mu_2\mu_3}}. \tag{26}$$

$l/L_c \ll 1$ is realized when the parameters are chosen such that $\Delta k'$ is small, or even better, vanishes, i.e., if

$$\frac{2\Delta k}{N} \approx \frac{\mu_2|c_2|^2 - \mu_3|c_1|^2}{\delta_3}.$$

This can be achieved in different ways: (i) by tuning the wave vector k_1 of the atomic coherence (e.g., by introducing a detuning δ_2, as in Ref. [3]), (ii) by selecting the appropriate detuning δ_3, as in Ref. [7], and/or (iii) by preparation of atoms in a superposition with suitable amplitudes c_1, c_2.

In order for the absorption losses to be negligible within the optimum propagation distance, the following condition has to be satisfied:

$$\frac{\gamma}{\delta_3}\frac{1}{\rho_{12}} \ll 1,$$

which indicates once again that it is indeed advantageous to prepare a large atomic coherence ρ_{12}.

C. Conventional nonlinear optics: Weak excitation

Now, we discuss the case of a weak excitation of atoms which corresponds to the regime of conventional nonlinear optics. The weak excitation takes place, for example, when both detunings are very large:

$$|\delta_3|, |\delta_2| \gg |\Omega_j|, \gamma.$$

Under this condition, the atomic probability amplitudes are

$$|c_1| \approx 1,$$

$$c_2 = -\frac{\Omega_1\Omega_2 e^{-i\theta + i\Delta kz} - \Omega_3\delta_2}{\delta_2(\delta_3 + i\gamma)},$$

$$c_3 = -\frac{\Omega_3\Omega_2^* e^{i\theta - i\Delta kz} - \Omega_1(\delta_3 + i\gamma)}{\delta_2(\delta_3 + i\gamma)}.$$

Since $|c_2|,|c_3| \ll |c_1|$, the medium polarization at frequency ω_2 is much smaller than for the ω_1 and ω_3 components: $|\mathcal{P}_2| \ll |\mathcal{P}_1|,|\mathcal{P}_3|$. Therefore, we can assume that the ω_2 field is almost undepleted, $|\Omega_2| \approx \mathrm{const}(z)$.

The propagation equations read for this case

$$\frac{\partial \mathcal{E}_1}{\partial z} = -i\frac{\pi N d_1^2}{\hbar\delta_2}\frac{\omega_1}{c}\mathcal{E}_1 + i\frac{\pi N d_1 d_3 \omega_1}{\hbar c} \times \frac{|\Omega_2|}{\delta_2\delta_3}\left(1 - i\frac{\gamma}{\delta_3}\right)e^{i\theta - i\Delta kz}\mathcal{E}_3, \tag{27}$$

$$\frac{\partial \mathcal{E}_3}{\partial z} = -i\frac{\pi N d_3^2}{\hbar}\frac{\omega_3}{c}\frac{\delta_3 - i\gamma}{\delta_3^2 + \gamma^2}\mathcal{E}_3 - i\frac{\pi N d_1 d_3 \omega_3}{\hbar c} \times \frac{|\Omega_2|}{\delta_2\delta_3}\left(1 - i\frac{\gamma}{\delta_3}\right)e^{-i\theta + i\Delta kz}\mathcal{E}_1. \tag{28}$$

The solution of this set of propagation equations has exactly the same form as in the maximum-coherence and EIT cases, Eq. (14), with parameters

$$\kappa = \frac{N}{2}\frac{|\Omega_2|\sqrt{\mu_1\mu_3}}{\delta_2\delta_3}, \tag{29}$$

$$\Gamma = \frac{\gamma}{\delta_3}\sqrt{\frac{\mu_1}{\mu_3}}\frac{\delta_2}{|\Omega_2|}, \tag{30}$$

$$\Delta k' = \Delta k + \frac{N}{2}\left(\frac{\mu_3}{\delta_3} - \frac{\mu_1}{\delta_2}\right). \tag{31}$$

We see that the conversion length $l = \kappa^{-1}$ here is much larger than both in the maximum-coherence, Eq. (22), and in the EIT, Eq. (15), schemes. In fact, the conversion length is also much larger than the (non-phase-matched) coherence length $L_c = 2/\Delta k'$ as $\delta_{2,3}/|\Omega_2| \gg 1$. Therefore, it is not possible in conventional nonlinear optics to get the complete conversion, $\eta_3 \sim \eta_{10}$, without careful phase matching.

If such a compensation is performed, i.e., if $(\kappa/2\Delta k')^2 \gg 1$ (by proper choice of detunings δ_2 and δ_3), optimum conversion occurs for a density-length product

$$Nz|_{\mathrm{opt}} = \frac{\pi}{2}Nl = \pi\frac{\delta_2\delta_3}{\sqrt{\mu_1\mu_3}|\Omega_2|}. \tag{32}$$

At this optimum propagation distance, the relative absorption losses are given by the parameter Γ, Eq. (30).

Thus, we conclude that in the limit of undepleted drive field(s) both EIT and maximum-coherence schemes perform much better than conventional nonlinear optics, regarding the number of atoms $Nz|_{\mathrm{opt}}$ necessary for optimum conversion and robustness to phase mismatch. The aim of the present paper is to investigate whether the EM energy can be transferred to the generated wave from both pump fields, and whether the attractive features of EIT- and maximum-coherence-assisted conversion survive when the drive field is depleted. This requires the solution of the complete nonlinear propagation problem, which we treat with the Hamiltonian formalism outlined in the following section.

III. HAMILTONIAN APPROACH

The resonant interaction of EM fields with atomic systems is described by Maxwell's equations for the fields and a set of master equations for the density matrix of the atoms. The solution of this coupled set is rather difficult and except for some very special cases impossible analytically. In order to derive analytical solutions for the field propagation, approximations are needed that allow to eliminate the atomic degrees of freedom and to express the polarization in terms of field variables. A common approximation, which is not perturbative in the fields, is the adiabatic solution of the density-matrix equations. This straightforward approach provides, when applicable, full information about both atoms and fields in the adiabatic limit.

A. General formalism

Instead of the usual explicit scheme of adiabatic elimination which is rather cumbersome, we use here a different, implicit approach [11,12] which yields directly effective-field equations. This approach is based on the representation of the medium polarization P as a partial derivative of the time-averaged free-energy density of a dielectric with respect to the electric field E [17]:

$$P = -\left\langle N\frac{\partial \hat{H}}{\partial E}\right\rangle, \tag{33}$$

where $\langle\cdots\rangle$ denotes quantum-mechanical averaging, and $\hat{H}$ is the single-atom interaction Hamiltonian. For the field given by Eq. (1), we can write

$$P = -\left\langle N\sum_j \frac{\partial \hat{H}}{\partial \mathcal{E}_j^*}\exp[-i(\omega_j t - k_j z)] + \mathrm{c.c.}\right\rangle,$$

so that the propagation equation becomes

$$\frac{\partial \mathcal{E}_j}{\partial z} = -i2\pi\frac{\omega_j}{c}N\left\langle\frac{\partial \hat{H}}{\partial \mathcal{E}_j^*}\right\rangle. \tag{34}$$

We here consider light-atom interaction processes that are adiabatic, that is, the atomic system can be assumed to follow the evolution of the instantaneous eigenstates. For example, if the atomic system is at some initial time t_0 in the nondegenerate eigenstate $|\psi_0(t_0)\rangle$ of the interaction Hamiltonian, i.e.,

$$\hat{H}|\psi_0\rangle = \hbar\lambda_0|\psi_0\rangle, \tag{35}$$

which is at $t = t_0$ usually identical to the ground state of the atoms, it will remain in this state at all times. Furthermore, we disregard irreversible dissipation processes. As we show later, for the processes we consider here, this is justified even for rather long pulses, i.e., longer than the natural lifetime γ^{-1} of the excited state $|3\rangle$. In this limit one finds

$$\left\langle \frac{\partial \hat{H}}{\partial \mathcal{E}_j^*} \right\rangle = \left\langle \psi_0 \left| \frac{\partial \hat{H}}{\partial \mathcal{E}_j^*} \right| \psi_0 \right\rangle = \hbar \frac{\partial \lambda_0}{\partial \mathcal{E}_j^*}.$$

Hence the propagation equation can be written as

$$\frac{\partial \mathcal{E}_j}{\partial z} = -i 2\pi \frac{\hbar \omega_j}{c} N \frac{\partial \lambda_0}{\partial \mathcal{E}_j^*}. \tag{36}$$

For the following it is useful to express the field amplitude $\mathcal{E}_j$ in terms of photon flux η_j, Eq. (12), and phase φ_j:

$$\mathcal{E}_j = |\mathcal{E}_j| \exp\{-\varphi_j\}.$$

Separating the real and imaginary parts, we find from Eq. (36),

$$\frac{\partial \eta_j}{\partial z} = -\frac{\partial \mathcal{H}'}{\partial \varphi_j}, \tag{37}$$

$$\frac{\partial \varphi_j}{\partial z} = \frac{\partial \mathcal{H}'}{\partial \eta_j}.$$

These equations have the form of Hamilton equations of classical canonical mechanics with action and angle variables η_j, and φ_j "time," z, and the Hamiltonian function $\mathcal{H}' = \frac{1}{2} N \lambda_0$.

B. Constants of motion for resonant three-wave mixing

For atomic systems with a closed loop of transitions, the set of canonical equations (37) can be further simplified and in fact under some conditions explicitly integrated. This is the case when the number of coherent fields involved is not too large. To illustrate the procedure, let us consider the eigenvalue equation (35) for the three-level system in Fig. 1. In the rotating-wave approximation, the light-atom interaction Hamiltonian is given by

$$\hat{H} = -\hbar[\delta_2 |2\rangle\langle 2| + \delta_3 |3\rangle\langle 3|] - \hbar \Omega_1 |1\rangle\langle 2| - \hbar \Omega_2 e^{i\varphi} |2\rangle\langle 3| - \hbar \Omega_3 |1\rangle\langle 3| + \text{H.c.}, \tag{38}$$

and the eigenvalues are determined by the characteristic equation

$$\lambda_0(\delta_2 + \lambda_0)(\delta_3 + \lambda_0) - (\Omega_1^2 + \Omega_2^2 + \Omega_3^2)\lambda_0 - \Omega_1^2 \delta_3 - \Omega_3^2 \delta_2 = -2\Omega_1 \Omega_2 \Omega_3 \cos\varphi. \tag{39}$$

Here Ω_j are the Rabi frequencies, related to the photon flux via the coefficients μ_j, Eq. (13), as $\Omega_j = \sqrt{\mu_j \eta_j}$.

The relative phase φ of the EM waves is

$$\varphi = \varphi_1 + \varphi_2 - \varphi_3 - \Delta k z, \tag{40}$$

which includes the residual phase mismatch Δk. Also the multiphoton resonance condition

$$\omega_3 = \omega_1 + \omega_2 \tag{41}$$

has been used.

One can see from Eq. (39) that λ_0 and, hence $\mathcal{H}'$, depend on the field phases φ_j only through the relative phase φ. Therefore, we have

$$\frac{\partial \mathcal{H}'}{\partial \varphi_1} = \frac{\partial \mathcal{H}'}{\partial \varphi_2} = -\frac{\partial \mathcal{H}'}{\partial \varphi_3} \left(= \frac{\partial \mathcal{H}'}{\partial \varphi} \right). \tag{42}$$

An immediate consequence of this symmetry of $\mathcal{H}'$ is the existence of constants of motion. Substituting the above Eqs. (42) into the first line of Eqs. (37) yields the well-known Manley-Rowe relations [9]:

$$\frac{\partial \eta_1}{\partial z} = \frac{\partial \eta_2}{\partial z} = -\frac{\partial \eta_3}{\partial z}, \tag{43}$$

which correspond to two independent constants of motion:

$$\eta_1 + \eta_3 = \eta_{10} + \eta_{30}, \tag{44}$$

$$\eta_1 - \eta_2 = \eta_{10} - \eta_{20}.$$

Here $\eta_{j0} = \eta_j(z=0)$ are the photon flux values at the entrance to the medium. Taking into account the multiphoton resonance condition (41), one finds, furthermore, that the total intensity of the EM fields is conserved: $I_1 + I_2 + I_3 = \text{const}(z)$. The Manley-Rowe relations and the constants of motion tell us that in the process under consideration, the energy is transferred from the frequency components ω_1, ω_2 into ω_3 and back, with equal rates and without losses, which of course is expected for a dissipationless nonlinear medium.

The relations Eq. (44) enable us to rewrite η_j as

$$\eta_1(z) = \eta_{10} - J(z),$$

$$\eta_2(z) = \eta_{20} - J(z), \tag{45}$$

$$\eta_3(z) = \eta_{30} + J(z).$$

$J(z)$ characterizes the amount of energy exchange between the waves and has the initial condition $J(z=0) = 0$.

Thus the original problem with six amplitude and phase variables can be reduced to two variables J and φ by a canonical transformation. This leads to

$$\frac{\partial J}{\partial z} = -\frac{\partial \mathcal{H}}{\partial \varphi}, \tag{46}$$

$$\frac{\partial \varphi}{\partial z} = \frac{\partial \mathcal{H}}{\partial J}, \tag{47}$$

with the new Hamiltonian function

$$\mathcal{H} = \frac{1}{2} N \lambda_0 + \Delta k J \equiv \frac{1}{2} N \lambda. \tag{48}$$

As can be seen from Eqs. (48) and (39), $\mathcal{H}$ (or λ) does not depend on the coordinate z explicitly. Therefore, $\mathcal{H}$ (or λ) is a fourth constant of motion expressing the conservation of the energy density of the medium with respect to z.

C. Solution of the wave-propagation problem

To solve the remaining two equations of motion for $J(z)$ and $\varphi(z)$, the Rabi frequencies Ω_j are expressed in terms of η_{j0} and J, and the characteristic equation (39) is written in the form

$$G(\lambda,J)=g(J)\cos\varphi. \tag{49}$$

Differentiating both sides with respect to φ yields

$$\frac{\partial G}{\partial\varphi}=\frac{\partial G}{\partial\lambda}\frac{\partial\lambda}{\partial\varphi}=-g\sin\varphi=\pm\sqrt{g^2-G^2}.$$

Substituting this relation into Eq. (46), we find

$$\frac{\partial J}{\partial z}=\pm\frac{N}{2}\frac{\sqrt{g^2-G^2}}{\partial G/\partial\lambda}. \tag{50}$$

The choice of the sign in Eq. (50) depends on the sign of $\sin\varphi$ at $z=0$. Integration of Eq. (50) gives an implicit solution for $J(z)$:

$$\pm\frac{N}{2}z=\int_0^J\frac{\partial G(J')}{\partial\lambda}\frac{dJ'}{\sqrt{g^2(J')-G^2(J')}}. \tag{51}$$

To analytically evaluate the remaining integral, we note that both functions g^2-G^2 and $\partial G/\partial\lambda$ are polynomials in J:

$$g=-2\sqrt{\mu_1\mu_2\mu_3}\sqrt{(\eta_{10}-J)(\eta_{20}-J)(\eta_{30}+J)},$$

$$G=G_0+\sum_{m=1}^{3}A_mJ^m,$$

$$\frac{\partial G}{\partial\lambda}=\sum_{m=0}^{2}a_mJ^m.$$

Therefore, Eq. (50) describes a one-dimensional finite motion of a pendulum in an external potential. The solution is, in general, given by some combination of elliptic functions [18] with parameters determined mainly by the roots J_n of the polynomial equation:

$$g^2(J)-G^2(J)=0. \tag{52}$$

The allowed range of J, corresponding to the region of classically allowed motion of the pendulum, lies between zero and the smallest positive root J_1 of the polynomial (52).

The eigenvalue λ is a constant of motion [cf. Eq. (48)], and can thus be found from the characteristic equation (49) with parameters taken at the medium entrance $z=0$:

$$G_0(\lambda)=g(z=0)\cos\varphi(z=0). \tag{53}$$

Thus, we have reduced the propagation problem to solving two algebraic equations: Eq. (53) for λ and Eq. (52) for the roots J_n. If this can be done explicitly, the Hamiltonian method provides an analytical solution to the propagation problem. But even if an explicit solution is not possible, it considerably simplifies numerical calculations. Apart from the advantage of being convenient from the formal point of view, the Hamiltonian method allows a deeper insight into the nature of the wave-propagation process. For example, the method provides a direct access to the stability analysis of the solution by referring to a well-developed theory of Hamiltonian systems [14].

To understand the dynamics of the system, in particular, interesting quantities such as the conversion efficiency, it is useful to discuss the physical meaning of the coefficients A_m and a_m. Considering the canonical equation (47) for the relative phase:

$$\frac{\partial\varphi}{\partial z}=\frac{N}{2}\frac{\partial\lambda}{\partial J}=\frac{N}{2}\frac{\partial G/\partial J}{\partial G/\partial\lambda}=\frac{N}{2}\frac{A_1+2A_2J+3A_3J^2}{a_0+a_1J+a_2J^2}, \tag{54}$$

one recognizes that the A_m and a_m describe the linear and nonlinear refraction coefficients of the medium. For example, if J is sufficiently small, the first term $NA_1/2a_0$ on the right-hand side of Eq. (54) can be identified with the phase mismatch induced by the linear refraction, including both contributions from the three-level interaction and the residual mismatch Δk.

D. Generation of field with frequency ω_3

In the context of the present discussion we are most interested in the generation of the ω_3 mode from vacuum $\eta_{30}=0$. In this case, the eigenvalue equation (53) reduces to

$$G_0(\lambda)=\lambda(\lambda+\delta_2)(\lambda+\delta_3)-\mu_1\eta_{10}(\lambda+\delta_3)-\mu_2\eta_{20}\lambda=0. \tag{55}$$

The nonvanishing coefficients in the expansion of G and $\partial G/\partial\lambda$ are given by

$$A_1=q[(\lambda+\delta_2)(\lambda+\delta_3)+\lambda(\lambda+\delta_2)+\lambda(\lambda+\delta_3)-\mu_1\eta_{10}-\mu_2\eta_{20}]+\mu_1(\lambda+\delta_3)+\mu_2\lambda-\mu_3(\lambda+\delta_2), \tag{56}$$

$$A_2=q^2(3\lambda+\delta_2+\delta_3)+q(\mu_1+\mu_2-\mu_3), \tag{57}$$

$$A_3=q^3, \tag{58}$$

$$a_0=3\lambda^2+2\lambda(\delta_2+\delta_3)+\delta_2\delta_3-\mu_1\eta_{10}-\mu_2\eta_{20}, \tag{59}$$

$$a_1=2q(3\lambda+\delta_2+\delta_3)+(\mu_1+\mu_2-\mu_3), \tag{60}$$

$$a_2=3q^2, \tag{61}$$

where

$$q=-2\Delta k/N. \tag{62}$$

With this one finds for the denominator in the integral (51), which determines the allowed values of J:

$$g^2-G^2=4\mu_1\mu_2\mu_3J(\eta_{10}-J)(\eta_{20}-J)-(A_1+A_2J+A_3J^2)^2J^2=0. \tag{63}$$

The second term in this expression is nonpositive, so the smallest positive root of the polynomial is bounded by the minimum of η_{10} and η_{20}. This reflects the fact that the conversion process stops when the energy of the weaker of the two pump fields is entirely depleted. In order to reach this limit and thus in order to attain maximum conversion efficiency, the second term in Eq. (63) should be small, which corresponds to the phase mismatch to be negligible:

$$A_1+A_2J+A_3J^2\approx 0. \qquad (64)$$

In order to see what parameters are required to approximately satisfy this condition, we have to analyze the coefficients A_m.

It is worthwhile to point out that the behavior of the total phase is described by Eq. (49):

$$\cos\varphi(z)=\frac{G}{g}=\frac{A_1J+A_2J^2+A_3J^3}{2\sqrt{\mu_1\mu_2\mu_3 J(\eta_{10}-J)(\eta_{20}-J)}}. \qquad (65)$$

When the total phase mismatch is compensated, $A_1+A_2J+A_3J^2\approx 0$, the phase follows the equation

$$\cos\varphi(z)=0,$$

that is, the phase is constant and equal to $+\pi/2$ or $-\pi/2$ when all of the waves are present, and jumps by π to $-\pi/2$ or $+\pi/2$ when one of the wave intensities approaches zero.

As it is seen from Eqs. (57), (58), the terms A_2, A_3 responsible for the intensity-dependent refractive index are both proportional to the residual phase mismatch Δk. One can show that in atomic (molecular) media, where the background refractive index is given by Eq. (25), Δk is always sufficiently small so that the leading term in the phase mismatch is A_1, while A_2J and A_3J^2 are negligibly small. It is also true that the influence of the intensity-dependent phase mismatch remains insignificant even when the linear refraction is compensated: $A_1=0$. Therefore, in what follows, we will disregard the terms A_2J and A_3J^2. In this case, the solution of the propagation equation (51) gives the following dependence of $J(z)$ in implicit form:

$$\pm\kappa z+\chi_0=F[\gamma_1(J),p]+\frac{a_1J_2}{a_0}\left\{F[\gamma_2(J),p]+\left(1-\frac{J_1}{J_2}\right)\Pi[\gamma_2(J),p^2,p]\right\}, \qquad (66)$$

where χ_0 is an integration constant, and $F(\gamma,p)$ and $\Pi(\gamma,r,p)$ are the elliptic integrals of the first and third kind, respectively [18]. κ is the nonlinear conversion coefficient defined as

$$\kappa=\frac{N}{2}\frac{\sqrt{\mu_1\mu_2\mu_3J_2}}{a_0}. \qquad (67)$$

The parameters of the elliptic integrals are

$$\gamma_1(J)=\arcsin\sqrt{\frac{J}{J_1}},$$

$$\gamma_2(J)=\arcsin\sqrt{\frac{J_2(J_1-J)}{J_1(J_2-J)}},$$

$$p=\sqrt{\frac{J_1}{J_2}} \qquad (68)$$

with J_1 and J_2 being the roots of Eq. (63):

$$J_{2,1}=\frac{1}{2}(\eta_{10}+\eta_{20}+B_1)\pm\frac{1}{2}\sqrt{(\eta_{10}+\eta_{20}+B_1)^2-4\eta_{10}\eta_{20}}, \qquad (69)$$

$$B_1=\frac{A_1^2}{4\mu_1\mu_2\mu_3}.$$

In the following section we discuss the solution of the propagation problem for several field-atom interaction configurations resulting in the EIT-assisted, maximum-coherence, and conventional nonlinear-optical regimes of the frequency conversion.

IV. ANALYTICAL SOLUTIONS FOR RESONANT THREE-WAVE MIXING

We assume in our simple three-level model system that the transitions $|1\rangle-|3\rangle$, $|2\rangle-|3\rangle$ as well as $|1\rangle-|2\rangle$ are allowed. To take into account the much weaker coupling between levels $|1\rangle$ and $|2\rangle$, which in reality is a two-photon transition, we assume $\mu_1\ll\mu_2$. An analytic solution for the more general four-wave-mixing system can also be obtained within the Hamiltonian formalism [15]. The explicit results are, however, rather lengthy and not very instructive.

A. EIT-based up-conversion

The EIT scheme implies two-photon resonance between the states $|1\rangle$ and $|3\rangle$: $\delta_3=0$. According to the idea of EIT-assisted nonlinear optics, we suppose here that $\eta_{20}\gg\eta_{10}$. Since $\mu_2\gg\mu_1$, we always have $\Omega_2\gg\Omega_1$. We allow, for the moment, for spontaneous decay from state $|3\rangle$ out of the system, but require the initial Rabi frequency Ω_{20} to be much larger than the decay rate: $\Omega_{20}\gg\gamma$. Under these conditions, the eigenstate that asymptotically connects to the ground state of atoms the for $t\rightarrow-\infty$ is the state

$$|\psi_0(z=0)\rangle\approx\frac{\Omega_{20}}{\sqrt{\Omega_{10}^2+\Omega_{20}^2}}|1\rangle-\frac{\Omega_{10}e^{-i\varphi}}{\sqrt{\Omega_{10}^2+\Omega_{20}^2}}|3\rangle,$$

corresponding to the complex eigenvalue

$$\lambda\approx i\gamma\frac{\Omega_{10}^2}{\Omega_{20}^2}.$$

Since $\Omega_{10}\ll\Omega_{20}$, one has $|\lambda|\ll\gamma$. Correspondingly, the probability of spontaneous decay of the state $|\psi_0\rangle$, which can be defined as $\mathrm{Im}(\lambda)\tau$ with τ being the characteristic pulse length, remains small even for rather long pulses: $\tau \ll \tau_0 \equiv (\Omega_{20}^2/\Omega_{10}^2)\gamma^{-1}$ with $\tau_0 \gg \gamma^{-1}$. For such pulses, we can safely disregard the spontaneous emission and put $\lambda \approx 0$, which means that the corresponding adiabatic state is the usual dark state of EIT. Moreover, since $\Omega_{10} \ll \Omega_{20}$ the eigenstate $|\psi_0(z=0)\rangle$ is an only slightly disturbed ground state $|1\rangle$. Therefore, the requirement of adiabaticity of the process is satisfied automatically at the medium entrance, $z=0$.

The coefficients A_1 and a_m are given for the regime of EIT by the following expressions:

$$A_1 \simeq -q\mu_2\eta_{20} - \mu_3\delta_2, \tag{70}$$

$$a_0 \simeq -\mu_2\eta_{20}, \tag{71}$$

$$a_1 = 2q\delta_2 + (\mu_1 + \mu_2 - \mu_3). \tag{72}$$

1. Undepleted EIT-generating field

In the original proposal of EIT-assisted frequency conversion, the ω_2 field is assumed to be very strong and undepleted. The latter condition implies $\eta_{10} \ll \eta_{20}$. In this situation the solution (66) can be well approximated by

$$J = \frac{\eta_{10}}{1+\left(\frac{\Delta k'}{2\kappa_e}\right)^2} \sin^2\left[\kappa_e z\sqrt{1+\left(\frac{\Delta k'}{2\kappa_e}\right)^2}\right], \tag{73}$$

$$\kappa_e = \frac{N}{2}\sqrt{\frac{\mu_1\mu_3}{\mu_2\eta_{20}}}, \tag{74}$$

$$\Delta k' = \Delta k - \frac{N}{2}\frac{\mu_3\delta_2}{\mu_2\eta_{20}}, \tag{75}$$

which, of course, coincides with that obtained in Sec. II, Eqs. (14) and (15), under the undepleted drive approximation. The influence of the intensity-dependent phase mismatch (terms A_2J and A_3J^2) is negligible in this case, as was discussed above.

One can immediately see from Eqs. (70), (57), and (58) that for vanishing residual phase mismatch $q=0$ (which can be realized by adding a buffer gas with proper dispersion [19]) and for $\delta_2=0$, there is perfect phase matching: $A_1 = A_2 = A_3 = 0$. Consequently maximum energy transfer into the generated wave ω_3 is possible. This applies also for the case when linear refraction is compensated by a detuning δ_2:

$$\delta_2 = \frac{2}{N}\frac{\mu_2\eta_{20}}{\mu_3}\Delta k. \tag{76}$$

For both of these phase-matching procedures, the maximum value of $\eta_3 = J$ achieved at $z_e = \pi/2\kappa_e$ is equal to η_{10}. Thus the instantaneous fractional conversion efficiency defined as

$$\epsilon \equiv \frac{J_{\max}}{\min(\eta_{10},\eta_{20})} \tag{77}$$

can become unity. It should be noted that $\epsilon = 1$ is, in general, only achieved at a specific instant of time since κ_e depends on time via $\eta_{20}(t-z/c) \equiv \overline{\eta_{20}} f_{20}(t-z/c)$ [$\overline{\eta_{20}}$ is the amplitude and $f_{20}(t-z/c)$ is the temporal envelope of the $\eta_{20}(t-z/c)$ pulse]. Hence the transfer of energy is complete only for the part of the ω_1 pulse which satisfies the condition $\sqrt{f_{20}(t-z/c)} \approx 1$. A high overall conversion requires a flat temporal profile of the "coupling" pulse ω_2. This is easy to implement, however, since always $\eta_3 = J \ll \eta_2$ ($\approx \eta_{20}$) and $\eta_1 \ll \eta_{20}$. We have then $\Omega_1, \Omega_3 \ll \Omega_2$ for the whole medium, which means, in particular, that all the atoms throughout the medium are essentially in the ground state. Therefore, the ω_2 pulse can be arbitrarily long.

In contrast to the fractional conversion efficiency, Eq. (77), the total conversion efficiency defined as

$$W(z) \equiv \frac{\int dt\,\omega_3\eta_3(z,t)}{\int dt[\omega_1\eta_1(z,t) + \omega_2\eta_2(z,t)]} \tag{78}$$

remains very small because $\eta_{10} \ll \eta_{20}$.

2. Depletion of the pump fields

Conversion with $\varepsilon \approx 1$ can be achieved, however, also for the case when both pump fields ω_1 and ω_2 are of comparable intensity. For example, for exact phase matching and $\eta_{10} = \eta_{20} \equiv \eta_0$ (which would correspond, e.g., to the second harmonic generation), the solution Eq. (66) can be written as

$$\frac{\mu_2}{\mu_3}\kappa_e z = \arctan\left(\sqrt{\frac{J}{\eta_0}}\right) - \frac{(\mu_1+\mu_2-\mu_3)}{\mu_3}\sqrt{\frac{J}{\eta_0}}, \tag{79}$$

which demonstrates that J monotonically approaches η_0 as z increases. The form of this solution for given retarded time is shown in Fig. 2. The monotonic dependence ensures that after a sufficiently long propagation length all parts of the pulse $\eta_{10} = \eta_{20}$ are converted into the ω_3 wave so that not only $\varepsilon \approx 1$, but also the total energy conversion efficiency W reaches unity.

When the phase mismatch is not compensated, the solution for $\eta_{10} = \eta_{20} \equiv \eta_0$ is given by a more general expression, Eq. (66), with $B_1 = \eta_0(\Delta k'/2\kappa_e)^2$ and $J_{2,1} \approx \eta_0(1 \pm \Delta k'/2\kappa_e)$. The spatial dependence of the generated field intensity (Fig. 3) differs only slightly from

$$J = \eta_0\left(1 - \frac{\Delta k'}{2\kappa_e}\right)\mathrm{sn}^2\left[\kappa_e z\sqrt{1+\frac{\Delta k'}{2\kappa_e}};\sqrt{\frac{J_1}{J_2}}\right],$$

where $\mathrm{sn}[x;p]$ is the Jacobi elliptic sine function. The period of intensity oscillations seen in Fig. 3 can be estimated as [18]

$$z_e \simeq \ln(16\kappa_e/\Delta k').$$

Since in the EIT regime we have always $\Delta k' \ll 2\kappa_e$, very high total conversion efficiency is achieved without phase matching also for $\eta_{10} \approx \eta_{20}$ when the drive fields are substantially depleted.

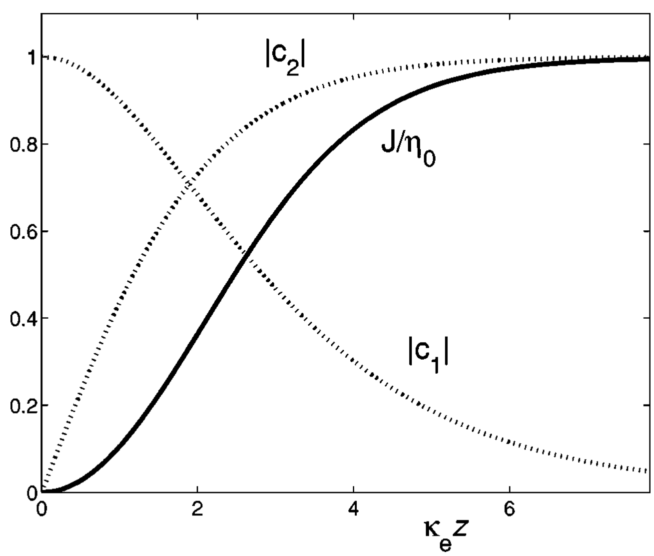

FIG. 2. EIT up-conversion for perfect phase matching. Solid line: spatial evolution of J according to Eq. (79) for given retarded time. For sufficiently large propagation length there is complete conversion. Dotted lines: corresponding spatial evolution of the atomic bare state probabilities. The amplitude of the excited state $|3\rangle$ is negligibly small (not shown). Parameters: $\mu_2/\mu_3=0.5$, $\mu_1/\mu_3=0.05$.

3. Evolution of the atomic state

In the case of EIT with comparable intensities of the ω_1 and ω_2 pump fields, the intensity of the ω_3 wave increases with z and eventually exceeds that of the ω_1,ω_2 waves. Therefore, the adiabatic state of the atomic system will no longer approximately coincide with the ground state $|1\rangle$ over the entire medium length at a given retarded time. It will evolve via different superpositions of the bare states $|1\rangle$ and $|2\rangle$ following the change of the fields (Figs. 2 and 3) [20]. We see that the "conventional" EIT regime (one strong and one weak field, and thus one highly populated and one slightly populated state) does not hold over the full conversion cycle. We should stress, however, that the atoms remain in the adiabatic state corresponding to $\lambda=0$, i.e., in the dark state. Therefore, all the features of the EIT-assisted conversion process such as vanishing linear absorption and refraction as well as resonantly enhanced nonlinearity are present even when the pump field is depleted.

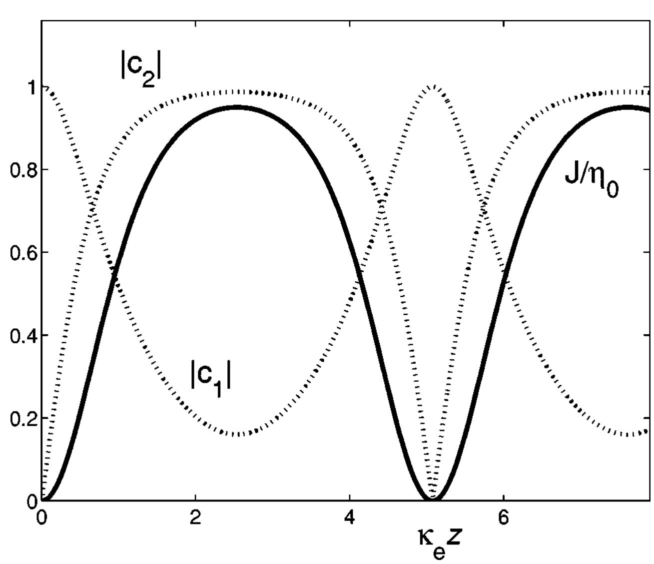

FIG. 3. EIT up-conversion with finite phase mismatch. Solid line: spatial evolution of J. Dotted lines: corresponding spatial evolution of the atomic bare state probabilities. The amplitude of the excited state $|3\rangle$ is negligibly small (not shown). Parameters are the same as in Fig. 2, $\Delta k'/2\kappa_e=0.06$.

When most of the input energy is transferred to the ω_3 pulse, i.e., for larger propagation distances into the medium, the dark state $|\psi_0\rangle$ approaches the bare state $|2\rangle$,

$$|\psi_0\rangle \rightarrow |2\rangle,$$

which does not coincide with the initial atomic state (i.e., the ground state $|1\rangle$). Thus we encounter two problems: First, since all atoms are in the ground state before the arrival of the pulses, there must be an adiabatic transfer in the initial phase from $|1\rangle$ to $|\psi_0\rangle$. This requires a specific time order of the pulses. In particular, Ω_2 should arrive after the other pulses. This time ordering is, however, not reflected in our solutions. Second, after the interaction with the pulses, there is energy left in the atomic system. This is in apparent contradiction to the fact that full conversion of photon energy has taken place and no energy got lost. It should be noted though that the size of the effect is rather small in the considered limit of a large initial number of photons. This contradictory behavior is a result of the limited validity of the assumptions made. We have here completely neglected nonadiabatic corrections, which are small on the level of individual atoms, but add up in the field evolution when integrated over the whole sample. The most important effect ignored by this is the group-velocity delay of the weak fields.

Although the group delay cannot be treated within the current approach, it may resolve the problems. In the first part of the medium, i.e., for small Nz the generated field is still small. Thus the group velocities of the pulses Ω_1 and Ω_3 should be smaller than that of the strong drive field Ω_2. Then the leading edge of the Ω_2 pulse will always propagate ahead of the other pulses, guaranteeing that the adiabatic state $|\psi_0\rangle$ asymptotically matches with the ground state at early times for all values of z. For larger propagation distances into the medium and for times in the central part of the pulses, a substantial portion of the energy is transferred to the field Ω_3. Now the situation is reversed. Ω_3 is strong and Ω_2 is weak. Thus the pulse Ω_2 will have a smaller group velocity and should lag behind at the trailing edge of the pulses. Consequently also for large times, the adiabatic eigenstate $|\psi_0\rangle$ approaches asymptotically the ground state $|1\rangle$.

A full quantitative account of the leading-order nonadiabatic corrections requires, however, a reformulation of the Hamiltonian approach. This is beyond the scope of the present work and will be presented in a future publication.

B. Maximum-coherence case

The mechanism of nonlinear optics with maximum coherence is realized when the detuning δ_3 is much larger than the other parameters including all Rabi frequencies and δ_2. In this case the coefficients A_1 and a_m are as follows:

$$A_1 \simeq q[(2\lambda+\delta_2)\delta_3-\mu_1\eta_{10}-\mu_2\eta_{20}]+\mu_1\delta_3+\mu_2\lambda - \mu_3(\lambda+\delta_2), \tag{80}$$

$$a_0 \simeq (2\lambda+\delta_2)\delta_3-\mu_1\eta_{10}-\mu_2\eta_{20}, \tag{81}$$

$$a_1 = 2q\delta_3+(\mu_1+\mu_2-\mu_3). \tag{82}$$

1. Preparation of maximum atomic coherence

The energy eigenvalue for large δ_3 is given by

$$\lambda = -\frac{1}{2}\left(\delta_2-\frac{\Omega_{20}^2}{\tilde{\delta}_3}\right)+\frac{1}{2}\sqrt{\left(\delta_2-\frac{\Omega_{20}^2}{\tilde{\delta}_3}\right)^2+4\Omega_{10}^2}, \tag{83}$$

where $\tilde{\delta}_3=\delta_3+i\gamma$. The amplitudes of the adiabatic state corresponding to this eigenvalue at the entrance to the medium, $z=0$, are

$$c_1=\frac{\Omega_{10}}{\sqrt{\lambda^2+\Omega_{10}^2}}, \tag{84}$$

$$c_2=-\frac{\lambda}{\sqrt{\lambda^2+\Omega_{10}^2}}, \tag{85}$$

$$c_3 \ll c_1, c_2.$$

Similar to the EIT case, the probability of spontaneous decay of the state $|\psi_0\rangle$ is small: $\mathrm{Im}(\lambda)\tau \ll 1$ for large detuning assumed here, $\delta_3 \gg \gamma$, even for pulses that are longer than γ^{-1}: $\tau \ll (\delta_3^2/\Omega_{20}^2)\gamma^{-1}$. Again, we will consider only such pulses and neglect spontaneous emission.

The situation of maximum coherence, $|c_1|=|c_2|=1/\sqrt{2}$, takes place when the Rabi frequency exceeds the detuning on transition $|1\rangle-|2\rangle$ including the ac Stark shift induced by the ω_2 field: $\Omega_{10} \gg |\delta_2-\Omega_{20}^2/\delta_3|$. In this case $\lambda \approx \Omega_{10}$ and the atomic state at the medium entrance is the superposition: $|\psi_0(z=0)\rangle=(|1\rangle-|2\rangle)/\sqrt{2}$. However, in order for the atoms to get there from the ground state, one first has to have the inverse situation $\delta_2 \gg \Omega_{10}$, and then adiabatically decrease δ_2, simultaneously increasing Ω_{10}. This can be accomplished by, e.g., Stark-chirped rapid adiabatic passage [8]. We will not include this process explicitly in the consideration, but assume that when the ω_2 pulse arrives, the condition $\Omega_{10} \gg \delta_2$ is already satisfied. As a consequence, the energy taken from the leading edge of the ω_1 pulse for the superposition preparation is not taken into account. However, if the number of photons in the ω_1 pulse is much larger than the total number of atoms along the field propagation path, these preparation energy losses ($\hbar\omega_1/2$ per atom) can be neglected.

2. Undepleted coherence-generating field

When a strong, undepleted ω_1 field is supposed: $\eta_{20} \ll \eta_{10}$ (implying also undepleted coherence on $|1\rangle-|2\rangle$ transition), the generated field intensity experiences sinusoidal oscillations with respect to the propagation length:

$$J=\frac{\eta_{20}}{1+\left(\frac{\Delta k'}{2\kappa_m}\right)^2}\sin^2\left[\kappa_m z\sqrt{1+\left(\frac{\Delta k'}{2\kappa_m}\right)^2}\right], \tag{86}$$

$$\kappa_m=\frac{N}{4}\frac{\sqrt{\mu_2\mu_3}}{\delta_3}, \tag{87}$$

$$\Delta k'=\Delta k-\frac{N}{4}\left(\sqrt{\frac{\mu_1}{\eta_{10}}}+\frac{(\mu_2-\mu_3)}{\delta_3}\right), \tag{88}$$

in correspondence with the analysis of Sec. II, Eqs. (14), (22), and (24), and assuming $|c_1|^2=|c_2|^2=\rho_{12}=1/2$.

Since κ_m is of the order of $\Delta k'$, substantial transfer of energy from the ω_2 field into the ω_3 field occurs even without compensation of the phase mismatch. If one can manage to make $\Delta k'$ small: $\Delta k' \ll \kappa_m$, then the best possible regime for conversion can be attained. Not only complete transfer for the pulse maximum is realized: $J_1 \approx J_{max}=\eta_{20}$, but also the conversion length $l=\kappa_m^{-1}$ does not depend on time, and hence, all parts of the ω_2 pulse are homogeneously converted into the ω_3 wave.

There are several possibilities to reach this regime. First, one may set Δk to zero by use of the buffer gas, and choose the input laser parameters such that

$$\delta_3=-\frac{(\mu_2-\mu_3)}{\mu_1}\sqrt{\mu_1\eta_{10}}. \tag{89}$$

This condition can be satisfied only for the time interval when $\sqrt{f_{10}(t-z/c)} \approx 1$. That is, a flat temporal profile of the ω_1 pulse is required. Since this is not always possible, another method can be used. One may simply increase the intensity η_{10} so that the refraction contribution of the $|1\rangle-|2\rangle$ transition becomes small: $\sqrt{\mu_1/\eta_{10}} \ll (\mu_2-\mu_3)/\delta_3$, and choose the detuning δ_3 as

$$\delta_3=\frac{(\mu_2-\mu_3)}{2q}. \tag{90}$$

Since in this regime $\eta_3=J \ll \eta_{10}$, we have at any propagation distance, $\Omega_1 \gg \Omega_2^2/\delta_3$, Ω_3^2/δ_3. Therefore, the atoms remain in a superposition $|\psi_0\rangle=(|1\rangle-|2\rangle)/\sqrt{2}$ throughout the medium.

It should be noted that although the instantaneous conversion efficiency ϵ can reach unity, the total energy conversion efficiency is always small, $W \ll 1$, because $J_1 \approx \eta_{20} \ll \eta_{10}$.

3. Depletion of the pump fields

The situation is completely different for comparable intensities of the two pump waves: $\eta_{20} \approx \eta_{10}$.

For the maximum coherence regime, $|c_1|=|c_2|=1/\sqrt{2}$, to be attained, the condition $\Omega_1 \gg \Omega_2^2/\delta_3$ must be satisfied (see above), which reduces to $\sqrt{\mu_2\eta_{10}} \ll \delta_3\sqrt{\mu_1/\mu_2}$ at $\eta_{20} \approx \eta_{10}$. That is, the intensity of the pump pulses cannot be large, and the refraction contribution of the $|1\rangle-|2\rangle$ transition ($\sim\sqrt{\mu_1/\eta_{10}}$) is not small, so that $\Delta k' \approx (N/4)\sqrt{\mu_1/\eta_{10}} \gg \kappa_m$. Thus, the "exact maximum-coherence" regime fails when the drive field is depleted.

However, the phase matching can still be performed for $\eta_{20}\approx\eta_{10}$ if the requirement for $|c_1|=|c_2|=1/\sqrt{2}$ is lifted. One of the possibilities is to cancel the residual refraction by the addition of the buffer gas: $\Delta k=0$ (then $A_2=A_3=0$), and to choose the interaction parameters such that $A_1\approx 0$. The latter condition can be satisfied for specific relations between the detuning δ_2 and/or δ_3, and the input intensities η_{10},η_{20}. For example, for $\eta_{10}=\eta_{20}\equiv\eta_0$ and when $\mu_2<\mu_3$, one may choose the detunings $\delta_2=0$ and δ_3 such that

$$\mu_3\eta_0=\frac{\mu_1}{(\mu_3-\mu_2)}\delta_3^2. \tag{91}$$

Taking into account the condition (91), the eigenenergy λ is reduced in this regime to

$$\lambda=\sqrt{\mu_1\eta_0}\sqrt{\frac{\mu_3}{(\mu_3-\mu_2)}}=\Omega_{10}\zeta, \tag{92}$$

and the corresponding adiabatic state at the entrance to the medium is

$$|\psi_0(z=0)\rangle=\frac{1}{\sqrt{1+\zeta^2}}(|1\rangle-\zeta|2\rangle). \tag{93}$$

Thus, the atoms are prepared not with maximum coherence, $\rho_{12}=1/2$, but in a specific superposition with $\rho_{12}=\zeta/(1+\zeta^2)$, which is also not small.

The propagation problem solution in this case resembles that of the EIT regime, Eq. (79):

$$\kappa_m\frac{2}{\zeta}z=\arctan\left(\sqrt{\frac{J}{\eta_0}}\right)-\frac{\mu_1+\mu_2-\mu_3}{\mu_3}\sqrt{\frac{J}{\eta_0}}, \tag{94}$$

$$\zeta=\sqrt{\frac{\mu_3}{(\mu_3-\mu_2)}}, \tag{95}$$

which is demonstrated in Fig. 4. The conversion coefficient differs from that of the undepleted coherence case κ_m, Eq. (87), only by a numerical factor $2\sqrt{(\mu_3-\mu_2)/\mu_3}$. As we can see, although the total conversion efficiency may approach unity, the robustness of the maximum coherence scheme to the phase mismatch is lost. One has to fulfill the phase-matching conditions [like Eq. (91)] with sufficient accuracy to get a large conversion. A considerable transfer of energy into the generated wave can be expected when the envelope of the pump pulse satisfies the condition:

$$1-\sqrt{f_{10}(t-z/c)}\leqslant\sqrt{\frac{4\mu_2}{(\mu_3-\mu_2)}},$$

and the detuning $\delta_3=\delta_3^0+\delta_3'$ differs from the value δ_3^0, given by condition (91), by not more than

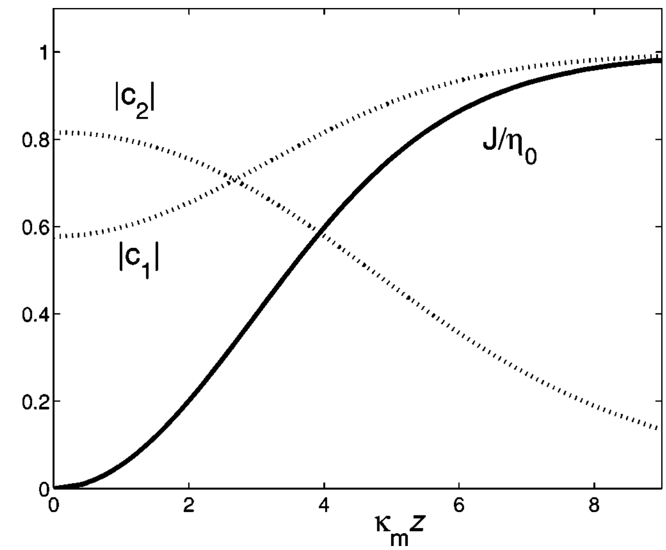

FIG. 4. Up-conversion with initial maximum coherence. Solid line: spatial evolution of J according to Eq. (94) for given retarded time. For sufficiently large propagation length there is complete conversion. Dotted lines: corresponding spatial evolution of the atomic bare state probabilities. The amplitude of the excited state $|3\rangle$ is negligibly small (not shown). Parameters: $\mu_2/\mu_3=0.5$, $\mu_1/\mu_3=0.05$.

$$\delta_3'\leqslant\sqrt{\frac{\mu_2}{(\mu_3-\mu_2)}}\delta_3^0.$$

Both of these conditions require that the values of the transition coupling constants, μ_2 and μ_3, be close.

Similar to the EIT regime, the adiabatic superposition does not coincide with the state $|\psi_0(z=0)\rangle$, Eq. (93), over the entire medium, but will evolve in the course of propagation, following the change of the fields (Fig. 4). For large density-length products Nz when $\Omega_1\ll\Omega_3^2/\delta_3$, the atomic state tends to the ground state: $|\psi_0\rangle\to|1\rangle$. Thus, "the maximum coherence" is not maintained throughout the light propagation path, although it is still sufficiently large for most part of the medium. One can see from Fig. 4 that as the coherence gets smaller, the conversion slows down, and for very large Nz (hence, for small atomic coherence), the generated intensity only slowly approaches its maximum.

Here again we encounter an apparent contradiction related to the limited validity of the adiabatic approximation. The atoms do not return to the initial state despite the overall energy conservation. Now the preparation energy for the atomic coherence is extracted as well. Under the conditions discussed here, i.e., large photon number as compared to number of atoms, this is however a small effect. A resolution of this problem should be found when nonadiabatic corrections are taken into account, which will be the subject of further investigations.

C. Conventional nonlinear optics (weak excitation)

Finally, we consider the regime corresponding to conventional nonlinear optics. This regime takes place at weak excitation when both detunings δ_2 and δ_3 are much larger than all Rabi frequencies. Then we have $\lambda \approx \Omega_{10}^2/\delta_2$ and $|\psi_0\rangle \approx |1\rangle$ for all atoms in the medium.

The coefficients A_1 and a_m do not depend on laser intensities in this case:

$$A_1 \simeq q\delta_2\delta_3 + \mu_1\delta_3 - \mu_3\delta_2, \tag{96}$$

$$a_0 \simeq \delta_2\delta_3, \tag{97}$$

$$a_1 = 2q(\delta_2+\delta_3) + (\mu_1+\mu_2-\mu_3). \tag{98}$$

Since $a_1 \min(\eta_{10},\eta_{20}) \ll a_0$, the solution Eq. (66) can be well approximated by

$$J = J_1 \mathrm{sn}^2[\kappa_n z; \sqrt{J_1/J_2}], \tag{99}$$

$$\kappa_n = \frac{N}{2}\frac{\sqrt{\mu_1\mu_2\mu_3 J_2}}{\delta_2\delta_3}, \tag{100}$$

with $J_{2,1}$ determined from Eqs. (69) and (96). This formula is valid for any relation between η_{10} and η_{20}.

When the phase mismatch is compensated: $A_1 \approx 0$, which is made by proper tuning,

$$q = \frac{\mu_3}{\delta_3} - \frac{\mu_1}{\delta_2}, \tag{101}$$

the roots $J_{2,1}$ are $J_1 = \min(\eta_{10},\eta_{20})$, $J_2 = \max(\eta_{10},\eta_{20})$. Therefore, the optimum conversion can be realized also in the case of the off-resonant nonlinear optics. In particular, for $\eta_{10} \approx \eta_{20}$ the complete transfer of energy to the generated wave occurs:

$$J = \eta_0 \mathrm{sn}^2[\kappa_n z; p \to 1] \approx \eta_0 \tanh^2(\kappa_n z),$$

leading to perfect total conversion efficiency, $W \approx 1$. However, the nonlinear conversion coefficient κ_n is much smaller in this case than in the EIT and maximum-coherence regimes [κ_e and κ_m, Eqs. (74) and (87), respectively]. Therefore, the energy transfer is accomplished for much larger density-length products Nz.

At last, we note that if the phase mismatch is not compensated, we have $B_1 \gg (\eta_{10}+\eta_{20})$ and the solution is reduced to the traditional formula of nonlinear optics [9]:

$$J = N^2 \frac{\mu_1\mu_2\mu_3\eta_{10}\eta_{20}}{4\delta_2^2\delta_3^2}\frac{\sin^2(\Delta k' z/2)}{(\Delta k'/2)^2}, \tag{102}$$

with the total phase mismatch $\Delta k' = \Delta k - N\mu_1/2\delta_2 + N\mu_3/2\delta_3$.

V. SUMMARY

Resonant optical processes based on atomic coherence effects such as EIT or maximum coherence allow for a maximum nonlinear conversion of photons within a much smaller density-length product than possible in schemes of conventional off-resonant nonlinear optics. Since degrading mechanisms such as phase mismatch and absorption scale with the same density length product, EIT or maximum coherence can thus substantially reduce the requirements for efficient nonlinear optics. As a measure for the efficiency of using atoms for nonlinear conversion processes, one can introduce the ratio of the number of converted photons $n \sim \mathcal{A}\tau_j \overline{\eta_{j0}}$ to the number of atoms $N_{at} = N\mathcal{A}l \sim N\mathcal{A}/\kappa$ needed ($\mathcal{A}$ is a cross-sectional area and τ_j is the duration of the pulse). This figure of merit n/N_{at} is largest for the EIT scheme, $n/N_{at} = \tau_1\Omega_{10}$; considerably smaller for the maximum coherence method, $n/N_{at} = \tau_2\Omega_{20}(\Omega_{20}/\delta_3)$ (with $\Omega_{20}/\delta_3 \ll 1$); and is very small for conventional nonlinear optics, $n/N_{at} = \tau_2\Omega_{20}(\Omega_{10}\Omega_{20}/\delta_3\delta_2)$ (with $\Omega_{20}/\delta_3 \ll 1$ and $\Omega_{10}/\delta_2 \ll 1$).

In previous theoretical studies of EIT- and maximum-coherence based nonlinear optics, undepleted drive fields or a constant coherence was assumed. If one takes into account the resources to maintain the drive field or the constant coherence, the overall efficiency of the processes is in this limit tiny. We have shown in the present paper that when this restriction is lifted and coherence- or transparency-maintaining fields with comparable intensities to the pump fields are considered, it is possible to achieve also a maximum overall conversion. To study the conditions for this, we have derived analytical solutions for the pulse interaction in the adiabatic limit using an approach that maps the propagation problem to that of a nonlinear pendulum. Under certain conditions this problem could be explicitly integrated allowing for a simple discussion of the physical processes involved.

We have found that when the coherence- or transparency-maintaining fields are depleted, the atomic state does not remain constant but evolves along the propagation path following the change of the fields. Therefore, the conversion process cannot be associated with maximum coherence or EIT in the traditional sense alone. Nevertheless, the main features of EIT—reduced linear absorption and refraction, and enhanced nonlinearity—are still present in the latter mechanism. Although in the maximum-coherence mechanism the robustness to the phase mismatch is lost, the nonlinear conversion coefficient remains much larger than in conventional nonlinear optics. Thus, for both mechanisms, a complete conversion can be achieved within a small density-length product.

We have also encountered several limitations of the strong adiabatic assumption used in the Hamiltonian approach. An extension of the approach to take into account nonadiabatic corrections and thus such important effects as group delay are currently under investigation and the corresponding results will be presented elsewhere.

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