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Study of the maximum conversion efficiency of the four-wave mixing process based on the Hamiltonian approach

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ABSTRACT

In the four-wave mixing process, since the intensity of the generated coherence field is related closely to the third-order nonlinear susceptibility. With the help of a Hamiltonian approach, we obtain the expression of third-order nonlinear susceptibility $\chi^{(3)}$, and the special relation between $\chi^{(3)}$ and the nonlinear conversion coefficient. They are very useful to analyze the phase mismatch due to the Kerr effect and linear refraction contributions, which affect the maximum conversion efficiency under the condition taking account into pump field depletion. The investigative results show that energy conversion efficiency from the weak idler field to the generated field may exceed 100% when the phase mismatch induced by linear refraction is compensated, though the phase mismatch contribution from the Kerr effect can not be compensated simultaneously. Moreover, the conversion efficiency of the photon numbers fast reaches a plateau value.

1. Introduction

Frequency conversion techniques, based on the nonlinear optical response of a gaseous medium with coherent radiation, provide a well-established powerful tool for the generation of tunable coherent short-wavelength vacuum ultraviolet (UVU) radiation [1]. These light sources are of considerable interest for laser-lithography high resolution microcopy and spectroscopy. Due to the rather small third-order nonlinear susceptibility, such conventional techniques suffer from relatively poor conversion efficiencies, typically in the range of 10^{-6} to 10^{-4}[2]. If resonances are used to enhance the nonlinear optical response, re-absorption of the generated radiation strongly limits the attainable efficiency.

Recently, a new technique has been implemented widely, which substantially enhances frequency conversion efficiency. The technique usually is referred to as "nonlinear optics with maximum coherence" [3–5]. In a simple two-level atom system, the polarization leading to nonlinear mixing process depends upon the coherence between the ground and excited states. This property reaches a maximum of 1/2 for an equal amplitude coherent superposition of ground and excited states. To generate shorter wave radiation and to apply the concept of maximum coherence to systems, the technique of stark-chirped rapid adiabatic passage (SCRAP) is a very efficient method at present [6–9]. Korsunsky et al. [10] have discussed the three-wave mixing process in a Λ-type three-level system and derived the full analytic solution with the help of Hamiltonian approach [11–15] when taking into account pump field depletion. When the phase matching is compensated, in order to obtain large overall conversion efficiency, the physical nature of the conversion efficiency is no longer associated with maximum coherence. As the light propagation path gets longer (for very large Nz) the generated intensity of the mixing field only slowly approaches its maximum. However, such conversion efficiency considers only the phase mismatch contribution from the linear refraction. With the generated coherent field intensity becoming larger, it is bound to generate the phase mismatch induced by the intensity-dependent index of refraction, such as Kerr effect. Therefore, considering the maximum conversion efficiency, we not only take into account the phase mismatch contribution from the linear refraction and also must ponder the nonlinear refraction induced by Kerr effect.

In the present paper, we take into account a process of difference-frequency mixing involving a two-photon pump field as shown in Fig. 1. In the three-level system, a two-photon transition $|1\rangle \leftrightarrow |2\rangle$ resonantly excited by a strong pump field with frequency ω_1, a strong Stark pulse inducing a dynamic Stark shift, and a dipole-allowed transition $|2\rangle \leftrightarrow |3\rangle$ excited off-resonance by an idler field with frequency ω_2. In this case, the energy of the

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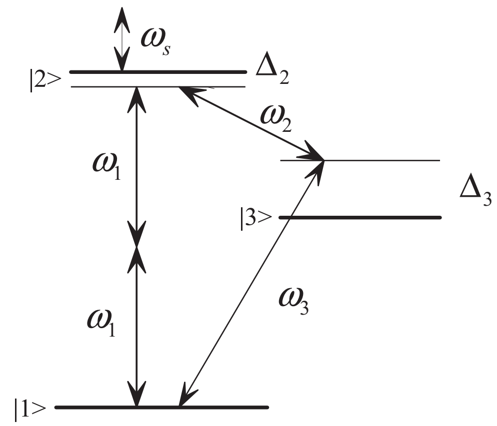

Fig. 1. Resonant four-wave mixing with maximum coherence between $|1\rangle$ and $|2\rangle$ adiabatically prepared by a strong pump with ω_1 and a "Stark-shifting" far-off-resonance field with ω_s.

generated field is taken only from the pump field, which at the same time participates in the preparation of the coherence. Therefore, it will unavoidably be depleted if considerable conversion efficiency is expected. In order to obtain full analytical solution taking into account the pump depletion, we apply the so-called Hamiltonian approach to discuss the conversion efficiency of four-wave mixing (FWM) involving preparation of an atomic system driven to maximum coherence by SCRAP. The enhancement of the nonlinear conversion efficiency with maximum coherence has been demonstrated for the regime of undepleted pump field [4,10]. In the present paper, we discuss mainly the conversion process for depleted pump field in the process of difference-frequency mixing. The same Λ-type scheme employing a two-photon transition was previously studied by Korsunsky et al. [10]. We deduce the expression of third-order nonlinear susceptibility with a Hamiltonian approach to discuss these factors, which affect the maximum conversion efficiency, and analyze the phase mismatch due to the Kerr effect and linear refraction contributions. The result shows that the phase mismatch from the two contributions cannot be compensated simultaneously. Therefore, under the condition of considering the depleted pump field, the conversion efficiency can be large improved, but not be able to approach unity, which is different from the results of Ref. [10].

The paper is organized as follows: in Section 2 we deduce the general expression of the nonlinear susceptibility with a Hamiltonian approach in a multilevel system, and discuss the relation between the nonlinear conversion coefficient and third-order nonlinear susceptibility. In Section 3 we outline the solutions for resonant four-wave mixing with depleted pump field, and discuss the influence of the linear and nonlinear phase mismatch on the conversion efficiency, respectively. In Section 4, through the nonlinear third-order susceptibility, we discuss the conditions for phase matching induced by linear refraction and the Kerr effect simultaneously with respect to the compensation, Conclusions are presented in Section 5.

2. The deduction of three-order nonlinear susceptibility

We first consider the propagation of pulsed electromagnetic fields in a medium of three-level atoms (Fig. 1), The electromagnetic field propagating in the z direction is assumed to consist of three components with carrier frequencies ω_1, ω_2 and $\omega_3 = 2\omega_1 - \omega_2$:

$$E(z,t) = \sum_j (\varepsilon_j(z,t)\exp(-i(\omega_j t - \vec{K}_j\vec{r})) + c.c.) \quad (1)$$

Here $|K_i| = n_i\omega_i/c$ with the refractive index n_j at frequency ω_i describing refraction due to levels outside the three-level system in Fig. 1. The radiation pulses are supposed to be shorter than the relaxation times in the atomic system. The waves $\vec{k}_2$ and $\vec{k}_3$ propagate at small angles with respect to the vector $\vec{k}_1$ (the z-axis).

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

$$\frac{\partial \varepsilon_j}{\partial z} = i2\pi\frac{\omega_j}{c}P_j \quad (2)$$

where ε_j and p_j ($j = 1, 2, 3$) are functions of the coordinate z and the retarded time $\tau = t - z/c.P_j$ are the components of the medium polarization:

$$P = \sum (P_j(z,t)\exp[-i(\omega_j t - \vec{K}_j\cdot\vec{r})] + c.c) \quad (3)$$

For the FWM scheme as shown in Fig. 1, the light-atom interaction Hamiltonian of this system in a rotating frame can be written as

$$\hat{H} = -\hbar[\Delta_2\,|2\rangle\langle 2| + \Delta_3\,|3\rangle\langle 3| + \Omega_1\,|1\rangle\langle 2| + \Omega_2 e^{i\varphi}\,|2\rangle\langle 3| + \Omega_3\,|1\rangle\langle 3|] + h.c. \quad (4)$$

where the multi-photon resonance condition

$$\omega_3 = 2\omega_1 - \omega_2 \quad (5)$$

has been used. Ω_1 corresponds to the two-photon process, Ω_2 and Ω_3 are the Rabi frequencies for transitions $|1\rangle \leftrightarrow |2\rangle$, $|2\rangle \leftrightarrow |3\rangle$ and $|3\rangle \leftrightarrow |1\rangle$, respectively, which are related with the coupling strength μ_j and photon flux η_j ($j = 1, 2, 3$) defined as:

$$\Omega_1 = \mu_1\eta_1, \quad \Omega_j = \sqrt{\mu_j\eta_j}\,(j = 2, 3) \quad (6)$$

The frequency detuning $\Delta_n (n = 2, 3)$ include the "static" detuning δ_{no}, ac Stark shifts $\beta_{nj}\eta_j$ induced by the $\omega_j (j = 1, 2, 3)$ fields, and the shifts $S_n = \beta_{ns}\eta_s$ induced by an intense far-off-resonant "SCRAP laser pulse" with frequency ω_s and photon flux η_s:

$$\Delta_n = \delta_n + \sum_{j=1,2,3}\beta_{nj}\eta_j (n = 2, 3) \quad (7)$$

$$\delta_n = \delta_{n0} + S_n \quad (8)$$

$$\beta_{nj} = \frac{2\pi\omega_j}{C}(\alpha_{nn}(\omega_j) - \alpha_{11}(\omega_j)) \quad (9)$$

$$\delta_{30} = \omega_3 - \omega_{31}, \quad \delta_{20} = 2\omega_1 - \omega_{21}$$

In the present paper, we consider adiabatic light-atom interaction processes, i.e. 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.*

$$\hat{H}\,|\psi_0\rangle = \hbar\lambda_0\,|\psi_0\rangle \quad (10)$$

it will remain in this state $|\psi_0(t_0)\rangle$ at all times. Eq. (10) yields the characteristic equation for the eigenvalues:

$$\lambda_0(\Delta_2 + \lambda_0)(\Delta_3 + \lambda_0) - (\Omega_1^2 + \Omega_2^2 + \Omega_3^2)\lambda_0 - \Omega_1^2\Delta_3 - \Omega_3^2\Delta_2 = -2\Omega_1\Omega_2\Omega_3\ \cos\varphi \quad (11)$$

where the relative phase φ of the electromagnetic waves is:

$$\varphi = 2\varphi_1 - \varphi_2 - \varphi_3 - \Delta kz \quad (12)$$

This includes the residual phase mismatch $\Delta k = 2k_1 - k_2 - k_3$

To obtain the general expression of the nonlinear susceptibility in a multilevel system, here we will present an outline of the Hamiltonian approach in nonlinear optics. This approach is based on the representation of the medium polarization P as a partial derivative