Enhancement of nonlinear-optical signals under coherent-population-trapping conditions

W. Harshawardhan$^{1,2}$ and G. S. Agarwal$^1$

$^1$Physical Research Laboratory, Ahmedabad 380 009, India
$^2$University of Hyderabad, Hyderabad 500 046, India

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We develop a nonperturbative approach to study the generation of nonlinear signals in a coherently prepared medium. Specifically, we study the process $\omega_1 - \omega_2 + (\omega_2 + \Omega)$, in a system prepared in a coherent-population-trapping state. We calculate enhancement factors of the order of $\sim 10^2$ in the generated signal, under a variety of conditions thus supplementing the work of Jain et al. [Phys. Rev. Lett. 77, 4326 (1996)]. Our approach also enables us to study other related issues. We demonstrate the existence of pulse matching at higher probe powers. [S1050-2947(98)05505-X]

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I. INTRODUCTION

In nonlinear optics one of the goals has been to improve the efficiency of the generation. Several proposals have been made. These include the early work of Tewari and Agarwal [1] on the control of phase matching and nonlinear generation by using additional fields and the work of Harris et al. [2] on electromagnetic-field induced transparency (EIT) for the enhancement of third harmonic generation. The EIT has been extensively applied in many different situations [3]. Hakuta et al. [4] used a dc electric field in atomic hydrogen to produce large second harmonic generation. Later Zhang et al. [5] used EIT to produce large extreme ultraviolet (XUV) radiation. Jain et al. [6] demonstrated enhancement of vacuum ultraviolet (VUV) using a control laser. Use of a control laser has also been suggested for the inhibition as well as enhancement of two-photon absorption [7]. In many of the proposals atomic coherence plays an important role. Indeed, the ideas of atomic coherence have been used in a very wide variety of applications such as to lasing without inversion [8–11], large refractive index, and magnetometry [12]. We know that the atomic coherence is maximized in a coherent-population-trapping state [13]. An ensemble of Pb atoms prepared in this maximum coherence state was utilized for efficient conversion of blue to ultraviolet light [14]. Jain et al. obtained an efficiency of $\sim 40\%$ in the generated signal; we augment their findings in the nonperturbative regime. In an optical parametric oscillator pumped by population trapped atoms, Harris and Jain [15] have enhanced second order nonlinear susceptibility. Earlier Jain et al. [16] had shown elimination of propagation distortion due to self-focusing and defocusing. They achieved this by forcing the atoms into a coherent-population-trapping (CPT) state. Furthermore, unusually high phase for the high conjugate signal was observed by Hemmer et al. [17] in a double-$\Lambda$ system. At CPT they obtained both high gain and low spatial distortion simultaneously [18]. They also reported much lower quantum noise due to the effects of quantum interference which produce quenching of spontaneous emission [19]. Here, we study in detail the enhancement of nonlinear signal by utilizing the maximal coherence of the CPT state.

In our scheme we essentially have two fields acting simultaneously on each transition in a $\Lambda$ system. By choosing the detunings of the pump fields such that we operate at the coherent-population-trapping condition, we create the required atomic coherence. We study the enhancement of $\omega_1 + \Omega$ generated by the nonlinear process $(\omega_1 + \Omega) = \omega_1 - \omega_2 + (\omega_2 + \Omega)$, under CPT conditions in a three level $\Lambda$ system. Figure 1(a) depicts this process where $\omega_1$ and $\omega_2 + \Omega$ are absorbed and $\omega_2$ is emitted, to generate the field at $\omega_1 + \Omega$. Our analysis is nonperturbative, hence all the fields at $\omega_1$, $\omega_2$, and $\omega_2 + \Omega$ can be strong.

Moreover, our nonperturbative analysis also enables us to answer, if for a thick medium pulse matching will occur at high probe powers. We find the remarkable result that even at higher probe intensities pulse matching takes place.

In Sec. II we describe our nonperturbative treatment and derive the equations that govern the nonlinear polarization of the medium. In Sec. III we undertake the dressed-state analysis in the perturbative limit, and obtain the line shape at the CPT condition. In Sec. IV we compare and contrast the generation of the signal at two-photon resonance and away from it, and predict that enhancement is due to the CPT condition. In Sec. V we study the generation of the signal in a medium and study the pulse matching phenomenon for higher probe powers inside a thick medium.

II. NONPERTURBATIVE FORMULATION

We consider a $\Lambda$ system, Fig. 1(b). The excited state $|1\rangle$ is coupled, to $|3\rangle$ ($|2\rangle$) by the monochromatic field $\vec{E}_1$ ($\vec{E}_2$), with frequency $\omega_1$ ($\omega_2$). We have another set of fields $\vec{E}_1'$ ($\vec{E}_2'$) of frequency $\omega_1 + \Omega$ ($\omega_2 + \Omega$) acting on the $|1\rangle \leftrightarrow |3\rangle$ ($|1\rangle \leftrightarrow |2\rangle$) transition. The total Hamiltonian of the system is given as

$$
H_T = \hbar \omega_{13} A_{11} + \hbar \omega_{23} A_{22} - \vec{d}_{12} \cdot (\vec{E}_2 e^{-i(\omega_2 t - \vec{k}_2 \cdot \vec{r})}) + \vec{E}_1' e^{-i(\omega_1 + \Omega) t - \vec{k}_1' \cdot \vec{r})} A_{12} - \vec{d}_{13} \cdot (\vec{E}_1 e^{-i(\omega_1 t - \vec{k}_1 \cdot \vec{r})}) + \vec{E}_2' e^{-i(\omega_2 + \Omega) t - \vec{k}_2' \cdot \vec{r})} A_{13} + \text{H.c.},
$$

where $A_{ij}$ is the atomic transition operator $|i\rangle \langle j|$. The energy is measured from $|3\rangle$, where $\hbar \omega_{13}$ and $\hbar \omega_{23}$ are the energy difference between levels $|1\rangle$ and $|2\rangle$ from $|3\rangle$, respectively.