Large Enhancements in Nonlinear Generation by External Electromagnetic Fields

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We show how the generation of vacuum uv radiation can be doubly enhanced by using strong external electromagnetic fields so that one can simultaneously make use of (a) the amplification of the fundamental as it propagates through the medium and (b) the near-zero absorption of the generated vacuum uv radiation. We include in the analysis the effects of propagation and pump depletion.

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Recently Harris and coworkers [1] have emphasized the usage of electromagnetic-field-induced transparency [2] for enhancing the efficiency of vacuum ultraviolet (VUV) generation. Tewari and Agarwal [3] earlier considered the question of controlling the efficiency of VUV generation using external resonant electromagnetic fields. They found that the field can change the phase matching conditions and one could even generate VUV in regions where it is normally forbidden. The relationship between the two works has been pointed out [4]. Clearly the external fields change not only the nonlinear susceptibilities but also the absorption and dispersion characteristics [5,6] of the medium which in turn changes the VUV generation [7]. It is now known that the external fields can also be used to amplify signals and this idea has been used for a possible laser system without population inversion [8–10].

In this Letter we show how the amplification of the fundamental and the electromagnetic-field-induced transparency can be used *together* to produce much larger enhancements in the VUV generation. *Our mechanism amplifies the fundamental field as it propagates through the medium.* This is clearly different from the case where the input has been amplified before it enters the medium.

Consider the energy level scheme shown in Fig. 1. The levels $|1\rangle$ and $|3\rangle$ are connected by a three-photon transition through the intermediate state $|2\rangle$. We assume that the levels $|1\rangle$ and $|2\rangle$ are connected by a weak two-photon transition with the two-photon matrix element κ. The interaction Hamiltonian for such a system can be written as

$$H_1 = \hbar(\kappa|1\rangle\langle 2|e^{-2i\omega t} + g|2\rangle\langle 3|e^{-i\omega t} + \text{H.c.})\,, \qquad (1)$$

where g is the one-photon coupling between the levels $|2\rangle$ and $|3\rangle$. Within the framework of the perturbation theory it is straightforward to calculate the nonlinear susceptibility $\chi^{(3)}(\omega,\omega,\omega)$ responsible for the third-harmonic generation,

$$\chi^{(3)}(\omega,\omega,\omega) \propto \frac{-\kappa g n \mathcal{E}^{-3}}{[\Gamma_{13}+i(\omega_{13}-3\omega)][\Gamma_{23}+i(\omega_{23}-\omega)]}\,. \qquad (2)$$

Here Γ_{ij} gives the decay width associated with the decay of the off-diagonal element ρ_{ij} of the density matrix and n is the density of atoms; $\mathcal{E}$ is the envelope of the fundamental field at the frequency ω. This is the usual situation regarding the generation of the radiation at 3ω. The VUV generation has been studied over a very wide range of wavelengths [11]. The method of the present paper will also work over a wide range, depending on the choice of the atom and the transition.

We now modify the system. We apply an external electromagnetic field of frequency ω_l at the transition $|2\rangle \leftrightarrow |3\rangle$. This field modifies the absorption and dispersion characteristics of the medium; we will refer to this field as the control field. In order to study systematically the generation of the third harmonic, we have to examine the susceptibilities $\chi_d^{(1)}(3\omega)$, $\chi_d^{(1)}(\omega)$, and $\chi_d^{(3)}(\omega,\omega,\omega)$, where the subscript d indicates that such susceptibilities are to be obtained *in the presence of the control field* of frequency ω_l. Such susceptibilities depend on the strength (to all orders) and frequency of the additional electromagnetic field.

Note that $\chi_d^{(1)}(3\omega)$ gives the absorption and dispersion

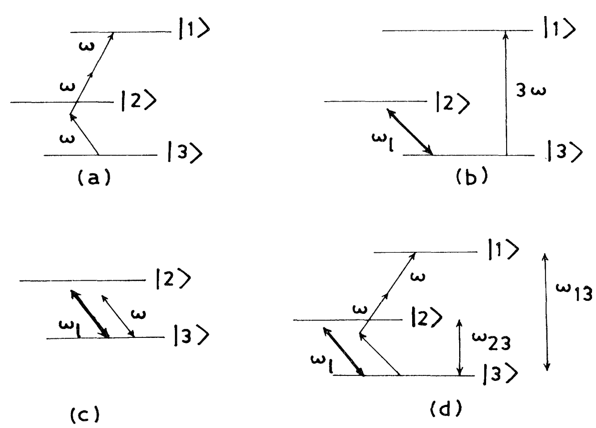

FIG. 1. Schematic illustration of the model for producing double enhancement of the VUV generation.

of the generated third harmonic. The calculation based on Fig. 1(b) determines this susceptibility. Further, $\mathrm{Im}\chi_d^{(1)}(\omega)$ gives the amplification or absorption of the field at ω. This is to be calculated using the scheme of Fig. 1(c). The susceptibility $\chi_d^{(3)}(\omega,\omega,\omega)$ is obtained by using the level scheme of Fig. 1(d). The Hamiltonian describing the interaction shown in Fig. 1(d) is

$$H=\hbar\omega_{23}|2\rangle\langle 2|+\hbar\omega_{13}|1\rangle\langle 1|+\hbar(\kappa|1\rangle\langle 2|e^{-2i\omega t}+g|2\rangle\langle 3|e^{-i\omega t}+G|2\rangle\langle 3|e^{-i\omega_l t}+\mathrm{H.c.})\,, \tag{3}$$

where $2G$ is the Rabi frequency of the control field on the transition $|2\rangle\leftrightarrow|3\rangle$. The corresponding density matrix equations follow from (3). As usual we will also *include various radiative and collisional relaxation terms* while writing the density matrix equations. In particular each off-diagonal element of the density matrix will have a decay term Γ_{ij}. The different susceptibilities can be calculated using the density matrix equations. The calculations can be done to all orders in the field G by working in a frame rotating with the frequency ω_l so that (3) transforms to

$$H=\hbar(\omega_{23}-\omega_l)|2\rangle\langle 2|+\hbar(\omega_{13}-3\omega_l)|1\rangle\langle 1|+\hbar(\kappa|1\rangle\langle 2|e^{-2i\delta t}+g|2\rangle\langle 3|e^{-i\delta t}+G|2\rangle\langle 3|+\mathrm{H.c.})\,, \tag{4}$$

where

$$\delta=\omega-\omega_l\,. \tag{5}$$

To obtain $\chi_d^{(1)}(\omega)$ we set $\kappa=0$ and calculate the induced polarization at ω to first order in g and to all orders in G. This induced polarization will be given in terms of the response of the density matrix element ρ_{23}. The susceptibility $\chi_d^{(3)}(\omega,\omega,\omega)$ is obtained from the induced polarization at 3ω (density matrix element ρ_{13}) to first order in κ and to first order in g but to all orders in G. To obtain the susceptibility $\chi_d^{(1)}(3\omega)$ we set $\kappa=0$, $g=0$, and include an additional interaction $\hbar\tilde{g}|1\rangle\langle 3|\times e^{-3i(\omega-\omega_l)t}+\mathrm{H.c.}$ in (4). Note that $2\tilde{g}$ is the Rabi frequency of the field at 3ω. We calculate the induced polarization at 3ω to first order in $\tilde{g}$ but to all orders in G. The analytical results for susceptibilities are rather long and will be presented elsewhere. In this Letter we present our final results.

Harris has argued that the external electromagnetic field can be used to create a transparency region and thus the reabsorption of the generated third harmonic can be reduced considerably; thereby the efficiency of the generation of the third harmonic can be enhanced. In this communication we demonstrate the possibility of *further enhancement* of VUV generation by effectively utilizing the *amplification* of the electromagnetic field at the fundamental frequency ω.

The nonlinear generation is quite sensitive to the various choice of parameters and we have to make a judicious choice in order to obtain optimal generation. A clue to the choice of the parameters is provided by the behavior of the susceptibilities. From the work of Mollow it is known that the imaginary part of the susceptibility $\chi_d^{(1)}(\omega)$ shows amplification regions given by

$$\omega-\omega_l\sim-[(\omega_{23}-\omega_l)^2+4G^2]^{1/2}\ \text{if}\ \omega_{23}>\omega_l\,. \tag{6}$$

If we look for VUV generation at the line center, then $3\omega=\omega_{13}$. Note that the condition (6) implies that in order to achieve amplification of the fundamental, the fundamental must be detuned from the intermediate state $|2\rangle$; thus $\omega\neq\omega_{23}$. Let us denote this detuning by δ_0,

$$\delta_0=\omega_{23}-\omega,\quad \omega_{13}=3\omega,\quad \omega_{12}^{-2\omega}=-\delta_0\,. \tag{7}$$

Hence (6) and (7) lead to

$$\delta_0^2=2\Delta\delta_0+4G^2,\quad \Delta=\omega_{23}-\omega_l\,. \tag{8}$$

Thus for a fixed δ_0, one should fix the parameters of the control field so that (8) is satisfied. We will see that this gives us a lot of flexibility and hence the possibility of varying the intensity of the VUV generation.

It should be borne in mind that the susceptibilities $\chi_d^{(1)}(\omega)$, $\chi_d^{(1)}(3\omega)$, and $\chi_d^{(3)}(\omega,\omega,\omega)$ depend on the intensity I_l of the control field. The intensity I_l changes throughout the medium because as the fundamental field is amplified it draws energy from the control field. Thus the above susceptibilities also depend on the spatial coordinate inside the medium. We thus have to examine the *propagation* equations for different fields. The propagation equations for different fields can be obtained in slowly varying envelope approximation as

$$\frac{\partial\varepsilon(\omega_l)}{\partial z}=\frac{2\pi i\omega_l}{c}\{\chi_d^{(1)}(\omega_l)+\chi_d^{(3)}(\omega_l,\omega,-\omega)\times|\varepsilon(\omega)|^2\}\varepsilon(\omega_l)\,, \tag{9}$$

$$\frac{\partial\varepsilon(\omega)}{\partial z}=\frac{2\pi i\omega}{c}\chi_d^{(1)}(\omega)\varepsilon(\omega)\,, \tag{10}$$

$$\frac{\partial\varepsilon(3\omega)}{\partial z}=\frac{6\pi i\omega}{c}\{\chi_d^{(1)}(3\omega)\varepsilon(3\omega)+\chi_d^{(3)}(\omega,\omega,\omega)\varepsilon^3(\omega)\}\,. \tag{11}$$

In Eq. (9), $\chi_d^{(1)}(\omega_l)$ just gives the absorption of ω_l by the medium; $\chi_d^{(3)}(\omega_l,\omega,-\omega)$ gives the absorption of the field ω_l in the presence of the field at ω. These two susceptibilities depend on all orders of the field at ω_l. Both these susceptibilities can be calculated from the configuration shown in Fig. 1(c). In writing (9)-(11) we ignore the reaction of the generated third harmonic on the fundamental. We also assume that the fundamental is such that the saturation effects on the transition $1\leftrightarrow 2$ are not important. The procedure is now as follows—various susceptibilities are calculated using the fields at a given point. The nonlinear Eqs. (9)-(11) are integrated numerically to obtain the growth or depletion of various fields.

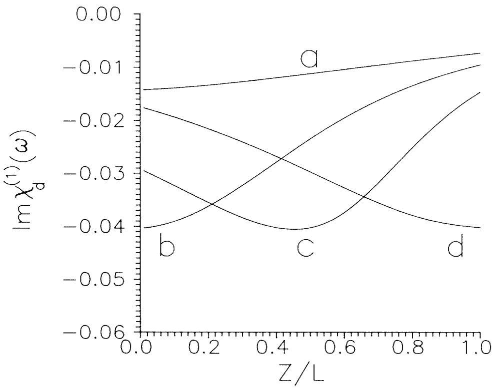

FIG. 2. The behavior of the $\hbar\Gamma \mathrm{Im}\chi_d^{(1)}(\omega)/n|d_{23}|^2$ inside the sample as a function of z/L for $3\omega=\omega_{13}$. We have set $\Gamma_{12}=\Gamma_{13}=\Gamma_{23}=\Gamma$. The states $|1\rangle$ and $|2\rangle$ decay to the state $|3\rangle$ at the rate 2Γ. All Rabi frequencies and detuning are normalized with respect to Γ. The other parameters for different curves are (a) $\Delta=21$, $G=10$; (b) $\Delta=G$, $G=15.45$; (c) $\Delta=15.45$, $G=16.00$; (d) $\Delta=15.45$, $G=16.45$.

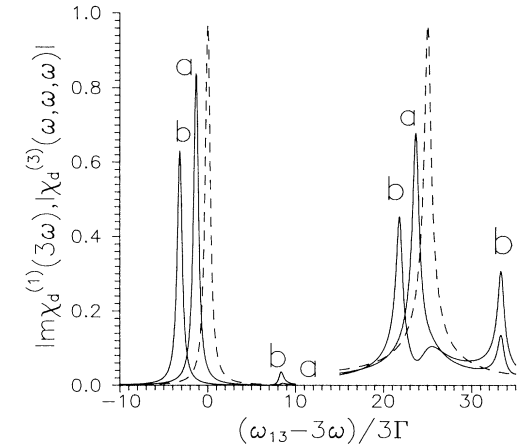

FIG. 3. The behavior of $\hbar\Gamma \mathrm{Im}\chi_d^{(1)}(3\omega)/n|d_{13}|^2$ (on the left) and $\Gamma^2|\chi_d^{(3)}|/n|d_{13}|\kappa g\mathcal{E}^{-3}$ (on the right) as a function of the dimensionless parameter $(\frac{1}{3}\omega_{13}-\omega)/\Gamma$ for the parameters corresponding to the cases *a* and *b* in Fig. 2. The dashed curves show the results in the absence of the control field. For clarity, the curves on the right were obtained by multiplying the susceptibility by 50 and adding 25 to the abscissa.

We have analyzed a number of situations. In what follows we present some typical results to demonstrate how the amplification of the fundamental can be used to enhance the VUV generation. For the range of parameters for which we present numerical results, we find that the control field Rabi frequency reduces by about 10% over the length L of the sample assuming $\alpha L=120$, where $\alpha=2\pi n|d_{13}|^2T_2(3\omega)/\hbar c$. This depletion changes various susceptibilities. In Fig. 2 we show how the gain of the fundamental changes as the field propagates through the medium. Different curves correspond to choices of parameters so that the thin-sample susceptibility (equivalent to taking the value at $z=0$) $\chi_d^{(1)}(\omega)$ exhibits gain in all cases. For certain Rabi frequencies of the control field the gain is maximum in the middle of the sample. In Fig. 3 (on the left) we present the behavior of $\mathrm{Im}\chi_d^{(1)}(3\omega)$ for a thin sample. We exhibit different situations for $\mathrm{Im}\chi_d^{(1)}(3\omega)$: dashed curve, no control field, i.e., $G=0$; (a),(b) control field off resonant with the transition $2\leftrightarrow 3$. The two peaks in the absorption curve are just the Autler-Towne's doublets which occur at

$$\omega_{13}-3\omega=\frac{\Delta}{2}\pm\frac{1}{2}(\Delta^2+4G^2)^{1/2}. \qquad (12)$$

Even the reabsorption of 3ω changes as a function of z because the control field gets depleted and the sidebands come closer, leading to an increase in the reabsorption of the generated third harmonic. Our calculations shown in Fig. 5 take *into account this change in the reabsorption as a function of z,* though for want of space we do not give graphs here for $\chi_d^{(1)}(3\omega,z)$. In Fig. 3 we have also shown (on the right) the thin-sample susceptibility $|\chi_d^{(3)}(\omega,\omega,\omega)|$ responsible for the harmonic generation. The curves demonstrate resonant enhancement of $|\chi_d^{(3)}(\omega,\omega,\omega)|$ at the modified eigenvalues of the *dressed atom.* In Fig. 4 we show the z dependence of $|\chi_d^{(3)}(\omega,\omega,\omega,z)|$ for different operational points. Having seen the behavior of various susceptibilities that determine the behavior of the fundamental and the third-harmonic fields, we are in a position to discuss the intensities of the VUV generation under different strengths and detunings of the control field. The intensity of the VUV field *as a function of z/L* is shown in Fig. 5. We show different cases [12]—the dashed curve gives the usual third-harmonic generation in the absence of the control field; curves *a–d* give the generation in the presence of a control field which is off resonant with the tran-

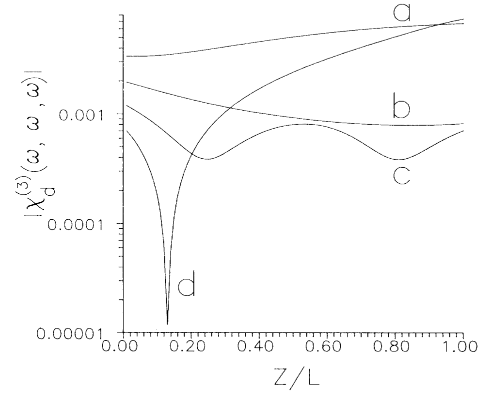

FIG. 4. The figure demonstrates the variation of the nonlinear susceptibility $\Gamma^2|\chi_d^{(3)}|/n|d_{13}|\kappa g\mathcal{E}^{-3}$ along the length of the sample for the same cases as in Fig. 2.

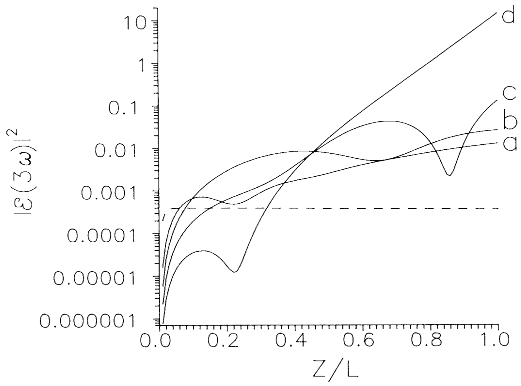

FIG. 5. The magnitude of the generated third harmonic $|\varepsilon(3\omega)|^2$ in units of $|\hbar\kappa g/\Gamma|d_{13}||^2$ as a function of z/L for $\alpha L = 120$ and for the cases corresponding to a–d in Fig. 2. The dashed curve gives the values in the absence of the control field.

sition $|2\rangle \leftrightarrow |3\rangle$. In all cases a–d the fundamental is amplified as it propagates through the medium. However, as shown in Fig. 2, the gain maximum [of $\chi_d^{(1)}(\omega)$] lies at different points inside the medium. Besides, the reabsorption of the generated third harmonic is different in different cases. For example, if we compare cases b and a then we find that the reabsorption in case b is about a factor 10 smaller than in case a. The gain features in different cases a–d are also different. For example, if we compare cases b and c then we have large gain (cf. Fig. 2) over a rather large region of the sample. This coupled with smaller reabsorption is responsible for *very large* enhancement in the VUV generation. In case d the gain persists over the entire length of the sample, leading to much larger generation.

It is thus clear from the various curves in Fig. 5 that the generated VUV can be *doubly enhanced* by using both (i) amplification of the fundamental and (ii) the negligible readsorption of the generated third harmonic. It is also clear that larger enhancements in VUV generation will be obtained for bigger values of αL.

Thus, in conclusion, our work shows the tremendous possibilities for enhancing VUV generation by suitable use of control fields and by judicious choice of the strength and the frequency of the control field.

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[12] In our model the depletion of the control laser makes $\varepsilon(3\omega)$ a nonlinear function of $\varepsilon^3(\omega)$. Note further that the energy is being taken from both the control field as well as the fundamental. In view of this it is more involved to define the efficiency of the process and, besides, all physical quantities depend on the spatial parameter z. It may be noted that previous works (Refs. [1,4]) considered the steady-state situation, i.e., much larger αL values.