Pump-induced transparency and enhanced third-harmonic generation near an autoionizing state

S. J. van Enk,[1,2] Jian Zhang,[2] and P. Lambropoulos[1,2]
[1]Foundation for Research and Technology–Hellas, Institute of Electronic Structure and Laser, P.O. Box 1527, Heraklion 71110, Crete, Greece
and Department of Physics, University of Crete, Crete, Greece
[2]Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Strasse 1, D-85748 Garching, Germany
(Received 28 December 1993)

We analyze an electromagnetically induced transparency near an autoionizing state, which is at the same time stabilized against ionization. We show that the effect is fairly general as long as only one continuum is involved. We compare the efficiency for third-harmonic generation in this system with that in an intermediate two-photon resonance system, and discuss the advantages of the former. Explicit calculations are presented for a specific autoionizing resonance in Ca.

PACS number(s): 42.65.Ky, 32.80.Dz, 42.50.Gy

According to the traditional understanding of third-harmonic generation (THG), a single- or three-photon resonance of the pump frequency with atomic states is undesirable as it leads to absorption of either the pump or the generated harmonic radiation [1]. An intermediate two-photon resonance (TPR) [2] is, on the other hand, considered helpful as it enhances the susceptibility $\chi^{(3)}$ without appreciable loss; up to the point where significant accumulation of population at the TPR state as well as ionization therefrom limit the benefit. Three-photon resonance with an autoionizing state [3,4] as a means of enhancing the susceptibility has also been explored a number of years ago, but it does not seem to have led to crisp conclusions and widespread use.

More recently, considerable discussion [5,6] has been devoted to the investigation of the possible benefits of electromagnetically induced transparency on nonlinear optical processes and a host of other effects in radiation-matter interaction. In a recent paper [7], the idea was demonstrated experimentally in sum-frequency generation.

Our purpose in this paper is to show that an autoionizing state in three-photon resonance with a pump field inherently develops a transparency in a certain range of intensity leading thus to enhanced THG, which turns out to be more significant than that from a TPR system. The first obvious advantage of our scheme is that no additional field is needed for the transparency. A second advantage comes from a rather subtle and surprising effect, namely, the stabilization [8–10] of the autoionizing state (against ionization) under the very same conditions that establish the transparency. We have thus the combination of an enhanced nonlinear susceptibility with the prolonging of the stability of an atomic system in moderately strong electromagnetic fields. The old question of the usefulness of three-photon resonance with an autoionizing state is now found to have an unexplored dimension that allows its exploitation under significantly higher pump intensity, and hence larger harmonic production.

The practical side of our theory is demonstrated by a quantitative application to atomic Ca with a complete description and specification of all atomic and radiation parameters necessary for experimental implementation. We consider an atomic system that has an isolated autoionizing state $|1\rangle$, which can be reached from the ground state $|0\rangle$ by a three-photon transition. The three-photon pump field is treated as an external field with prescribed time dependence and has frequency ω_3. In the presence of the third harmonic field with frequency $\omega_1=3\omega_3$, one-photon transitions are also possible between the two states. Both fields also couple the ground state directly to the continuum. We assume that the autoionizing state decays into the same part of the continuum and that couplings of the two states to other parts can be neglected.

The atomic density-matrix equations for this system have been given in [11]. Here we do not consider any incoherent processes, which allows us to give a discussion in terms of amplitude equations. After elimination of the continuum and nonresonant bound states, the reduced state vector of the atom is represented by $|\psi\rangle=G|0\rangle+A|1\rangle$, whose evolution is governed by an effective Hamiltonian H of the form ($H_{ij}=\langle i|H|j\rangle$),

$$\begin{aligned}
H_{00}&=-\tfrac{1}{2}i\gamma\ ,\\
H_{01}&=\Omega_3\left[1-\frac{i}{q_3}\right]+e^{i\phi}\Omega_1\left[1-\frac{i}{q_1}\right]\ ,\\
H_{10}&=\Omega_3\left[1-\frac{i}{q_3}\right]+e^{-i\phi}\Omega_1\left[1-\frac{i}{q_1}\right]\ ,\\
H_{11}&=-\tfrac{1}{2}i\Gamma-\delta\ .
\end{aligned}\tag{1}$$

Here, three-photon transitions from $|0\rangle$ to $|1\rangle$ are described by the (real) three-photon Rabi frequency Ω_3, while the strength of transitions from state $|0\rangle$ directly into the continuum is determined by the asymmetry parameter q_3. The one-photon Rabi frequency is complex and is written as $\Omega_1\exp(-i\phi)$, where ϕ is the phase difference between the electric-field components of pump and harmonic field. Direct one-photon transitions in the continuum are described by another asymmetry parameter q_1. The ionization rate from the ground state γ is given in terms of the autoionizing width Γ by

$$\gamma=\frac{4}{\Gamma}\left|\frac{\Omega_3}{q_3}+e^{i\phi}\frac{\Omega_1}{q_1}\right|^2 , \tag{2}$$

and depends on the strength of both the pump and the harmonic field. The detuning of the harmonic field from resonance is denoted by δ.

The instantaneous ionization rate of the atom is determined by the time derivative of the norm of the wave function $|\psi\rangle$,

$$\frac{\partial}{\partial t}\langle\psi|\psi\rangle=-i\langle\psi|(H-H^\dagger)|\psi\rangle . \tag{3}$$

The Hermitian matrix $i(H-H^\dagger)$ has two real nonnegative eigenvalues, one of which is zero because of (2); the other is the sum of the two decay rates γ and Γ. The corresponding instantaneous nondecaying and decaying eigenstates of $i(H-H^\dagger)$ are denoted by $|\psi_{ND}\rangle$ and $|\psi_D\rangle$, respectively, and have the form

$$\begin{aligned}|\psi_{ND}\rangle&=\frac{1}{N}\begin{bmatrix}-\Gamma/2\\ \Omega_3/q_3+\exp(-i\phi)\Omega_1/q_1\end{bmatrix} ,\\ |\psi_D\rangle&=\frac{1}{N}\begin{bmatrix}\Omega_3/q_3+\exp(i\phi)\Omega_1/q_1\\ \Gamma/2\end{bmatrix} ,\end{aligned} \tag{4}$$

with N the normalization factor $N=\sqrt{\Gamma(\Gamma+\gamma)}/2$. In general, a nondecaying state exists for any detuning for any intensities in any number of fields, as long as the decay rates obey a relation like (2). This condition is fulfilled if the relevant states are coupled to the same part of the continuum. With the help of (2), we see that a nondecaying state $|\psi_{ND}\rangle$ with inversion can be created if the atom is pumped sufficiently hard, such that the ionization rate γ from the ground state exceeds the autoionizing rate Γ.

If the atom is in the state $|\psi_{ND}\rangle$ at some instant of time, then the atom does not ionize at that particular moment. However, when the system evolves, the atom will in general not remain in the state $|\psi_{ND}\rangle$ since the state is not an eigenstate of the Hamiltonian. In the case where both the pump and harmonic field are time independent, we find two necessary and sufficient conditions for $|\psi_{ND}\rangle$ to be an eigenstate of H. The first is $|H_{01}|^2=|H_{10}|^2$, which implies that the phase ϕ between the pump and harmonic field must equal either 0 or π. The second condition determines the detuning δ in terms of the Rabi frequencies and asymmetry parameters, according to

$$\delta=-\frac{1}{2}\frac{(\Gamma-\gamma)(\Omega_3\pm\Omega_1)}{\Omega_3/q_3\pm\Omega_1/q_1} . \tag{5}$$

The $\pm$ sign refers to $\phi=0,\pi$, respectively. If the atom is pumped at a slightly different detuning or intensity, such that (5) is not satisfied, it will not reach the exact nondecaying state. Nevertheless, the atom will be ionized at a rate that is considerably lower than Γ. For instance, when the harmonic field is weak the stable state is determined by the pump field alone, and (5) reduces to $\delta=-\frac{1}{2}q_3(\Gamma-\gamma)\equiv\delta_0$, which has been derived before for a similar (laser-induced continuum structure) system [10]. By examining the eigenvalues of the Hamiltonian H, we can show that in this case the effective ionization rate γ_{eff} for detunings not too far from δ_0 is given by

$$\gamma_{\text{eff}}=\frac{4(\delta-\delta_0)^2\gamma\Gamma}{(\gamma+\Gamma)^3(1+q_3^2)} . \tag{6}$$

This expression is valid as long as $\gamma_{\text{eff}}\ll\Gamma,\gamma$.

We describe the harmonic field by a dimensionless complex amplitude $\langle a\rangle$, whose definition is such that $N\equiv|\langle a\rangle|^2$ is the number of harmonic photons per atom. The equation for the field amplitude $\langle a\rangle$ is given by

$$\begin{aligned}\frac{\partial}{\partial t}\langle a\rangle=&-i\sqrt{\bar{n}}\,\Gamma\left[1-\frac{i}{q_1}\right]\sigma_{10}\\ &-2\frac{\sqrt{\bar{n}}}{q_1}\left[\frac{\Omega_3}{q_3}+e^{-i\phi}\frac{\Omega_1}{q_1}\right]\sigma_{00} .\end{aligned} \tag{7}$$

The coherence and the ground-state population are related to the amplitudes A and G by $\sigma_{10}=AG^*$ and $\sigma_{00}=|G|^2$. Here $\bar{n}$ is the reduced atomic density

$$\bar{n}=n/n_0 , \quad n_0=\frac{2\epsilon_0\hbar\Gamma^2}{\omega_1|\langle 1|\hat{d}|0\rangle|^2} , \tag{8}$$

with $\hat{d}$ the dipole operator and n the atomic density. When the atom is in the nondecaying state $|\psi_{ND}\rangle$, the equation for $\langle a\rangle$ becomes

$$\frac{\partial}{\partial t}\langle a\rangle=2i\sqrt{\bar{n}}\left[\frac{\Omega_3}{q_3}+e^{-i\phi}\frac{\Omega_1}{q_1}\right]\frac{p\Gamma}{\Gamma+\gamma} , \tag{9}$$

where p is the total trapped population in $|\psi_{ND}\rangle$. The equation for the number of photons per atom is then

$$\frac{\partial}{\partial t}N=-4\sin\phi(t)\sqrt{\bar{n}}\sqrt{N}\frac{\Omega_3}{q_3}\frac{p\Gamma}{\Gamma+\gamma} . \tag{10}$$

The right-hand side is proportional to the pump-field Rabi frequency Ω_3, which shows that there is only parametric down or up conversion in the nondecaying state, depending on the sign of q_3 and $\sin\phi(t)$. The right-hand side being proportional to the square root of N rather than to N also implies that there is no amplification or absorption of the harmonic field by stimulated transitions. In other words, the state $|\psi_{ND}\rangle$ is transparent to the harmonic field radiation. This transparency is independent of inversion between the state $|0\rangle$ and $|1\rangle$. Thus we have here a case of inversion without amplification where $\gamma>\Gamma$. This property often goes together with the phenomenon of amplification without inversion. For this system, amplification without inversion has indeed been predicted [12].

The two equations (9) and (10) can be solved exactly. Here we only note that from (9) it follows that for a weak harmonic field the harmonic photons are produced at a phase $\phi=\pi/2$ for $q_3<0$ and $\phi=-\pi/2$ for $q_3>0$. The two-field nondecaying state, therefore, cannot be maintained when the harmonic field grows, since this would require a phase difference 0 or π. Although wrong in this sense, this phase difference is the optimum phase difference for the THG, as is clear from (10). We investigate the THG in more detail now.

We solved the set of coupled differential equations for the atom and for the harmonic field in time for a prescribed pulsed pump field for several values of the pump intensity, detuning, and pulse length. The atomic parameters chosen here correspond to transitions between a specific autoionizing doubly excited state $(3d5p)^1P$ and the ground state $(4s)^1S$ in Ca. Details can be found in [13]. The unit of time used is the inverse autoionization width $\Gamma^{-1}=8.38$ fs. The density n_0 as defined in (8) is relatively large in this case, $n_0 \approx 1.3\times 10^{21}$ cm $^{-3}$, so that the scale factor $\bar{n}$ for realistic atomic densities is small. In all of the following we take $n=1.3\times 10^{17}$ cm^{-3}, so that $\bar{n}=10^{-4}$. The units of intensity for the three-photon pump field I_{p0} and for the third harmonic field I_{t0} are defined by the relations $\Omega_3/\Gamma=(I_p/I_{p0})^{3/2}$ and $\Omega_1/\Gamma=(I_t/I_{t0})^{1/2}$, with I_p and I_t the pump and harmonic intensities. For Ca, one has the values $I_{p0}=1.96\times 10^{12}$ W/cm^3 and $I_{t0}=7.23\times 10^{12}$ W/cm^2. Finally, the q parameters are $q_3=-0.12$ and $q_1=4.38$.

We consider pumping by a Gaussian pulse. The intensity of the pump field as a function of time is given by $I_p(t)=I_0\exp[-(t/\tau)^2]$, where τ is referred to as the length of the pulse and I_0 as the strength or peak intensity. The condition (5) for a steady state can be fulfilled only at two instants of time during a pulse. However, for appropriate pulse strength and length, the detuning and intensities are not too far away from that condition, so that the ionization of the atoms is slowed down considerably during the major part of the pulse, with the decay rate approximately given by (6).

In Fig. 1, we plot the total number of photons produced per atom by a pulse of length $\tau=100\Gamma^{-1}\approx 0.8$ ps and peak intensity $I_0=0.19I_{p0}\approx 3.7\times 10^{11}$ W/cm^2 (being the optimum intensity for this given pulse length) as a function of the detuning. This intensity is sufficiently low so that ac Stark shifts can be neglected. The graph shows that the third-harmonic production is not very sensitive to the detuning, the width being given by $\approx 10^{13}$ Hz. For this pulse the maximum number of harmonic photons produced at the optimum detuning $\delta=0.014\Gamma$ corresponds to an intensity of $I_t\approx 6.4\times 10^8$ W/cm^2.

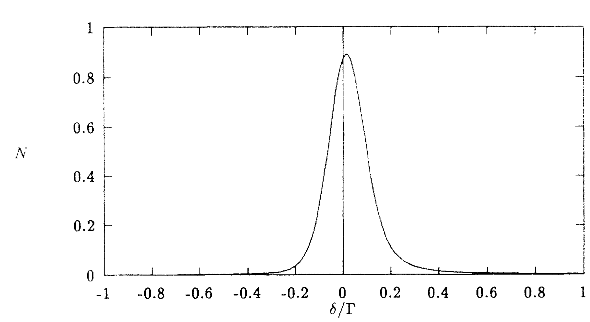

FIG. 1. Number of photons per atom, N, produced in an autoionizing system by a pulse of length $\tau=100\Gamma^{-1}$ and peak intensity $I_0=0.19I_{p0}$ as a function of the detuning δ in units Γ. Atomic parameters are given in the text.

Then we plot in Fig. 2(a) as a function of the dimensionless peak intensity $J\equiv I_0/I_{p0}$ for several values of τ, the maximum number of photons produced at the optimum detuning. The optimum field strength for these pulses is seen to be $J\approx 0.2$. For larger intensities the production of photons decreases, since the atoms then do not reach a nondecaying state and hence are more rapidly ionized.

We now wish to compare the total THG in the autoionizing (AI) system with the THG in a bound system coupled to the continuum by a three-photon transition through an intermediate two-photon resonance (TPR). This TPR system is the usual one used to produce harmonic photons. The attractive feature of this system is that in weak fields the ionization rate of the atoms is small compared to the rate in the AI system, since the ionization of *both* states is field induced. To make a meaningful comparison, the atomic parameters are chosen to correspond to the state $(4s5s)^1S$ in Ca as the intermediate state [13]. This state is coupled to the ground state by two-photon transitions, where the pump photons are in the same frequency range as for the AI system. The decay rate of this state as a function of the pump intensity is $\Gamma_1=0.415J\Gamma$, which is indeed relatively small for the intensities $J\approx 0.2$ considered above. The two-photon Rabi frequency is given by $\Omega_2=0.848J\Gamma$, while

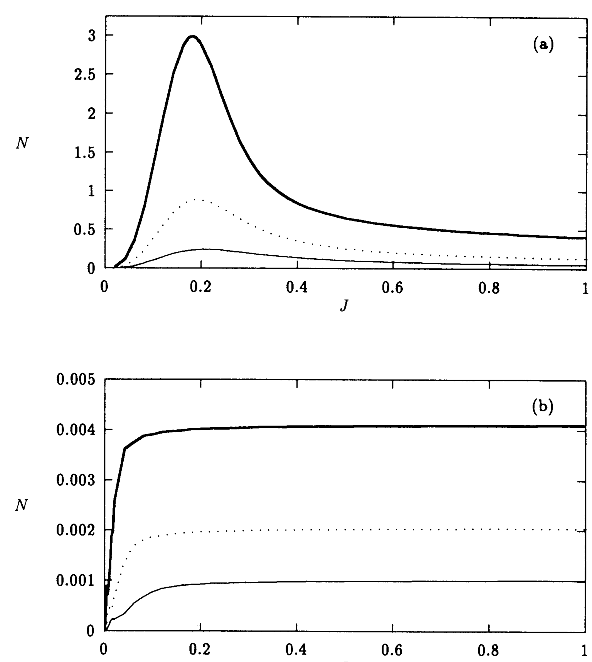

FIG. 2. Maximum number of photons per atom N produced at optimum detuning by a pulse of length $\tau=50\Gamma^{-1}$ (thin line), $\tau=100\Gamma^{-1}$ (dotted line), and $\tau=200\Gamma^{-1}$ (thick line), as a function of the peak intensity $J=I_0/I_{p0}$. (a) For an autoionizing system and (b) for an intermediate two-photon resonance system. Atomic parameters are given in the text.

the Rabi frequency $\Omega_{(1,-1)}$, corresponding to the transition from the intermediate state to the ground state through the absorption of one pump and the emission of one harmonic photon, is determined by $4.3\Omega_2\Omega_{(1,-1)}=\Omega_3\Omega_1$. Finally, the two-photon q parameter is $q_2=1.867$.

In Fig. 2(b) we plot for this TPR system the maximum number of photons produced at the optimum detuning as a function of the intensity J. The plots show the comparison for the same values of the pulse length τ as in Fig. 2(a). First we note that the intensity dependence is different from the AI system. The oscillations for small intensities are due to Rabi oscillations, which can occur since the decay of both discrete states is small. Furthermore, the number of photons produced saturates for large intensity, there is no optimum intensity. The TPR system does not have the possibility to remain trapped in a nondecaying state. This implies that the AI system will be better in terms of THG when the pulses get longer. Indeed, we find that the total harmonic generation grows linearly with τ for the TPR system, but quadratically for the AI system for the range of pulse lengths considered. For the atomic parameters chosen here, the TPR system is better only for very short pulses with $\tau<0.2\Gamma^{-1}$, but the autoionizing system is superior for larger τ. For realistic pulse length $\tau\approx 100\Gamma^{-1}$ the latter is better by three orders of magnitude. For other atomic systems the numbers may be more favorable for the TPR system, but the general conclusion that longer pulses favor the AI system remains the same.

In conclusion, we have shown how the combination of induced transparency with a stabilized state can be exploited to produce a large third-harmonic field. This stable state can often be present in autoionizing systems when irradiated by coherent light sources. If an incoherent decay channel is present, or if the pumping is incoherent, the stable state will be affected due to the loss of coherence between the ground and autoionizing state. In our specific case of Ca, however, the influence of incoherent transitions is small, so that the results are not severely affected by them. Furthermore, although we have not included the effects of propagation here, they are expected to add more physics to the phenomena but not to interfere destructively under the proper phase-matching conditions. Propagation and incoherence effects will be discussed elsewhere.

The work of S.J.v.E. is supported by the Human Capital and Mobility program under Contract No. ERBCHBGCT920192.

---

[1] D. C. Hanna, M. A. Yuratich, and D. Cotter, *Nonlinear Optics of Free Atoms and Molecules* (Springer-Verlag, Berlin, 1979).

[2] A. T. Georges, P. Lambropoulos, and J. H. Marburger, Phys. Rev. A **15**, 300 (1977), and references therein.

[3] S. C. Wallace and G. Zdasiuk, Appl. Phys. Lett. **28**, 449 (1976).

[4] G. Alber and P. Zoller, Phys. Rev. A **27**, 1373 (1983).

[5] S. E. Harris, J. E. Field, and A. Imamoğlu, Phys. Rev. Lett. **64**, 1107 (1990), and references therein.

[6] M. O. Scully, Phys. Rev. Lett. **67**, 1855 (1991).

[7] G. Z. Zhang, K. Hakuta, and B. P. Stoicheff, Phys. Rev. Lett. **71**, 3099 (1993).

[8] P. Lambropoulos and P. Zoller, Phys. Rev. A **24**, 379 (1981).

[9] K. Rzążewski and J. H. Eberly, Phys. Rev. Lett. **47**, 408 (1981); J. H. Eberly, K. Rzążewski, and D. Agassi, *ibid.* **49**, 693 (1982); Z. Deng and J. H. Eberly, J. Opt. Soc. Am. B **1**, 102 (1984).

[10] P. E. Coleman and P. L. Knight, J. Phys. B **15**, L235 (1982); **15**, 1957(E) (1982); P. E. Coleman, P. L. Knight, and K. Burnett, Opt. Commun. **42**, 171 (1982).

[11] T. Nakajima and P. Lambropoulos, Phys. Rev. Lett. **70**, 1081 (1993).

[12] Jian Zhang, P. Lambropoulos, and X. Tang, Phys. Rev. A **50**, 1935 (1994).

[13] Jian Zhang, Ph.D. thesis, University of Southern California, 1991 (unpublished); P. Lambropoulos, Jian Zhang, and X. Tang, *Coherence Phenomena in Atoms and Molecules in Laser Field* (Plenum, New York, 1992).