Phase Control of Photoabsorption in Optically Dense Media

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We present a self-consistent theory, as well as an illustrative application to a realistic system, of phase control of photoabsorption in an optically dense medium. We demonstrate that, when propagation effects are taken into consideration, the impact on phase control is significant. Independent of the value of the initial phase difference between the two fields, over a short scaled distance of propagation, the medium tends to settle the relative phase so that it cancels the atomic excitation. In addition, we find some rather unusual behavior for an optically thin layer.

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After the initial ideas [1] and experimental demonstration [2,3] of the feasibility of the control of photoabsorption and its products through the control of the relative phase of two fields, much work in atoms [4] and molecules [3] has explored a variety of processes. Many interesting issues [3–5] have been raised and clarified, establishing thus the idea as a useful tool. With the exception of one paper [6], however, theory and experiment have dealt only with single-atom (molecule) situations. But, if the ideas are to be contemplated for applications, the issue of propagation is crucial. Addressing this issue is the purpose of this Letter.

We have chosen the fundamental scheme [2,5] which has served as a benchmark for much of the initial and the continuing work. We consider the excitation of a bound transition through the combined effect of a single- and a three-photon transition via two fields whose relative phase is controlled externally. We formulate and examine the propagation of a bichromatic electromagnetic field E through an optically dense medium consisting of Xe atoms. This electric field is a function of time t and space coordinate z and is composed of the fundamental and its third harmonic fields that have the same (linear) polarization and frequencies ω_f and $\omega_h = 3\omega_f$, respectively. It is expressed as

$$E(z,t) = \frac{1}{2}\left[E_f e^{i(k_f z-\omega_f t)} + E_h e^{i(k_h z-\omega_h t)} + \text{c.c.}\right], \tag{1}$$

where $E_j = \mathcal{E}_j e^{-i\phi_j}$, $j = f, h$, with $\mathcal{E}_j$ and ϕ_j the slowly varying in time and space real amplitude and phase of the corresponding field, and $k_j = \omega_j n_j c^{-1}$, with n_j the refraction index of the host medium at frequency ω_j. Although in our present treatment the host medium is vacuum, and thus $n_f = n_h = 1$ and $k_h = 3k_f$, for the sake of generality, e.g., presence of a buffer gas, we shall keep in the formalism the refraction index. The frequencies $\omega_{h,f}$ are chosen so that one harmonic photon and three fundamental photons are at near resonance with the transition from the ground state ($|1\rangle$) to the $6s$ state ($|2\rangle$) of Xe. A two-photon transition due to the strong fundamental or a one-photon transition due to the harmonic fields lead to the ionization continuum (states $|c\rangle$) of the atom. As we intend to explore intensities of the fields for which the one- and three-photon transition amplitudes between states $|1\rangle$ and $|2\rangle$ are of comparable magnitude so as to maximize the modulation depth, the transition $|2\rangle \rightarrow |c\rangle$ would be dominated by the two-photon process, and the one-photon ionization due to harmonic photon can be neglected. Experimental contexts for the situation we are considering have been detailed in the literature.

Beginning with the second order wave equation for the field $E(z,t)$, in the slowly varying (during an optical cycle) amplitude approximation one neglects all second derivatives and, after projecting onto the corresponding mode function $\exp[i(\omega_j t - k_j z)]$, $j = f, h$, one arrives at

$$\frac{\partial E_j}{\partial z} + \frac{n_j}{c}\frac{\partial E_j}{\partial t} = \frac{1}{c\epsilon_0 n_j}\left[i\frac{\omega_j}{2}P_j - \frac{\partial P_j}{\partial t}\right], \tag{2}$$

where $P_j = P_j' e^{-i\phi_j}$ (P_j' being complex) is the slowly varying in time and space field-induced medium polarization at frequency ω_j. Consistently with Eq. (1), it can be expressed as

$$P(z,t) = \frac{1}{2}\left[P_f e^{i(k_f z-\omega_f t)} + P_h e^{i(k_h z-\omega_h t)} + \text{c.c.}\right]. \tag{3}$$

The most general approach to the calculation of the response of the medium is through the atomic density matrix ρ which obeys the equation $\partial_t \rho = -i\hbar^{-1}[H_{\text{atom}} + D, \rho]$, with H_{atom} the free atomic Hamiltonian and $D = -\mu E$ the atom-field interaction in the dipole approximation where μ is the electric dipole operator. Introducing the rotating wave approximation and adiabatically eliminating the continuum and all virtual (nonresonant) bound states (connecting by the lowest-order paths the states $|1\rangle$ and $|2\rangle$), the slowly varying density matrix elements of the two remaining states $\sigma_{11} \simeq \rho_{11}$, $\sigma_{22} \simeq \rho_{22}$, and $\sigma_{21} \simeq \rho_{21}\exp[i3(\omega_f t + \phi_f - k_f z)]$ are found to obey the following set of equations:

$$\frac{\partial}{\partial t}\sigma_{11} = \gamma\sigma_{22} - \mathrm{Im}\left[\left(\frac{\mu_{12}^{(3)}}{\hbar}\mathcal{E}_f^3 + e^{i\theta}\frac{\mu_{12}}{\hbar}\mathcal{E}_h\right)\sigma_{21}\right], \tag{4a}$$

$$\frac{\partial}{\partial t}\sigma_{22} = -(\gamma + \gamma_{\mathrm{ion}})\sigma_{22} + \mathrm{Im}\left[\left(\frac{\mu_{12}^{(3)}}{\hbar}\mathcal{E}_f^3 + e^{i\theta}\frac{\mu_{12}}{\hbar}\mathcal{E}_h\right)\sigma_{21}\right], \tag{4b}$$

$$\frac{\partial}{\partial t}\sigma_{21} = -\left[\frac{\gamma + \gamma_{\mathrm{ion}}}{2} + i\left(\Delta - 3\frac{\partial\phi_f}{\partial t}\right) + i\frac{s_1 - s_2}{2\hbar}\mathcal{E}_f^2\right]\sigma_{21} + i\left(\frac{\mu_{12}^{(3)}}{2\hbar}\mathcal{E}_f^3 + e^{-i\theta}\frac{\mu_{12}}{2\hbar}\mathcal{E}_h\right)(\sigma_{11} - \sigma_{22}), \tag{4c}$$

where γ is the radiative decay rate of level $|2\rangle$, $\gamma_{\mathrm{ion}} \propto (\mu_{2c}^{(2)} I_f)^2$ is the two-photon ionization rate of $|2\rangle$ being proportional to the square of the intensity $I_f \propto \mathcal{E}_f^2$ of the fundamental ($\mu_{2c}^{(2)}$ is the effective two-photon matrix element for the fundamental field on the transition $|2\rangle \rightarrow |c\rangle$), $\mu_{12}^{(3)}$ is the effective three-photon matrix element for the fundamental field on the transition $|1\rangle \rightarrow |2\rangle$, s_1 and s_2 are the lowest-order Stark shift coefficients (polarizabilities) of levels $|1\rangle$ and $|2\rangle$, respectively, and $\mu_{12} \equiv \langle 1|\mu|2\rangle$ is the matrix element of the electric dipole operator μ. Finally Δ is the detuning of both fields from the $|1\rangle \rightarrow |2\rangle$ transition resonance and $\theta = (\phi_h - 3\phi_f) - (k_h - 3k_f)z$ is their relative phase.

Consider now the polarization $P(z,t) = N\,\mathrm{Tr}[\mu\rho]$ of a medium of atomic density N. In expanding the trace of this equation, we again follow the same procedure as in obtaining Eqs. (4), i.e., we use the adiabatic approximation to express all density matrix elements that do not refer to the states $|1\rangle$ and $|2\rangle$ in terms of the three main elements σ_{11}, σ_{22}, and σ_{21}. Equating the result with Eq. (3), identifying and grouping together terms oscillating with the same frequencies, we obtain

$$P_f' = 2N[\mathcal{E}_f(s_1\sigma_{11} + s_2\sigma_{22}) + 3\mu_{12}^{(3)}\mathcal{E}_f^2\sigma_{21} + i\pi\hbar^{-1}|\mu_{2c}^{(2)}|^2\mathcal{E}_f^3\sigma_{22}], \tag{5a}$$

$$P_h' = 2N\mu_{12}\sigma_{21}e^{i\theta}. \tag{5b}$$

Those equations, together with the Maxwell equation (2) and the atomic density matrix equations (4), provide a complete description of our system in terms of a closed set of equations.

To present the numerical results for Xe, we use the parameters calculated previously [5] via multichannel quantum defect theory and appropriately converted to conform to the present definitions. For illustration purposes, it is desirable to have a maximally pronounced interference of the fundamental and harmonic fields. The respective Rabi frequencies are given by the first and second terms in the parentheses of Eq. (4a). To obtain, for example, complete cancellation at $\theta = \pi$ when these two terms are purely real and have opposite signs, it is obvious that the peak values and the temporal widths of both Rabi frequencies should be equal so as to overlap completely. Let the strong fundamental field have a Gaussian temporal profile with a peak amplitude $\mathcal{E}_f^{\max} \equiv \mathcal{E}_f(t = t_{\max})$ and width τ_f. Then the peak amplitude and width of the weak harmonic field should satisfy the relations

$$\mathcal{E}_h^{\max} = \frac{\mu_{12}^{(3)}}{\mu_{12}}(\mathcal{E}_f^{\max})^3, \qquad \tau_h = \frac{\tau_f}{\sqrt{3}}. \tag{6}$$

In Fig. 1 we plot the ion yield $Q = [1 - \sigma_{11}(t) - \sigma_{22}(t)]_{t\rightarrow\infty}$ at $z = 0$ as a function of the relative phase θ for three different intensities I_f of the fundamental. In all cases, the detuning Δ is taken such that it compensates the relative Stark shift of levels $|1\rangle$ and $|2\rangle$ at the maximum $t_{\max}$ of the pulse, the harmonic pulse duration $\tau_h = 1$ ns, and the conditions (6) are satisfied. In this figure, for all intensities and relative phase $\theta = \pi$, the ionization vanishes completely since the two transition amplitudes interfere destructively and the second term on the right-hand side of Eq. (4a), responsible for the stimulated transition from $|1\rangle$ to $|2\rangle$, is equal to zero throughout the duration of the pulses. Consequently, the medium practically does not interact with the fields and the atoms are "trapped" in their ground state $|1\rangle$. The more surprising result, however, is that in the case $I_f^{\max} = 8 \times 10^{10}$ W/cm^2, maximal ionization is found not for $\theta = 0, 2\pi$, as one would expect and is the case for the other intensities. This is a manifestation of the quantum-mechanical interference resulting from the fact that for this set of parameters in Eqs. (4) the terms responsible for the stimulated transition reach the maxima at $\theta \simeq \pi \pm 0.28\pi$ where the ionization peaks are located. The numerical simulations also show that, while keeping the conditions (6) satisfied, with decreasing pulse duration τ_f, the ion yield reduces and its peaks at $\theta \neq 0, 2\pi, \ldots$ gradually disappear, which is analogous to the decreasing of intensity since the total energy of the pulse lessens. Increasing the intensity, however, results in a narrower dip in the ionization profile and a shift of its peaks towards the values of θ that are closer to π.

Let us turn now to the propagation effects. The results presented below are obtained for a density of atoms $N = 10^{13}$ cm^{-3}. This, however, does not imply any limitation on the generality of the discussion since, as one can easily verify, the parameter $zN\Sigma$, where Σ is the laser beam cross section, is a propagation constant, and thus it is always possible to rescale the problem to any desired density and propagation length z. Conditions (6) are assumed at the entrance to the medium. As we have noted above, in

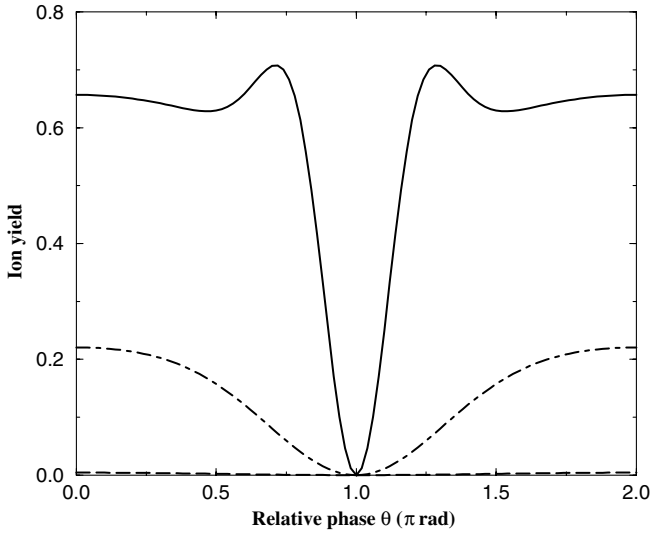

FIG. 1. Ion yield $Q = (1 - \sigma_{11} - \sigma_{22})_{t\to\infty} \simeq 1 - \sigma_{11}(t \to \infty)$ versus relative phase θ for three different peak intensities of the fundamental: $I_f^{\max} = 1 \times 10^{10}$ W/cm^2 (dashed line), $I_f^{\max} = 3 \times 10^{10}$ W/cm^2 (dot-dashed line), $I_f^{\max} = 8 \times 10^{10}$ W/cm^2 (solid line).

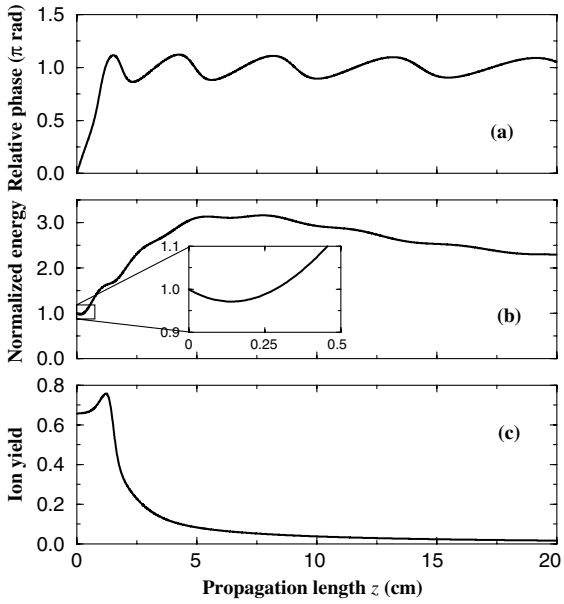

FIG. 2. Relative phase $\theta(z, t = t_{\max} + z/c)$ (a), normalized energy $S_h(z)/S_h(0)$ of harmonic field (b), and ion yield $Q(z)$ (c) versus propagation length z for the case $I_f^{\max} = 8 \times 10^{10}$ W/cm^2.

the case of initial phase difference $\theta(0, t) = \pi$, the atoms stay in the ground state and the medium appears to be "transparent" to both fields; neither the fundamental nor the harmonic experience any remarkable distortion of their shapes or total energy $S_j(z) \propto \int dt |\mathcal{E}_j(z, t)|^2$, $j = f, h$, over distances of propagation z as large as ~50 cm. The accumulated over this distance change of the relative phase is only $\sim 10^{-3}\pi$ rad, which is due to the field independent phase shift of the fundamental, given by the term in the parentheses of Eq. (5a).

Consider next the case $\theta(0, t) = 0$; i.e., at the entrance to the cell the two fields interfere constructively. The results corresponding to the parameters of Fig. 1 with $I_f^{\max} = 8 \times 10^{10}$ W/cm^2 are collected in Figs. 2 and 3. One can see in Fig. 2 that, in the course of propagation, the relative phase θ (taken at the dynamic pulse maximum $t_{\max} + z/c$) grows rapidly and over a distance of the order of 1 cm reaches the value π, at which the initial constructive interference of the two fields turns to destructive. At the same time, the total energy of the harmonic pulse, after a small reduction over a short interval of z, begins to increase as a result of the energy transfer from the strong fundamental field, in the parametric conversion process. This small reduction of the harmonic takes place only at the beginning of the propagation, when the relative phase is still close to 0 and the two fields interfere constructively, in the process of excitation of atoms from the ground state $|1\rangle$ to the state $|2\rangle$, while the generated part of the harmonic field is out of phase with the fundamental approximately by π and continues to build up with a slight oscillation around the value π of the phase. It is important to mention that throughout the propagation the amplitude and the phase of the fundamental field do not change significantly. This is because the number of photons contained in that pulse exceeds by many (≥ 6) orders of magnitude the number of atoms the pulse interacts with over the distance of $z \leq 20$ cm. Comparing the three graphs of Fig. 2, one can see that with increasing θ and S_h the ionization probability first also grows, which is consistent with the previous discussion related to that intensity of the fundamental field. But as θ approaches π, the ion yield drops almost exponentially until $Q \simeq 10\%$. This residual ionization that is present even at $\theta \simeq \pi$ (and tends to 0 rather slowly) is caused by the fact that, because of the significant increase of the total energy of harmonic field, conditions (6) are not completely satisfied and the upper atomic level $|2\rangle$ acquires population due to that fraction of the generated field which exceeds the initial. Since the temporal widths of the pulses are less than the (radiative) relaxation time of the atomic coherence σ_{21} ($\gamma^{-1} \simeq 2$ ns), a significant fraction of the harmonic pulse amplitude is generated behind the fundamental (Fig. 3). That part of the amplitude is then attenuated due to the atomic relaxation. Thus the total energy of the harmonic, after passing a maximum at $z \simeq 5$–7 cm, then decays slowly back. Under these conditions, the leading part of the harmonic pulse that falls under the temporal shape of the fundamental is by $\theta \simeq \pi$ out of phase with the latter and therefore the ionization vanishes, while the generated tail is continuously scattered by the atoms in the process of radiative decay. The oscillations of the relative phase around π are also slowly damped and the propagation reaches a "dynamic equilibrium."

We note finally that a similar behavior of the system is obtained for a range of intensities we have explored. The

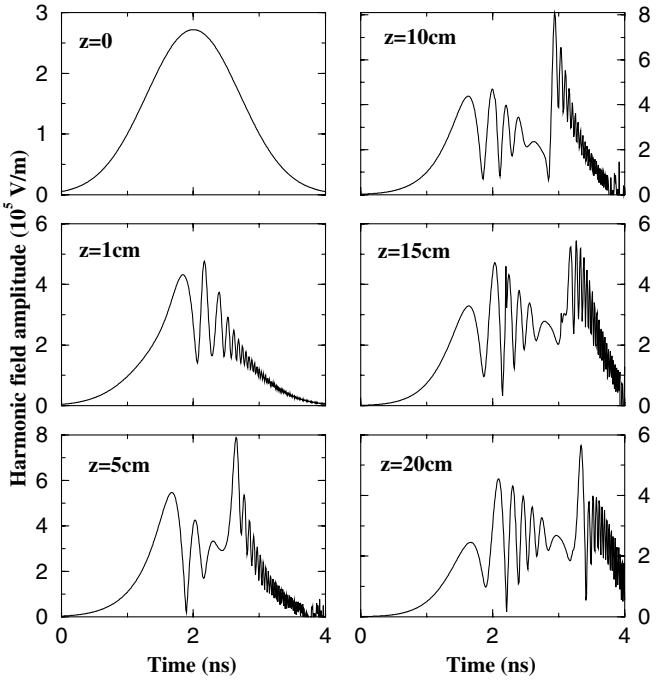

FIG. 3. Temporal profile of the amplitude $\mathcal{E}_h$ of harmonic field at different z. All parameters are as in Fig. 2.

main difference is that for weaker fields ($I_f^{\max} = 3 \times 10^{10}$ and 1×10^{10} W/cm^2) the ion yield does not exhibit a maximum other than at $z = 0$ and drops to zero much faster as z increases, which is consistent with the discussion above.

We have examined the problem of propagation in the simplest context of phase control, namely, the excitation of a bound state, which has been used as a prototype in much of the initial work [1,2]. As discussed here, the problem bears resemblance to earlier works [7–10] on cancellation in third harmonic generation experiments, and so does the whole issue of phase control. The relevance and possible impact of propagation has been recognized by Chen and Elliott [6] who presented data and an interpretation in terms of rate equations [10]. Their study showed evidence of nonlinear coupling, such as those discussed above, and called for "more rigorous techniques" in the approach to this basic problem. In the limit of validity of rate equations, our results do indeed recapture the equations employed in their analysis. It will be interesting to explore this issue under more general conditions, such as the excitation of states embedded in continua, on which we expect to report elsewhere. The basic features of our analysis should, however, remain valid.

In summary, we have shown that the propagation of a bichromatic field, with a preselected initial relative phase, has a profound effect. Over a rather short scaled distance and independent of its initial value, the relative phase settles to a value that makes the medium transparent to the radiation, thus precluding further excitation and consequently control. The scaled distance $zN\Sigma$ does of course involve the density of the species and the cross section of the laser beam, which suggests some flexibility on the choice of these parameters. In any case, however, the actual length of the interaction region over which control can be active will be defined and limited by the combination of the above parameters, as well as by the geometry of the focused or unfocused laser beam. Briefly, for not very low atomic densities ($N > 10^{12}$ cm^{-3}), the harmonic field settles to the steady-state value within a thin layer where a focused beam is well approximated by a plane wave. In the presence of large ac Stark shifts, however, a detailed analysis including specific experimental parameters is mandatory.

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