Harmonic-generation control

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In this paper we examine some effects of quantum interference on high harmonic generation. We demonstrate in particular that preparing the initial state in a coherent superposition of bound states leads to a harmonic spectrum with distinct plateaus with different conversion efficiencies. We show how this scheme may provide a way of controlling the coherent output that is produced in an experiment. [S1050-2947(96)03611-6]

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I. INTRODUCTION

While some aspects of harmonic generation are both theoretical and experimentally quite well understood, the way in which the coherent radiation output can be controlled for practical purposes is still a matter of debate. By controlling harmonic generation we mean the possibility of modifying the spectrum to our convenience, for example, by enhancing only a narrow range of harmonics around a desired energy in the total spectrum or extending the harmonic spectrum to higher orders than the ones predicted by the well-known cutoff law U_i+3U_p [1] (where $U_p=E^2/4\omega^2$ is the ponderomotive energy expressed in atomic units, ω is the frequency of the driving laser field, and U_i refers to the ionization potential of the atomic state).

The semiclassical model of harmonic generation [1,2] is, in spite of its simplicity, correct in showing the importance of the electron's classical returning trajectories in the harmonic-generation process. In particular, it suggests that it should be possible to change the spectrum by modifying these trajectories. Early work [3] using two different lasers with commensurate frequencies interacting simultaneously with an atom reinforces this idea. For that combination of fields $E_1\sin(\omega t)+E_2\sin(3\omega t+\varphi)$, the plateau structure extends up to U_i+kU_p with $k>3$ (here U_p still refers to the ponderomotive potential of the fundamental field). The value of k is determined by the intensity of the fields as well as their relative phase. The two-color field modifies the returning trajectories and a classical analysis of those trajectories once again gives an accurate prediction of the position of the cutoff. Furthermore, when a second color is superimposed it not only extends the spectrum to higher orders but also produces a clear enhancement (up to 2–3 orders of magnitude) in the low-energy region of the spectrum (with energies below U_i+U_p) [4]. To our knowledge, however, it is not possible to enhance the higher-energy harmonics by modifying the relative phase or the intensity ratio between both fields. Achieving such a goal would require the selective injection of electrons into the continuum only (or mainly) at those times that correspond to returning trajectories with maximum kinetic energy. At present this does not look as though it can be straightforwardly achieved [5].

It appears at first sight easier and more beneficial to modify the classical aspects of the process, i.e., the recolliding trajectories, than to modify the steps that deal with the intrinsically quantum aspects of it. In this paper, however, we shall demonstrate that more profound effects are produced when the ionization and rescattering events are modified. This can be achieved, for example, by tailoring the initial state to allow different paths in the recombination event. If the initial state is prepared in a coherent superposition of different bound states [6,7], in which only the more loosely bound state becomes ionized by the action of the field, the harmonic spectrum contains two distinct set of harmonic plateaus [8]. Our aim is to show how one may take advantage of this feature (each plateau presents a distinct conversion efficiency) to manipulate the harmonic spectrum.

The paper is organized as follows. In the next section we shall introduce the model we use and discuss the main features of the recombination event in terms of the values and phases of the dipole matrices. In Sec. III we analyze the dependence of the conversion efficiency on the initial states. This aspect is further discussed in the Appendix. In Sec. IV we shall demonstrate how to use these features to control the harmonic generation. Finally, in Sec. V we discuss the feasibility of the practical realization of the scheme we propose.

II. COHERENT SUPERPOSITIONS

Our results are based on quantum interference effects in recombination via different states. We prepare the initial state in a superposition of the ground state $|g\rangle$ and some excited state denoted by $|e\rangle$ with a fixed though arbitrary phase difference between both states

$$\Psi(r,t)=\{\alpha|g\rangle+\beta e^{-i\phi}|e\rangle\} \tag{1}$$

($|\alpha|^2+|\beta|^2=1$). We shall take $\phi=0$ to simplify the notation. The laser parameters (I and ω) are chosen such that only the excited state is depleted by ionization. We do this because it is sufficient, and requires much lower intensities, to promote the electron into the continuum from the excited state. Since we aim to describe a rather general way of possible control over the harmonic emission, we perform our calculations for a simple hydrogenic ion He^+. At this point it is worth pointing out that the results we present are not directly related to the structure of the atomic potential and therefore can be straightforwardly extended to any type of ion or atom.

In a previous paper [8] we demonstrated that the harmonic spectrum obtained from an initial state similar to the one mentioned in Eq. (1) consists of two distinct plateaus. We can see why this should be so by splitting up the various contributions to the dipole acceleration for the coherent superposition as

$$\begin{aligned}\ddot{d}(t) &\equiv \langle \Psi(r,t)|\ddot{z}|\Psi(r,t)\rangle \\ &= \langle \psi_g(r,t)|\ddot{z}|\psi_g(r,t)\rangle + \langle \psi_e(r,t)|\ddot{z}|\psi_e(r,t)\rangle \\ &\quad + \langle \psi_g(r,t)|\ddot{z}|\psi_e(r,t)\rangle + \text{c.c.} \end{aligned} \tag{2}$$

The first two terms on the right-hand side of Eq. (2) are simply the dipole accelerations one would obtain starting in the ground and excited states, respectively. The third term can be thought of as an interference between the two parts of the solution. If we assume that the ground state is not depleted and the excited state is not coupled to any other bound state during the pulse, we can write the time-dependent wave functions of the ground and excited states in the form

$$|\psi_g(r,t)\rangle = \alpha e^{-iU_g t}|g\rangle,$$

$$|\psi_e(r,t)\rangle = \beta\left(b_e(t)e^{-iU_e t}|e\rangle + \int dc\, b_c(t)e^{-iU_c t}|c\rangle\right). \tag{3}$$

Here $b_e(t)$ and $b_c(t)$ are the time-dependent amplitudes of the excited and continuum states and we have factorized out the energy dependence of the bare states (atomic units are used throughout) [9]. That means that only the last two terms of Eq. (2) will contribute to the harmonic generation, and since continuum-continuum transitions have no significant influence to harmonic generation, we can rewrite the relevant contributions to the acceleration as

$$\langle \psi_e(r,t)|\ddot{z}|\psi_e(r,t)\rangle = |\beta|^2 \int dc\, b_e^*(t)b_c(t)\langle e|\ddot{z}|c\rangle e^{i(U_e-U_c)t} + \text{c.c.}, \tag{4}$$

$$\langle \psi_g(r,t)|\ddot{z}|\psi_e(r,t)\rangle = \alpha^*\beta\left(b_e(t)\langle g|\ddot{z}|e\rangle e^{i(U_g-U_e)t} + \int dc\, b_c(t)\langle g|\ddot{z}|c\rangle e^{i(U_g-U_c)t}\right) + \text{c.c.} \tag{5}$$

An inspection of the above equations show that if the ionization of the excited state is small, at least in one optical cycle, the time-dependent integrals of Eqs. (4) and (5) differ only by a factor $e^{i(U_g-U_e)t}$. In other words, the third term in the dipole acceleration gives rise to a similar plateau to the second term, but shifted in energy by an amount equal to the gap between the ground and excited energies. This is the second plateau observed in the harmonic spectra. [The term proportional to $\langle g|\ddot{z}|e\rangle$ in Eq. (5) simply corresponds to the transition between the initial states of the superposition. If both states share the same parity, this term is equal to zero; otherwise, the spectrum will contain a peak corresponding to this transition.] We can gain further insight into our interpretation of these expressions by comparing the spectrum obtained directly from the dipole acceleration

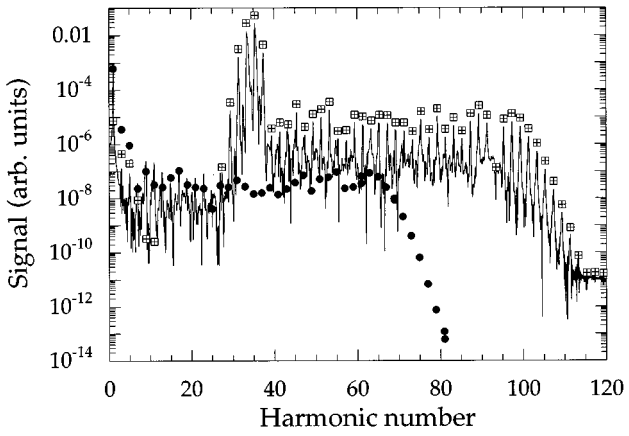

FIG. 1. Harmonic-generation spectra from the He^+ ion at $I = 1.15\times10^{14}$ W/cm^2 and $\omega = 0.042$ a.u. ($\lambda = 1$ μm) starting from a coherent superposition of the ground and the first excited state ($2s$) with equally weighted populations $(1/\sqrt{2})(|g\rangle + |2s\rangle)$ (solid line). Full dots correspond to the harmonic spectrum computed from the dipole projection onto the excited state and the squares correspond to the dipole projection onto the ground state. (The pulse lasts 16 cycles and is linearly ramped in the two first cycles.)

$\langle \Psi(r,t)|\ddot{z}|\Psi(r,t)\rangle$ with the spectra obtained by projecting the dipole acceleration [10] onto the ground and excited field-free states, respectively, i.e.,

$$\begin{aligned}\ddot{d}(t)_{pg} &= \langle \Psi(r,t)|g\rangle\langle g|\ddot{z}|\Psi(r,t)\rangle \\ &= \alpha^\star\beta\left(b_e(t)\langle g|\ddot{z}|e\rangle e^{i(U_g-U_e)t} + \int dc\, b_c(t)\langle g|\ddot{z}|c\rangle e^{i(U_g-U_c)t}\right), \end{aligned} \tag{6}$$

$$\begin{aligned}\ddot{d}(t)_{pe} &= \langle \Psi(r,t)|e\rangle\langle e|\ddot{z}|\Psi(r,t)\rangle \\ &= |\beta|^2 \int dc\, b_c(t)b_e(t)\langle e|\ddot{z}|c\rangle e^{i(U_e-U_c)t}. \end{aligned} \tag{7}$$

In Fig. 1 we show the harmonic-generation spectrum computed numerically from the dipole acceleration $\langle \Psi(r,t)|\ddot{z}|\Psi(r,t)\rangle$ (full line) alongside the spectra obtained numerically from the dipole projections $\langle \Psi(r,t)|g\rangle\langle g|\ddot{z}|\Psi(r,t)\rangle$ (squares) and $\langle \Psi(r,t)|e\rangle\langle e|\ddot{z}|\Psi(r,t)\rangle$ (full dots). The figure clearly shows that the projection onto the ground state is mainly responsible for the second (higher-energy) plateau, while recombination into the excited state gives the main contribution to the first one. Figure 1 also shows that the projection onto the ground state also gives a contribution to the first plateau and therefore some interference between both contributions is present. Finally, projecting the dipole acceleration onto any other atomic bare state produces a harmonic signal several orders of magnitude smaller than those shown in Fig. 1.

To fully understand the underlying physics of the harmonic generation starting from a coherent superposition we need a more complete description of the process than the one provided by the dipole projections. It is important to keep track of the fact that only those states that remain populated during the pulse will contribute to the harmonic generation.

It is, after all, a coherent dipole transition between the continuum and the bound state that produces the emission [11]. A bound-state amplitude is therefore a critical element of the phase-locked dipole amplitude needed for coherent harmonic-generation. To see this point more clearly we need to invoke the periodicity of the harmonic-generation process [12,13] as well as the phase of each harmonic (relative to the fundamental) in the dipole acceleration [13–15]. We shall use now the terminology of the semiclassical recollision model, although the analysis of the process will be purely quantum mechanical. We assume that in the tunneling regime a wave packet is produced in the continuum every half a laser cycle. In the conditions we have considered the continuum wave packet is only due to ionization from the excited state. We can characterize each single continuum wave-packet function in the form

$$\Psi_f(r,t)=\Psi_{f0}(r,t)\exp(-iU_e t_i), \tag{8}$$

where Ψ_{f0} is identical for each wave packet created along the whole pulse and there is a phase that keeps track of the phase of the excited state at the time such wave packet was created. Each wave packet is then driven by the laser field and scatters off the core emitting a short burst of radiation. To simplify the description of the tunneling and recollision event, we do not consider here the spreading of the wave packet along one recollision nor the effects due to multiple recollisions with the core by the same wave packet. We shall rather focus on the dynamics of single recollisions along the whole pulse [13]. As we previously saw, the coherent part of the radiation emitted comes from the induced dipoles between the continuum wave packet and each bound state of the initial superposition. We shall analyze each of them separately. In the recombination event, characterized by a time t_r, the induced dipole between the continuum wave packet [Eq. (8)] and the excited state will acquire a global phase factor $\exp[iU_e(t_r-t_i)]=\exp(iU_e t_c)$, where t_c is simply the time between the wave packet is created and recollides, in other words, the recollision period. Equivalently, the induced dipole between the continuum wave packet and the ground state will acquire a global phase $\exp[i(U_g t_r-U_e t_i)]$. In this case, the phase can be factorized as $\exp(iU_e t_c)\exp(i\Omega t_r)$, where $\Omega=U_g-U_e$, that is, a constant phase plus a time-dependent phase factor. Due to the periodic nature of the driving field, the overall process of escape and recollision is repeated every half a cycle. Because successive wave packets are created and therefore recombine at intervals $\tau_l=T/2=\pi/\omega$, each successive burst of radiation due to recombination into the ground state differs from the recombination into the excited state by a phase factor $\exp(i\Omega\tau_l)$. The effect of this phase factor is to shift the harmonic peaks in the second plateau by Ω, and unless Ω corresponds exactly to an odd number of laser photons, they no longer occur at the harmonic frequencies.

III. CONVERSION EFFICIENCIES

For an atom initially in the ground state irradiated by a single laser field, the height of the plateau is found to be proportional to the ionization rate, with a system-dependent proportionality constant [10]. The effect of the depletion of the ground state in the harmonic efficiency has been analyzed in some detail by Lewenstein *et al.* [14], showing how the ground-state depletion modulates the harmonic strength. Although in our case the harmonics in each plateau are due to the same recolliding wave packet, it is only the excited state that becomes depleted. The depletion rate can be simply thought of as $|b_e(t)|^2\sim e^{-\gamma t}$, where γ is the ionization rate. Such an approach allows us to introduce the effect of the depletion on the conversion efficiencies straightforwardly. However, for our choice of initial state, efficiencies clearly depend also on the weights α and β of the initial preparation, as shown in Eqs. (3) and (4). As the power spectrum is proportional to the square of the dipole acceleration, the first plateau (we denote it as S_e) scales as $|\beta|^4$, while the second one (S_g) scales as $|\alpha|^2|\beta|^2$. It is worth noting that since the second plateau scales as $|\alpha|^2|\beta|^2$, the maximum conversion efficiency occurs for $\alpha=\beta=1/\sqrt{2}$. However, even for an equally weighted initial superposition ($\alpha=\beta$), S_g is clearly more intense than the S_e. The reason for this is the distinct strength of the dipole amplitudes $\langle g|\ddot{z}|c\rangle$ and $\langle e|\ddot{z}|c\rangle$ contributing to each plateau. Rather than evaluating these matrix elements, we are interested in their ratio, i.e., $\langle g|\ddot{z}|c\rangle/\langle e|\ddot{z}|c\rangle$. For relatively low-order harmonics we can obtain a reasonable estimate of this quantity using the bare atomic states and Coulomb wave functions for the bound and continuum states, respectively, and assuming that the acceleration can be approximated by $\ddot{z}\simeq-\partial V_c/\partial z$, where V_c denotes the Coulomb potential. To simplify the analysis we further assume both states of the initial superposition ($|g\rangle$, $|e\rangle$) to be in s states, so that the only contribution from the continuum comes from the states with angular momentum $l=1$, i.e., $|c_{k,l=1}\rangle$. One should note that if the energy of the continuum state is large compared to the binding energies of the atomic states, i.e., $k^2/2\gg U_e,U_g$, the continuum states can be approximated by plane waves and the ratio between the matrix elements involved in the coherent superposition behaves then as $|\langle g|\ddot{z}|c_{k,1}\rangle|/|\langle e|\ddot{z}|c_{k,1}\rangle|\rightarrow n^{3/2}$, where n is the principal number of the excited state [16]. This result, however, cannot be used in the present case. This is because when the relevant continuum states have energies close to the threshold, i.e., $k^2/2\leqslant U_e$, and therefore the ratio between matrix elements has to be calculated using exact Coulomb wave functions, it then behaves more closely to $|\langle g|\ddot{z}|c_{k,1}\rangle|/|\langle e|\ddot{z}|c_{k,1}\rangle|\rightarrow n^{5/2}$. It is, of course, in that region that the lower harmonics of each plateau are produced. From the initial-state amplitudes, the distinct dipole strengths and the depletion of the excited state we can derive a ratio for the plateaus' conversion efficiencies

$$\frac{S_g}{S_e}\approx 2\frac{(1-e^{-\gamma T})}{(1-e^{-2\gamma T})}\frac{|\alpha|^2}{|\beta|^2}\left(\frac{U_g}{U_e}\right)^{5/2}, \tag{9}$$

where in the case of small ionization the γ prefactor reduces to 1, whereas for a complete depletion it reduces to a factor 2.

In the Appendix we show that the same result is also obtained in the limit where $k^2/2\gg U_e,U_g$. In such a case we use the strong-field approximation of Ref. [14] to calculate the ratio (9). Both of these results suggest that the relation (9) holds always in practice, in excellent agreement with our numerical observations.

IV. RESULTS

So far we have demonstrated that different plateaus can be generated by a purely quantum interference effect in the recombination process. We shall now focus more specifically on the issue of controlling the radiation output. It is important to keep in mind that the recolliding wave packet determines the overall shape of the harmonic structure and since both plateaus are generated by the same single wave packet they share the same structure. In other words, they both extend up to $3U_p$ presenting the same structure, but the second plateau is shifted by Ω; the energy gap between the states of the initial superposition. As a consequence, both plateaus will overlap only if $3U_p > \Omega$; otherwise the total spectrum will consist of two well-separated sets of harmonic peaks.

To check the dependence on the binding energies as well as on the population weights $|\alpha|^2$ and $|\beta|^2$ of the initial superposition, we solve numerically the time-dependent Schrödinger equation for different cases. For example, the results presented in Fig. 1 are obtained for an equally weighted superposition $|\Psi(r,t=0)\rangle = (|1s\rangle + |2s\rangle)/\sqrt{(2}$ irradiated by a 1 μm wavelength laser at a peak intensity of 1.5×10^{14} W/cm^2. The intensity we use is above the saturation intensity (the excited state is completed depleted before the end of the pulse $\gamma T \approx 1$) for this wavelength and pulse. For this choice of laser parameters the predicted cutoffs for S_e and S_g are approximately at the 62th and 96th harmonic orders, respectively, and $3U_p = 1.98 > U_g - U_e$ $(=1.50)$, so the second plateau S_g overlaps with the first one S_e. Because the initial condition corresponds to an equally weighted superposition, the efficiencies according to Eq. (9) depend only on the ratio between the binding energies $2(U_g/U_{2s})^{5/2} \simeq 32$, in good agreement with the numerical results.

To test the relevance of the dipole strengths we now modify the initial state to be $|\Psi(r,t=0)\rangle = (|1s\rangle + |4s\rangle)/\sqrt{(2}$, where again ground and excited states initially have the same amplitude. The laser field parameters are chosen such that we reach the saturation intensity for the excited state, i.e., $\omega = 0.020$ a.u $(\lambda = 2$ μm) and $I = 8.75\times10^{11}$ W/cm^2. For this case $3U_p = 0.355 \ll \Omega = 1.88$ and the plateaus are therefore clearly separated, as shown in Fig. 2. Given the ratio between the binding energies for this case, we expect a factor of $2\times(U_g/U_{4s})^{5/2} \simeq 2\times(4)^5 \simeq 2\times10^3$ between the conversion efficiencies. The numerical simulations once again show good agreement with this scaling law. As a final case, we repeat the previous calculation but modifying the initial-state amplitudes so $\alpha = 7\beta$. The scaling law of Eq. (9) predicts now a difference in intensity between both plateaus of $2(\alpha/\beta)^2(U_g/U_e)^{5/2} \approx 10^5$. The numerical results are displayed in Fig. 3. It is easier to compare the relative height of the plateaus by using the spectra obtained from the dipole projections P_{pg} and P_{pe} instead of the dipole acceleration itself. In these (Fig. 4) we observe that the harmonic set corresponding to recombination into the excited state is almost entirely hidden by the background of the second plateau and, in fact, the second plateau is approximately 10^5 times more intense than the first one.

The above examples show the versatility of the scheme we propose for controlling the harmonic generation. It is important to point out that the intensities we have used in the simulations correspond to rather weak lasers. For instance, it would not be possible to generate harmonics up the order 100 (with $\lambda = 1$ μm) from the ground state of the ion He^+ (Fig. 1) unless it is irradiated with very high intensities (up to 10^{16} W/cm^2) and even in such a case the effect of the turn on of the pulse will smear out any harmonic structure generated [17]. Although in the examples we have used the excited state shares the same parity of the ground state, this is not an essential condition. The same effects will be present if the excited state has an angular momentum $l \geqslant 0$. The reason for this is that the continuum is highly degenerate in angular momentum states and many transitions from an energy level of the continuum back to states with different angular momentum are therefore allowed.

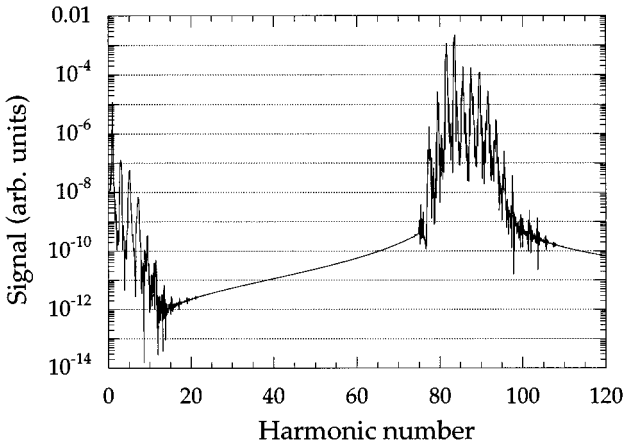

FIG. 2. Harmonic-generation spectra from the He^+ ion at $I = 8.7\times10^{11}$ W/cm^2 and $\omega = 0.02$ a.u. $(\lambda = 2$ μm) starting from a coherent superposition of the ground and the fourth excited state $(4s)$ with equally weighted populations $(1/\sqrt{2})(|g\rangle + |4s\rangle)$. (The pulse lasts 16 cycles and is linearly ramped in the two first cycles.)

V. PRACTICAL REALIZATION

The main difficulty in the experimental realization of the scheme we propose arises from the coherence condition on the initial preparation, that is, the requirement that all atoms must have the same phase in the initial superposition

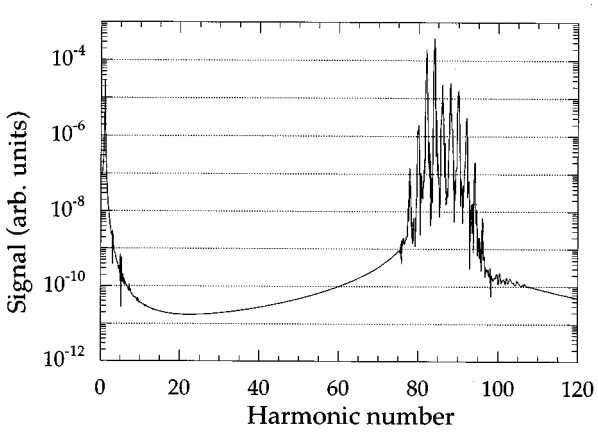

FIG. 3. Same parameters as in Fig. 2, but now with an initial state $\Psi(t=0) = (1/\sqrt{50})(7|g\rangle + |4s\rangle)$, i.e., $\alpha = 7\beta$.

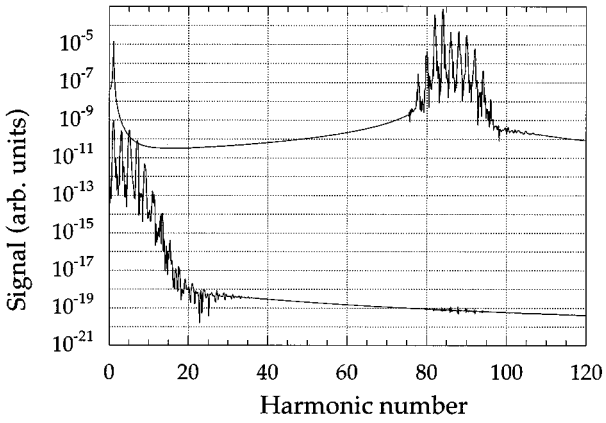

FIG. 4. Conversion efficiency comparison between the plateaus corresponding to the parameters of Fig. 3 by using the dipole projections spectra [see Eqs. (6) and (7)]. The scaling law displayed in Eq. (9) predicts for this case a conversion efficiency factor of 10^5 between both plateaus. The results obtained numerically are in a very good agreement with such scaling law.

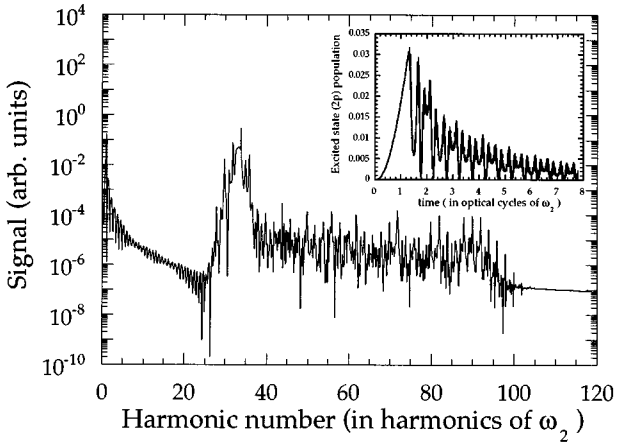

FIG. 5. Inset: Time-dependent population of the ($2p$) excited state. In the simulation the seventh harmonic of a KrF laser ($\omega=0.19$ a.u.) operating at 8×10^{14} W/cm^2 is used to transfer population from the ground state to the $2p$ state (the intensity of the harmonic is kept fixed at 8×10^{10} W/cm^2). Immediately after the coherent superposition has been created a second laser ($\omega_2=0.0420$ a.u., $I_2=1.5\times10^{14}$ W/cm^2) is switched on to ionize the excited ($2p$) state. In the main figure the corresponding harmonic spectrum is shown with a strong peak at the transition energy gap plus a set of harmonics corresponding to recombination back to the ground state.

($|g\rangle+e^{i\phi}|e\rangle$). This is necessary in order to obtain a coherent N^2 enhancement, where N is the number of atoms in the sample. As we showed above, the second plateau is generated because the dipole amplitudes of the transitions corresponding to recombination into the ground state differ by a phase $\exp(i\Omega\tau_l)\exp(i\phi)$ from the dipole amplitudes corresponding to recombination into the excited state. If different atoms have different phases in the initial superposition the interference coming out from the different atoms will substantially reduce any contribution to the second plateau. For the same reason, in order to have a second plateau the precise value of this phase is not important as long as it is shared by all the atoms. Although there are several well-established techniques for preparing coherent superpositions (e.g., adiabatic transfer and $\pi/2$ pulses), they may not be suitable in cases where the energy gap between the two initial states is large, as it is in our case.

Recently, Véniard *et al.* [18] have shown that two-color multiphoton ionization of atoms can be driven using a strong low-frequency laser field along with one of its (weaker) high-order harmonics simultaneously. We can, in principle, apply a similar procedure to prepare the initial coherent state, but using now only a low-intensity harmonic pulse that matches the energy gap between the ground and the desired excited state. In this way we can transfer population from the ground to the excited state with a single-photon Rabi transition. In theory, one could also prepare the initial superposition by means of multiphoton Rabi transitions; however, that should require much higher intensities than in the single-photon Rabi case. Once the initial state has been prepared, a second low-frequency, relatively-low-intensity laser is used to ionize the excited state, hence producing the two set of harmonics.

We should remember that the initial preparation has to be achieved before the population in the excited state starts to ionize. Ions with large binding energies are the best candidates to fulfill these conditions. We have done some numerical simulation of the preparation of the initial superposition with this technique. The following case is just an example. In our simulation we first irradiated He^+ with KrF ($\omega=0.19$ a.u.) at intensities $I=8\times10^{14}$ W/cm^2. At this intensity the depletion of the ground state is negligible and very few harmonics are produced. The seventh harmonic pulse with photons of energy 1.4 a.u (38 eV) is (after the fundamental has been already switched off) used to pump population from the ground to the $2p$ state ($|U_g-U_e|=40.8$ eV; in our numerical results this energy gap happens to be of 38.5 eV), creating, therefore, the coherent superpostion. The intensity of the seventh harmonic pulse is kept fixed at 8×10^{10} W/cm^2, i.e., four orders of magnitude below the laser field intensity, which is a reasonable assumption. Finally, a second very-low-frequency laser ($\omega_2=0.042$, $\lambda=1$ μm) with a moderate intensity ($I_2=1.5\times10^{14}$ W/cm^2) is used to ionize the excited state, producing the two sets of harmonics. The results of the simulation are shown in Fig. 5. The inset shows the time-dependent population of the corresponding excited state. At $t=0$ the atom is initially in the ground state; when the harmonic field is switched on a Rabi flopping between $1s$ and $2p$ takes place. When approximately 3% of the population is in the excited state we switch on the low-frequency field (ω_2, I_2), which ionizes the excited state, producing the two sets of harmonics. The harmonic spectrum shows the main features previously pointed out: a second set of harmonics starting at the energy gap between the excited and the ground state ($\Omega=1.4$ a.u$=33\omega_2$) that extends up to $3.2U_{2p}+U_g$ (around the 91th harmonic). One should note that even for such a small population in the $2p$ state, the interference effects are clearly present.

VI. CONCLUSION

In this paper we have presented some effects related to quantum interferences in strong atom-laser interactions. In

particular we have shown that modifying the ionization and recombination events induces dramatic changes in the harmonic spectrum. For instance, if the initial state is prepared as a coherent superposition of different bound states, the harmonic-generation spectrum is composed by different plateaus with different conversion efficiencies. Our aim has been to control the harmonic spectrum by means of this feature. Although the scheme we present is very simple and has great versatility, we consider it just a first step in the control of harmonic generation. We want to stress here that our method does not allow one to generate shorter harmonic wavelength than the ones obtained directly from the ground state of the atom or ion (the cutoff will always be at U_g+3U_p). It does, however, require much weaker intensities than in the later case, and the signal corresponding to transitions back to the initial state are greatly enhanced compared to the case in which all the population is initially in the ground state. Finally, we have discussed the practical realization of our scheme, showing that by means of a single field and one of its harmonics it is possible to prepare the initial coherent state.

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APPENDIX: OTHER ESTIMATES OF CONVERSION EFFICIENCIES

It is possible to generalize the approach of Ref. [14] (see also [19,20]) to the case when initially the ground and excited states have nonvanishing probability amplitudes α and β and only the excited state is essentially depleted. The generalization is straightforward when there is no coupling of these bound states other than via the continuum. One can then follow the lines of [19] and derive the expression for the relevant parts of the time-dependent dipole moment

$$d_{pe}(t)=i|\beta|^2e^{-\gamma t}\int_0^t dt'\int d^3\vec{p}\,d_e^*[\vec{p}-\vec{A}(t)/c]\times\exp[-iS(\vec{p},t,t')]\mathcal{E}(t')d_e[\vec{p}-\vec{A}(t')/c]+\text{c.c.} \tag{A1}$$

In this expression $d_e[\vec{p}-\vec{A}(t)/c]$ is the z component of the field-free dipole transition matrix element between the excited state and the continuum state characterized by the velocity $\vec{v}=\vec{p}-\vec{A}(t)$, $\vec{p}$ denoting the canonical momentum, $\vec{A}(t)=[0,0,A(t)]$ the vector potential, and $E(t)=-(1/c)\partial A(t)/\partial t$ is the electric field. Finally, $S(\vec{p},t,t')$ is the *quasiclassical action*, describing the motion of an electron moving in the laser field with a constant momentum $\vec{p}$,

$$S(\vec{p},t,t')=\int_{t'}^t dt''\left(\frac{(\vec{p}-\vec{A}(t'')/c)^2}{2}+U_e\right). \tag{A2}$$

Note that damping rate of the part of the dipole moment (A1) is equal to the depletion rate of the population the excited state γ.

Similarly, we obtain

$$d_{pg}(t)=i\alpha^*\beta e^{-i(U_g-U_e)t-\gamma t/2}\int_0^t dt'\int d^3\vec{p}\,d_g^*[\vec{p}-\vec{A}(t)/c]\times\exp[-iS(\vec{p},t,t')]\mathcal{E}_z(t')d_e[\vec{p}-\vec{A}(t')/c]+\text{c.c.} \tag{A3}$$

Here $d_e[\vec{p}-\vec{A}(t)/c]$ is the corresponding z component of the field-free dipole transition matrix element between the ground state and the continuum. Note that this part of the dipole moment is damped as the probability amplitude of the excited state, i.e., with the rate half that of (A1). Note also that above expressions are, strictly speaking, only valid in the tunneling limit, i.e., when $U_p\geqslant U_e,U_g$.

To obtain the harmonic spectrum we have to calculate the Fourier transform of the second time derivatives of the above equation. A good estimate of the fivefold integral over $t,t',\vec{p}$ can be obtained using the method of stationary phase [20]. To do that we replace the integral by a sum of contributions corresponding to stationary points of the Legendre-transformed quasiclassical action

$$S(\vec{p},t,t')-Kt \tag{A4}$$

in the case (A1) and

$$S(\vec{p},t,t')-(K'-U_g+U_e)t \tag{A5}$$

in the case (A3), where K and K' denote the corresponding harmonic orders.

To compare the plateau levels we take $K'=U_g-U_e+K$. That means that the same electron trajectories provide the stationary points of the actions (A4) and (A5). As a result we obtain

$$\frac{S_g}{S_e}\simeq 2\frac{|\alpha|^2}{|\beta|^2}\left|\frac{d_g[\vec{p}-\vec{A}(t)/c]}{d_e[\vec{p}-\vec{A}(t)/c]}\right|^2\left(\frac{K'}{K}\right)^4, \tag{A6}$$

where the field-free dipole moments are calculated at the stationary points, i.e., for the continuum state corresponding to the electron returning to the nucleus at time t with the appropriate velocity $\vec{p}-\vec{A}(t)/c$. The factor 2 in above formula comes, as in Eq. (9), from the fact that $d_{pe}(t)$ is damped twice as fast as $d_{pg}(t)$, whereas the last factor in (A6) is a consequence of the second time derivative.

To proceed further we limit our attention to the case when the kinetic energy of the returning electron $k^2/2=[\vec{p}-\vec{A}(t)/c]^2/2$ is much larger than U_e,U_g. In particular

we consider the harmonics at the end of the plateaus, $K \simeq 3U_p + U_e$ and $K' \simeq U_g + 3U_p$. In such a case the field-free matrix elements can be calculated using the plane waves as the continuum states [16]

$$d_{e,g}(\vec{p}) = i \frac{2^{7/2}(2U_{e,g})^{5/4} p_z}{(\vec{p}^{\,2} + 2U_{e,g})^3}. \tag{A7}$$

From the stationary point equation for the action (A4), $\partial(S - Kt)/\partial t = 0$, we obtain

$$[\vec{p} - \vec{A}(t)]^2 + 2U_e = 2K, \tag{A8}$$

so that

$$\frac{S_g}{S_e} \simeq 2 \frac{|\alpha|^2}{|\beta|^2} \left(\frac{U_g}{U_e} \right)^{5/2} \left(\frac{K}{K'} \right)^2. \tag{A9}$$

Note that in the considered case the two plateaus overlap ($3U_p > U_g - U_e$) and $K \simeq K'$, so that the expression (A9) becomes, in fact,

$$\frac{S_g}{S_e} \simeq 2 \frac{|\alpha|^2}{|\beta|^2} \left(\frac{U_g}{U_e} \right)^{5/2}. \tag{A10}$$

In this way we recover the expression (9) in the limit opposite the one considered in Sec. III. This suggests strongly that the result (A10) is universally valid.

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