Harmonic generation from a coherent superposition of states

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(Received 21 July 1995)

In this Rapid Communication we present a method of extending the harmonic generation plateau to high energies with very high conversion efficiencies using moderate intensities. Preparing the initial state as a coherent superposition of the ground state and an excited state we can induce dipole transitions between the continuum and the ground state via the intermediate (excited) state responsible for the ionization. We solve numerically the Schrödinger equation and show that the spectrum contains two distinct plateaus, with cutoffs at U_e+3U_p and U_g+3U_p. Using an essential states method we demonstrate that the plateaus are due to recombination into the excited and ground states. We briefly discuss possible experimental realizations of the scheme we present.

PACS number(s): 42.50.Hz, 32.80.Rm

It is now well established that an atom driven by a strong laser field radiates high odd harmonics of the incident radiation. The strongly nonlinear response of the atom to the field produces a large conversion efficiency from the fundamental wavelength to the harmonic wavelength up to very high orders. The result is a set of odd harmonics extending to high orders with approximately constant efficiency, followed by a sharp cutoff. This is commonly referred to as the harmonic generation plateau. There is an upper limit to the harmonic plateau determined by the maximum kinetic energy that an electron (that has been previously promoted to the continuum) could have if it is to be driven back to the core by the action of the field. This maximum kinetic energy is approximately $3.2U_p$ ($U_p=E^2/4\omega^2$) and depends only on the intensity and frequency of the incident radiation, since at such intensities the interaction of the electron in the continuum with the external field dominates over the Coulomb interaction. This is the origin of the well known cutoff rule $U_i+3.2U_p$ [1] of the harmonic plateau, where U_i refers to the ionization potential. Harmonics corresponding to higher photon energies are produced with an exponentially diminishing efficiency, thus becoming rapidly indistinguishable from the background.

The two-step quasiclassical model has been very successful in interpreting this plateau [1,2]. In this model the electron escapes into the continuum by a tunneling process. The electron is then driven by the field, and returns to the atomic core emitting a photon in the transition back to the ground state. The efficiency of the harmonic signal is found to be directly proportional to the ionization rate, with a system-dependent proportional constant. Therefore this suggests that at best, for a given ionization potential, the highest harmonic order achievable would be the one produced just below the saturation intensity for each wavelength.

The quadratic scaling of the ponderomotive energy with fundamental laser wavelength also suggests that atomic systems with a large ionization potential driven by long laser wavelengths should generate the most energetic harmonics. In principle, ions can produce higher-energy harmonics due to both their higher ionization potential and the correspondingly high ponderomotive energy at their saturation intensity. However, at long laser wavelengths, harmonic signal for ions has been shown to be, at best, very weak in comparison to the neutral response [3], and short wavelengths are needed to generate harmonics from ions with an efficiency comparable to that of neutral species. In addition, numerical simulations using the full pulse response, rather than the steady-state response, have shown that the harmonic signal in the plateau disappears when ions are driven by long wavelengths [4]. This effect is related to the intensity of the laser changing with time, and will be discussed in more detail in a future paper [5]. Thus, the shortest harmonic wavelengths generated to date are the 141st harmonic (75 Å) of a Nd:YAG (neodymium-doped yttrium aluminum garnet) laser (1.053 μm) using He [6], the 109th harmonic (74 Å) of a Ti:sapphire laser (800 nm) using Ne [7], and the 37th harmonic (67 Å) of a KrF laser (248 nm) using helium [8], with only the final example clearly demonstrating high efficiency harmonic generation from ions.

To date, most of the theoretical and experimental work done on harmonic generation has focused on the response of atoms initially prepared in the ground state. It is, however, well known that quantum interference in atomic systems can lead to new and unexpected effects [10]. The work that involves superposition effects that is most closely related to ours is that of Gauthey *et al.* [9]. There are, however, some distinct features that relate to harmonic generation by superposition states that we want to emphasize. In particular, we want to show that if the initial state is prepared as a coherent superposition of the ground state and an excited state, it is possible to generate very high-order harmonics with high conversion efficiency using moderate laser intensities. By moderate intensities we mean that the intensity is high enough to ionize the excited state, but the field is too weak to directly ionize the ground state. In this case we found that the harmonic spectrum is composed of two distinct sets of harmonics. The first (lowest energy) plateau corresponds to transitions back to the initial state, and as one might expect, has a cutoff at U_e+3U_p. The second plateau begins at an energy U_g-U_e and has a cutoff at energy U_g+3U_p. We shall show that this set of harmonics is generated by transitions from the excited state into the continuum and then back to the ground state. These harmonics cannot be obtained by computing the responses starting from the ground and ex-

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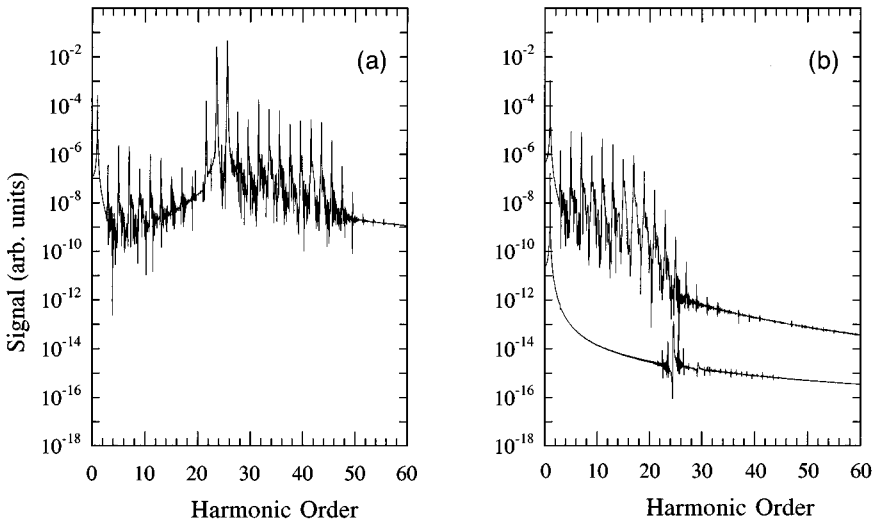

FIG. 1. (a) Harmonic generation spectra from a He^+ ion at $I=8.85\times10^{13}$ W/cm^2 and $\omega=0.06$ a.u. ($\lambda=746$ nm) starting from a coherent superposition of ground and first excited state ($2s$) with equally weighted populations; $1/\sqrt{2}(|g\rangle+|e\rangle)$. (b) Harmonic spectra corresponding to the ground state alone (lower line), and excited state alone (upper line), for the same pulse as in case (a).

cited states separately, and therefore it is essential to have coherence between the ground- and excited-state populations.

Our results are based on numerical solutions of the time-dependent Schrödinger equation for a superposition of ground and excited states of He^+. We use ions to take advantage of their higher ionization energy, but the process we wish to present here is very general, and is not restricted to ions.

To solve the Schrödinger equation we use a partial-wave decomposition of the wave function, together with a split operator and a Crank-Nicholson algorithm as described elsewhere [11]. We have also carried out calculations using a one-dimensional model and have found the same qualitative results. We prepare the initial state as a coherent superposition of the ground state $1s$ ($U_g=54.4$ eV) and the first excited state $2s$ ($U_e=13.6$ eV):

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

where α and β are the amplitudes of the ground and excited states, and $|\alpha|^2+|\beta|^2=1$. We have varied the initial relative phase between the states, ϕ, and found that the results are not modified, so we shall take $\phi=0$ for the remainder of this Rapid Communication. We solve the Schrödinger equation in the length gauge, with the laser field linearly polarized. The interaction term is written as

$$H_{int}=f(r)\vec{r}\cdot\vec{E}\sin(\omega t), \tag{2}$$

where $\vec{E}$ is the electric-field amplitude, ω is the laser frequency, and $f(t)$ is the pulse shape function that is chosen to be a 32-cycle pulse with a linear turn-on of 12 cycles followed by a flat part for the rest of the pulse.

We first present results for a coherent superposition of the He^+ ion ($\alpha=\beta=1/\sqrt{2}$) irradiated by a laser frequency of $\omega=0.060$ a.u. ($\lambda=746$ nm) at an intensity of $I=8.8\times10^{13}$ W/cm^2. For this choice of intensity and wavelength the ionization of the ground state is negligible, and there is only transference of population from the excited state to the continuum. The harmonic spectrum corresponding to this case is shown in Fig. 1(a). The spectrum clearly shows two different sets of harmonics, the second one starting after the two strong peaks corresponding to the transition energy between the $1s$ and $2s$ states. In this case, the energy gap between the ground and excited states does not satisfy a multiphoton resonance condition. As a consequence the two plateaus are not exactly separated by an integer number of photons. To avoid phase-matching problems during the propagation the resonant condition with an integer (odd) number of photons will be necessary. For the sake of comparison we also display in Fig. 1(b) the harmonic spectra corresponding to an initial state of the ground ($\alpha=1$, $\beta=0$) and excited ($\alpha=0$, $\beta=1$) states alone. Figure 2 shows the populations of the ground and excited states together with the total ionization for the case of an equally weighted initial superposition.

We can produce a simple picture of the processes using a simple three-level system. In Fig. 3 we schematically represent our process. Our population is initially a coherent superposition of the ground $|0\rangle$ and excited state $|1\rangle$. The continuum level involved in the generation of the qth harmonic is represented as level $|2\rangle$. We assume that the coupling between the ground state and the continuum (Ω_{02}) is much

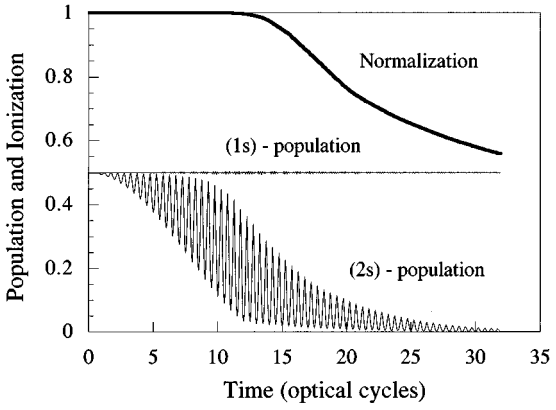

FIG. 2. Ground state, excited-state ($2s$) population, and ionization (normalization) corresponding to an initial coherent superposition of ground and excited state with equally weighted populations [Fig. 1(a)].

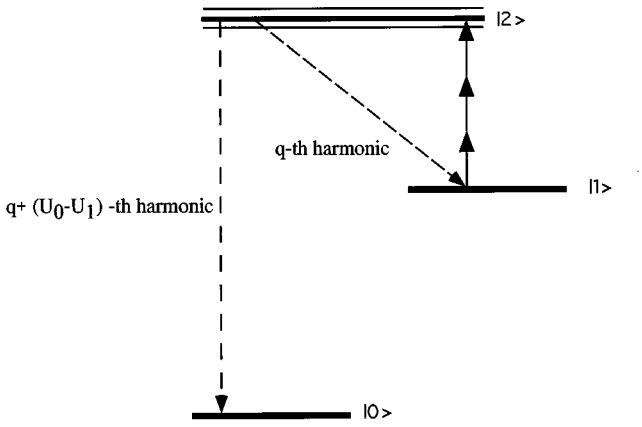

FIG. 3. Three-level Λ configuration representing schematically the emission of the qth harmonic and the $q+(U_g-U_e)$th harmonic. The ground state $|0\rangle$ is not coupled to the excited state $|1\rangle$, and is very weakly coupled to the continuum $|2\rangle$. The excited state $|1\rangle$, responsible for ionization, is strongly coupled to the continuum $|2\rangle$, inducing a dipole moment between the continuum and the ground state. This induced dipole momentum allows transitions from the continuum back to the ground state.

smaller than the coupling between the excited state and the continuum, i.e., $\Omega_{02} \ll \Omega_{12}$, and finally we also assume that the coupling between ground and excited state Ω_{02} is also much smaller than Ω_{12}.

A simple analytical model based on essential states can be applied to explain the main features of both harmonic plateaus. Since the time-dependent Schrödinger equation is linear, we can write its solution as the sum of two separate wave functions:

$$\Psi(r,t) = \psi_g(r,t) + \psi_e(r,t). \tag{3}$$

If we choose the initial conditions such that $\psi_g(r,t=0) = \alpha|g\rangle$ and $\psi_e(r,t=0) = \beta|e\rangle$, then it is clear that the wave function $\Psi(r,t)$ at any subsequent time is equal to the sum of the wave functions we would have obtained starting in the ground and excited states independently multiplied by a numerical factor. To calculate the harmonic generation we look at the dipole acceleration:

$$\begin{aligned}\ddot{d}(t) &= \langle \Psi(r,t)|\hat{\ddot{x}}|\Psi(r,t)\rangle \\ &= \langle \psi_g(r,t)|\hat{\ddot{x}}|\psi_g(r,t)\rangle + \langle \psi_e(r,t)|\hat{\ddot{x}}|\psi_e(r,t)\rangle \\ &\quad + \langle \psi_g(r,t)|\hat{\ddot{x}}|\psi_e(r,t)\rangle + \text{c.c.}\end{aligned} \tag{4}$$

The first two terms in this expression 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. This term is only relevant if we start from a coherent superposition, and is, as we shall demonstrate, responsible for the second plateau. At the intensities we are considering the response starting from the ground state is essentially negligible [as it can be seen in Fig. 1(b)]; therefore, we shall only describe the second and third terms in detail.

In order to simplify our analysis we make the following assumptions: (a) We assume that since the laser intensity is small (relative to the ground-state saturation intensity), the ground-state population is not changed by the field. This is supported by calculations of the response starting from the ground state only for these conditions. (b) We neglect the coupling between the excited state and any other bound states. These approximations are similar to the ones made by Lewenstein *et al.* [12] in explaining the origin of the plateau. We can use them to write the wave functions in the following form:

$$\psi_g(r,t) = \alpha|g\rangle e^{-i\omega_g t}, \tag{5}$$

$$\psi_e(r,t) = \beta\left(a(t)|e\rangle e^{-i\omega_e t} + \int dn b_n(t)|n\rangle e^{-i\omega_n t}\right), \tag{6}$$

where $a(t)$ and $b_n(t)$ are time-dependent complex coefficients, and the integral is over all the continuum states $|n\rangle$. If we neglect continuum-continuum interactions the second term in Eq. (4) is given by

$$\begin{aligned}&\langle \psi_e(r,t)|\hat{\ddot{x}}|\psi_e(r,t)\rangle \\ &\quad = \beta^2 \int dn a^*(t) b_n(t) \langle e|\hat{\ddot{x}}|n\rangle e^{i(\omega_e-\omega_n)t} + \text{c.c.},\end{aligned} \tag{7}$$

and the final terms by

$$\begin{aligned}\langle \psi_g(r,t)|\hat{\ddot{x}}|\psi_e(r,t)\rangle = \alpha\beta\Big(& a(t)\langle g|\hat{\ddot{x}}|e\rangle e^{i(\omega_g-\omega_e)t} \\ & + \int dn b_n(t)\langle g|\hat{\ddot{x}}|n\rangle e^{i(\omega_g-\omega_n)t}\Big) + \text{c.c.}\end{aligned} \tag{8}$$

If we assume that $a(t)$ is a slowly varying function of time, then the time-dependent parts of Eq. (7) and the second term in Eq. (8) differ only by a factor $e^{i(\omega_g-\omega_e)t}$. 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 states. This is the second plateau observed in the harmonic spectra. This simple analysis also demonstrates the dependence of the relative efficiencies of the two plateaus on the weighting of the initial state. Since the power spectrum scales as the square of the dipole acceleration, it is clear from Eqs. (7) and (8) that the first plateau efficiency scales as β^4 while the second scales as $\alpha^2\beta^2$. We demonstrate this in Fig. 4, where we show the harmonic spectra for (a) $\alpha=\sqrt{3}/2$, $\beta=1/2$ (squares), and (b) $\alpha=1/2$, $\beta=\sqrt{3}/2$ (circles), calculated for the same parameters as Fig. 1. As predicted above, the efficiencies for cases (a) and (b) are in the ratio 1:9 for the first plateau and 1:1 for the second plateau. Finally, we would like to stress that in our example the choice of the $2s$ excited state is only due to the fact that this is a metastable state, but in general, both states do not need to have the same parity. The reason for this is that the continuum is highly degenerate in angular momentum and therefore transitions from a energy level of the continuum back to states with different angular momenta are allowed.

In conclusion, we have presented a method of extending the single atom harmonic plateau to higher energies with higher efficiencies than it is possible to obtain starting the

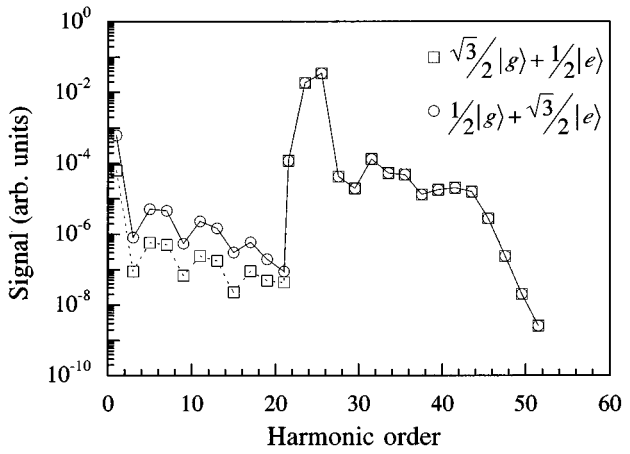

FIG. 4. Relative efficiencies of the two plateaus on the population weight of the initial state: $\alpha|g\rangle+\beta|e\rangle$. The first plateau scales as β^4 and the second one as $\alpha^2\beta^2$. Case (a) $\alpha=(\sqrt{3}/2)$ and $\beta=1/2$ (squares), and case (b) $\alpha=1/2$ and $\beta=\sqrt{3}/2$ (circles).

process from the ground or excited state. If we have a collection of atoms, to obtain a coherent N^2 enhancement, all of the atoms have to be prepared in the same superposition, i.e., with the same initial phase. However, any common phase will be appropriate for the effects to be present. To prepare such an initial superposition may not be an easy task to do if the energy gap between the two initial states is large. This is, of course, a substantial issue for this technique. We do not believe, however, it is an insuperable problem and could be achieved, for example, by resonant multiphoton excitation [13]. The phase of the initial superposition will then depend on the intensity of the laser used, but it will obviously be the same for all the atoms [14]. The best way to optimize this for an experiment will depend on the ions and laser sources available. This is a matter we intend to pursue in the future.

We have explained our results in terms of an essential states method demonstrating the origin of the two sets of harmonics we have observed. Since both cutoffs are separated by an energy corresponding the the ground-excited transition, i.e., (U_g-U_i), this method of generating high-order harmonics is particularly suitable for experiments on ions driven by long wavelengths. We get the advantage of the high ionization energy of (the ground state of) the ion, while still using moderate intensities and avoiding the pulse-shape-related difficulties mentioned previously. Finally, to improve the phase matching between the two harmonic sets it is enough to choose a resonant condition (with an odd number of photons) between the two states of the initial preparation.

This work is financially supported by the UK Engineering and Physical Science Research Council, as well as the Human Capital and Mobility program of the European Community. A.S. also thanks the Fleming/MEC program for financial support and DGICYT (Contract No. PB92-0600).

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