Strong-field multifrequency electromagnetically induced transparency

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Based on *ab initio* simulations of the interaction of the hydrogen atom with a multifrequency ultrashort laser pulse in the strong-field regime, coherent population trapping is found. It leads to the inhibition of the ionization process and, hence, electromagnetically induced transparency, provided that certain two-photon resonance conditions are satisfied. This effect opens additional possibilities to detect phase-locked trains of attosecond pulses.

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I. INTRODUCTION

Coherent population trapping (CPT) is a nonlinear optical effect which has been widely studied during recent decades [1]. The first observations of CPT [2] were made using a multimode laser. Similar techniques have given rise to a lot of interesting phenomena such as electromagnetically induced transparency (EIT) [3,4], lasing without inversion [5], and ultraslow light propagation [6]. An explanation of atomic stabilization in Rydberg atoms using coherent trapping effects has also been given [7].

Population trapping has already been reported in hydrogen [8,9] and other atomic gases such as helium [10] with two different moderately intense (tens of $\mathrm{MW/cm^2}$) laser sources and long pulses (in the nanosecond regime). Here we report the appearance of a coherent strong-field process leading to coherent population trapping due to the destructive interference of competing ionization channels when an atom interacts with an intense (irradiances above 10^{12} $\mathrm{W/cm^2}$), ultrashort (subpicosecond) laser pulse composed of several harmonic frequencies. The atomic electron is thus placed in a superposition state that is difficult to ionize and the system becomes more transparent if the phases of different harmonics are locked. The use of ultrashort pulses implies broadband fields. As a consequence, trapping cannot be exact and permanent as it is with three-level systems. However, it is clearly present.

The superposition of harmonic frequencies appears naturally in high-order harmonic generation processes when a strong laser field interacts with matter. The radiation is generated in most of the cases as a long plateau with many atomic components [11,12]. When these harmonics are phase locked, a train of attosecond pulses is created [13–15].

As opposed to the measure of the intensities of the different harmonics, the detection of their respective phases is not straightforward. Several nonlinear techniques have been proposed, most of them based on the ionization dynamical effects induced by the multiharmonic field [16–18]. The effect we have found can be an alternative technique to detect whether the phases are locked or not. It has, of course, several additional difficulties, but it opens the way to a signal to noise ratio much higher than other techniques due to the intrinsic characteristics of CPT.

There are other phenomena related to a suppression of the ionization in the strong-field regime, such as the widely discussed atomic stabilization [19]. Whether it can be achieved or not, atomic stabilization is radically different from the coherent effects we report in this paper because it happens in the tunnel or in the over-the-barrier ionization regime instead of in the multiphoton regime and, even more important, stabilization does not critically depend on the frequencies and phases of the laser pulses.

II. ATOMIC MODEL

Let us start by describing our quantum system, the atom. The time-dependent three-dimensional Schrödinger equation (TDSE) in the dipole approximation for a hydrogen atom reads

$$i\hbar\frac{\partial}{\partial t}\Psi(\vec{r},t)=\left(\frac{1}{2m}\vec{p}^2-\frac{e^2}{r}-ezE(t)\right)\Psi(\vec{r},t). \tag{1}$$

We assume that the laser field $E(t)$ is linearly polarized along the z axis and it is a sum of ten odd harmonics of the fundamental frequency ω_L,

$$E(t)=\frac{1}{2}\sum_{q=1}^{q=10}E_q(e^{i((2q-1)\omega_L t+\varphi_q)}+\text{c.c.}). \tag{2}$$

The pulse envelope has a flat shape during 20 cycles of ω_L. We choose the partial amplitudes E_q to be equal, i.e., $E_q=E_1$ for all values of q. This is a realistic assumption for fields obtained by high-order harmonic generation in gases, where there exists a long plateau of harmonic components with roughly the same intensities corresponding to the central part of the emitted spectrum. In our calculations we will take a value for $E_1=E_L/\sqrt{10}$ such that the average amplitude of the total field is $E_L=\sqrt{\Sigma E_q^2}=0.04$ a.u., which corresponds to an average irradiance of 5.6×10^{13} $\mathrm{W/cm^2}$.

In the phase-locked case (say $\varphi_q=0$ for simplicity) the field is a train of attosecond pulses, while for arbitrary phases such high-field spikes do not appear. It is thus reasonable to expect a different response in the phase-locked case because of the high field existing at times of global constructive interference.

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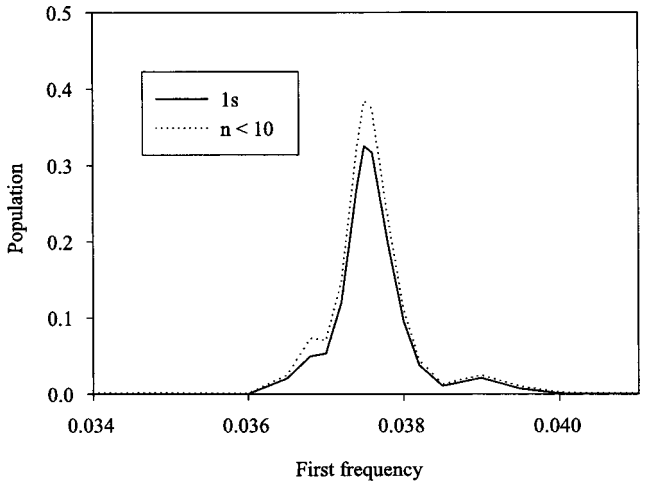

FIG. 1. Ground-state population (full line) and population in the atomic states with $n_r \leqslant 10$ (dotted line) at $t=40\pi\omega_L$ (i.e., 20 cycles of the ω_L frequency).

The numerical solution of the TDSE in the dipole approximation for the hydrogen atom submitted to a linearly polarized field is nowadays a routine task. We expanded the wave function in the spherical harmonics basis and solved the resulting set of coupled differential equations by means of a Crank-Nicholson algorithm [20]. We used a radial grid of 5000 points separated by a space step of 0.1 a.u., giving a maximum radius $r_{\max}=500$ a.u. (26.5 nm). The maximum angular number considered in the expansion is $l_{\max}=72$. The time step used is $1/200$ of the fundamental frequency cycle and the total simulation time is 20 cycles of that frequency.

III. POPULATION TRAPPING

We simulated the response of the atom to such multifrequency laser fields. In order to point out the basic mechanisms leading to population trapping we have chosen laser frequencies between 0.034 and 0.041 a.u. (from 1343 to 1114 nm wavelengths). The results of these simulations are shown in Fig. 1, where we have depicted the population that remains in the ground state after 20 cycles for the different values of frequencies. We show also the population remaining in the bound states up to $n_r=10$, which can be considered the total bound population after that time.

Figure 1 shows a clear maximum close to $\omega_L=0.0375$ a.u. Therefore, for this frequency a mechanism for inhibition of ionization appears in the phase-locked case. To understand this, it is necessary to observe that $\omega_L=0.0375$ a.u. corresponds to a ten-photon $1s$-$2s$ interference. In fact, this is a two-photon resonance mediated by an $l=1$ state (resonant or not). Since it may lead to atomic wave function interference, the CPT, we refer to this frequency as the $1s$-$2s$ interference (of course, for atomic hydrogen the $1s$-$2s$ energy difference is 0.375 a.u.). Let us recall that we are dealing with multifrequency fields. Absorption of an $11\omega_L$ photon from the $1s$ state is resonant with the absorption of a $1\omega_L$ photon from the $2s$ state for $\omega_L=0.0375$ a.u. Simultaneously, using $13\omega_L$ and $3\omega_L$ photons, the same two-photon resonance between these states is achieved. This also happens for the couple $15\omega_L$ and $5\omega_L$ and so on. Hence, there are several paths to get the $1s$-$2s$ two-photon resonance (some of them with a resonant intermediate state and some of them without it). Each one of these paths is a typical three-level scheme leading to CPT.

In principle, all these resonances are not coupled. However, as we can see, when the harmonics are phase locked all these two-photon resonances behave in the same way, enhancing the trapping effect. For random phases the resonances interfere among them in a destructive way, and there is no longer population trapping.

IV. RESONANCES AND INTERFERENCES

To explain the inhibition of ionization at certain frequencies, we must take into account the interactions of the different harmonic components, which can give rise to interference phenomena, coupling states of the same parity via continuum states to form some kind of dark states which trap part of the population and, as a final result, yield ionization much lower than expected.

The interference at $\omega_L=0.0375$ a.u. occurs obviously between two resonances. For $\omega_L=0.0341$ a.u. one has ionization with one intermediate $11\omega_L$ resonance for the $1s$-$2p$ transition. Analogously, for $\omega_L=0.0417$ a.u., there is a $9\omega_L$ resonance for the $1s$-$2p$ transition. In both cases, np states with n higher than 2 are nonresonant, in this frequency interval.

Resonances appear also for monochromatic fields, but in our case, the coupling of the transition probabilities associated with the different paths can enhance the ionization yield provided this coupling is constructive, which happens when the different harmonics are phase locked. As a consequence, the ionization will happen faster when all harmonics are phase locked than when their phases are random, as stated in [16]. This can be observed in Fig. 2, where we represent the evolution of the ground-state population and the total bound population ($n_r \leqslant 10$) for a phase-locked pulse with $\omega_L=0.042$ a.u. (wavelength 1141 nm) and the result of averaging 20 different sets of random phase pulses with the same frequencies. Anyway, the final population after 10 cycles is not noticeably different in either case.

Interferences consist of couplings of two states of the same angular momentum via an upper level, usually in the continuum, which is resonant with both of them. These interactions are strong when the two coupled states are separated by an energy equivalent to an even number of photons, n, because in this way there will always exist a common resonant upper level which is reachable from the lower one with m photons, m being odd—a transition allowed in the dipole approximation—and from the upper one with $m-n$, also odd and thus allowed.

This kind of resonant interference phenomenon generates population trapping in a state which is a linear combination of the original two, but it is very difficult to observe with monochromatic fields unless the energy difference between the two coupled states is negligible as compared with the photon energy (degenerate levels). Otherwise we need ex-

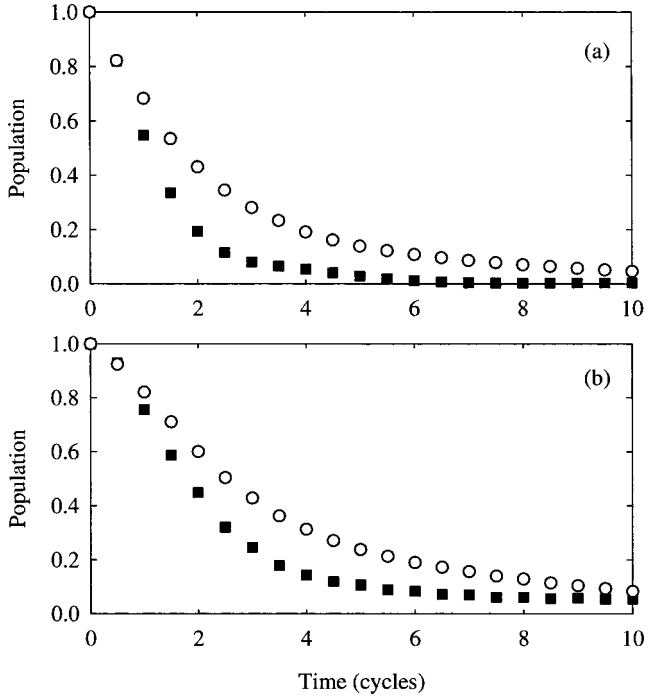

FIG. 2. Time evolution of the ground state (a) and total bound population (b) for $\omega_L = 0.042$ a.u. in the phase-locked (black squares) and random phase (white circles) cases. For this frequency, phase locking is not fundamental in the ionization dynamics.

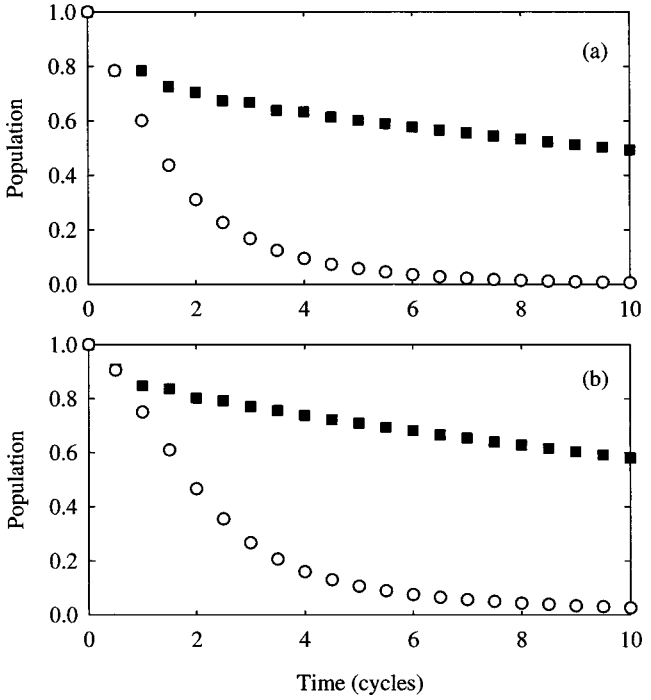

FIG. 3. Time evolution of the ground state (a) and total bound population for $\omega_L = 0.0375$ a.u. in the phase-locked (black squares) and random phase (white circles) cases. For this frequency, the influence of phase locking is fundamental: it prevents ionization due to a CPT process.

tremely intense fields for which multiphoton transitions occur with similar probability as one-photon transitions or, alternatively, two- or many-color fields as in pump-probe experiments. In our case, the field is naturally many color and the different frequencies are harmonics of a fundamental one, so that the even-resonant condition perfectly satisfies our needs.

The interference that appears at frequency $\omega_L = 0.0375$ a.u. (1217 nm) is very clear since it represents an island of anomalously low ionization. For this frequency, the levels involved in the coupling are the ground state, 1*s*, and the first excited state, 2*s*, separated by a distance of ten fundamental photons, plus resonant continuum states which are 15, 17, or 19 photons above the ground state. Hence, there is not only one but three processes which can generate such interference: the first one implies a continuum level with energy 0.0625 a.u. coupled to the ground state through the $15\omega_L$ component of the field and to the 2*s* state through the $5\omega_L$ component; the second one implies a continuum level with energy 0.1375 a.u. coupled to the ground state through the $17\omega_L$ component of the field and to the 2*s* state through the $7\omega_L$ component; the last one couples the 1*s* and 2*s* states to a continuum state with energy 0.2125 a.u. through the $19\omega_L$ and $9\omega_L$ components, respectively.

These three different couplings can add coherently or incoherently depending on their relative phases so that when the phases of all the harmonics are phase locked, they will combine constructively and the result will be enhanced. On the contrary, when the phases are random, they will combine destructively and the effect will disappear. We can observe this behavior in Fig. 3, where we compare the results obtained with phase-locked and random phase pulses when $\omega_L = 0.0375$ a.u. The result is more impressive than in the case of the resonance commented on above because the bound population after ten cycles is close to 60% of the total in the case of the phase-locked pulse, whereas in the random case it is around 2.5%. Eventually, all the population will be ionized also in the phase-locked case due to transitions to different hydrogen levels, but it can take much more time than expected.

This behavior is in principle counterintuitive since one would tend to think that the phase-locked pulse is more suitable for ionizing the atom due to its sharp and intense peaks. It is clear, however, that this is not a problem linked to peak power but to the different phases: it is purely a coherence problem. As strange as this result may seem, it can be used as a test of the phase locking of fields composed of several harmonic frequencies.

V. ESSENTIAL-STATES MODEL

In order to get a better understanding about the coherent effects which prevent ionization, we can study a simple four-level model that, although not completely realistic, contains the basic elements we need for our purpose.

Following Fig. 4, the two lowest levels in our model represent two bound states whose energy difference is equal to an even number of photons of a given frequency, $\varepsilon_2 - \varepsilon_1 = 2n\hbar\omega_L$. Above them, there are two upper levels, which will represent two continuum states of the hydrogen atom. Level

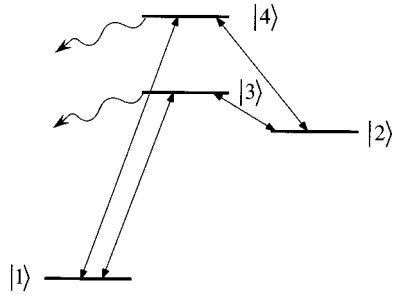

FIG. 4. Schematic representation of the model atom.

3 is considered resonant with both bound levels, with photon energies $\varepsilon_3-\varepsilon_1=(2n+2m-1)\hbar\omega_L$, $\varepsilon_3-\varepsilon_2=(2m-1)\hbar\omega_L$, and the same happens for level 4, with energies $\varepsilon_4-\varepsilon_1=(2n+2m+1)\hbar\omega_L$, $\varepsilon_4-\varepsilon_2=(2m+1)\hbar\omega_L$. These perfect resonance conditions among upper and lower levels are not strictly necessary, and the existence of detunings would not affect the final result, but for simplicity we keep it this way, which is not too bad when ε_3 and ε_4 lie in the continuum.

We also assume that the coupling with the states in the continuum is independent of their energy, i.e., $d_{13}=d_{14}$ and $d_{23}=d_{24}$. The upper levels have also decay rates γ_3, γ_4 which represent the loss of population going out of the system into the continuum and never coming back. In this sense, this is an open model and not a closed one.

After eliminating all counter-rotating components, our multifrequency laser field will only contain the four resonant components which couple each pair of levels:

$$E=\frac{1}{2}\{E_{2m-1}+E_{2m+1}+E_{2n+2m-1}+E_{2n+2m+1}\}+\text{c.c.} \quad (3)$$

with $E_q=E_0e^{iq\hbar\omega_Lt}e^{i\phi_q}$. The Rabi frequencies associated with each transition are $\Omega_{jk}e^{i\phi_q}=E_0e^{i\phi_q}d_{jk}/\hbar$. Under these conditions, neglecting the effect of all nonresonant interactions, the Hamiltonian for the four-level system is

$$H=-\frac{1}{2}\begin{pmatrix} 0 & 0 & \Omega_{13}e^{i\phi_{2n+2m-1}} & \Omega_{14}e^{i\phi_{2n+2m+1}} \\ 0 & 0 & \Omega_{23}e^{i\phi_{2m-1}} & \Omega_{24}e^{i\phi_{2m+1}} \\ \Omega_{13}e^{-i\phi_{2n+2m-1}} & \Omega_{23}e^{-i\phi_{2m-1}} & i\gamma_3/2 & 0 \\ \Omega_{14}e^{-i\phi_{2n+2m+1}} & \Omega_{24}e^{-i\phi_{2m+1}} & 0 & i\gamma_4/2 \end{pmatrix}, \quad (4)$$

which is a straightforward extension of the well-known case of a three-level system with two resonant frequencies [4].

As can be easily verified, the condition to obtain a dark state is $\Omega_{13}/\Omega_{23}e^{i(\phi_{2n+2m-1}-\phi_{2m-1})}=\Omega_{14}/\Omega_{24}e^{i(\phi_{2n+2m+1}-\phi_{2m+1})}$, and this happens only when $e^{i(\phi_{2n+2m-1}-\phi_{2m-1})}=e^{i(\phi_{2n+2m+1}-\phi_{2m+1})}$. Hence, when the fields are phase locked (at least by pairs of components), we have a dark state

$$|d\rangle=\frac{\Omega_{23}e^{-i\phi_{2m-1}}|1\rangle-\Omega_{13}e^{-i\phi_{2n+2m-1}}|2\rangle}{\sqrt{|\Omega_{13}|^2+|\Omega_{23}|^2}}=\frac{\Omega_{24}|1\rangle e^{-i\phi_{2m+1}}-\Omega_{14}e^{-i\phi_{2n+2m+1}}|2\rangle}{\sqrt{|\Omega_{14}|^2+|\Omega_{24}|^2}}. \quad (5)$$

If the phases of the different harmonics are random, there will not be a dark state and the atom will be rapidly ionized.

When we consider more than two pairs of resonant frequencies, as we have in the hydrogen simulation, the previous arguments can be extrapolated considering more than two upper levels, and the phase-locking condition is needed for all the pairs in order to obtain a trapped state.

Undoubtedly, the four-level model is an oversimplification of the real hydrogen atom, but this simple model gives us a nice explanation of why the ionization in the case of phase-locked harmonics is much slower than when the harmonics are absolutely random, and it constitutes the physical mechanism responsible for the reduction of ionization at certain frequencies shown in the much more realistic case of Fig. 1.

The four-level model shows the basic process implied. The 1-3-2 system allows a dark state, while the 1-4-2 system allows another dark state. Whether these two dark states are the same or not depends crucially on the harmonic phases. In the phase-locked case both dark states coincide and thus CPT is preserved for a multiharmonic field. This can be easily extrapolated to the case 1-ε-2, $|\varepsilon\rangle$ being an upper level such that the two-photon $|1\rangle$-$|2\rangle$ transition is resonant.

VI. DISCUSSION AND CONCLUSIONS

Regarding the experimental feasibility of this kind of strong-field CPT, the intensities of the harmonics are not a major problem since energies of the order of a microjoule can be reached now for high harmonics of the Ti:sapphire laser in several laboratories around the world [21,22], although getting the same conversion efficiency for several harmonics may be difficult. It is also clear that, so far, there do not exist ultrashort intense fields for the wavelengths we have used in our simulation. In addition, the hydrogen atom is not the most suitable system to start with in a new experimental technique. However, a rescaling to available laser sources and appropriate atomic or molecular systems is straightforward since the mechanisms involved have a general nature. For instance, atomic argon, which is a gas widely used in strong-field experiments, has several transitions close

to 80 and 100 nm, corresponding to eight- and ten-photon processes for the frequency of a Ti:sapphire laser.

In conclusion, based on a time-dependent numerical simulation and on a naive essential-states model, we have shown that strong-field multifrequency CPT in atomic systems is possible for ultrashort, multiharmonic laser pulses. We suggest that this kind of ultrafast CPT is a physical effect related to phase locking of the different harmonics. This insight into coherent processes in strong-field physics may open additional ways to measure phases of different harmonics in high-order harmonic generation processes. Moreover, the interference effect presented here opens the road to conceptually different ways of partial EIT in the propagation of strong ultrashort multifrequency laser pulses.

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