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High-intensity two-color interactions in the tunneling and stabilization regimes

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Two-color excitation of a hydrogen atom has been studied by solving the time-dependent Schrödinger equation. We have chosen the two laser frequencies to be the fundamental and its third harmonic with an adjustable phase between the two laser fields. This has been proposed and successfully implemented in the past for "coherent control" of atomic and chemical processes. Here we have extended the study to the high-intensity domain and well into the tunneling and the stabilization regimes. We show that in the tunneling regime the two-color excitation can enhance the intensity of harmonics by more than two orders of magnitude compared to a single color at the same effective intensity. In the stabilization regime we confirm that two-color excitation can be used to generate higher harmonics.

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The dynamics of an atom driven simultaneously by two frequencies displays important novel features that cannot be seen with single-frequency driving [1–3]. As an example, an enhancement of high-order harmonic generation has been demonstrated in the irradiation of rare-gas atoms with a combination of the fundamental and third harmonic of a laser field [4]. Also, early work on two-color experiments using the fundamental and the second harmonic of a laser field showed that the relative phase between two fields can have a significant influence on the ionization yield, the photoelectron spectra, and the associated angular distributions [5]. The effects associated with the relative phase between two fields should be more evident for the cases in which the electron wave packet is strongly driven by the laser field. One should expect, therefore, to observe larger effects in the tunneling regime and in the stabilization regime. With all these premises in mind, we certainly would like to know not only how two-color excitation of neutral atoms boosts the overall harmonic generation but whether one can drive the higher-order harmonics more efficiently.

In this paper we shall address these questions by calculating the harmonic generation from a hydrogen atom interacting with a two-color laser field. The two laser frequencies are chosen to be the fundamental and its third harmonic at the same intensity and with a relative phase between them. Our goal has been to study the effects of the two colors and the influence of the relative phase in the tunneling and the stabilization regimes. Our results confirm some of the previous findings using one-dimensional models [6], and show how, in the tunneling regime, the harmonics can be enhanced by more than three orders of magnitude compared to a single color at the same field strength. In the stabilization regime the harmonic spectra generated can be extended to higher orders than those achievable using one-color excitation. Our study is based on numerical solutions of the time-dependent Schrödinger equation, which we solve using a partial-wave decomposition of the electron's wave function, together with a split operator and Crank-Nicholson algorithm, as described elsewhere [7–10]. Application of this method to two-color excitation is straightforward as long as both fields have the same linear polarization. We choose to work in the length gauge, where the interaction term can be written in the form

$$H_{in}=\vec{r}\cdot[\vec{E}_{\omega}\sin(\omega t)+\vec{E}_{3\omega}\sin(3\omega t+\phi)]f(t), \qquad (1)$$

where $\vec{E}_{\omega}$ and $\vec{E}_{3\omega}$ are the fundamental and third harmonic field amplitudes, ω is the fundamental frequency, and ϕ the relative phase between the two fields. The function $f(t)$ is the pulse shape function, which is chosen to be $f(t)=\sin^2(\pi t/\tau)$, where τ is the pulse length; here we consider a pulse length of 32 optical cycles of the fundamental frequency. To produce the maximum enhancement of the harmonic conversion efficiency we choose the same field amplitude for both fields, i.e., $E_{\omega}=E_{3\omega}$. With these conditions, the time varying electric field has a maximum for a relative phase difference between the two fields of $\phi=\pi$, when the maximum field strength of the combined fields reaches the value $E=2E_{\omega}$ twice each cycle. For a phase difference of $\phi=\pi/2$ the maximum amplitude achieved is $E=1.8E_{\omega}$, and finally for $\phi=0$ the maximum field amplitude is $E=1.6E_{\omega}$.

The discussion of the results will be separated into two main parts, one devoted to the tunneling regime and the other to the stabilization regime. Each part will be preceded by a brief introduction of its associated dynamics.

The tunneling regime is conventionally characterized by the Keldysh parameter being smaller than unity, i.e., $\sqrt{U_i/2U_p}<1$ (here U_i refers to the ionization potential and $U_p=E^2/4\omega^2$ is the ponderomotive potential). Physically this means that the laser field can be considered as a quasistatic field (high-intensity, low-frequency) that lowers the atomic potential so that the electron is able to tunnel out. The subsequent evolution in the continuum of the wave packet that has tunneled out can then be described almost "classically" since the interaction with the electric field would dominate and the influence of the Coulomb potential could therefore be neglected. Harmonic generation is understood in this context as due to the rescattering of the electron being driven back by the field to the nucleus and emitting a photon. This view is supported, among other facts, by the success of the classical model in explaining the origin of the harmonic cut-

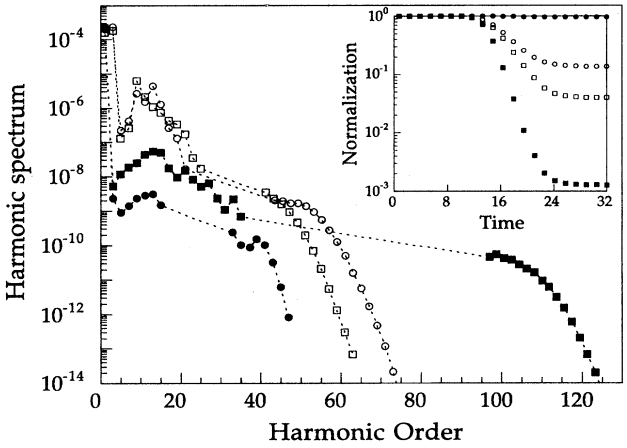

FIG. 1. Comparisons between one- (filled symbols) and two-color (open symbols) harmonic spectra in hydrogen in the tunneling regime. Filled circles correspond to $E_\omega=0.05$ a.u.; filled squares to $E_\omega=0.10$ a.u. Two colors $E_\omega=E_{3\omega}=0.05$ a.u. and $\phi=0$ (open circles), and $\phi=\pi/2$ (open squares). The inset window shows the corresponding ionization yields.

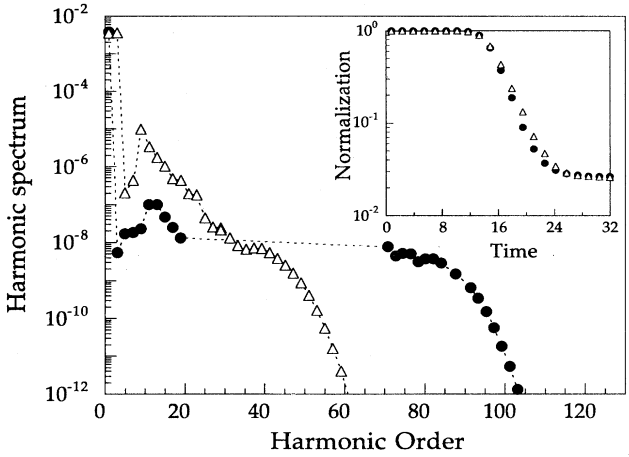

FIG. 2. Comparisons between one- (filled symbols) and two-color (open symbols) harmonic spectra in hydrogen ($\omega=0.042$ a.u.). Filled circles correspond to $E_\omega=0.09$ a.u. Open triangles correspond to two colors $E_\omega=E_{3\omega}=0.05$ a.u. and $\phi=\pi$ (open triangles). The inset shows the corresponding ionization yields.

off [11,12]. In the two-color tunneling regime we expect the ionization yield to be dependent not only on the maximum field strength of the combined fields but also on the relative phase ϕ, since the potential barrier is significantly modified by this relative phase. In order to study the effect of the two-color interference, we calculate ionization yields and the harmonic spectra for one single color at different intensities and compare them with the two-color results.

We first present results for a hydrogen atom irradiated by the combination of the Nd:YAG (neodymium-doped yttrium aluminum garnet) laser frequency ($\lambda=1064$ nm) and its third harmonic ($\lambda_3=354$ nm) at $E_\omega=E_{3\omega}=0.05$ a.u. ($I=8.3\times10^{13}$ W/cm^2). For the sake of comparison we also include the ionization produced by the fundamental laser alone at E_ω and at twice the electric field amplitude $E=2E_\omega$ ($I=3.5\times10^{14}$ W/cm^2). In the latter case we have already surpassed the saturation intensity for this frequency and pulse shape. [The saturation intensity is conventionally understood as the intensity (for a given wavelength and pulse shape) at which a sample of atoms is mostly ionized.] Since surpassing the saturation intensity diminishes the efficiency of generating harmonics, one should be careful in comparing the effects of one and two colors at these high intensities. In Fig. 1 we show the harmonic spectra in the tunneling regime for one- and two-color cases (the corresponding ionization yields are shown in the inset). All the spectra have been normalized to the first harmonic so that a better comparison of conversion efficiencies can be made (since the first harmonic is proportional to the term $E\sin\omega t$). For clarity, only the harmonic peak intensities at the frequencies encountered are plotted. We use a dotted line to join the first harmonics with the cutoff region where the harmonics are clearly produced again [13].

The two-color harmonic spectra show formally the same pattern as their one-color partner: a plateau of harmonics and an abrupt cutoff, where the intensity of the harmonics drops off rapidly. However, a closer inspection reveals significant differences. First of all, the plateau structure extends to higher orders than the fundamental alone at $E=E_\omega$, although the overall plateau structure appears less clearly defined. The most striking difference, however, is the different conversion efficiency in both cases. Generally speaking, the conversion efficiency is much better for the two-color case, regardless of the relative phase. This is especially true for low-energy harmonics, those with energies below U_i+U_p, which are enhanced by almost four orders of magnitude compared to the one-color case at $E=E_\omega$. Furthermore, if we compare conversion efficiencies between two colors ($E_\omega=E_{3\omega}$) and the fundamental alone at $E=2E_\omega$, we still find harmonics with intensities approximately two orders of magnitude larger in the bichromatic case. Since the residual ionization for two colors is smaller than the one produced by the strong field ($E=2E_\omega$), as is clearly shown in Fig. 1, the conversion efficiency increase seems to be partially related to the interference between the two colors. To reinforce this idea we compare the harmonics generated by one color and two colors, when both cases produce the same ionization yield. This is accomplished, for example, with a single laser operating at $E_\omega=0.09$ a.u. ($I=2.8\times10^{14}$ W/cm^2) and the two-color lasers at $E_\omega=E_{3\omega}=0.05$ a.u. and $\phi=\pi$. For both cases the residual ionization at the end of the pulse is practically the same but the enhancement of the harmonics is drastically different, as can be seen in Fig. 2. These results suggest that the enhancement of the harmonic generation is produced by the interference between both colors, and it is independent, to some extent, of the amount of ionization. Two different reasons could, *a priori*, cause this enhancement. The first one is related to the transverse spreading of the wave packet on its way back to the nucleus [14]. The spreading of the free wave packet is intensity independent but wavelength dependent. The longer the wave packet takes to return to the nucleus the larger the spreading and therefore the weaker the interaction with the nucleus. However, the second color impinges fast oscillations on the quiver motion of the free electron that oscillates at ω, slowing down the velocity of the electron. Since the spreading of the wave

packet is proportional to time, this enhancement seems not to be related to this. A semiclassical reason could perhaps account for the enhancement. We know that harmonics are produced in transitions involving the ground state [15], and that implies that harmonics should be produced dominantly by the part of the wave packet closest to the nucleus. Diminishing the velocity of the particle when it "crosses" the nucleus increases the interaction time and therefore the cross section of scattering. The higher-order harmonics, which correspond to electrons returning with high velocities, would be less affected by the retardation produced by the second color and the enhancement would become smaller.

The cutoff in the harmonics can be understood by applying the classical two-step model to the two-color case. From this classical model the predicted cutoff energies agree reasonably well with the quantum results: $4.8U_p$ for $\phi=0$, $5.1U_p$ for $\phi=\pi/2$, and 4.1 for $\phi=\pi$ (U_p refers to the ponderomotive potential of the fundamental). The classical model also predicts a uniform enhancement in conversion efficiency along the whole range of allowed returning kinetic energies. However, this enhancement comes from the fact that the instantaneous electric field strength of the combined fields increases. This dependence on the instantaneous electric field arises from the definition of the classical collision probability [16]. We define the collision probability as the allowed returning trajectories weighted by the corresponding dc ionization rate, $\Gamma(t)\propto[4U_i/\xi(t)]e^{-2/3\xi(t)}$, corresponding to the instantaneous electric field, $\xi(t)$, at which the electron escapes through or over the barrier [17].

We have repeated our calculations for higher field intensities, and the results are in agreement with what we have already presented. However, the improvement in the conversion efficiency is obviously reduced if we are above the saturation intensity or over barrier ionization. Finally, for lower ratios between the two fields $E_\omega/E_{3\omega}>1$ the enhancement in the harmonics is also smaller.

Next, we analyze the effect of the bichromatic field in the adiabatic stabilization regime. By stabilization regime we mean [18,19] that the ionization is suppressed for increasing laser intensities; in other words the ionization rate becomes a decreasing function of the laser intensity. The presence of a two-color laser field does not modify significantly the range of intensities at which stabilization appears, although it modifies the residual ionization. In principle one should expect larger stabilization in a two-color excitation because of the larger time-dependent field strength compared to one-color excitation. Nevertheless, the survival of the atom to the turn-on is far more important than the peak laser intensity itself in the stabilization regime. This survival condition is usually achieved in numerical simulations by using a very short and fast turn-on of the radiation, which is then held constant in amplitude. For the cases presented here, however, we study the full pulse response (32 $\sin^2$ pulse profile) with a laser peak amplitude of $E_\omega=18.4$ a.u. ($I=1.2\times10^{19}$ W/cm^2) and frequency $\omega=1$ a.u. The residual ionization at the end of the pulse is around 40%. (The temporal step necessary to obtain numerical convergence for such superstrong fields is around 2^{14} steps per optical cycle.) Calculations for lower field intensities ($E<E_\omega$) but otherwise identical parameters (frequency and pulse shape) result in larger residual ionizations, so we are indeed in the stabilization regime. Another manifestation of the stabilization in these superstrong fields is the localization of the electron wave packet in space [19,20]. The electron density probability tends to be a peaked structure (two or more peaks) along the polarization axis that simply moves back and forth following the laser oscillations without spreading in time [20]. The time-averaged electron density occupies a range of roughly $2\alpha_0=2E/\omega^2$ with a central peak at the origin and some weaker peaks around α_0. The structure of the stabilized wave packet depends on the pulse shape that creates it [21], and therefore should also depend on the phase difference between the two fields for the bichromatic case. In Fig. 3 we show the harmonic spectra corresponding to (a) one color at $E_\omega=18.4$ a.u, (b) two colors at $E_\omega=E_{3\omega}=18.4$ and $\phi=0$, and (c) two colors at $E_\omega=E_{3\omega}=18.4$ and $\phi=\pi$. We observe that very few har-

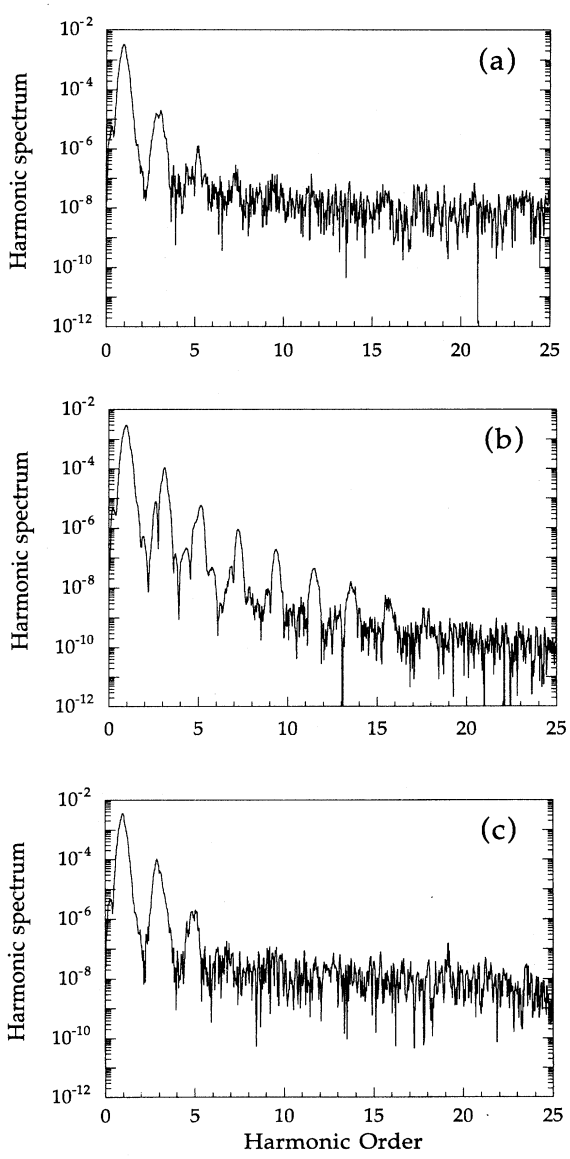

FIG. 3. Harmonic generation in the stabilization regime for one and two colors at $\omega=1$ a.u. (a) One color at $E_\omega=18.4$ a.u., (b) two colors at $E_\omega=E_{3\omega}=18.4$ a.u. and $\phi=0$, and (c) two colors at $E_\omega=E_{3\omega}=18.4$ a.u. and $\phi=\pi$.

monics (up to the fifth order) are generated in one color and two colors for a relative phase of $\phi=\pi$; however, the harmonic generation is extended to the 15–17th order when the relative phase between the fields is set equal to zero. The explanation of the large differences in the harmonic spectra arises from the differences in shape of the stabilized wave packet [6]. A richer structure on the stabilized wave packet is obtained for a phase difference $\phi=0$. The stabilized wave packet moves backward and forward along the classical excursion α_0, "crossing" the nucleus in its excursion. The richer the structure the larger the interaction with the nucleus, and therefore the better the harmonic generation.

As a conclusion to this Rapid Communication we summarize our results. We have demonstrated that two commensurate frequencies (1:3) interacting simultaneously with an atom modify substantially the generated harmonic spectra in the high-intensity domain. For instance, in the tunneling regime the two-color excitation extends and enhances the intensity of the produced harmonics. This enhancement is particularly clear for harmonics below U_i+U_p. In the stabilization regime the harmonic generation depends strongly on the relative phase between the two fields. The next step will be to apply two-color excitation to ions.

After submitting this paper we have learned about the work done by Eichmann *et al.* [22] on polarization-dependent high-order two-color mixing. Their results using two commensurate frequencies (1:2) at the same intensity and linear polarization also show the enhancement of harmonics in the two-color case compared to the one single color. The enhancement in our case is significantly stronger, probably caused by our choice of frequencies (1:3), which generates only odd harmonics.

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