Attosecond control of electron-ion-recollision dynamics

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(Received 15 April 2008; published 29 October 2008)

As is now well known, when an atom or molecule is ionized by an intense linearly polarized multicycle laser pulse, an electron wave packet is launched into the continuum at each half optical cycle and returns to the ion core several times, forming a train of attosecond electron bunches. In this paper, the possibility of controlling the relative proportion between the different electron bunches in the train using a two-harmonic multicycle laser field is demonstrated. This offers a highly promising method for disentangling the contribution of the different electron bunches to the electron-ion-recollision dynamics and eventually for probing ultrafast atomic or molecular dynamics.

DOI: 10.1103/PhysRevA.78.043420 PACS number(s): 32.80.Rm, 32.80.Qk, 32.80.Fb, 33.80.−b

I. INTRODUCTION

During the last decade, using the process of high-order harmonic generation (HHG) [1,2], significant efforts have been made to generate and characterize isolated attosecond laser pulses [3–6]. The HHG process is well explained by the three-step model introduced by Corkum in 1993 [7]. In HHG, the driving field is an intense linearly polarized near-infrared laser pulse, and the three steps are as follows. (1) Tunnel ionization launches an electron wave packet in the continuum at every half optical cycle of the driving laser field, (2) the electron gains kinetic energy from the laser field and returns to the ion core with a kinetic energy whose upper limit is equal to $3.17U_p$ (U_p being the ponderomotive energy), and (3) through recombination, extreme ultraviolet (xuv) light is generated.

To produce a single highly reproducible attosecond light pulse per driving laser pulse, the electron should return only once to the ion core [6]. The key to satisfying this condition is that the laser cycle phase relative to the few-cycle wave amplitude must be stabilized, so the key technology is the carrier envelope phase-stabilized few-cycle titanium-sapphire laser system [8,9], recently extended into the infrared spectral range [10]. Shorter attosecond pulses have been produced by using polarization gating of the HHG process [5,11,12].

In addition to high-order harmonic generation, other physical processes can be induced by the recollision, such as elastic scattering [13,14] and inelastic scattering inducing excitation or nonsequential ionization [15,16]. The recollision dynamics associated with these processes is highly complex. For every half optical cycle of a multicycle laser pulse, tunnel ionization launches an electron wave packet in the continuum and returns to the ion core through a train of attosecond electron bunches. Since the electron wave packet expands with time, the overall probability of returning to the ion core decreases with the return time. Therefore, one should expect that the first recollision is predominant. This argument is valid only if the atomic or molecular parent ion is time independent after ionization. For various atomic and molecular systems such as H_2, tunnel ionization induces dynamics [17–20], meaning that the cross section for the collision processes is time dependent. This is an important complication for understanding the dynamics induced by electron-ion recollision.

In this paper, a scheme for evading these complications by controlling the relative proportion between the different electron bunches is described and analyzed. The control scheme proposed requires a laser field composed of two coherently related harmonic frequencies. (The control wave is at half the frequency of the driving field at 800 nm.) The level of control is independent of the pulse duration. This is highly promising for disentangling the contribution of the different attosecond electron bunches to the electron-ion-recollision dynamics.

The difficulties in understanding the electron-ion-recollision dynamics are well illustrated by work on H_2 which has been interpreted in two quite different ways. The temporal structure of the train of attosecond electron bunches has been well characterized using an H_2^+ molecular clock (see Niikura *et al.* [17]). The attosecond molecular clock is the well-understood vibrational motion of H_2^+ in its ground electronic state ($^2\Sigma_g^+$). Briefly, tunnel ionization of H_2 launches two correlated wave packets; an electron one which leaves and a vibrational one onto the H_2^+ ground electronic state. The electron ejection starts the clock. When the electron returns to the ion core, inelastic scattering induced the dissociation or nonsequential ionization of H_2^+ and the kinetic energy of the H^+ fragments is a direct readout of the electron return time. Inelastic scattering stops the clock.

From the H^+ kinetic energy spectra they obtained, Niikura *et al.* [17] concluded that the first electron bunch returning to the ion core contributes the most to the electron-H_2^+-recollision dynamics. This contention has been opposed by Tong *et al.* [18,19] who suggested that it is the third recollision that is important here. The difference lies in the dynamics assumed. On one hand, Niikura and co-workers concluded that most of the H^+ fragments are produced via the first recollision, which induced the electronic excitation $^2\Sigma_g^+ \rightarrow {}^2\Sigma_u^+$, leading to the dissociation of $H_2^+ \rightarrow H^+ + H$. On the other hand, Tong and co-workers [18,19] concluded that it was the third recollision that was the most efficient in producing H^+ fragments, inducing also the elec-

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tronic excitation of H_2^+, but followed by ionization toward the Coulombic state. The debate remains, as shown by the recent paper by Hu *et al.* [20] followed by the comment of Tong *et al.* [21]. However, with the control scheme proposed in this paper, it should be experimentally possible to provide a clear answer to this question, by, for instance, destroying the possibility of any third-return collision. This approach offers the capability of controlling the relative contribution of the different electron bunches to the electron-ion-recollision dynamics.

II. METHODOLOGY

The control scheme requires a laser field composed of two harmonic frequencies that are phase locked. It is the phase difference between the two colors which is critical for the control scheme. The most intense component is at frequency ω_{driving} (most likely at a wavelength around 800 nm) and is elliptically polarized. The control is provided by the half-harmonic component at $\omega_{\text{driving}}/2$, which is also elliptically polarized. The electric field components are

$$E_x(t) = E_0 f(t)\left[\cos(\omega_{\text{driving}} t) + \frac{1}{\kappa_1}\cos\left(\frac{\omega_{\text{driving}}}{2}t + \phi_1\right)\right], \tag{1}$$

$$E_y(t) = E_0 f(t)\left[\frac{1}{\kappa_2}\sin(\omega_{\text{driving}} t) + \frac{1}{\kappa_3}\sin\left(\frac{\omega_{\text{driving}}}{2}t + \phi_2\right)\right]. \tag{2}$$

The frequency of the driving field corresponds to an 800 nm wavelength [$\omega_{\text{driving}}=0.057$ a.u. (atomic units)]. Such a laser field can be obtained experimentally by doubling the signal (or the idler) pulses of a high-energy optical parametric amplifier seeded by a high peak power Ti-sapphire laser [10]. This will assure that the phase difference between the two colors is locked.

For all simulations, $|E_0|^2(1+1/\kappa_1{}^2+1/\kappa_2{}^2+1/\kappa_3{}^2)=5.7\times10^{-3}$ a.u., corresponding to laser intensity of 2×10^{14} W/cm^2. By adjusting the free parameters of this two-harmonic laser field, i.e., κ_i and ϕ_i, it will be demonstrated that the temporal structure of the electron-ion-recollision dynamics can be controlled with a high level of precision.

Using the three-step model of Corkum [7], the control scheme is validated by modeling the electron-Ar^+-recollision dynamics of argon. This intuitive approach is now widely used to model recollision in atomic and molecular systems (see [17–19,22] for more details). In particular, it has proved its validity by application to nonsequential double ionization of helium [23].

It is convenient to begin with argon because its ionization potential (15.8 eV) is very similar to that of H_2 (15.6 eV) and to avoid grappling with molecular dynamics. The following procedure is applied for every time of creation. This approach is identical to the one described by Kitzler and Lezius [22]. In the first step, the ionization probability is calculated using the usual Ammosov-Delone-Krainov (ADK) theory [24]. In the second step, a large number of electron trajectories are calculated by solving Newton's equations of motion. To avoid an unphysically strong singularity at the origin, the model softens the two-dimensional Coulomb potential to $V(x,y)=1/\sqrt{x^2+y^2+1}$ [25]. In atomic units, the coupled Newtonian equations of motion are

$$\ddot{x} = -E_x(t) - \frac{x}{(x^2+y^2+1)^{3/2}}, \tag{3}$$

$$\ddot{y} = -E_y(t) - \frac{y}{(x^2+y^2+1)^{3/2}}. \tag{4}$$

The second step takes into account (i) the initial position of the electron at the time of creation (set at the classical creation point [22]), (ii) the initial velocity distributions parallel and perpendicular to the instantaneous laser polarization [26], and (iii) the Coulomb potential. In the third step, the electron trajectories returning to the ion core are counted if they pass through a radius equal to $r_\sigma=\sqrt{\sigma(E_k)/\pi}$, where $\sigma(E_k)$ is the Ar^+ ground-state electron impact excitation cross section for impact energy E_k [27]. [At this stage, we assume that the atomic system left after tunnel ionization is time independent (no dynamics). Also, we neglect the effects of excitation of Ar^+ after the recollision.] The probability of recollision is calculated using the ionization probability and those associated with the velocity distributions (parallel and perpendicular). The ability to control the relative proportion between the different electron bunches is demonstrated by comparing the results with a numerical simulation considering a linearly polarized driving laser field at 800 nm with a peak intensity of 2×10^{14} W/cm^2. In this calculation, the effect of the laser pulse envelope is not taken into account (i.e., one ignores any possible changes in the oscillating field from one cycle to another), which is equivalent to the regime of multicycle laser pulses [22].

III. RESULTS AND DISCUSSION

Figure 1(a) shows the temporal structure of the electron-Ar^+-recollision dynamics for this specific case of a single-color linear polarization laser field ($1/\kappa_1=1/\kappa_2=1/\kappa_3=0$). A train of attosecond electron bunches is observed.

In Figs. 1(b)–1(e), $\kappa_2=\kappa_3$, $\kappa_1=\infty$, and $\phi_2=0$. As the strength of E_y ($\sim1/\kappa_2=1/\kappa_3$) increases, the contribution of the electron bunches with return time longer than about 4 fs is considerably suppressed, and the temporal structure of the electron-Ar^+-recollision dynamics is greatly simplified. The probability of recollision decreases by about a factor of 2 when the strength of E_y increases to the maximum value ($\kappa_2=\kappa_3=2.5$). In Fig. 1(e), for $\kappa_2=\kappa_3=2.5$, the impact of the laser pulse envelope has been addressed. In this calculation, a 40 fs full width at half maximum (FWHM) pulse duration has been assumed. No significant difference is observed in comparison with calculation using the continuous wave (cw) approximation [Fig. 1(d)].

One more important parameter of the control scheme is the phase difference (ϕ_2) between the Y components of the 800 and 1600 nm laser fields. In Fig. 1, the phase difference

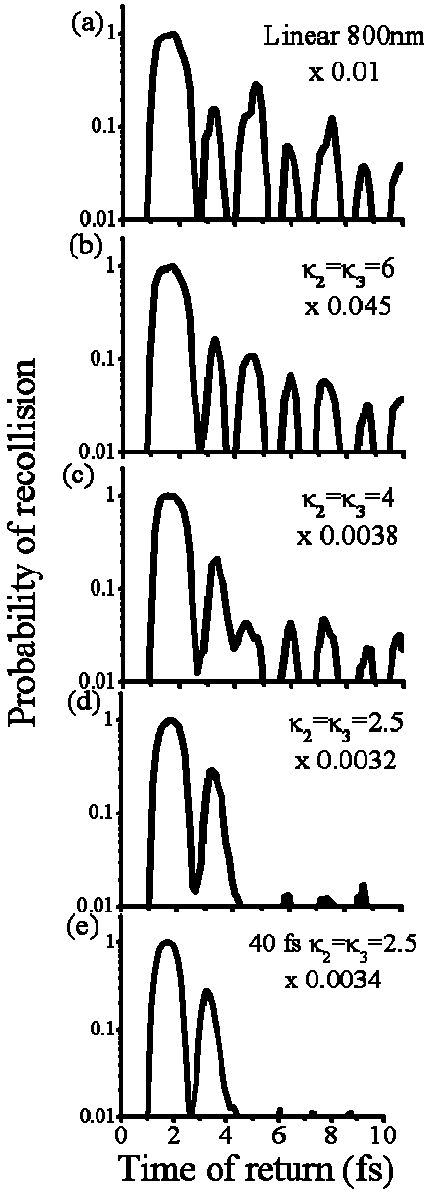

FIG. 1. Temporal structure of the electron-Ar^+-recollision dynamics as a function of E_y strength. The laser field is the combination of an elliptically polarized 800 nm laser field with a linearly polarized 1600 nm field with its polarization parallel to the minor component of the driving field ($\kappa_2=\kappa_3$, $\kappa_1=\infty$, and $\phi_2=0$).

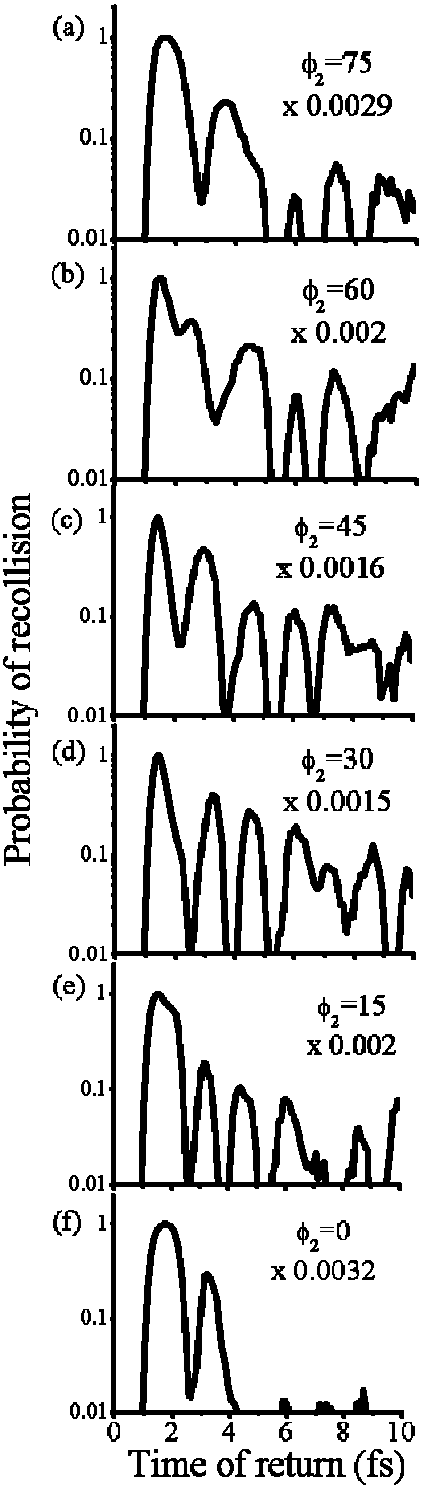

FIG. 2. Temporal structure of the electron-Ar^+-recollision dynamics as a function of ϕ_2. ϕ_2 is the phase difference between the two frequencies of E_y [see Eq. (2)].

was set to $\phi_2=0$. In Fig. 2, its impact on the temporal structure of the electron-Ar^+-recollision dynamics is shown. For those calculations, the parameters are $\kappa_2=\kappa_3=2.5$ and $\kappa_1=\infty$ [unchanged from Fig. 1(d)]. The temporal structure is made of only two bunches when the phase difference ϕ_2 equals $n\pi/2$ (n being an integer). The value of ϕ_2 strongly influences the return time of the first bunch, and for $\phi_2 \neq n\pi/2$ multiple electron bunches contribute to the electron-Ar^+-recollision dynamics. Those results with ϕ_2 demonstrate that its control is necessary, so the proposed control scheme requires a laser field composed of two harmonic frequencies that are phase locked.

By combining an elliptically polarized 800 nm driving laser field with a linearly polarized 1600 nm field, it is shown that the recollision dynamics can be highly simplified. In particular, the contribution of the electron bunches with return time longer than ~4 fs can clearly be suppressed. This simple control scheme thus offers the capability to answer the debate raised by the papers of Tong *et al.* [18,19], where it was urged that the third recollision (return time of ~4.6 fs)

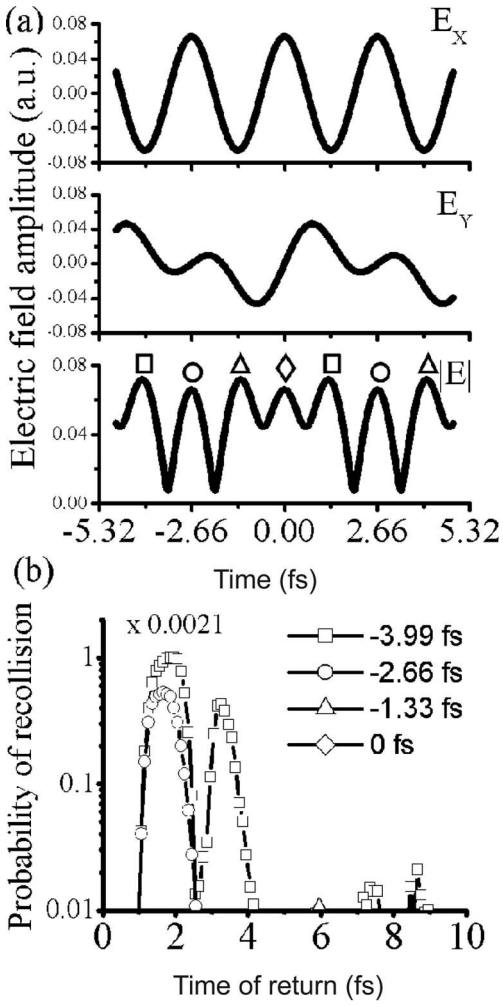

FIG. 3. (a) Electric field components for the laser parameters $\kappa_1=\infty$, $\kappa_2=\kappa_3=2.5$, and $\phi_2=0$. (b) Temporal structure of the electron-Ar^+-recollision dynamics as a function of the half optical cycle. The −1.33 fs and 0 fs half cycles are not contributing to the electron-Ar^+-recollision dynamics.

is the most important for the electron-$H_2{}^+$-recollision dynamics, as opposed to the paper of Niikura *et al.* emphasizing the first recollision (return time of ~1.8 fs) [17].

In order to understand the origin of the control scheme, the electric field components [for the case of Fig. 1(d) $\kappa_1=\infty$, $\kappa_2=\kappa_3=2.5$, and $\phi_2=0$] are plotted in Fig. 3(a). As shown in Fig. 3(b), only the first cycle contributes significantly to the recollision dynamics. The electron trajectories launched within the −3.99 fs half cycle contribute in the highest proportion to the recollision dynamics, and the temporal profile of the electron-Ar^+-recollision dynamics is characterized by two electron bunches. For the −2.66 fs half cycle, the recollision dynamics contains only the first bunch and the total probability of returning to the core is slightly lower (~2.6 times less). The contribution of the −3.99 fs half cycle is higher than the −2.66 fs half cycle primarily because of a higher ionization probability (~3.3 times more; $|E|$ is larger in the −3.99 fs half cycle). For the −1.33 fs and 0 fs half cycles, the low values reflect rather the fact that the recollision probability is very small (approximately 100 times less), since already after their creation in the continuum the electron trajectories experience a large modulation of the E_y field. This also explains the difference between the electron-Ar^+-recollision dynamics for the −3.99 fs half cycle and that associated with the −2.66 fs half cycle. The electron trajectories launched within the −2.66 fs half cycle experience the modulation of the E_y field sooner after their creation than the trajectories associated with the −3.99 fs half cycle. This explains why the suppression of the long trajectories is incomplete for the −3.99 fs half cycle and that the temporal structure of the electron-Ar^+-recollision dynamics is made of two attosecond bunches.

In the cw approximation, valid for multicycle laser pulses, these recollision dynamics repeat at half the frequency of the driving laser field, i.e., electron trajectories launched within the 1.33 fs half cycle are equivalent to those launched within the −3.99 fs half cycle (the same for the −2.66 and 2.66 fs half cycles).

In principle, by adding a third color at twice the frequency of the driving field, one can promote the −2.66 fs half cycle by increasing the ionization rate while suppressing it within the −3.99 fs half cycle (not shown). Then the temporal structure of the electron-ion-recollision dynamics will be made up of a single electron bunch without the requirement of few-cycle laser pulses. However, having three phase-locked harmonic colors will be extremely challenging experimentally.

The results presented so far were obtained using a linearly polarized control field with its polarization axis perpendicular to the main component of the driving field [i.e., $\kappa_1=\infty$; see Eqs. (1) and (2)]. In Fig. 4, the laser parameters are $\kappa_1=2.5$ and $\phi_1=\pi$. For the specific case of Fig. 4(a), $\kappa_2=\kappa_3=\infty$, i.e., the total electric field is linearly polarized along the X axis and composed of two harmonic frequencies. The electric field is plotted in the inset of Fig. 4(a). Under those laser parameters, multiple electron bunches contribute to the temporal structure of the electron-Ar^+-recollision dynamics. In comparison with the temporal structure obtained with a linearly polarized 800 nm laser field (see the dotted curve), one can observe that, while the main electron bunch remains at ~1.8 fs, the recollision dynamics is modified for return times longer than 2.66 fs. In Fig. 4(b), the recollision dynamics associated with each half optical cycle is presented. The temporal structure of the electron-Ar^+-recollision dynamics, as well as their relative contribution to the overall dynamics, depends on the half optical cycle. In particular, the electron trajectories launched within the −2.66 fs half cycle contribute the most to the recollision dynamics, primarily due to a higher ionization probability.

In Fig. 5(a), the temporal structure of the electron-Ar^+-recollision dynamics is presented for the following laser parameters: $\kappa_1=\kappa_2=\kappa_3=2.5$, $\phi_1=\pi$, and $\phi_2=0$. In that case, the Y component of the electric field is plotted in the inset and the X component is the same as the one shown in the inset of Fig. 4(a). The contribution of the electron bunches with return time longer than ~2.66 fs is suppressed. [The validity of the cw approximation was verified by repeating the calculation with a 40 fs FWHM pulse envelope. No differences were observed (not shown).] In Fig. 5(b), the recollision dynamics for each half optical cycle is presented. Similar to the results of Fig. 4(b), the electron trajectories

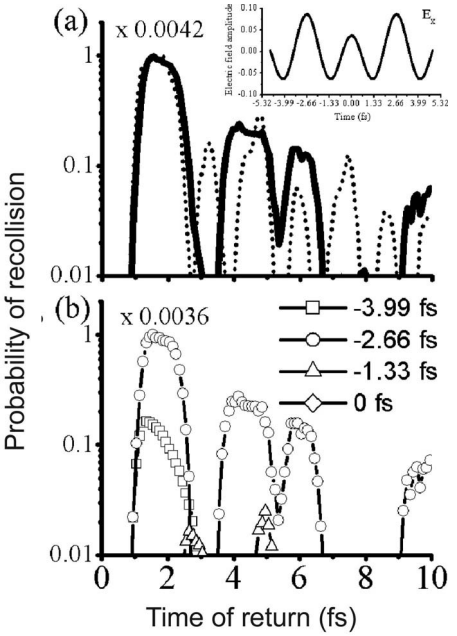

FIG. 4. Dotted curve represents the electron-Ar^+-recollision dynamics for a linearly polarized 800 nm laser field. (a) Temporal structure of the electron-Ar^+ recollision for the laser parameters $\kappa_1=2.5$, $\phi_1=\pi$, and $\kappa_2=\kappa_3=\infty$. The laser field is linearly polarized along the X axis (shown in the inset). (b) Temporal structure of the electron-Ar^+-recollision dynamics as a function of the half optical cycle.

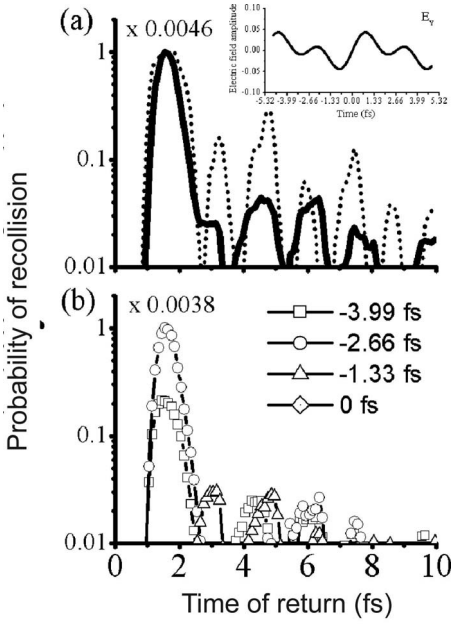

FIG. 5. Dotted curve represents the electron-Ar^+-recollision dynamics for a linearly polarized 800 nm laser field. (a) Temporal structure of the electron-Ar^+ recollision for the laser parameters $\kappa_1=\kappa_2=\kappa_3=2.5$, $\phi_1=\pi$, and $\phi_2=0$. The E_y component is shown in the inset and the E_x component is the same as in the inset of Fig. 4(a). (b) Temporal structure of the electron-Ar^+-recollision dynamics as a function of the half optical cycle.

launched within the −2.66 fs half cycle contribute the most to the recollision dynamics due to the higher ionization probability. With this simple control scheme, the temporal structure of the electron-Ar^+-recollision dynamics is highly simplified as the first recollision becomes the most predominant and remains at the same return time. Those results show that the X component of the control field provides an extra knob for modifying the temporal structure of the electron-ion-recollision dynamics.

IV. CONCLUSION

The first electron bunch has been used to probe ultrafast molecular dynamics [28]. Despite the use of a multicycle laser pulse (50 fs, 800 nm), 200 as temporal resolution and 0.05 Å spatial resolution have been obtained. This counterintuitive result is possible because of the correlation between the electron wave packet launched in the continuum by tunnel ionization and the molecular vibrational wave packet. In analogy to a pump-probe spectroscopy experiment, tunnel ionization can be seen as the pump while the recollision is the probe. During the delay between the pump and the probe, the molecular wave packet evolves. Using few-cycle laser pulses, one can extract cleaner information about the molecular dynamics since the first recollision becomes predominant [22,29].

Using the control scheme proposed in this paper, one can possibly extract more information about the molecular dynamics than with few-cycle laser pulses. One can see the train of attosecond electron bunches as multiple attosecond probes fixed at various pump-probe delays. By controlling the relative proportion between the electron bunches, one can possibly extract the molecular dynamics at various delays simultaneously. The ability to control the relative contribution of the second and third recollisions has been clearly demonstrated and offers the possibility to fully extract the recollision dynamics associated with those returns. Finally, as proposed in the paper, by using an optical parametric amplifier (OPA) laser source, it will give the opportunity to tune the frequencies of the driving and control laser fields, which will change the return time of the different electron bunches [28] while the ability to control the temporal structure of the electron-ion-recollision dynamics remains.

ACKNOWLEDGMENTS

F.L. acknowledges financial support from INRS-EMT, Canada's Natural Science and Engineering Research Council, and Le Fonds Québécois de la Recherche sur la Nature et les Technologies. F.L. also thanks Dr. P. B. Corkum at NRC Canada and Dr. T. W. Johnston at INRS-EMT for stimulating discussions.

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