Effects of the carrier-envelope phase on atomic ionization by the pulse train in the multiphoton-ionization regime

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We theoretically investigate the effects of the carrier-envelope phase on atomic ionization by the pulse train in the multiphoton-ionization regime, and show that regardless of the number of pulses, the phase dependence is striking. Similarity and the difference between the phase-dependent phenomena by the pulse train and isolated pulse are clearly understood in the frequency domain.

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It is now well understood that the carrier-envelope phase (CEP) of a laser field plays a very important role in laser-matter interactions when the duration of the pulse becomes as short as a few cycles or less. Although some theoretical works have indicated the emergence of phase-dependent ionization [1–4], it was not until the first experimental demonstrations by Paulus *et al.* [5,6] and Baltuska *et al.* [7] who have, respectively, utilized few-cycle pulses to observe the phase-dependent ionization and produced phase-stabilized few-cycle pulses that the importance of the carrier-envelope phase is recognized and lots of works start to appear in the literature [8–15]. Note that all the above experimental as well as theoretical works for the CEP in the tunneling ionization (TI) [2–12] and multiphoton-ionization (MI) regimes [1,13–15] refer to the *isolated pulse*.

A completely different approach has been proposed and experimentally demonstrated by the Stanford group [16–18] to produce ultrashort pulses through the synthesis of Raman sidebands with extremely high efficiency. Naturally, the synthesized pulses form a train with a time interval given by the inverse of the energy difference of Raman sidebands. Although the vibrational states of deutrium (D_2) gas has been employed for the first experimental demonstration [17], rotational as well as vibrational states of other kinds of gases can be alternatively used [19,20]. A natural but not yet answered question is whether and how the *pulse train* with a controlled CEP interacts with a matter. The above question is also related to the recent significant advancement of laser technology to stabilize the CEP of mode-locked lasers [21,22] where the carrier-envelope offset rather than the CEP itself is of great importance. However, if one is to utilize this light source which naturally forms a pulse train, the CEP can play a crucial role as well during light-matter interactions.

At first glance one might naively consider that the persistence of phase-dependent ionization by a pulse train instead of an isolated pulse is obvious if one is to make a prediction in the time domain utilizing, say, S matrix under strong-field approximation. Actually this can be true in the TI regime. We must, however, bear in mind that we are assuming the MI regime where it is well understood that the phase dependence by few-cycle pulses appears at the *valleys* in the angle-resolved photoelectron spectra (AR-PES) [1,14,15]. In contrast, as we will show later on in this paper, we find that the phase dependence appears at the *peaks* in the AR-PES if a pulse train with a sufficient bandwidth is employed. Clearly we need a unified interpretation which is applicable for both isolated pulses and pulse trains.

The purpose of this paper is to theoretically investigate the effects of the CEP by a pulse train and show, with a clear physical picture in the frequency domain, that the phase dependence can be clearly seen even if the pulses form a train. Related to this work, we note that the use of a pulse train, in particular with a duration of monocycle or sub-optical-cycle for each subpulse, certainly has an advantage in the TI regime since it offers far more flexibility to *steer the electron motion* through the relative phases and the intensity ratios of different frequency components, and of course the CEP, etc., which could result in the efficient generation of (nearly isolated) attosecond pulses with a sufficient time interval and at a high repetition rate. In this paper, however, we focus on the moderate intensity range (MI regime) since such an intensity range would be within reach in the very near future, and moreover, the origin of the phase dependence in the MI regime [13–15] has been found to be quite different from that in the TI regime. Along this line, a successful generation of sub-single-cycle optical pulse train with a constant CEP through the Raman sidebands has been reported very recently [23]. In order to obtain a clear physical insight, we carry out the theoretical calculations by considering two kinds of pulse trains from the synthesis of vibrational and rotational Raman sidebands, and find that the quantum-mechanical interference, which can be more clearly seen in the *frequency domain* rather than the *time domain*, is the physical origin of the phase dependence.

In this paper we assume that a pulse train is generated by the synthesis of vibrational (rotational) Raman sidebands of D_2 (parahydrogen) gas, which are referred to the *vibrational (rotational) pulse train* from now on. In either case, provided the two applied laser frequencies ω_0 and ω_1, the synthesized electric field can be written in the form of

$$E(t) = f(t)\sum_q \varepsilon_q \cos(\omega_q t + \phi_q), \tag{1}$$

where $f(t)$ is the envelope function (cos-squared function in this paper), and ω_q and ϕ_q are the frequency and the phase of the Raman sidebands of order q, i.e., $\omega_q = \omega_0 + q\omega_m$ with ω_m

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being the modulation frequency and ε_q the corresponding maximum field amplitude, respectively. Although the fundamental frequency, ω_0, is generally written as $\omega_0 = n\omega_m + \delta_{\text{off}}$, where n is an integer and δ_{off} is a so-called frequency offset, δ_{off} is required to be zero in this paper, unless otherwise noted, so that the CEP of each pulse becomes constant throughout the pulse train [21–23]. That is, $\omega_q = (n+q)\omega_m$ and accordingly $\phi_q = \phi + (n+q)\Delta\phi$, where both ϕ and $\Delta\phi$ are constants. Similar to the Fourier decomposition of a pulse with a continuous frequency spectrum [24] (i.e., isolated pulse), the zeroth- and first-order terms with respect to the frequency, ϕ and $\Delta\phi$, determine the CEP and group delay of the pulse, respectively.

The target atom we have employed is a Na atom which is represented by the Hartree-Fock potential with a core-polarization correction [25]. The calculation has been carried out by solving the three-dimensional time-dependent Schrödinger equation in a spherical box of radius 5000–12000 atomic units. All field components are assumed to be linearly polarized along the z axis. We note that when we refer to the pulse duration it is the *envelope duration* defined by the number of cycles [full width at half maximum (FWHM)] at the fundamental frequency ω_0. Without loss of generality we have assumed that $\Delta\phi = 0$ unless otherwise mentioned.

To start with, we consider the case of *vibrational pulse train* $(-2 \le q \le 13)$ for the D_2 gas ($\omega_m = 0.3712$ eV) with a peak intensity of 10^{10} W/cm^2 for $\omega_0 = 1.4848$ eV (and accordingly $n=4$), and the ratios of ε_q's are taken from the circled experimental data in Fig. 3 of Ref. [17]: If normalized with respect to ε_0, they are $\varepsilon_{-2}=0.55$, $\varepsilon_{-1}=1$, $\varepsilon_0=1$, $\varepsilon_1=0.34$, $\varepsilon_2=0.29$, $\varepsilon_3=0.18$, $\varepsilon_4=0.18$, $\varepsilon_5=0.15$, $\varepsilon_6=0.13$, $\varepsilon_7=0.12$, $\varepsilon_8=0.1$, $\varepsilon_9=0.065$, $\varepsilon_{10}=0.058$, $\varepsilon_{11}=0.04$, $\varepsilon_{12}=0.032$, and $\varepsilon_{13}=0.013$. First we investigate the variation in the AR-PES [up ($+z$) and down ($-z$) directions] for different CEP's and envelope durations. Representative results are shown in Fig. 1, where the inset in each graph shows the temporal electric fields at $\phi=0$ (solid) and $\pi/2$ (dashed), respectively. Since $\omega_m = 0.3712$ eV, the time interval between successive pulses is 11.1 fs. For six cycles the AR-PES is clearly phase dependent, although the envelope duration is already longer than a few cycles. Note that the phase dependence clearly persists even if the envelope duration becomes as long as 24 cycles. We point out that the angle-integrated signal (or equivalently total ion yield) shows very little phase dependence as it is so for the Cs atom by the isolated nearly monocycle pulse in the MI regime [13]. The total ionization yield, however, may also exhibit a phase dependence if different atoms (such as rare gases) are employed [14].

In the above calculations we have assumed the vibrational pulse train for the order of $-2 \le q \le 13$, which results in an enormous (discretized) spectral bandwidth. One may wonder if a similar phase dependence can be seen for the vibrational pulse train with a *reduced order*. In order to answer this question, we introduce an asymmetry parameter, A, defined as $A(\varepsilon,\phi) \equiv [S_{\text{up}}(\varepsilon,\phi) - S_{\text{down}}(\varepsilon,\phi)]/[S_{\text{up}}(\varepsilon,\phi) + S_{\text{down}}(\varepsilon,\phi)]$, where ε stands for the photoelectron energy and S_{up} and S_{down} are the angle-resolved photoelectron signals to the upward and downward directions, and carry out

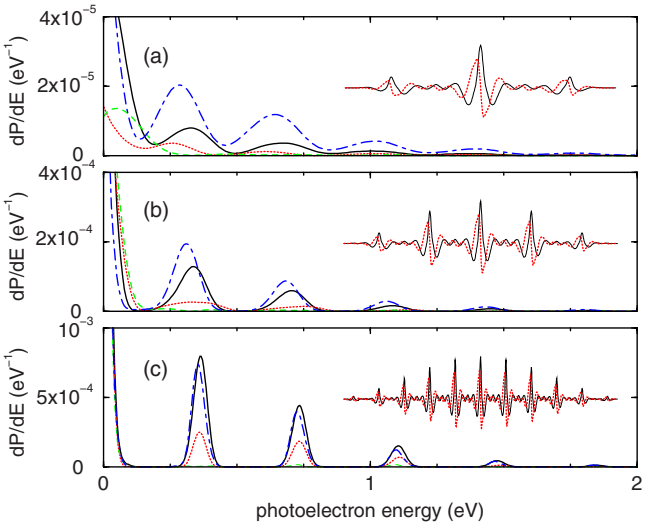

FIG. 1. (Color online) AR-PES by the vibrational pulse train $(-2 \le q \le 13)$ obtained from the D_2 gas with different CEP's and envelope durations, (a) six cycles, (b) 12 cycles, and (c) 24 cycles where solid and dashed (long-dashed and dot-dashed) lines in each graph represent photoelectron signals to the upward and downward directions at $\phi = 0(\pi/2)$, respectively. Inset in each graph shows the corresponding temporal electric fields for $\phi=0$ (solid) and $\phi = \pi/2$ (dashed).

calculations for $-2 \le q \le 3$ with a 24 cycles envelope duration. The variation in the asymmetry parameter is shown in Fig. 2(a) by a false-color plot as functions of photoelectron energy and the CEP together with AR-PES shown in Figs. 2(b) and 2(c) at $\phi=0$ and $\pi/2$, respectively. It is now clear that the AR-PES show a strong phase dependence even for the vibrational pulse train with a reduced order. One may still argue that the phase dependence we have seen above can be a consequence of accidental interference since the temporal variation in the electric field is perfectly symmetric with respect to $t=0$ because we have set $\Delta\phi=0$. In order to check this, we have introduced a few fs group delay by setting a nonzero value for $\Delta\phi$ to temporally shift the pulse train relative to the envelope and performed the calculations, finding practically identical results. This indicates that practically the same phase dependence would be seen for any group delay if the envelope duration is not too short.

Another interesting question is how much the phase dependence survives if there is a *phase slip* in the pulse train. Such a pulse-to-pulse phase slip will take place if the frequency offset δ_{off} is not exactly zero. To investigate this, we have carried out several calculations for different values of δ_{off} with different envelope durations (results are not shown here due to the space limitation), and found that for a given envelope duration the phase dependence gradually disappears as δ_{off} more deviates from zero, and consistently for a longer envelope duration the phase dependence disappears at a smaller value of δ_{off}.

The last question we would like to address is whether and how much phase dependence we can see if we employ a *rotational pulse train*. Typically the energy interval between rotational states is one order of magnitude smaller than that between the vibrational ones, and hence the time interval

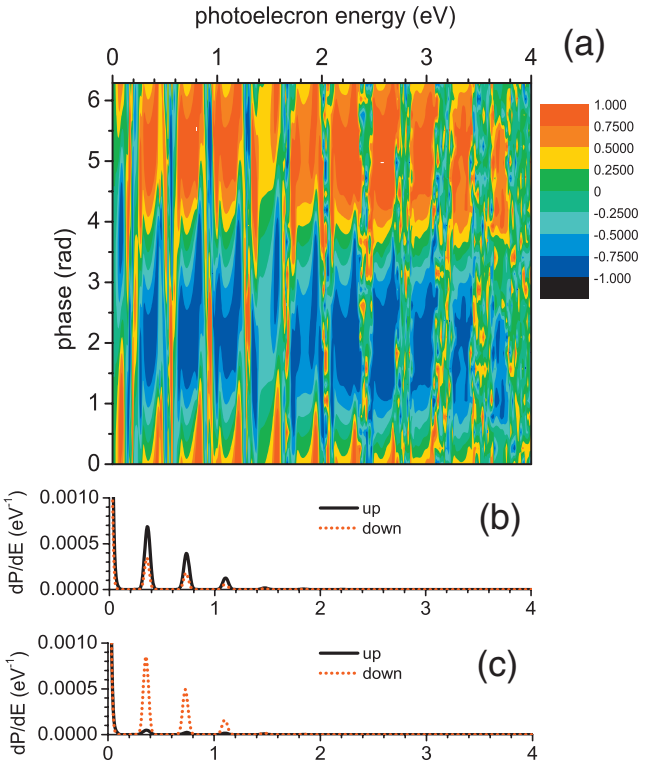

FIG. 2. (Color) (a) False-color plot of the asymmetry parameter, A, as functions of photoelectron energy and CEP, and AR-PES at (b) $\phi=0$ and (c) $\pi/2$ by the vibrational pulse train with a reduced order ($-2\leq q\leq 3$). The envelope duration is 24 cycles. In graphs (b) and (c) solid and dashed lines represent the signals to the upward and downward directions, respectively.

becomes about one order of magnitude longer. Representative results are shown in Fig. 3 for the parahydrogen gas ($\omega_m=0.0439$ eV) with $\omega_0=1.6243$ eV (and accordingly $n=37$) for the order of $-8\leq q\leq 8$ and $-30\leq q\leq 30$ with a 95 cycles envelope duration and the respective peak intensities of 10^{10} W/cm^2 and 3×10^9 W/cm^2 for $\omega_0=1.6243$ eV.

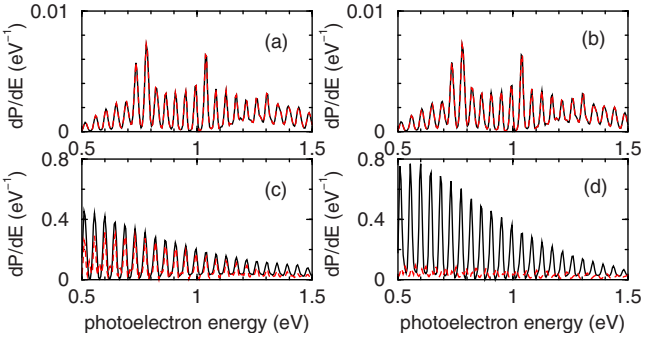

FIG. 3. (Color online) AR-PES by the rotational pulse train obtained from the parahydrogen gas with different orders of sidebands. The order of sidebands and CEP's are (a) $-8\leq q\leq 8$ and $\phi=0$, (b) $-8\leq q\leq 8$ and $\phi=\pi/2$, (c) $-30\leq q\leq 30$ and $\phi=0$, and (d) $-30\leq q\leq 30$ and $\phi=\pi/2$, respectively. The envelope duration is 95 cycles. In each panel, the photoelectron signals to the upward and downward directions are shown by solid and dashed lines, respectively.

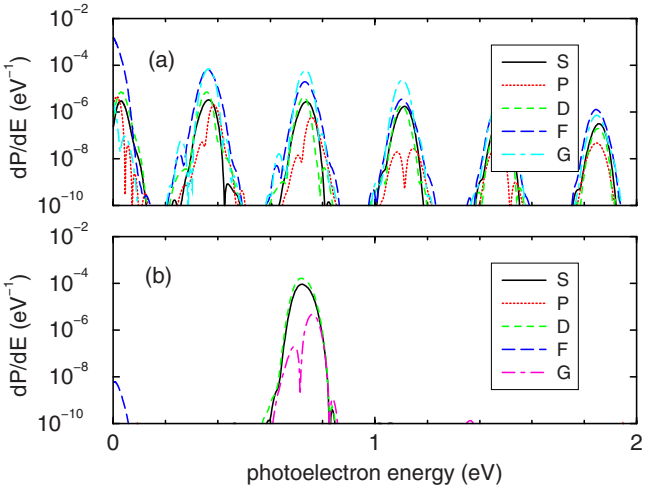

FIG. 4. (Color online) Partial angle-integrated photoelectron spectra by the (a) vibrational pulse train ($-2\leq q\leq 13$) and (b) isolated pulse with an envelope duration of 36 cycles. Solid, dotted, dashed, long-dashed, and dot-dashed lines in each panel represent partial ionization spectra into the S, P, D, F, and G waves, respectively. The CEP is $\phi=\pi/2$ for both graphs.

ε_q's are assumed to have values such that the full width at half maximum of the spectrum approximately becomes half of the entire orders. Although we find no phase dependence when the spectral bandwidth is relatively narrow ($-8\leq q\leq 8$), the phase dependence becomes significant when it is increased up to $-30\leq q\leq 30$. This result clearly indicates that whether a vibrational or rotational pulse train is employed, the phase dependence can be seen under certain conditions, and it is neither the time intervals nor spectral interval but the *spectral bandwidth* that is important.

To justify the above statement regarding the origin of phase dependence induced by the pulse train, we now compare the partial photoelectron spectra by the pulses with a same envelope duration but different spectral bandwidths, i.e., polychromatic and quasimonochromatic fields. The results are shown in Fig. 4 for the vibrational pulse train representing a polychromatic field ($-2\leq q\leq 13$) and the isolated pulse representing a quasimonochromatic field ($q=0$ only) at $\phi=\pi/2$ with a same envelope duration, which is chosen to be 36 cycles. What we can learn from Fig. 4 is that because

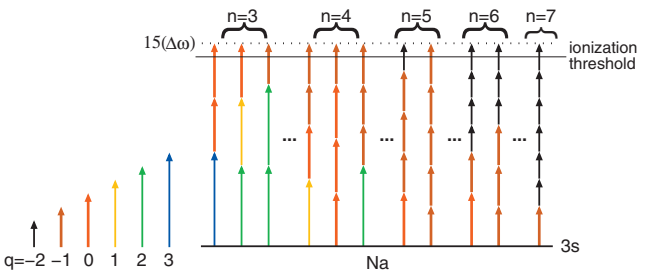

FIG. 5. (Color) Frequency domain interpretation of the phase dependence at the first peak for the vibrational pulse train with a reduced order ($-2\leq q\leq 3$). n represents the number of photons involved for ionization. Note that the first peak corresponds to the continuum energy of 15 $\Delta\omega$ from the ground state.

of the different spectral bandwidths the partial photoelectron spectra are entirely different for the pulse train and isolated pulse even if the envelope duration is the same. For instance, we consider the peak located at ~0.7 eV which appears in both Figs. 4(a) and 4(b). For the isolated pulse [Fig. 4(b)], this peak originates from four-photon ionization ($4\omega_0$) starting from the $3s$ ground state, and hence only even-parity continua (S, D, and also G waves) contribute to the peak. The situation is quite different for the case of pulse train [Fig. 4(a)]. Now the peak at the same energy originates from *different orders* of the MI processes (three- to eight-photon processes for the case of $-2 \leq q \leq 3$) by the combination of photons with different photon energies in the polychromatic field, some of which are written as $4\omega_0$, $\omega_{-1}+2\omega_0+\omega_1$, etc., for the four-photon processes with an accumulated CEP of 4ϕ, and $2\omega_1+\omega_2$, $\omega_0+\omega_1+\omega_3$, etc., for three-photon processes with an accumulated CEP of 3ϕ. Because the even-/odd-number-photon ionization processes coexist with different accumulated CEP's, it is now clear why the phase-dependent asymmetric photoelectron ejection occurs at the *peaks* for the pulse train with a sufficient bandwidth. Note that this interpretation is fully consistent with all numerical results presented in this paper. Obviously which channels make more contribution than the others strongly depend on the spectral distribution of the pulse.

From the above consideration we have clarified that the physical origin of the phase dependence by the pulse train in the MI regime can be most easily understood in the frequency domain as *phase-dependent interference by the phase-locked polychromatic field*, which is most clearly shown in Fig. 5 for the vibrational pulse train with a reduced order ($-2 \leq q \leq 3$). Although not shown in Fig. 5, the ionization paths may include the stimulated emission processes with a contribution to the accumulated phase of $-\phi$ as well as the absorption processes with a contribution to the accumulated phase of ϕ as mentioned before. This interpretation is actually reminiscent of the two-color ($\omega-2\omega$) schemes with ns pulses [26,27]. Based on this interpretation, we can say that as long as the spectral bandwidth is sufficient, i.e., *suboctave* so that MI channels with both even- and odd-photon numbers coexist and interfere at the same continuum energies, a significant phase dependence, similar to Figs. 1 and 2, should be observed no matter how long (>ps) the envelope duration of the pulse train is. Under these conditions the CEP effects by the train of subpulses with a *sub-optical-cycle* duration is clearly seen at the *peaks* of the AR-PES.

For better understanding it would be instructive to consider the pulse train consisting of subpulses with a *few-cycle* duration in the MI regime. For such a case, the CEP effects is still there (not shown in this paper) but they appear only at the *valleys* between the successive peaks in the AR-PES. This is similar to the case of the isolated few-cycle pulse in the MI regime with or without a chirp [1,14,15]. The difference, however, is that for the pulse train consisting of few-cycle pulses, *rapid oscillations* originating from the discretized spectral profile are superposed to the photoelectron signals at the *valleys*, which would make the experimental observation quite difficult.

In summary, we have theoretically investigated the effects of the carrier-envelope phase by the pulse train in the MI regime, and clarified that the physical origin of the phase-dependent effects is its spectral bandwidth. Namely, if the bandwidth is as broad as suboctave which is sufficient to support sub-optical-cycle subpulses, the phase dependence appears at the peaks in the AR-PES due to the phase-dependent interference by the phase-locked polychromatic field, irrespective of the envelope duration (>ps) of the pulse train and the mode spacing (vibrational or rotational pulse train).

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[1] E. Cormier and P. Lambropoulos, Eur. Phys. J. D **2**, 15 (1998).
[2] I. P. Christov, Opt. Lett. **24**, 1425 (1999).
[3] I. P. Christov, Appl. Phys. B: Lasers Opt. **70**, 459 (2000).
[4] P. Dietrich, F. Krausz, and P. B. Corkum, Opt. Lett. **25**, 16 (2000).
[5] G. G. Paulus *et al.*, Nature (London) **414**, 182 (2001)
[6] G. G. Paulus, F. Lindner, H. Walther, A. Baltuska, E. Goulielmakis, M. Lezius, and F. Krausz, Phys. Rev. Lett. **91**, 253004 (2003).
[7] A. Baltuska *et al.*, Nature (London) **421**, 611 (2003).
[8] T. Brabec and F. Krausz, Rev. Mod. Phys. **72**, 545 (2000).
[9] D. B. Milosevic, G. G. Paulus, and W. Becker, Phys. Rev. Lett. **89**, 153001 (2002).
[10] S. Chelkowski and A. D. Bandrauk, Phys. Rev. A **65**, 061802(R) (2002).
[11] F. Lindner, G. G. Paulus, H. Walther, A. Baltuska, E. Goulielmakis, M. Lezius, and F. Krausz, Phys. Rev. Lett. **92**, 113001 (2004).
[12] C. Altucci, V. Tosa, R. Velotta, and C. H. Nam, Phys. Rev. A **70**, 065402 (2004).
[13] T. Nakajima and S. Watanabe, Phys. Rev. Lett. **96**, 213001 (2006).
[14] T. Nakajima and S. Watanabe, Opt. Lett. **31**, 1920 (2006).
[15] T. Nakajima and E. Cormier, Opt. Lett. **32**, 2879 (2007).
[16] S. E. Harris and A. V. Sokolov, Phys. Rev. Lett. **81**, 2894 (1998).
[17] A. V. Sokolov, D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. E. Harris, Phys. Rev. Lett. **85**, 562 (2000).
[18] A. V. Sokolov, D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. E. Harris, Phys. Rev. Lett. **87**, 033402 (2001).
[19] M. Katsuragawa, K. Yokoyama, T. Onose, and K. Misawa, Opt. Express **13**, 5628 (2005).
[20] W.-J. Chen and A. H. Kung, Opt. Lett. **30**, 2608 (2005).
[21] D. J. Jones *et al.*, Science **288**, 635 (2000).

[22] R. K. Shelton *et al.*, Science **293**, 1286 (2001).
[23] W.-J. Chen *et al.*, Phys. Rev. Lett. **100**, 163906 (2008).
[24] J.-C. Diels and W. Rudolph, *Ultrashort Lasr Pulse Phenomena* (Academic, New York, 1996).
[25] T. Nakajima, Phys. Rev. A **75**, 053409 (2007).
[26] Y.-Y. Yin, C. Chen, D. S. Elliott, and A. V. Smith, Phys. Rev. Lett. **69**, 2353 (1992).
[27] E. Dupont, P. B. Corkum, H. C. Liu, M. Buchanan, and Z. R. Wasilewski, Phys. Rev. Lett. **74**, 3596 (1995).