Prepare for J. Phys. B, version 2.0 2 November 2009

Carrier-envelope phase dependence of nonsequential double ionization of H_2 by few-cycle laser pulses

X. M. Tong[1] and C. D. Lin[2]

[1] Institute of Materials Science, Graduate School of Pure and Applied Sciences, and Center for Computational Sciences, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8573, Japan

[2] Physics Department, Kansas State University, Manhattan, KS 66506-2601

Abstract. We studied the dependence of double ionization of H_2 molecules on the carrier-envelope phase of a few-cycle laser pulse in the nonsequential double ionization regime. For short pulses at low intensities the strong dependence is due to the return energy of the rescattering electrons and the result may be used for determining the absolute value of the carrier envelope phase.

PACS numbers: 34.50.Rk, 31.70.Hq, 95.55.Sh

Submitted to: *J. Phys. B: At. Mol. Phys.*

1. Introduction

Recent advances in femtosecond laser technology have opened the door for studying laser-matter interaction on the time scale of a few optical cycles [1, 2]. For such few-cycle pulses, the electric field $E(t) = E_0(t)cos(\omega t + \delta)$ depends on the phase of the carrier wave with respect to the pulse envelope $E_0(t)$, the so-called carrier-envelope phase (CEP) δ. Since the feedback control of the CEP was first established [3, 4], it has become possible to perform laser-matter interactions where the outcome depends on the CEP. Such measurements, in turn, provide means for determining the carrier-envelope phase of a few-cycle pulse.

Experimentally, the high harmonic generation (HHG) [5] and the left-right asymmetry of the electron yields from the above-threshold ionization (ATI) [6, 7] have been shown to depend on the CEP. While these experiments demonstrate how the results depend on the CEP of the few-cycle pulse, the determination of its actual value has to depend on the predictions of theoretical models.

To optimize the contrast of the CEP dependence it is clear that one would employ high energy electrons generated in laser-matter interactions directly (as in ATI electrons)

or indirectly (as in HHG). While both HHG and ATI from atoms are considered to be well understood, the yields of high energy electrons and high order harmonics are many orders of magnitude smaller and more difficult to calculate them accurately. Thus other processes which are initiated by high-energy electrons are of interest. These high-energy electrons, for example, can initiate nonsequential double ionization, and they are expected to depend on the carrier-envelope phase, as demonstrated in the double ionization of Ar by Liu *et al* [8]. Most recently, it has been further demonstrated in the asymmetric D^+ ejection in the dissociative ionization of D_2 molecules by Kling *et al* [9], where the dissociative ionization was initiated by the rescattering process.

The dependence of ATI spectra on the carrier-envelope phase of a few-cycle pulse has been studied in many papers recently [10, 11, 12, 13, 14, 15, 16]. The spectra were calculated either by directly solving the time-dependent Schrödinger equation for "standard" system such as atomic hydrogen, or by using second-order S-matrix theory with additional approximations [17]. Alternatively, the dissociation of molecular ions by few-cycle pulses has been shown to depend on the carrier-envelope phase as well [18, 19]. Finally, the carrier-envelope phase dependence can also be determined by ionization from metallic surfaces [4, 20]. The latter may be mostly useful for pulses with intensity of the order of 10^{12} W/cm^2 or lower, but the determination of the CEP depends critically on the theoretical simulation with no alternative means for checking its validity.

In this paper we studied the nonsequential double ionization of H_2 by few-cycle pulses. For such studies, full *ab initio* quantum mechanical calculations is impossible. Previously we have studied nonsequential double ionization and dissociative ionization of H_2 (or D_2) by a long pulse with duration of the order of 25-40 fs [21, 22] and the resulting predictions were shown to agree well with experiments [23, 24, 25]. The theoretical model [21, 22] employed there can be easily extended to study nonsequential double ionization by the few-cycle pulses. In fact, the previously developed rescattering theory, with some modifications, have been used recently by us [26] to interpret the dissociative ionization experiment of Kling *et al* [9] where it was found that the asymmetric D^+ ejection depends on the carrier-envelope phase of the laser pulse.

The paper is organized as following. A brief review of the physical processes and our theoretical model is given in Sec. 2. The theoretical results are presented and analyzed in Sec. 3. We have also examined how the energies of the rescattered electrons depend on the carrier-envelope phase which are important for determining the results of the calculated double ionization probabilities. The last section summarizes with a short conclusion.

2. Theory of nonsequential double ionization in H_2

Figure 1 depicts schematically the physical processes in the nonsequential double ionization of H_2 in the laser field. Note that the present theory can be directly carried over to D_2. The detailed theoretical modelling has been described previously for the

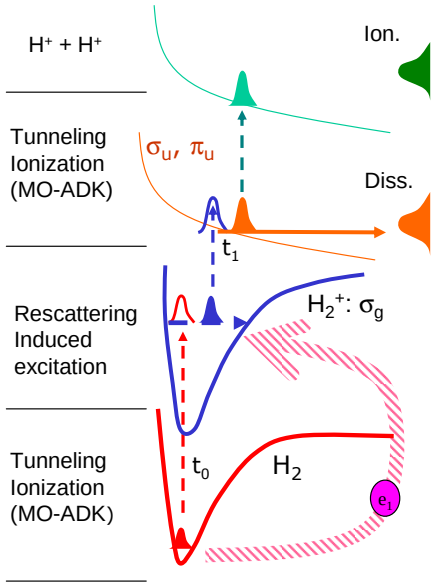

Figure 1. (Color online) Schematic of the major processes leading to Coulomb breakup of the H_2 in an intense laser field.

longer pulses [21, 22]. Here we summarize the basic elements in the model and the modification for sub-10 fs pulses. The laser field first ionizes H_2 to H_2^+ via tunneling ionization at time t_0. The ionized electron is accelerated by the laser but it may be driven back as the electric field changes direction in the next half cycle. If the returned electron acquires enough energy it may recollide with the parent ion H_2^+, to ionize it or to excite it to higher electronic states. By adjusting the laser to proper lower intensity, the ionization can be neglected and only excitation to the two lowest 2p states, $2p\sigma_u$ and $2p\pi_u$, are significant. Once in the excited states, the H_2^+ may be further ionized by the laser field via tunneling ionization. Upon the second ionization, the two protons break apart and the kinetic energy release (KER) from the Coulomb explosion is characteristic of the initial breakup distance. Detailed description of the theoretical modelling of these elementary processes can be found in our earlier works [21, 22]. Basically we first used the molecular tunneling ionization (MO-ADK) theory [27] to calculate the ionization rates. The motion of the electron in the laser field is treated classically and the motion of the nuclear wave packet is followed quantum mechanically. From the calculated rescattering energy of the electron we used empirically fitted electron impact excitation cross sections to calculate the probability for exciting H_2^+ to the excited electronic states. From these excited states, H_2^+ is readily ionized by the laser which is again calculated using the MO-ADK theory. The internuclear distance when this happens is reflected in the kinetic energy of the ions at the end of Coulomb explosion.

Here we describe the modifications in order to incorporate the carrier-envelope phase into the simulation. For the long pulses, we only need to simulate tunneling ionization within one half-cycle of the laser and then follow the electron trajectory in the laser field for a few cycles. For the long pulses we can neglect the pulse shape as

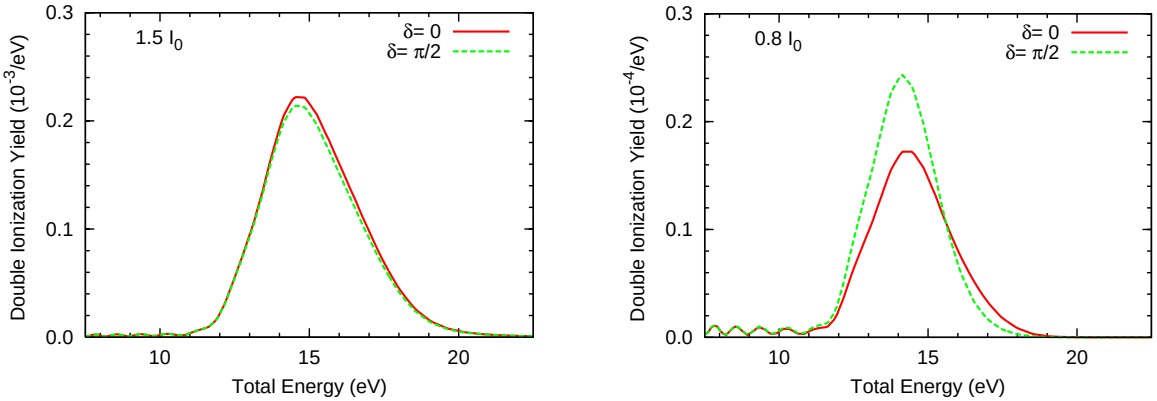

Figure 2. (Color online) Kinetic energy release spectra of the double ionization of H_2 in the 5 fs laser pulses with two different carrier-envelope phases. The laser intensities are (a) 1.5×10^{14} W/cm^2 (left panel) and (b) 0.8×10^{14} W/cm^2 (right panel).

well as the carrier-envelope phase. For few-cycle pulses we calculate the ionization from the whole pulse and for each ionization we follow its time evolution till the end of the pulse. For each simulation, the time-dependent electric field is explicitly used such that the carrier-envelope phase is fully incorporated. Consider a few-cycle pulse written as

$$E(t) = E_0 e^{-2\ln 2\ t^2/\tau^2} \cos(\omega t + \delta). \tag{1}$$

(Atomic units are used throughout the paper unless stated otherwise). Here, E_0 is the peak of the field envelope, τ the laser pulse duration (FWHM), ω the carrier frequency, and δ the carrier-envelope phase. Following equation (26) in [21] the kinetic energy release spectra $\frac{dP_{ion}(E,\delta)}{dE}$ is obtained, and the total double ionization yield can be calculated as

$$Y(\delta) = \int \frac{dP_{ion}(E,\delta)}{dE} dE. \tag{2}$$

To optimize the carrier-envelope phase dependence, ionization by the tail of the laser pulse should be minimized. By choosing molecules which are perpendicular to the laser polarization, charge resonance enhanced ionization (CREI) [28] is eliminated. This is accomplished experimentally by measuring the two protons in coincidence in the direction perpendicular to the laser field direction [24, 25, 29]. Such experiment can be done for phase stabilized few-cycle laser pulses too.

3. Results and discussion

Based on the above theory, we have calculated the kinetic energy release spectra of H_2 in few-cycle laser pulses. Figure 2 shows the typical KER spectra at two peak intensities for 5 fs pulses at two carrier-phases δ=0 and $\pi/2$. Clearly, the KER spectra are not sensitive to the CEP at the higher intensity (1.5×10^{14} W/cm^2). At the lower intensity (0.8×10^{14} W/cm^2), the KER yield shows dependence on the CEP. The KER yield is higher for $\delta = \pi/2$. To understand these results, we examine the energy distribution of the electrons that return to recollide with the H_2^+ core, see Fig. 3. Note that for intensity

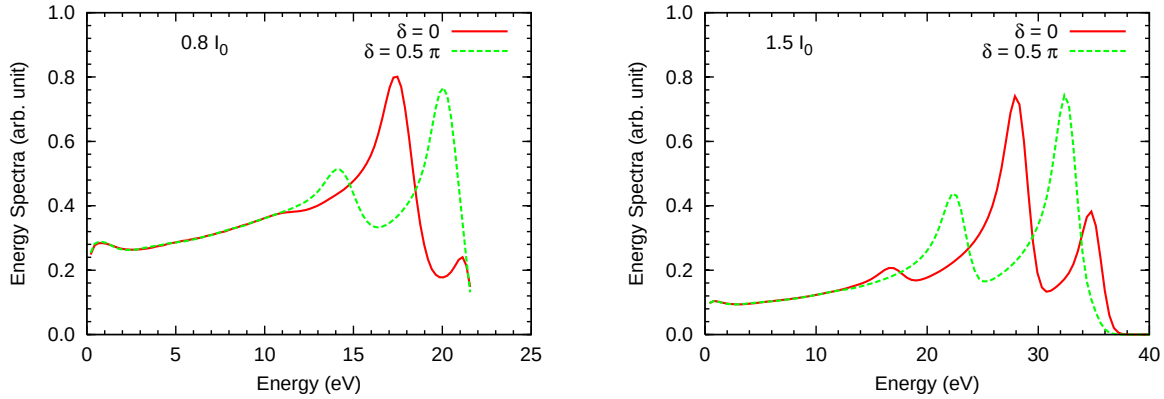

Figure 3. (Color online) The returning electron energy spectra for a 5 fs pulse at (a) 0.8×10^{14} W/cm^2 (left panel) and (b) 1.5×10^{14} W/cm^2 (right panel).

at 0.8×10^{14} W/cm^2, the electron energy distribution is dominated by a single peak at about 17 eV for $\delta = 0$. For $\delta = \pi/2$, the main peak has energy at 20 eV, with a minor one at 14 eV. The excitation energy from $1s\sigma_g$ to $2p\pi_u$ state for H_2^+ after the first return is about 13 eV at an internuclear distance of about 2 a.u. Since the excitation cross section is small near the threshold and rises rapidly with excessive energy, this explains the difference in the strength of the KER spectra for the two CEP's in Fig. 2(b) at 0.8×10^{14} W/cm^2. At the higher intensity of 1.5×10^{14} W/cm^2, on the other hand, the return energy of the electrons are much higher. For such higher energy electrons, the electron impact excitation cross sections are relatively flat, thus resulting in nearly identical yield in the KER spectra, as seen in Fig. 2(a). We comment that the kinetic energy quoted here refer to the electron energy when it is far away from the ion, as in the typical incident energy in electron-atom collisions.

The above explanation clearly demonstrates that the return energy of the electrons is responsible for the difference in the KER spectra. The knowledge of these energy distributions and their dependence on the CEP are important in their own right. Note that these returned electrons have been proposed for self imaging of molecules [23]. For this purpose, electrons that return to recollide with the molecule only once would be much easier to analyze. The electron spectra shown in Fig. 3 would favor 5 fs pulse with CEP chosen at $\delta = \pi/2$. On the other hand, for diffraction experiment, one would like to use higher energy electrons. At higher intensities the second peak (for a given CEP) becomes not negligible. However, these two peaks involve electrons "incident" from opposite sides and thus probably can be separated, especially for higher return energy since the electron scattering will be restricted to small angles.

To understand the calculated electron energy distribution, we make further analysis. Fig. 3(b) is replotted in Fig. 4(a), which is accompanied by Fig. 4(b) showing the electric fields for the two CEP's. First consider peak A_1 in Fig. 4(a). These electrons are generated by tunneling ionization when the laser field strength is near the maximum, labelled A_1 in Fig. 4(b). According to the rescattering model, the electrons that return to the core are those ionized at about 17° after the peak at A_1. These electrons, once

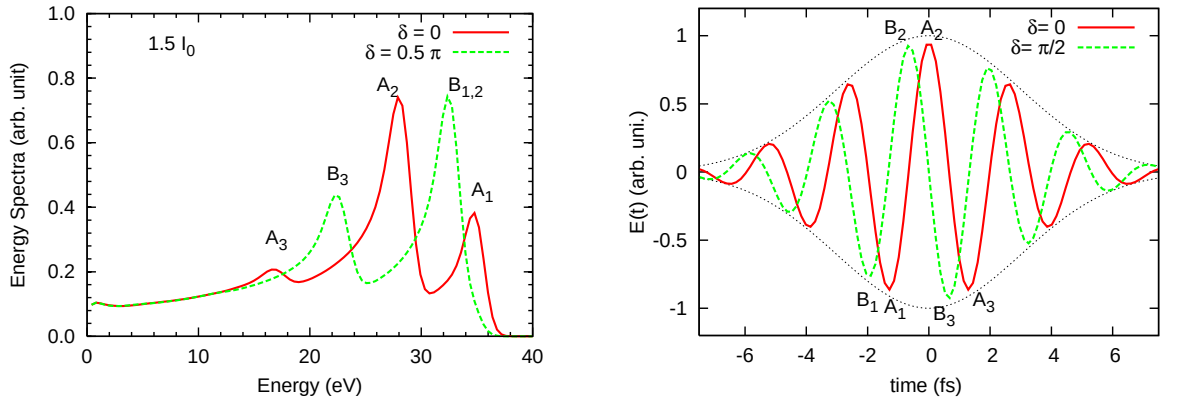

Figure 4. (Color online) (a) The returning electron energy spectra (left panel) and (b) time-dependent laser field strength (right panel) for I= 1.5×10^{14} W/cm^2 .

ionized, are accelerated in the oscillating electric field. The maximum kinetic energy they will gain depends strongly on the laser's electric field the next time it reaches the maximum. For electrons "born" near A_1, this occurs at the field near A_2. Back to Fig. 4a, the peak A_2 is due to electrons that are ionized near peak A_2 and accelerated by the field near A_3 in Fig. 4b. The field near A_3 is smaller than the field near A_2, thus the smaller returning energies. On the other hand the yield at A_2 of Fig. 4a is larger since the electrons are ionized by the larger electric field.

For $\delta=\pi/2$, the peak labelled as $B_{1,2}$ in Fig. 4a indicates that it is the sum of contributions of tunneling ionization from B_1 and B_2 in Fig. 4b. Note that electrons born near B_1 are accelerated by the electric field near B_2, and those born near B_2 are accelerated by the field near B_3. Since the field strengths at B_2 and B_3 are identical, the return energies are identical, except that they are in opposite directions. Since the electric field at B_2 and B_3 is smaller than that at A_2, the peak return energy for $\delta=\pi/2$ is smaller. By analyzing the subcycle dynamics, the energy distributions of the rescattering electrons and how they depend on the CEP can be understood.

For completeness, we show in Fig. 5 the calculated energy of the returned electrons for the 5 fs pulse at peak intensity of 0.8×10^{14} W/cm^2. At each CEP, there is only one dominant peak energy. The highest return energy occurs at near CEP=$\pi/2$. We next return to examine the CEP dependence of the non-sequential double ionization of H_2. In Fig. 6 we show the ratio of double ionization yield with respect to single ionization yield, for a 5 fs laser pulse for different peak intensities, to examine how the ratio depends on the CEP. Clearly at higher intensities the "normalized" ratio is very flat with respect to the CEP. Only at the lowest peak intensity of 0.8×10^{14} W/cm^2 do we see some variation of the ratio with respect to the CEP. It shows that nonsequential double ionization of H_2 can be used for determining the carrier-envelope phase only at low laser intensities. In this case the double to single ionization ratio is about 10^{-3} [21, 30] (Note that single ionization is essentially independent of CEP). For the lower peak intensity, shorter pulses would produce even stronger contrast and more accurate

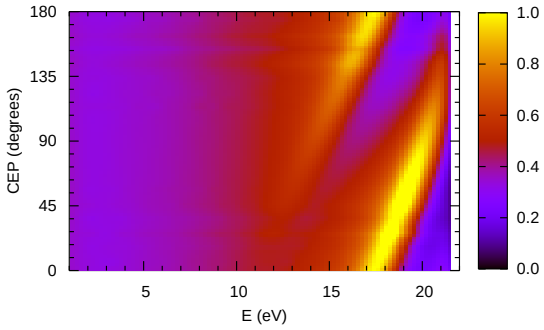

Figure 5. (Color online) The returning electron energy spectra for a 5 fs pulse at 0.8×10^{14} W/cm^2 vs the carrier envelope phase.

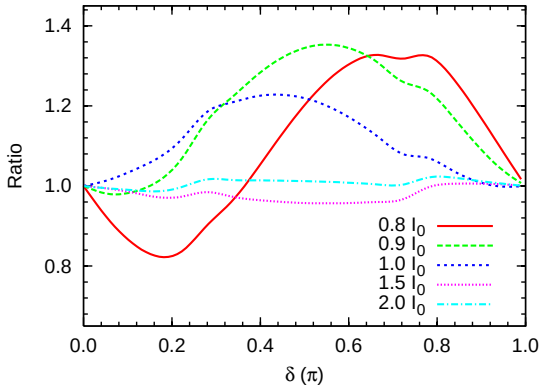

Figure 6. (Color online) CEP-dependence of normalized ratios of double ionization vs single ionization probabilities of H_2 by a 5 fs laser pulse with different laser intensities. The double ionization is initiated by the rescattering process. Here $I_0 = 10^{14}$ W/cm^2.

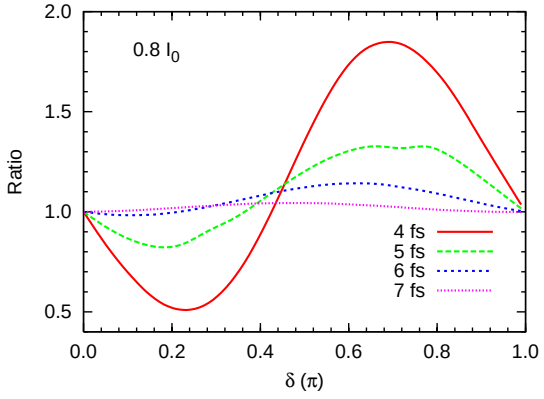

Figure 7. (Color online) Same as Fig. 6 but with a fixed laser intensity for different pulse durations.

determination of the CEP, as shown in Fig. 7 for peak intensity at 0.8×10^{14} W/cm^2 and laser pules ranging from 4 fs to 7 fs. Note that at lower intensities, on the other hand, the counting rate would be smaller.

So far, we have studied the CEP dependence of the KER yield of the double ionization process. In this case, we can not distinguish the field directions. In other

words, we can not distinguish carrier-envelope phase δ from $\pi + \delta$. Such distinction can be made if one looks at the dissociation channel, i.e., the yield of H^+ to the left, or to the right, as demonstrated in Kling *et al* for the dissociative ionization of D_2 in a CEP stabilized 5 fs pulse, and interpreted theoretically by us recently [26]. There are other ways to distinguish δ from $\pi + \delta$ if the inverse symmetry along the field polarization direction can be broken. For example, one can use H_2^+ ion beam, with the beam traveling along the direction of polarization [18].

4. Summary and Conclusions

In summary, we calculated the ratio of nonsequential double ionization vs single ionization of H_2 molecules with respect to the carrier-envelope phase of a few-cycle laser pulse. For short pulses we observed significant CEP dependence. For different carrier-envelope phases, electrons are ionized by tunneling at different field strengths and accelerated by the subsequent electric fields differently, thus resulting in different kinetic energies of the returned electrons. These different kinetic energies lead to different excitation cross sections. Once in the excited states, the molecular ions are subsequently ionized by the remaining electric field of the same pulse, and resulting in CEP dependent double ionization cross sections. No such measurements have been carried out as yet, but such measurement is expected to be straightforward once CEP stabilized pulses are available. The measurements would complement the experiment of Kling *et al* – to illustrate subcycle ionization dynamics of molecular ionization which can be controlled by varying the carrier-envelope phase. At present they serve as examples of phase control in laser-molecule interaction dynamics at the shortest time scale.

5. Acknowledgments

CDL was supported in part by the Chemical Sciences, Geosciences and Biosciences Division, Office of Basic Energy Sciences, Office of Science, US Department of Energy.

References

[1] Brabec T and Krausz F 2000 *Rev. Mod. Phys.* **72** 545
[2] Nisoli M *et al* 2003 *Phys. Rev. Lett.* **91** 213905
[3] Apolonski A *et al* 2000 *Phys. Rev. Lett.* **85** 740
[4] Apolonski A *et al* 2004 *Phys. Rev. Lett.* **92** 073902
[5] Baltuska A *et al* 2003 *Nature* **421** 611
[6] Paulus G G *et al* 2001 *Nature* **414** 182
[7] Paulus G *et al* 2003 *Phys. Rev. Lett.* **91** 253004
[8] Liu X and de Morisson Faria C F 2004 *Phys. Rev. Lett.* **92** 133006
[9] Kling M F *et al* 2006 *Science* **312** 246
[10] Chelkowski S and Bandrauk A D 2002 *Phys. Rev. A* **65** 061802
[11] Bandrauk A D, Chelkowski S and Shon N H 2002 *Phys. Rev. Lett.* **89** 283903
[12] Chelkowski S, Bandrauk A D and Apolonski A 2004 *Phys. Rev. A* **70** 013815
[13] Milosevic D B, Paulus G G and Becker W 2002 *Phys. Rev. Lett.* **89** 153001

[14] Milosevic D B, Paulus G G and Becker W 2003 *Optics Express* **11** 1418
[15] Milosevic D B, Paulus G G and Becker W 2005 *Phys. Rev. A* **71** 061404(R)
[16] Tong X M, Hino K and Toshima N 2006 *Phys. Rev. A* **74** 031405(R)
[17] Becker A and Faisal F H M 2005 *J. Phys. B* **38** R1
[18] Roudnev V, Esry B D and Ben-Itzhak I 2004 *Phys. Rev. Lett.* **93** 163601
[19] Kamta G L and Bandrauk A D 2005 *Phys. Rev. Lett.* **94** 203003
[20] Lemell C, Tong X M, Krausz F and Burgdorfer J 2003 *Phys. Rev. Lett.* **90** 076403
[21] Tong X M, Zhao Z X and Lin C D 2003 *Phys. Rev. A* **68** 043412
[22] Tong X M, Zhao Z X and Lin C D 2003 *Phys. Rev. Lett.* **91** 233203
[23] Niikura H *et al* 2003 *Nature* **421** 826
[24] Alnaser A S *et al* 2003 *Phys. Rev. Lett.* **91** 163002
[25] Alnaser A S *et al* 2004 *Phys. Rev. Lett.* **93** 183202
[26] Tong X M and Lin C D 2006 *Phys. Rev. Lett.* (submitted)
[27] Tong X M, Zhao Z X and Lin C D 2002 *Phys. Rev. A* **66** 033402
[28] Zuo T and Bandrauk A D 1995 *Phys. Rev. A* **52** R2511
[29] Niikura H *et al* 2002 *Nature* **417** 917
[30] Yudin G L and Ivanov M Y 2001 *Phys. Rev. A* **63** 033404