

# Carrier-envelope phase dependence of nonsequential double ionization of H<sub>2</sub> by few-cycle laser pulses

X M Tong<sup>1</sup> and C D Lin<sup>2</sup>

<sup>1</sup> Institute of Materials Science, Graduate School of Pure and Applied Sciences, Center for Computational Sciences, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8573, Japan

<sup>2</sup> Physics Department, Kansas State University, Manhattan, KS 66506-2601, USA

Received 25 October 2006, in final form 6 December 2006

Published 24 January 2007

Online at [stacks.iop.org/JPhysB/40/641](http://stacks.iop.org/JPhysB/40/641)

## Abstract

We studied the dependence of double ionization of H<sub>2</sub> 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.

## 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)  $\delta$ . Since the feedback control of 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. For intensity of the order of  $10^{12}$  W cm<sup>-2</sup> or lower, ionization from metallic surfaces [4, 8] has been shown to depend on the CEP as well. While these experiments demonstrated how the results vary with the CEP of the few-cycle pulse, the determination of its actual value has to rely on the prediction of theoretical calculations.

Theoretically the dependence of ATI spectra on the carrier-envelope phase of a few-cycle pulse for atomic targets has been studied in many papers recently [9–15]. The spectra were calculated either by directly solving the time-dependent Schrödinger equation, or by using the *S*-matrix theory [16]. For molecular targets the carrier-envelope phase dependence of the dissociation of one-electron molecular ions in a few-cycle laser pulse has also been studied by solving the time-dependent Schrödinger equation in [17, 18].

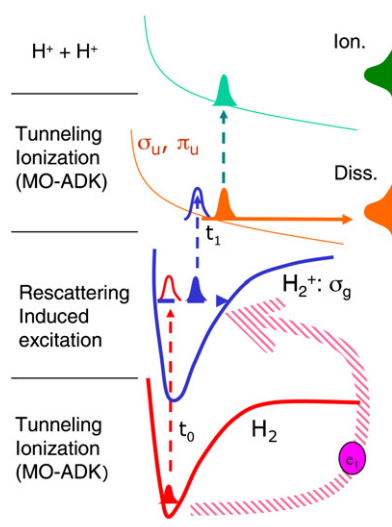
To optimize the contrast of the CEP dependence it is clear that one would employ high-energy electrons generated in laser–matter interactions directly (as in ATI electrons) or indirectly (as in HHG). While both HHG and ATI from atoms are considered to be well understood, the yields of high-energy electrons and higher HHG are many orders of magnitude smaller and they are difficult to calculate accurately. Thus other processes initiated by high-energy electrons are of interest. For example, the nonsequential double ionization of Ar by few-cycle pulses has been experimentally demonstrated to depend on the carrier-envelope phase by Liu *et al* [19]. While it is in principle possible to solve the time-dependent Schrödinger equation of the two-electron systems directly to predict the carrier-envelope dependence of the double ionization yields, in reality such calculations are very difficult and results have been reported only for two-electron systems with reduced dimensions or with very short pulse durations [20, 21].

In a recent paper, Kling *et al* [22] reported the dissociative ionization of D<sub>2</sub> molecules in a few-cycle laser pulse with duration of about 5 fs. They found that the D<sup>+</sup> ions are ejected asymmetrically along the laser polarization direction and the asymmetry depends on the carrier-envelope phase of the laser. In order to interpret their measurement, we have modified a previously developed rescattering theory that was used for describing the nonsequential double ionization and dissociative ionization of H<sub>2</sub> and D<sub>2</sub> molecules in a long pulse [23, 24]. We were able to show that the asymmetry of the D<sup>+</sup> ejection is a consequence of tunnelling ionization and rescattering occurring within one optical cycle [25], and the nature of the electric field within this optical cycle depends critically on the carrier-envelope phase of the laser pulse. In the present paper, we report that the yield of the nonsequential double ionization of D<sub>2</sub> (or H<sub>2</sub>) molecules also depends on the carrier-envelope phase. Such experiments have not been carried out yet, but the same experimental methods for studying the nonsequential double ionization for longer pulses can be used for such measurement once CEP-stabilized short laser pulses become available. We note that for pulses of durations of 25–40 fs the kinetic energy release spectra of D<sup>+</sup> (or H<sup>+</sup>) have been determined in previous experiments [26–28], and the results have been interpreted by the rescattering theory developed in [23, 24].

The paper is organized as follows. A brief review of the physical processes and our theoretical model is given in section 2. The theoretical results are presented and analysed in section 3. We have also examined how the energies of the rescattered electrons depend on the carrier-envelope phase which is 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<sub>2</sub>

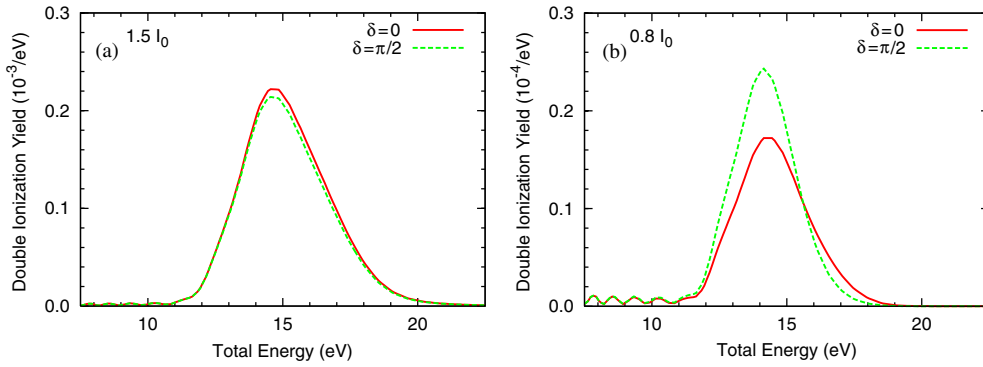
Figure 1 depicts schematically the physical processes in the nonsequential double ionization of H<sub>2</sub> in the laser field. Note that the present theory can be directly applied to D<sub>2</sub>. The detailed theoretical modelling has been described previously for the longer pulses [23, 24]. Here we summarize the basic elements in the model and the modification for sub-10 fs pulses. In the nonsequential double ionization of H<sub>2</sub>, the laser field first ionizes H<sub>2</sub> to H<sub>2</sub><sup>+</sup> via tunnelling 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<sub>2</sub><sup>+</sup> to ionize it or to excite it to higher electronic states. Once in the excited states, the H<sub>2</sub><sup>+</sup> may be further ionized by the laser via tunnelling 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, where the KER can be determined experimentally.



**Figure 1.** Schematic of the major processes leading to Coulomb breakup of the H<sub>2</sub> molecule in an intense laser field.

In the simulation, at time  $t_0$  near the peak of the laser's electric field, an electron is released. The ionization rate at each instantaneous electric field is treated using the static molecular tunnelling ionization theory (MO-ADK) [29]. We assumed that the electron released from tunnelling ionization initially has a Gaussian velocity distribution [30]. For each initial condition the trajectory of the ionized electron in the laser field and the potential of the H<sub>2</sub><sup>+</sup> ion is calculated. By monitoring the distance of closest approach of the electron, the return time  $t_1$  and the kinetic energy for each trajectory are recorded. Using semi-empirically fitted excitation cross sections, the excitation probability density by a rescattering electron at the time  $t_1$  of collision is calculated, for H<sub>2</sub><sup>+</sup> at the internuclear distances  $R$  [23]. This probability is weighted by the density of the vibrational wave packet  $|\chi(R, t_0, t_1)|^2$ , where  $\chi(R, t_0, t_1)$  is the wave packet at time  $t_1$  that was created at time  $t_0$ , assuming that Franck–Condon principle is applicable in the ionization process. From these excited states, H<sub>2</sub><sup>+</sup> is readily ionized by the laser which is again calculated using the MO-ADK theory.

An essential step in this rescattering modelling is the calculation of excitation and ionization probabilities of H<sub>2</sub><sup>+</sup> ions by the returning electrons. These probabilities are needed for a range of internuclear separations, for the different alignment of the H<sub>2</sub><sup>+</sup> ions, for different excited states, and for the whole range of energies of the returned electrons. Such scattering cross sections are not available. On the other hand, electron impact excitation and ionization cross sections are available in the united-atom limit, He<sup>+</sup>, and the separated-atom limit, H. By fitting these cross sections to some analytical form with electron energies given in units of excitation energies, semi-empirical cross sections for excitation and ionization of H<sub>2</sub><sup>+</sup> at any internuclear separation and alignment have been obtained. The detailed procedure can be found in [23]. Note that if the return energy is large, ionization by the returning electron is possible and such processes are included in the simulation as well. Note also that the fitted cross sections are for electron impact excitation or ionization without the laser field. The 'incident' impact energy of the electron is obtained by solving the classical equation of motion backward from the position and velocity of the rescattering electron at time  $t_1$  without the laser field. The energy at large distance then is the 'incident' energy of the electron in electron impact excitation of H<sub>2</sub><sup>+</sup> without the laser field.



**Figure 2.** Kinetic energy release spectra of the double ionization of  $\text{H}_2$  molecules in a 5 fs laser pulse with two different carrier-envelope phases. The peak laser intensities are (a)  $1.5 \times 10^{14} \text{ W cm}^{-2}$  (left panel) and (b)  $0.8 \times 10^{14} \text{ W cm}^{-2}$  (right panel).

Previously to simulate nonsequential double ionization processes for long pulses, we only needed to consider tunnelling ionization within one half-cycle of the laser and then follow the electron trajectory in the laser field for a few cycles. For few-cycle pulses we calculate the ionization from the whole pulse and for each ionization we follow its time evolution until the end of the pulse. In 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

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

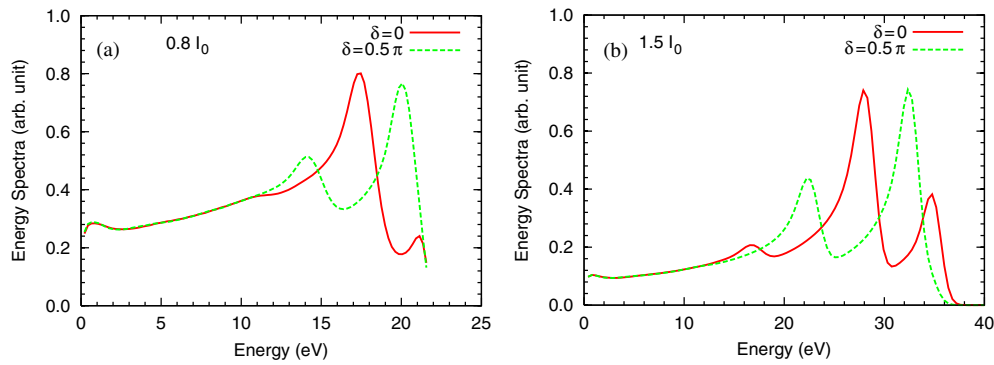
(Atomic units are used throughout the paper unless stated otherwise.) Here, we assume that the temporal intensity profile is Gaussian with pulse duration  $\tau$  (FWHM).  $E_0$  is the peak of the field envelope,  $\omega$  the carrier frequency, and  $\delta$  the carrier-envelope phase. Following equation (26) in [23] the kinetic energy release spectra  $\frac{dP_{\text{ion}}(E, \delta)}{dE}$  is obtained, and the total double ionization yield can be calculated as

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

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) [31] is eliminated. This is accomplished experimentally by measuring the two protons in coincidence in the direction perpendicular to the laser field direction [27, 28, 32]. Such an 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  $\text{H}_2$  in few-cycle laser pulses. Figure 2 shows the typical KER spectra at two peak intensities for 5 fs pulses at two carrier phases  $\delta = 0$  and  $\pi/2$ . Clearly, the KER spectra are not sensitive to CEP at the higher intensity ( $1.5 \times 10^{14} \text{ W cm}^{-2}$ ). At the lower intensity ( $0.8 \times 10^{14} \text{ W cm}^{-2}$ ), the KER yield shows dependence on 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  $\text{H}_2^+$  core, see figure 3. Note that for intensity of  $0.8 \times$

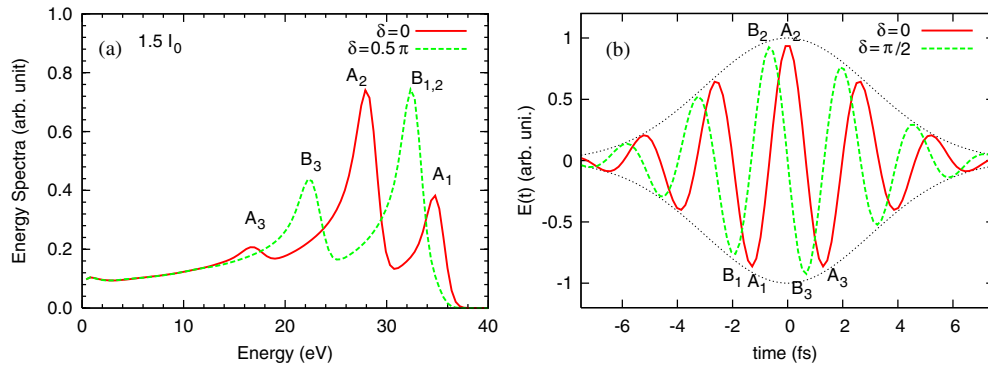


**Figure 3.** The returning electron energy spectra for a 5 fs pulse at (a)  $0.8 \times 10^{14} \text{ W cm}^{-2}$  (left panel) and (b)  $1.5 \times 10^{14} \text{ W cm}^{-2}$  (right panel).

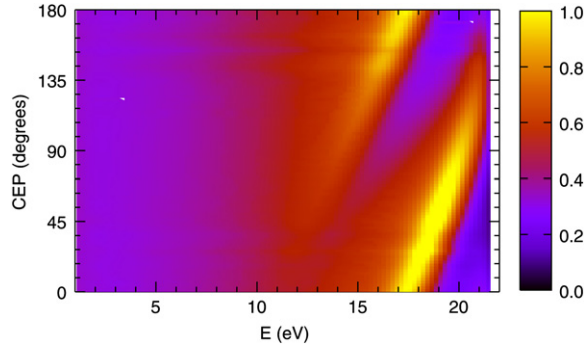
$10^{14} \text{ W cm}^{-2}$ , the electron energy distribution is dominated by a single peak at about 17 eV for  $\delta = 0$ . For  $\delta = \pi/2$ , the main peak has energy of 20 eV, with a minor one at 14 eV. Note that due to the Coulomb focusing effect, the simulated returning energy is higher than  $3.2U_p$ , with  $U_p = I/(4\omega^2)$  the ponderomotive energy. The excitation energy from  $1s\sigma_g$  to  $2p\pi_u$  state for  $\text{H}_2^+$  after the first return is about 13 eV at an internuclear distance of 2 au. 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 figure 2(b) at  $0.8 \times 10^{14} \text{ W cm}^{-2}$ . At the higher intensity of  $1.5 \times 10^{14} \text{ W cm}^{-2}$ , the return energy of the electrons is much higher. For such higher energy electrons, the energy dependence of electron impact excitation cross sections is relatively flat, thus resulting in nearly identical yield in the KER spectra, as seen in figure 2(a). Recall that the kinetic energy quoted here refers to the electron energy as in the typical incident energy in electron–ion collisions. The impact excitation cross sections of  $\text{H}_2^+$  ions by these electrons without the laser field are taken to be the electron impact excitation cross sections by the rescattering electrons in the laser field.

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 CEP are important in their own right. Note that these returned electrons have been proposed for self imaging of molecules [26]. For this purpose, electrons that return to recollide with the molecule only once would be much easier to analyse. The electron spectra shown in figure 3 would favour a 5 fs pulse with CEP chosen at  $\delta = \pi/2$ . On the other hand, for diffraction experiments, one would like to use higher energy electrons. At higher intensities the electrons from the second peak (for a given CEP) would contribute to the rescattering to introduce interference in the electron spectra, making the analysis more complicated. Furthermore, with even higher energies the sequential double ionization process is more important [33, 34] and rescattering becomes insignificant for double ionization.

To understand the calculated electron energy distribution, we make further analysis. Figure 3(b) is replotted in figure 4(a), which is accompanied by figure 4(b) showing the electric fields for the two CEP's. First consider peak A<sub>1</sub> in figure 4(a). These electrons are generated by tunnelling ionization when the laser field strength is near the maximum, labelled A<sub>1</sub> in figure 4(b). According to the rescattering model, the electrons that return to the core with the maximum kinetic energy are those ionized at about  $17^\circ$  after the peak at A<sub>1</sub>. These electrons, once 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



**Figure 4.** (a) The returning electron energy spectra (left panel) and (b) time-dependent laser field strength (right panel) for  $11.5 \times 10^{14} \text{ W cm}^{-2}$ , at two carrier-envelope phases.

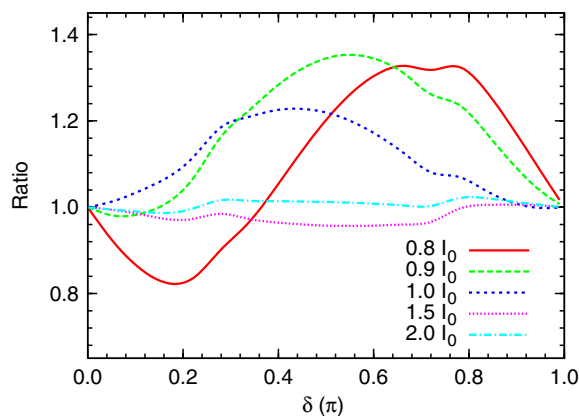


**Figure 5.** The returning electron energy spectra for a 5 fs pulse at  $0.8 \times 10^{14} \text{ W cm}^{-2}$  versus the carrier envelope phase.

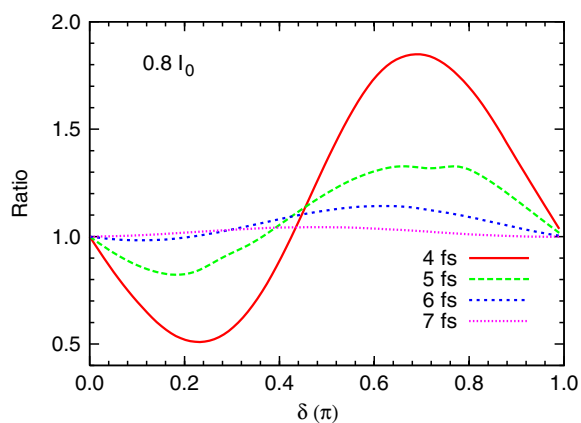
the maximum. For electrons ‘born’ near  $A_1$ , they are accelerated by the field near  $A_2$ . In figure 4(a), the peak  $A_2$  is due to electrons that are ionized near peak electric field at  $A_2$  and accelerated by the field near  $A_3$  in figure 4(b). This explains why in figure 4(a), the peak  $A_1$  has higher energy but smaller yield, but the peak  $A_2$  has more yield but lower energy. The tunnelling ionization rate is larger for peak  $A_2$  than for  $A_1$  due to its larger electric field at  $A_2$ .

For  $\delta = \pi/2$ , the peak labelled as  $B_{1,2}$  in figure 4(a) indicates that it is the sum of contributions of tunnelling ionization from  $B_1$  and  $B_2$  in figure 4(b). 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 analysing the subcycle dynamics, the energy distributions of the rescattering electrons and how they depend on CEP can be understood.

For completeness, we show in figure 5 the calculated energy of the returned electrons for a 5 fs pulse at peak intensity of  $0.8 \times 10^{14} \text{ W cm}^{-2}$ . At each CEP, there is only one dominant peak energy. The highest return energy occurs near  $\text{CEP} = \pi/2$ . We next return to examine the CEP dependence of the non-sequential double ionization of  $\text{H}_2$ . In figure 6 we show the ratio of double ionization yield with respect to single ionization yield, for a 5 fs



**Figure 6.** CEP-dependence of normalized ratios of double ionization versus single ionization probabilities of H<sub>2</sub> by a 5 fs laser pulse with different laser intensities. The double ionization is initiated by the rescattering process. Here  $I_0 = 10^{14} \text{ W cm}^{-2}$ .



**Figure 7.** Same as figure 6 but with a fixed laser intensity for different pulse durations.

laser pulse for different peak intensities, to examine how the ratio depends on CEP. Clearly at higher intensities the ‘normalized’ ratio is very flat with respect to CEP. Only at a lowest peak intensity of  $0.8 \times 10^{14} \text{ W cm}^{-2}$  do we see some variation of the ratio with respect to CEP. It shows that nonsequential double ionization of H<sub>2</sub> 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  $10^{-3}$  [23, 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 determination of CEP, as shown in figure 7 for peak intensity at  $0.8 \times 10^{14} \text{ W cm}^{-2}$  and laser pulses 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 cannot distinguish the field directions. In other words, we cannot distinguish carrier-envelope phase  $\delta$  from  $\pi + \delta$ . Such distinction can be made if one looks at the dissociation channel, i.e., the yield of H<sup>+</sup> to the left, or to the right, as demonstrated in Kling *et al* for the dissociative ionization of D<sub>2</sub> in a CEP-stabilized 5 fs pulse, and theoretically



interpreted by us recently [25]. There are other ways to distinguish  $\delta$  from  $\pi + \delta$  if the inversion symmetry along the field polarization direction can be broken. For example, one can use an  $H_2^+$  ion beam, with the beam travelling along the direction of polarization [17].

#### 4. Summary and conclusions

In summary, we calculated the ratio of nonsequential double ionization versus 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 tunnelling 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 result 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 work of Kling *et al*—to illustrate subcycle 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.

#### 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] Lemell C, Tong X M, Krausz F and Burgdorfer J 2003 *Phys. Rev. Lett.* **90** 076403
- [9] Chelkowski S and Bandrauk A D 2002 *Phys. Rev. A* **65** 061802
- [10] Bandrauk A D, Chelkowski S and Shon N H 2002 *Phys. Rev. Lett.* **89** 283903
- [11] Chelkowski S, Bandrauk A D and Apolonski A 2004 *Phys. Rev. A* **70** 013815
- [12] Milosevic D B, Paulus G G and Becker W 2002 *Phys. Rev. Lett.* **89** 153001
- [13] Milosevic D B, Paulus G G and Becker W 2003 *Opt. Express* **11** 1418
- [14] Milosevic D B, Paulus G G and Becker W 2005 *Phys. Rev. A* **71** 061404
- [15] Tong X M, Hino K and Toshima N 2006 *Phys. Rev. A* **74** 031405
- [16] Becker A and Faisal F H M 2005 *J. Phys. B: At. Mol. Opt. Phys.* **38** R1
- [17] Roudnev V, Esry B D and Ben-Itzhak I 2004 *Phys. Rev. Lett.* **93** 163601
- [18] Kamta G L and Bandrauk A D 2005 *Phys. Rev. Lett.* **94** 203003
- [19] Liu X *et al* 2004 *Phys. Rev. Lett.* **93** 263001
- [20] Bandrauk A D and Lu H Z 2005 *Phys. Rev. A* **72** 023408
- [21] Baier S, Ruiz C, Plaia L and Becker A 2006 *Phys. Rev. A* **74** 033405
- [22] Kling M F *et al* 2006 *Science* **312** 246
- [23] Tong X M, Zhao Z X and Lin C D 2003 *Phys. Rev. A* **68** 043412
- [24] Tong X M, Zhao Z X and Lin C D 2003 *Phys. Rev. Lett.* **91** 233203
- [25] Tong X M and Lin C D 2006 *Phys. Rev. Lett.* (submitted)
- [26] Niikura H *et al* 2003 *Nature* **421** 826



- [27] Alnaser A S *et al* 2003 *Phys. Rev. Lett.* **91** 163002
- [28] Alnaser A S *et al* 2004 *Phys. Rev. Lett.* **93** 183202
- [29] Tong X M, Zhao Z X and Lin C D 2002 *Phys. Rev. A* **66** 033402
- [30] Yudin G L and Ivanov M Y 2001 *Phys. Rev. A* **63** 033404
- [31] Zuo T and Bandrauk A D 1995 *Phys. Rev. A* **52** R2511
- [32] Niikura H *et al* 2002 *Nature* **417** 917
- [33] Tong X M and Lin C D 2004 *Phys. Rev. A* **70** 023406
- [34] Legare F *et al* 2003 *Phys. Rev. Lett.* **91** 093002