Carrier Envelope Phase (CEP) dependence of double ionization in molecules at high laser intensities – 2-D model for H_2 and HeH^+.

by

André D. Bandrauk*, Huizhong Lu
Laboratoire de chimie théorique
Université de Sherbrooke (Québec) J1K 2R1

***Canada Research Chair**
email: andre.bandrauk@USherbrooke.ca

Recent advances in ultrashort intense laser pulse technology allows for unprecedented study of laser matter interaction on the time scale of a few optical cycles [1]. For few cycle pulses the electric field, $E(t)=E_0(t)\cos(\omega t+\phi)$ depends strongly on the phase ϕ of the carrier wave of frequency ω with respect to the pulse envelope $E_0(t)$. ϕ is called the carrier envelope phase, CEP. Spatiotemporal variation of electromagnetic pulses E(t) consisting of very few cycles are now precisely known and can be shaped with attosecond precision via control over ϕ.

We have previously demonstrated by numerical simulations that intense few cycle laser pulses produce asymmetries in strong-field ionization of one electron atomic 3-D model systems [2] and that such asymmetry follows a universal CEP dependence which can be used to measure the duration of subfemtosecond pulses [3]. We investigate in the present work double ionization in a 2-D model of H_2 and HeH^+ and its dependence on CEP effects. The 2-D model allows for calculation of the angular dependence of the double electron ejection by intense 800 nm laser pulses in molecules and to compare to recent experimental results in Ar atoms [4]. Comparisons are also made at large internuclear distances where Charge Resonance Enhanced Ionization, CREI dominates [5].

[1] T. Brabec, F. Krausz, Rev. Mod. Phys. 72, 545 (2000).
[2] S. Chelkowski, A.D. Bandrauk, A. Apolonski, Phys. Rev. A71, 053815 (2005); Opt. Lett. 29, 1557 (2004).
[3] A.D. Bandrauk, S. Chelkowski, H.S. Nguyen, Phys. Rev. A68, 041802 (2003).
[4] X. Liu et al., Phys. Rev. Lett. 93, 263001 (2004).
[5] G. Lagmago Kamta, A.D. Bandrauk, Phys. Rev. Lett. 94, 203003 (2005).

1

.org
the online photonics resource

home | buyer's guide | news | employment | resources | feature articles | events | new products | OLE magazine | contact us | find | advanced site search | IoP

news

Browse the archive
2004 August
Show summaries
Go

quick search
Search the archive
Find

news

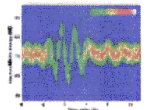
<< previous article more articles next article >>

Looking inside a laser pulse

31 August 2004

Physicists have directly measured the electric field of a light pulse for the first time.

The oscillation of the electric field in a laser pulse has been measured for the first time by physicists in Austria and Germany. The technique could be used to study ultrafast dynamics in atoms and molecules (*Science* **305** 1267).



Light pulse

Ferenc Krausz and co-workers at the University of Vienna, the University of Bielefeld and the Max Planck Institute for Quantum Optics sent an extreme-ultraviolet laser pulse with a duration of just 250 attoseconds (250 x 10^{-18} s) into a gas of neon atoms, along with the longer femtosecond (10^{-15} s) pulse that they wanted to measure. This second pulse contains only a few cycles of the electromagnetic field. The attosecond pulse ionises the neon atoms, and the electrons that are released are then accelerated by the electric field of the longer pulse. The duration of the electron bunch is much shorter that the timescale over which the electric field of the femtosecond pulse changes.

The energy of the accelerated electrons - which can be measured with a spectrometer - depends on the strength of the electric field in the femtosecond pulse. By varying the relative timing of the two pulses and measuring how the electron energy changes, it is possible to build up a picture of the electric field in the longer pulse. The method reveals that the light pulse has a duration of 4.3 femtoseconds.

"Our technique can measure the dynamic evolution and the exact value of the electric field for few-cycle light

NewsAlert
Sign up to our FREE news alerting service or, if you are already a subscriber, you can update or unsubscribe

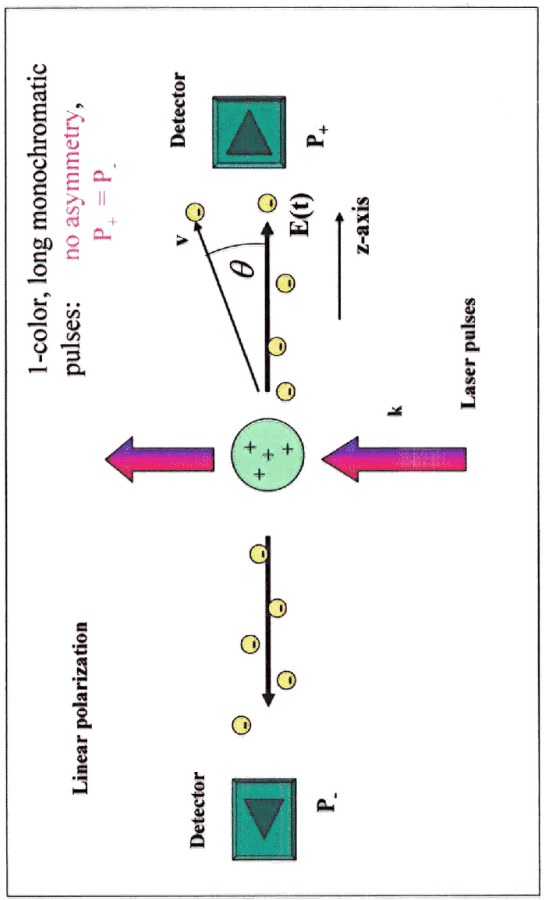
links
User comments
Add a comment

Related Links
Ferenc Krausz homepage
Photonics at the University of Vienna
Max Planck Institute of Quantum Optics

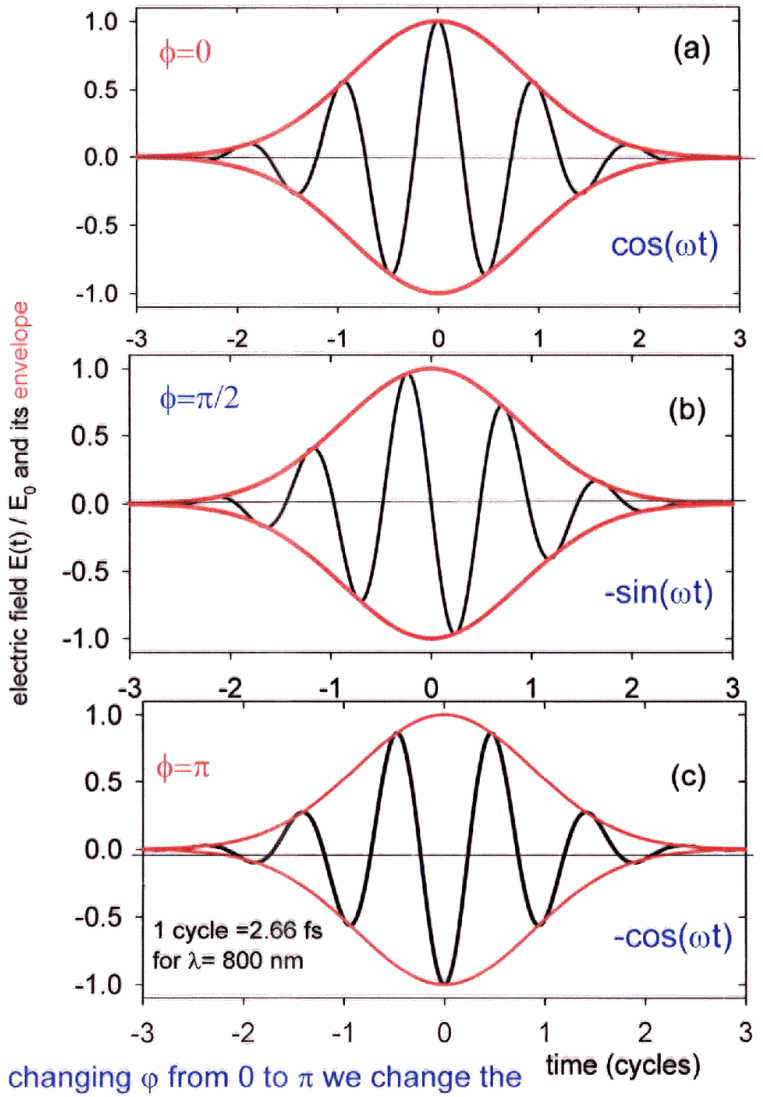
Restricted Links
Science **305** 1267

Related Stories
First light for attophysics (October 2002)
Attosecond pulses "see" light waves (November 2001)

2



Long, 1-color pulses: angular distributions are symmetric, $f(\theta)=f(\pi-\theta)$, i.e. $P_+ = P_-$, e.g. n-photon absorption leads to odd or even states, 1-photon: $f-\cos^2(\theta)$



By changing φ from 0 to π we change the direction of photoemission

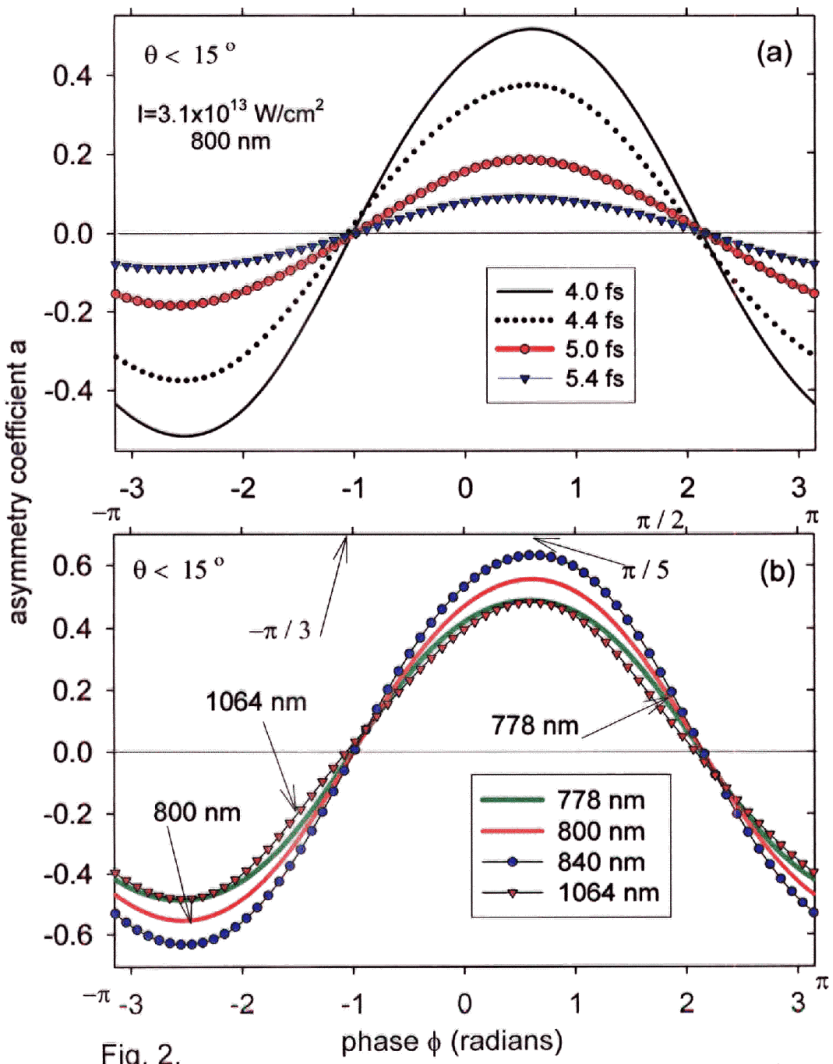
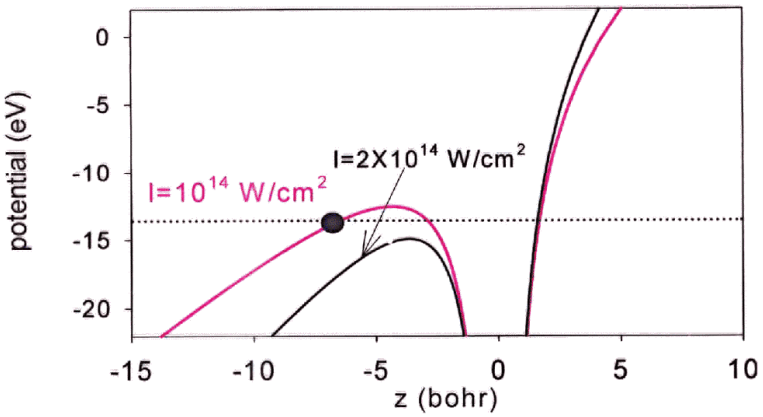
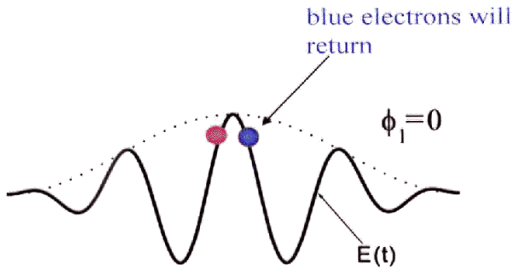


Fig. 2.





E(t) is symmetric. Newton equations:

$$\frac{dv}{dt}=\frac{1}{c}\frac{\partial A}{\partial t} \rightarrow v_f=-\frac{1}{c}A(t_0)=\frac{\varepsilon_0(t_0)}{\omega}\sin(\omega t_0+\varphi)$$

if $\varphi=0$, $v_f(-t_0)=-v_f(t_0) \Rightarrow$ symmetric photoelectrons

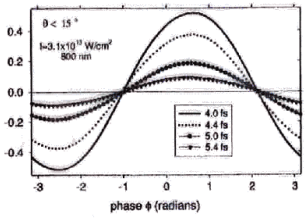


Figure 31. The asymmetry coefficient $a(\phi)$ as a function of the absolute phase ϕ for the laser intensity $I = 3.1 \times 10^{13}$ W cm^{-2} and $\lambda = 800$ nm for various pulse durations as given in the inset, obtained from a solution of the TDSE. From [61].

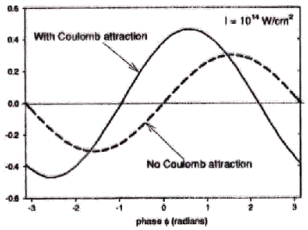


Figure 32. The asymmetry coefficient $a(\phi)$ from a semiclassical model with (solid line) and without (dashed line) the Coulomb attraction by the ion, as a function of the CE phase ϕ, for the laser intensity $I = 10^{14}$ W cm^{-2} and pulse duration of 3.9 fs. From [66].

Since $E(t_+) = E(t_-)$, the quasistatic emission rates at these two times are identical provided the envelope functions at these two times have identical values (as is the case for envelope 1). The lower part of figure 33 shows the electron trajectories starting at t_+ and t_- calculated from Newton's equation in the absence of the Coulomb field. Initially, they take off in the same direction. However, the one that departs at the later time t_- turns around after the field $E(t)$ has changed sign. Thereby, this trajectory returns to the ion and experiences (in the spacetime region shown shaded) strong Coulomb attraction by the ion [112], in contrast to the electron that was set free at the earlier time t_+. The Coulomb field will deflect the electron from its original direction. Since the electronic wave packet can be envisaged as a divergent bundle of trajectories, the net effect will be a focusing in the direction of the laser polarization. This destroys the original b-f symmetry for the case of the envelope 1, which has the same value at the two times t_+ and t_- and corresponds to the cosine pulse. In order to compensate the

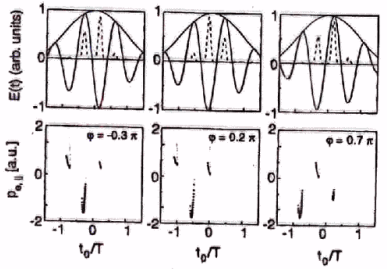
W. Becker et al, J. Phys B 39, R203-262 (2006).

Nonsequential Double Ionization at the Single-Optical-Cycle Limit

. Liu,[1] H. Rottke,[1] E. Eremina,[1] W. Sandner,[1] E. Goulielmakis,[2] K. O. Keeffe,[2] M. Lezius,[2] F. Krausz,[2,3] F. Lindner,[3] M. G. Schätzel,[3] G. G. Paulus,[4,5] and H. Walther[3,4]

[1]*Max-Born-Institut, Max-Born-Strasse 2a, D-12489 Berlin, Germany*
[2]*Institut für Photonik, Technische Universität Wien, Gusshausstrasse 27, A-1040 Wien, Austria*
[3]*Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Strasse 1, D-85748 Garching, Germany*
[4]*Ludwig-Maximilians-Universität München, Am Coulombwall 1, D-85748 Garching, Germany*
[5]*Department of Physics, Texas A&M University, College Station, Texas 77843-4242, USA*

(Received 15 June 2004; published 20 December 2004)



G. 3. Upper panel: electric field $E(t)$ (solid line) and the iasistatic field ionization rate [18] (dashed line) for a four-cycle ser pulse (760 nm center wavelength, 3.5×10^{14} W/cm^2 peak tensity). Lower panel: classically accessible final phase space on recollision ($p_{1\parallel} = p_{2\parallel} = p_{e,\parallel}$) for electrons liberated at nneling time t_0 (in units of the optical period $T = 2.6$ fs). φ notes the corresponding CE phase.

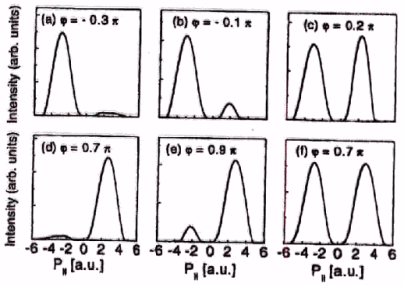


FIG. 4. Numerical simulation of the ion momentum distribution induced by few-cycle pulses at different CE phases. A four-cycle laser pulse is used in the calculation for (a)–(e) and an eight-cycle pulse in (f). The other parameters are the same as in Fig. 3.

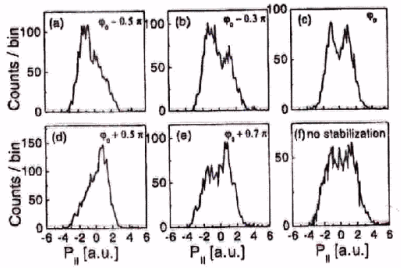


FIG. 2. Experimental momentum distribution of Ar^{2+} ions from NSDI of Ar at different CE phases φ. The laser pulse parameters are ≈ 5 fs pulse width and ≈ 350 TW/cm^2 peak light intensity. The distributions are integrated over the Cartesian momentum components perpendicular to the light beam polar-

Centre de Recherches Mathématiques
CRM Proceedings and Lecture Notes

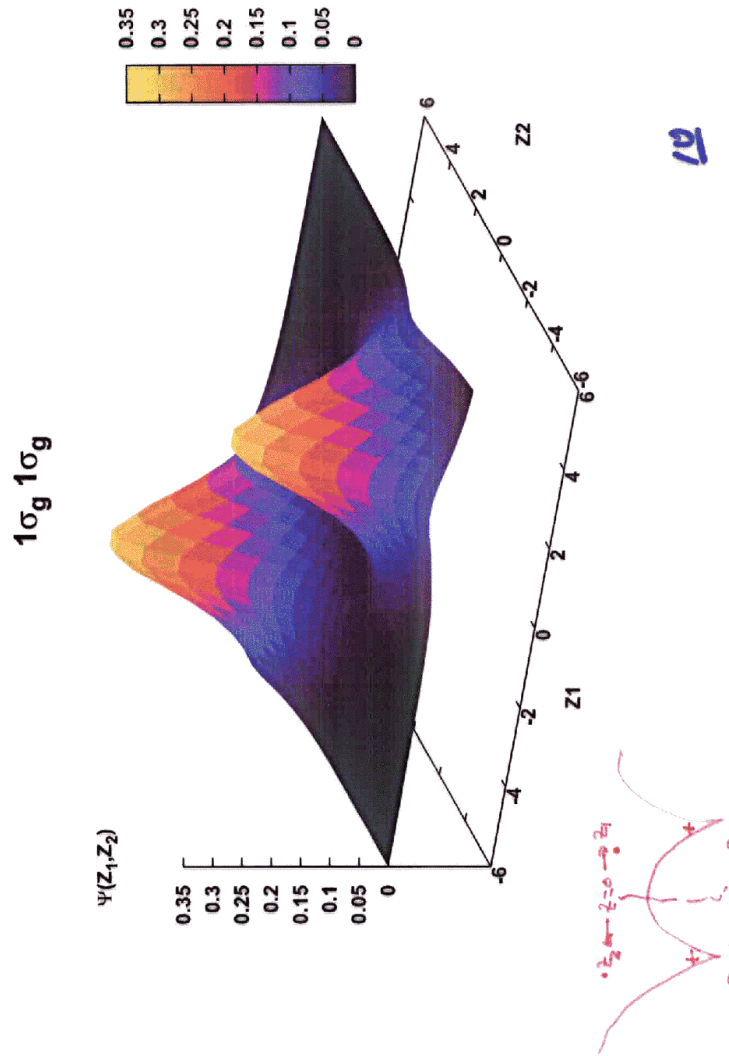
Singularity-Free Methods for the Time-Dependent Schroedinger Equation for Nonlinear Molecules in Intense Laser Fields — A Non-Perturbative Approach

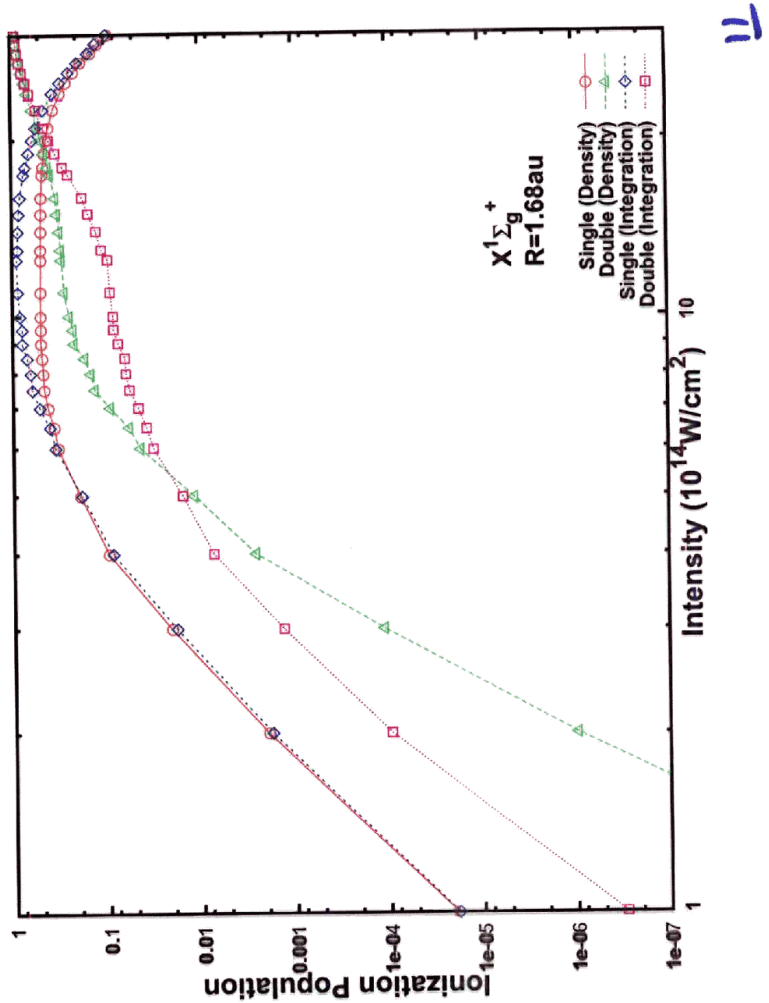
André D. Bandrauk*, HuiZhong Lu

ABSTRACT. We present a new method for the nonperturbative numerical solution of the Time-Dependent Schroedinger Equation, TDSE, for molecules in intense laser fields, using cylindrical/polar coordinates systems. For cylindrical coordinates systems, after use of a split-operator method which separates the z direction propagation and the (x, y) plane propagation, we approximate the wave function in each (x, y) section by a Fourier serie $(\sum c_m(\rho)e^{im\phi})$ which offers an exponential convergence in the ϕ direction and is naturally applicable for polar coordinates system. The coefficients $(c_m(\rho))$ are then calculated by a Finite Difference Method (FDM) in the ρ direction. We adopt the Crank-Nicholson method for the temporal propagation. The final linear system consists of a set of independent one-dimensional (ρ) linear systems and the matrix for every one-dimensional linear systems is *sparse*, so the whole linear system may be very efficiently solved. The most important advantage of the new method is that it is *unitary* at all steps and is analytically convergent every where even near the $\rho = 0$ origin, in contrast to most other numerical method based on cylindrical/polar coordinates systems which are not unitary and introduce a numerical singularity at $\rho = 0$. We also illustrate its generalization to the case of dimension D=4, to treat two electrons in the H_2 molecule.

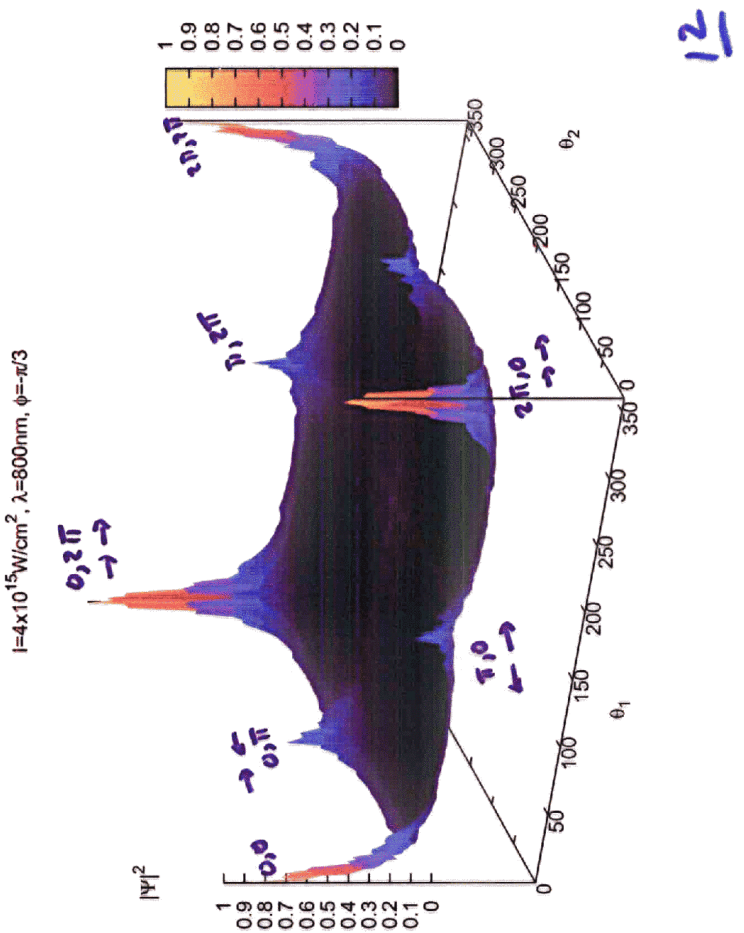
1. Introduction

The current advent of ultrashort intense laser pulses [1] necessitates the development of nonperturbative numerical methods to solve problems of interaction of lasers with molecules [2]. Because of efficiency, in many cases one uses cylindrical coordinates for the numerical solution of the 3-D Time Dependent Schroedinger Equation, TDSE, for linear molecules exposed parallel to an intense laser pulse: H_2^+ [3, 4, 5, 6], H_2 [7]. Generally, we use the split-operator method or the ADI (Alternating Direction Implicit Method [8]) method which separate the propagation in the different directions. This process reduces a lot of computing time and is very precise [8]. After splitting, one must propagate the wave function in each section plane in the polar coordinate. Polar coordinates have also been used in 2-D simulations of molecular alignment [5]. But in the polar coordinate system,

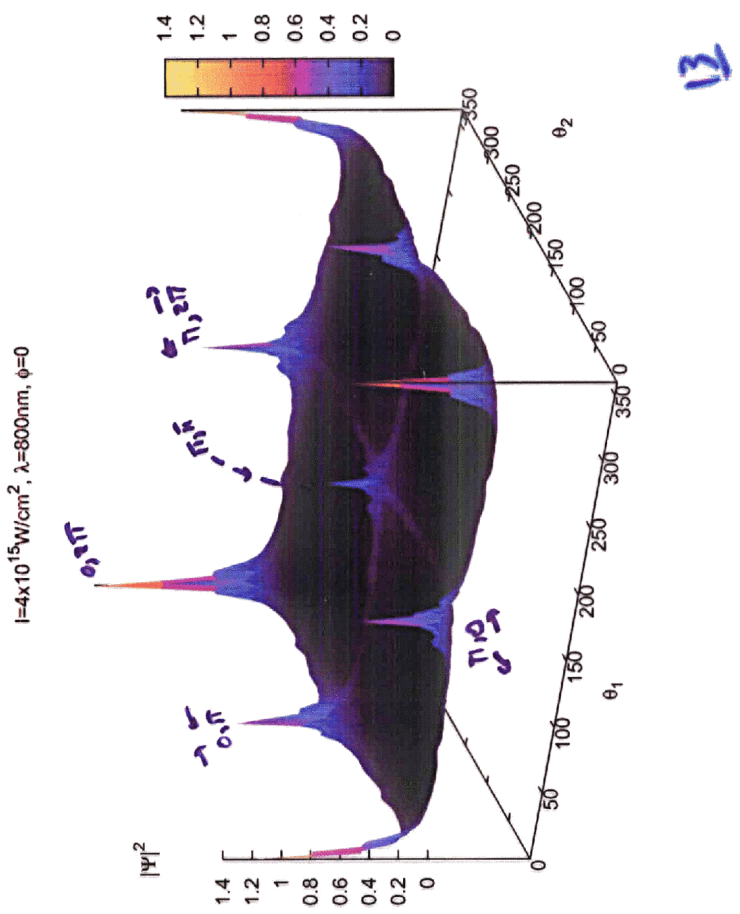


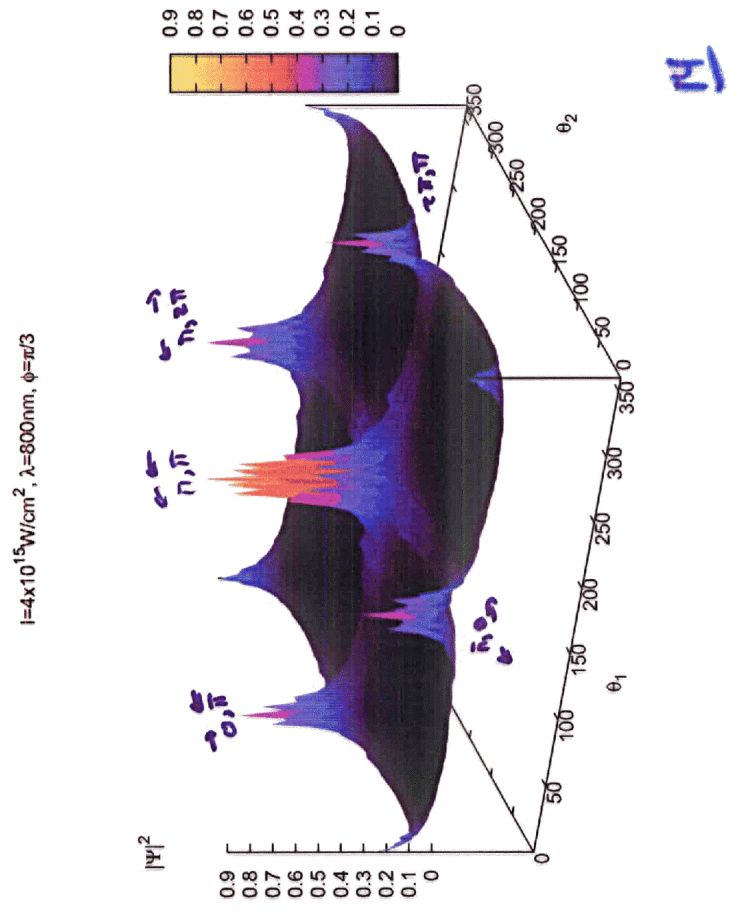

Ionization Population
Intensity (10^{14}W/cm^2)
$X^1\Sigma_g^+$
R=1.68au
Single (Density)
Double (Density)
Single (Integration)
Double (Integration)

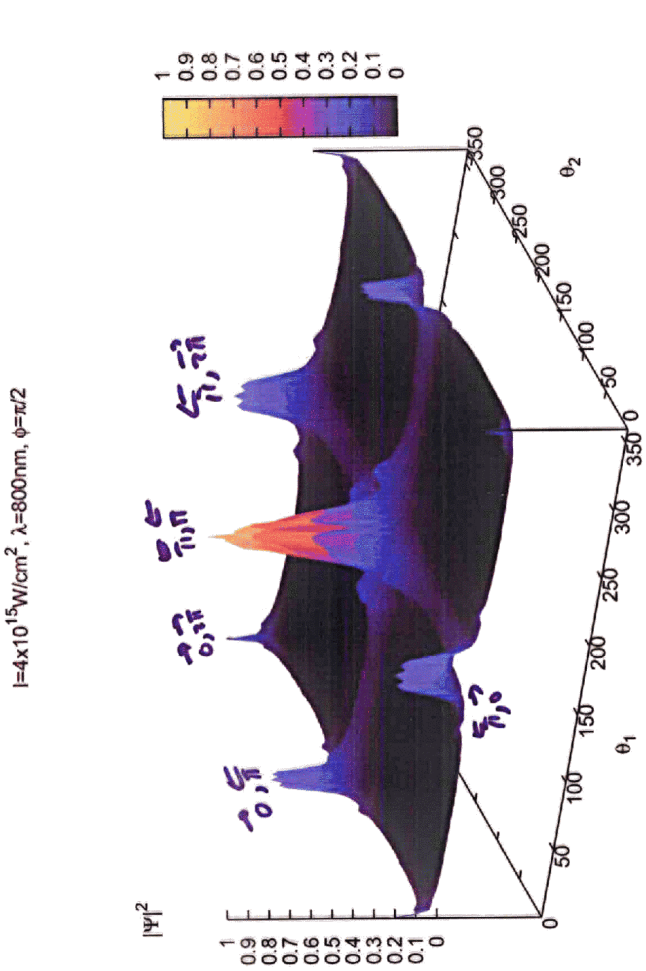
11

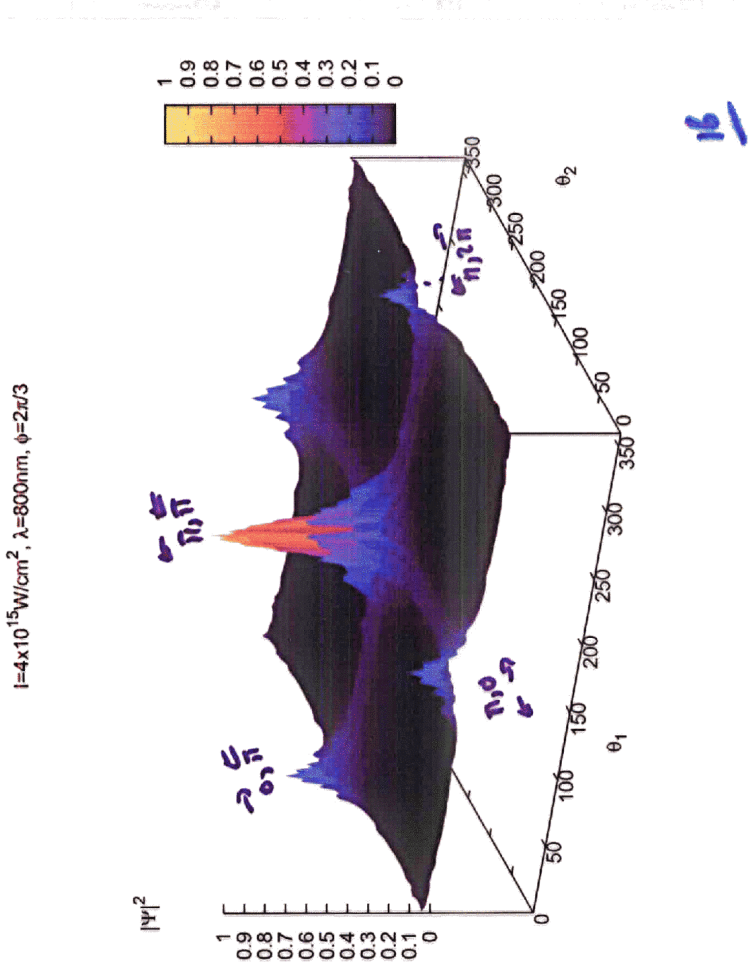

I=4x10^{15}W/cm^2, λ=800nm, φ=-π/3
$|\Psi|^2$
θ_1
θ_2

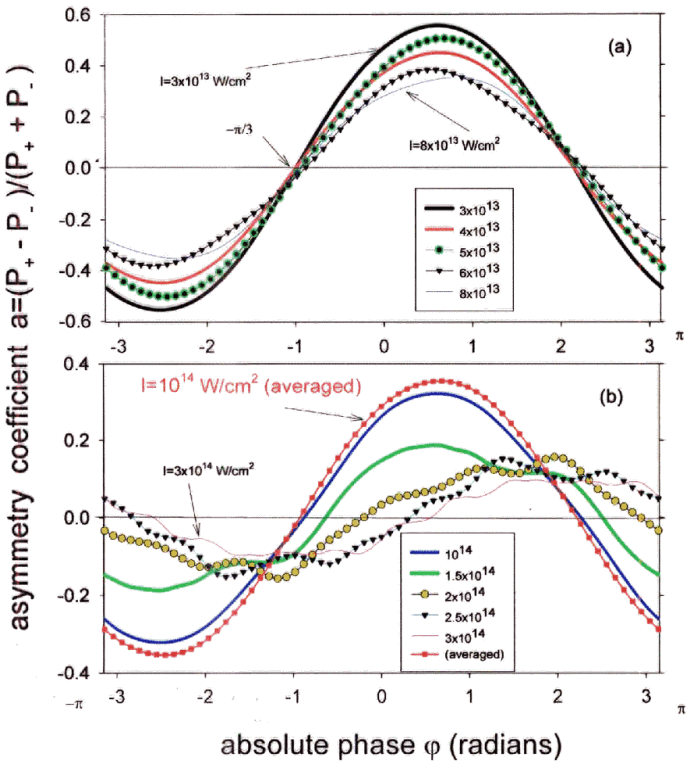
12


I=4x10^15 W/cm^2, λ=800nm, φ=0
|Ψ|^2
θ1
θ2


I=4x10^15 W/cm^2, λ=800nm, φ=π/3
|Ψ|^2
θ1
θ2


I=4x10^15 W/cm^2, λ=800nm, φ=π/2
|Ψ|^2
θ1
θ2


I=4x10^15 W/cm^2, λ=800nm, φ=2π/3
|Ψ|^2
θ1
θ2



* Phys. Rev. A 70, 013815 (2004).
Opt. Lett. 29, 1557 (2004).

17

18

week ending
29 AUGUST 2003

VOLUME 91, NUMBER 9 PHYSICAL REVIEW LETTERS

Time-Resolved Double Ionization with Few Cycle Laser Pulses

F. Légaré,[1,2] I. V. Litvinyuk,[1] P. W. Dooley,[1,3] F. Quéré,[1] A. D. Bandrauk,[2] D. M. Villeneuve,[1] and P. B. Corkum[1]
[1]*National Research Council of Canada, Ottawa, Ontario, Canada K1A 0R6*
[2]*Département de Chimie, Université de Sherbrooke, Sherbrooke, Québec, Canada*
[3]*Department of Physics & Astronomy, McMaster University, Hamilton, Ontario, Canada*
(Received 24 January 2003; published 29 August 2003)

Ionization of D_2 launches a vibrational wave packet on the ground state of D_2^+. Removal of the second electron places a pair of D^+ ions onto a Coulombic potential. Measuring the D^+ kinetic energy determines the time delay between the first and the second ionization. Caught between a falling ionization and a rapidly rising intensity, the typical lifetime of the D_2^+ intermediate is less than 5 fs when an intense 8.6 fs laser pulse is used. We simulate Coulomb explosion imaging of the ground state wave function of D_2 by a 4 fs optical pulse and compare with our experimental observations.

DOI: 10.1103/PhysRevLett.91.093002 PACS numbers: 33.80.Rv, 34.50.Gb, 42.50.Hz

Femtosecond pump-probe spectroscopy is an extremely powerful technique for studying molecular dynamics [1]. However, inferring the mechanisms responsible for the dynamics from these measurements is difficult. Imaging molecular structure would be much more direct and intuitive. We show that laser induced Coulomb explosion for direct imaging of molecular structure is within reach.

Accurate Coulomb explosion imaging of molecular structure requires that the molecule reach a sufficiently high charge state for the interionic potential to be approximated by Coulomb's law [2]. This charge state must be reached while the ions are inertially confined in their original configuration. Inertial confinement is readily achieved for small molecular ions moving at megavolt complete ionization in less than two optical cycles. Therefore, we can suppress (or control) processes that occur at larger internuclear separations even for a light molecule such as D_2.

To generate our ~9 femtosecond pulses, the output of a Ti:sapphire regenerative amplifier (810 nm, 40 fs, 250 μJ, 500 Hz repetition rate) amplifier was coupled into a hollow core fiber (250 μm diameter, 100 cm long) filled with 1 atm of argon. The self-phase modulation [10] during propagation of the optical pulses through the hollow fiber broadens the spectral bandwidth from 30 to 200 nm. The pulse was compressed using multiple reflections from a pair of chirped mirrors. The resulting pulses were characterized using spectral phase interfer-

VOLUME 89, NUMBER 28 PHYSICAL REVIEW LETTERS 31 DECEMBER 2002

Measuring the Electric Field of Few-Cycle Laser Pulses by Attosecond Cross Correlation

Andre D. Bandrauk,* Szczepan Chelkowski, and Nguyen Hong Shon†
Laboratoire de Chimie Theorique, Faculte des Sciences, Université de Sherbrooke, Quebec, J1K 2R1, Canada
(Received 5 August 2002; published 31 December 2002)

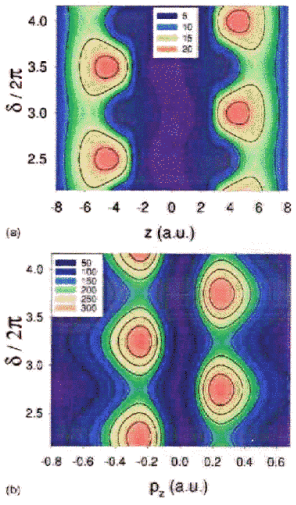
A new technique for directly measuring the electric field of linearly polarized few-cycle laser pulses is proposed. Based on the solution of the time-dependent Schrödinger equation (TDSE) for an H atom in the combined field of infrared (IR) femtosecond (fs) and ultraviolet (UV) attosecond (as) laser pulses we show that, as a function of the time delay between two pulses, the *difference* (or equivalently, *asymmetry*) of photoelectron signals in opposite directions (along the polarization vector of laser pulses) reproduces very well the profile of the electric field (or vector potential) in the IR pulse. Such ionization asymmetry can be used for directly measuring the *carrier-envelope phase difference* (i.e., the relative phase of the carrier frequency with respect to the pulse envelope) of the IR fs laser pulse.

DOI: 10.1103/PhysRevLett.89.283903 PACS numbers: 42.65.Re, 32.80.Rm, 42.65.Ky

To exploit attosecond technology FP1

Measuring electron wave packets

1. Attosecond pulses are fast enough to observe electron wave packets.
2. Electron wave packets are resolved through changes to the photo-electron spectrum as a function of pump-probe time delay.
3. The attosecond pulse projects the momentum distribution into the continuum.



Yudin et al, Phys Rev A 72, 51401(R) (2005)