Paper IV

Jan Petter Hansen, Morten Førre, Sølve Selstø and Ingrid Sundvor

Atoms and Molecules in Strong, High-frequency Fields

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ATOMS AND MOLECULES IN STRONG, HIGH-FREQUENCY FIELDS

J. P. Hansen, M. Førre, S. Selstø, I. Sundvor

Dept. of Physics and Technology, University of Bergen, Allégt. 55, N-5007 Bergen, Norway

ABSTRACT

A method for solving the time dependent Schrödinger equation in three dimensions based on a split operator method in spherical coordinates has been developed. This method is used to study photo-ionisation of atoms and molecules in the presence of intense, high-frequency, ultra-short laser pulses. We have investigated how the ionisation dynamics of *H(2p)* and H_2^+ depend on geometrical factors. In general, it is found that the relative orientation of the linearly polarised laser field and the initial orientation of the system is crucial. Furthermore, effects of atomic stabilisation is studied. From a generalisation of the Kramers-Henneberger formulation of the Hamiltonian, the laser ionisation of *H(1s)* is investigated without applying the dipole approximation.

INTRODUCTION AND METHOD

Laser technology has gone through continuous improvements ever since the beginning. By now, ultra intense lasers pulses with photon energies as high as 100 eV and with duration on the atto second time scale has been demonstrated [1,2]. The interaction between such fields and matter is a highly non perturbative one which calls for accurate, ab initio modelling methods on the theoretical side.

The method for solving the Schrödinger equation that we apply, is an extension of the original scheme of Hermann and Fleck [3]. Through a uniformly distributed spherical quadrature made public by Wommersley and Sloan [4], the approach is able to treat the electron dynamics in all three spatial dimensions [5]. The reduced wave function is expanded in Spherical Harmonics,

$$\Theta(\vec{r},t) = \sum_{l,m} f_{l,m}(r,t) Y_{l,m}(\hat{r}), \tag{1}$$

and the Schrödinger equation (in atomic units),

$$\left\{ -\frac{1}{2}\frac{\partial^2}{\partial r^2} + \frac{L^2}{2r^2} + V_s(r) + W(\vec{r},t) \right\} \Theta(\vec{r},t) = i\frac{\partial}{\partial t}\Theta(\vec{r},t),$$

(2)

is solved by writing the time propagator as

$$\Theta(\vec{r},t+\Delta t) = \mathrm{e}^{-iA\Delta t/2}\,\mathrm{e}^{-iB\Delta t/2}\,\mathrm{e}^{-iC\Delta t}\,\mathrm{e}^{-iB\Delta t/2}\,\mathrm{e}^{-iA\Delta t/2}\,\Theta(\vec{r},t)+\mathrm{O}(\Delta t^2),$$
$$A \equiv -\frac{1}{2}\frac{\partial^2}{\partial r^2},$$
$$B \equiv l(l+1)/(2r^2)+V_s(r),$$
$$C \equiv W(\vec{r},t)$$

(3)

The spatial differentiations are carried out through fast Fourier transforms. While the spherical part of the potential amounts to a straight forward multiplication of the spherical components of the wave function, it is necessary to construct and afterwards decompose the entire wave function to propagate the anisotropic, time-dependent potential W.

In principle any effective one electron dynamic system may be described by this method. In atomic and molecular physics, the most obvious applications would be collisions [6] and interaction with light. In the following we will focus on three examples of the latter.

APPLICATIONS

In general we will take the field to be of the form:

$$\vec{A}(t) = \frac{\vec{E_0}}{\omega}\sin^2\left(\frac{\pi t}{T}\right)\sin\left(\omega t+\varphi\right)$$

(4)

where E_0 is the maximum field strength, ω is the central frequency, T is the pulse duration and φ is carrier envelope phase.

In the following we will focus on the dependence of geometry in the interaction between the laser pulse and anisotropic systems. Furthermore, we will study non dipole effects and its importance to atomic stabilisation.

Laser Ionisation of the Hydrogen Molecular Ion

This most simple of molecular systems has been subject to intense theoretical investigation. Still, no ab initio, non perturbative description of this system including all spatial variables has been achieved. This has to do with the system's high number of degrees of freedom. In addition to the three spatial dimensions for the electronic, the internuclear separation R and the relative orientation θ between the internuclear axis and the polarisation of the field need to be considered, making the description far more complex than the corresponding atomic one.

Validity of the Fixed Nuclei Approach

Due to the long rotational period of the molecule it is usually quite safe to assume the orientation of the internuclear axis to be fixed during the time of interaction. In our case, we are focusing on pulses as short as 500 as. Therefore it seems reasonable to assume that also the internuclear separation R may be held fixed. Figure 1 confirms this [7]. It shows the total ionisation probability for a one dimensional electron with fixed internuclear separation, with classical internuclear dynamics and with full quantum mechanical internuclear dynamics.

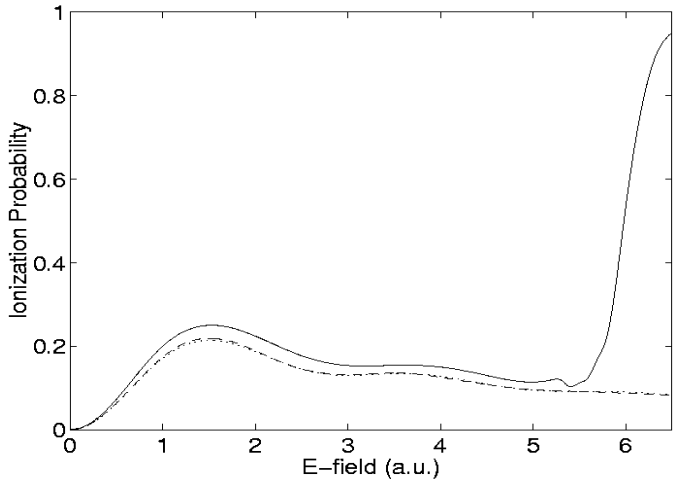

Figure 1: Ionisation probability with fixed nuclei (dotted curve), classical internuclear dynamics (dashed curve) and full quantum mechanical description (full curve). The laser has a central frequency of ω = 1 a.u. (about 27 eV), and the pulse duration T is 2 fs.

Results

The Schrodinger equation is solved with the Hamiltonian

$$H_l = -\frac{1}{2}\nabla^2 - \frac{1}{\left|\vec{r}+\vec{R}/2\right|} - \frac{1}{\left|\vec{r}-\vec{R}/2\right|} + \vec{E}(t)\cdot\vec{r} \tag{6}$$

in the length gauge. The electric field ***E(t)*** defines the angle θ with the internuclear axis. Figure 2 shows the total ionisation probability P_I as a function of both the internuclear distance R and the orientation θ [8].

The ionisation probability exhibits strong dependence on both R and θ. In particular, for θ=0° the ionisation probability P_I oscillates with R. For θ=90° these oscillations are absent, however. This phenomenon can be understood in terms of interference between outgoing waves originating from each of the scattering centres. Assuming that each of the outgoing waves essentially travels in the direction of the field, two outgoing waves in the direction parallel to the internuclear axis will have an initial phase difference depending on their separation R making the total outgoing wave subject to destructive interference. In the direction perpendicular to the internuclear axis, there is no initial

phase difference, and hence no such interference effect either.

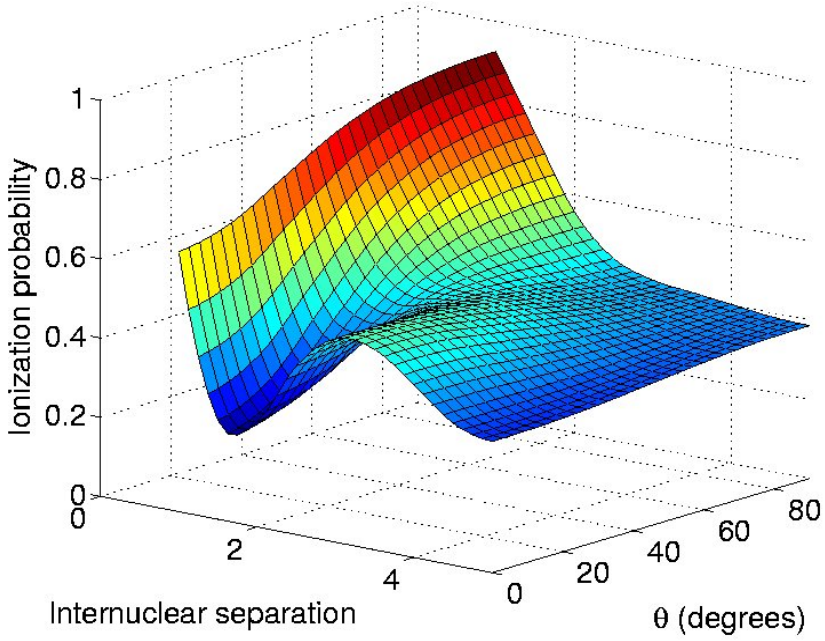

Figure 2: Ionisation probability as a function of internuclear distance R and the angle between the field and the internuclear axis. The laser field is given by ω=2 a.u., E_0=3 a.u. and the field duration T corresponds to 6 optical cycles.

Also the angular distribution of the photo electron can be understood within the same idea. Figure 3 shows the probability of ionisation as a function of direction for parallel (θ=0°), intermediate (θ=45°) and perpendicular (θ=90°) polarisation. The first two rows correspond to R=2 and 3, respectively, whereas the third row represents an average over R corresponding to the vibrational ground state.

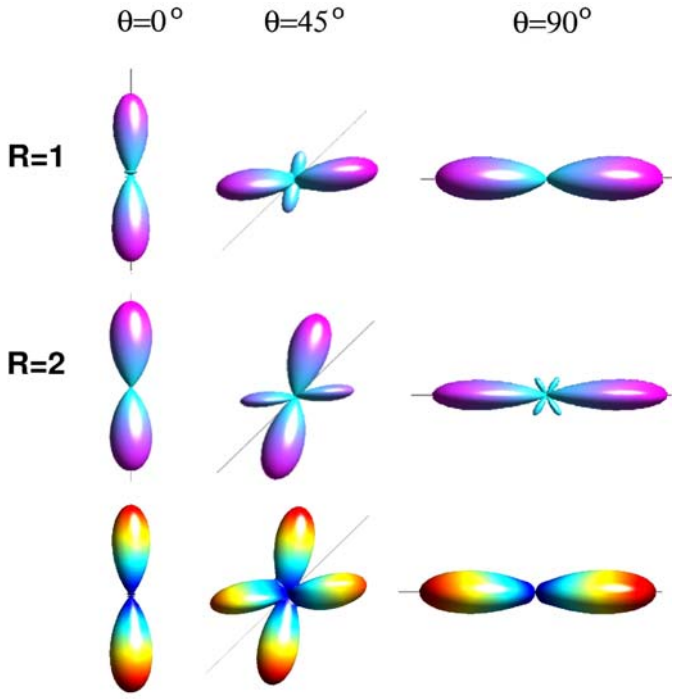

Figure 3: Angular distributions of the photo electron for various orientations and internuclear separations. The lowest row of figures corresponds to an initial R-distribution given by the vibrational ground state. The field parameters are the same as in Fig. 2.

Orientational Dependence in Photo-Ionisation of *H(2p)*

We may expect that the relative orientation θ will influence the ionisation probability also in the case of non isotropic atomic states. We have investigated this influence for a hydrogen atom initially prepared in the n=2, l=1, m=0-state exposed to laser field of energy ω=1 a.u. [9]. Results are displayed in Fig. 4. This time θ refers to the angle between the field and the quantisation axis. We find a rather strong dependence on this angle.

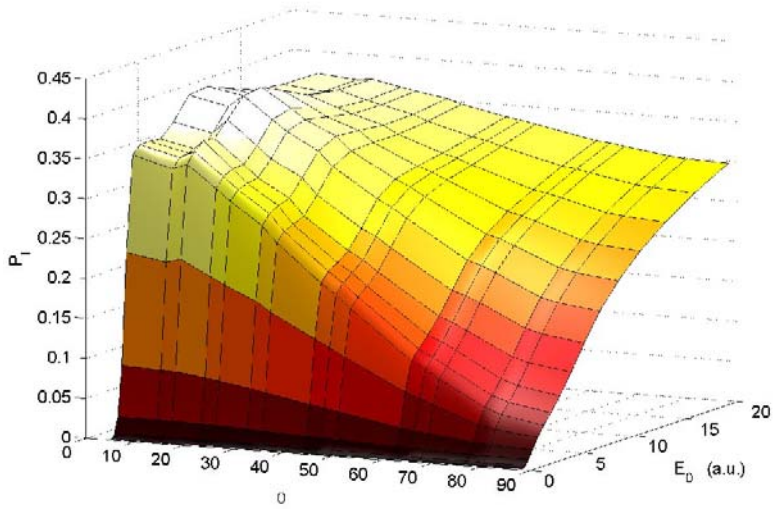

Figure 4: Probability of ionizing *H(2p)* as a function of the orientation of the electric field (θ) and the maximum field strength (E_0). The central frequency is ω=1 a.u., and the pulse duration corresponds to 5 optical cycles.

It is well known that the multi photon ionisation channels close as the photon energy becomes very large compared to the effective ground state binding energy [10]. For θ=90°, the *1s*-state is not accessible through a one photon transition (Δm=1) causing the effective binding energy to be lower than in the θ=0°-case. Consequently, the perpendicular ionisation is suppressed compared to the parallel one. Furthermore, as the intensity of the field increases, the effective binding energy decreases and the multi photon channels close for all values of θ. Due to this phenomena, the ionisation probability ceases to increase with the field intensity, and the system is *stabilised*. These phenomena explains the behaviour in Fig. 4 at higher field intensities.

Atomic Stabilisation and None-Dipole Effects

The effect of stabilisation has also been studied in more detail for a hydrogen atom initially in the ground state. In the stabilisation limit, the effective ionisation potential is small compared to the photon energy. This is manifested in the fact that only the zeroth order Floquet term in the potential in the Kramers Henneberger formulation of the Hamiltonian [11-13] contributes to the interaction,

$$V_{KH}(\vec{r},t) = -\frac{1}{\left|\vec{r}+\vec{\alpha}(t)\right|} \rightarrow V^{0}_{KH}(\vec{r}) = -\frac{1}{T}\int_{0}^{T}\frac{1}{\left|\vec{r}+\vec{\alpha}(t)\right|}dt. \tag{7}$$

This potential represents the time average of the field. Thus, the dynamics in this limit arises as a consequence of the non-adiabatic turn on and off of the laser pulse. We expect that as the intensity of the field increases, the energy spectra of the photo electron loose the peaks corresponding integer numbers of the photon energy, and only the strong maximum near the threshold survives. This is exactly what we find, as demonstrated in Fig. 5 [14].

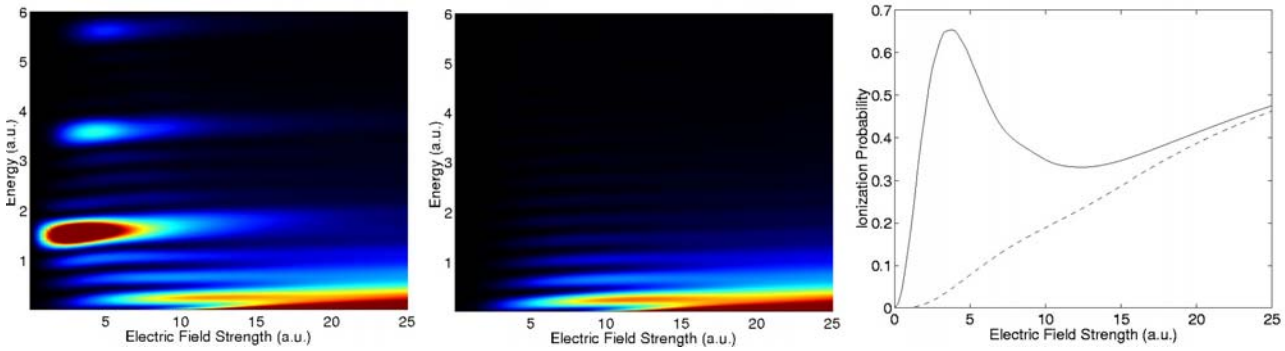

Figure 5: The figure to the left and in the middle show the energy distribution of the ionisation probability for various field strengths with the full interaction with the field and with the time averaged potential, respectively. We see that they coincide as E_0 becomes large. The figure to the right shows the total ionisation probability in the same two cases (full curve and dotted curve, respectively). Here we have ω=2 a.u. and T corresponds to 5 optical cycles. The shape of the pulse is slightly different from Eq. 4.

It has been claimed that atomic stabilisation is an artefact of the dipole approximation and that the effect will be strongly reduced by inclusion of the magnetic field [15]. This claim can be checked using the non-dipole version of the Kramers Henneberger Hamiltonian:

$$H^{ND}_{KH} = -\frac{1}{2}\nabla^2 + V(\vec{r}+\vec{\alpha}(\eta)) + \frac{1}{2}\left(A(\eta)\right)^2,$$
$$\eta \equiv \vec{k}\cdot\vec{r} - \omega t. \tag{8}$$

This formula is valid as long as the condition

$$\frac{E_0}{\omega c} \quad << \quad 1 \tag{9}$$

where E_0 is given in atomic units, is fulfilled. From Fig. 6 we see that as long as E_0 stays below 20 a.u. (corresponding to an intensity of about 10^{19} W/cm^2), the total ionisation probability is essentially unaltered by the inclusion of the magnetic field. The figure shows the ionisation probability and the survival probability of the ground state within and without the dipole approximation. It should be noted, however, that other quantities, such as e.g. the angular distribution of the photo electron, may be influenced by the spatial variation of the field, even though the total ionisation probability is not.

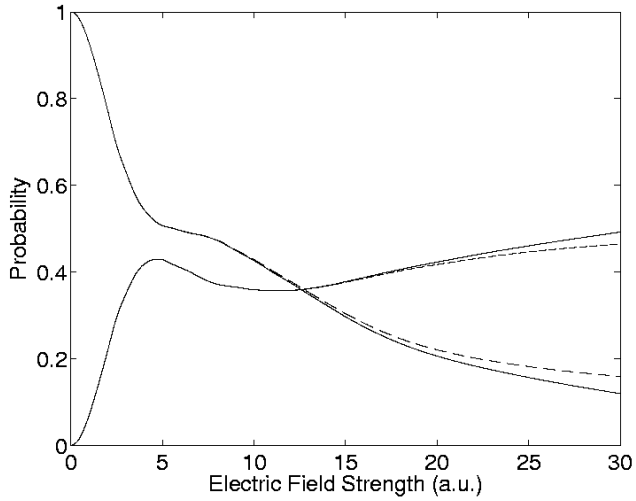

Figure 6: Ionisation probability and survival probability of the ground state with ω=2 a.u. and T= 380 a.s.. The full curve corresponds to the full interaction, and the dashed one to the dipole approximation.

CONCLUDING REMARKS

We have demonstrated that for non-isotropic molecular and atomic systems, geometry is crucial, and hence any adequate description of the dynamics should include all three spatial degrees of freedom of the electron. Furthermore, the effect of atomic stabilisation is found to sustain inclusion of the magnetic field of the laser. It has been demonstrated that the phenomena is a consequence of the closing down of multi photon ionisation channels. Consequently, photo electrons are found to have a very low energy after being ionized. The effect of the magnetic field on the photo electron, in particular its angular distribution, is currently being investigated. Furthermore, methods to include more particles in the scheme is being developed.

REFERENCES

[1] A. Baltuska, Th. Udem, M. Uiberacker, M. Hentschel, E. Goulielmakis, Ch. Gohle, R. Holzwarth, V. S. Yakovlev, A. Scrinzi, T. H. Hänsch, F. Kraysz, *Nature (London)* **412**, 611 (2003).

[2] R. Kienberger, E. Goulielmakis, M. Uiberacker, A. Baltuska, V. Yakovlev, F. Bammer, A. Scrinzi, Th. Westerwalbesloh, U. Kleinberg, U. Heinzmann, M. Drescher, F. Krausz, *Nature (London)* **427**, 817 (2004).

[3] M. R. Hermann, J. A. Fleck, *Phys. Ref. A* **38**, 6000 (1988).

[4] I. H. Sloan, R. S. Wommersley, Adv. Comput. Math. (2003), http://www.maths.unsw.edu.au/rsw/Sphere.

[5] J. P. Hansen, T. Matthey, T. Sørevik, "A parallel split operator mehtod for the time dependent Schrödinger equation", *10th Euro PVM/MPI* (2003).

[6] J. P. Hansen, T. Sørevik, L. B. Madsen, Phys. Rev. A **68**, 031401(R) (2003).

[7] I. Sundvor, J. P. Hansen, R. Taïeb, submitted to *Phys. Rev. A*.

[8] S. Selstø, M. Førre, J. P. Hansen, L. B. Madsen, *Phys. Rev. Lett.* **95**, 093002 (2005).

[9] T. Birkeland, M. Førre, J. P. Hansen, S. Selstø, *J. Phys. B* **37**, 4205 (2004).

[10] M. Gavrila, "Atoms in Intense Laser Fields" ed Gavrila (San Diego: Academic), 1992.

[11] W. Pauli, M. Fierz, *Nuovo Cimento* **15**, 167 (1938).

[12] H. A. Kramers, "Collected Scientific Papers" (Amsterdam: North Holland), 1956.

[13] W. C. Hennebeberger, *Phys. Rev. Lett.* **21**, 838 (1968).

[14] M. Førre, S. Selstø, J. P. Hansen, L. B. Madsen, *Phys. Rev. Lett.* **95**, 043601 (2005).

[15] A. Bugacov, M. Pont, R. Shakeshaft, Phys. Rev. A 48, R4027 (1993).