Ionization and dissociation dynamic of H2 molecule driven by a laser field

E. Fiordilino[1], G. Camiolo[1], G. Castiglia[1], P.P. Corso[1], R. Daniele[1],
J.P. Marangos[2], F. Morales[3], G. Orlando[1] and F. Persico[1]

[1]CNISM and Dipartimento di Scienze Fisiche ed Astronomiche, Università di Palermo, via Archirafi 36,
90123 Palermo, Italy
[2]Blackett Laboratory, Imperial College London, Prince Consort Road, South Kensington,
SW7 2AZ, United Kingdom
[3]CNISM and Dipartimento di Fisica e Tecnologie Relative, Università di Palermo, Ed. 18, Viale delle Scienze,
Parco d'Orleans, 90128 Palermo, Italy

We study the single and double ionization yields and the dissociation dynamics of a one-dimensional two-electron molecule exposed to an intense laser pulse. The double ionization yields are systematically investigated for both fixed and moveable nuclei. At low intensities the ionization curves have a power law shape, followed by a knee profile for higher intensities. A temporal correlation between the nuclear motion and the electron ionization shows that the nuclear motion deeply affects the ionization yield.

1. Introduction

The interaction of a strong laser pulse with atoms and molecules gives the opportunity to observe processes, like high order harmonic generation and non-sequential multiple ionization, which are non linear in the laser intensity and cannot be described by the standard perturbation theory.

In particular, the exact mechanism of multiple ionization is object of debate: the crucial experimental point is that the yield for double ionization is very large and its magnitude cannot be obtained by simple considering the double extraction process as produced by two individually independent and sequential steps [1]. The outputs of the suggested, non-sequential, ionization models are rather similar and agree sufficiently well with the experiments although differ from their microscopic temporal evolution [2-11]. They can be loosely grouped into two main families: the first one (shake-off models [1]) assumes that the extraction of one of the two electrons is such an abrupt process that the remnant electron does not have time to adapt to the new ionic potential and is left in an excited state from where it is extracted shortly later by the laser.

The second family (rescattering models [12]) assumes that the first extracted electron is pushed by the same laser field, that in the meanwhile inverted its phase, back to the parent ion where after a collision scatters the second electron into the continuum. Actually this second mechanism is strongly dependent upon the laser polarization; in fact a circularly polarized field does not allow a return trajectory to the ion and so prevents the double ionization [3,13].

Experiments and theoretical curves of multiple ionization yields as a function of the laser intensity present an initial power law profile explained by a n-th order perturbation regime followed by a knee flagging the breakdown of the perturbative approach. The challenge set by the problem of multiple ionization to the theorists resides in the fact that it couples the difficulties of non linear processes with those of treating satisfactorily correlation effects [14]. The scenario becomes rather more complicated if the dissociation of the molecule during the laser operation is taken into consideration. The presence of the laser field induces oscillations of the wave function of the electrons [15-16] that result in a force acting over the nuclei; accordingly such a force induce oscillations of the nuclei [17-18]. Since the electrons provide the only binding mechanism for the molecule, it is to expected that electron extraction induces molecular dissociation.

Recently it has been suggested that the dynamics of molecules can be directly accessible to real-time observation by detecting the harmonics diffused by the molecule acted upon by a laser field. The basic reason of this possibility is related to the fact that the acceleration of the charge that produces the harmonics is a quantity intrinsically determined by the physical environment where the charge is located; it is therefore to be expected that the generated field carries information on the dynamics of the charge. One of the main goals of today's science is the possibility of real time observation and control of chemical reaction. In these scenarios the knowledge of the dynamics of the molecule acted upon by a laser field is of paramount importance.

By generalizing a method proposed in [19], this paper discusses the problem of the dynamics of a one-dimensional H2 molecule acted upon by a laser

field. All the ionization channels are analyzed with particular care to understand the different routes that can be undertaken by the electrons in their ionization path. The nuclei are considered both fixed and moveable, their motion being evaluated by means of classical Newton equations of motion.

2. The model

We study the dynamics of a one-dimensional H_2 molecular system driven by an external laser field. In what follows X_1 and X_2 indicate the nuclei positions, x_1 and x_2 the electron positions; furthermore M and m indicate respectively the nuclei and the electron masses while the electron charge is -e. The temporal evolution of such a system is described by the time-dependent Schroedinger equation

$$i\hbar\frac{\partial}{\partial t}\Phi(X_1,X_2,x_1,x_2,t)=H\,\Phi(X_1,X_2,x_1,x_2,t) \quad (1)$$

where the Hamiltonian is given by

$$H = -\frac{\hbar^2}{2m}\left(\frac{\partial^2}{\partial x_1^2}+\frac{\partial^2}{\partial x_2^2}\right)-\frac{\hbar^2}{2M}\left(\frac{\partial^2}{\partial X_1^2}+\frac{\partial^2}{\partial X_2^2}\right)+$$

$$+U(X_1,X_2,x_1,x_2,t)+V(X_1,X_2,x_1,x_2,t); \quad (2)$$

being

$$U(X_1,X_2,x_1,x_2,t)=-\frac{e^2}{\sqrt{A+(x_1-X_1)^2}}-\frac{e^2}{\sqrt{A+(x_1-X_2)^2}}-$$

$$-\frac{e^2}{\sqrt{A+(x_2-X_1)^2}}-\frac{e^2}{\sqrt{A+(x_2-X_2)^2}}+$$

$$+\frac{e^2}{\sqrt{A_{12}+(x_1-x_2)^2}}+-\frac{e^2}{\sqrt{A_N+(X_1-X_2)^2}} \quad (3)$$

the system potential energy and

$$V(X_1,X_2,x_1,x_2,t)=$$

$$e(-X_1-X_2+x_1+x_2)E_0 f(t)\sin(\omega_L t) \quad (4)$$

the laser-molecule interaction energy in the dipole approximation, with E_0 the peak laser electric field amplitude and $f(t)$ the laser pulse profile. As it is evident from Eq. (3) we use a soft-core potential to model the electrostatic interactions among the electrons and the nuclei. The screening parameters A, A_{12} and A_N are responsible respectively for the softening of the electron-nuclei, electron-electron and nucleus-nucleus interaction.

In order to simplify the treatment of the system, it is possible to introduce some approximations which allow us to deal with only two quantum coordinates (the electron ones) and with two classical coordinates (the nuclei ones). This approximation relies upon the fact that the proton mass is much larger than the electron one, so we may assume that it is possible to decouple the nuclei dynamics from the electron dynamics; this is the well known adiabatic approximation [20].

3. Results

The results concern the dynamics the one-dimensional H_2 molecule, initially in its ground state, for different values of the internuclear distance R, interacting with a linearly polarized laser electric field, whose photon energy is $\hbar\omega = 3.18\,\text{eV} = 0.117\,e^2/a_O$ in the range of laser intensity $I_L = 10^{13} - 10^{15}\,\text{W/cm}^2$. We use a trapezoidal laser pulse, which linearly turns on for 8 optical cycles (o.c.), remains constant for 16 o.c. and then linearly turns off for 8 o.c..

We start by showing the results obtained in the fixed-nuclei approximation. In Fig. 1 we show the single and double ionization yields for three different internuclear distances, namely 3, 6 and 10 a.u., as function of laser intensity I_L.
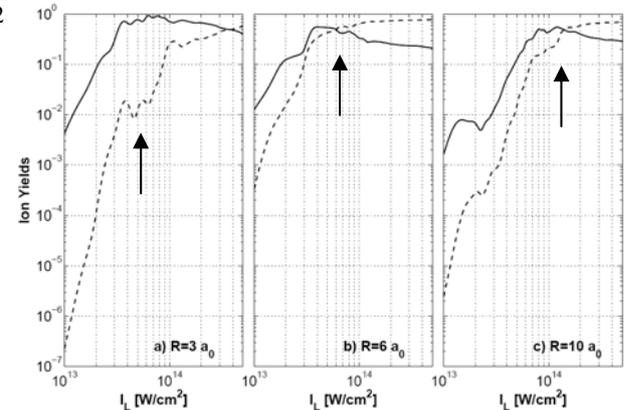
Figure 1 - Logarithmic plot of single (solid line) and double (dashed line) ionization signals versus I_L for three different internuclear distances. The arrows indicate approximately the position of the knee structure in the double ionization signals.

From Fig. 1 it is evident that the ionization yields strongly depend on the value of the internuclear distance, showing a strong enhancement for $R =|X_2-X_1|=6 a.u.$.

In Fig. 2 we show analogous results obtained in the case of moveable nuclei, assuming the system starting from the equilibrium internuclear distance $R = 2.62 \, a_O$.
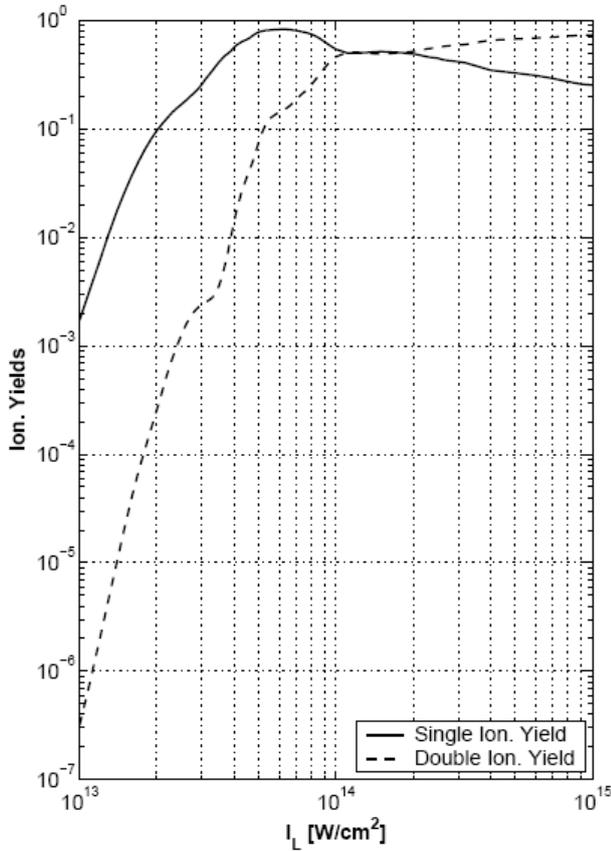

Figure 2 - Logarithmic plot of single (solid line) and double (dashed line) ionization signals versus I_L for the moveable nuclei system.

For small intensities the single ionization signal is in a perturbative regime in fact it has a trend of the kind I^n_L, where I_L is the laser intensity, till $I_L=2x10^{13}$ W/cm^2, then we observe a break of the power law as we see in Fig. 2. This behaviour is due to the nuclei motion. In fact, as apparent in Fig. 3(a)-(b), for small laser intensities the molecular system is bounded; the nuclei oscillate around a new equilibrium position and their distance is always less than $3a_0$ so that the ionization remains always small or negligible. Beyond $I_L=2x10^{13}$ W/cm^2 a small ionization causes the two nuclei to move apart; during the laser operation R attains the critical value $3a_0$ and causes an increased ionization: the final result is molecular dissociation.

This temporal relation becomes evident if we compare Fig. 4(a)-(c), where we plot the ionization signals versus the time, and Fig. 4(b)-(d), where we plot the detail of the internuclear distance R versus the time for the same laser intensities.
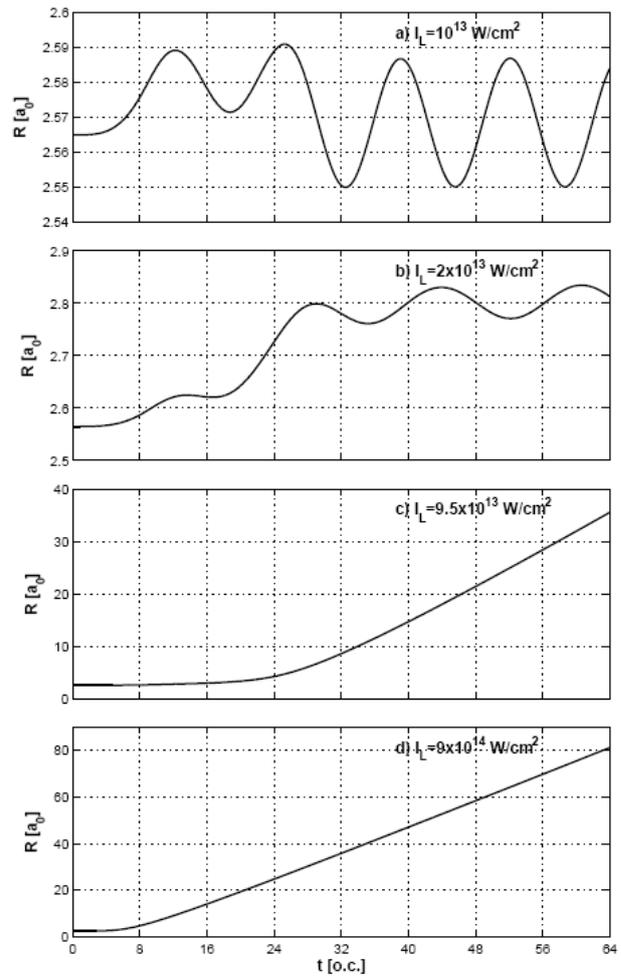
Figure 3 - Plot of the internuclear distance R as a function of time for various laser intensities I_L: a) I_L is 10^{13}, b) $2x10^{13}$, c) $9.5x10^{13}$, d) $9x10^{14}$ W/cm^2.
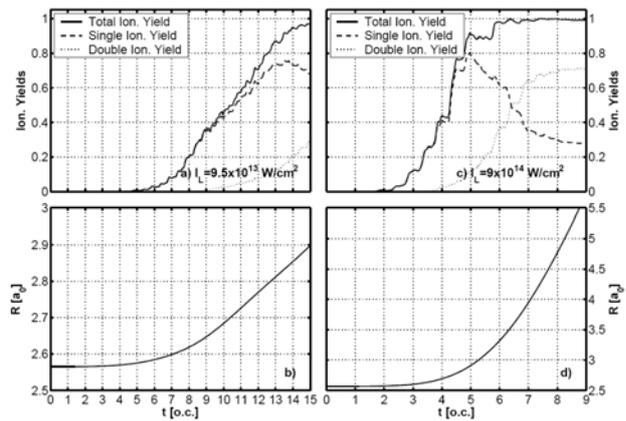
Figure 4 - Plot of the total (solid line), of the single (dashed line) and of the double (dotted line) ionization signals as a function of time for two laser intensities, a) $9.5x10^{13}$, c) $9x10^{14}$ W/cm^2. Plot of the internuclear distance R as a function of time for two laser intensities, b) $9.5x10^{13}$, d) $9x10^{14}$ W/cm^2.

4. Conclusions

In conclusion we have carried out a systematic and detailed investigation on the ionization and

dissociation of a one-dimensional H_2 molecule driven by a strong laser field. The first step has been the study of the ionization yields in the fixed nuclei approximation as a function of the internuclear distance R. The ionization channels are thoroughly examined both for the single and the double ionization yield and present the well known knee structure marking the breakdown of perturbative and/or single active electron approaches.

As a second step the nuclei have been allowed to move; now the laser intensity plays an important role. At low intensity the oscillation of the electrons wave function induces vibrations of the nuclei which are left excited. At higher laser intensity, a small ionization induces a stretching of the molecular axis: if the molecular axis length reaches the value $R = 3a_O$ the single ionization yield sudden increases and a quick dissociation starts in a sort of chain reaction; of course the same line of reasoning can be repeated for the double ionization probability. Finally the process results is a non linear ionization and dissociation dynamics. The motion of the nuclei, therefore, seems to play a role in the mechanism of ionization.

5. Acknowledgements

6. References

[1] D. N. Fittinghoff, P. R. Bolton, B. Chang, and K. C. Kulander, Phys. Rev. Lett. 69, 2642 (1992).

[2] B. Walker, B. Sheehy, L. F. DiMauro, P. Agostini, K. J. Schafer, and K. C. Kulander, Phys. Rev. Lett 73, 1227 (1994).

[3] Th. Weber, H. Giessen, M. Weckenbrock, G. Urbasch, A. Staudte, L. Spielberger, O. Jagutzki, V. Mergel, M. Vollmer, and R. Dörner, Nature 405, 658 (2000).

[4] H. W. van der Hart and K. Burnett, Phys. Rev. A 62, 013407 (2000).

[5] M. Lein, E. K. U. Gross, and V. Engel, Phys. Rev. Lett. 85, 4707 (2000).

[6] A. Becker and F. H. M. Faisal, Phys. Rev. Lett. 89, 193003 (2002).

[7] C. F. de Morisson Faria, H. Schomerus, X. Liu, and W. Becker, Phys. Rev. A 69, 043405 (2004).

[8] M. Dörr, Opt. Express 6,111 (2000).

[9] J. L. Chaloupka, R. Lafon, L. F. DiMauro, P. Agostini and K. C. Kulander, Opt. Express 8, 352 (2001).

[10] M. Lein, V. Engel, E. K. U. Gross, Opt. Express 8, 411 (2001).

[11] R. Moshammer, J. Ullrich, B. Feuerstein, D. Fischer, A. Dorn, C. D. Schröter, J. R. Crespo López-Urrutia, C. Höhr, H. Rottke, C. Trump, M. Wittmann, G. Korn, K. Hoffmann, and W. Sandner, J. Phys. B: Atom. Mol. Phys. 36, L113 (2003).

[12] P. B. Corkum, Phys. Rev. Lett. 71, 1994 (1993).

[13] D. N. Fittinghoff, P. R. Bolton, B. Chang, and K. C. Kulander, Phys. Rev. A 49, 2174 (1994).

[14] W.-C. Liu, J. H. Eberly, S. L. Haan, and R. Grobe, Phys. Rev. Lett. 83, 520 (1999).

[15] G. Castiglia, P. P. Corso, R. Daniele, E. Fiordilino, F. Morales, and F. Persico, J. Mod. Optics 51, 1163 (2004).

[16] R. Daniele, G. Camiolo, G. Castiglia, P. P. Corso, F. Morales, and E. Fiordilino, Appl. Phys. B 78, 813 (2004).

[17] P. P. Corso, R. Daniele, E. Fiordilino, J. P. Marangos, F. Morales, and R. Velotta, Phys Rev A 70, 053410 (2004).

[18] G. Castiglia, G. Camiolo, P. P. Corso, R. Daniele, E. Fiordilino, and F. Morales, Laser Phys. 14, 1185 (2004).

[19] M. Lein, E. K. U. Gross, and V. Engel, J. Phys B: At. Mol. Opt. Phys. 33, 433 (2000).

[20] M. Weissbluth Atoms and molecules (Academic press, New York, 1978).