

Molecules in intense laser fields : From Coulomb explosion to high order harmonic generation

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The nonperturbative response of molecules in ultrashort ($t < 20$ femtoseconds), intense ($I \geq 10^{14}$ W/cm²) laser fields can only be described by accurate numerical solutions of the corresponding time-dependent Schrödinger equation (non-relativistic) or Dirac (relativistic) equations. Different time scales, attosecond (10^{-18} s) for electrons, femtosecond (10^{-15} s) for nuclei make the accuracy of such numerical solutions strongly dependent on the algorithms. We present various versions of the split-operator method used in our work with concrete applications to calculating kinetic energy spectra of Electrons (ATI), and of protons (ATD, Coulomb explosion). Possible experimental and technological applications such as electron and nuclear imaging, high order harmonic generation and laser control of electron-nuclear dynamics are suggested

Introduction

The advent of short ($t \leq 20$ fs), intense ($I \geq 10^{14}$ W/cm²) laser pulses¹⁾ has led to the investigation of the regime of nonlinear, nonperturbative laser-matter interaction. The atomic case is now well documented in a recent review²⁾ that emphasizes the discovery of new nonlinear multiphoton processes such as above threshold ionization, ATI, and high order harmonic generation, HOHG. The behavior of molecules in intense laser fields offers a new challenge due to the presence of the nuclear degrees of freedom. Thus as early as 1980, we predicted using the dressed molecule picture of nonperturbative laser-molecule interaction, that new laser-induced molecular bound states would be created whenever such radiative interactions exceeded molecular vibrational energies.³⁾ These laser-induced molecular bound states have now been experimentally confirmed^{4,5)} and are the first concrete examples of nonperturbative molecular interactions.

With current high laser intensities approaching or exceeding the atomic unit of field strength:

$$\varepsilon_0 = e/a_0^2 = 5.15 \times 10^9 \text{ V/cm} \quad (1)$$

$$I_0 = c\varepsilon_0^2/8\pi = 3.5 \times 10^{16} \text{ W/cm}^2, \quad (2)$$

where $a_0 = 0.052$ nm is the Bohr radius, the atomic unit (a.u.) of length, ionization is a predominant process. Thus in a dissociative-ionization molecular process, one is dealing with an initial electron-nuclear bound state where electronic time scales correspond to the atomic unit of time 24.7 attoseconds (atts, one attosecond = 10^{-18} s), whereas nuclear vibrational time scales correspond to femtoseconds (fs), *e.g.* H_2^+ vibrational period = 15 fs. The final electronic state corresponds to an ionized electron which acquires energy through the ponderomotive energy,

$$U_p = \frac{e^2 \varepsilon^2}{4m\omega^2} = \frac{I/I_0}{4\omega^2} \text{ a.u.} \quad (3)$$

In the last formula ω and U_p are in atomic units of energy, 1 a.u. = 27.2 eV. U_p is a classical concept from plasma physics.^{6,7)} With current lasers such as YAG laser, $\lambda =$

1064 nm ($\omega = 0.0428$ a.u.) and at the a.u. intensity, $I = I_0 = 3.5 \times 10^{16}$ W/cm², one obtains $U_p = 1/4\omega^2 = 136.5$ a.u. = 3.7 keV. The corresponding electron-oscillatory (quiver) radius defines the maximum displacement of a free electron in a field of maximum amplitude ε ,

$$\alpha_0 = e\varepsilon/m\omega^2 = (\varepsilon/\varepsilon_0\omega^2)(\text{a.u.}) \quad (4)$$

where 1 a.u. of length $a_0 = 0.052$ nm. Thus one obtains an effective, classical radius $\alpha_0 = 546$ a.u. = 28.4 nm at intensity I_0 . Such large electronic energies and displacements suggest that classical models might be applicable to describe such nonperturbative phenomena, but the initial bound state requires quantum calculations.

Surprisingly, although high laser intensities produce highly nonlinear nonperturbative phenomena, simple classical models can sometimes be applied. In the case of atomic ionization in the long wavelength regime a quasistatic laser field induced Coulomb barrier suppression model allows for predicting atomic tunneling ionization probabilities⁶⁾ and even HOHG.⁷⁾ A similar quasistatic model which include the multilevel Coulomb potential of electrons in molecules can be used to explain a new molecular nonperturbative phenomenon, CREI, Charge Resonance Enhanced Ionization, originally discovered in numerical simulations of the ionization and HOHG in the simplest molecule,⁸⁾ H_2^+ . Such a quasistatic model explains the critical internuclear distances⁹⁻¹¹⁾ R_c and angles θ_c where CREI occurs, as well as, molecular HOHG plateaus.^{12,13)}

A useful parameter, γ , called the Keldysh parameter¹⁴⁾ helps to separate nonlinear multiphoton processes, either atomic or molecular, into two regimes, the multiphoton and tunneling regimes. γ is basically the ratio of atomic (molecular) electronic energy and field induced energies,

$$\gamma = \sqrt{\frac{I_p}{2U_p}}, \quad (5)$$

where U_p is the ponderomotive energy defined in Eq. (3) and I_p is the ionization potential. Thus for the hydrogen atom, $I_p = 13.6$ eV = 0.5 a.u., so that at $I_0 = 1$ a.u. ($3.5 \times$

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10^{16} W/cm^2), for $\omega = 1 \text{ a.u.}$ (a photon having such energy has a wavelength $\lambda = 45.6 \text{ nm}$), then $\gamma = 1.0$. Thus for $\gamma < 1$ and $\omega < I_p$, where field effects as measured by U_p , exceed atomic (molecular) energies I_p , it is found generally that ionization rates become independent of wavelength and that a tunneling model can be adequate for describing ionization and HOHG.⁷⁾ For $\gamma > 1$, typical of high frequencies, *e.g.* UV, X-Ray, one has the regime of multiphoton ionization, where the ionization rate is usually proportional to I^n , where n is the photon order and I is the maximum field intensity. In both cases, *i.e.* $\gamma > 1$ and $\gamma < 1$, one has to deal with multiphoton processes, so that even in any perturbative procedure one has to sum over large number of states, resonant and non-resonant, bound and continuum, electronic and nuclear. Thus a numerical method based on accurate solutions of the time-dependent Schrödinger equation, TDSE, is the most appropriate avenue to study intense field-molecule interactions. Recent proposals to construct Free Electron Lasers,¹⁵⁾ FEL's, predict attaining intensities of $I = 10^{26} \text{ W/cm}^2$ and wavelength of 1 Å ($\lambda = 0.1 \text{ nm}$, $\omega = 455.6 \text{ a.u.} = 12.4 \text{ keV}$). In such extreme cases, the achievable ponderomotive energy in the X-Ray beam will be according to Eq. (3), $U_p = 10^5 \text{ eV} = 0.1 \text{ MeV}$. This approaches the rest mass of the electron $mc^2 = 0.51 \text{ MeV}$, thus necessitating solutions of the time-dependent Dirac equation, TDDE, in such extreme conditions.

As stated above, the molecular case is quite different from the atomic case, since in the former now one has to deal with nuclear motion which occurs in slower time scales than electronic motion (atts.) in the ground state. Excited electronic states however have similar time scales as nuclear motion, so that the usual Born-Oppenheimer approximation of molecular physics¹⁶⁾ is no longer applicable. We have performed the first non-Born-Oppenheimer calculations of dissociative-ionization of the simplest molecule in 3-D¹⁷⁾ and 1-D.¹⁸⁾ In the latter 1-D simulations, complete electron and nuclear kinetic energy spectra were obtained using a new technique which avoids using absorbing boundaries. This advance in computational method has been extremely useful and is allowing us to predict new imaging methods for electrons¹⁹⁾ and nuclei²⁰⁾ using ultrashort intense pulses. Extensions to multi-electron, multi-nuclear systems is required to address the efficient laser-heating and Coulomb Explosion of clusters leading to table-top production of neutrons,²¹⁾ and even to understand the astrophysical phenomena.²²⁾

A numerical method for linearly polarized laser fields

An intense, linearly polarized laser field aligns molecules,²³⁾ thus allowing to use a simple model, in which the molecular axis is parallel to the laser polarization. This allows to reduce considerably the computer time and computer memory. A numerical method of integrating the time-dependent Schrödinger equation, for aligned molecule, was first presented for the H_2^+ molecule using cylindrical coordinates and expressing the wave function as a Bessel-Fourier series.²⁴⁾ This expansion allows one to eliminate singularities present in the Hamiltonian and to use unitary split operator method²⁵⁾ to evaluate numerically multiphoton transitions. Thus in cylindrical coordinates and in atomic units ($e = \hbar = m = 1$), the TDSE for static (Born-Oppenheimer) nuclei describing

the interacting matter with linearly polarized laser field along the z -axis coinciding with the internuclear R axis, is written as,

$$i \frac{\partial}{\partial t} \Psi(z, \rho, t) = -\frac{1}{2} \frac{\partial^2}{\partial z^2} \Psi + D_\rho \Psi + V_c(z, \rho) \Psi + z \varepsilon(t) \Psi \quad (6)$$

where

$$D_\rho = \frac{1}{2} \left(\frac{\partial^2}{\partial \rho^2} + \frac{1}{\rho} \frac{\partial}{\partial \rho} \right) \quad (7)$$

and

$$V_c = -(\rho^2 + (z - R/2)^2)^{-1/2} - (\rho^2 + (z + R/2)^2)^{-1/2} \quad (8)$$

for H_2^+ and

$$V_c = -(\rho^2 + z^2)^{-1/2} \quad \text{for H - atom,} \quad (9)$$

where ρ and z ($z = 0$ is the center of the molecule) are the cylindrical electron coordinates in H_2^+ . Numerical integration of Eq. (6) using the finite-difference scheme requires special care near $\rho = 0$ because of the singularity in the cylindrical Laplacian D_ρ and in the Coulomb potential $V_c(\rho, z)$. Using Bessel-Fourier series built from the functions²⁶⁾

$$v_n(\rho) = \frac{\sqrt{2}}{L J_1(x_n)} J_0(x_n \rho / L) \quad (10)$$

where J_0 and J_1 are Bessel functions, x_n are zero's of J_0 and L is the maximal ρ value, one can eliminate efficiently these singularities, thus transforming the parabolic partial differential equations ppde, Eq. (6) in three coordinates (R is fixed) t, ρ , into coupled 2-D ppde's by eliminating the cylindrical coordinate ρ . Thus since the v_n 's, Eq. (10) are eigenfunctions of D_ρ ,

$$D_\rho v_n(\rho) = -\left(\frac{x_n}{L}\right)^2 v_n(\rho), \quad (11)$$

then the v_n 's constitute a complete basis for functions $f(\rho)$ defined on the interval $0 \leq \rho \leq L$ and have continuous second derivatives at the origin²⁶⁾ ($\rho = 0$) and ensured the condition $f(L) = 0$. Using the orthonormality of v_n 's, then one can expand the total wave function Ψ in this basis,

$$\Psi(z, \rho, t) = \sum_{n=0}^N \Phi_n(z, t) v_n(\rho) \quad (12)$$

resulting (after inserting (12) into TDSE (6)) in the following set of coupled 2-D ppde's in z and t :

$$i \frac{\partial}{\partial t} \Psi(z, t) = -\frac{1}{2} \frac{\partial^2}{\partial z^2} \Psi + A(z) \Psi + \varepsilon(t) z \Psi(z, t), \quad (13)$$

where Ψ is an N -dimensional vector and $A(z)$ is a $N \times N$ matrix with elements defined by

$$A_{n,k}(z) = \frac{1}{2} \left(\frac{x_n}{L}\right)^2 \delta_{n,k} + \int_0^L v_n(\rho) V_c(z, \rho) v_k(\rho) \rho d\rho. \quad (14)$$

We note that the matrix potential $A(z)$ which replaces the Coulomb potential $V_c(z, \rho)$ is now nonsingular. The system of ppde's Eq. (13) being parabolic can be integrated using split-operator methods.²⁵⁾ To second order accuracy, the time-evolution algorithm is based on the relation

$$\begin{aligned} \Psi(t + \delta t, z) = & \exp(D_z \delta t) \exp(-iE(t') z \delta t) \\ & \exp(-iA(z) \delta t) \exp(D_z \delta t) \Psi(t, z) \end{aligned} \quad (15)$$

where $D_z = \frac{i}{4}\partial/\partial z$ and $t' = t + \delta t/2$. The operation $\exp(D_z \delta t)\Psi(t, z)$ is performed with the help of FFT and the exponential matrix $B(z) = \exp(-iA(z)\delta t)$ was evaluated using the formula valid for the Hermitian matrix A ,

$$B = U \exp(-iA_D \delta t) U^T \quad (16)$$

where U is a unitary diagonalizing matrix (it contains eigenvectors of A as its columns), and A_D is the corresponding diagonalized matrix. The matrix $\exp(-iA_D \delta t)$ is simply a diagonal matrix containing the exponentials of the eigenvalues of A . Calculating of the matrix B for each z is time consuming, but needs to be done only once since B is time independent for a fixed R . The propagation scheme (15) has the advantage of being always unitary and can be easily extended beyond second order accuracy using symmetric products which preserve Hermiticity and time reversibility.²⁵⁾ We have also applied the scheme to coupled nonlinear Schrödinger equations²⁷⁾ and shown it to reproduce well known soliton structures of such nonlinear equations.

The above integration scheme is used to obtain ionization rates by applying a best exponential fit to the function

$$P_V(t) = \int_{-z_M}^{z_M} dz \int_0^L d\rho \rho |\Psi|^2 \quad (17)$$

which represents the probability of finding the electron in the integration cylinder with absorbing boundaries at the limits $\pm z_M$. Alternatively, we can calculate the ground state population $P_0(t) = |\langle \Psi_0 | \Psi(t) \rangle|^2$ from which the ionization rate Γ can be calculated as $-\ln[P_0(t)]$. Such a procedure fails in the case of H_2^+ due to resonant multiphoton processes, especially between the ground state and first excited state which have divergent transition dipole moments $\mu = R/2$, leading to large radiative couplings³⁾ $\mu \varepsilon(t)$. Thus rapid Rabi oscillations appear in P_0 which make it difficult to define proper ionization rate.

Equation (6) has been generalized to include nuclear, *i.e.* R motion.¹⁷⁾ This increases the dimensionality of the problem to four, t, z, ρ, R , which using the Fourier-Bessel series expansion Eq. (12) reduces Eq. (13) to a 3-D set of coupled ppde's in t, z and R . This has allowed us to study the effect of nuclear motion on ionization rates and to discover the phenomenon of critical distances for Coulomb explosions, via CREI.⁸⁻¹⁰⁾ Exact solution of Eq. (6) with static (Born-Oppenheimer) and moving (non-Born Oppenheimer) nuclei has also allowed us to calculate a highly nonperturbative phenomenon, high order harmonic generation (HOHG). The HOHG spectrum is obtained by calculating the power spectrum generated by the highly nonlinear laser-induced dipole moment,

$$z(t) = \langle \Psi(t) | z | \Psi(t) \rangle \quad (18)$$

which is Fourier transformed to give the power spectrum $|z_F(\omega)|^2$. Such a spectrum is generated by recollisions of an electron with its parent ion⁷⁾ giving maximum photon energy of $3U_p$ or by recollision with neighbouring ions giving maximum energies up to $12U_p$ and beyond.^{12,13)} In the latter the maximum displacement of the electron from its initial bound state position can attain easily $\pi\alpha_0$. Clearly one is dealing with high kinetic energy electrons, having very short wavelengths $\lambda = h/p$ and large radii as measured by α_0 . Thus highly accurate numerical algorithms and extensive grids are

required in intense field laser-molecule interactions.

One serious draw-back of absorbing boundary methods is the loss of high energy particle informations, as these are absorbed by the boundaries and never return. In the case of HOHG, return of the electron to its initial position constitutes the fundamental physics, so extremely large grids are required to obtain reliable HOHG spectra. Furthermore, multiphoton electron kinetic energy spectra, called ATI, and photo dissociation called ATD with concomitant Coulomb explosion requires collecting and analyzing all asymptotic fragments. This is impossible with absorbing boundary methods. In order to alleviate this problem, we have recently developed a new algorithm which circumvents the loss of information contained in the asymptotic wave function, by projecting, in a well defined domain, of size larger than α_0 , see Eq. (4), the exact asymptotic numerical wave function onto the exact solutions of the electron in a laser pulse, called Volkov states.¹⁸⁾ Such a procedure can only be implemented in 1-D H_2^+ models, *i.e.* $\Psi = \Psi(z, R, t)$, due to the large amount of data which needs to be stored for a complete study of the electron-nucleus dynamics of the simplest molecule H_2^+ in an intense laser pulse.

A numerical method for a molecule in elliptically and linearly polarized laser fields

Let us consider the H_2^+ molecule in two laser fields: one is elliptically polarized in the xy -plane, another is polarized linearly along the z -axis. The corresponding time-dependent Schrödinger equation has now the following form (in atomic units):

$$\begin{aligned} i \frac{\partial \psi}{\partial t} = & -\frac{2m_p + 1}{4m_p} \Delta_{x,y,z} \psi \\ & - \left[\frac{\psi}{|(x, y, z + R/2)|} + \frac{\psi}{|(x, y, z - R/2)|} \right] \\ & + \left[\left(1 + \frac{1}{2m_p + 1} \right) E_l(t) z \sin(\omega_z t) \right] \psi \\ & + \left[\left(1 + \frac{1}{2m_p + 1} \right) E_c(t) x \cos(\omega_c t) \right] \psi \\ & + \left[\left(1 + \frac{1}{2m_p + 1} \right) \epsilon E_c(t) y \sin(\omega_c t) \right] \psi. \end{aligned} \quad (19)$$

We will solve numerically the above equation using the finite difference method. We will present, in the following, our method of spatial and temporal discretization.

Spatial discretization

Because of the singularity of the potential at nuclei, we will use grids with spacings smaller near nuclei. Our mesh is defined *via* the following transformation of coordinates: $X = F(\tilde{x})$, $Y = G(\tilde{y})$, $Z = H(\tilde{z})$ and we use a homogeneous mesh in plane $(\tilde{x}, \tilde{y}, \tilde{z})$. Here, F , G and H are the functions having same form:

$$T(u) = u * \frac{u^n + s * p^n}{u^n + p^n} \quad (20)$$

where n is an even integer, $p \geq 0$ represents the refinement range and $0 \leq s \leq 1$ equal to $\min(DT/Du)$.

To improve the conservation of the norm (L_2), we use the following new function $\tilde{\psi} = \sqrt{F'G'H'}\psi$ as in Ref. 28. By multiplying (19) by $\sqrt{F'G'H'}$, we get $\frac{\partial}{\sqrt{F'}\partial\tilde{x}}\frac{\partial}{\sqrt{F'}\partial\tilde{x}}\frac{\tilde{\psi}}{\sqrt{F'}}$ instead of $\frac{\partial^2\psi}{\partial x^2}$. We discretize this operator using the finite-difference scheme:

$$\begin{aligned} & \frac{\partial}{\sqrt{F'}\partial\tilde{x}}\frac{\partial}{\sqrt{F'}\partial\tilde{x}}\frac{\tilde{\psi}}{\sqrt{F'}}(\tilde{x}_i) \\ & \simeq \left[\frac{1}{90h_x^2\sqrt{F'_iF'_{i-3/2}}\sqrt{F'_{i-3}}} \right] \tilde{\psi}_{i-3} \\ & - \left[\frac{3}{20h_x^2\sqrt{F'_iF'_{i-1}}\sqrt{F'_{i-2}}} \right] \tilde{\psi}_{i-2} \\ & + \left[\frac{3}{2h_x^2\sqrt{F'_iF'_{i-1/2}}\sqrt{F'_{i-1}}} \right] \tilde{\psi}_{i-1} \\ & + \left[\frac{3}{2h_x^2\sqrt{F'_{i+1}F'_{i+1/2}}\sqrt{F'_i}} \right] \tilde{\psi}_{i+1} \\ & - \left[\frac{3}{20h_x^2\sqrt{F'_{i+2}F'_{i+1}}\sqrt{F'_i}} \right] \tilde{\psi}_{i+2} \\ & + \left[\frac{1}{90h_x^2\sqrt{F'_{i+3}F'_{i+3/2}}\sqrt{F'_i}} \right] \tilde{\psi}_{i+3} \\ & - \frac{1}{90h_x^2F'_i} \left[\frac{1}{F'_{i+3/2}} + \frac{1}{F'_{i-3/2}} \right] \tilde{\psi}_i \\ & + \frac{3}{20h_x^2F'_i} \left[\frac{1}{F'_{i+1}} + \frac{1}{F'_{i-1}} \right] \tilde{\psi}_i \\ & - \frac{3}{2h_x^2F'_i} \left[\frac{1}{F'_{i+1/2}} + \frac{1}{F'_{i-1/2}} \right] \tilde{\psi}_i. \end{aligned} \quad (21)$$

This 7 point scheme is of order 5 and is symmetric. At singular points we use an average value of the potential $\frac{\int_V \frac{1}{r} dV}{V}$ in a small sphere surrounding the nucleus. This is equivalent to redistributing the nucleus charge within this sphere but is superior to using regularized potentials.

Temporal discretization

We use the following notation:

$$\begin{aligned} \alpha &= -\frac{2m_p + 1}{4m_p} & \beta &= -\frac{1}{m_p} \\ C_x &= \frac{\alpha\partial}{\sqrt{F'}\partial\tilde{x}}\frac{\partial}{\sqrt{F'}\partial\tilde{x}}\frac{1}{\sqrt{F'}} + \frac{2m_p + 2}{2m_p + 1} E_c(t)x \cos(\omega_c t) \\ C_y &= \frac{\alpha\partial}{\sqrt{G'}\partial\tilde{y}}\frac{\partial}{\sqrt{G'}\partial\tilde{y}}\frac{1}{\sqrt{G'}} + \frac{2m_p + 2}{2m_p + 1} E_c(t)y \sin(\omega_c t) \\ C_z &= \frac{\alpha\partial}{\sqrt{H'}\partial\tilde{z}}\frac{\partial}{\sqrt{H'}\partial\tilde{z}}\frac{1}{\sqrt{H'}} + \frac{2m_p + 2}{2m_p + 1} \epsilon E_l(t)z \cos(\omega_l t) \\ C_V &= -\frac{1}{|(x, y, z - R/2)|} - \frac{1}{|(x, y, z + R/2)|} \\ P_x &= \left[1 + i\frac{\delta t}{4}C_x \right]^{-1} \exp(-i\frac{\delta t}{6}C_V) \left[1 - i\frac{\delta t}{4}C_x \right] \\ P_y &= \left[1 + i\frac{\delta t}{4}C_y \right]^{-1} \exp(-i\frac{\delta t}{6}C_V) \left[1 - i\frac{\delta t}{4}C_y \right] \\ P_z &= \left[1 + i\frac{\delta t}{4}C_z \right]^{-1} \exp(-i\frac{\delta t}{6}C_V) \left[1 - i\frac{\delta t}{4}C_z \right]. \end{aligned} \quad (22)$$

We propagate (in time) the wave function using the SPLIT operator method²⁵⁾ and the Crank-Nicholson scheme²⁹⁾

$$\tilde{\psi}(t + \delta t) = P_z P_y P_x^2 P_y P_z \tilde{\psi}(t). \quad (23)$$

These are second order schemes in δt (see Ref. 25) and show complete separability of the integration scheme thus facilitating future projection onto Volkov states.

Tests for H_2^+ for a linear polarization case

We apply, in the following, our new numerical method for computation of the ionization rates of H_2^+ in a linearly polarized laser field. We initialize our simulation using the electronic ground state of H_2^+ obtained numerically with the help of imaginary time propagation method. The cylindrical symmetry of the problem simplifies much the problem. Because we have $\psi(x, y, z, t) = \psi(-x, y, z, t)$ and $\psi(x, y, z, t) = \psi(x, -y, z, t)$ for all (x, y, z, t) , we can use the following conditions to save the computer time:

$$\begin{aligned} \frac{\partial\psi}{\partial x}(y=0) &= 0 \\ \frac{\partial\psi}{\partial y}(x=0) &= 0. \end{aligned} \quad (24)$$

This saves the computer time by factor 4. We used the laser intensity $I = 10^{14}$ W/cm² and the wavelength $\lambda = 228$ nm. We have studied first the convergence in one direction, by using rather narrow ranges, *i.e.*: $|x|_{\max} = |y|_{\max} = 10.25$, $|z|_{\max} = 125$ a.u. We used the following values of other parameters: $\min(\Delta x) = \min(\Delta y) = 0.5$, $\text{ratio}_x = \text{ratio}_y = 1$, $\Delta t = 0.02$, and the refinement range of our mesh was $|z| \leq 26$. Our laser was smoothly turned-on during 5 cycles. We use the following notation: $\text{ratio}_x = \frac{\min(\Delta x)}{\max(\Delta x)}$. The ionization rates given in the first line of Table 1 were obtained without using the coordinate transformation (*i.e.* equidistant grid), and required two times more points in z direction than in the second line which implicate consuming two times more CPU-time. The third result which use a more finer grid with the same transformation parameter as the second one has been added to verify the convergence. We conclude that coordinate transformation reduced the computation time without decreasing the accuracy. For comparison, we present also results obtained using the alternation-direction implicit method (ADI)²⁸⁾ which gives the same result. Next, we increased the xy range ($|x|_{\max}, |y|_{\max}$) to demonstrate the convergence of calculated rates (see Table 2). Here, the last result has been obtained with absorption in all three directions (x, y, z) , but the other calculations have not used absorption in the transverse direction (x, y) . Our ionization rates are close to that obtained by S.Chelkowski *et al.*²⁴⁾ using a 3-D code with absorption in the z -direction only, and we have used 574 points in the z -direction in the

Table 1. Effect of the coordinate transformation in z direction.

Method	$\min(\Delta z)$	ratio_z	Ionization rate (s ⁻¹)
SPLIT	0.214	1.0	$5.2 * 10^{13}$
SPLIT	0.214	0.2	$5.2 * 10^{13}$
SPLIT	0.107	0.2	$5.2 * 10^{13}$
ADI	0.214	0.2	$5.2 * 10^{13}$

Table 2. Comparison of different computation methods.

$\max(x) = \max(y)$	Ionization rate
10.25	$5.2 * 10^{13}$
15.25	$3.6 * 10^{13}$
20.75	$2.98 * 10^{13}$
62.75	$2.83 * 10^{13}$

place of 2048 points used by S.Chelkowski *et al.*²⁴⁾ (they have used the Bessel functions in the xy direction). We note that due to expansion of the wave function in the transverse (x, y) direction, convergence requires at least half as many points in the (x, y) -direction as in the z -direction parallel to the field.

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