The dynamics of the ionization of atoms exposed to strong low-frequency fields

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Abstract. We present a theoretical study of strong-low-frequency-field ionization of atoms in the intermediate regime where the Keldysh parameter is close to one. For that purpose we have developed a novel method based on an *explicit* algorithm for the propagation of the time-dependent Schrödinger equation which is known to be very stiff. This method overcomes the stiffness by taking into account the eigenfrequencies of the interacting system. With the help of this approach we are able in particular to investigate the ionization of the hydrogen atom by ultra-short (25 fs) 790 nm laser pulses at intensities 0.5–6×10^{14} W/cm^2.

1. Introduction

Atomic photoionization under intense laser radiation has been intensively investigated. It is widely perceived that the ionization process occurs through multiphoton absorption or tunnelling [1–4]: in the low-intensity/short-wavelength limit, Above-Threshold Ionization (ATI) is the absorption of photons beyond the minimum required for ionization. In the high-intensity/long-wavelength limit, Keldysh theory suggests that the valence electron escapes by tunnelling through the barrier created by the time-dependent bending of the Coulomb potential by the field followed by an oscillating motion of the free electron induced by the field of frequency ω and intensity I. These two limits are linked through the dimensionless parameter $\gamma = \sqrt{I_p/2U_p}$, where I_p is the ionization potential and $U_p = I^2/4\omega^2$ is the pondemorotive energy.

At intensities of the order of $10^{14} - 10^{15}$ W/cm^2 and infrared frequencies, there is no clear separation between the regime where multiphoton processes dominate the ionization of atoms exposed to such fields and the strong field limit dominated by tunnel ionization. In fact, there is a subtle interplay between the two mechanisms as indicated by the fact that the Keldysh parameter γ is close to one. From the theoretical point of view, de Bohan *et al.* [5] have shown, in the case of the interaction of atomic hydrogen with such fields, that the low-energy part of the ATI spectrum does not result from tunnel ionization but rather involves some multiphoton transitions. This explains why the tunnelling formula for the total ionization rate gives results that disagree with those obtained by solving the Time-Dependent Schrödinger Equation (TDSE). From the experimental side, this was confirmed by Rudenko *et al.* [6] who found that the ionization of rare atoms in this regime shows evidence of resonance structures in the electron ATI spectrum which are thought to be characteristic of the multiphoton regime. By contrast,

another experiment [7] in the same regime shows that the ionization rate follows adiabatically the oscillations of the field. Understanding the interplay of such mechanisms poses a challenge for both experiment and theory. From the theoretical point of view, one of the main difficulties to deal with this regime is the solution of the TDSE, which requires an accurate treatment of the very high-order angular momentum coupling in this regime of low frequency, rather long pulse duration and high field intensity. Due to this and the infinite range of the coulombic interactions, this requires either very large grids or big bases. In both cases, one has to solve large systems of coupled first order differential equations that are well known to be stiff [8]. By this, it is meant that the time step decreases rapidly with the increasing size of the system. The origin of the stiffness is clear: by increasing the size of the grid or the basis, the diagonalization of the atomic Hamiltonian generates large positive energy eigenvalues. It is precisely the highest eigenvalue that controles the time step while leading to highly oscillating solutions. In order to overcome this problem, one can use implicit time propagation methods. This requires to solve very large systems of algebraic equations at each time step. A typical example of such an implicit scheme is the Crank-Nicholson algorithm which is used in grid methods based on finite differences [9]. This propagation method is tractable when the system is banded but as soon as the complexity of the atomic system increases, the bandwidth of the system increases rapidly together with the computation time. An alternative way of overcoming the stiffness of the problem is to propagate in the atomic basis where the atomic Hamiltonian is diagonal. Time propagating in the atomic basis has three advantages: first, it is possible to eliminate from the propagation, very high energy eigenstates that play a minor role in the dynamics. Second, it allows one to work in the interaction picture where the free evolution of the atomic system is somehow subtracted. And finally, explicit methods which involve only matrix-vector products are numerically stable in this case. However, this method requires the diagonalization of large matrices which is a computationally very demanding problem.

In this contribution, we present a theoretical study of strong-field ionization of atoms in the intermediate regime where the Keldysh parameter is close to 1. We consider the interaction of atomic hydrogen with a 790 nm laser pulse which duration is of the order of 25 femtoseconds and the intensity ranges from 5×10^{13} to 6×10^{14} W/cm^2. The propagation of the spectral representation of the TDSE is accomplished through a very efficient novel *explicit method* which is briefly described in Sec. 2. Our results are presented in Sec. 3.

2. Time propagation algorithm

In this section, a brief review of the method proposed in [10, 11] is given. Let us start with the m-dimensional *stiff* first order differential equation

$$\boldsymbol{y}' = \boldsymbol{f}(x, \boldsymbol{y}), \qquad \boldsymbol{y} = (y_1, y_2, \ldots, y_m), \tag{1}$$

where $\boldsymbol{f}(x, y)$ is in general a complex m-dimensional function.

The stiffness of the equation leads to a solution $\boldsymbol{y}(x)$ which is an oscillating function. In a given interval (x_n, x_{n+1}), $x_{n+1} = x_n + h$, with h a small number, $\boldsymbol{y}(x)$ is approximated by the function

$$\boldsymbol{F}(x) = (I - e^{\Omega_1 x})\boldsymbol{a} - (I - e^{-\Omega_2 x})\boldsymbol{b} + \boldsymbol{c}, \tag{2}$$

with I the identity matrix, $\Omega_i = \mathrm{diag}(\omega_1^{(i)}, \ldots, \omega_m^{(i)})$, $i = 1, 2$, and $\boldsymbol{a}, \boldsymbol{b}, \boldsymbol{c}$ constant vectors. The complex numbers $\omega_1^{(i)}, \ldots, \omega_m^{(i)}$, $i = 1, 2$ are called *stiffness parameters*. Assuming that $\boldsymbol{F}(x)$ coincides with $\boldsymbol{y}(x)$ at x_n and x_{n+1}, that $\boldsymbol{F}'(x)$ coincides with $\boldsymbol{f}(x, \boldsymbol{y})$ at x_n, and that $\boldsymbol{F}''(x)$ coincides with $\boldsymbol{f}'(x, \boldsymbol{y})$ at x_n, the solution $\boldsymbol{y}_{n+1} = \boldsymbol{y}(x_{n+1})$ at x_{n+1} can be expressed recursively in terms of $\boldsymbol{y}_n = \boldsymbol{y}(x_n)$, $\boldsymbol{f}_n = \boldsymbol{f}(x_n, \boldsymbol{y}_n)$ and $\boldsymbol{f}_n^{(1)} = d\boldsymbol{f}/dx|_{x=x_n}$ according to

$$\boldsymbol{y}_{n+1} = \boldsymbol{y}_n + R\boldsymbol{f}_n + S\boldsymbol{f}_n^{(1)}. \tag{3}$$

R and S are diagonal matrices which can be written in terms of the stiffness parameters:

$$R = \Omega_2\Phi - \Omega_1\Psi, \qquad S = \Phi + \Psi, \qquad (4)$$

where Φ and Ψ are diagonal matrices whose nonzero entries are

$$\Phi_i = \frac{e^{\omega_i^{(1)}h} - 1}{\omega_i^{(1)}(\omega_i^{(1)} + \omega_i^{(2)})} \quad \text{and} \quad \Psi_i = \frac{e^{-\omega_i^{(2)}h} - 1}{\omega_i^{(2)}(\omega_i^{(1)} + \omega_i^{(2)})}. \qquad (5)$$

The recursive relation (3) depends on the so far unknown stiffness matrices Ω_1 and Ω_2. However, these matrices can be written in terms of the function $\boldsymbol{f}(x_n, \boldsymbol{y}_n)$ and its derivatives up to 3rd order at x_n. This is obtained after substituting the Taylor expansion of $\boldsymbol{y}_{n+1} = \boldsymbol{y}(x_n + h)$, and the Maclaurin series of $\exp(\Omega_1 h)$ and $\exp(-\Omega_2 h)$ in (3) and equating the coefficients of h^k, $k = 0, \ldots, 4$. The components of the stiffness matrices obtained after solving these equations read [10]

$$\omega_i^{(1)} = \frac{1}{2}\left[-D_i + \sqrt{D_i^2 + 4E_i}\right] \quad \text{and} \quad \omega_i^{(2)} = \omega_i^{(1)} + D_i, \qquad (6)$$

where D_i and E_i, $i = 1, \ldots, m$ are given in terms of the respective components $f_{in}^{(k)}$ of the derivatives $\boldsymbol{f}_n^{(k)}$, $k = 0, 1, 2, 3$, of $\boldsymbol{f}(x, \boldsymbol{y})$ at $x = x_n$ by

$$D_i = \frac{f_{in}^{(0)}f_{in}^{(3)} - f_{in}^{(1)}f_{in}^{(2)}}{f_{in}^{(1)}f_{in}^{(1)} - f_{in}^{(0)}f_{in}^{(2)}}, \quad \text{and} \quad E_i = \frac{f_{in}^{(1)}f_{in}^{(3)} - f_{in}^{(2)}f_{in}^{(2)}}{f_{in}^{(1)}f_{in}^{(1)} - f_{in}^{(0)}f_{in}^{(2)}}, \quad i = 1, \ldots, m. \qquad (7)$$

If the denominator of the previous expressions is zero, special care has to be taken into account [12].

The implementation of the recursion (3) is now rather simple. It requires the calculation of the function $\boldsymbol{f}_n$ and its derivative $\boldsymbol{f}_n^{(1)}$ at each value of x_n. For the stiffness matrices Ω_1 and Ω_2, and thus also for the matrices R and S, the derivatives $\boldsymbol{f}_n^{(2)}$ and $\boldsymbol{f}_n^{(3)}$ are also needed. Ω_1 and Ω_2 have to be calculated in principle at each integration step, since they characterize the local frequencies of the solution $\boldsymbol{y}(x)$ [12]. In addition, the truncation error [10, 12]

$$T_{n+1} = \frac{h^5}{5!}\frac{1}{\omega_1 + \omega_2}\left[(\omega_1 + \omega_2)f_n^{(4)} + (\omega_2^4 - \omega_1^4)f_n^{(1)} - (\omega_1^4\omega_2 + \omega_1\omega_2^4)f_n^{(0)}\right] + \mathcal{O}(h^6) \qquad (8)$$

can be used to control the size of the integration step, e.g., by imposing a boundary criterion for $|T_n|$. For this also the derivative $\boldsymbol{f}_n^{(4)}$ must be provided.

3. Resonance structures in the tunnelling regime of ATI of hydrogen atoms

In this section we consider hydrogen atoms exposed to intense low frequency femtosecond laser pulses. We consider linearly polarized light, and apply the dipole approximation. In the velocity gauge the interaction operator reads

$$V(t) = \boldsymbol{A}(t) \cdot \boldsymbol{p}, \qquad (9)$$

where the vector potential takes the form $\boldsymbol{A}(t) = A_0(t)\sin(\omega t)\boldsymbol{e}_z$, for $-\tau/2 \leq t \leq \tau/2$, and zero outside this time interval. $A_0(t) = A_0 \cos^2(\pi t/\tau)$ is the envelope containing n_c optical cycles, ω is the pulse angular frequency, $\tau = 2\pi n_c/\omega$ is the pulse duration and $\boldsymbol{e}_z$ is the unit vector along the z-axis in the laboratory frame. The Hamiltonian of the driven atom is thus

$$H(t) = H_0 + V(t), \qquad (10)$$

with

$$H_0 = \frac{p^2}{2} - \frac{Z}{r}. \tag{11}$$

We use a spectral representation of the Hamiltonian in terms of spherical harmonics and Coulomb Sturmian functions defined by

$$S_{n\ell}^{(\alpha)}(r) = \sqrt{\frac{(n-\ell-1)!}{2(n+\ell)!}} \left(\frac{2r}{\alpha}\right)^{\ell+1} L_{n-\ell-1}^{2\ell+1}\left(\frac{2r}{\alpha}\right) e^{-r/\alpha}, \tag{12}$$

where $L_n^{(\beta)}(x)$ are the Laguerre polynomials and α is a real number called the dilation parameter.

Substitution of the expansion

$$\psi(\boldsymbol{r}, t) = \sum_{n\ell} c_{n\ell}(t) \frac{S_{n\ell}^{(\alpha)}(r)}{r} Y_{\ell m}(\theta, \varphi) \tag{13}$$

of the wave packet $\psi(\boldsymbol{r}, t)$ in the TDSE leads to the following matrix representation:

$$iS\frac{d\boldsymbol{y}}{dt} = \mathcal{A}\boldsymbol{y} + g(t)\mathcal{V} \tag{14}$$

where S is the overlap matrix, $\mathcal{A}$ is the matrix representation of the unperturbed Hamiltonian of the system, $g(t) = A_0 \cos^2(\pi t/\tau) \sin(\omega t)$ is the scalar time-dependent part of the field interaction (9) and $\mathcal{V}$ is the matrix representation of the dipole operator p_z. $\boldsymbol{y}$ is the vector of the coefficients $c_{n\ell}(t)$.

In order to solve numerically the TDSE, the basis (12) has to be truncated: $n_{\max}$ is the number of Sturmian functions per angular configuration for each of the values of $\ell = 0, \ldots, \ell_{\max}$.

We consider the ionization process from the ground state of atomic hydrogen exposed to a laser pulse of 790 nm ($\omega = 0.057$ a.u.), which is chosen to match the Ti:sapphire lasing frequency, and peak intensities: from 5×10^{13} to 6×10^{14} W/cm^2. The Keldysh parameter γ for these choices ranges from $\gamma = 1.510$ to $\gamma = 0.435$. We consider a pulse of $n_c = 10$ optical cycles which corresponds to a pulse duration of about 25 fs (1102.3 a.u.). The laser parameters are chosen in this way in order to match those of Rudenko's experiment [6]. The minimum number of photons needed for ionization ranges from 10 to 31, which implies a high-order angular momentum coupling. Therefore, an accurate solution of the TDSE requires a rather big basis containing a large number of Sturmian functions (up to $n_{\max} = 800$) and large number of angular configurations (up to $\ell_{\max} = 70$).

Fig. 1 displays the energy spectrum for $I = 5 \times 10^{13}$ (a), 10^{14} (b), 4×10^{14} (c), and 6×10^{14} W/cm^2 (d) obtained after propagation of the TDSE with the help of the method described above. The accuracy and efficiency of this method has been confronted with a diagonally implicit Runge-Kutta propagator [12]. Our approach is typically one order of magnitude faster than the implicit Runge-Kutta method. The results for $I = 5 \times 10^{13}$ and 10^{14} W/cm^2 were obtained with $\ell_{\max} = 50$, and $n_{\max} = 300$ and 400, respectively. For higher field intensities some features of the energy spectrum at low energy can be already seen by coupling at least 40 angular momenta with $n_{\max} = 400$, however the resolution of structures at higher energies require larger bases (Fig. 1(c), (d)).

For a field intensity $I = 5 \times 10^{13}$ W/cm^2 the ionization process is a multiphoton process as expected. The peaks in the spectrum are precisely located at positions given by
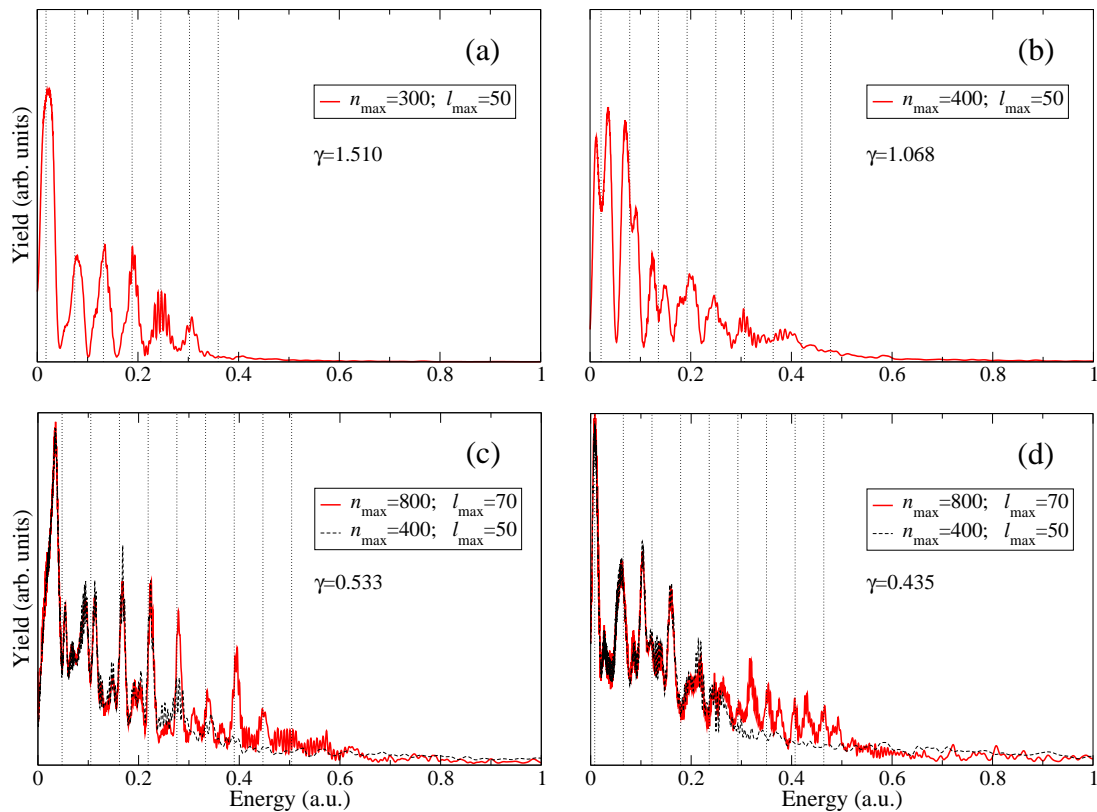
$$E_n = n\omega - U_p - I_p, \tag{15}$$

Figure 1. Electron energy distributions for single ionization of the hydrogen atom by 25 fs pulses at $I = 5 \times 10^{13}$ (a), 10^{14} (b), 4×10^{14} (c), and $6 \times 10^{14}\,\mathrm{W/cm^2}$ (d). The dotted vertical lines denote the expected position of the peaks according to Eq. (15).

which are highlighted in Fig. 1 by the vertical dotted lines. As the intensity increases new structures appear at the low-energy part of the spectrum. At $I = 10^{14}\,\mathrm{W/cm^2}$ the peaks associated to $n = 1, 2$ and 3 are splitted (Fig. 1(b)). At higher intensities the spectrum consists of peaks which are rather irregularly distributed, specially in the case of $I = 6 \times 10^{14}\,\mathrm{W/cm^2}$.

Understanding the origin of these structures is not the purpose of the present contribution. It requires further investigation and will be presented elsewhere. Indeed, we are also exploring this problem under two other approaches: a Floquet formalism combined with complex rotation which allows us to access the resonances induced by the field [13] and a model calculation based on series of separable potentials in momentum space, each of them supporting a bound state of the hydrogen atom [14].

4. Conclusions

We presented a study of the ionization process of hydrogen atoms by intense fields in a regime where the Keldysh parameter is close to 1. For this purpose we developed a method for the propagation of the TDSE which includes an efficient implementation of an explicit algorithm for the solution of stiff differential equations. Some structures in the ATI energy spectrum were found which can be only partially associated to multiphoton ionization peaks. The nature of the remaining structures still remains unclear and deserves further investigation.

Our calculations show a stupendous efficiency compared to diagonally implicit propagation algorithms. This opens new perspectives for the treatment of more complicate ionization processes like two-photon ionization of helium [15–23], where the non-linearity of this process

poses tremendous experimental difficulties that so far prevent the measurement of accurate total and differential cross sections [20, 21].

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