

On the role of the Coulomb potential in strong field atomic ionization dynamics

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Abstract

In this paper, we present a model aimed at exploring the role of the Coulomb potential in the mechanism of ionization of atomic hydrogen exposed to a strong low frequency pulsed laser field. Our approach is based on the solution of the time-dependent Schrödinger equation in momentum space. Although we are in a frequency and intensity regime where tunnelling is expected to dominate, our results indicate that the atomic structure associated to the Coulomb potential plays a significant role for low energy ejected electrons.

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1. Introduction

In his pioneering work [1] on the ionization dynamics of atoms exposed to strong low frequency electric fields, Keldysh introduced the so-called adiabaticity parameter $\gamma = (\omega/E)\sqrt{2I_p}$ where ω is the laser field frequency with amplitude E and I_p is the ionization potential of the atom. This parameter allows to distinguish two regimes of field intensity: the perturbative regime ($\gamma > 1$) where multiphoton processes dominate, and the strong field limit ($\gamma < 1$) where tunnel ionization is assumed to set in. For $\gamma < 1$ however, the pure tunnel ionization rate does not agree at all with the ionization rate calculated by solving numerically the corresponding time-dependent Schrödinger equation. de Bohan et al. [2] have shown that the slow ejected

electrons whose energy is less than twice the ponderomotive potential do not experience tunnelling. In other words, the tunnel ionization mechanism that satisfactorily accounts for many features of the high energy part of the electron spectrum fails to explain the low energy part that in fact gives the dominant contribution to the ionization rate. This raises the question of the actual role of the Coulomb potential in the strong field limit. If tunneling does not take place for slow electrons, the influence of the atomic structure must be taken into account to understand the ejection mechanism in that case. The role of the atomic structure has been clearly demonstrated in recent experiments by Rudenko and co-workers [3]. They measured above-threshold ionization (ATI) spectra for various atoms in a regime where tunnelling is in principle the dominant ionization mechanism. The first ATI peaks exhibit a fine structure which can be unambiguously attributed to intermediate resonances.

In this paper, we analyze the role of the Coulomb potential (and therefore the atomic structure) in the strong field limit by means of a model. Our approach is based on the solution of the time-dependent Schrödinger equation (TDSE) in momentum

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space. The kernel associated to the Coulomb potential which, in this space, is non local, is replaced by a series of N separable potentials that support the first N exact hydrogen bound states. Within this approximation, the solution of the TDSE reduces to the solution of N coupled 1D Volterra integral equations of the second kind. An extremely fast and accurate numerical code has been developed to solve this system [4].

2. Theory

2.1. Construction of separable potentials

We present here a general scheme for the factorization of the Coulomb potential. It consists in replacing the Coulomb potential in momentum space by a sum of non-local separable potentials built in such a way that they keep the correct characteristics of the exact Coulomb potential for the bound electrons. It is therefore providing a way of analyzing how the atomic structure associated to the Coulomb potential influences the ionization dynamics.

We actually consider a symmetric attractive separable potential (the atomic units $\hbar = e = m_e = 1$ are used throughout):

$$V(\mathbf{p}, \mathbf{p}') = - \sum_{n=1}^N v_n(\mathbf{p}) v_n^*(\mathbf{p}'), \quad (1)$$

which we are going to use as a separable analogue of the Coulomb potential $V(r) = -Z/r$ in momentum-space. We demand from this potential to reproduce N eigenfunctions and eigenenergies of the Coulomb spectrum. The momentum space representation of the bound state φ_j satisfies the following equation:

$$\left(\varepsilon_j - \frac{1}{2} p^2 \right) \varphi_j(\mathbf{p}) + \sum_{n=1}^N a_{jn} v_n(\mathbf{p}) = 0, \quad (2)$$

where

$$a_{jn} = \int \frac{d\mathbf{p}'}{(2\pi)^3} v_n^*(\mathbf{p}') \varphi_j(\mathbf{p}'). \quad (3)$$

Eq. (2) may be written in matrix form as follows:

$$\Phi = -\mathbf{A}\mathbf{V}, \quad (4)$$

where \mathbf{A} is a symmetric $N \times N$ matrix of elements a_{jn} . Φ and \mathbf{V} are $N \times 1$ vectors whose elements are the $v_n(\mathbf{p})$ and $(\varepsilon_j - (1/2)p^2)\varphi_j(\mathbf{p})$, respectively. Consequently,

$$\mathbf{V} = -\mathbf{A}^{-1}\Phi. \quad (5)$$

On the other hand, from Eq. (3) we have:

$$\mathbf{A} = (\mathbf{A}^T)^{-1}\mathbf{\Gamma}, \quad \text{or} \quad \mathbf{A}\mathbf{A}^T = \mathbf{\Gamma}, \quad (6)$$

where \mathbf{A}^T denotes the transpose matrix \mathbf{A} . The elements of the symmetric $N \times N$ matrix $\mathbf{\Gamma}$ are given by:

$$\begin{aligned} \gamma_{ij} &= - \int \frac{d\mathbf{p}}{(2\pi)^3} \varphi_i^*(\mathbf{p}) \left(\varepsilon_j - \frac{1}{2} p^2 \right) \varphi_j(\mathbf{p}) \\ &= Z \int \frac{d\mathbf{r}}{r} \tilde{\varphi}_i^*(\mathbf{r}) \tilde{\varphi}_j(\mathbf{r}), \end{aligned} \quad (7)$$

where $\tilde{\varphi}_j(\mathbf{r})$ is the wavefunction associated to a Coulomb bound state in the configuration space.

The obtained potential is not obligatory unique. The whole family of equivalent potentials can be generated in the same way.

2.2. One bound state potential

In this section and for the sake of clarity, we describe our model in more details in the case where $N = 1$ (only the ground state taken into account). The TDSE in momentum space for atomic hydrogen exposed to a laser field linearly polarized along the unit vector \mathbf{e} writes:

$$\begin{aligned} \left[i \frac{\partial}{\partial t} - \frac{p^2}{2} - \frac{1}{c} A(t)(\mathbf{e}\mathbf{p}) \right] \Phi(\mathbf{p}, t) \\ - \int \frac{d\mathbf{p}'}{(2\pi)^3} V(\mathbf{p}, \mathbf{p}') \Phi(\mathbf{p}', t) = 0. \end{aligned} \quad (8)$$

In (8), $\Phi(\mathbf{p}, 0) \equiv \varphi_0(p) = 8\sqrt{\pi}/(p^2 + 1)^2$, and the kernel $V(\mathbf{p}, \mathbf{p}')$ is given by:

$$V(\mathbf{p}, \mathbf{p}') = - \frac{4\pi}{|\mathbf{p} - \mathbf{p}'|^2}. \quad (9)$$

The wavepacket is normalized in the following way:

$$\int \frac{d\mathbf{p}}{(2\pi)^3} |\Phi(\mathbf{p}, t)|^2 = 1. \quad (10)$$

We assume that the laser pulse has a sine square envelope and define the potential vector as follows:

$$\begin{aligned} A(t) &= \frac{c}{\omega} \sqrt{\frac{I}{I_0}} \sin^2 \left(\pi \frac{t}{T} \right) \sin(\omega t), \quad (0 \leq t \leq T); \\ A(t) &= 0, \quad (t \geq T). \end{aligned} \quad (11)$$

In Eq. (11), I is the peak intensity of the laser pulse and $I_0 = 3.5 \times 10^{16} \text{ W/cm}^2$ is the atomic unit of intensity. ω is the laser frequency and $T = 2\pi n/\omega$, the total pulse duration with n indicating the total number of optical cycles.

Let us perform the following substitution for the Coulomb kernel in Eq. (8):

$$V(\mathbf{p}, \mathbf{p}') \rightarrow - \frac{16\pi}{(p^2 + 1)(p'^2 + 1)}. \quad (12)$$

This potential is unique and reproduces the ground state of hydrogen. We obtain instead of (8)

$$\left[i \frac{\partial}{\partial t} - \frac{p^2}{2} - \frac{1}{c} A(t)(\mathbf{e}\mathbf{p}) \right] \Phi(\mathbf{p}, t) + F(t) \frac{16\pi}{(p^2 + 1)} = 0, \quad (13)$$

where

$$F(t) = \int \frac{d\mathbf{p}'}{(2\pi)^3} \frac{\Phi(\mathbf{p}', t)}{(p'^2 + 1)}.$$

Its solution takes the form:

$$\begin{aligned} \Phi(\mathbf{p}, t) = & \exp\left(-i\frac{p^2}{2}t + ib(t)(\mathbf{ep})\right) \\ & \times \left[\Phi(\mathbf{p}, 0) + \frac{16i\pi}{(p^2 + 1)} \int_0^t F(\xi) \right. \\ & \left. \times \exp\left(i\frac{p^2}{2}\xi - ib(\xi)(\mathbf{ep})\right) d\xi \right], \end{aligned} \quad (14)$$

with $b(t) = -(1/c) \int_0^t A(\xi) d\xi$. By inserting (14) into (13), we get a Volterra integral equation for the function $F(t)$:

$$F(t) = F_0(t) + \int_0^t K(t, s)F(s) ds, \quad (15)$$

where

$$F_0(t) = \int \frac{d\mathbf{p}}{(2\pi)^3} \frac{\Phi(\mathbf{p}, 0)}{(p^2 + 1)} \exp\left(-i\frac{p^2}{2}t + ib(t)(\mathbf{ep})\right), \quad (16)$$

and

$$\begin{aligned} K(t, s) = & 16i\pi \int \frac{d\mathbf{p}}{(2\pi)^3} \frac{1}{(p^2 + 1)^2} \\ & \times \exp\left(-i\frac{p^2}{2}(t - s) + i(b(t) - b(s))(\mathbf{ep})\right). \end{aligned} \quad (17)$$

The function $F(t)$ contains the complete information about the dynamics of the ionization process.

All integrals present in the above equations can be calculated analytically with the basic formula:

$$\begin{aligned} I(x, y, \gamma) = & \int_{-\infty}^{+\infty} \frac{p dp}{p^2 + \gamma} e^{-ixp^2 + iyp} \\ = & \frac{i\pi}{2} e^{ix\gamma} \left\{ e^{-y\sqrt{\gamma}} \operatorname{erfc}\left(e^{i\pi/4} \sqrt{\gamma x} - e^{-i\pi/4} \frac{y}{2\sqrt{x}}\right) \right. \\ & \left. - e^{y\sqrt{\gamma}} \operatorname{erfc}\left(e^{i\pi/4} \sqrt{\gamma x} + e^{-i\pi/4} \frac{y}{2\sqrt{x}}\right) \right\}. \end{aligned} \quad (18)$$

Here,

$$\operatorname{erfc}(z) = 1 - \frac{2z}{\sqrt{\pi}} \int_0^1 e^{-z^2 t^2} dt,$$

is one of the definitions of the complementary error function. Let us define

$$\mathcal{I}_n(x, y, \gamma) = \frac{1}{2iy} \frac{(-1)^{n-1}}{(n-1)!} \frac{\partial^{n-1}}{\partial \gamma^{n-1}} I(x, y, \gamma). \quad (19)$$

All observable values may be written in term of this function. By projecting the wavepacket on the ground state $\varphi_0(p)$, we obtain the amplitude $c_0(t) = \langle \varphi_0 | \Phi(t) \rangle$ and the probability $|c_0(t)|^2$ for our “hydrogen atom” to stay in the ground state. The ionization amplitude $c(\mathbf{k}, t) = \langle \varphi^+(\mathbf{k}) | \Phi(t) \rangle$ can also be written in terms of this function. Finally, the differential probability for an electron to have the energy E can be determined in terms of the spectral density $D(E, t)$:

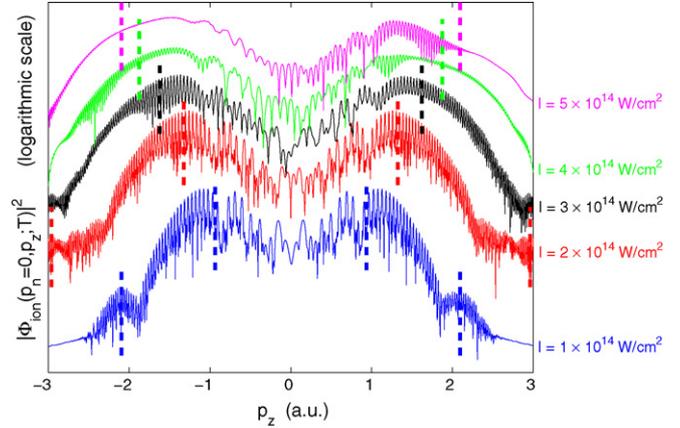


Fig. 1. Ionization probability density $|\Phi_{\text{ion}}(p_n = 0, p_z; T)|^2$ at the end of the pulse for different intensities, and $\omega = 0.057$ a.u. The vertical dashed lines indicate the value of p_z corresponding to $2U_p$ and $10U_p$. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of the article.)

$$dP = D(E, t) dE, \quad D(E, t) = \frac{\sqrt{2E}}{(2\pi)^3} \int |c(\mathbf{k}, t)|^2 d\Omega_k. \quad (20)$$

Obviously, the total ionization yield is $P(t) = 1 - |c_0(t)|^2$.

3. Results and discussion

In all our results, the frequency is fixed at 0.057 a.u. (Ti-Saphir laser) and the duration of the pulse is 8 optical cycle.

We first consider the case where only one bound state (the ground state) is included. This model actually reproduces all well known features of the above-threshold ionization spectra and of the ionization probability. There are however some interesting and unexpected observations requiring a deeper analysis.

In Fig. 1, we show the longitudinal density $|\Phi_{\text{ion}}(p_n = 0, p_z; T)|^2 = |\Phi(\mathbf{p}, T) - c_0(T)\varphi_0(p)|^2$ in logarithmic scale, as a function of the canonical momentum along the polarization axis

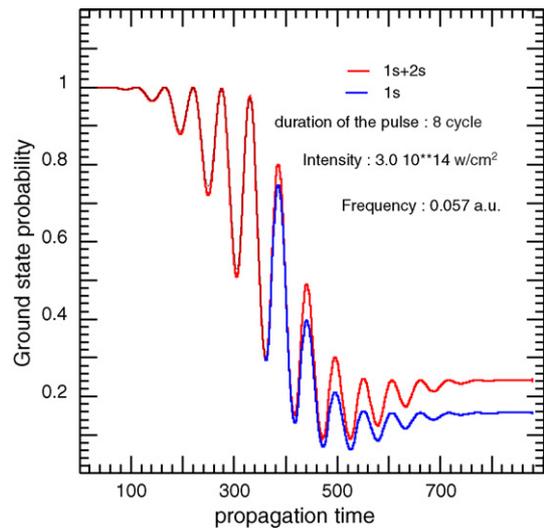


Fig. 2. Probability $|C_0(t)|^2$ that the atomic system stays in its ground state. Separable potential supports respectively $-1s$ state for blue curve and $1s+2s$ states for red curve. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of the article.)

p_z at the end of the interaction. The pulse intensity varies between $I = 1 \times 10^{14}$ W/cm² and $I = 5 \times 10^{14}$ W/cm². The vertical dashed lines indicate in each case, the value of the canonical momentum corresponding to $2U_p$ and $10U_p$ where $U_p = I/(4\omega^2 I_0) = 1/(4\gamma^2)$ is the ponderomotive potential. Above $2U_p$, the results are in qualitative agreement with those corresponding to the exact Coulomb potential. Below $2U_p$ however, the results obtained with our model strongly underestimate what is expected with the exact Coulomb potential [2]. This raises the question of the role of the other atomic hydrogen bound states in the mechanism of ejection of low energy electrons. This question stays unsolved within this one-level model and calls for the generalization of the present calculation to the case where more than one component are included in the expansion of the Coulomb potential kernel. Note that the asymmetry that is observed at high peak intensity is due to the fact that the time it takes to the system to ionize is very short.

The extension of the present approach to a multi-level model is in progress. Our first results show clear evidences of the role of the atomic structure on the ionization mechanism. In order to illustrate this point we show in Fig. 2, the time evolution of the ground state population obtained within a one-state (1s) and a two-state (1s, 2s) model. The presence of the 2s-state

leads already to a significant trapping of population into the ground state. Clearly, further investigations are necessary. We are however confident that our approach will eventually shed some light on the actual role of the Coulomb potential in the emission of slow electrons. So far all the calculations that have been performed to solve this problem were fully numerical making the extraction of valuable information on the dynamics very difficult. Here, numerics reduce to the solution of 1D integral equations.

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