

Extraction of the absolute value of the photoelectron spectrum probability density by means of the resolvent technique

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In this work we consider the theoretical treatment of the interaction of a laser pulse with an atom and focus on the methodology used to extract the quantity that is observed experimentally, i.e., the electron probability density. We solve the time-dependent Schrödinger equation (TDSE) using two techniques: the spectral method, which makes use of an expansion of the wave function onto the field-free atomic eigenstates, and the wave-function method, where the TDSE is numerically solved on a grid. In this work we show that the electron probability density can be extracted from the final wave function, obtained by the wave-function method, by using the resolvent technique with a proper choice of the energy resolution. The advantage of this approach is that it does not require the calculation of the field-free atomic eigenstates. As an illustration of this method we compare the spectral and wave-function methods in the case of atomic hydrogen interacting with a linearly polarized mid-IR laser pulse. The comparison shows excellent agreement.

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I. INTRODUCTION

The interaction of a short intense mid-IR laser pulse with atoms and molecules is analyzed by means of various observables. The photoelectron spectrum from which the above-threshold ionization (ATI) peaks have been observed [1] is used as an observable. This spectrum is obtained by electron time-of-flight measurement for which a time binning is used [2]. The conversion from time to energy is made easily in the case of the field-free propagation of electrons and is more complex to achieve in the case of the velocity map imaging. The number of events per unit of energy is extracted from the number of events per unit of time by using the Jacobian transformation [3]. Consequently, the quantity that is actually measured is a density of probability, which will be called density in the following unless stated otherwise. As a consequence, when comparing experimental results and calculations, one has to be concerned about the absolute value of the density.

From the theoretical point of view, depending on the method used to solve the time-dependent Schrödinger equation (TDSE), the determination of the electron spectrum continuum density relies on the proper normalization of the final wave function. Here it is worth noting that the TDSE being solved in a box of finite-radial-length atomic spectra are fully discretized [4].

The first class of calculations that can be distinguished is called the spectral method [5], where the eigenstates are extracted from the diagonalization of the Hamiltonian without including the interaction term with the field [6]. From these eigenstates, the dipoles are calculated and the TDSE is solved by integrating the equation satisfied by the coefficients describing the expansion of the wave function in the field-free eigenstates basis. The electron density probability is then obtained by projecting the wave function at the end of the pulse onto the eigenstates describing the continuum properly normalized.

In the second class, which we call the wave-function method (also referred to as the grid method), the TDSE is solved directly on the wave function [7,8]. In the latter case, at the end of the laser pulse we could also project the resulting wave function onto the eigenstates describing the continuum, but it is preferable to avoid this procedure since the eigenstates are not calculated in the wave-function method. In order to calculate the electron spectrum probability we use the resolvent technique for which the calculation of eigenstates is avoided. The resolvent technique was introduced in strong-field physics by Schafer and Kulander [9] and allows for the calculation of the probability of having an electron of energy E after the interaction with the laser field (in the continuum or bound eigenstates meaning). However, as explained above, to compare with experiments one needs to calculate the density instead of the probability *per se*. That is why most of the comparisons between theories, based on the resolvent technique, and experiments are performed in arbitrary units [10–12].

The purpose of this work is to show how the density can be extracted from the probability calculated using the resolvent technique and we focus on the case of atomic targets described by one active electron. The paper is organized as follows. First we recall the main properties of the resolvent technique. Then we show how to transform the probability into the probability density. Finally, we illustrate this transformation for one example and compare the result to the spectral method.

II. RESOLVENT OPERATOR AND PROBABILITY DENSITY

In this section we first recall the main properties of the resolvent operator in order to extract the electron spectrum probability. Then we show how the density of probability can be extracted from the electron spectrum probability obtained by using the resolvent operator. The interaction of a laser pulse with an isolated target is formally studied by solving

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the TDSE

$$i \frac{\partial \Psi}{\partial t} = [H_0 + D(t)]\Psi. \quad (1)$$

In this expression, H_0 is the field-free Hamiltonian such that written in atomic units (a.u.) it is given by $H_0 = \frac{1}{2}p^2 + V(r)$, where p is the momentum operator and $V(r)$ is a pseudopotential describing the potential experienced by the active electron of the target; $D(t)$ is the interaction term given by $\vec{E}(t) \cdot \vec{r}$ in the length gauge and by $\vec{A}(t) \cdot \vec{p}$ in the velocity gauge, with $\vec{E}(t)$ being the electric field of the laser pulse and $\vec{A}(t)$ the corresponding vector potential. The initial state is the $|\Psi_0\rangle$ solution of the equation $H_0|\Psi_0\rangle = E_0|\Psi_0\rangle$, where E_0 is the lowest eigenenergy corresponding to the ground state. The TDSE (1) is then solved and the wave function at the end of the pulse $|\Psi_f\rangle$ is obtained. Two methods are used to solve the TDSE: the spectral method and the wave-function method. The spectral method consists in calculating the eigenstates of the field-free Hamiltonian, from which the dipole matrix is computed. The wave function is then expanded on the basis of the field-free Hamiltonian states so that the coefficients representing the projection of the wave function onto these states satisfies a linear first-order differential equation solved by using the fourth-order Runge-Kutta technique (see Sec. 5.2.3 in Ref. [5]). The wave-function technique consists in expanding the wave function on spherical harmonics. The split-operator technique is used to calculate the evolution operator and the well-known Crank-Nicholson technique is applied on each split operator so that the r -dependent term is a solution of l -coupled differential equations [13]. There are advantages of using either method; however, the main advantage of using the wave-function technique lies in the fact that the memory used by the latter technique is not as big as for the spectral method case. Once the convergence of the TDSE is achieved, a simple unitary relation between $|\Psi_f\rangle$ in the velocity and length gauge exists so that the physical content, in particular the electron spectrum, is not changed. From the calculation of $|\Psi_f\rangle$, the photoelectron spectrum is obtained by projecting the latter wave function onto the eigenfunctions solution of $H_0|\varphi_E\rangle = E|\varphi_E\rangle$. The latter equation is solved, with E being either positive or negative, such that $|\varphi_E\rangle$ is normalized to 1. Finally, we obtain the coefficients $c(E) = \langle \varphi_E | \Psi_f \rangle$ so that the modulus squared of c corresponds to the probability of having the state $|\varphi_E\rangle$ at the end of the pulse. This method is convenient when the eigenstates $|\varphi_E\rangle$ are known beforehand, e.g., for the spectral technique. In the case of the wave-function technique, the latter projection is not as suitable since the eigenstates are not calculated. In order to resolve this issue, one can use the resolvent operator. The Green's operator is noted: $G(z) = \frac{1}{z - H_0}$ where H_0 , as previously mentioned, is the Hamiltonian of the target, from which the resolvent of degree n is defined by

$$R_\epsilon^n(E) = \frac{\epsilon}{E - q_n\epsilon - H_0} \cdots \frac{\epsilon}{E - q_1\epsilon - H_0} \\ = \epsilon G(E - q_n\epsilon) \epsilon G(E - q_{n-1}\epsilon) \cdots \epsilon G(E - q_1\epsilon), \quad (2)$$

with $q_j = i^{1/n} z_j$ and z_j the j th root of unity. The latter operator can also be written $R_\epsilon^n(E) = \frac{\epsilon^n}{(E - H_0)^n - i\epsilon^n}$. Let us define the ket $|\Psi'\rangle = R_\epsilon^n(E)|\Psi_f\rangle$, which depends on ϵ , n , and E .

The term

$$\langle \Psi' | \Psi' \rangle = \langle \Psi_f | R_\epsilon^n(E)^\dagger R_\epsilon^n(E) | \Psi_f \rangle \\ = \left\langle \Psi_f \left| \frac{\epsilon^{2n}}{(E - H_0)^{2n} + \epsilon^{2n}} \right| \Psi_f \right\rangle$$

can then be calculated. The physical meaning of the latter quantity is better understood when the ket $|\Psi_f\rangle$ is expanded in the basis of eigenvectors: $|\Psi_f\rangle = \sum_{|b\rangle} c_b |b\rangle + \int_{e>0} de c(e) |e\rangle$. Here it should be noted that this procedure is performed to aid in understanding, but is not done in practice. Let us introduce the quantity $K(E, \epsilon, n) = \frac{\epsilon^{2n}}{E^{2n} + \epsilon^{2n}}$. One can show that

$$\langle \Psi' | \Psi' \rangle = \left\langle \Psi_f \left| \frac{\epsilon^{2n}}{(E - H_0)^{2n} + \epsilon^{2n}} \right| \Psi_f \right\rangle \\ = \sum_{|b\rangle} |c_b|^2 K(E - E_b, \epsilon, n) \\ + \int_{e>0} de |c(e)|^2 K(E - e, \epsilon, n) = P(E, \epsilon, n). \quad (3)$$

The function K is actually a window function similar to a door function (with a width of a few ϵ depending on n) as shown in Ref. [14]. Thus, for a fixed value of E , only the state of energy E contributes to the total sum defined by $\langle \Psi' | \Psi' \rangle$ in Eq. (3). For an energy E close to a bound-state energy E_b we find $\langle \Psi' | \Psi' \rangle = |c_b|^2$. Similarly, for E close to an energy e in the continuum, we get $\langle \Psi' | \Psi' \rangle \propto |c(e)|^2$. So the term $\langle \Psi' | \Psi' \rangle$ is assimilated to the probability that the wave function contains the energy E , which we denote $P(E, \epsilon, n)$. In practice, this probability is calculated in two steps. (i) The operator

$$R_\epsilon^n(E) = \frac{\epsilon}{E - q_n\epsilon - H_0} \cdots \frac{\epsilon}{E - q_1\epsilon - H_0} \\ = \epsilon G(E - q_n\epsilon) \epsilon G(E - q_{n-1}\epsilon) \cdots \epsilon G(E - q_1\epsilon) \quad (4)$$

is applied on $|\Psi_f\rangle$ such that $|\Psi'\rangle = R_\epsilon^n(E)|\Psi_f\rangle$. An expansion of the wave function on the basis of the spherical harmonics makes the application of the Green's function $G(E - q_j\epsilon)$ equivalent to an inverse problem [15]. (ii) Then the modulus squared of the resulting ket $|\Psi'\rangle$ is calculated.

Of course the larger n is, the more cumbersome the calculations are. However, if n is too small, then the function K is not really well represented by a door function and the energy resolution is deteriorated. A value of n satisfying good energy resolution and calculation efficiency is $n = 2$, for which $q_1 = \sqrt{i}$ and $q_2 = -\sqrt{i}$. In the region of $E > 0$, if we choose ϵ small enough such that $c(e)$ does not evolve too much in an arbitrarily large region of energy (but not larger than any physical evolution of energy such as ATI peaks, so typically a region of few ϵ), then

$$P(E, \epsilon, n) \approx \int_{e>0} de |c(e)|^2 K(E - e, \epsilon, n) \\ \approx |c(E)|^2 \int_{e>0} de K(E - e, \epsilon, n) \\ \approx |c(E)|^2 \int_{-\infty}^{\infty} de K(E - e, \epsilon, n) \\ = |c(E)|^2 \frac{\pi}{n} \csc\left(\frac{\pi}{2n}\right) \epsilon. \quad (5)$$

Formula (5) makes the link between the density of probability $|c(e)|^2$ and the probability defined by Eq. (3), which can be written

$$P(E, \epsilon, n) = \langle \Psi_f | R_\epsilon^n(E)^\dagger R_\epsilon^n(E) | \Psi_f \rangle, \quad (6)$$

where P is calculated by means of the resolvent operator given by Eq. (4). More generally, we define the density of probability by the relation

$$\rho(E, \epsilon, n) = \frac{P(E, \epsilon, n)}{\epsilon \frac{\pi}{n} \csc\left(\frac{\pi}{2n}\right)}, \quad (7)$$

which is extended to the case of bound eigenstates since, by using the resolvent technique, P becomes continuous for all energies E .

The proper choice of ϵ depends on two properties. The first one corresponds to the fact that if ϵ is too small, the electron spectrum would exhibit isolated peaks corresponding to the discretization of the continuum [14] and the approximation used in Eq. (5) is no longer valid. Moreover, a good value of ϵ should not be too large if one wants to be able to extract any physical information from the electron spectrum such as ATI peaks and resonances. A proper choice of ϵ satisfies $\rho_{st}\epsilon \approx 1$, where ρ_{st} is the density of states in the discretized continuum. The density of states is given by $\rho_{st} = \frac{1}{\pi\sqrt{2}} \frac{r_{\max}}{\sqrt{E}}$ [5], with r_{\max} being the box size on which the calculation has been performed and E the energy of the most energetic electron of the photoelectron spectrum (typically $10U_p$, with U_p being the ponderomotive energy defined by $U_p = \frac{F^2}{4\omega_0^4}$ in a.u. with F the peak amplitude of the field and ω_0 the central frequency). In that case, as will be shown later, the density no longer depends on ϵ and n in the region where $E > 0$ and can simply be denoted $\rho(E)$. For $E < 0$, the density is no longer meaningful; only its integration over a few ϵ around the energy of a bound state E_b is meaningful leading to the probability $|c_b|^2 \approx \int_{E_b-\epsilon}^{E_b+\epsilon} dE \rho(E, \epsilon, n)$.

The normalization of the wave function $|\Psi_f\rangle$ is given by $\sum_{|b\rangle} |c_b|^2 + \int_{e>0} de |c(e)|^2 = 1$ and the integral is then defined by

$$\begin{aligned} & \int_{-\infty}^{\infty} dE \rho(E) \\ &= \sum_{|b\rangle} |c_b|^2 \frac{\int dE K(E - E_b, \epsilon, n)}{\epsilon \frac{\pi}{n} \csc\left(\frac{\pi}{2n}\right)} \\ &+ \int_{e>0} de |c(e)|^2 \frac{\int dE K(E - e, \epsilon, n)}{\epsilon \frac{\pi}{n} \csc\left(\frac{\pi}{2n}\right)} \\ &= \sum_{|b\rangle} |c_b|^2 + \int_{e>0} de |c(e)|^2 = 1. \end{aligned} \quad (8)$$

The latter quantity represents the probability of having an electron in the whole range of energy that is 1 regardless of the value of n and ϵ , as expected. The previous expansion of $|\Psi_f\rangle$ was performed in the case of a true continuum. Recalling that we deal with a finite box to solve the TDSE, it is easy to show that the last result is still valid since the integral over the continuous positive energy is replaced by a discrete continuum so that the same method can be used.

So far, the calculations have been performed for integrated spectra over angles. The previous analysis can be extended to the case of differential spectra over angles. In that case the resolvent technique allows for the determination of the probability of having one electron of energy E emitted in a given direction. The wave function $|\Psi_f\rangle$ is expanded on the spherical harmonic functions basis so that $\Psi_f(\vec{r}) = \sum_{l,m} f_{lm}(r) Y_l^m(\theta, \phi)$. When the scalar product $\langle \Psi_f | \Psi_f \rangle$ is performed, the integration over angles and r is achieved. If one wants to calculate the differential probability spectrum, then only the integration over r is performed. The term that is calculated is then

$$\begin{aligned} P(E, \epsilon, n, \Omega) &= \langle \Psi_f | R_\epsilon^n(E)^\dagger R_\epsilon^n(E) | \Psi_f \rangle_r \\ &= \int dr r^2 \sum_{l,m,l',m'} f_{l'm'}(r)^* Y_{l'}^{m'}(\theta, \phi)^* \\ &\quad \times R_\epsilon^n(E)^\dagger R_\epsilon^n(E) f_{lm}(r) Y_l^m(\theta, \phi). \end{aligned} \quad (9)$$

The normalization procedure is strictly the same since the link between $P(E, \epsilon, n, \Omega)$ and $P(E, \epsilon, n)$ is simply $\int d\Omega P(E, \epsilon, n, \Omega) = P(E, \epsilon, n)$, leading to $\int d\Omega \rho(E, \epsilon, n, \Omega) = \rho(E, \epsilon, n)$. Consequently, the differential probability density of having an electron of energy $E > 0$ ejected in the direction Ω is given by

$$\rho(E, \epsilon, n, \Omega) = \frac{P(E, \epsilon, n, \Omega)}{\epsilon \frac{\pi}{n} \csc\left(\frac{\pi}{2n}\right)} = \frac{dP}{dE d\Omega}. \quad (10)$$

Of course, by integrating over Ω and E , the result is 1, as expected. Formula (10), leading also to Eq. (7), is the main result of this work and allows us to link the probability $P(E, \epsilon, n, \Omega)$ to the density $\rho(E, \epsilon, n, \Omega)$ simply by a coefficient of proportionality that depends on ϵ .

III. APPLICATIONS

In this section we use and illustrate the previous definition of the density for two cases. The first one corresponds to the calculation of the probability of ionization, which is a key quantity in strong field physics. Then we will compare the differential density of probability in energy and angle calculated by two different methods: the spectral method and the wave-function method for which the electron spectrum is extracted by means of the method previously explained. All the calculations are performed for a linearly polarized laser.

A. Ionization probability

For the sake of illustration, the laser field is set to a central frequency of $\omega_0 = 0.057$ a.u. ($\lambda = 800$ nm) and a field strength of $F = 0.053$ a.u. ($I = 10^{14}$ W/cm²) for six optical cycles (flat-top envelope) including a turn-on and turn-off duration of half a cycle for each. The target is chosen to be the atomic hydrogen. These conditions are close to those encountered experimentally [12]. We choose a value of ϵ equal to 4×10^{-3} a.u., which is of the order of the state density in the continuum, and $n = 2$, which optimizes the resolution and the time calculation. The plot of the probability spectra integrated over angles, obtained by using Eqs. (6) and (4), is given in Fig. 1.

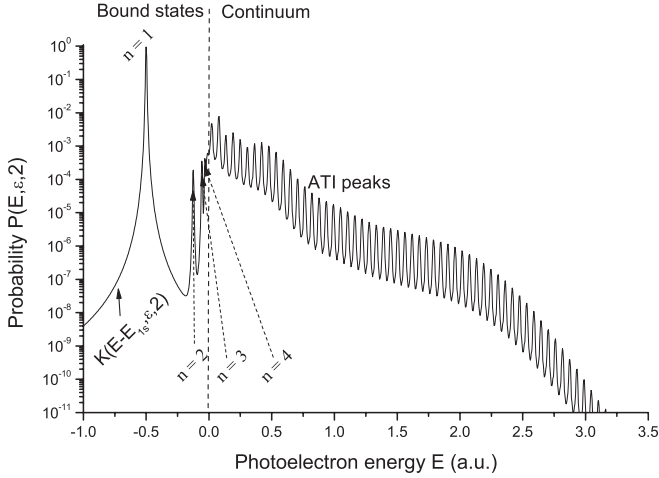


FIG. 1. Plot of the probability spectrum integrated over angles obtained for hydrogen by using Eqs. (6) and (4). The laser pulse has a central wavelength of 800 nm ($\omega_0 = 0.057$ a.u.), a peak intensity of 10^{14} W/cm 2 ($F = 0.053$ a.u.), a pulse duration of six cycles, and a flat-top envelope. The order of the window function is $n = 2$ and the energy resolution $\epsilon = 4 \times 10^{-3}$ a.u. In this example the inverse of the density of states is roughly 7×10^{-4} a.u. These calculations have been performed in the velocity gauge, a discretization of space $dx = 0.1$ a.u., and a grid size of 1500 a.u. The highest angular momentum used is $l = 80$ and the time step is 2.2×10^{-3} a.u.

To convert these spectra into a probability density we use Eq. (7) for which the conversion coefficient from probability to density is the inverse of $\frac{\pi}{\sqrt{2}}\epsilon$. The probability of ionization is given by the formula $P_{\text{ion}} = \int_0^\infty dE \rho(E)$, with $\rho(E)$ the density. From our calculations, we get $\int_0^\infty dE P(E) = 4.195788 \times 10^{-4}$ a.u. To convert this quantity into the probability of ionization we need to divide by $\frac{\pi}{\sqrt{2}}\epsilon$, leading to $P_{\text{ion}} = 0.04722$. One can compare this result to the remaining population in the ground state. A simple projection of the wave function at the end of the pulse onto the ground state gives $P_{n=1} = 0.951013$. This quantity is in excellent agreement with the value of $P(E)$ extracted from Fig. 1. Indeed, according to Eq. (3), the value of $|c_b|^2$ is retrieved from $P(E = E_b)$, where E_b is the energy of the bound state $|b\rangle$.

The ionization probability can be approximated by $P_{\text{ion}} = 1 - P_{n=1} = 0.04898$, which is close to the value found from the integration of the spectrum. The latter value is a bit larger than the one from the integration of the spectra in the continuum part since the populations of the excited states have not been taken into account. From Fig. 1 we get $P_{n=2} = 2 \times 10^{-4}$, $P_{n=3} = 3 \times 10^{-4}$, $P_{n=4} = 4 \times 10^{-4}$, and $P_{n=5} = 6 \times 10^{-4}$; the other contributions are too small to be extracted from this figure. Including the population of the excited states, we get $P_{\text{ion}} = 1 - P_{n=1} - P_{n=2} - \dots = 0.04748$, which is close to the one calculated from the integration of the spectra within 0.5%.

B. Comparison of spectra

Here we compare two methods to calculate the electron spectrum and the TDSE. The first method, as previously explained, consists in solving the TDSE by the spectral method.

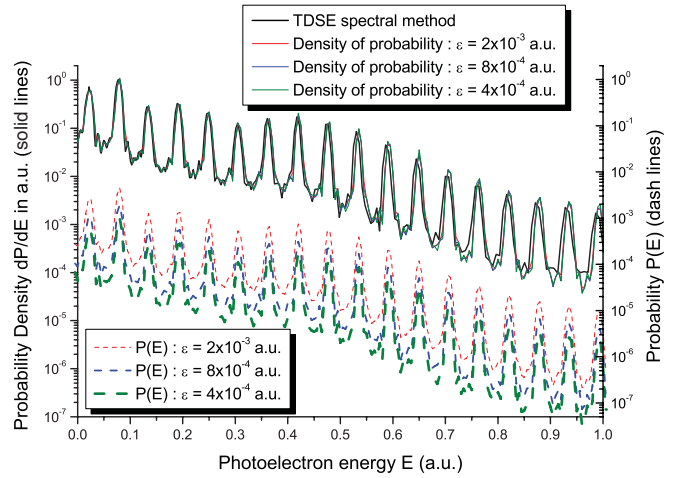


FIG. 2. (Color online) Plot of the probability density and the probability for hydrogen as a function of the energy of the ejected electron integrated over angles. The laser parameters are the same as the one defined in Fig. 1. The solid curves correspond to the density of probability $\rho(E) = \frac{dP}{dE}$, while the dashed curves are the probability distributions $P(E, \epsilon, n)$. The red curves correspond to a value of $\epsilon = 2 \times 10^{-3}$ a.u., the blue curves correspond to $\epsilon = 8 \times 10^{-4}$ a.u., and the green curves correspond to $\epsilon = 4 \times 10^{-4}$ a.u. All the calculations are performed for $n = 2$. The solid black curve is the result of the calculation performed by the spectral method. The numerical parameters are the same as in Fig. 1.

The use of this method implies an explicit calculation of the eigenstates. Since a box in radial coordinates r is considered to calculate these states, the continuum becomes a discretized continuum. Here special care is needed to normalize the states in the continuum. As previously noted, all the states are normalized to 1 and the coefficients $|c(E)|^2 = |\langle \varphi_E | \Psi_f \rangle|^2$, extracted after solving the TDSE, correspond to the probability of having the state $|\varphi_E\rangle$ at the end of the pulse. In order to calculate the density of probability $\frac{dP}{dE}$, the probability $|c(E)|^2$ is multiplied by the state density of the discretized continuum, which is a straightforward procedure (see Sec. 3.3 in Ref. [5]).

By using the wave-function method, the electron spectrum is calculated by the resolvent method, as presented in the preceding section. The main difference lies in the fact that the whole photoelectron spectrum is continuous by using the resolvent technique, even in the bound-state region, while for the spectral method, the latter region is discretized. In Fig. 2, three electron probabilities are plotted (dashed curves) for three different values of the parameter ϵ , which somehow mimics an experimental energy resolution. The range of energy differs by a factor of 5 between the smallest and the largest value. The ATI peaks are shown clearly and are spaced by the photon energy as expected. Furthermore, the width of each ATI peak is equal to 0.0142 a.u. and does not depend on the choice of ϵ , as pointed out by Eq. (5). The width is due to the finite duration of the pulse. A simple Fourier analysis allows one to link the width in energy ΔE of the ATI peaks and the pulse duration Δt by $\Delta E \times \Delta t = 8$ for a flat-top envelope. If we set Δt equal to five cycles at 800 nm, we get $\Delta E = 0.0145$ a.u., in excellent agreement with the data extracted from Fig. 2. If we now calculate the density from the probability spectra by using Eq. (7), the solid curves of

Fig. 2 are obtained. The three distributions are in accordance with each other, showing the independence of ρ as a function of ϵ , as previously pointed out. The results obtained by the spectral method are given by the solid black curve, in excellent agreement with the ones issued from the resolvent technique. The differential probability density is then obtained. All the probability spectra obtained from the resolvent technique are proportional to each other in the continuum region for a proper value of ϵ . This last criterion is checked afterward to validate a proper choice of ϵ .

IV. CONCLUSION

A method allowing for the calculation of the probability density from the probability by means of the resolvent technique has been presented. We show that those two quantities are simply proportional to each other as long as the resolution energy is properly chosen. The resolution has to be small enough to be able to exhibit the energy variation due to the physics and large enough to avoid a resolution of the discrete states of the continuum: These

two conditions are fulfilled for an energy resolution of the order of the inverse of the density of states. The calculation of the probability density allows for a direct comparison to perturbative calculations that are intrinsically normalized and experiments. In the example investigated in this work, corresponding to the ionization of hydrogen in the infrared regime, we have shown that the ionization probability and the population of the excited states can easily be extracted from the resolvent technique. We have also shown that the electron density spectra extracted from the TDSE solved by using the wave-function method or the resolvent technique agree perfectly with the result coming from the spectral method. A hydrogen target has been chosen for the sake of simplicity, but the method can be applied to any target described by one active electron, any laser polarization, or any gauge. The use of the resolvent technique was particularly convenient since the eigenstates (bound and continuum) are not calculated and the result proved to be useful for a complex target where these states cannot be expressed analytically [16]. Further work should consist in extending this method to the multielectron case.

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