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Monte Carlo calculation of the above-threshold ionization of atomic hydrogen

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Abstract. We present a systematic study on the above-threshold ionization of the hydrogen atom using an adaptive Monte Carlo algorithm. The ionizations from both the ground state and the excited states are discussed in the weak-field regime. It is found that the Monte Carlo approach is quite efficient and may be useful to treat the high-order multiphoton ionization of other atoms.

1. Introduction

During the past few years, many calculations have been performed to study the above-threshold ionization (ATI) of atoms, particularly for the hydrogen atom where the analytic Coulomb wavefunctions can be obtained and the atomic Green function can be constructed in terms of the regular and irregular Coulomb wavefunctions (Karule 1978, Fainshtein *et al* 1984, Chu and Cooper 1985, Shakeshaft 1986a). The problem of calculating the multiphoton ionization cross section (in lowest-order perturbation) reduces to that of evaluating a multidimensional integral. Recently, Edwards *et al* (1986) have taken advantage of the fact that the atomic Green function can be split into a product of functions of only a single variable. Using this property the N-dimensional integral is expressible as a sum of $N!$ integrals, each of which is a product of N one-dimensional integrals. The shortcoming of this approach is that $N!$ grows very rapidly with N, and the computational time grows exponentially. The question naturally arises as to whether the Monte Carlo scheme is an appropriate method to be used to evaluate this multidimensional integral. In order to investigate this point, a preliminary calculation (Han and Shakeshaft 1989) has been performed for multiphoton ionization matrix elements of hydrogen as a testing example for the case of absorbing only one photon above threshold. It was found that the computing time for integral evaluations is reduced a considerable amount, with the total computational time increasing with N only as (roughly) N^2. In this paper, we present a systematic study on the above-threshold ionization of hydrogen using an adaptive Monte Carlo algorithm. The ionizations from both the ground state and several excited states will be discussed.

2. Method

Consider an atom, initially in state $|i\rangle$, irradiated by a monochromatic radiation field of frequency ω and intensity I. We adopt the length gauge for the electromagnetic

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field. Using the dipole approximation for the interaction between atom and field, the generalized cross section for the atom to absorb N photons, in lowest-order perturbation theory, is

$$\sigma_N = \int d\Omega_k \, 2\pi[2\pi\alpha\omega]^N \left| \sum_{LM_z} i^L Y_{LM_z}(\hat{\boldsymbol{K}}) \sum_{l_{N-1}m_{N-1}} \cdots \sum_{l_1 m_1} A(l_{N-1}m_{N-1}; \ldots l_1 m_1) M^{(N)} \right|^2 \tag{1}$$

where $\alpha = e^2/\hbar c \simeq \frac{1}{137}$ is the fine structure constant, $\boldsymbol{K}$ is the wavevector of the outgoing photoelectron, and the integration is over all angles of propagation of the photoelectron. L and M_z are the angular momentum quantum numbers of the final state, l_j and m_j are the allowed intermediate angular momentum quantum numbers, and $A(l_{N-1}m_{N-1}\ldots)$ is a geometrical factor given as

$$A(l_{N-1}m_{N-1}\ldots) = \prod_{n=1}^{N} \int d\hat{\boldsymbol{r}} \, Y^*_{l_n m_n}(\hat{\boldsymbol{r}})(\hat{\boldsymbol{\varepsilon}}\cdot\hat{\boldsymbol{r}}) \, Y_{l_{n-1}m_{n-1}}(\hat{\boldsymbol{r}}) \tag{2}$$

where $\hat{\boldsymbol{\varepsilon}}$ is the polarization vector of the radiation field, $\boldsymbol{r}$ the position operator of the electron and $l_N = L$, $m_N = M_z$. The transition matrix element $M^{(N)}$ is given by

$$M^{(N)} = \int_0^\infty dr_N \ldots \int_0^\infty dr_1 \, u_{\nu_N}(r_N) r_N g^+_{\nu_{N-1}}(r_N, r_{N-1}) \ldots r_1 u_i(r_1) \tag{3}$$

where $\nu_n = (E_n, l_n)$, $E_n = E_i + n\hbar\omega$; E_i is the energy of the electron in the initial state. $u_i(r)$ and $u_{\nu_N}(r)$ are, respectively, the initial and final radial wavefunctions normalized on the energy scale. The quantity $g^+_\nu(r, r')$ is the radial Green function, for the hydrogen atom, it can be expressed analytically in terms of hypergeometric functions as follows:

$$g^+_{\nu_n}(r, r') = -\frac{i}{k_n} \frac{\Gamma(l_n + 1 - i\gamma_n)}{(2l_n+1)!} W_{i\gamma_n, l_n+1/2}(-2ik_n r_>) M_{i\gamma_n, l_n+1/2}(-2ik_n r_<) \tag{4}$$

where W and M are the Whittaker functions, $r_>$ ($r_<$) denotes the larger (smaller) of r and r', and $\gamma_n = Z e^2 m/\hbar^2 k_n$ with k_n given by

$$k_n = (2m/\hbar^2)^{1/2}(E_i + n\hbar\omega)^{1/2}. \tag{5}$$

For the above-threshold ionization, where the atom absorbs more photons than the minimum number N_0 of photons required to ionize, it can be shown that $M^{(N)}$ satisfies the following relation so that the divergent part of the integral involving the continuum wavefunction can be circumvented (Shakeshaft 1986b),

$$M^{(N)} = 2\,\mathrm{Re}\{M^{+(N)}\} - 2\pi i \sum_{j=1}^{N-N_0} J_j^{(N)} M^{(N-j)} \tag{6a}$$

$$J_j^{(N)} = [\langle u^+_{\nu_N} | r g^+_{\nu_{N-1}} r \ldots g^+_{\nu_{N-j+1}} r | u_{\nu_{N-j}} \rangle]^* \tag{6b}$$

with

$$J_1^{(N)} = [\langle u^+_{\nu_N} | r | u_{\nu_{N-1}} \rangle]^* \tag{6c}$$

where u^+_ν is the outgoing wave component of the continuum wavefunction and $M^{+(N)}$ is obtained from $M^{(N)}$ by replacing $u_{\nu_N}(r)$ by $u^+_{\nu_N}(r)$. Therefore, the problem reduces to that of evaluating the multidimensional integrals of $M^{+(N)}$ and $J_j^{(N)}$.

3. Results and discussion

To evaluate the integrals of $M^{+(N)}$ and $J_j^{(N)}$, an adaptive Monte Carlo method (Lepage 1978) is used. We first rotate the integration contour into the upper right quadrants of the complex plane, that is, we replace r by $s\,e^{i\theta}$ where $0 \leqslant s \leqslant \infty$ and $0 < \theta < \pi/2$. This ensures the integrand being damped exponentially along this path. We chose $\theta = \pi/4$ in our calculation. We then convert the range of integration from $[0, \infty]$ to $[0, 1]$ via the transformation $r_n = a_n y_n/(1-y_n)$. The scaling factor a_n is chosen as $|a_n| = 1/(|k_{n-1}| + |k_n|)$, which will roughly cover the most significant range of integration over r_n. As shown in (4) the Green function is factorized as a product of Coulomb functions, thus we set up two tables for the Whittaker functions W and M, respectively, for the smaller region of r, with $|2k_n r| < 2(l_n + 1 + |\gamma_n|)$. Then the Green function $g_l^+(r, r')$ can be evaluated by interpolation between the tabulated points. For the larger region of r, we can calculate the Coulomb functions very rapidly using the asymptotic expansions.

In the adaptive Monte Carlo algorithm for calculating the multi-dimensional integrals, an iterative scheme is used. The distribution of the integration points is varied that will give a non-uniform probability density function, which weights the region where the integrand is largest in magnitude. The grid points are selected at random in the first calculation, then the iterative procedure is repeated until the optimal grid is achieved. Table 1 gives the results of the multiphoton ionization cross section of hydrogen in the ground state by circularly polarized light. For each wavelength of the radiation field, we have calculated several ATI cross sections, up to seven-photon ionization for $\lambda = 5063$ Å. In table 1, the previous results are also listed for comparison.

Table 1. Generalized cross sections $\hat{\sigma}_N$ for N-photon ionization of ground-state hydrogen by circularly polarized light of wavelength λ. N_0 represents the minimum of photons required for ionization. n_i is the number of integration points used in the Monte Carlo calculation. The numbers in brackets represent powers of 10.

				$\hat{\sigma}_N$ ($\mathrm{cm}^{2N}\,\mathrm{s}^{N-1}$)			
					Previous work		
λ (Å)	N_0	N	n_i	Present work	a	b	c
700	1	1	6 000	3.1 (−18)	3.1 (−18)	3.1 (−18)	2.4 (−18)
		2	15 000	2.2 (−52)	2.2 (−52)	1.1 (−52)	1.3 (−52)
		3	25 000	9.1 (−87)	8.7 (−87)	3.9 (−87)	4.1 (−87)
1400	2	2	15 000	1.8 (−50)	1.8 (−50)	1.8 (−50)	5.3 (−52)
		3	25 000	4.7 (−84)	4.8 (−84)	2.8 (−84)	1.7 (−85)
		4	50 000	7.3 (−118)	7.4 (−118)	3.9 (−118)	2.4 (−119)
2650	3	3	25 000	2.1 (−83)	2.1 (−83)	2.1 (−83)	1.3 (−87)
		4	50 000	4.4 (−116)	4.5 (−116)	2.1 (−116)	2.6 (−118)
		5	80 000	3.2 (−149)	3.2 (−149)	1.0 (−149)	2.5 (−151)
3200	4	4	50 000	3.6 (−116)	4.0 (−116)	4.0 (−116)	4.2 (−119)
		5	80 000	8.0 (−149)	8.8 (−149)	4.9 (−149)	3.4 (−151)
4339	5	5	90 000	2.2 (−149)	2.5 (−149)		
		6	120 000	2.3 (−181)	2.3 (−181)		
5063	6	6	120 000	4.2 (−182)	5.0 (−182)		
		7	160 000	5.0 (−214)	5.7 (−214)		

The results in columns a, b and c are obtained from (exact) lowest-order perturbation theory by representing the Coulomb Green function on a Sturmian basis (Grontier *et al* 1986, Shakeshaft 1986b), from a non-perturbative approximation method (Shakeshaft and Potvliege 1987) and from a Reiss approximation (Reiss 1980), respectively. It is clear to see that the present results are in satisfactory agreement with the exact ones and those obtained by the non-perturbative theory. However, the Reiss approximation yields results which are generally much smaller than the others. It is also interesting to note that, as shown in table 1, the integration points n_i used in our calculations for the multidimensional integrals are very small, even for $N = 6$ (7), we used only 1.2×10^5 (1.6×10^5) points corresponding to only about seven points per dimension. Another advantage of the adaptive Monte Carlo scheme is that the convergence for the iterative procedure is quite good. The optimal grid can generally be reached within ten iterations. Table 2 illustrates the variation in the values of the integrals $M^{+(N)}$ from one iteration to the next, which shows the general behaviour of convergence is fast and stable. The last row shows the cumulative standard deviation (defined by equation (5) of Lepage (1978)), which is, in general, less than 8% of the calculated results.

Table 2. Values of $M^{+(N)}$ at the ith iteration of the Monte Carlo calculation. n_i is the number of integration points used. The numbers in brackets represent powers of 10.

	$\lambda = 3200$ Å		$\lambda = 4339$ Å	
i	$N = 4$ ($n_i = 50\,000$)	$N = 5$ ($n_i = 80\,000$)	$N = 5$ ($n_i = 90\,000$)	$N = 6$ ($n_i = 120\,000$)
1	1.92 (5)	0.22 (8)	0.16 (8)	0.88 (9)
2	0.75 (5)	0.99 (6)	0.14 (8)	0.89 (9)
3	0.69 (5)	0.12 (7)	0.53 (7)	0.71 (9)
4	0.64 (5)	0.14 (7)	0.53 (7)	0.72 (8)
5	0.62 (5)	0.13 (7)	0.53 (7)	0.13 (9)
6	0.65 (5)	0.14 (7)	0.52 (7)	0.11 (9)
7	0.66 (5)	0.13 (7)	0.53 (7)	0.12 (9)
8	0.65 (5)	0.13 (7)	0.52 (7)	0.12 (9)
9	0.65 (5)	0.13 (7)	0.51 (7)	0.13 (9)
10	0.65 (5)	0.13 (7)	0.51 (7)	0.13 (9)
Cumulative standard deviations	0.20 (4)	0.47 (5)	0.39 (6)	0.97 (7)

We have also calculated the ATI cross sections of the hydrogen atom from the ns excited states with $2 \leqslant n \leqslant 6$. Two-photon ionization cross sections of hydrogen in ns states have been studied by Aymar and Crance (1980) using a method based on numerical computations in the framework of the central field approximation, and by Karule (1985) employing above threshold analytic continuation of the Sturmian expansion for the transition matrix elements. Table 3 shows our results and the previous ones. The agreement between the values computed by us and others is quite good. In those previous studies, only two-photon ionization of the hydrogen in ns states with one photon above threshold is calculated. We have extended the calculation for the case of three-photon ionization with two photons above threshold. The results are also listed in table 3.

Table 3. Generalized cross section $\hat{\sigma}_N$ ($cm^{2N}\ s^{N-1}$) for N-photon ionization of the hydrogen atom in ns states by circularly polarized light.

λ/n^2 (Å)	N	$n=2$		$n=3$		$n=4$		$n=5$		$n=6$	
		a	b	a	b	a	b	a	b	a	b
100	2	1.06 (−54)	1.04 (−54)	2.05 (−53)	2.03 (−53)	1.63 (−52)	1.61 (−52)	7.88 (−52)	7.75 (−52)	2.75 (−51)	2.71 (−51)
	3	1.30 (−89)		1.52 (−87)		4.58 (−86)		6.45 (−85)		5.56 (−84)	
300	2	3.28 (−52)	3.23 (−52)	5.58 (−51)	5.50 (−51)	3.83 (−50)	3.77 (−50)	1.62 (−49)	1.59 (−49)	5.07 (−49)	5.00 (−49)
	3	4.79 (−86)		5.68 (−84)		1.64 (−82)		2.15 (−81)		1.71 (−80)	
500	2	4.54 (−51)	4.48 (−51)	6.82 (−50)	6.71 (−50)	4.26 (−49)	4.20 (−49)	1.68 (−48)	1.65 (−48)	8.41 (−48)	4.94 (−48)
	3	2.28 (−84)		2.56 (−82)		6.91 (−81)		8.47 (−80)		1.32 (−78)	

[a] This work.

[b] Aymar and Crance (1980).

The method used in this work can be generalized for the case of non-hydrogen atoms, such as alkali-metal atoms where the Green function can still be constructed in a factorized form by using quantum defect theory. Therefore, our method is applicable for this case. A detailed calculation will be reported later.

In conclusion, we have applied an adaptive Monte Carlo algorithm to study the above-threshold ionization of the hydrogen atom from both the ground state and excited states. It is found that the Monte Carlo approach is quite efficient and may be useful in treating the high-order multiphoton ionization of non-hydrogen atoms.

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