

Monte Carlo integration of multiphoton ionization matrix elements in the weak-field regime

C. S. Han

Department of Electrophysics, Chiao-Tung University, Hsin-Chu, Taiwan, Republic of China

Robin Shakeshaft

Department of Physics, University of Southern California, Los Angeles, California 90089-0484

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An adaptive Monte Carlo algorithm is applied, as a test case, to the evaluation of multiphoton ionization matrix elements for hydrogen. This study indicates that a Monte Carlo approach may be useful in treating high-order multiphoton ionization of alkali-metal atoms, and possibly other atoms, by weak fields.

I. INTRODUCTION

Within lowest-order perturbation theory there are numerous methods for calculating cross sections for multiphoton ionization of atoms. Many of these methods rely on the analytic form of the electron wave function, and hence apply only to one-electron atoms. More general methods, which apply to multielectron atoms, are often limited by the order N of the multiphoton process, with the computation time increasing rapidly as N increases. Thus, so far, applications to alkali-metal atoms,¹⁻⁵ to carbon,⁶ to the rare-earth metals,^{7,8} and to the rare gases,⁹⁻¹¹ have been restricted to $N \leq 6$.

For the alkali-metal atoms, an approximate atomic Green's function can be constructed in terms of the regular and irregular Coulomb wave functions, either by using experimentally derived information on the quantum defects^{4,5,12} or by approximating $rW(r)$, where $W(r)$ is the Herman-Skillman potential, by a series of straight lines.³ For other multielectron atoms it might become possible to construct an atomic Green's function, perhaps

by using multichannel quantum defect theory or by other means. To the extent that an atomic Green's function can be constructed, the problem of calculating multiphoton ionization cross sections (in lowest-order perturbation theory) reduces to that of evaluating a multidimensional integral. The dimensionality of the integral increases linearly with N , and the question naturally arises as to whether Monte Carlo integration is appropriate. As a test case, we have applied an adaptive Monte Carlo algorithm¹³ to the integration of multiphoton ionization matrix elements for hydrogen (exact results are available for comparison). We report some results below, and observe that if only, say, 10% accuracy is required, this algorithm is quite efficient.

II. METHOD

Assuming a single-particle radial Green's function $g_{\alpha}^{+}(r_1, r_2)$, where the subscript α includes the energy, orbital angular momentum, and perhaps other quantum numbers, the problem at hand is to evaluate an integral of the form¹⁴

$$M^{(N)} = \int_0^{\infty} dr_N \int_0^{\infty} dr_{N-1} \cdots \int_0^{\infty} dr_1 u_{\alpha_N}(r_N) r_N g_{\alpha_{N-1}}^{+}(r_N, r_{N-1}) r_{N-1} \cdots g_{\alpha_1}^{+}(r_2, r_1) r_1 u_{\alpha_0}(r_1), \quad (2.1)$$

where $u_{\alpha_0}(r)$ and $u_{\alpha_N}(r)$ are, respectively, the initial and final radial wave functions. If E_0 is the initial energy of the electron, and if ω is the frequency of the field, the energy associated with the n th step of the ionization process, that is, the energy appearing in $g_{\alpha_n}^{+}(r_{n+1}, r_n)$, is

$$E_n = E_0 + n\hbar\omega.$$

For $r \rightarrow \infty$ we have, ignoring a factor independent of r ,

$$g_{\alpha_n}^{+}(r, r') \sim e^{ik_n r + i\gamma_n \ln(2k_n r)}, \quad (2.2)$$

where, for $n \geq 0$,

$$k_n = (2\mu/\hbar^2)^{1/2} (E_0 + n\hbar\omega + i\eta)^{1/2}, \quad (2.3)$$

where $\gamma_n = Ze^2\mu/(\hbar^2 k_n)$, and where μ is the electron mass, η is positive but infinitesimal, and Z is the effective atomic number. If N_0 is the smallest integer m for which

$E_0 + m\hbar\omega \geq 0$, we see that as r increases $g_{\alpha_n}^{+}(r, r')$ decreases exponentially if $n < N_0$, or behaves as an oscillating outgoing wave if $n \geq N_0$. We can rotate the paths of integration into the upper right quadrants of the r_n planes to make $g_{\alpha_n}^{+}(r_{n+1}, r_n)$ vanish exponentially for $|r_{n+1}| \sim \infty$ even when $n \geq N_0$. However, if we do this we cause $u_{\alpha_N}(r_N)$, which is a standing wave, to explode exponentially for $|r_N| \sim \infty$. To circumvent this, we write

$$u_{\alpha_N}(r) = u_{\alpha_N}^{+}(r) + u_{\alpha_N}^{-}(r),$$

where $u_{\alpha_N}^{+}(r)$ and $u_{\alpha_N}^{-}(r)$ are, respectively, the outgoing and ingoing wave components, and we make use of the relation^{5,15}

$$M^{(N)} = 2 \operatorname{Re}(M^{+(N)}) - 2\pi i \sum_{j=1}^{N-N_0} J_j^{(N)} M^{(N-j)}, \quad (2.4a)$$

where $M^{+(N)}$ is obtained from $M^{(N)}$ by replacing $u_{\alpha_N}(r)$ by $u_{\alpha_N}^+(r)$ and where

$$J_j^{(N)} = \left[\int_0^\infty dr_N \cdots \int dr_{N-j+1} u_{\alpha_N}^+(r_N) r_N g_{\alpha_{N-1}}^+(r_N, r_{N-1}) \cdots r_{N-j+1} u_{\alpha_{N-j}}(r_{N-j+1}) \right]^*, \quad (2.4b)$$

where $u_{\alpha_n}(r)$ is the continuum radial wave function of the electron corresponding to the energy E_n , $n \geq N_0$. Note that for $j=1$ Eq. (2.4b) becomes

$$J_1^{(N)} = \left[\int_0^\infty dr u_{\alpha_N}^+(r) r u_{\alpha_{N-1}}(r) \right]^*. \quad (2.4c)$$

Consequently, we have only to evaluate the integrals $M^{+(n)}$ and $J_j^{(n)}$, with $N_0 \leq n \leq N$. For these integrals we can convert the range of integration from $[0, \infty]$ to $[0, 1]$ via the transformation

$$r_n = a_n y_n / (1 - y_n), \quad (2.5)$$

where y_n is now the (real) integration variable. Choosing a_n to be complex, that is,

$$a_n = |a_n| \exp(i\theta_n),$$

with $0 < \theta_n < \pi/2$, amounts to rotating the paths of integration into the upper right quadrants of the r_n planes; this ensures that the integrand vanishes exponentially as any of the y_n approach 1. We choose $\theta_n = \pi/4$, for all n . The integration points were initially selected at random throughout the intervals $0 \leq y_n \leq 1$. However, the distribution of points in r space is affected by our choice of the magnitudes of the a_n , and this choice must be made judiciously in order to make the integration efficient. We chose

$$|a_n| = 1 / (|k_{n-1}| + |k_n|), \quad (2.6)$$

recognizing that, after complex rotation of the r_n , the right-hand side of (2.6) is roughly the significant range of integration over r_n .

III. DISCUSSION

We used the adaptive Monte Carlo algorithm described by Lepage.¹³ This is an iterative procedure; the integration points (in y space) are selected at random in the first iteration, but in subsequent iterations these points are selected with a nonuniform probability distribution which weights the regions where the integrand is largest. In Tables I and II we show some results for hydrogen, initially in its ground state. [We normalized $u_{\alpha_0}(r)$ to unity, and we normalized $u_{\alpha_N}(r)$ on the energy scale.] Table I illustrates the variation in the values of the integrals $M^{+(N)}$ from one iteration to the next. (The number of integration points was held fixed from one iteration to the next.) The values begin to settle down, though quite slowly, as the number of iterations increases; we did not attempt to extrapolate beyond the last iteration, but of course, in general, extrapolation would be useful. We also show in Table I the cumulative standard deviation at each iteration, where this quantity is defined by Eq. (5) of Lepage.¹³ The values for the cumulative standard deviation may seem large, but relative to the magnitudes of $M^{(N)}$ they are not so large. In Table II we give estimates of the complete matrix elements at different orders and wavelengths.¹⁶ Generally speaking, it is not difficult to achieve an accuracy of 10%. Note that even for $N=7$ we used only 1.6×10^5 integration points. Were we to evaluate this seven-dimensional integral by using a product of one-dimensional quadrature rules, with, say, 20 points per dimension (a modest number, considering the integrand is oscillatory) we would use a total of 1.3×10^9 points. Of course, one could take advantage of the fact that $g_\alpha(r, r')$ can be expressed as a product (or sum of

TABLE I. Values of $M^{+(N)}$ and of the cumulative standard deviation (SD) at the i th iteration of the Monte Carlo calculation. The wavelength is $\lambda = 4339.3 \text{ \AA}$ and the polarization is circular. The numbers in square brackets represent powers of 10. We used 90 000 integration points for $N=5$, and 120 000 points for $N=6$.

i	$\text{Re}(M^{+(N)})$	$N=5$		$\text{Re}(M^{+(N)})$	$N=6$	
		$\text{Im}(M^{+(N)})$	SD		$\text{Im}(M^{+(N)})$	SD
1	1.53[7]	-6.07[6]	1.9[7]	-6.35[8]	6.06[8]	8.5[8]
2	1.41[7]	-3.19[6]	5.1[6]	-7.99[8]	3.87[8]	6.2[8]
3	3.73[6]	-3.48[6]	2.9[6]	-7.00[8]	1.14[8]	5.2[8]
4	2.37[6]	-4.71[6]	1.4[6]	2.28[7]	6.87[7]	9.8[7]
5	2.91[6]	-4.40[6]	1.2[6]	7.69[7]	9.97[7]	3.7[7]
6	2.06[6]	-5.48[6]	7.9[5]	8.43[7]	7.26[7]	3.1[7]
7	1.97[6]	-4.92[6]	6.4[5]	9.36[7]	7.61[7]	2.3[7]
8	1.77[6]	-4.89[6]	5.5[5]	9.75[7]	7.51[7]	1.7[7]
9	1.77[6]	-4.77[6]	4.8[5]	1.08[8]	7.29[7]	1.4[7]
10	1.92[6]	-4.66[6]	4.4[5]	1.05[8]	7.70[7]	1.2[7]
11	2.09[6]	-4.72[6]	4.2[5]	1.08[8]	7.72[7]	1.0[7]
12	2.21[6]	-4.68[6]	3.9[5]	1.05[8]	7.64[7]	9.7[6]

TABLE II. Values of the complete matrix elements $M^{(N)}$ at the n_i th iteration of the Monte Carlo calculation, using n_p points, compared to results that are exact (to the figures given) for different wavelengths λ of circularly polarized light. The numbers in square brackets represent powers of 10.

λ (Å)	N	n_p	n_i	Monte Carlo		Exact	
				$\text{Re}M^{(N)}$	$\text{Im}M^{(N)}$	$\text{Re}M^{(N)}$	$\text{Im}M^{(N)}$
3200	4	50 000	12	1.24[5]	0	1.26[5]	0
	5	80 000	12	3.99[6]	1.27[6]	3.99[6]	1.22[6]
4339	5	90 000	12	4.41[6]	0	4.72[6]	0
	6	120 000	12	3.68[8]	4.03[7]	3.70[8]	4.08[7]
5063	6	120 000	10	2.53[8]	0	2.76[8]	0
	7	160 000	10	2.39[10]	2.53[9]	2.57[10]	2.60[9]

products) of functions of only a single integration variable, r or r' . Using this property, and splitting the total N -dimensional volume into $N!$ subregions inside each of which the integration variables satisfy a permutation of the inequality

$$r_1 < r_2 < \dots < r_N,$$

the integral $M^{+(N)}$ can be expressed as a sum of $N!$ integrals, each of which is a product of N one-dimensional integrals whose integrands depend on only one variable. This simplification was exploited earlier.⁵ The shortcoming of this procedure is that $N!$ grows very rapidly with N , and the computational time grows exponentially. In our Monte Carlo calculations reported here, we exploited the factorization of $g_\alpha(r, r')$ as a product of Coulomb functions, without breaking the total integration volume into $N!$ subregions, by forming a relatively sparse grid of n_{int} points in r space on which the regular and irregular Coulomb wave functions were evaluated. Thus we set up two tables, each of dimensions $N \times n_{\text{int}}$, for the Coulomb wave functions. [For $|2kr| > 2(l+1+|\gamma|)$ we rapidly evaluated the Coulomb wave functions by asymptotic expansion.] The Green's function $g_\alpha(r, r')$, at any points r

and r' , was then evaluated by interpolation between tabulated points. This technique reduced the time for integrand evaluations to a reasonable level, with the total computational time increasing with N only as (roughly) N^2 .

There should be no difficulty in applying this method to alkali-metal atoms, where the quantum defect is nonzero; the (approximate) Green's function still factorizes—see, for examples, Refs. 4 and 5. However, for other multielectron atoms there remains the problem of developing a tractable approximation to the Green's function. We hope that this study will stimulate further exploration of Monte Carlo methods in the treatment of high-order multiphoton ionization.

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