High-order above-threshold ionization of hydrogen in perturbation theory

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We describe a very efficient basis-set method for calculating cross sections for high-order above-threshold ionization of a hydrogenic atom in perturbation theory. We present results for up to 20-photon ionization of H(1*s*) and D(1*s*) by 1064 nm circularly, linearly, or elliptically polarized radiation. We show the rapid decrease in the integrated cross sections as the polarization passes from linear to circular, and we illustrate the asymmetry in the angular distributions for elliptic polarization.

During the last few years many calculations have been performed for multiphoton-ionization rates of atoms, particularly hydrogen, within perturbation theory. With the notable exception of the work of Gontier *et al.*,[1] no calculations have been carried out for the case where many photons are absorbed in the continuum, although there have been numerous studies[2–7] involving the absorption of a few photons (up to three) above the ionization threshold. In this paper we show that, within perturbation theory, rates for relatively high-order "above-threshold" ionization can be obtained rather easily by modifying a technique introduced by Klarsfeld and Maquet.[3] We present cross sections for 20-photon ionization of hydrogen and deuterium by light of wavelength 1064 nm; a minimum of 12 photons must be absorbed for ionization to occur. To our knowledge, these are the first reported results for above-threshold ionization at this wavelength, the fundamental wavelength of the Nd:YAG laser.[8] We consider cases where the light is linearly, circularly, or elliptically polarized; the dependence of the cross sections, particularly the angular distributions, on the polarization of 1064-nm light has been the subject of recent experimental investigations.[9–12]

We proceed to describe our method, assuming an atomic system with only one electron. We separate the center-of-mass motion from the internal motion, and consider only the latter. We make the dipole approximation and express the interaction $V(t)$ of the electron with the radiation field, in either the length or velocity gauge, as $V(t)=V_+e^{-i\omega t}+V_-e^{i\omega t}$, where ω is the frequency of the field. (In the velocity gauge we omit the term in the square of the vector potential; it yields no contribution to the on-shell ionization matrix element in lowest-order perturbation theory.) The electron wave vector $|\phi(t)\rangle$ has the harmonic expansion

$$|\phi(t)\rangle=e^{-iE_it}\sum_p e^{-ip\omega t}|\phi_p\rangle\ , \tag{1}$$

where p is an integer. Within perturbation theory E_i is the initial binding energy of the electron, $|\phi_0\rangle$ is the initial bound-state vector, and the harmonic components with $p>0$ satisfy the coupled differential equations

$$(E_i+i\eta+p\hbar\omega-H_a)|\phi_p\rangle=V_+|\phi_{p-1}\rangle\ , \tag{2}$$

where η is positive but infinitesimal and H_a is the field-free Hamiltonian of the atom; we use the *reduced* mass for the atom in defining H_a and $V(t)$. We expand the harmonic components on a finite set of basis functions $S_{nl}^{\kappa}(r)Y_{lm}(\hat{\mathbf{r}})$, where the $Y_{lm}(\hat{\mathbf{r}})$ are the usual spherical harmonics and the $S_{nl}^{\kappa}(r)$ are the radial Sturmian functions,[3–6] which behave as $r^n\exp(i\kappa r)$ for $r\sim\infty$,

$$S_{nl}^{\kappa}(r)=A_{nl}(-i\kappa r)^{l+1}e^{i\kappa r}\times{}_1F_1(l+1-n,2l+2,-2i\kappa r)\ , \tag{3}$$

where A_{nl} is a normalization factor. We choose the wave number κ to be the same for all basis functions. An important advantage to this is that $E-H_a$ is *tridiagonal* on the basis set when the atomic potential is pure Coulombic, a special property of Sturmian functions.[13] Consequently, the linear equations for the expansion coefficients of the $|\phi_p\rangle$, obtained by projecting Eq. (2) onto the basis set, can be solved very rapidly. We must, however, choose κ appropriately. To understand this, note first that any well-behaved function of r which vanishes as r^{l+1} for $r\sim0$ and behaves as $r^{\nu}\exp(ikr)$ for $r\sim\infty$

TABLE I. Relative difference between the results obtained by expanding the radial part of each harmonic component on n_M Sturmian functions, and those obtained with $n_M=150$. The initial state is H(1*s*), the wavelength is 1064 nm, the polarization is linear, and $|\kappa|^2/2=0.3$. (a) Integrated cross sections for $N=12$, calculated in the velocity gauge with $\arg(\kappa)=80°$; (b) same as (a) but for $N=16$; (c) same as (a) but for $N=20$; (d) same as (b), but results calculated in the length gauge; (e) rate for generation of 31st harmonic, calculated in the velocity gauge with $\arg(\kappa)=60°$.

	n_M = 60	90	120
(a)	2.6×10^{-9}	4.1×10^{-9}	-6.3×10^{-12}
(b)	5.6×10^{-4}	-1.7×10^{-6}	1.2×10^{-8}
(c)	1.7×10^{-1}	3.4×10^{-4}	-4.0×10^{-6}
(d)	-7.9×10^{-2}	-1.0×10^{-2}	4.3×10^{-5}
(e)	8.4×10^{-11}	1.7×10^{-13}	-5.9×10^{-15}

TABLE II. Integrated (σ) and differential ($d\sigma$) cross sections for N-photon ionization of H(1s) and D(1s) by 1064- or 532-nm linearly (LP) or circularly (CP) polarized light. $N=N_0+S$. (a) σ/I^{N-1}, where I is the intensity, for H(1s) and LP; (b) σ/I^{N-1} for D(1s) and LP; (c) σ/I^{N-1} for H(1s) and CP; (d) σ/I^{N-1} for D(1s) and CP; (e) $d\sigma/I^{N-1}$ for H(1s), LP, and ejection along the direction of polarization; (f) $d\sigma/I^{N-1}$ for H(1s), CP, and ejection in the polarization plane. Units are $\mathrm{cm}^{2N}/\mathrm{W}^{N-1}$ for (a)–(d), $\mathrm{cm}^{2N}/\mathrm{sr\,W}^{N-1}$ for (e) and (f).

	1064 nm ($N_0=12$)						532 nm ($N_0=6$)	
S	(a)	(b)	(c)	(d)	(e)	(f)	(a)	(c)
0	1.62×10^{-167}	1.77×10^{-167}	6.08×10^{-175}	5.75×10^{-175}	1.51×10^{-167}	1.95×10^{-175}	4.63×10^{-88}	1.76×10^{-90}
1	3.57×10^{-180}	3.97×10^{-180}	1.37×10^{-185}	1.33×10^{-185}	1.69×10^{-180}	4.55×10^{-186}	9.58×10^{-102}	1.55×10^{-103}
2	4.40×10^{-193}	4.93×10^{-193}	5.21×10^{-198}	5.12×10^{-198}	1.80×10^{-193}	1.80×10^{-198}	9.87×10^{-116}	2.00×10^{-117}
3	4.17×10^{-206}	4.69×10^{-206}	5.62×10^{-211}	5.53×10^{-211}	2.15×10^{-206}	2.00×10^{-211}	9.46×10^{-130}	1.28×10^{-131}
4	3.36×10^{-219}	3.79×10^{-219}	3.13×10^{-224}	3.08×10^{-224}	2.13×10^{-219}	1.15×10^{-224}	9.08×10^{-144}	5.62×10^{-146}
5	2.36×10^{-232}	2.68×10^{-232}	1.15×10^{-237}	1.13×10^{-237}	1.79×10^{-232}	4.35×10^{-238}	8.02×10^{-158}	1.95×10^{-160}
6	1.46×10^{-245}	1.67×10^{-245}	3.18×10^{-251}	3.14×10^{-251}	1.31×10^{-245}	1.24×10^{-251}	6.31×10^{-172}	5.67×10^{-175}
7	8.10×10^{-259}	9.24×10^{-259}	7.14×10^{-265}	7.05×10^{-265}	8.46×10^{-259}	2.85×10^{-265}	4.42×10^{-186}	1.45×10^{-189}
8	4.04×10^{-272}	4.63×10^{-272}	1.36×10^{-278}	1.34×10^{-278}	4.82×10^{-272}	5.56×10^{-279}	2.79×10^{-200}	3.31×10^{-204}

can be expanded in terms of the $S_{nl}^{\kappa}(r)$, with coefficients that vanish rapidly as n increases, provided that[6]

$$|\arg(k)-\arg(\kappa)|<\pi/2\ . \tag{4}$$

Now let us define $\hbar k_p=(2\mu E_p)^{1/2}$, where $E_p=E_i+p\hbar\omega+i\eta$ and μ is the reduced electron mass. If N_0 is the minimum number of photons which the atom must absorb to ionize we have $k_p=i|k_p|$ if $p<N_0$ and $k_p=|k_p|$ if $p\geq N_0$. Each harmonic component $|\phi_p\rangle$ has[14] in perturbation theory $p+1$ subterms $|\phi_{pj}\rangle$ $(j=0,1,\ldots,p)$, where $\langle\mathbf{r}|\phi_{pj}\rangle$ behaves as $r^{\nu}\exp(ik_jr)$ for $r\sim\infty$; thus, each $|\phi_p\rangle$ with $p\geq N_0$ contains some subterms whose wave numbers point along the real axis, *and* other subterms whose wave numbers point along the imaginary axis. In view of inequality (4), it follows that we must choose κ so that $0<\arg(\kappa)<\pi/2$. This is an important distinction between our technique and that of Klarsfeld and Maquet.[3] They expanded $|\phi_p\rangle$ using $\kappa=k_p$; for this choice the Green's function $(E_p-H_a)^{-1}$ is diagonal on the Sturmian basis set.[15] However, the inequality (4) is violated for the subterms $|\phi_{pj}\rangle$, $j<N_0$, when we set $\arg(\kappa)=\arg(k_p)=0$, $p\geq N_0$; consequently, the expansion of $|\phi_p\rangle$ does not converge for $p\geq N_0$. In contrast, our expansion of $|\phi_p\rangle$ has convergent coefficients.

Once we have solved Eqs. (2) we can calculate many quantities of interest in a variety of processes, e.g. multiphoton ionization, harmonic generation, etc. The matrix element for the electron to absorb $N\geq N_0$ photons and emerge with momentum $\hbar\mathbf{k}_N$ in the continuum state represented by $|\Phi_{\mathbf{k}_N}^-\rangle$ is $M_N=\langle\Phi_{\mathbf{k}_N}^-|V_+|\phi_{N-1}\rangle$ in lowest order perturbation theory. The corresponding photoionization cross section is

$$(2\pi/\hbar)\rho'(E_p)|M_N|^2/F\ ,$$

where ρ' is the density of final states and F is the photon flux. Since $|\phi_{\mathbf{k}_N}^-\rangle$ is composed of *standing* waves, it cannot be expanded on the Sturmian basis set because inequality (4) cannot be simultaneously satisfied for $k=k_N$ and $k=-k_N$. Consequently, the expansion of M_N obtained by expanding the $|\phi_p\rangle$ on the Sturmian basis set diverges for most cases of interest. However, following Klarsfeld and Maquet[3,5] we sum this divergent series using the Padé method.

In Table I we illustrate the rate of convergence of ionization cross sections with respect to basis size. Note that convergence is more rapid in the velocity gauge than in the length gauge. Presumably this is because the harmonic components $\langle\mathbf{r}|\phi_p\rangle$, $p\geq N_0$, extend over a larger region in the length gauge than in the velocity gauge, and are therefore more difficult to represent on a discrete basis. We also include in Table I results for the rate for generation of the 31st harmonic at 1064 nm; since it involves the overlap of a harmonic component with a bound state, rather than a continuum wave function, its

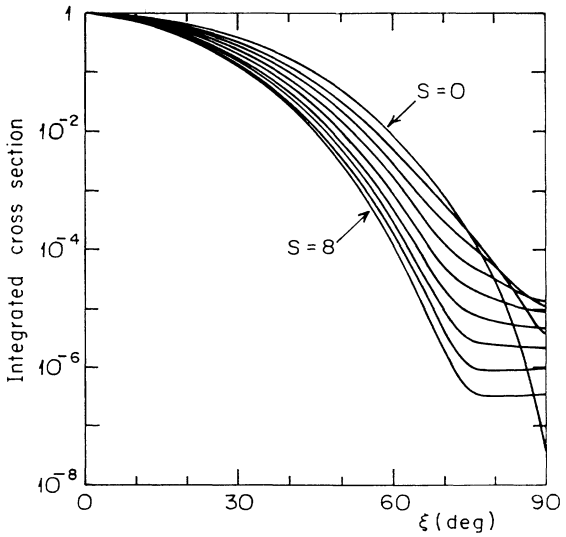

FIG. 1. Integrated cross sections for N-photon ionization of H(1s) by 1064-nm elliptically polarized radiation, as functions of the retardation angle ξ. Results are normalized to 1 at $\xi=0°$. $N=12+S$; S increases monotonically from 0 to 8 when one passes from the uppermost curve to the lowest one, when $\xi\leq70°$.

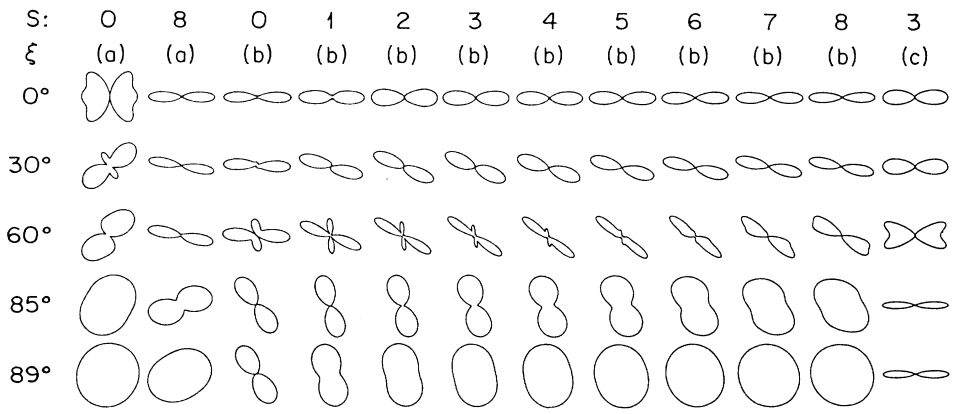

FIG. 2. Angular distributions for N-photon ionization of H($1s$) by elliptically polarized radiation, for different values of the retardation angle ξ (a) in the $0xy$ plane at 265 nm, (b) in the $0xy$ plane at 1064 nm, (c) in the $0xz$ plane at 1064 nm. $N=N_0+S$, where $N_0=3$ for 265 nm or 12 for 1064 nm. The horizontal axis is the $0x$ axis; the vertical axis is the $0y$ axis for (a) and for (b), the $0z$ axis for (c). The $0xy$ angular distributions are viewed from above in a right-handed coordinate system.

Sturmian expansion converges without Padé summation and hence it is far easier to calculate than an ionization matrix element. More detailed results for harmonic generation, and Raman scattering, will be presented elsewhere.[16]

In Table II we show cross sections for N-photon ionization of H($1s$) by linearly polarized (LP) or circularly (CP) polarized light of 532-nm or 1064-nm wavelengths, with $S=N-N_0$ ranging from 0 to 8. As is well known, LP is more efficient than CP in multiphoton ionization. For LP the number of accessible intermediate atomic states is greater, and accordingly a small energy denominator is more likely to occur, especially at long wavelengths. At 1064 nm we include results for ionization of D($1s$); although there is no "exact" resonance, the sensitivity of the cross sections on the atomic energy spectrum, through the reduced mass, is evident in the significant difference between the results for H and D.[17] (The difference is much smaller at 265, 355, and 532 nm.) From Table II we can compute the strengths of consecutive above-threshold peaks. At the wavelength 1064 nm and an intensity of 10^{12} W/cm^2 (perturbation theory is inapplicable at intensities much higher than this) we find that for LP the strengths of the above-threshold peaks diminish slowly but monotonically as S increases, while for CP the $S=1$ peak strength is about a factor of 23 times larger than the $S=0$ peak strength. This relative weakness of the $S=0$ peak in CP and not LP is not observed when N_0 is small, but otherwise we found it to be quite general. A probable explanation for this feature has been given by Bucksbaum *et al.*[9]

In Fig. 1 we show cross sections, integrated over angle, for ionization by elliptically polarized light whose electric field vector is $\mathrm{Re}[F_0\hat{\epsilon}\exp(-i\omega t)]$, where $\hat{\epsilon}=\cos(\xi/2)\hat{\mathbf{x}}+i\sin(\xi/2)\hat{\mathbf{y}}$. The cross sections diminish rapidly as the retardation angle ξ increases from 0° (LP) to 90° (CP). The angular distributions of the ejected electron, which are illustrated in Fig. 2 for different values of ξ, are invariant under reflection about the origin or about the polarization plane, since the initial state of the atom is spherically symmetric. As noted recently in both experiment[11] and theory,[18] the azimuthal angular distributions (for ejection in the polarization plane $0xy$) are of lower symmetry for elliptic polarization than for LP or CP; however, as seen from Fig. 2, there is often an approximate axis of symmetry whose orientation depends on ξ, the wavelength, and S. Note that neither the asymmetry nor the azimuthal anisotropy disappear until ξ is very close to 90°, although at 1064 nm these effects weaken fairly rapidly as S increases. Angular distributions for ejection in the $0xz$ plane are also shown in Fig. 2.

We end by noting that a Sturmian basis set is useful not only in treating high-order above-threshold ionization in perturbation theory, but also in the study of free-free processes, a topic currently under investigation, and in going beyond perturbation theory,[19] though the computational difficulties there are far more severe.

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