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Two-photon ionization of atomic hydrogen above the one-photon ionization threshold

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Abstract

An alternative method is presented for the evaluation of the two-photon ionization transition amplitude and transition rates of atomic hydrogen in the ground state above the one-photon ionization threshold. In this approach it is straightforward to calculate the angular distribution of the emitted electrons. These angular distributions are plotted and calculated transition rates are compared with the previously reported results.

1. Introduction

Studies of multiphoton processes in atoms and molecules provide detailed information on the structural properties and dynamics of the interaction of intense electromagnetic radiation with matter. One such elementary process is two-photon absorption which has become a useful tool for the experimental study of various spectroscopic characteristics of atoms [1–3]. For the case of two-photon ionization of atomic hydrogen there are many known ways of calculating the transition matrix elements [4–13]. The major difference in all such calculations is the treatment of the infinite summation over the complete set of intermediate states. Earlier, we presented an efficient alternative approach [14] to evaluate the sum over intermediate states, which is a variation of the Dalgarno and Lewis [15] treatment of the second-order Stark effect in the hydrogen atom. A variational method based on this approach for the calculation of multiphoton transition rates is used in [16]. In this paper we consider the two-photon ionization of atomic hydrogen from the ground state for photon energies above the one-photon ionization threshold. This method is an extension of our earlier work [14], where it was used to study two-photon transitions in the hydrogen atom from its ground state to discrete final states. The advantage of using this method is that it is very simple to include the effect of the infinite sum over the complete set of intermediate states. Our results are in excellent agreement with the results reported previously [5, 10, 12, 17].

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2. Transition matrix elements

In a two-photon absorption process, the differential cross section per unit intensity may be derived by a straightforward application of second-order perturbation theory [4]. The result is

$$\frac{1}{I} \frac{d\sigma}{d\Omega} = \frac{\alpha}{4\pi I_0} |D_{fg}|^2 \omega k a^2 \quad (1)$$

where

$$D_{fg} = \sum_i \frac{\langle f | \boldsymbol{\varepsilon} \cdot \mathbf{r} | i \rangle \langle i | \boldsymbol{\varepsilon} \cdot \mathbf{r} | g \rangle}{E_g - E_i + \omega}. \quad (2)$$

Here I is the light intensity in W cm^{-2} , $\frac{d\sigma}{d\Omega}$ is the differential cross section in $\text{cm}^2 \text{sr}^{-1}$, I_0 is $7.019 \times 10^{16} \text{ W cm}^{-2}$, α is the fine-structure constant and a is the Bohr radius. The remaining terms are all dimensionless multiples of atomic units. Thus E_g and E_i are, respectively, the energies of the initial and intermediate states of the atom in units of (e^2/a) , ω is the photon energy in units of (e^2/a) , k is the wavenumber of the emitted electron in units of $(1/a)$ and $\boldsymbol{\varepsilon}$ is the unit polarization vector of the incident radiation. The intermediate states $|i\rangle$ and the final continuum states $|f\rangle$ are normalized such that the dipole matrix elements $\langle i | \boldsymbol{\varepsilon} \cdot \mathbf{r} | g \rangle$ and $\langle f | \boldsymbol{\varepsilon} \cdot \mathbf{r} | i \rangle$ are in units of a and $a^{5/2}$, respectively, and $\sum_i |i\rangle \langle i| = \hat{I}$. The energy of the emitted electron E_e in atomic units is given by

$$E_e = 2\omega - \frac{1}{2}. \quad (3)$$

Using $E_e = \frac{1}{2}k^2$ we get

$$k = \sqrt{4\omega - 1}. \quad (4)$$

The method depends on an auxiliary dimensionless operator U which enables us to perform the summation over the whole spectrum exactly. The operator U is determined by a certain inhomogeneous differential equation which depends functionally on the initial wavefunction of the atom. We define U such that

$$\boldsymbol{\varepsilon} \cdot \mathbf{r} | g \rangle = [U H_0 - H_0 U + \omega U] | g \rangle \quad (5)$$

where H_0 is the unperturbed Hamiltonian of the hydrogen atom. In atomic units

$$H_0 = \frac{-\nabla^2}{2} - \frac{1}{r}. \quad (6)$$

Substituting this in equation (2) we get

$$D_{fg} = \langle f | \boldsymbol{\varepsilon} \cdot \mathbf{r} U | g \rangle. \quad (7)$$

Using [18], $|g\rangle = \exp(-r)/\sqrt{\pi}$ and writing U as $U(\mathbf{r}) = u(r)\boldsymbol{\varepsilon} \cdot \mathbf{r}$ in equation (5) we find that $u(r)$ satisfies the differential equation

$$r u''(r) + (4 - 2r)u'(r) + 2(\omega r - 1)u(r) = 2r. \quad (8)$$

This equation and the solution for $\omega < \frac{1}{2}$ was used in our earlier work [14]. Now for ionization above the one-photon threshold, $\omega > \frac{1}{2}$. In this case the integral representation of the solution is given by

$$u(r) = \frac{1}{\omega} \left\{ 1 - \frac{(1+i\beta)}{\omega} e^{(1+i\beta)r} \int_0^1 e^{-(1+i\beta)rt} t^p \left(\frac{\alpha-t}{\alpha-1} \right)^q dt \right\} \quad (9)$$

with $\beta = \sqrt{2\omega - 1}$, $p = 1 - i/\beta$, $q = 1 + i/\beta$ and $\alpha = 2i\beta/(1+i\beta)$. By direct substitution we can show that equation (9) is the solution of equation (8) with correct asymptotic behaviour for

the first order correction to the wavefunction. It is easy to see that $\beta \rightarrow -\beta$ is also a solution to this equation.

The final-state wavefunction [18] can be expanded as

$$|f\rangle = 4\pi \sum_{l=0}^{\infty} \sum_{m=-l}^l i^l R_{lk}(r) Y_{lm}(\hat{r}) Y_{lm}^*(\hat{k}) \quad (10)$$

where

$$R_{lk}(r) = e^{\frac{\pi\gamma}{2}} \frac{\Gamma(l+1-i\gamma)}{\Gamma(2l+2)} (2kr)^l e^{ikr} F(l+1-i\gamma, 2l+2, -2ikr) \quad (11)$$

where $\gamma = 1/k$, for attractive coulomb potential. In the case of linear polarization along the z axis we have

$$\boldsymbol{\varepsilon} \cdot \mathbf{r} = z = \sqrt{\frac{4\pi}{3}} r Y_{10}(\hat{r}). \quad (12)$$

Then, using equations (9)–(12) in (7) we obtain

$$D_{fg}^{(l)} = D_0 Y_{00}(\hat{k}) + D_2 Y_{20}(\hat{k}) \quad (13)$$

where $D_{fg}^{(l)}$ is the transition matrix element for linear polarization and

$$D_0 = \frac{8\pi}{3} e^{\frac{\pi}{2k}} \Gamma\left(1 + \frac{i}{k}\right) \int_0^{\infty} dr e^{-r} r^4 u(r) e^{-ikr} F\left(1 + \frac{i}{k}, 2, 2ikr\right) \quad (14)$$

and

$$D_2 = \frac{64\pi}{3\sqrt{5}} \left(\frac{k^2}{\omega}\right) \frac{\Gamma(3 + \frac{i}{k})}{\Gamma(6)} e^{\frac{\pi}{2k}} \int_0^{\infty} dr e^{-r} r^6 u(r) e^{-ikr} F\left(3 + \frac{i}{k}, 6, 2ikr\right). \quad (15)$$

For the case of circular polarization,

$$\boldsymbol{\varepsilon} \cdot \mathbf{r} = \sqrt{\frac{4\pi}{3}} r Y_{1\pm 1}(\hat{r}) \quad (16)$$

and by performing the algebra involving Clebsch–Gordon coefficients the transition matrix element of circular polarization is found to be

$$D_{fg}^{(c)} = \sqrt{\frac{3}{2}} D_2 Y_{2\pm 2}(\hat{k}). \quad (17)$$

Using the standard integral [19]

$$\int_0^{\infty} dr e^{-sr} r^{2l+1} F\left(l+1 + \frac{i}{k}, 2l+2, 2ikr\right) = \Gamma(2l+2) \frac{s^{\frac{1}{k}-l-1}}{(s-2ik)^{\frac{1}{k}+l+1}}. \quad (18)$$

With the relation $\int_0^{\infty} dr r^n e^{-sr} f(r) = (-\frac{\delta}{\delta s})^n \int_0^{\infty} dr e^{-sr} f(r)$, we can simplify equations (14) and (15) to obtain

$$D_0 = \frac{8\pi}{3\omega} e^{\frac{\pi}{2k}} \Gamma\left(1 + \frac{i}{k}\right) \left\{ \Phi_0[1+ik] - \frac{(1+i\beta)}{\omega} \int_0^1 dt t^p \left(\frac{\alpha-t}{\alpha-1}\right)^q \Phi_0[t+i(k-\beta+\beta t)] \right\} \quad (19)$$

and

$$D_2 = -\frac{8\pi}{45\sqrt{5}} \left(\frac{k^2}{\omega}\right) e^{\frac{\pi}{2k}} \Gamma\left(3 + \frac{i}{k}\right) \times \left\{ \Phi_2[1+ik] - \frac{(1+i\beta)}{\omega} \int_0^1 dt t^p \left(\frac{\alpha-t}{\alpha-1}\right)^q \Phi_2[t+i(k-\beta+\beta t)] \right\} \quad (20)$$

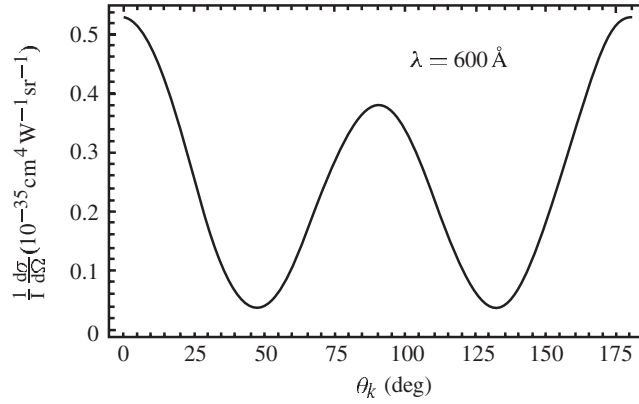


Figure 1. Two-photon ionization differential cross section per unit intensity for atomic hydrogen in ground state for linearly polarized light with wavelength $\lambda = 600 \text{ \AA}$.

Table 1. Two-photon ionization rate per unit intensity for atomic hydrogen in ground state above one-photon ionization threshold (numbers in parentheses indicate powers of 10).

λ (\AA)	$\frac{\sigma^{(l)}}{I}$ ($\text{cm}^4 \text{W}^{-1}$)				$\frac{\sigma^{(c)}}{I}$ ($\text{cm}^4 \text{W}^{-1}$)			$\frac{\sigma^{(c)}}{\sigma^{(l)}}$
	Zernik and Klopfenstein [5]	Klasfeld [10]	Karule [12]	Present work	Karule [12]	Aymar and Crance [17]	Present work	
20	—	—	1.61(-44)	1.6150(-44)	1.10(-44)	1.10(-44)	1.1003(-44)	0.6813
100	—	—	4.09(-40)	4.0896(-40)	3.02(-40)	3.02(-40)	3.0228(-40)	0.7391
200	—	—	3.02(-38)	3.0153(-38)	2.44(-38)	—	2.4409(-38)	0.8095
300	—	—	3.67(-37)	3.6683(-37)	3.22(-37)	3.22(-37)	3.2187(-37)	0.8774
400	—	—	2.15(-36)	2.1511(-36)	2.03(-36)	2.03(-36)	2.0278(-36)	0.9426
500	8.7(-36)	—	8.49(-36)	8.4931(-36)	8.53(-36)	8.54(-36)	8.5345(-36)	1.0048
600	2.7(-35)	2.61(-35)	2.62(-35)	2.6189(-35)	2.79(-35)	2.79(-35)	2.7859(-35)	1.0637
700	7.0(-35)	6.80(-35)	6.83(-35)	6.8265(-35)	7.64(-35)	7.64(-35)	7.6393(-35)	1.1190
800	1.6(-34)	1.57(-34)	1.58(-34)	1.5774(-34)	1.85(-34)	1.85(-34)	1.8463(-34)	1.1704
900	3.3(-34)	3.32(-34)	3.33(-34)	3.3317(-34)	4.06(-34)	4.06(-34)	4.0572(-34)	1.2177

where

$$\Phi_0(s) = 8 \left(\frac{s}{s-2ik} \right)^{\frac{1}{k}} \left\{ \frac{3s^3 - 9s^2(1+ik) + 6s(1+3ik-2k^2) - 1 - 6ik + 11k^2 + 6ik^3}{s^4(s-2ik)^4} \right\} \quad (21)$$

$$\Phi_2(s) = 240 \left(\frac{s}{s-2ik} \right)^{\frac{1}{k}} \frac{3s - 3ik - 1}{s^4(s-2ik)^4}. \quad (22)$$

3. Results and discussion

Using equation (13) in (1) the differential cross section per unit intensity for linearly polarized light can be written as

$$\frac{1}{I} \frac{d\sigma^{(l)}}{d\Omega} = \frac{\alpha}{4\pi I_0} \omega k a^2 |D_0 Y_{00}(\hat{\mathbf{k}}) + D_0 Y_{20}(\hat{\mathbf{k}})|^2. \quad (23)$$

Now for a given wavelength λ of the radiation, we can compute the complex amplitudes D_0 and D_2 by performing a numerical integration in equations (19) and (20). With these values of D_0 and D_2 for $\lambda = 600 \text{ \AA}$ we have plotted the angular distribution of the ejected electron in figure 1. Since the complex amplitudes are known we can easily find the value of θ_k for the minimum of the angular distribution and it occurs when $\theta_k = 47.65^\circ$. By integrating equation (23) over the solid angles the total two-photon ionization cross section per unit intensity for the linearly polarized light can be written as

$$\frac{\sigma^{(l)}}{I} = \frac{\alpha}{4\pi I_0} \omega k a^2 \{|D_0|^2 + |D_2|^2\}. \quad (24)$$

Similarly, for the case of circular polarization the total ionization cross section per unit intensity is obtained as

$$\frac{\sigma^{(c)}}{I} = \frac{\alpha}{4\pi I_0} \omega k a^2 \frac{3}{2} |D_2|^2. \quad (25)$$

Again, by doing a simple one-dimensional numerical integration over the variable t in equations (24) and (25), the calculation of the two-photon ionization rate above the single-photon ionization threshold with linearly and circularly polarized light has been carried out in the region of radiation wavelength $0 < \lambda < 911 \text{ \AA}$. Our results are compared with previous ones and are presented in table 1.

For a system other than atomic hydrogen the solution may not be a closed expression of the form given in equation (9). For that case we have to solve a differential equation similar to equation (8) numerically for the given initial state of the system. But the entire contribution of the infinite sum of intermediate states is treated exactly in this formalism.

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