Quantum calculations for photodetachment cross sections of H^- in an inhomogeneous electric field*

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We calculate the photodetachment cross sections of H^- in a gradient electric field based on traditional quantum approach. The system provides a rare example that the formulas for the cross sections can be explicitly derived by both the quantum approach and closed-orbit theory. The quantum results are compared with those of the closed-orbit theory. The correct phase values in the closed-orbit theory are essential and necessary to produce accurate cross sections. Our quantum results remove some previous ambiguities in assigning the phase values in the closed-orbit theory (G. C. Yang and M. L. Du 2007 *Phys. Rev.* A **75** 029904E).

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1. Introduction

The experiment of the photodetachment of H^- in a homogeneous static electric field has been reported in 1987.[1] The measurements show that the detachment cross sections have "ripple" structures. The theoretical investigation was carried out by Rau and Wong.[2] Du and Delos[3] carried out the theoretical studies based on wave functions in momentum space. There has been considerable interest in the photodetachment of H^- in different environments in recent years, such as near metal surfaces,[4−8] near interfaces,[9−11] in electric and magnetic fields,[12−16] in static electric field near an elastic wall or a metal surface,[17,18] and in a microcavity.[19] These systems have been investigated theoretically either using closed-orbit theory (COT)[20−22] or quantum approach or both.

Yang *et al.*[23,24] have studied the photodetachment of H^- in a gradient field a few years ago. Their derivation of the photodetachment was based on the closed-orbit theory. There are numerous closed orbits in the system and all closed orbits have contributions to the photodetachment cross section. So the obtained formula involved a summation of infinite terms. Closed-orbit theory provides a clear physical picture and a quantitative tool for the oscillations in the photodetachment or photoionization cross sections. However, a complete quantum solution can provide all possible knowledge of a system and a different physical picture.[25] Those two perspectives are complementary.

We apply quantum approach to calculate the photodetachment cross sections of H^- in the same inhomogeneous external fields of Refs. [23] and [24]. This system provides a rare example that cross section formula can be explicitly derived by both COT and quantum approach. We find that the quantum results agree well with the closed-orbit theory. The first purpose of the present studies is to provide an alternative physical picture for the photodetachment process and to understand the correspondence between the quantum and semiclassical physical pictures. In the closed-orbit theory the correct phase values related to Maslov indexes are essential and necessary to produce accurate cross sections. Previously there were some ambiguities in assigning the phase values in the closed-orbit theory approach of the problem and there is no independent method to easily verify which set of phase values is correct.[23,24] Our present quantum calculations confirm the set of phase values and clarify the ambiguities in assigning the phase values in the closed-orbit theory

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approach previously.

In the quantum approach, the cross section is directly related to the dipole matrix element. In calculations of the dipole matrix elements we use the method of frame transformation. The method of obtaining the cross section is given in Section 2 and it may also be found in Refs. [2], [15], and [17]. Throughout the article we use atomic units unless otherwise specified.

2. Photodetachment cross section by quantum approach

We use a quantum approach to derive a formula for the photodetachment cross section of H^-. Let the applied electric field be in the z direction and given by[23,24]

$$F = F_0 + \alpha z, \tag{1}$$

where F_0 is the electric field intensity at $z = 0$, α is the field gradient in z. Here we assume both F_0 and α are positive.

In the cylindrical coordinates (ρ, z, ϕ), the Hamiltonian governing the electron motion is

$$H = \frac{1}{2}(p_\rho^2 + p_z^2) + \left(F_0 + \frac{1}{2}\alpha z\right) z + V_{\rm b}(r). \tag{2}$$

Because of the cylindrical symmetry of the system the ϕ motion has been separated. The z component of the angular momentum is a constant of motion due to the symmetry and has been set to be zero. When the electron is far away from the nucleus so that the short-range potential $V_{\rm b}(r)$ is negligible.

The photodetachment cross section is given in terms of the dipole matrix elements by the following expression[15]

$$\sigma(E, F) = \frac{4\pi^2 E_{\rm p}}{c} \sum_{n=1}^{n_{\max}} |\langle f|D|i\rangle|^2, \tag{3}$$

where c is the speed of light, $E_{\rm p}$ is the photon energy, and $E_{\rm p} = E + E_{\rm b}$. E is the kinetic energy of the photo-detached electron. $E_{\rm b}$ is the binding energy which equals $k_{\rm b}^2/2$ and is approximately 0.754 eV. D is the dipole operator, for photons polarized along the direction z, $D = z$. $\langle f|$ is the final-state wave function of the electron after detachment. The final state is energy-normalized according to $\langle f_E|f_{E'}\rangle = \delta(E - E')$. $|i\rangle$ is the initial state of wave function of H^- and a good approximation to it is given by[3]

$$\psi_{\rm i} = \frac{B_{\rm s}\, {\rm e}^{-k_{\rm b} r}}{r}, \tag{4}$$

where $B_{\rm s}$ is a scale factor and with the value 0.31552.

The final state wave functions can be taken to be the wave functions of an electron in the field $F_0 z + \frac{1}{2}\alpha z^2$. This exact energy-normalized wave function for an electron of energy $E = k^2/2$, with $E_z = q_n^2/2$ apportioned to the z motion can be written as

$$\begin{aligned} &\Psi^F_{E,n,m}(\rho, z, \phi) \\ &= (2\pi)^{-1/2}\, {\rm e}^{{\rm i}\, m\phi}\, {\rm J}_m[(k^2 - q_n^2)^{1/2}\rho] \left(\frac{\alpha^{1/4}}{\pi^{1/2} 2^n n!}\right)^{1/2} \\ &\quad \times \exp\left[-\frac{\alpha^{1/2}}{2}\left(z + \frac{F_0}{\alpha}\right)^2\right] \\ &\quad \times {\rm H}_n[\alpha^{1/4}(z + F_0/\alpha)], \end{aligned} \tag{5}$$

where $\rm J_m$ is the regular Bessel function[26] and $\rm H_n$ is the Hermite polynomials.[27] The wave functions in Eq. (5) are correctly energy normalized as required and shown in Appendix A. The eigenenergies of z motion are

$$E_z = \left(n + \frac{1}{2}\right)\alpha^{1/2} - \frac{F_0^2}{2\alpha}. \tag{6}$$

For the purpose of calculations, we transform the final wave function from spherical coordinate to cylindrical coordinate. The detailed procedure is as follows: the energy-normalized solutions for the free field in cylindrical coordinates are[2]

$$\begin{aligned} \Psi^{\pm}_{E,n,m} &= (\pi q_n)^{-1/2}(2\pi)^{-1/2}\, {\rm e}^{{\rm i}\, m\phi}\, {\rm J}_m[(k^2 - q_n^2)^{1/2}\rho] \\ &\quad \times \begin{cases} \cos(q_n z), & \Pi_z = +\\ \sin(q_n z), & \Pi_z = -, \end{cases} \end{aligned} \tag{7}$$

where Π_z is the parity of the solution. Writing the field-free wave function as an expansion in spherical functions,[15] we can obtain

$$\psi^{+}_{E,n,m} = {\sum_l}' U_{q_n,l}(n, m) f_{lm}(\boldsymbol{r}), \tag{8}$$

and similarly

$$\psi^{-}_{E,n,m} = {\sum_l}'' U_{q_n,l}(n, m) f_{lm}(\boldsymbol{r}), \tag{9}$$

where

$$\begin{aligned} f_{lm}(\boldsymbol{r}) &= \left(\frac{1}{2\pi}\right)^{1/2} N_{lm}\, {\rm e}^{{\rm i}\, m\phi}\, {\rm P}_{lm}(\cos\theta) \\ &\quad \times \left(\frac{2k}{\pi}\right)^{1/2} j_l(kr) \end{aligned} \tag{10}$$

with

$$N_{lm} = (-1)^{(|m|+m)/2} {\rm i}^l \left[\frac{2l+1}{2}\frac{(l - |m|)!}{(l + |m|)!}\right]^{1/2} \tag{11}$$

are the energy-normalized wave functions of an free electron in spherical coordinates.

The summation $\sum_l'$ in Eq. (8) should include all the l's such that $(-1)^l = (-1)^m$ and the summation $\sum_l''$ in Eq. (9) should include all the l's such that $(-1)^l = (-l)^{m+1}$. The expansion coefficient is

$$U_{q_n,l}(n,m) = \left(\frac{2l+1}{kq_n}\right)^{1/2} \left(\frac{(l-m)!}{(l+m)!}\right)^{1/2} \times (-1)^{[(l-1-m)/2]} \mathrm{P}_{lm}(q_n/k), \quad (12)$$

where the $[x]$ in the $(-1)^{[x]}$ denotes the smallest integer which is bigger than or equal to x.

Equation (5) can be written as a linear combination of Eqs. (8) and (9),[2]

$$\Psi^F_{E,n,m}(\rho,z,\phi) = A\psi^+_{E,n,m}(\rho,z,\phi) + B\psi^-_{E,n,m}(\rho,z,\phi). \quad (13)$$

Insetting Eqs. (8) and (9) into Eq. (13), we obtain

$$\Psi^F_{E,n,m}(\rho,z,\phi) = A\sum_l{}' U_{q_n,l}(n,m) f_{lm}(\boldsymbol{r}) + B\sum_l{}'' U_{q_n,l}(n,m) f_{lm}(\boldsymbol{r}). \quad (14)$$

Upon evaluating both sides at $z = 0$, we have

$$A = \left(\frac{\alpha^{1/4}}{\pi^{1/2}2^n n!}\right)^{1/2} \times \mathrm{e}^{-F_0^2/2\alpha^{3/2}} \mathrm{H}_n[F_0\alpha^{-3/4}](\pi q_n)^{1/2}, \quad (15)$$

$$B = \left(\frac{\pi}{q_n}\right)^{1/2} \left(\frac{\alpha^{1/4}}{\pi^{1/2}2^n n!}\right)^{1/2} \times \mathrm{e}^{-F_0^2/2\alpha^{3/2}} \left[\alpha^{1/4}\,\mathrm{H}'_n(F_0\alpha^{-3/4}) - \frac{F_0}{\alpha^{1/2}}\,\mathrm{H}_n(F_0\alpha^{-3/4})\right]. \quad (16)$$

Inserting Eqs. (15) and (16) into Eq. (14), the final wave functions can be obtained.

The initial state ψ_i is a loosely bound S state. According to the dipole selection rule, only the $l = 1$ term in Eq. (14) contributes to the cross section $\sigma(E,F)$ for laser polarized along z direction. The resulting analytical formula derived from the traditional quantum approach for the photodetachment cross section is

$$\sigma(E,F) = \sigma_0(E)\frac{3\pi^{1/2}\alpha^{1/4}}{(2E)^{3/2}} \times \mathrm{e}^{-(F_0\alpha^{-3/4})^2} \sum_{n=1}^{n_{\max}} \left[\alpha^{1/4}\,\mathrm{H}'_n(F_0\alpha^{-3/4}) - \frac{F_0}{\alpha^{1/2}}\,\mathrm{H}_n(F_0\alpha^{-3/4})\right]^2 \left(\frac{1}{2^n n!}\right), \quad (17)$$

where $\sigma_0(E)$ is the free field cross section and can be expressed as

$$\sigma_0(E) = \frac{16\pi^2 2^{1/2} B_{\mathrm{s}}^2}{3c} \frac{E^{3/2}}{(E_{\mathrm{b}}+E)^3}. \quad (18)$$

In Eq. (17), $n_{\max}$ is the maximum number of energy levels in the potential well for a given energy E and α and it can be written as

$$n_{\max} = \mathrm{Int}\left(\frac{E}{\alpha^{1/2}} + \frac{F_0^2}{2\alpha^{3/2}} - \frac{1}{2}\right), \quad (19)$$

where Int denotes the integer part of the expression. The detailed derivation of the cross section formula in Eq. (17) is given in Appendix B.

3. Results and discussions

The photodetachment cross sections in Eq. (17) can be calculated for different values of F_0 and α. In Figs. 1 and 2, we compare our quantum results (solid lines) and the COT results with the corrected phase values provided later.[23,24] The quantum results confirmed that the correct phase values are important in COT. The cross sections apparently display staircase structures. The slight deviation between the two results may be attributed to the approximations used in both methods.

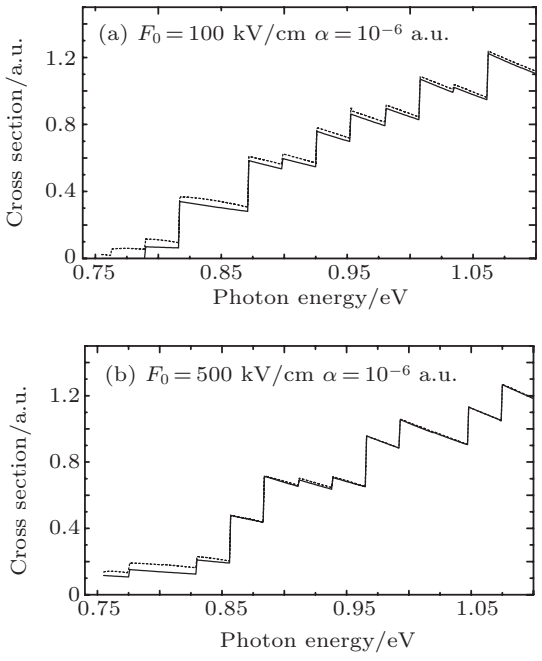


Fig. 1. Our quantum cross sections (solid lines) are compared with the COT results (dashed lines). The gradient of the field is $\alpha = 1 \times 10^{-6}$ a.u. The constant term of the field is (a) $F_0 = 100$ kV/cm and (b) $F_0 = 500$ kV/cm respectively.

In Fig. 2, we show the influence of α on the photodetachment cross section for a fixed value of F_0. We can see that as α increases, the sizes of the staircases increase. This can be understood readily from the present quantum approach. As α increases, the potential in the z direction is steeper, the corresponding energy levels become more separated.

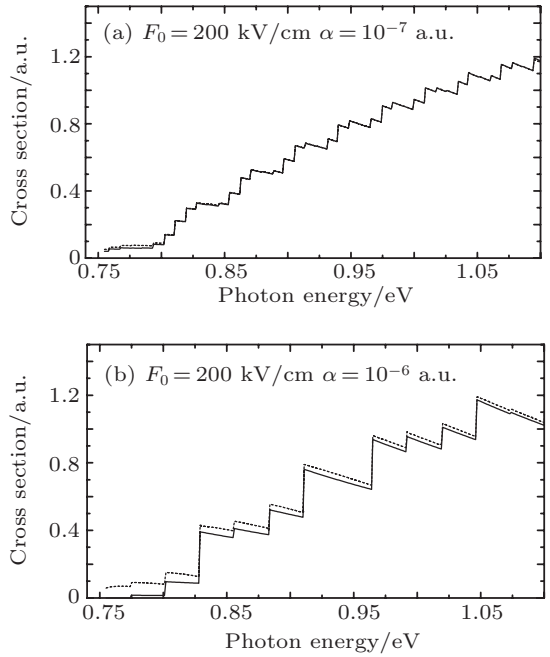


Fig. 2. Our quantum cross sections (solid lines) are compared with the COT results (dashed lines). The constant term of the electric field is $F_0 = 200$ kV/cm. The gradient of the field is (a) $\alpha = 10^{-7}$ a.u. and (b) $\alpha = 10^{-6}$ a.u. respectively.

Finally, we compare the efficiency of the quantum method with the COT. The COT formula is a sum over all closed-orbits and each closed-orbit corresponds to one oscillatory term in the cross section. In Fig. 3, we show how the finite sum of closed-orbits converges to our formula in Eq. (17) for a typical case with $F_0 = 500$ kV/cm and $\alpha = 10^{-6}$ a.u. Figure 3(a) demonstrates that four closed-orbits already give a main structure in the quantum cross sections. When we use 100 closed-orbits as shown in Fig. 3(b), the COT result essentially reproduces the quantum cross section with sharp edges. The number of required terms in the quantum formula varies with the photon energy, F_0 and α. In the case of Fig. 3, when $E_{\rm P} = 1.1$ eV, the number of required terms in the quantum formula is 16. But if we set $\alpha = 10^{-9}$ a.u. and keep the same F_0 and $E_{\rm P} = 1.1$ eV, the number of required terms in the quantum formula in Eq. (17) reaches about 1.5×10^5! Therefore the present quantum method is less economical for small α.

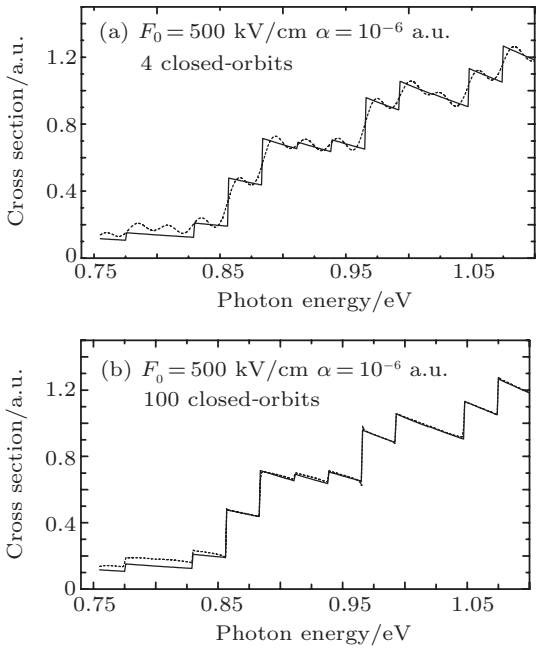


Fig. 3. The quantum cross sections (solid lines) are compared with the COT results (dashed lines) using a limited number of closed orbits for (a) four closed orbits and (b) 100 closed orbits. The constant term of the electric field is $F_0 = 500$ kV/cm. The gradient of the field is $\alpha = 10^{-6}$ a.u.

Appendix A: Energy normalization

To prove that the wave functions given in Eq. (5) are correctly energy normalized, we calculate

$$
\begin{aligned}
&\langle \Psi^F_{E,n,m} | \Psi^F_{E',n',m'} \rangle \\
&= \int_0^{2\pi} \frac{1}{2\pi} \mathrm{e}^{-\mathrm{i}\, m\phi} \mathrm{e}^{-im'\phi} \mathrm{d}\phi \int_{-\infty}^{+\infty} \sqrt{\frac{\alpha^{1/4}}{\pi^{1/2} 2^n n!}} \sqrt{\frac{\alpha^{1/4}}{\pi^{1/2} 2^{n'} n'!}} \exp\left\{ - \left[\alpha^{1/4} \left(z + \frac{F_0}{\alpha} \right) \right]^2 \right\} \\
&\quad \times \mathrm{H}_n \left[\alpha^{1/4} \left(z + \frac{F_0}{\alpha} \right) \right] \mathrm{H}_{n'} \left[\alpha^{1/4} \left(z + \frac{F_0}{\alpha} \right) \right] \mathrm{d}z \int_0^{+\infty} \mathrm{J}_m(\sqrt{k^2 + q_n^2}\rho)\, \mathrm{J}_{m'}(\sqrt{k'^2 + q_{n'}^2}\rho) \rho \mathrm{d}\rho \\
&= \delta_{mm'} \int_{-\infty}^{+\infty} \mathrm{d}z \sqrt{\frac{1}{\pi^{1/2} 2^n n!}} \sqrt{\frac{1}{\pi^{1/2} 2^{n'} n'!}} \exp\left\{ - \left[\alpha^{1/4} \left(z + \frac{F_0}{\alpha} \right) \right]^2 \right\} \mathrm{H}_n \left[\alpha^{1/4} \left(z + \frac{F_0}{\alpha} \right) \right] \\
&\quad \times \mathrm{H}_{n'} \left[\alpha^{1/4} \left(z + \frac{F_0}{\alpha} \right) \right] \mathrm{d} \left[\alpha^{1/4} \left(z + \frac{F_0}{\alpha} \right) \right] \int_0^{+\infty} \mathrm{J}_m(\sqrt{k^2 - q_n^2}\rho)\, \mathrm{J}_m(\sqrt{k'^2 - q_{n'}^2}\rho) \rho \mathrm{d}\rho. \qquad \text{(A1)}
\end{aligned}
$$

After using the following property of Hermite polynomials,

$$\int_{-\infty}^{+\infty} \frac{1}{\sqrt{\pi}2^n n!}\,\mathrm{e}^{-\varepsilon^2}\,\mathrm{H}_n(\varepsilon)\,\mathrm{H}_{n'}(\varepsilon)\,\mathrm{d}\varepsilon = \delta_{nn'},$$

the above result is reduced to

$$\begin{aligned}&\langle \Psi^F_{E,n,m}|\Psi^F_{E',n',m'}\rangle \\ &= \delta_{mm'}\delta_{nn'}\int_0^{+\infty} \mathrm{J}_m(\sqrt{k^2-q_n^2}\rho) \\ &\quad\times\ \mathrm{J}_m(\sqrt{k'^2-q_{n'}^2}\rho)\rho\,\mathrm{d}\rho. \end{aligned} \tag{A2}$$

Now applying the orthogonality relation for Bessel functions[28]

$$\int_0^{+\infty} \mathrm{J}_m(\lambda\rho)\,\mathrm{J}_m(\lambda'\rho)\rho\,\mathrm{d}\rho = \frac{1}{\lambda}\delta(\lambda-\lambda'), \tag{A3}$$

we then have

$$\begin{aligned}&\langle \Psi^F_{E,n,m}|\Psi^F_{E',n',m'}\rangle \\ &= \delta_{mm'}\delta_{nn'}\frac{1}{\sqrt{k^2-q_n^2}}\,\delta\left[\sqrt{k^2-q_n^2}-\sqrt{k'^2-q_{n'}^2}\right] \\ &= \delta_{mm'}\delta_{nn'}2\delta(k^2-k'^2) = \delta_{mm'}\delta_{nn'}\delta\left(\frac{k^2}{2}-\frac{k'^2}{2}\right) \\ &= \delta_{mm'}\delta_{nn'}\delta(E-E'). \end{aligned} \tag{A4}$$

Therefore the wave functions $\Psi^F_{E,n,m}$ in Eq. (5) are energy-normalized.

Appendix B: Derivation of cross section formula

Here we derive the cross section formula in Eq. (17). Inserting the wave function Eq. (14) and the initial state wave function Eq. (4) into Eq. (3), for laser polarized in z direction, $D = z$, we have

$$\begin{aligned}\langle f|D|i\rangle &= \int\left[A\sum_l{}' U_{q_n,l}(n,m)f^*_{lm}(\boldsymbol{r}) + B\sum_l{}'' U_{q_n,l}(n,m)f^*_{lm}(\boldsymbol{r})\right] r\cos\theta\frac{B_\mathrm{s}\,\mathrm{e}^{-k_\mathrm{b}r}}{r}\,\mathrm{d}V \\ &= \int\left[A\sum_l{}' U_{q_n,l}(n,m)\mathrm{Y}^*_{lm}(\theta,\phi)\sqrt{\frac{2k}{\pi}}\,\mathrm{j}_l(kr) + B\sum_l{}'' U_{q_n,l}(n,m)\mathrm{Y}^*_{lm}(\theta,\phi)\sqrt{\frac{2k}{\pi}}\mathrm{j}_l(kr)\right] \\ &\quad\times\ r\cos\theta\frac{B_\mathrm{s}\,\mathrm{e}^{-k_\mathrm{b}r}}{r}\,\mathrm{d}V \\ &= \int\left[A\sum_l{}' U_{q_n,l}(n,m)\mathrm{Y}^*_{lm}(\theta,\phi)\sqrt{\frac{2k}{\pi}}\,\mathrm{j}_l(kr) + B\sum_l{}'' U_{q_n,l}(n,m)\mathrm{Y}^*_{lm}(\theta,\phi)\sqrt{\frac{2k}{\pi}}\,\mathrm{j}_l(kr)\right] \\ &\quad\times\ r\sqrt{\frac{4\pi}{3}}\mathrm{Y}_{10}(\theta,\phi)\frac{B_\mathrm{s}\,\mathrm{e}^{-k_\mathrm{b}r}}{r}\,\mathrm{d}V \\ &= A\sum_l{}'\int U_{q_n,l}(n,m)\mathrm{Y}^*_{lm}(\theta,\phi)\sqrt{\frac{2k}{\pi}}\,\mathrm{j}_l(kr)r\sqrt{\frac{4\pi}{3}}\mathrm{Y}_{10}(\theta,\phi)\frac{B_\mathrm{s}\,\mathrm{e}^{-k_\mathrm{b}r}}{r}\,\mathrm{d}V \\ &\quad+\ B\sum_l{}''\int U_{q_n,l}(n,m)\mathrm{Y}^*_{lm}(\theta,\phi)\sqrt{\frac{2k}{\pi}}\,\mathrm{j}_l(kr)r\sqrt{\frac{4\pi}{3}}\mathrm{Y}_{10}(\theta,\phi)\frac{B_\mathrm{s}\,\mathrm{e}^{-k_\mathrm{b}r}}{r}\,\mathrm{d}V, \end{aligned} \tag{B1}$$

where $\mathrm{Y}_{lm}(\theta,\phi)$ is the spherical harmonic function. Applying the orthogonality relation of the spherical harmonic functions,

$$\int \mathrm{Y}^*_{lm}(\theta,\phi)\,\mathrm{Y}_{l'm'}(\theta,\phi)\sin\theta\,\mathrm{d}\theta\,\mathrm{d}\phi = \delta_{ll'}\delta_{mm'},$$

we have

$$\begin{aligned}\langle f|D|i\rangle &= A\sum_l{}'\delta_{l1}\delta_{m0}U_{q_n,l}(n,m)\sqrt{\frac{2k}{3}}2B_\mathrm{s} \\ &\quad\times\int \mathrm{j}_l(kr)\,\mathrm{e}^{-k_\mathrm{b}r}r^2\,\mathrm{d}r \\ &\quad+\ B\sum_l{}''\delta_{l1}\delta_{m0}U_{q_n,l}(n,m)\sqrt{\frac{2k}{3}}2B_\mathrm{s} \\ &\quad\times\int \mathrm{j}_l(kr)\,\mathrm{e}^{-k_\mathrm{b}r}r^2\,\mathrm{d}r. \end{aligned} \tag{B2}$$

Note that $\sum'_l$ includes all the l's such that $(-1)^l = (-1)^m$ and $\sum''_l$ includes all the l's such that $(-1)^l = (-1)^{m+1}$, then the matrix element is

$$\begin{aligned}\langle f|D|i\rangle &= BU_{q_n,1}(n,0)\sqrt{\frac{2k}{3}}2B_\mathrm{s}\int \mathrm{j}_1(kr)\,\mathrm{e}^{-k_\mathrm{b}r}r^2\,\mathrm{d}r \\ &= B\sqrt{\frac{3q_n}{k}}\frac{1}{k}\sqrt{\frac{2k}{3}}2B_\mathrm{s}\frac{2k}{(k^2+k_\mathrm{b}^2)^2} \\ &= B\sqrt{2q_n}\frac{4B_\mathrm{s}}{(k^2+k_\mathrm{b}^2)^2}. \end{aligned} \tag{B3}$$

Inserting this dipole matrix element into Eq. (3) and

using Eq. (16), we obtained the cross section formula Eq. (17). The value for $n_{\max}$ in Eq. (19) is equivalent to the requirement that the transverse energy $E - E_z \geq 0$ which is assumed in the approximation in Eq. (14).

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