Orientation effects in thermal collisions between "circular"—Rydberg-state atoms and ground-state helium

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A general formulation for thermal collisions between a Rydberg-state atom and a ground-state rare-gas atom is developed within the framework of the impulse approximation. This formulation allows calculation of cross sections for state-to-state transitions for an arbitrary initial orientation between the Rydberg-state atom and the relative velocity of the two partners of the collision. It also allows a direct computation of these cross sections averaged over all orientations, a situation corresponding to a cell experiment. In this formulation, the differential cross sections with respect to the modulus of the momentum transfer are obtained analytically in terms of rotation matrix elements. Numerical applications are made for the case of a sodium atom in a "circular" Rydberg state ($|n,l,m\rangle$, with $l=m=n-1$) colliding with helium. The collisional processes are found to be highly anisotropic. In particular, a selection rule may appear, or disappear, depending upon the initial orientation. The extension to the case where an external electric field is present is also discussed, with special emphasis on symmetry properties.

I. INTRODUCTION

A state-to-state transition in thermal collisions between a Rydberg-state atom A and a ground-state neutral atom B, with relative momentum q, can be written as

$$(A(i)+B,(\mathbf{q}))\rightarrow(A(i')+B,(\mathbf{q'})) , \qquad (1)$$

where i denotes all quantum numbers of A in its initial state. The initial and final atomic states $|i\rangle$, $|i'\rangle$ are assumed to be eigenstates of the atomic angular momentum relative to some z axis, i.e.,[1]

$$L_z|i\rangle=m|i\rangle , \qquad (2a)$$

$$L_z|i'\rangle=m'|i'\rangle . \qquad (2b)$$

Process (1) generally depends on the orientation angle α between the relative momentum vector q of the two partners of the collision and the z axis. The collisional process (1) is invariant if both the planes $(\mathbf{q},z),(\mathbf{q'},z)$, are rotated the same arbitrary angle along the z axis. Such a rotation corresponds to modification of $|i\rangle$, $|i'\rangle$, only by phase factors, and is therefore irrelevant for a change in the physical processes.

The more anisotropic the state $|i\rangle$ is, the more important the orientation effect is expected to be. The so-called "circular" states $|n,l=n-1,m=n-1\rangle$ therefore appear to be good candidates for a study of the role of the orientation.

Collisional processes involving circular Rydberg states already have suggested theoretical works.[2,3] The interest for these processes increases as the experimental production of circular Rydberg states has now been demonstrated to be feasible.[4,5] Process (1) has been considered in Ref. 3 for parallel orientation ($\alpha=0$), within the framework of the impulse approximation.

In this paper we provide a general formulation for state-to-state thermal collisions (1), according to the impulse approximation in its simplest form [Eq. (5) below]; this formulation allows computations of cross sections for an arbitrary angle α, as well as a direct computation of the cross sections averaged over all orientations. Numerical applications are done for the case:

$$(\mathrm{Na}(n=20,l=19,m=19)+\mathrm{He},\mathbf{q})$$

$$\rightarrow(\mathrm{Na}(n'=20,l',m')+\mathrm{He},\mathbf{q'}) . \qquad (3)$$

The influence of the angle α is found to be important, as will be seen in Sec. IV. Section II gives the basic formulation of the problem. Sec. II A considers the differential cross section $dS/d\Omega$ for process (1); $d\Omega$ denotes the solid angle corresponding to $\mathbf{q'}$. Section II B gives a method for obtaining the differential cross section dS/dK in the case $\alpha=0$, where K is the modulus of the momentum transfer $\mathbf{K}$ defined as

$$\mathbf{K}=\mathbf{q'}-\mathbf{q} . \qquad (4)$$

Section II C gives the method for obtaining dS/dK for an arbitrary value of α; it also provides a method for obtaining directly dS/dK averaged over all orientations. Section III gives the expression of the hydrogenic form factors in terms of rotation matrix elements. Section IV discusses the results obtained for process (3). Section V extends this work for the case where an external electric field is present.

II. BASIC FORMULATION

A. $dS/d\Omega$ and its symmetries

According to the impulse approximation in its simplest form, the differential cross section for process (1) is given by

$$dS/d\Omega = (q'/q)[(M_1/M_{12})L \, |\, \langle i' \, | \exp(-i\mathbf{K}\cdot\mathbf{r}) | i \rangle \, |\,]^2 \, ,$$

$$(5)$$

where M_1 is the reduced mass of A and B, M_{12} the reduced mass of B and e^-, and L the scattering length associated with a zero-energy collision between B and a free electron. q' and q are related by energy conservation:

$$q^2/(2M_1) + \epsilon = (q')^2/(2M_1) + \epsilon' \, ,$$

$$(6)$$

where ϵ (ϵ') denotes the negative binding energy of A in its initial (final) state.

For a derivation of Eq. (5), its relation to Faddeev-Watson expansion, and its validity for the thermal process (1), see, e.g., Ref. 6. We only recall here that the impulse approximation is expected to be valid when the eccentricity $[1 - l(l+1)/n^2]^{1/2}$ is small; the circular states therefore appear particularly suitable for this study. At this stage it is worthwhile to note two symmetry relations (see the Appendix) that are of significant importance for state-to-state collisions:

$$|\, \langle n',l',m' \, | \exp(-i\mathbf{K}\cdot\mathbf{r}) | n,l,m \rangle \, |$$

$$= |\, \langle n',l',m' \, | \exp(i\mathbf{K}\cdot\mathbf{r}) | n,l,m \rangle \, |\ , \quad (7)$$

$$|\, \langle n',l',m' \, | \exp(-i\mathbf{K}\cdot\mathbf{r}) | n,l,m \rangle \, |$$

$$= |\, \langle n',l',-m' \, | \exp(i\mathbf{K}\cdot\mathbf{r}) | n,l,-m \rangle \, |\ . \quad (8)$$

Both Eqs. (7) and (8) refer to symmetry properties of the isolated atom A. Equation (7) is a direct consequence of space inversion symmetry and reduces the fundamental range of variation of α from $[0,\pi]$ to $[0,\pi/2]$ since the two differential cross sections dS/dK corresponding to α and to $\pi - \alpha$ are equal. Equation (8) can be obtained either from time reversal or rotational symmetry (see the Appendix). Equations (7) and (8) show explicitly that reversing simultaneously both the sign of m and m' leaves the differential cross section dS/dK invariant.

The method for computing dS/dK for an arbitrary value of α will be given in Sec. II C. The following paragraph gives another method valid only for $\alpha = 0$. In all cases, the integrated cross section S is obtained from dS/dK by numerical integration:

$$S = \int_{|q-q'|}^{q+q'} dK \, dS/dK \ . \quad (9)$$

B. dS/dK for $\alpha = 0$

Several numerical applications based on relation (5) or on closely related expressions have been done (see, e.g., Refs. 6–12). These works only consider the cross sections dS/dK averaged over the initial magnetic quantum numbers m and summed over the final m'. The resulting cross section is independent of the angle α. The method used in Refs. 6–11 involves an expansion of $\exp(-i\mathbf{K}\cdot\mathbf{r})$ in Eq. (5) in terms[13] of spherical Bessel functions j_λ and spherical harmonics y_λ^μ, i.e.,

$$\exp(-i\mathbf{K}\cdot\mathbf{r}) = 4\pi \sum_{\lambda,\mu} (-i)^\lambda j_\lambda(Kr)[Y_\lambda^\mu(\hat{r})]^* Y_\lambda^\mu(\hat{K}) \quad (10)$$

and leads to the following expression:

$$\frac{1}{2l+1} \sum_{m,m'} dS(n,l,m \to n',l',m')/dK$$

$$= 2\pi(2l'+1)[(M_1/M_{12})L/q]^2$$

$$\times K \left[\sum_\lambda (2\lambda+1) \begin{bmatrix} l' & \lambda & l \\ 0 & 0 & 0 \end{bmatrix}^2 \right.$$

$$\times \left| \int_0^\infty dr \, r^2 j_\lambda(Kr) R_{n'l'}(r) R_{nl}(r) \right|^2 \right] .$$

$$(11)$$

$R_{n'l'}(r)$ and $R_{n,l}(r)$ are the radial wave functions of atom A. In the case where α is zero, it is possible to go from $dS/d\Omega$ to dS/dK in a similar way.

More precisely, we now define two orthonormal right-hand systems of axes denoted I and II. System I admits a orthonormal right-hand basis $\hat{e}_1,\hat{e}_2,\hat{q}$, where $\hat{q}$ means $\mathbf{q}/q$ (see Fig. 1). The orientation of the pair $\hat{e}_1,\hat{e}_2$ with respect to $\hat{q}$ is determined by requiring that a positive rotation of angle α ($0 \le \alpha \le \pi$) along $\hat{e}_2$ carry $\hat{q}$ into the axis z of Eq. (2). (If $\hat{q}$ is parallel to z, the orientation of the pair $\hat{e}_1,\hat{e}_2$ is arbitrary.) System II is defined to be the system I transformed by the previous rotation. The axis z of Eq. (2) is therefore also an axis for system II. This means that system II coincides with the system relative to which the states $|i\rangle$, $|i'\rangle$ of Eqs. (2) are defined, except for a possible rotation along the axis z; such a rotation affects $|i\rangle$, $|i'\rangle$ only by phase factors and is therefore irrelevant for the physical processes (1) (see Sec. I). Let $\theta_{q'}, \varphi_{q'}$ be the polar and azimuthal angles of $\mathbf{q}'$ relative to system I ($\mathbf{q}$ being the polar axis). One has

$$\cos\theta_{q'} = [q^2 + (q')^2 - K^2]/(2qq') \ . \quad (12)$$
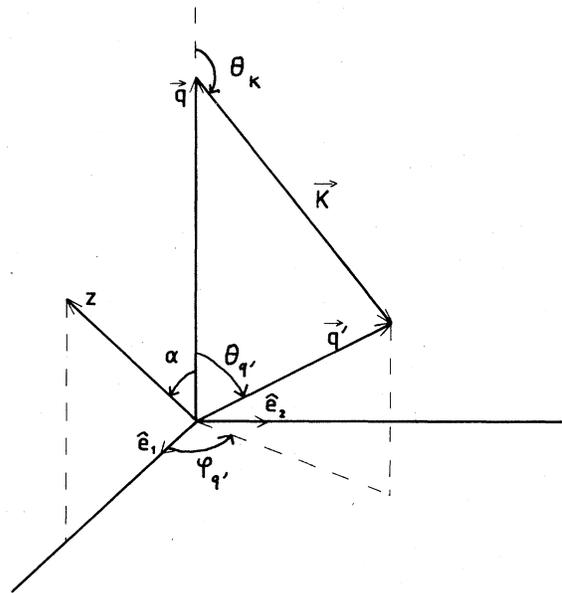
FIG. 1. Schematic diagram of the coordinate system.

It is clear that for the case $\alpha=0$, the two systems I and II coincide. It follows that the azimuthal angle φ of **K** in Eq. (10) is equal to $\varphi_{q'}$. The solid angle $d\Omega$ can therefore be written as

$$d\Omega = -d\cos\theta_{q'}\,d\varphi_{q'} = (K/qq')dK\,d\varphi \ . \tag{13}$$

Using Eq. (10), the integration over φ can be performed. Using the following expression[13] of an integral of three spherical harmonics in term of 3-j symbols:

$$\int d\hat{r}\, Y_{l'}^{m'}(\hat{r})Y_{\lambda}^{\mu}(\hat{r})Y_{l}^{m}(\hat{r})$$

$$= \left[\frac{(2l'+1)(2\lambda+1)(2l+1)}{4\pi}\right]^{1/2}$$

$$\times \begin{bmatrix} l' & \lambda & l \\ 0 & 0 & 0 \end{bmatrix}\begin{bmatrix} l' & \lambda & l \\ m' & \mu & m \end{bmatrix} \tag{14}$$

one obtains after some calculation

$$dS(n,l,m \to n',l',m';\alpha=0)/dK$$

$$= 8[\pi(M_1/M_{12})L/q]^2(2l'+1)(2l+1)$$

$$\times K\left|\int_0^\infty dr\sum_\lambda(-i)^\lambda\sqrt{2\lambda+1}\,r^2 R_{n'l'}(r)j_\lambda(Kr)R_{nl}(r)\begin{bmatrix} l & l' & \lambda \\ 0 & 0 & 0 \end{bmatrix}\begin{bmatrix} l & l' & \lambda \\ -m & m' & m-m' \end{bmatrix}Y_\lambda^{m-m'}(\theta,0)\right|^2 \tag{15a}$$

with

$$\cos\theta = [(q')^2 - q^2 - K^2]/2qK \ . \tag{15b}$$

In the case of arbitrary values of α, the preceding formulation is no longer valid, since the azimuthal angle φ of **K** relative to system II will be different from the azimuthal angle, denoted φ_K, of **K** relative to system I. Another method allowing the computation of dS/dK for arbitrary values of α as well as an averaged value of dS/dK over all orientations is given in the following paragraph.

C. dS/dK for arbitrary α and averaged over α

A third system of axis, III, having one of its three basis vectors parallel to **K**, is defined as follows. Let θ_K and φ_K be the polar and azimuthal angles of **K** relative to system I. φ_K is then equal to $\varphi_{q'}$ defined in Sec. II B. System I is first rotated an angle θ_K along the vector $\hat{e}_2$; the resulting system (say I') is then rotated an angle φ_K along $\hat{q}$. The resulting system defines system III. The key point is now to express the state $|n,l,m\rangle$ of Eq. (2) in terms of the states $|n,l,m;\text{III}\rangle$. The notation $|n,l,m;\text{III}\rangle$ indicates an eigenstate of the component of the atomic angular momentum along **K**:

$$L_K|n,l,m;\text{III}\rangle = m|n,l,m;\text{III}\rangle \ . \tag{16}$$

In view of the above definitions of the three systems I, II, and III, the transformation is given with obvious notation by

$$|n,l,m\rangle = \sum_{m_1,m_2}|n,l,m_2;\text{III}\rangle\langle l,m_2;\text{III}|l,m_1;\text{I}\rangle$$

$$\times\langle l,m_1;\text{I}|l,m\rangle \ . \tag{17}$$

The coefficients in Eq. (17) are the following rotation matrix elements:[14]

$$\langle l,m_2;\text{III}|l,m_1;\text{I}\rangle$$

$$= \langle l,m_2;\text{I}|\exp(i\theta_K L_2)\exp(i\varphi_K L_{\hat{q}})|l,m_1;\text{I}\rangle \tag{18a}$$

$$= \exp(im_1\varphi_K)d_{m_2,m_1}^l(-\theta_K) \ , \tag{18b}$$

$$\langle l,m_1;\text{I}|l,m\rangle = \langle l,m_1;\text{I}|\exp(-i\alpha L_2)|l,m;\text{I}\rangle \tag{19a}$$

$$= d_{m_1,m}^l(\alpha) \ . \tag{19b}$$

Relative to the system I, the solid angle $d\Omega$ is equal to $(K/qq')dK\,d\varphi_K$. The integration of $dS/d\Omega$ over $\varphi_{q'}=\varphi_K$ is now easy to perform since the form factors $\langle n',l',m';\text{III}|\exp(-i\mathbf{K}\cdot\mathbf{r})|n,l,m;\text{III}\rangle$ are by construction independent of the **K** orientation [see Eq. (16)]. These form factors are clearly diagonal with respect to the magnetic quantum numbers. Using the relations

$$d_{m',m}^j(-\gamma) = [d_{m',m}^j(-\gamma)]^* = d_{m,m'}^j(\gamma) \tag{20}$$

one obtains

$$dS/dK = 2\pi K[(M_1/M_{12})L/q]^2\sum_{t=-l-l'}^{l+l'}\left|\sum_{m_1,m_2}d_{m_1,m_2}^l(\theta_K)d_{m_1-t,m_2}^{l'}(\theta_K)d_{m_1,m}^l(\alpha)d_{m_1-t,m'}^{l'}(\alpha)\right.$$

$$\times\langle n',l',m_2;\text{III}|\exp(-i\mathbf{K}\cdot\mathbf{r})|n,l,m_2;\text{III}\rangle\Bigg|^2 \tag{21}$$

with $\cos\theta_K$ given by the right-hand side of Eq. (15b). Using the relation[15]

$$d_{m',m}^j(\gamma) = (-1)^{m-m'}d_{-m',-m}^j(\gamma) \ , \tag{22}$$

the sum over m_1 in Eq. (21), to be denoted by W, can be rewritten as

$$W=(-1)^{m_2+m'}\sum_{m_1}d^l_{m_1,m_2}(\theta_K)d^{l'}_{t-m_1,-m_2}(\theta_K)d^l_{m_1,m}(\alpha)d^{l'}_{t-m_1,-m'}(\alpha) \ . \tag{23}$$

Using the reduction of the product of two rotation matrix elements in a sum of rotation matrix elements multiplied by Clebsch-Gordan coefficients.[15]

$$d^{j_1}_{v'_1,v_1}(\gamma)d^{j_2}_{v'_2,v_2}(\gamma)=\sum_{J}\langle j_1,v'_1,j_2,v'_2\,|\,j_1,j_2,J,M'\rangle\langle j_1,j_2,J,M\,|\,j_1,v_1,j_2,v_2\rangle d^J_{M',M}(\gamma) \ , \tag{24}$$

one obtains

$$W=(-1)^{m_2+m'}\sum_{J}\langle l,l',J,0\,|\,l,m_2,l',-m_2\rangle\langle l,l',J,m-m'\,|\,l,m,l',-m'\rangle d^J_{t,0}(\theta_K)d^J_{t,m-m'}(\alpha) \ . \tag{25}$$

Finally, using the orthogonality relation,[15]

$$\int_0^\pi d\alpha\,\sin(\alpha)d^J_{t,m-m'}(\alpha)d^{J'}_{t,m-m'}(\alpha)=\delta_{JJ'}[2/(2J+1)] \ . \tag{26}$$

The differential cross section dS/dK averaged over all solid angles, denoted $\langle dS/dK\rangle$, can be thus obtained:

$$\langle dS/dK\rangle=\tfrac{1}{2}\int_0^\pi d\alpha\,\sin(\alpha)\,dS/dK \tag{27a}$$

$$=2\pi K[(M_1/M_{12})L/q]^2$$

$$\times\sum_{t=-l-l'}^{l+l'}\sum_{J=\max\{|t|,|m-m'|\}}^{l+l'}\{[d^J_{t,0}(\theta_K)]^2/(2J+1)\}$$

$$\times\left|\sum_{m_2}(-1)^{m_2}\langle l,l',J,0\,|\,l,m_2,l',-m_2\rangle\langle l,l',J,m-m'\,|\,l,m,l',-m'\rangle\right.$$

$$\times\left.\langle n',l',m_2;\text{III}\,|\,\exp(-i\mathbf{K}\cdot\mathbf{r})\,|\,n,l,m_2;\text{III}\rangle\right|^2 \ . \tag{27b}$$

The integral of $\langle dS/dK\rangle$ over K corresponds to the cross section pertaining to a cell-type experiment.

Equations (21) and (27b) are the starting points of the numerical applications to be discussed in Sec. IV. These numerical computations are made by expressing the hydrogenic form factors in Eqs. (21) and (27b) in terms of rotation matrix elements.

III. THE HYDROGENIC FORM FACTORS IN TERMS OF ROTATION MATRIX ELEMENTS

In this section the notation $\langle n',l',m;\text{III}\,|\,\exp(-i\mathbf{K}\cdot\mathbf{r})\,|\,n,l,m;\text{III}\rangle$ will be replaced by the standard one, $\langle n',l',m\,|\,\exp(-iKz)\,|\,n,l,m\rangle$. An analytic expression of the above form factor has been derived on the basis of the noninvariance algebra $o(4,2)$ of the hydrogen atom.[16,17] The method involves first a unitary change of basis vectors by means of Clebsch-Gordan coefficients:

$$|\,n,l,m\,\rangle=\sum_{f,g}|\,n,f,g\,\rangle\langle F,f,F,g\,|\,F,F,l,m=f+g\rangle \tag{28a}$$

with

$$F=(n-1)/2 \ . \tag{28b}$$

The wave functions $\langle\mathbf{r}\,|\,n,f,g\rangle$ are, except for phase factors, the usual hydrogenic wave functions separable in parabolic coordinates.[17] The usual parabolic quantum numbers n_1,n_2 are related to f and g by[17]

$$n_1=F-g \quad\text{if } m\geq 0 \tag{29a}$$

$$n_2=F-f \quad\text{if } m\geq 0 \tag{29b}$$

$$n_1=F+f \quad\text{if } m\leq 0 \tag{29c}$$

$$n_2=F+g \quad\text{if } m\leq 0 \tag{29d}$$

Within Englefield phase convention, one has the symmetry relation

$$\langle n',-f',-g'\,|\,\exp(-iKz)\,|\,n,-f,-g\rangle$$

$$=(-1)^{n-n'}\langle n',f',g'\,|\,\exp(iKz)\,|\,n,f,g\rangle \ . \tag{30}$$

From now on, all the formulas of this section [Eqs. (31)—(38)] refer to the case $m\geq 0$, the case $m\leq 0$ resulting from Eq. (30). The result quoted from Englefield[17] is

$\langle n',f',g' \mid \exp(iKz) \mid n,f,g \rangle$

$$= \frac{1}{2} K \left[\frac{(n'_1+m)!(n'_2+m)!(n_1+m)!(n_2+m)!}{n'_1!n'_2!n_1!n_2!} \right]^{1/2} \left[\frac{4nn'}{(n+n')^2+K^2n^2n'^2} \right]^{m+2}$$

$$\times \frac{(-1)^{n'_1+n_2}(n-n'-iKnn')^{n_1+n'_2-1}(n-n'+iKnn')^{n'_1+n_2-1}}{(m!)^2(n+n'-iKnn')^{n_1+n'_1}(n+n'+iKnn')^{n_2+n'_2}}$$

$$\times \{ [2Kn^2n'^2+2inn'(n_2-n_1)+i(n^2+n'^2+K^2n^2n'^2)(n'_1-n'_2)]\Phi_m(n_1,n'_1)\Phi_m(n_2,n'_2)+i[(n+n')^2+K^2n^2n'^2]$$

$$\times [n'_2\Phi_m(n_1,n'_1)\Phi_m(n_2,n'_2-1)-n'_1\Phi_m(n_1,n'_1-1)\Phi_m(n_2,n'_2)]\} \ . \tag{31}$$

The function $\phi_m(b,c)$ is defined in terms of the hypergeometric function $_2F_1$ by

$$\phi_m(b,c)=_2F_1(-b,-c;m+1;y) \tag{32a}$$

with

$$y=-4nn'/[(n-n')^2+(Knn')^2] \ . \tag{32b}$$

Using the relation[18]

$$_2F_1(b,c;d;y)=(1-y)^{-b} {}_2F_1(b,d-c;d;y/(y-1)) \tag{33}$$

one obtains

$$\phi_m(b,c)=[\sin^2(\omega/2)]^{-b} {}_2F_1(-b,m+1+c;m+1;(1+\cos\omega)/2) \tag{34}$$

with

$$\omega=\arccos[(y+1)/(y-1)] \ . \tag{35}$$

Using the relation[15] between a rotation matrix element and the hypergeometric function involved in Eq. (34), one obtains

$$\phi_m(b,c)=\frac{m![b!c!/((b+m)!(c+m)!)]^{1/2}}{\{(-1)^c[\cos(\omega/2)]^m[\sin(\omega/2)]^{b+c}\}} d^{(b+c+m)/2}_{(c-b+m)/2,(b-c+m)/2}(\omega) \ . \tag{36}$$

Finally, using the well-known recursion relations[19] between the rotation matrix elements $d^j_{m',m}(\omega)$, one obtains after some calculation

$\langle n',f',m-f' \mid \exp(iKz) \mid n,f,m-f \rangle$

$$= \{(-1)^{2\mu_2-m}K[\cos(\omega/2)]^6/[2\sin(\omega/2)]^2\}\exp[2i(\beta(\mu_2-\mu_1)+\rho(j_1-j_2))]$$

$$\times (d^{j_1}_{m-\mu_1,\mu_1}(\omega)d^{j_2}_{m-\mu_2,\mu_2}(\omega)[Knn'+i2(\mu_1-\mu_2)]$$

$$+2i\tan(\omega/2)\{ d^{j_1}_{m-\mu_1,\mu_1}(\omega)[(j_2-m+\mu_2)(j_2+m-\mu_2+1)]^{1/2}d^{j_2}_{m-\mu_2+1,\mu_2}(\omega)$$

$$-d^{j_2}_{m-\mu_2,\mu_2}(\omega)[(j_1-m+\mu_1)(j_1+m-\mu_1+1)]^{1/2}d^{j_1}_{m-\mu_1+1,\mu_1}(\omega)\}) \tag{37}$$

with

$$\exp(2i\beta)=(n-n'+iKnn')/(n-n'-iKnn') \ , \tag{38a}$$

$$\exp(2i\rho)=(n+n'+iKnn')/(n+n'-iKnn') \ , \tag{38b}$$

$$j_1=(n_1+n'_1+m)/2 \ , \tag{38c}$$

$$j_2=(n_2+n'_2+m)/2 \ , \tag{38d}$$

$$\mu'_1=(n'_1-n_1+m)/2 \ , \tag{38e}$$

$$\mu_1=(n_1-n'_1+m)/2 \ , \tag{38f}$$

$$\mu'_2=(n'_2-n_2+m)/2 \ , \tag{38g}$$

$$\mu_2=(n_2-n'_2+m)/2 \ . \tag{38h}$$

Equation (37) has some advantages compared to Eq. (31). The nontrivial part of Eq. (37) indeed involves only rotation matrix elements. These elements have a direct physical interpretation, and satisfy many special properties that are useful for numerical computations.

IV. NUMERICAL RESULTS

Three situations are considered for process (3): $\alpha=0$ (q parallel to the axis z corresponding to the eigenstate of atomic angular momentum); $\alpha=\pi/2$ (q perpendicular to the axis z); and, finally, an average over α corresponding to the situation of a cell experiment. The calculations are made for the same value of the relative velocity between helium and sodium as used in Ref. 3, the thermal averaged velocity for a temperature of 296 K. The value used for the scattering length L for the He-electron collision is 1.19. The results for $l'=19,18$ are shown in Table I and

TABLE I. Cross sections S, in atomic units, for the processes Na($n=20$, $l=19$, $m=19$)+He→Na($n'=20$, l', m')+He. Angle α is either equal to zero (columns 1 and 4) or $\pi/2$ (columns 2 and 5). Results obtained when averaging over all orientations [$\langle S \rangle$, see Eq. (27)] are shown in columns 3 and 6. The number enclosed in parentheses signifies powers of 10 by which the prefactor is multiplied.

m'	$l'=19$ $\alpha=0$	$\alpha=\pi/2$	$\langle S \rangle$	$l'=18$ $\alpha=0$	$\alpha=\pi/2$	$\langle S \rangle$
19	0.263(+3)	0.689(+3)	0.406(+3)			
18	0.966(−3)	0.181(+3)	0.100(+3)	0.135(+3)	0.890(+2)	0.107(+3)
17	0.196(+1)	0.994(+2)	0.582(+2)	0.235(−2)	0.394(−2)	0.379(+2)
16	0.120(−3)	0.663(+2)	0.410(+2)	0.301(+1)	0.272(+2)	0.246(+2)
15	0.751(−1)	0.487(+2)	0.319(+2)	0.418(−3)	0.207(+2)	0.182(+2)
14	0.197(−4)	0.379(+2)	0.262(+2)	0.196	0.166(+2)	0.146(+2)
13	0.607(−2)	0.307(+2)	0.224(+2)	0.909(−4)	0.138(+2)	0.122(+2)
12	0.444(−5)	0.257(+2)	0.196(+2)	0.228(−1)	0.117(+2)	0.106(+2)
11	0.826(−3)	0.219(+2)	0.175(+2)	0.257(−4)	0.102(+2)	0.936(+1)
10	0.135(−5)	0.191(+2)	0.159(+2)	0.410(−2)	0.899(+1)	0.844(+1)
−10	0.243(−4)	0.651(+1)	0.894(+1)	0.412(−2)	0.339(+1)	0.474(+1)
−11	0.834(−3)	0.656(+1)	0.915(+1)	0.571(−3)	0.344(+1)	0.488(+1)
−12	0.164(−3)	0.667(+1)	0.945(+1)	0.228(−1)	0.353(+1)	0.509(+1)
−13	0.612(−2)	0.686(+1)	0.986(+1)	0.412(−2)	0.368(+1)	0.537(+1)
−14	0.170(−2)	0.715(+1)	0.104(+2)	0.196	0.390(+1)	0.578(+1)
−15	0.753(−1)	0.760(+1)	0.112(+2)	0.442(−1)	0.427(+1)	0.640(+1)
−16	0.309(−1)	0.832(+1)	0.125(+2)	0.298(+1)	0.490(+1)	0.745(+1)
−17	0.193(+1)	0.957(+1)	0.145(+2)	0.751	0.613(+1)	0.944(+1)
−18	0.163(+1)	0.121(+2)	0.187(+2)	0.134(+3)	0.124(+2)	0.194(+2)
−19	0.261(+3)	0.239(+2)	0.374(+2)			

in Fig. 2. The results for $l'=9,8$ are shown in Table II and in Fig. 3. Finally, Table III shows the results for $l'=3,2$. The cases $l'=1,0$ corresponding to large quantum defects require nonhydrogenic form factors and are not considered here.

A. The case $\alpha=0$

This case was considered with $l'=19,18$ in Ref. 3. The slight differences between the results of Yosizawa and Matsuzawa[3] and our results originate in the introduction

TABLE II. Cross sections S, in atomic units, for the processes Na($n=20$, $l=19$, $m=19$)+He→Na($n'=20$, l', m')+He. Angle α is either equal to zero (columns 1 and 4) or $\pi/2$ (columns 2 and 5). Results obtained when averaging over all orientations [$\langle S \rangle$, see Eq. (27)] are shown in columns 3 and 6.

m'	$l'=9$ $\alpha=0$	$\alpha=\pi/2$	$\langle S \rangle$	$l'=8$ $\alpha=0$	$\alpha=\pi/2$	$\langle S \rangle$
9	0.443(+2)	0.795(+1)	0.122(+2)			
8	0.166(−1)	0.250(+1)	0.366(+1)	0.415(+2)	0.708(+1)	0.109(+2)
7	0.949(+1)	0.271(+1)	0.403(+1)	0.171(−1)	0.205(+1)	0.303(+1)
6	0.137(−1)	0.198(+1)	0.288(+1)	0.102(+2)	0.246(+1)	0.370(+1)
5	0.388(+1)	0.189(+1)	0.276(+1)	0.156(−1)	0.170(+1)	0.250(+1)
4	0.115(−1)	0.164(+1)	0.237(+1)	0.474(+1)	0.172(+1)	0.254(+1)
3	0.223(+1)	0.156(+1)	0.227(+1)	0.146(−1)	0.145(+1)	0.212(+1)
2	0.112(−1)	0.144(+1)	0.208(+1)	0.310(+1)	0.144(+1)	0.212(+1)
1	0.171(+1)	0.140(+1)	0.203(+1)	0.154(−1)	0.130(+1)	0.191(+1)
0	0.127(−1)	0.133(+1)	0.193(+1)	0.270(+1)	0.131(+1)	0.194(+1)
−1	0.171(+1)	0.131(+1)	0.193(+1)	0.191(−1)	0.122(+1)	0.181(+1)
−2	0.171(−1)	0.128(+1)	0.188(+1)	0.310(+1)	0.128(+1)	0.191(+1)
−3	0.223(+1)	0.131(+1)	0.194(+1)	0.276(−1)	0.121(+1)	0.181(+1)
−4	0.273(−1)	0.129(+1)	0.192(+1)	0.473(+1)	0.136(+1)	0.206(+1)
−5	0.387(+1)	0.140(+1)	0.211(+1)	0.454(−1)	0.127(+1)	0.192(+1)
−6	0.502(−1)	0.138(+1)	0.208(+1)	0.102(+2)	0.174(+1)	0.268(+1)
−7	0.947(+1)	0.180(+1)	0.275(+1)	0.770(−1)	0.135(+1)	0.207(+1)
−8	0.958(−1)	0.154(+1)	0.235(+1)	0.414(+2)	0.448(+1)	0.698(+1)
−9	0.442(+2)	0.470(+1)	0.732(+1)			

S (a.u.)

4×10²

10²

4×10¹

10¹
8
6
4

2

10⁰
20   0   -20   m'
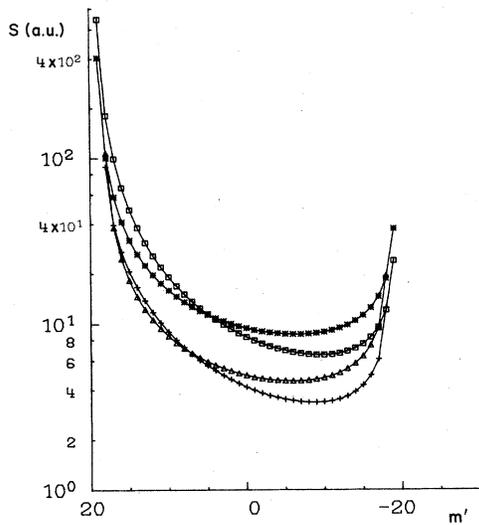
FIG. 2. Semilogarithmic plot of the cross sections S corresponding to the processes $Na(n=20, l=19, m=19)+He$ $\rightarrow Na(n'=20, l', m')+He$. Atomic unit: a_0^2. Abscissa: m'. $\square$: $l'=19$, $\alpha=\pi/2$. $*$: $l'=19$, average over α [see Eq. (27a)]. $+$: $l'=18$, $\alpha=\pi/2$. $\triangle$: $l'=18$, average over α [see Eq. (27a)].

by these authors of an additional term in Eq. (5) involving the He polarizability. When looking at the present results, the most striking feature is the appearance of a selection rule which nearly forbids transitions when $l'-m'$ is odd. This rule is most easily explained by considering Eqs. (15). The angle θ is nearly equal to $\pi/2$ for the K values which contribute significantly to the integral of dS/dK over K. $Y_\lambda^{m-m'}$ is then different from zero only for $\lambda-m+m'$ even. In this case the sum $l+l'+\lambda$ has the parity of $l'-m'$. The 3-j symbol $\begin{pmatrix} l & l' & \lambda \\ 0 & 0 & 0 \end{pmatrix}$ of Eq. (15a) is therefore zero if $l'-m'$ is odd.

TABLE III. Cross sections S, in atomic units, for the processes $Na(n=20, l=19, m=19)+He\rightarrow Na(n'=20, l', m')$ $+He$. Angle α is either equal to zero (column 1) or $\pi/2$ (column 2). Results obtained when averaging over all orientations [$\langle S\rangle$, see Eq. (27)] are shown in column 3.

l'	m'	$\alpha=0$	$\alpha=\pi/2$	$\langle S\rangle$
3	3	0.275(+02)	0.387(+01)	0.599(+01)
3	2	0.105(−01)	0.515	0.775
3	1	0.134(+02)	0.186(+01)	0.287(+01)
3	0	0.139(−01)	0.530	0.802
3	−1	0.134(+02)	0.176(+01)	0.273(+01)
3	−2	0.158(−01)	0.460	0.698
3	−3	0.275(+02)	0.330(+01)	0.512(+01)
2	2	0.239(+02)	0.326(+01)	0.505(+01)
2	1	0.714(−02)	0.303	0.457
2	0	0.144(+02)	0.188(+01)	0.291(+01)
2	−1	0.874(−02)	0.286	0.434
2	−2	0.239(+02)	0.293(+01)	0.455(+01)

Another striking fact is the quasiequality of the cross section S for processes differing only by the sign of m', when $l'-m'$ is even. It should be emphasized that this concerns only the cross section S and not the differential cross section dS/dK. This is illustrated in Fig. 4 where it is seen that dS/dK for $m'=-19$ takes significant values for larger values of K than does dS/dK for $m'=19$. We do not presently have a simple explanation why the area of the two curves of Fig. 4 should be nearly equal. The fact that the dotted curve $(m'=-19)$ is centered on larger values of K than the solid line curve $(m'=19)$ sup-

S (a.u.)
12

8

4

0
10   0   -10   m'

FIG. 3. As in Fig. 2 but linear plot. $\square$: $l'=9$, $\alpha=\pi/2$. $*$: $l'=9$, average over α [see Eq. (27a)]. $+$: $l'=8$, $\alpha=\pi/2$. $\triangle$: $l'=8$, average over α [see Eq. (27a)].

40000

30000

dS/dK (a.u.)

20000

10000

0
0.00   0.05   0.10   0.15
K (a.u.)
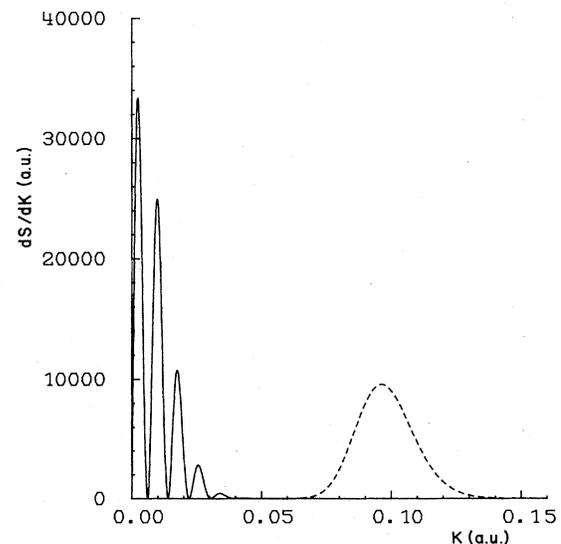
FIG. 4. Differential cross sections dS/dK (a.u.), as a function of K (a.u.), for the processes $Na(n=20, l=19, m=19)$ $+He\rightarrow Na(n=20, l'=19, m')+He$ with $\alpha=0$. Solid line curve, $m'=19$; dotted line curve, $m'=-19$.

ports a classical interpretation: reversing the direction of motion of an electron on a classical circular trajectory requires large momentum transfers.

B. The case $\alpha = \pi/2$ and the average over α

It is seen in Figs. 2 and 3 and Table III that the cross section S averaged over α are generally not too different from those corresponding to $\alpha = \pi/2$. The reason for that is a geometrical one: for a given value of $d\alpha$, Eq. (27a), the solid angle encompassed is the greatest for $\alpha = \pi/2$, as indicated by the factor $\sin(\alpha)$ in Eq. (27a). A systematic study of the cross sections S as a function of α would be time consuming. When calculating dS/dK as a function of α, for some fixed values of K, not other general selection rules were found. As an illustration, Fig. 5 shows the α dependence for two particular cases: dS/dK corresponding to the transition towards the states $|20,18,18\rangle$ and $|20,18,17\rangle$ for $K = 0.02$.

The cross sections S are found to be maximum when $|m'| = l'$ as for the case $\alpha = 0$. The minimum of S as a function of m' decreases from about $9a_0^2 - 10a_0^2$ to about a fraction of a_0^2 as l' varies from 19 to 3. For large l' values the behavior of S as a function of m' is smooth, whereas it presents structure, with unit period, as for the case $\alpha = 0$, for low l' values.

V. INFLUENCE OF AN EXTERNAL ELECTRIC FIELD

The previous formulation of state-to-state collisions can be extended in the presence of an external electric field $\mathbf{E}$. The direction of $\mathbf{E}$ is assumed to be parallel to L_z [see Eqs. (2)]. Then, it is known that the states $|n,f,g\rangle$ [see Eq. (28a)],
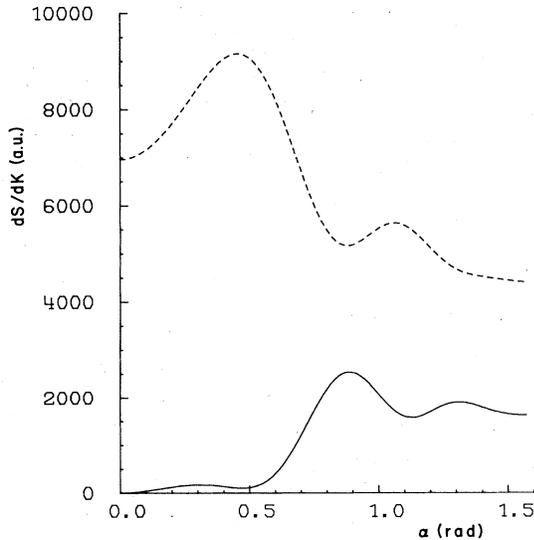
FIG. 5. Differential cross sections dS/dK (a.u.), as a function of the angle α (rad), for $K = 0.02$ (a.u.), and for the processes $\mathrm{Na}(n = 20, l = 19, m = 19) + \mathrm{He} \rightarrow \mathrm{Na}(n' = 20, l' = 18, m')$ $+$ He. Solid line curve, $m' = 17$; dotted line curve, $m' = 18$.

$$|n,f,g\rangle = \sum_l |n,l,m = f + g\rangle\langle F,F,l,m | F,f,F,g\rangle \ , \quad (39)$$

are the correct eigenstates of the hydrogenic Hamiltonian in the limit of zero external field. The differential cross section $dS/d\Omega$ can therefore be expressed as in Eq. (5), the states $|n,f,g\rangle$ in place of the states $|n,l,m\rangle$. This is the starting point used by Hickman.[20] The method of Sec. II C for obtaining the differential cross sections dS/dK for an arbitrary value of α as well as for an average value of dS/dK over the angle α can be used after the change of basis given by Eq. (39) has been made. Hickman was able to obtain dS/dK from $dS/d\Omega$ for the case $\alpha = \pi/2$ by using another procedure; his procedure involves a further approximation[21] well justified when the energy gap $|\epsilon' - \epsilon|$ is not too large; it is not generalizable to arbitrary values of α.

The difficulty when using the expansion of $|n,f,g\rangle$ in terms of the $|n,l,m\rangle$, Eq. (39), is a serious increase of the computation time. It is therefore of interest to make a general study of the symmetry properties Sec. V A. In Sec. V B we shall concentrate on the results which can presently be obtained without any numerical computation in the case where the initial state is circular.

A. Symmetry properties

The states $|n,f,g\rangle$ are eigenvectors of the operators F^2, G^2, F_z, G_z with eigenvalues $F(F+1), F(F+1), f, g$ if F and G are defined by

$$\mathbf{F} = (\mathbf{L} - \mathbf{A}/\sqrt{-2H})/2 \ , \quad (40a)$$

$$\mathbf{G} = (\mathbf{L} + \mathbf{A}/\sqrt{-2H})/2 \ , \quad (40b)$$

with H the hydrogenic Hamiltonian, and $\mathbf{A}$ and Runge-Lenz[17] operator:

$$\mathbf{A} = (\mathbf{p} \wedge \mathbf{L} - \mathbf{L} \wedge \mathbf{p})/2 - \hat{r} \ . \quad (41)$$

It is clear that under space inversion $\mathbf{F}$ and $\mathbf{G}$ are interchanged, and that under time reversal $\mathbf{F}$ transforms into $-\mathbf{G}$, $\mathbf{G}$ into $-\mathbf{F}$. This allows us to conclude (see the Appendix) that

$$|\langle n',f',g' | \exp(-i\mathbf{K}\cdot\mathbf{r}) | n,f,g\rangle|$$
$$= |\langle n',g',f' | \exp(i\mathbf{K}\cdot\mathbf{r}) | n,g,f\rangle| \ , \quad (42)$$

$$|\langle n',f',g' | \exp(-i\mathbf{K}\cdot\mathbf{r}) | n,f,g\rangle|$$
$$= |\langle n',-g',-f' | \exp(i\mathbf{K}\cdot\mathbf{r}) | n,-g,-f\rangle| \ . \quad (43)$$

A further relation is obtained by considering rotational symmetry (see the Appendix):

$$|\langle n',f',g' | \exp(-i\mathbf{K}\cdot\mathbf{r}) | n,f,g\rangle|$$
$$= |\langle n',-f',-g' | \exp(i\mathbf{K}\cdot\mathbf{r}) | n,-f,-g\rangle| \ . \quad (44)$$

The differential cross section $dS/d\Omega$ is therefore invariant under the three following independent transformations:

(a) $\mathbf{q} \rightarrow -\mathbf{q}, \ \mathbf{q}' \rightarrow -\mathbf{q}', \ f' \leftrightarrow g', \ f \leftrightarrow g$.

(b) $\mathbf{q} \rightarrow -\mathbf{q}, \ \mathbf{q}' \rightarrow -\mathbf{q}', \ f' \leftrightarrow -g', \ f \leftrightarrow -g$.

(c) $\mathbf{q} \rightarrow -\mathbf{q}, \ \mathbf{q}' \rightarrow -\mathbf{q}', \ (f',g',f,g) \rightarrow (-f',-g',-f,-g)$.

These independent transformations can of course all be combined together. For example, the product of transformations (a) and (b) indicates that the differential cross section $dS/d\Omega$ is invariant if $\mathbf{q}$ and $\mathbf{q}'$ are unchanged whereas f',g',f,g all undergo a change of sign. These results for $dS/d\Omega$ of course also concern dS/dK. In particular dS/dK is invariant under the transformation which changes the angle α into $\pi-\alpha$.

These results are valid for an arbitrary initial state $|n,f,g\rangle$ and final state $|n',f',g'\rangle$. In Sec. V B some results obtained without an electric field for a circular initial Rydberg state are compared to the case where an external electric field is present.

B. Comparison with the zero-field case

The circular state $|n,l=n-1,m=n-1\rangle$ is identical to the state $|n,f=F,g=F\rangle$ [see Eq. (39)]. By expressing the projection operator onto the subspace characterized by fixed values of n' and m' in two different ways,

$$\sum_{\substack{f',g' \\ (f'+g'=m')}} |n',f',g'\rangle\langle n',f',g'| = \sum_{l'} |n',l',m'\rangle\langle n',l',m'|, \tag{45}$$

one thus obtains [see Eqs. (5), (9), and (45)]

$$\sum_{\substack{f',g' \\ (f'+g'=m')}} S(|n,f=F,g=F\rangle \rightarrow |n,f',g'\rangle)$$

$$\leq \sum_{l'} S(|n,l=n-1,m=n-1\rangle \rightarrow |n,l',m'\rangle) . \tag{46}$$

The inequality in relation (46) has its origin in an increase of the lower bound $|q'-q|$ [see Eq. (9)], which arises as the field $\mathbf{E}$ removes the degeneracy. The variation of the upper bound $q+q'$ does not matter since dS/dK is completely negligible for a value of K of the order of $q+q'$. The equality in Eq. (46) occurs for the zero-field limit.

Another relation can also be obtained in the case when $\alpha=0$. Due to the selection rule for the states $|n,l,m\rangle$, only the term corresponding to $l'=n-2$ has to be taken into account in the expression of $|n',f'=F-1,g'=F\rangle$ in terms of the states $|n',l',m'=2F-1\rangle$ [see Eq. (39)]. For $\alpha=0$, one then has

$$S(|n,f=F,g=F\rangle \rightarrow |n,f'=F-1,g'=F\rangle)/[S(|n,l=n-1,m=n-1\rangle \rightarrow |n,l'=n-2,m'=n-2\rangle)]$$

$$\lesssim |\langle F,F,l',m'|F,f',F,g'\rangle|^2 = \tfrac{1}{2} . \tag{47}$$

The inequality in relation (47) is transformed into a quasiequality in the zero-field limit.

Getting more detailed results requires numerical computation, and we are looking for procedures that could decrease the calculation time.

VI. CONCLUDING REMARKS

It has been shown that the impulse approximation, in its simplest form [Eq. (5)], predicts a highly anisotropic behavior of collisional processes involving a circular initial Rydberg state. A selection rule may appear, depending on the initial orientation. An experimental study of these orientation effects would be a crucial test of the validity of the impulse approximation for treating thermal collisions between Rydberg atoms and ground-state rare-gas atoms. Confrontations of the theory with experiment already exist when considering the so called l-mixing cross sections. These confrontations, as they concern summed and averaged cross sections, do not test the orientation effects. The experimental study of orientation effects of course represents a difficult experimental challenge, but, in view of the progress in crossed-beam techniques, this could be possible in the near future, especially by using the polarization properties of both the excitation and the detection.

ACKNOWLEDGMENTS

APPENDIX: SYMMETRIES OF THE MODULUS OF HYDROGENIC FORM FACTORS

The hydrogenic form factors A and B are defined, within the notations used in this work, by

$$A = \langle n',l',m'|\exp(-i\mathbf{K}\cdot\mathbf{r})|n,l,m\rangle , \tag{A1}$$

$$B = \langle n',f',g'|\exp(-i\mathbf{K}\cdot\mathbf{r})|n,f,g\rangle . \tag{A2}$$

Three symmetry operations concerning the isolated hydrogen atom (isolated means that $\mathbf{K}$ is not affected by these operations) will now be considered: reflection, rotation, and time reversal. From now on, the same letter s will denote different, unspecified, phase factors. The parity operator P has the following actions:

$$P|n,l,m\rangle = s|n,l,m\rangle , \tag{A3}$$

$$P|n,f,g\rangle = s|n,g,f\rangle , \tag{A4}$$

$$P\exp(i\mathbf{K}\cdot\mathbf{r})P = \exp(-i\mathbf{K}\cdot\mathbf{r}) . \tag{A5}$$

It follows that

$$|A| = |\langle n',l',m'|\exp(i\mathbf{K}\cdot\mathbf{r})|n,l,m\rangle| , \tag{A6}$$

$$|B| = |\langle n',g',f'|\exp(i\mathbf{K}\cdot\mathbf{r})|n,g,f\rangle| . \tag{A7}$$

A rotation along the z axis [see Eq. (2)] changes the states $|n,l,m\rangle, |n,f,g\rangle$ only by a phase factor. It is then possible to choose the x axis in the plane $(z,\mathbf{K})$, so that

$$\mathbf{K}\cdot\mathbf{r} = K_x x + K_z z . \tag{A8}$$

A rotation by π along the y axis then changes the sign of $\mathbf{K}\cdot\mathbf{r}$:

$$\exp(i\pi L_y)\exp(-i\mathbf{K}\cdot\mathbf{r})\exp(-i\pi L_y)=\exp(i\mathbf{K}\cdot\mathbf{r}) , \tag{A9}$$

whereas the hydrogenic states are transformed according to

$$\exp(-i\pi L_y)\,|\,n,l,m\,\rangle=s\,|\,n,l,-m\,\rangle , \tag{A10}$$

$$\exp(-i\pi L_y)\,|\,n,f,g\,\rangle=s\,|\,n,-f,-g\,\rangle . \tag{A11}$$

It follows that

$$|\,A\,|=|\,\langle n',l',-m'\,|\exp(i\mathbf{K}\cdot\mathbf{r})\,|\,n,l,-m\,\rangle\,| , \tag{A12}$$

$$|\,B\,|=|\,\langle n',-f',-g'\,|\exp(i\mathbf{K}\cdot\mathbf{r})\,|\,n,-f,-g\,\rangle\,| . \tag{A13}$$

The antiunitarity property of the time-reversal operator T gives

$$\langle i'\,|\,T^\dagger T\,|\,i\,\rangle=\langle i\,|\,i'\,\rangle , \tag{A14}$$

$$T\exp(-i\mathbf{K}\cdot\mathbf{r})=\exp(i\mathbf{K}\cdot\mathbf{r})T . \tag{A15}$$

T changes the hydrogenic states according to

$$T\,|\,n,l,m\,\rangle=s\,|\,n,l,-m\,\rangle , \tag{A16}$$

$$T\,|\,n,f,g\,\rangle=s\,|\,n,-g,-f\,\rangle . \tag{A17}$$

It follows that

$$[\langle i'\exp(-i\mathbf{K}\cdot\mathbf{r})\,|\,i\,\rangle]^*=\langle i'\,|\,T^\dagger\exp(i\mathbf{K}\cdot\mathbf{r})T\,|\,i\,\rangle \tag{A18}$$

and therefore

$$|\,A\,|=|\,\langle n',l',-m'\,|\exp(i\mathbf{K}\cdot\mathbf{r})\,|\,n,l,-m\,\rangle\,| , \tag{A19}$$

$$|\,B\,|=|\,\langle n',-g',-f'\,|\exp(i\mathbf{K}\cdot\mathbf{r})\,|\,n,-g,-f\,\rangle\,| . \tag{A20}$$

It should be noted that Eq. (A19) coincides with Eq. (A12), whereas Eq. (A20) differs from Eq. (A13). Equations (A6) and (A12) are not specific to the Coulomb problem, but are valid for any central potential.

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