Angular-momentum transfer in collisional ionization

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The double-differential cross sections for ionization in $e^-+\mathrm{H}(nl)$ collisions are reported as a function of the impact energy E of the projectile, final energy E_f, and angular momentum L_f of the ejected electron. This process is assumed to occur via an energy-changing and angular-momentum-changing binary collision between the Rydberg electron in a prepared state (nl) and the projectile electron e^- or H(1s). The atomic projectile can also be excited during this process. Systematic trends in the variation of the classical ionization cross sections with final angular momentum L_f of the ejected electron are discussed and are in accord with a previous quantal treatment, whereby the nondipole $(\Delta l>1)$ transitions are much more important in the low- and intermediate-energy range of relative motion, and that the value of the final angular momentum of the ejected electron depends mainly on the initial value of the principal quantum number n of the Rydberg atom.

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I. INTRODUCTION

Ionization of atoms and molecules is one of the most basic processes in physics, with fundamental applications in such different areas as astrophysics, plasma physics, fusion physics, surface science, etc. The primary goal of this work is the development of a classical theory of doubly differential cross sections $d^2\sigma^I/dE_f dL_f$ as a function of L_f and E_f, the angular momentum and energy of the ejected electron. The target atom is treated as two particles, the valence electron (labeled 1), with mass m_1 which orbits an ionic core (labeled 2) with mass m_2. In the basic binary-encounter approximation to the $e^-+\mathrm{H}(nl)$ inelastic collision, energy, momentum, and angular-momentum changes originate mainly from a collision between the Rydberg electron in prepared state (nl) and the incident projectile (labeled 3) with mass m_3. There is extensive literature on the excitation and ionization of Rydberg states of atoms by electrons (see Seaton [1], Percival and Richards [2], Flannery and McCann [3,4], Ton-That, Manson, and Flannery [5], Kunc [6], and MacAdam, Rofles, and Crosby [7]). In addition, the problem of the angular-momentum mixing of degenerate states of hydrogenic Rydberg atoms with fixed principal quantum number n by collision with electrons and heavy particles has been thoroughly investigated, within a semiclassical context by Percival and Richards [8], Vriens and Smeets [9], and within the Born approximation by Matsuzawa [10], Beigman and co-workers [11–13], and Lebedev [14,15]. Although a great deal of attention (Brauner, Briggs, and Klar [16], Berakdar *et al.* [17], Byron and Joachain [18], Zhang, Whelam, and Walters [19], and Lahmam-Bennani [20]) has recently been focused on the triple-differential cross sections $d^3\sigma/d\Omega_s d\Omega_e dE_f$, measured in an $(e,2e)$ coincidence experiments, the variations of the cross section with the angular momentum L_f of the electron ejected from the target has not been directly addressed previously. In this investigation, a basic formula for $d^2\sigma^I/dE_f dL_f$ will be developed from the classical binary encounter approximation. It will then be of interest to see whether or not the systematic trends previously observed [3,4] in a quantal Born approximation of angular momentum change in $e^-+\mathrm{H}(nl)$ collision will be predicted by a classical analysis such as the present classical binary-encounter approximation.

II. PRESENT BINARY-ENCOUNTER APPROXIMATION

Up to the present, the binary-encounter approximation has been developed only to provide $d\sigma^I/dE_f$ as a function of the energy of the ejected electron E_f for various impact energies of the projectile. In this paper, the binary encounter approximation is extended to provide $d^2\sigma^I/dE_f dL_f^2$ as a function of the added variable L_f, the angular momentum of the ejected electron.

A. Energy and angular-momentum changes

1. *Energy change*

The internal translational energy change of the (1) and (2) system, i.e., the target atom, is given by [21,22],

$$\epsilon_{fi}\equiv\epsilon_f-\epsilon_i=\omega\beta\sin\psi\cos\phi-\alpha(1-\omega\cos\psi)\ , \tag{1}$$

where ϵ_i and ϵ_f are the kinetic energies of the Rydberg electron before and after the collision respectively, where ψ is the scattering angle, the angle of rotation of the (1)–(3) relative velocity vector $\mathbf{g}$, and where α and β are

$$\alpha\equiv m_{13}V_{13}g\cos\eta+m_{13}^2g^2/(m_1+m_2)\ , \tag{2}$$

$$\beta\equiv m_{13}V_{13}g\sin\eta\ . \tag{3}$$

In Eq. (1) above $\omega\equiv g'/g$, where g and g' are being the relative speed of (1)–(3) system before and after collision, m_{13} being the reduced mass of the (1)–(3) subsystem and V_{13} the (1)–(3) center of mass velocity, where the projectile is assumed to be an electron.

2. *Angular-momentum change*

Let $\mathbf{L}_i$ and $\mathbf{L}_f$ be the initial and final angular momenta of the orbiting electron. The change in angular momentum due to the (1)–(3) collision is written as

$$L=\mathbf{L}_f-\mathbf{L}_i=m_{13}\mathbf{R}\times(\mathbf{g}'-\mathbf{g}) , \tag{4}$$

where $\mathbf{g}'$ is

$$\mathbf{g}'=(g\cos\psi)\mathbf{n}_0+(g\sin\psi)\mathbf{n} \tag{5}$$

and the unit vectors $\mathbf{n}$ and $\mathbf{n}_0$ are defined by

$$\mathbf{n}_0=\frac{(\mathbf{v}_1-\mathbf{v}_3)}{g} , \tag{6}$$

$$\mathbf{n}=\frac{(\mathbf{v}_1\times\mathbf{v}_3)}{(V_{13}g)} . \tag{7}$$

$\mathbf{R}$ is the radius vector of the valence electron. Therefore,

$$\mathbf{L}=m_{13}g\mathbf{R}\times[(\cos\psi-1)\mathbf{n}_0+\sin\psi\mathbf{n}] , \tag{8}$$

and since the rate coefficient depends on $\mathbf{L}^2$, then the latter is explicitly written as

$$\mathbf{L}^2=m_{13}^2g^2\{(\cos\psi-1)^2\mathbf{A}^2+(\sin\psi)^2\mathbf{B}^2 +2\sin\psi(\cos\psi-1)[\mathbf{A}\cdot\mathbf{B}]\} . \tag{9}$$

The vectors $\mathbf{A}$ and $\mathbf{B}$ are

$$\mathbf{A}=[\mathbf{R}\times\mathbf{n}_0] , \tag{10}$$

$$\mathbf{B}=[\mathbf{R}\times\mathbf{n}] , \tag{11}$$

and V_{13} and g satisfy the following relations,

$$V_{13}^2=\left[\frac{m_1^2v_1^2+m_3^2v_2+2m_1m_3v_1v_3\cos\theta_{13}}{(m_1+m_3)^2}\right] , \tag{12}$$

$$g^2=v_1^2+v_3^2-2v_1v_3\cos\theta_{13} \tag{13}$$

(see Fig. 1).

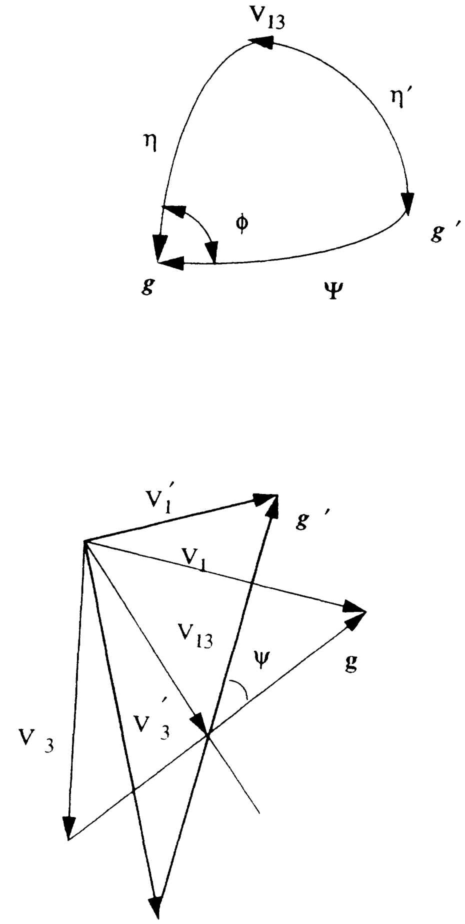

FIG. 1. The geometry of the collision.

B. Rate coefficient and cross section

The validity of the semiquantal treatment of electron-Rydberg atom collisions has been discussed extensively (Flannery [21,22], Vriens [23], Young and Märk [24], Percival and Richards [26], Burgess and Percival [27], McDowell and Coleman [28], Bates and Kingston [29], and Bates [30]), hence only a summary is required here. Therefore, in the present case the weakly bound electron in a highly excited state (nl), is assumed to follow a classical orbit. This assumption follows from the four points enumerated below.

As the principal quantum number n increases the electron becomes more and more localized in phase space, and its trajectory follows a classical orbit for which the quantal imprecision ΔR_n and ΔP_n are such that $\Delta R_n \ll R_n$ and $\Delta P_n \ll P_n$.

The reduced wavelength of the projectile $\hbar/p_3$ must be very small compared to R_n, so that the projectile interacts only with one particle at a time (binary collision), thereby minimizing the simultaneous overlap of the valence electron and the core of the target atom with the wave packet of the projectile. The (1)–(3) collision must be instantaneous, thus permitting the (1) and (2) potential energy to remain unaffected. The (1)–(3) collision time τ_{13} must be very much less than the typical time (τ_{orbit}) for the orbital electron to complete one orbit around the ionic core.

All contributions to the rate coefficient arising from (2) and (3) collisions are negligible with respect to those of the Rydberg electron–projectile collisions. This is mainly due to the inertiality of the ionic core. In the case where the projectile is a neutral atom or a molecule, the interaction between the atomic core and the projectile has a substantial contribution to the l-changing collision, especially at very low relative energies where the projectile and the core could form a bound state, and, therefore, a nonadiabatic energy exchange between the excited electron and the quasimolecule would result in an ionization of the Rydberg electron. The validity of this last criteria for atom–Rydberg atom collisions has been discussed previously, (Flannery [25,31], Matsuzawa [32,33], Hickman [34], Hahn [35], and Peach [36]). Therefore, the rate of collisions between the Rydberg electron in the state (nl) and the projectile, in the (1)–(3) center-of-mass frame, into the solid angle $d\Omega$ at a given speed v_3 of the projectile oriented with respect to particle 1 with speed

v_1 is given by

$$R_{if}d\Gamma_i d\Gamma_f = |\Psi_{nl}(\mathbf{R})|^2 d\mathbf{R} d\Gamma_i \times \left[g\sigma(g,\psi)d(\cos\psi)d\phi \frac{d(\cos\theta_{13})}{2} \right] \frac{d\Phi}{2\pi} , \tag{14}$$

where $\Gamma_f \equiv dE_f dL_f^2$. $\sigma(g,\psi)$ is the differential cross section in the (1)–(3) center-of-mass frame and for a Coulombic interaction it is written as

$$\sigma(g,\psi) = \frac{e^4}{4m_{13}^2[g\sin(\psi/2)]^4} , \tag{15}$$

and also in term of the momentum transfer P as

$$\sigma(P) = \frac{4m_{13}^2 e^4}{P^4} . \tag{16}$$

For nonhydrogenic targets, $\sigma(g,\psi)$ cannot in general be expressed in closed form analytically. However, in the nonhydrogenic case, $\sigma(g,\psi)$ can be obtained numerically (Berrington *et al.* [37], Bartschat and Burke [38]). Due to the Coulombic nature of the interaction, the quantal differential cross section coincides with that of the classical differential cross section. The $\frac{1}{2}$ factor in the square bracket accounts for the degeneracy of the rate coefficient where for $\theta_{13} = 0 - 2\pi$, $\cos\theta_{13}$ being an even function. $|\Psi_{nl}|^2 d\mathbf{R}$ is the probability associated with the orbital electron in state (nl), orbiting the ionic core within $d\mathbf{R}$ about $\mathbf{R}$. Since we are dealing with a highly excited state n, the WKB approximation to $\Psi_{nl}(\mathbf{R})$ would be appropriate. Therefore,

$$|\Psi_{nl}(\mathbf{R})|^2 d\mathbf{R} = \frac{2dR}{\tau_R v_R} , \tag{17}$$

where τ_R is the radial period for a Coulombic field and is written as

$$\tau_R(E_i, L_i^2) = 2\pi \left[\frac{e^2}{2}|E_i| \right]^{3/2} \left[\frac{m_1}{e^2} \right]^{1/2} , \tag{18}$$

and the radial speed v_R is

$$v_R = \left[\frac{2}{m_1} \left[E_i - V(R) - \frac{L_i^2}{2m_1 R^2} \right] \right]^{1/2} . \tag{19}$$

In the (1) and (2) center-of-mass frame, the number of particles 1, in initial state (E_i, L_i^2), scattered into the solid angle $d\Omega$, per incident flux of particle 3, with velocity $\mathbf{v}_3$ and relative initial and final speed g and g', respectively, is,

$$\sigma_{if}^c = R_{if} d\Gamma_i d\Gamma_f / v_3 , \tag{20}$$

which is given by

$$\frac{R_{if}}{v_3} d\Gamma_i d\Gamma_f = |\Psi_{nl}|^2 d\mathbf{R}\, d\Gamma_i \times \left[\frac{g}{v_3}\sigma(g,\psi)d(\cos\psi)d\phi \frac{d(\cos\theta_{13})}{2} \right] \frac{d\Phi}{2\pi} . \tag{21}$$

The expression in the square brackets was previously recast [21] in terms of P, g, and E_f and reduces to

$$\frac{\sigma(P)dPdg^2dE_f}{m_{13}^2 v_1 v_3[(g^2-g_-^2)(g_+^2-g^2)]^{1/2}} . \tag{22}$$

In Eq. (22), $g_\pm$ are the upper and lower limits to the speed of relative motion given by the following equations

$$g_+ = \min(g_3, G_3) , \tag{23}$$

$$g_- = \max(g_1, G_1) , \tag{24}$$

$$g_1 = |v_1 - v_3| , \tag{25}$$

$$g_3 = (v_1 + v_3) , \tag{26}$$

where

$$G_1^2 = (v_1 - v_3)^2 + 2\Delta_3/m_{13} , \tag{27}$$

$$G_3^2 = (v_1 + v_3)^2 + 2\Delta_3/m_{13} . \tag{28}$$

The final velocity of particle 1 and 3 are

$$V_1^2 = v_1^2 + 2aE_{12}/M , \tag{29}$$

$$V_3^2 = v_3^2 - 2(E_{12} + \Delta_3)/\mathcal{M} . \tag{30}$$

The masses M and $\mathcal{M}$ are defined as

$$M = m_1(1 + m_1/m_2) , \tag{31}$$

$$\mathcal{M} = \frac{(m_1+m_2)m_3}{[m_1+m_2+m_3]} . \tag{32}$$

We assume here that there is no change in the internal energy of the projectile as a result of the collision. The case of possible excitation of the projectile could be implemented in the same fashion. Hence, in this case we take $\Delta_3 = 0$. The element of angle $d\Phi$ can be written in terms of the momentum transfer P and the angular momentum of the ejected electron as

$$d\Phi = \frac{2L_i^{(\max)}L_f^{(\max)}}{R^2[(P_+^2 - P^2)(P^2 - P_-^2)]^{1/2}} \times \frac{d(L_f^2/(L_f^{(\max)})^2)}{[1 - L_f^2/(L_f^{(\max)})^2]^{1/2}} , \tag{33}$$

where $(L_{f(i)}^{(\max)})^2$ in Eq. (33) is $2m_1R^2[E_{f(i)} - V(R)]$. The upper and lower limits to the momentum transfer are defined by

$$P_+ = \min\{M(V_1 + v_1); \mathcal{M}(V_3 + v_3)\} , \tag{34}$$

$$P_- = \max\{M|V_1 - v_1|; \mathcal{M}|V_3 - v_3|\} . \tag{35}$$

Since $\sigma(P)$ does not depend on g, the integration over g^2 of the above expression for the rate coefficient can be done immediately and the doubly differential cross section (DDCS) associated with transfer of energy and angular momentum to the ejected electron reads

$$\frac{d^2\sigma_{if}^I}{dE_f dL_f^2} = \frac{C}{v_3^2}\int_{R_1}^{R_2} \frac{dR}{R^2[E_i + e^2/R - L_i^2/2m_1R^2]^{1/2}} \times \frac{1}{[E_f + e^2/R - L_f^2/2m_1R^2]^{1/2}} \times \int_{P_-}^{P_+} \frac{dP}{P^4[(P^2 - P_-^2)(P_+^2 - P^2)]^{1/2}} , \quad (36)$$

where $R_1(E_i, L_i^2)$ and $R_2(E_i, L_i^2)$ are the turning points of the orbiting electron and are given by the roots of the equation

$$[E_i + e^2/R - L_i^2/2m_1R^2]^{1/2} = 0 , \quad (37)$$

where C is a constant equals to $C = 2^{5/2}|E_i|^{3/2} m_1^{1/2} e^2/\pi$, and $E_i = -I_n = -13.605/n^2$ eV, being the initial energy of the Rydberg electron. Performing the P integration analytically, the above expression gives the DDCS associated with transfer of both energy and angular momentum, namely,

$$\frac{d^2\sigma_{if}^I}{dE_f dL_f^2} = \frac{C}{3v_3^2|E_i|^{1/2}}\int_{R_1}^{R_2} dR\,[2(P_+^2 + P_-^2)E(q) - P_-^2 K(q)] \times \frac{1}{[-R^2 + e^2R/|E_i| - L_i^2/2m_1|E_i|]^{1/2}} \frac{1}{[E_fR^2 + e^2R - L_f^2/2m_1]^{1/2}} , \quad (38)$$

where $K(q)$ and $E(q)$ are the complete elliptic integral of the first and the second kind respectively, and q is defined by

$$q = \frac{(P_+^2 - P_-^2)^{1/2}}{P_+} , \quad 0 \le q < 1 . \quad (39)$$

C. Results

Figure 2, gives the DDCS $d^2\sigma/dE_f dL_f^2$ as a function of the impact energy E for two sets of $[E_f = 1I_n, L_f = 0.1(n\hbar)]$ and $[E_f = 1I_n, L_f = 0.01(n\hbar)]$. We see that as the final angular momentum increases, the DDCS increases too, and in this case it does by a factor of 3.3.

In Fig. 3 we observe the variation of the classical ionization cross section as a function of the final angular momentum L_f. For an initial state ($n = 10$), the transition from an excited bound state to the continuum no longer follows the dipole transition rule. The maximum cross section occurs at a certain value L_f closer to 15, and then it drops sharply. Two major peaks emerge, one corresponding to the value $L_f = 1$, of dipole character and the other one to $L_f = 15$ of nondipole character. The magnitude of the DDCS corresponding to the latter one is much higher than the former one. The value $L_f^{(\max)}$ is strongly dependent on the initial value of the principal quantum number n, and is relatively insensitive to changes in the initial angular-momentum number l. For the ionization case we find that $L_f^{(\max)}$ is

$$L_f^{(\max)} = \lim_{n' \to \infty} \min \left\{ (n'-1), \sim n \left[\frac{2(n+3)}{n+1} \right]^{1/2} - \tfrac{1}{2} \right\} , \quad (40)$$

which is written in terms of n as

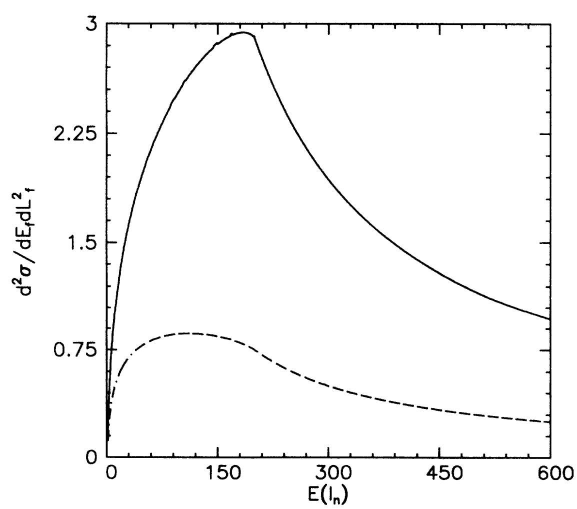

FIG. 2. Doubly differential cross section for H(10s) as a function of the impact energy E of the projectile electron; the upper curve corresponds to $E_f = 1I_n$, $L_f = 0.1(n\hbar)$, whereas the lower one corresponds to $E_f = 1I_n$ and $L_f = 0.01(n\hbar)$.

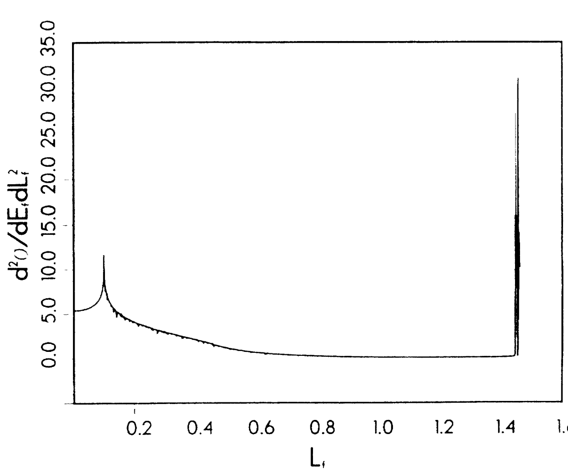

FIG. 3. Double differential cross section for H(10s) as a function of the final angular momentum $L_f(n\hbar)$ of the ejected electron, and a final energy $E_f = 0.1I_n$ for the ejected electron.

$$L_f^{(\max)} \simeq n \left[2\frac{(n+3)}{(n+1)} \right]^{1/2} - \frac{1}{2} . \tag{41}$$

This is related to the earlier work by Flannery and McCann [3] in which the predictions for the preferential population of the final discrete Rydberg states in collisions involving an initially excited atom. These features can be regarded as resulting from the drift of the pattern of the final radial orbital out of the effective range of the initial radial orbital. These trends do show up, for excitation to discrete states, in the Born and the multichannel eikonal treatments as well as in the early experimental work of Lake and Grascadden [39] and in a more recent work of Rall and co-workers [40]. As the impact energy increases the dipole transitions become much stronger. We see clearly that these trends depend on the value of the principal quantum number n of the initial state of the target, namely, the dipole transitions get stronger and stronger as n gets higher. This is due mainly to the fact that as n increases the radial period gets larger thereby decreasing the probability of having a collision.

III. CONCLUSION

In this investigation, we have developed a classical binary encounter approximation by deriving classical expressions for the energy and angular-momentum changes suffered by the orbiting electron together with the rate coefficient and various expressions for doubly differential cross sections as a function of L_f and E_f, the angular-momentum and energy of the ejected electron for ionization of Rydberg atom $H(nl)$.

This treatment is valid provided that all contributions to the rate coefficient arising from core-projectile collisions are negligible with respect to those of the Rydberg electron–projectile collisions. However, at low energy the interaction between the ionic core and the projectile could be substantial, and the Rydberg electron would be in a quasimolecular field of the ion core and neutral projectile.

The underlaying feature of this treatment is that it yields the cross section for processes in which the internal energy of an electron-ion pair, or of any ion-pair system, is changed by an amount between $\epsilon + d\epsilon$ via the binary e^{-}-A collision, and as such is, therefore, more suited to target ionization than to discrete excitation of the target atom from level n_i to n_f. This treatment does have advantage with respect to other theories because it provides much additional information concerning for instance the prediction of the final angular-momentum channels of the ejected electron. Quantum details of the interaction involved are furnished via the use of known and more refined electron-atom differential cross section such that various distortion effects would be automatically incorporated.

Our conclusions take the form of certain predictions which, as we have shown here, are fully substantiated by systematic trends in the ionization cross section of excited hydrogen atom induced by electron impact. These trends are different from the results of excitation from the ground-state target atoms, where excitation to levels optically connected to the initial level have larger cross sections. This study corroborates the previous experimental and theoretical calculations based on a Born-type approximation and on multichannel eikonal treatment. These trends would be maintained for Rydberg atoms other than the hydrogen atom H(nl), because the properties of all Rydberg atoms are similar to those of highly excited hydrogen atoms.

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