

Photoelectron angular distributions from electron-atom collisions

Michael G. White

Department of Chemistry, Brookhaven National Laboratory, Upton, New York 11973

(Received 14 April 1982)

The differential cross section for electron-impact ionization of atoms has been derived for the general case and the dipole ($E1$) limit of the Born approximation. Emphasis was placed on the theoretical description of the ($e,2e$) angular correlation between fast electrons scattered in the forward direction and electrons ejected by the ionization process. The general angular-momentum treatment of this work results in a more complete description of the dipole ($e,2e$) angular distribution than previous purely kinematic analyses.

I. INTRODUCTION

The dipole ($e,2e$) quantitative simulation of photoelectron spectroscopy¹⁻³ has proven to be an extremely valuable tool for the determination of energy-dependent photoionization cross sections of atoms and molecules.⁴ The technique utilizes the fact that, at vanishingly small momentum transfers, electron-impact excitation proceeds predominantly via electric dipole ($E1$) transitions and thereby follows optical selection rules.^{5,6} In addition to these cross-section measurements, which are taken at a fixed detector geometry, the dipole ($e,2e$) angular correlation can also be used to obtain the asymmetry parameter β which determines the photoelectron angular distribution for an $E1$ process.^{7,8} The first measurements of this kind by Brion and co-workers^{1,9} clearly demonstrated the feasibility of determining β parameters by dipole ($e,2e$), however, the data suffered from an incomplete, empirical analysis of the reaction kinematics. A later, more detailed kinematic study by Hamnett *et al.*,³ resulted in an approximate expression for the dipole ($e,2e$) differential cross section in terms of scattering parameters and slit geometries. Although their formula does not have a straightforward angular dependence, it does account for the existence of a "magic" angle ($\theta=54^\circ 44'$) between the two outgoing electrons at which the differential cross section is independent of the β parameter. The fact that this angle is identical to the magic angle for photoionization in the dipole approximation suggests that the underlying angular dependence has a more general form and is independent of the approximations used in Ref. 3. That this is so is clear from Yang's theorem⁷, which restricts the angular distribution of products from any cylindrically symmetric $E1$ process to even polynomials of $\cos\theta$ with a maximum exponent of two, i.e.,

$A_0 + A_2 P_2(\cos\theta)$.¹⁰ Hence, the existence of a magic angle at $\theta=54^\circ 44'$ for the dipole ($e,2e$) technique is a natural consequence of detecting (in a cylindrically symmetric fashion) only those collisions which have small momentum transfers and thus dominated by electric dipole transitions.¹¹ In a related kinematic study, Kim¹² derived the angular distribution of secondary electrons produced by charged particle impact for which the scattered particle is undetected. For the region of small momentum transfers Kim found that the angular distribution of secondary electrons was approximately that of photoelectrons produced by unpolarized light, i.e., $1 - \beta P_2(\cos\theta)/2$. However, the differential cross section derived by Kim also included an additional angularly dependent term which was left unspecified.

In this work, I report the results of a general angular-momentum derivation of the atomic ($e,2e$) differential cross section in the Born approximation. Particular attention is paid to the optical ($E1$) limit of the differential cross section and its application to the dipole ($e,2e$) technique. The dipole ($e,2e$) angular correlation is found to have the general form expected from symmetry arguments and explicit expressions for the coefficients A_0 and A_2 are given. This study is complementary to similar theoretical analyses of charged-particle impact ionization by Manson *et al.*¹³ and Theodosiou¹⁴ in which the optical limit and the ($e,2e$) reactions were not explicitly treated. The application of these results to the determination of β parameters from future dipole ($e,2e$) experiments and its extension to nondipole processes are discussed.

II. THEORY

A. General

Consider an ionizing collision between a fast electron with initial momentum $\hbar k$ (initial nonrelativis-

tic kinetic energy $T = mv^2/2$) with a stationary atom with atomic number Z . The kinematics of such a collision are shown in Fig. 1. The scattered and ejected electrons have final momenta of $\hbar\vec{k}'$ and $\hbar\vec{k}_e$, respectively, and θ represents the scattering angle relative to the incident beam direction. The momentum transferred in the collision is given by

$$\hbar\vec{q} = \hbar\vec{k} - \hbar\vec{k}',$$

$$\frac{d^3\sigma_{J_1 \leftarrow J_A}}{d\hat{k}' d\hat{k}_e d\epsilon} = \frac{4kk'}{(T/R)q^4 [J_A]} \delta(E - \epsilon - I) \sum_{M_A = -J_A}^{J_A} \sum_{M_I = -J_I}^{J_I} \sum_{\sigma = \pm 1/2} \left| \left\langle J_I M_I, \vec{k}_e \sigma \left| \sum_{i=1}^Z e^{i\vec{q} \cdot \vec{r}_i} \right| J_A M_A \right\rangle \right|^2, \quad (1)$$

where $d\hat{k}'$ and $d\hat{k}_e$ are the solid angles for the scattered and ejected electrons, respectively, ϵ is the kinetic energy of the ejected electron, $[J_A]$ represents the quantity $2J_A + 1$, and R is the Rydberg constant. The Born excitation matrix element gives the strength of the transition between a neutral atom in state $|J_A M_A\rangle$ to a final state consisting of an ion core in state $|J_I M_I\rangle$ and a continuum electron with linear momentum $\hbar\vec{k}_e$ and helicity σ .¹⁷ The summations over M_A , M_I , and σ represent averaging over initial-state orientations and summing over degenerate final states. The delta function $\delta(E - \epsilon - I)$ imposes energy conservation on the observed ionization event; here E is the energy loss of the scattered particle

$$[E/R = (ka_0)^2 - (k'a_0)^2]$$

and the symbol I represents the ionization energy

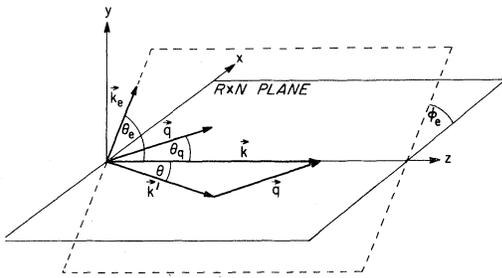


FIG. 1. Collision geometry for inelastic electron-atom scattering leading to the ejection of a bound electron. Vectors \vec{k} and \vec{k}' correspond to the initial and final momenta of the scattered electron, \vec{q} is the momentum transferred in the collision, and \vec{k}_e is the momentum of the ejected electron. Momenta \vec{k} , \vec{k}' , and \vec{q} all lie in the "reaction" plane defined by the xz axes.

where the three momenta lie in a "reaction" plane defined by the xz axes and where θ_q is the angle between the momentum-transfer direction and the beam axis. The trajectory of the ejected electron is described by the polar angles θ_e and ϕ_e as shown.

For fast electron-impact ionization (fast relative to the mean orbital velocity of the electron before ejection), the atomic $(e, 2e)$ differential cross section can be formulated in the Born approximation to give^{6,15,16}

given by the energy difference between the neutral and ionic atomic states, i.e., $I = E(J_I) - E(J_A)$. Exchange effects have been ignored in writing Eq. (1) as I am interested predominantly in describing inelastic collisions with energy losses up to ~ 100 eV. As the incident electron-beam energy is several kilovolts or higher, the scattered and ejected electrons have substantially different energies

$$[\epsilon/(T - E) < 0.05]$$

and, as a consequence, the effects of exchange are negligible.¹⁵

To evaluate the matrix element on the right-hand side of Eq. (1), an explicit form of the coupled final state is required. Following Eichler and Fritsch,¹⁸ the ejected continuum electron is written as a partial-wave expansion over one-electron angular-momentum eigenstates

$$\begin{aligned} |\vec{k}_e \sigma\rangle = & \sum_{l=0}^{\infty} \sum_{j=|l-s|}^{l+s} \sum_{m=-j}^j (-1)^{s-l-\sigma} i^l \tilde{f}_j \\ & \times \begin{pmatrix} l & s & j \\ 0 & \sigma & -\sigma \end{pmatrix} \\ & \times D_{m\sigma}^j(\hat{k}_e) e^{-i\delta_l^j} \left| \left(\frac{l}{2} \right) jm \right\rangle, \end{aligned} \quad (2)$$

where l is the orbital angular momentum, the quantity in parentheses is a 3- j symbol, $D_{m\sigma}^j(\hat{k}_e)$ is a rotation matrix element,¹⁹ and \tilde{a} represents the quantity $\sqrt{2a+1}$. The radial part of $|jm\rangle$ has the asymptotic form of a Coulomb wave with phase shift δ_l^j . The many-electron wave function is then obtained by coupling the ionic state wave function and the continuum electron state of Eq. (2) to give

$$|J_I M_I, \vec{k}_e \sigma\rangle = \sum_{ijm} \sum_{J=|J_I-j|}^{J_I+j} \sum_{M=-J}^J (-1)^{s-l+j-J_I-M-\sigma} i^l \tilde{l} \tilde{j} \tilde{J} \begin{pmatrix} l & s & j \\ 0 & \sigma & -\sigma \end{pmatrix} \begin{pmatrix} J_I & j & J \\ M_I & m & -M \end{pmatrix} D_{m\sigma}^j(\hat{k}_e) e^{-i\delta_j^I} |JM\rangle. \quad (3)$$

The $|JM\rangle$ represent continuum eigenfunctions of the full Z -electron Hamiltonian which are allowed by symmetry to contribute to the asymptotic ($r_e \rightarrow \infty$) dissociation channel specified by $|J_I M_I, \vec{k}_e \sigma\rangle$.

Use is also made of the multipole expansion of the exponential operator, i.e.,

$$\sum_{i=1}^Z e^{i\vec{q} \cdot \vec{r}_i} = \sum_{\lambda=0}^{\infty} \sum_{\nu=-\lambda}^{\lambda} i^\lambda [\lambda] C_{\lambda\nu}^*(\hat{q}) T_{\lambda\nu}(\hat{r}_i), \quad (4a)$$

where

$$T_{\lambda\nu}(\hat{r}_i) = \sum_{i=1}^Z j_\lambda(qr_i) C_{\lambda\nu}(\hat{r}_i) \quad (4b)$$

$$A_K(q) = \frac{[K]}{[J_A]} \sum_{l_1, l_2} i^{l_1+l_2} \tilde{l}_1 \tilde{l}_2 \begin{pmatrix} l_1 & l_2 & K \\ 0 & 0 & 0 \end{pmatrix} \\ \times \sum_{\lambda_1, \lambda_2} i^{\lambda_1+\lambda_2} [\lambda_1][\lambda_2] \begin{pmatrix} \lambda_1 & \lambda_2 & K \\ 0 & 0 & 0 \end{pmatrix} \\ \times \sum_{j_1 j_2} \sum_{J_1 J_2} (-1)^{-s-\lambda_2+l_2+j_1+j_2+J_1+2J_2-J_I} \tilde{j}_1 \tilde{j}_2 \tilde{J}_1 \tilde{J}_2 \\ \times \begin{pmatrix} l_1 & l_2 & K \\ j_2 & j_1 & s \end{pmatrix} \begin{pmatrix} \lambda_1 & \lambda_2 & K \\ J_2 & J_1 & J_A \end{pmatrix} \begin{pmatrix} j_1 & j_2 & K \\ J_2 & J_1 & J_I \end{pmatrix} \langle J_A || T_{\lambda_1} || J_1 \rangle \exp[-i(\delta_{j_2}^{l_2} - \delta_{j_1}^{l_1})] \\ \times \langle J_2 || T_{\lambda_2} || J_A \rangle, \quad (5b)$$

where the quantities in curly braces are 6- j symbols and the $\langle J_A || T_\lambda || J \rangle$ represent the reduced ionization matrix elements. The above expressions are completely general in that no particular coupling scheme has been chosen to describe the internal structure of the neutral atom or residual ion core. The use of J_j coupling ($\vec{J} = \vec{J}_I + \vec{j}$) in the construction of the $|JM\rangle$ continuum eigenstates (Eq. 3), however, implicitly assumes that the interaction of the ion core and continuum electron is weak. For $A_K(q)$ coefficients in alternative coupling schemes, the reader is referred to the single-particle results of Manson *et al.* (LS coupling)¹³ and Theodosiou (jj coupling).¹⁴

Of particular interest here is the measurement of the $(e,2e)$ angular correlations in which the scat-

and where $C_{\lambda\nu}$ are modified spherical harmonics ($Y_{\lambda\nu} \sqrt{4\pi/\lambda}$) and j_λ are spherical Bessel functions of order λ .

After substitution of Eqs. (3) and (4), Eq. (1) can be reduced by the application of standard angular-momentum algebra, after which the $(e,2e)$ differential cross section can be written as

$$\frac{d^3\sigma_{J_I \leftarrow J_A}}{d\hat{k}' d\hat{k}_e d\epsilon} = \frac{4kk'}{(T/R)q^4} \sum_{K=0}^{\infty} A_K(q) P_K(\hat{k}_e \cdot \hat{q}), \quad (5a)$$

where the coefficients $A_K(q)$ are given by

tered electron detector is fixed along the incident beam direction and collects only a cone of scattered electrons in the "forward" direction. The integration of Eq. (4a) over the scattered electrons' collected solid angle $d\hat{k}'$ in this detector geometry is simplified by the use of the relationship

$$dk' = \frac{q}{kk'} dq d\phi' \quad (6)$$

which follows from

$$q^2 = k^2 + k'^2 - 2kk' \cos\theta.$$

The integration over $d\phi'$ (corresponding at arbitrary choice of the reaction plane) is accomplished via the identity

$$\int P_K(\hat{k}_e \cdot \hat{q}) d\phi' = P_K(\cos\theta_e) P_K(\cos\theta_q) \int d\phi' \\ + 2 \sum_{p=1} \frac{(L-p)!}{(L+p)!} P_K^p(\cos\theta_e) P_K^p(\cos\theta_q) \int \cos[p(\phi_e - \phi')] d\phi', \quad (7)$$

where P_K^p are associated Legendre polynomials. As scattered electrons are collected in a cone symmetric about the beam axis, the integration is from $\phi' = 0^\circ$ to $\phi' = 2\pi$; the integral in the summation of Eq. (7) can be evaluated by noting that a rotation about $\phi_e - \phi'$ is equivalent to a rotation about ϕ' (see Fig. 1). Hence, this latter integral is identically zero and Eq. (7) becomes simply

$$\int_0^{2\pi} P_K(\hat{k}_e \cdot \hat{q}) d\phi' = 2\pi P_K(\cos\theta_e) P_K(\cos\theta_q).$$

The $(e, 2e)$ differential cross section can then be written as

$$\frac{d^2\sigma_{J_I \leftarrow J_A}}{d\hat{k}_e d\epsilon} = \frac{8\pi}{(T/R)} \sum_K P_K(\cos\theta_e) \\ \times \int_{q_{\min}}^{q_{\max}} A_K(q) P_K(\cos\theta_q) \\ \times \frac{d(\ln q)}{q^2}, \quad (8)$$

where the limits of integration for q are determined from

$$q^2 = k^2 + k'^2 - 2kk' \cos\theta,$$

with $\theta = 0^\circ$ and $\theta = \theta_0$, where θ_0 is the acceptance angle of the scattered electron detector. Hence, for a fixed collection solid angle in the scattered electron detector (fixed θ_0) and a given energy loss (which fixes k' by $k'a_0 = [(ka_0)^2 - E/R]^{1/2}$) the angular distribution of ejected electrons relative to the beam direction reduces to a Legendre-polynomial expansion of the form $\sum A_K P_K(\cos\theta_e)$. This angular distribution has the same general form as that for photoionization by unpolarized, multipole radiation for which the even K terms represent contributions (incoherent) from the various multipole waves, e.g., electric dipole ($E1$) and magnetic dipole ($M1$) processes contribute to the coefficients with $K=0, 2$, and 4 , while the odd- K terms give their interference, e.g., $K=1$ includes the ($E1, M1$) and ($E1, E2$) interference terms.²⁰ For electron excitation only electric multipoles exist, so that by analogy with radiation, excitation by electric L -pole processes contribute to the even- K terms up to $2L$ and the interference between these processes is given by the

odd- K terms. For photon excitation, the various multipole contributions are determined only by the relative magnitudes of dynamical coefficients similar in form to the A_K coefficients given by Eq. (5b).²¹ In the case of electron excitation, however, the relative magnitudes of these dynamical terms are multiplied by the purely kinematic factor $q^{-2} P_2(\cos\theta_q)$ which when integrated over q can emphasize different excitation multipolarities depending on the limits q_{\min} and q_{\max} . For example, for vanishingly small momentum transfers ($q_{\max}, q_{\min} \rightarrow 0$) only dipole ($E1$) excitations are readily observable (see Sec. IIB), while for larger momentum transfers electron impact becomes increasingly sensitive to quadrupole and higher-order excitations.²²

Finally, it should be noted that cylindrical symmetry is imposed on the ionization process, described by Eq. (8), by choosing the scattered electron detector to lie symmetrically about the beam axis. If instead, measurements were made off axis, the integration over $d\phi'$ [Eq. (7)] would no longer be straightforward as then ϕ' would be a function of the scattered electron detector's collection angles. This will, in general, introduce an azimuthal dependence into the $(e, 2e)$ differential cross sections, i.e.,

$$d\sigma \propto \sum b_{KN} Y_{KN}(\hat{k}_e).$$

For most scattering experiments this additional degree of freedom is highly undesirable, however, the binary $(e, 2e)$ coincidence technique makes exclusive use of this azimuthal dependence for the determination of momentum distributions.²³

B. Electric dipole limit ($q \rightarrow 0$)

For small momentum transfers and high incident electron velocities [conditions typical for the dipole $(e, 2e)$ experiment⁴], the Bethe approximation to the Born transition operator can be employed, i.e.,⁵

$$\lim_{K \rightarrow 0} e^{i\vec{q} \cdot \vec{r}_i} \sim 1 + i\vec{q} \cdot \vec{r}_i,$$

where the 1 term is appropriate for elastic scattering only. For ease of manipulation it is useful to put the Bethe-Born operator into spherical form as follows:

$$\begin{aligned} \sum_{i=1}^Z i \vec{q} \cdot \vec{r}_i &= iq \sum_{i=1}^Z r_i \cos(\theta_{qr_i}) \\ &= iq \sum_{\nu=-1}^1 C_{1\nu}^*(\hat{q}) T_{1\nu}(\hat{r}_i), \end{aligned} \quad (9a)$$

where

$$T_{1\nu}(\hat{r}_i) = \sum_{i=1}^Z r_i C_{1\nu}(\hat{r}_i) \quad (9b)$$

and where Eq. (9a) follows from the use of the spherical-harmonic addition theorem. Comparison of Eqs. (9a) and (9b) with the general multipole expansion [Eq. (4)] shows that the Bethe approximation is equivalent to restricting the sum over λ to the single term $\lambda=1$ and using the limiting form ($qr_i \rightarrow 0$) of the spherical Bessel functions

$$\lim_{qr_i \rightarrow 0} j_\lambda(qr_i) \sim \frac{(qr_i)^\lambda}{(2\lambda+1)!!}$$

which reduces to $qr_i/3$ for $\lambda=1$. The operator $T_{1\nu}(\hat{r}_i)$ defined by Eq. (9b) is the familiar spherical representation of the electric dipole ($E1$) operator in the length form.

The dipole limit of the general ($e,2e$) differential cross section can then be obtained by substituting $\lambda_1=\lambda_2=1$ in Eq. (5b) and replacing the reduced matrix elements $\langle J_A || T_\lambda || J \rangle$ by $q \langle J_A || T_1 || J \rangle$ as required by Eqs. (9a) and (9b). From the triangular condition

$$|\lambda_1 - \lambda_2| \leq K \leq \lambda_1 + \lambda_2,$$

imposed by the 3- j symbol $\begin{pmatrix} \lambda_1 & \lambda_2 & K \\ 0 & 0 & 0 \end{pmatrix}$ in Eq. (5b) and the fact that $\lambda_1 + \lambda_2 + K$ must be even for this coefficient to be nonzero, K is restricted to take the values of 0 and 2. Furthermore, the 3- j symbol $\begin{pmatrix} l_1 & l_2 & K \\ 0 & 0 & 0 \end{pmatrix}$ requires that l_1 and l_2 have the same parity. With these considerations the dipole ($e,2e$) "photoelectron" angular distribution is given by

$$\frac{d^2 \sigma_{J_I \leftarrow J_A}}{d\hat{k}_e d\epsilon} = \frac{8\pi}{(T/R)} \left[A_0(E1) \int_{q_{\min}}^{q_{\max}} d(\ln q) + A_2(E1) P_2(\cos \theta_e) \int_{q_{\min}}^{q_{\max}} P_2(\cos \theta_q) d(\ln q) \right], \quad (10a)$$

where

$$A_0(E1) = \frac{1}{\sqrt{30} [J_A]} \sum_{ijJ} |\langle J_A || T_1 || J[(ls)jJ_I] \rangle|^2, \quad (10b)$$

$$\begin{aligned} A_2(E1) &= \frac{10}{\sqrt{30} [J_A]} \sum_{l_1 l_2} \tilde{l}_1 \tilde{l}_2 \begin{Bmatrix} l_1 & l_2 & 2 \\ 0 & 0 & 0 \end{Bmatrix} \\ &\quad \times \sum_{j_1 j_2 J_1 J_2} \sum_{J} (-1)^{-s+l_2+j_1+j_2+J_1+2J_2-J_I} \tilde{j}_1 \tilde{j}_2 \tilde{J}_1 \tilde{J}_2 \\ &\quad \times \begin{Bmatrix} l_1 & l_1 & 2 \\ j_2 & j_1 & s \end{Bmatrix} \begin{Bmatrix} 1 & 1 & 2 \\ J_2 & J_1 & J_A \end{Bmatrix} \begin{Bmatrix} j_1 & j_2 & 2 \\ J_2 & J_1 & J_I \end{Bmatrix} \\ &\quad \times \langle J_A || T_1 || J_1 \rangle \exp[-i(\delta_{j_1}^{l_1} - \delta_{j_2}^{l_2})] \langle J_2 || T_1 || J_A \rangle \end{aligned} \quad (10c)$$

and where T_1 is defined by Eq. (9b). From Eq. (10a) it is clear that the dipole ($e,2e$) angular distribution is of the form $A_0 + A_2 P_2(\cos \theta_e)$ in accordance with Yang's theorem as discussed earlier. In addition, the magic angle for which the differential cross section is independent of β is confirmed to be that at which $P_2(\cos \theta_e)$ is zero, i.e., $\theta_e = 54^\circ 44'$.

By comparison of Eq. (10a) with the differential cross section for photoionization,²⁴ one finds the following relationships:

$$\begin{aligned} \frac{df^0}{dE} &= \frac{a_0^2}{(E/R)} A_0(E1), \\ \beta &= \frac{A_1(E1)}{A_0(E1)}, \end{aligned}$$

where df^0/dE is the integrated photoionization oscillator strength and β is the usual asymmetry parameter. In terms of these quantities, the dipole ($e,2e$) differential cross section becomes

$$\frac{d^2\sigma_{J_1 \leftarrow J_A}}{d\hat{k}_e d\epsilon} = \frac{8\pi a_0^2}{(E/R)(T/R)} \left[\int_{q_{\min}}^{q_{\max}} d(\ln q) \right] \frac{df^0}{dE} \times \left[1 - \frac{\beta}{2} P_2(\cos\theta_e) F(\vec{q}) \right], \quad (11a)$$

where

$$F(\vec{q}) = 1 - 3 \left[\int_{q_{\min}}^{q_{\max}} \cos^2\theta_q d(\ln q) \right] \times \left[\int_{q_{\min}}^{q_{\max}} d(\ln q) \right]^{-1}. \quad (11b)$$

Apart from the kinematic factor $F(\vec{q})$, the angular dependence for dipole ($e, 2e$) is identical to photoionization with unpolarized light.^{8,20,24} Furthermore, as the momentum transfer decreases, the angle θ_q approaches $\pi/2$ (\vec{q} becomes nearly perpendicular to the beam axis) and $F(\vec{q}) \rightarrow 1$, in which case the angular distributions for dipole ($e, 2e$) and photoionization becomes equivalent. Experimentally, this limit is unattainable as the acceptance angle of the forward detector (θ_0) must decrease to zero which would lead to a vanishingly small collection solid angle.

The kinematic factor $F(\vec{q})$ can be evaluated for typical experimental conditions by first expressing $\cos^2\theta_q$ in terms of momentum variables, i.e.,

$$\cos\theta_q = \frac{(k^2 - k'^2 + q^2)^2}{4k^2 q^2},$$

which can be substituted in Eq. (11b) and then readily integrated. Following the analysis of Hammett *et al.*³ for small angle scattering, the integration limits can be written analytically (to second order in the parameter $x = E/2T$) as $q_{\min} \sim kx$ and $q_{\max} \sim k(x^2 + \theta_0^2)^{1/2}$, where θ_0 is the acceptance angle of the forward detector. Utilizing these expressions, the integrated form of Eq. (11b) can be evaluated to give

$$F(\vec{q}) \sim 1 - 3x \left[\frac{x^2 + \theta_0^2 + 4}{x^2 + \theta_0^2} \right] \left[\ln \left[1 + \frac{\theta_0^2}{x^2} \right] \right]^{-1}. \quad (12)$$

In Fig. 2 the function $F(\vec{q})$ is plotted versus energy loss [for dipole ($e, 2e$) the energy loss is analogous to the photon energy] for incident electron energies of 1.5, 3.5, and 10 keV. The acceptance angle used to calculate these curves was that used in the dipole ($e, 2e$) apparatus of Brion and co-workers ($\theta_0 = 1.25 \times 10^{-2}$ rad).¹⁻⁴ For $T = 3.5$ keV, which

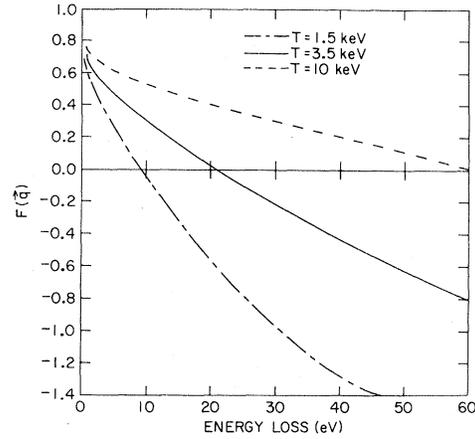


FIG. 2. Kinematic factor $F(\vec{q})$ as a function of energy loss (photon energy) for three different incident beam energies. These curves were calculated for $\theta_0 = 1.25 \times 10^{-2}$ rad (see Sec. II A of the text).

is the energy for which the early dipole ($e, 2e$) β measurements were made,^{1,9} one can see that the sensitivity of the dipole ($e, 2e$) technique to the photoelectron angular anisotropy [in comparison to photoionization experiments for which $F(\vec{q}) \equiv 1$ for all photon energies] is fair [$F(\vec{q}) > 0.2$] for low- ($E < 15$ eV) and high- ($E > 30$ eV) energy losses but very poor at intermediate energies, with $F(\vec{q}) = 0$ near ~ 21 eV. The curves for $T = 1.5$ and 10 keV illustrate that the sensitivity or anisotropy can be increased in certain spectral regions by changing the incident beam energy; in fact, at higher energies the $T = 1.5$ -keV curve approaches the anisotropy expected for linearly polarized light, i.e., $1 + \beta P_2(\cos\theta_e)$. The curves are shown to 60-eV energy loss as this represents the practical limit for which dipole ($e, 2e$) coincidence measurements can be made at present. This upper energy-loss limit results from the kinematic factor $E^{-1} \int d(\ln q)$ [see Eq. (11a)], which can be shown to behave as E^{-3} for $\theta_0 \ll x$.^{4,22} Hence, the observable dipole ($e, 2e$) intensity falls rapidly with increasing energy loss, making coincidence measurements at higher energy extremely difficult.

Comparison of Eq. (11a) with the kinematically derived dipole ($e, 2e$) differential cross section of Hammett *et al.*³ suggests that their function C_α can be related to the present work by

$$C_\alpha \sim -P_2(\cos\alpha)F(\vec{q})/2,$$

where α is equivalent to our angle θ_e . This approximate relationship is confirmed by suitably rearranging their C_α expression [Eq. (5b), Ref. 3] to give²⁵

$$\frac{2}{P_2(\cos\alpha)} C_\alpha = 1 - 3 \left[\frac{\theta_0^2}{x^2 + \theta_0^2} \right] \left[\ln \left[1 + \frac{\theta_0^2}{x^2} \right] \right]^{-1}$$

which is equivalent to Eq. (12) for $x^2 + \theta_0^2 \ll 4$.

III. CONCLUDING REMARKS

In this work, I have detailed the derivation of the ($e,2e$) angular correlation in both the general case and in the Bethe-Born dipole limit. The general differential cross section can be qualitatively interpreted by analogy with photoelectron angular distributions produced by multipole radiation. The relatively simple form of the general differential cross section, i.e., $\sum A_K P_K(\cos\theta_e)$, provides a basis for interpreting the angular distribution of ejected electrons produced via large momentum-transfer collisions for which nondipole processes are expected to be important.

The dipole ($e,2e$) photoelectron angular distribution derived here is consistent with and incorporates both the symmetry arguments inherent in Yang's theorem and the results of kinematic analyses. As expected, the dipole ($e,2e$) differential cross section has the same general form as that for photoionization, i.e., $A_0 + A_2 P_2(\cos\theta_e)$, and, in principle, can be used for the determination of β parameters. The most important difference between the two techniques, as regards β measurements, is that for most experimentally realizable conditions, dipole ($e,2e$) will exhibit smaller angular anisotropies than the corresponding photoionization experiment. Conse-

quently, it is more difficult to extract β parameters from dipole ($e,2e$) data. The viability of the dipole ($e,2e$) technique for making β measurements, then, depends on optimization of kinematic factors through the variation of the beam energy in order to maximize the observable anisotropy [maximize $F(\bar{q})$, see Fig. 2]. This disadvantage in sensitivity, however, is partly offset by the ability of the dipole ($e,2e$) technique to provide angular data over the entire uv spectral range; only with the highly dispersed radiation from laboratory rare-gas or hydrogen continuum emission sources and/or synchrotron radiation from electron-storage rings can this same energy range be covered by photoionization experiments. Thus, it appears that the dipole ($e,2e$) technique has the potential to provide valuable "survey" data on photoelectron angular distributions over wide spectral regions for which comparable photoionization data are particularly lacking.

ACKNOWLEDGMENTS

The author would like to thank Professor C. E. Brion for providing the stimulus for this work and Dr. Y.-K. Kim for helpful discussions and comments. Financial support for this work was provided in part by the National Sciences and Engineering Research Council of Canada and by the U.S. Department of Energy under Grant No. DE-AC02-76CH00016.

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²⁵The expression for C_α as published in Ref. 3 contains several typographical errors and their Eq. (5b) should read

$$C_\alpha = \left[4 \ln \left[\frac{x^2 + \theta_0^2}{x^2} \right]^{1/2} \right]^{-1} \\ \times \left[\frac{3 \cos^2 \alpha - 1 - 2\chi_0^2}{2} \left[\frac{\theta_0^2}{x^2 + \theta_0^2} \right] \right. \\ \left. - \frac{2 - 3 \sin^2 \alpha - 3\chi_0^2}{3} \ln \left[1 + \frac{\theta_0^2}{x^2} \right]^{1/2} \right].$$

Also, in relating this expression to Eq. (12), I have assumed that the quantity χ_0^2 is negligibly small (χ_0 represents the opening angle of the slit on the ejected electron detector). The appearance of this quantity results from integrating the differential cross section over $d\hat{k}_e$.