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Angular Distributions of Secondary Electrons in the Dipole Approximation*

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A close relationship between the angular distributions of secondary electrons produced by photoionization and by impact of fast charged particles is pointed out. The dipole-interaction term in the charged-particle impact cross section [which has $(\ln T)/T$ dependence, T being proportional to the incident energy] has essentially the same angular dependence as the photoelectrons ejected by unpolarized light. An analysis of recent electron-impact data on He and N_2 indicates consistency with the present theory.

I. INTRODUCTION

Recently an increasing number of experimental data on the angular distribution of electrons ejected from atoms and molecules either through photoionization[1–3] or through ionization by charged particles[4–7] have appeared in the literature. This paper shows a simple relationship between the photoelectron data and the dipole-interaction part of the charged-particle impact data (referred to as the secondary-electron data for brevity as opposed to the photoelectron data).

Ionizing collisions between a fast charged particle and an atom or molecule[8] can be qualitatively classified into two parts: "soft" or glancing collisions with large impact parameters and small momentum transfers, and "hard" or close collisions with small impact parameters and large momentum transfers. The soft collisions, because of their large impact parameters, impart on the atomic electrons an impulsive force (= momentum transfer) nearly perpendicular to the path of the incident particle. The effect of such a force upon the atomic electrons is equivalent to that of light propagating in the direction of the incident particle[9]; the electrons experience a force along the direction of the polarization of the light. In short, ionization by soft collisions is equivalent to photoionization, and hence the angular distribution of secondary electrons ejected by soft collisions should be essentially the same as that of photoelectrons. Our objective is to confirm this surmise by detailed analysis.

The first Born approximation provides an adequate basis for the evaluation of cross sections for ionization of atoms by fast charged particles. The soft-collision (dipole) part of the Born cross section contributes a $(\ln T)/T$ factor resulting from the integration over the momentum transfer, T being proportional to the kinetic energy of the incident particles.[10–12] Hence, the part of the ionization cross section which depends logarithmically on the incident energies should have an angular distribution closely related to that of photoelectrons. The precise relationship between the two angular distributions is derived in Sec. II, and an analysis of the electron-impact data on He and N_2 by Opal, Beaty, and Peterson[6] is presented in Sec. III.

II. THEORY

We begin with the basic expression for the Born cross section as given by Eqs. (2.7) and (2.21) of Ref. 12 (referred to as I71 hereafter), with a slight change; namely, the solid angle $d\omega$ in Eq. (2.1) of I71 is expressed as $(m/\hbar^2 kk')\times dQ\,d\Phi$, where Φ is the azimuth of the momentum transfer $\hbar\vec{K}=\hbar\vec{k}-\hbar\vec{k}'$ about the incident particle momentum $\hbar\vec{k}$, $\hbar\vec{k}'$ is the scattered particle momentum, m is the electron mass, and $Q=(\hbar K)^2/2m$. The differential cross section for the ionization of an atom to a continuum state $|E\Omega\rangle$ by a fast particle of charge ze and speed v is given by

$$d\sigma_{E\Omega}(T)=\frac{2a_0^2z^2}{T/\mathcal{R}}\,\frac{f_{E\Omega}(\vec{K})}{E/\mathcal{R}}\,\frac{dQ\,d\Phi}{Q}\,, \tag{1}$$

where a_0 is the Bohr radius, $\mathcal{R}$ is the Rydberg energy $(=me^4/2\hbar^2)$, $T=\frac{1}{2}mv^2$, and $f_{E\Omega}(\vec{K})$ is the generalized oscillator strength. The final state $|E\Omega\rangle$ is characterized by the excitation energy E measured from the initial state, and Ω stands for all other labels necessary to identify the collision products uniquely, such as the momentum $\hbar\vec{\kappa}$ and energy $W=(\hbar\kappa)^2/2m$ of the ejected electron. [Note that Eq. (2.21) of I71 refers to the "total" generalized oscillator strength, i.e., $f_{E\Omega}(\vec{K})$ summed over Ω.]

The experiments reported in Refs. 4–7 measured the energy and angular distributions of the ejected electrons, but not those of the scattered particle. The observed cross sections therefore correspond to Eq. (1) integrated over $\vec{K}$, or equivalently over Φ and Q, and also summed over all ionic states with fixed $\vec{\kappa}$. The summation over the ionic states is postponed to the final step.

The integration over $\vec{K}$ corresponds to the summation over the direction of polarization in the photoelectron case, and indeed our final result is very similar to the angular distribution for the photoelectrons ejected by *unpolarized* light propagating in the direction of the incident beam of the charged particles.

For a moment, in Eq. (1) we focus our attention on the integration over Q only. The integral $\int dQ/Q$ introduces a logarithmic dependence on the limits of Q; in particular, the lower limit [Eq. (2.17) of I71]

$$Q_{\min}=E^2/4T \tag{2}$$

gives the $\ln T$ dependence on the incident energy mentioned earlier. At low $Q(\ll\mathcal{R})$, $f_{E\Omega}(\vec{K})$ reduces to the dipole oscillator strength, which depends on the angle λ between the direction of the ejected electron and the direction $\hat{K}$ of the momentum transfer but not explicitly on $|\vec{K}|$. For ionization by charged-particle impact, the direction of the polarization of the photon is replaced by $\hat{K}$. The angular distribution of electrons ejected with energy W by a photon of energy E is well known[13]:

$$\lim_{K\to 0} f_{E\Omega}(\vec{K})=(f_{EW}/4\pi)\,[1+\beta_{EW}P_2(\cos\lambda)]\,, \tag{3}$$

where f_{EW} is the dipole oscillator strength, β_{EW} is the asymmetry parameter, and P_2 is the Legendre polynomial of the second order. It is sufficient for our purpose to carry out the integration of Eq. (1) in detail only in the vicinity of $Q_{\min}$ to bring out the essential feature of the angular distribution we are after.

As was done by Bethe,[10] Fano,[11] and Miller and Platzman,[14] we divide the range of Q between $Q_{\min}$ and $Q_{\max}\sim 4T$ into two parts: one for soft collisions $Q_{\min}\leq Q\leq Q_0$ and the other for the remainder $Q_0\leq Q\leq Q_{\max}$. Since the $\ln T$ dependence comes from $Q_{\min}$, the choice of Q_0 is immaterial, and we choose a small value which is independent of T such that the dipole term (3) dominates throughout the low-Q region. In the second region, $f_{E\Omega}(\vec{K})$ is finite and for $Q>E$ it decreases as some negative power of Q in such a way that the integral remains finite for large T. Details of the integral in the second region are irrelevant to our objectives, and symbolically we put

$$\int_{Q_0}^{Q_{\max}} dQ\int_0^{2\pi} d\Phi\, f_{E\Omega}(\vec{K})\,\mathcal{R}/EQ\equiv 2\pi B'_{E\Omega}(T,Q_0)\,. \tag{4}$$

In the low-Q region, we have from Eqs. (1) and (3)

$$\int_{Q_{\min}}^{Q_0} dQ\int_0^{2\pi} d\Phi\, f_{E\Omega}(\vec{K})/Q\cong\int_{Q_{\min}}^{Q_0} dQ\int_0^{2\pi} d\Phi \times(f_{EW}/4\pi)\,[1+\beta_{EW}P_2(\cos\lambda)]/Q\,. \tag{5}$$

To integrate Eq. (5) over Φ, we note from Fig. 1 that

$$\cos\lambda = \cos\theta\cos\psi + \sin\theta\sin\psi\cos\Phi ,$$

where θ is the angle between $\vec{\kappa}$ and $\vec{k}$, and from the addition theorem for the Legendre polynomials, we have

$$\int_0^{2\pi} P_2(\cos\lambda)\, d\Phi = 2\pi P_2(\cos\theta)\left(\tfrac{3}{2}\cos^2\psi - \tfrac{1}{2}\right) . \quad (6)$$

The $\cos^2\psi$ in Eq. (6) can be expressed in terms of momentum variables only (Fig. 1):

$$\cos^2\psi = \frac{(k^2 - k'^2 + K^2)^2}{4k^2K^2} = \frac{[E + (m/M)Q]^2}{4TQ} ,$$

where M is the reduced mass of the colliding system (see Secs. 2.1 and 2.2 of I71). Thus we have

$$\int_{Q_{\rm min}}^{Q_0} dQ\; Q^{-1}\cos^2\psi = 1 + \frac{mE}{2MT}\ln\left(\frac{4TQ_0}{E^2}\right) + O(T^{-1}) . \quad (7)$$

The right-hand side (rhs) of Eq. (7) remains finite for large T. The logarithmic term in Eq. (7) is *not* what we are after, because it gives a $(\ln T)/T^2$ [but not a $(\ln T)/T$] factor when substituted into Eq. (1).

The remainder of the rhs of Eq. (6) is independent of Q and it can be factored out of the Q integral in Eq. (5). *The resulting integral gives the* $\ln T$ *dependence we are looking for,* namely,

$$\int_{Q_{\rm min}}^{Q_0} dQ \int_0^{2\pi} d\Phi\, f_{E\Omega}(\vec{K})/Q \cong 2\pi\bar{f}_{EW}(\theta)\ln(4TQ_0/E^2) + [\text{contributions from Eq. (7)}]$$
$$= 2\pi\bar{f}_{EW}(\theta)\ln(T/\mathcal{R}) + (\text{terms finite for large } T) , \quad (8)$$

where

$$\bar{f}_{EW}(\theta) = (f_{EW}/4\pi)\left[1 - \tfrac{1}{2}\beta_{EW}P_2(\cos\theta)\right] . \quad (9)$$

Notice that Eq. (9) is the same formula that gives the angular distribution of electrons ejected by unpolarized light propagating parallel to $\vec{k}$. In fact, Eq. (9) comes from the component $\vec{K}_\perp$ of $\vec{K}$ perpendicular to $\vec{k}$, as can be verified by putting $\cos\lambda = (\vec{\kappa}\cdot\vec{K}_\perp)/\kappa K_\perp$ in Eq. (6) (see Fig. 1). The integration over Φ then corresponds to the summation over the direction of polarization of light propagating parallel to $\vec{k}$. The unit of energy used in separating the $\ln(T/\mathcal{R})$ term on the rhs of Eq. (8) is arbitrary, and we chose $\mathcal{R}$ only for convenience. We combine the terms in Eq. (8) that remain finite for large T together with $B'_{E\Omega}$ defined by Eq. (4), thus eliminating the dependence on the arbitrary constant Q_0. We shall denote the combined terms by $B_{EW}(T,\theta)$. Details of $B_{EW}(T,\theta)$ are again irrelevant for our purposes except for the fact that it remains finite for large T.

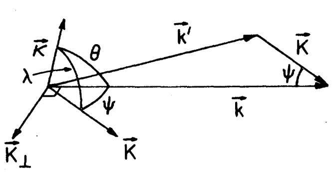

FIG. 1. Relations between various momenta in units of $\hbar$. The vectors $\vec{k}$ and $\vec{k}'$ are the momenta for the primary particle before and after collision, respectively, $\vec{K}$ is the momentum transferred to the target, $\vec{K}_\perp$ is the component of $\vec{K}$ perpendicular to $\vec{k}$, and $\vec{\kappa}$ is the momentum of the ejected electron. Note that all vectors except for $\vec{\kappa}$ are on the same plane.

Now, we have our final result by putting Eqs. (1), (4), (5), (8), and (9) together:

$$\sigma_{EW}(T,\theta) = \int_{Q=Q_{\rm min}}^{Q_0}\int_{\Phi=0}^{2\pi} d\sigma_{E\Omega}$$
$$= \frac{4\pi a_0^2 z^2}{T/\mathcal{R}}\left[A_{EW}(\theta)\ln\left(\frac{T}{\mathcal{R}}\right) + B_{EW}(T,\theta)\right] , \quad (10)$$

where

$$A_{EW}(\theta) = \bar{f}_{EW}(\theta)\,\mathcal{R}/E . \quad (11)$$

Equations (9)–(11) show that the angular dependence of the dipole term for secondary electrons, which appears with the $(\ln T)/T$ factor in the cross section, is related to the angular distribution of photoelectrons in a simple manner. The parameter $A_{EW}(\theta)$ is the analog of M_n^2 that appears as a coefficient of $\ln(T/\mathcal{R})$ in various integrated Born cross sections [see, for instance, Eqs. (2.11), (4.18), and (4.26) of I71].

When the energy $E_{\rm ion}$ transferred to the ion is undetermined, as in most experiments, the cross section must be summed over alternative final states of the ion. The values of $E_{\rm ion}$ (ionization potentials included) are usually discrete corresponding to discrete states of the ion, but $E_{\rm ion}$ becomes continuous for multiply ionized states. Thus *the angular distribution of the secondary electrons of a given energy* W *is*

$$\sigma(T,W,\theta) = \frac{4\pi a_0^2 z^2}{T/\mathcal{R}}\sum_{E_{\rm ion}}\left[A_{EW}(\theta)\ln\left(\frac{T}{\mathcal{R}}\right) + B_{EW}(T,\theta)\right] , \quad (12)$$

where $E_{\text{ion}} = E - W$, and θ is the angle between the direction of the secondary electron and that of the incident particle.

The asymmetry parameter β_{EW} in Eqs. (5) and (8) has been discussed frequently in the literature.[2,3] In principle, β_{EW} can be evaluated if the wave functions are known,[13,15] or it can be deduced from photoelectron experiments.[1-3] When β_{EW} is known, the integrated dipole oscillator strength f_{EW} may be deduced from secondary-electron experiments via Eq. (11) and compared with those obtained from photoabsorption experiment or from theory. Equation (11) relates three quantities, namely, $A_{EW}(\theta)$, f_{EW}, and β_{EW}, each of which can be deduced from experiments conducted independently of each other.

III. APPLICATIONS

The present theory is applicable when the Born approximation is valid. Furthermore, for practical reasons, the dipole interaction should be strong compared to the nondipole part. These conditions are usually satisfied when $E/T \ll 1$, that is, when small momentum transfers dominate. For instance, slow electrons of a few Ry in energy ejected from valence shells of atoms or molecules by incident electrons of several keV might satisfy both conditions.

Experimental data at a given angle may be plotted as a function of T in the usual Fano plot[11,12] (σT vs $\ln T$). The plot should approach a straight-line asymptotic behavior as T increases when the above conditions are met. The slope of the straight line is $\sum_{E_{\text{ion}}} A_{EW}(\theta)$ in Eq. (12), apart from a trivial constant. When the dipole interaction is weak, the slope will be too small (i.e., f_{EW} is too small) to make any meaningful comparison between theory and experiment.

The angular distribution given by Eq. (9) is symmetric with respect to $\theta = 90°$ direction, and hence,

$$A_{EW}(90° - \theta) = A_{EW}(90° + \theta) . \qquad (13)$$

This symmetry still holds after the summation over E_{ion}, and it can be used to check the consistency of secondary-electron data, whether absolute or relative, at different angles, without referring to any external data.

For the ionization of He, $\beta_{EW} = 2$ for all E so long as the He^+ ion is left in an s state.[13] Accurate values of β_{EW}, when the He^+ ion is left in $l \geq 1$ states, are not known. However, with the values of f_{EW} calculated from elaborate wave functions by Jacobs,[16] it is estimated that the excitations leaving the ion in excited states would contribute only a few percent to $\sum_{E_{\text{ion}}} A_{EW}(\theta)$. This estimate also agrees with that by Oldham and Miller.[17]

In Figs. 2(a)–2(d), we compare the theoretical slopes with the electron-impact data on He by Opal, Beaty, and Peterson,[6] for $W = 26.9$ eV. Because of the symmetry relation (13), the asymptotic slopes at supplementary angles (30° and 150°, and 60° and 120°, etc.) should be the same. The theoretical slopes were obtained using $\beta = 2$ (which gives the angular dependence of $\frac{3}{2}\sin^2\theta$) and $f_{EW} = 0.237$ for $E = 3.79\text{R}$ with the ion in the ground state.[16] Excited ion states are not included in the theoretical slopes shown. Inclusion of such states would increase the slopes by a few percent at the most. The experimental data for backward scattering seem to be more consistent with the theory than those for $\theta < 90°$.

It is difficult to deduce the slopes from the experimental data of Ref. 6 owing to a large uncertainty (~25%) in the absolute cross section. In principle, however, Eq. (9) may be used along with the Fano plot to determine f_{EW} and β_{EW}, if secondary-electron angular distributions are available with good statistics at various angles and for

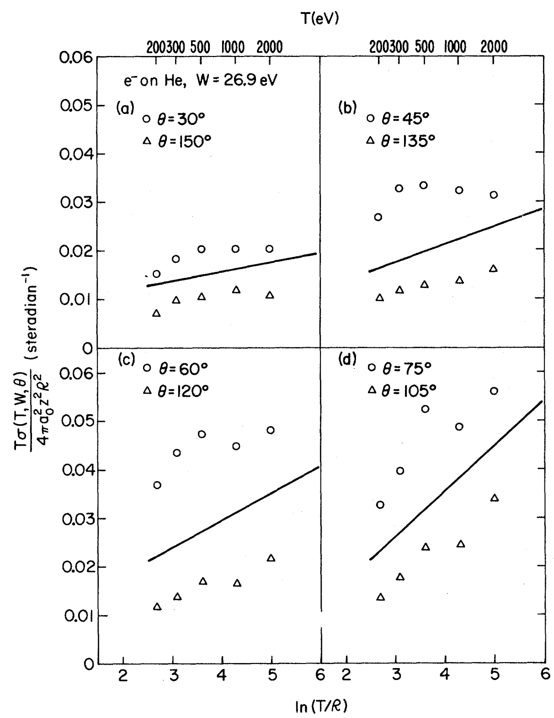

FIG. 2. Comparison of the slopes $A_{EW}(\theta)$ in the Fano plot ($T\sigma$ vs $\ln T$) for electron-impact data on He (Ref. 6). The circles and triangles are the experimental data, and the data for each pair of supplementary angles should approach the same asymptotic slope according to Eq. (13). The straight lines are the slopes based on the present theory. Theory predicts asymptotic slopes but not heights. Experimental uncertainty is ~25%, and the uncertainty in theoretical slopes is a few percent. The energies of primary and secondary electrons are denoted by T and W, respectively. For other notations, see the text.

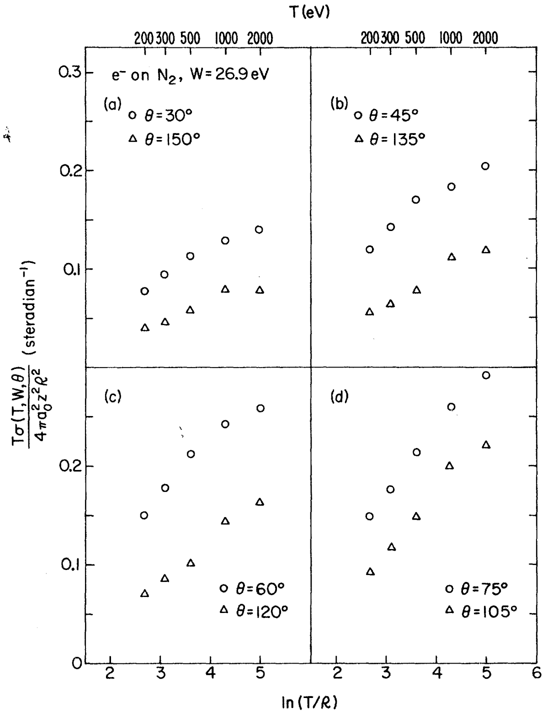

FIG. 3. The symmetry relation [Eq. (13)] for the slopes observed in the electron-impact data on N_2 (Ref. 6). Notations are the same as those in Fig. 2.

high enough incident energies. Equation (9) can also be used to normalize secondary-electron data when relevant f_{EW} and β_{EW} are accurately known. At the "magic" angle [where $P_2(\cos\theta)=0$], the β dependence in Eq. (9) is eliminated and the experimental data can be normalized from the appropriate sum of the optical oscillator strength only, that is,

$$\sum_{E_{\rm ion}} A_{EW}(\theta_M) = \sum_{E_{\rm ion}} f_{EW}\,\mathcal{R}/4\pi E$$

at $\theta_M = \cos^{-1}(1/\sqrt{3})$.

In Figs. 3(a)–3(d), we present the electron-impact data on N_2 also by Opal, Beaty, and Peterson.[6] In contrast to the He data in Figs. 2(a)–2(d), the asymptotic slopes of the N_2 data for the pairs of supplementary angles are remarkably similar as required by Eq. (13), though the actual cross sections are quite different at those angles.

For molecules, there are in general many ionic states (rotational, vibrational, and electronic) that are similar in $E_{\rm ion}$ but have widely different values of f_{EW} and β_{EW}.[2,3] Thus, the application of Eq. (12) to molecules requires more extensive data on photoelectrons.

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