molecule system $I=\eta(-1)^J$ is invariant of course. It follows then from (A9) that the parity $\bar{I}=\bar{\eta}(-1)^{\bar{J}}$ of the molecule alone is a collision invariant for homonuclear molecules. This means that in electron scattering by non-Σ homonuclear molecules, the rotational transition $\Delta\bar{J}=$ odd integer is *allowed*, but *only* with a simultaneous change of the quantum number $\bar{\eta}$. The occurrence of the $\Delta\bar{J}=$ odd-integer transition has been emphasized by Temkin and Faisal.[20]

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†Present address.

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Angular Momentum Transfer in the Theory of Angular Distributions*

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The angular distribution of collision products is expressed as a sum of incoherent contributions corresponding to different magnitudes of the angular momentum $\vec{j}_t$ transferred to an unpolarized target. For targets with a characteristic internal reference frame (e.g., molecules) the coefficients of this sum are interpreted as generalized 2^t-pole polarizabilities of the target in its internal reference frame, analogous to the scalar and quadrupole polarizabilities that determine the Raman effect. The theory is developed in the context of photoionization, but is applicable to more general collision processes as well. It is illustrated by use and extension of diagrammatic techniques.

Theoretical expressions of the angular distribution of radiations from single-collision (or decay) processes have repeatedly been given in terms of the angular momentum $\vec{j}_t$ transferred from one reactant to another.[1] In view of the increasing role of angular-distribution studies in atomic and mo-

lecular physics, it seems worthwhile to identify in what respects and by what procedures the theory may be usefully developed in terms of $\vec{j}_t$. The results of this investigation appear equally relevant to nuclear physics.

It will be shown that use of $\vec{j}_t$ plays two separate roles. First it simplifies the averaging over magnetic quantum numbers of reactants whose orientation is not controlled or observed. A subsidiary advantage is gained here in the frequent case that a single value of $|\vec{j}_t|^2$ is compatible with triangular conditions. In this role the use of $\vec{j}_t$ complements but need not replace altogether a consideration of the total angular momentum $\vec{J}$ of the system.

A second and more essential role is played by $\vec{j}_t$ in processes whose dynamics is properly treated in a rotating (or body-fixed) frame, e.g., in molecular processes or in processes involving nonspherical nuclei. Here use of $\vec{j}_t$ permits one to eliminate $\vec{J}$ altogether. Irreducible body-fixed parameters of a collision, such as scalar and quadrupole polarizabilities, are classified by values of the quantum number j_t.

These two aspects are treated separately in the following sections. For the sake of definiteness we will refer to the photoionization process

$$h\nu(j_r=1)+\mathrm{H}_2(^1\Sigma_g^+,\ N)\rightarrow \mathrm{H}_2^+(^2\Sigma_g^+,\ N')+e(l=1)\ , \qquad (1)$$

which is treated in detail in another paper[2] and whose study motivated the present one. We shall keep the formal development as general as we can without undue complications, having in mind application to more general collision processes.

As a by-product this paper presents a rather straightforward derivation of the relevant angular-distribution formula, which has a broad range of application and is amenable to further generalization. This derivation should make it particularly evident that theorems of Yang[3] on angular distributions are wholly independent of assumptions on the target structure and geometry, provided only that the target is unpolarized.

The calculations of this paper are done analytically, as usual, but are complemented by diagrammatic analogs drawn in accordance with a recent article by Briggs,[4] which we have extended slightly. For qualitative purposes, the diagrams may give a visual impression of the topological relationships utilized in the analytical calculation. However, the set of diagrams may also be regarded as constituting an equivalent self-contained quantitative calculation. As a further illustration the Appendix outlines a derivation of the final result of Sec. I carried out in the spirit of the Briggs article.

I. LABORATORY-FRAME TREATMENT

The probability amplitude for a reaction exemplified by (1) may be represented by a scattering matrix element

$$(j_2 m_2,\ lm\,|S|\,j_1 m_1,\ j_r m_r)\ . \qquad (2)$$

Here $|j_r m_r)$ indicates the state of a radiation incident on a "target" system which is itself in the state $|j_1 m_1)$, however, the orientation of the target system has not been preselected and m_1 will have to be averaged. Similarly $(lm|$ represents the state of the collision product to be detected, while $(j_2 m_2|$ indicates the state of the "residue" whose orientation remains unobserved, eventual summation over m_2 being required. In process (1), N and N' correspond to j_1 and j_2, respectively; the photoelectron's spin, which remains unobserved, can be regarded as still coupled in a singlet state with the electron of the H_2^+ residue.[2]

This aspect of process (1) raises a point of general significance for simplifying the treatment of the matrix element (2). We define the states $|j_1 m_1)$ and $(j_2 m_2|$ so as to include in them all elements of the process whose orientation is not controlled or observed, e.g., the photoelectron's spin in process (1). Should the matrix element (2) be introduced initially in a different form, its angular momenta can be recoupled to meet our definition. We shall deal explicitly only with collision processes whose matrix elements can take the form (2), but we have also in mind equivalent double-decay processes.

The scattering-matrix element (2) serves to calculate the differential cross section for emission of particles in a direction (θ, ϕ). The probability amplitude for this emission is proportional to

$$\sum_{lm} Y_{lm}(\theta,\phi)(j_2 m_2,\ lm\,|S|\,j_1 m_1, j_r m_r)\ , \qquad (3)$$

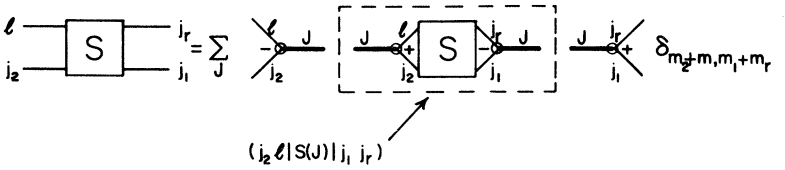
where Y_{lm} is a spherical harmonic. The cross section is proportional to the squared modulus of (3) averaged over m_1 and summed over m_2. The essence of the angular distribution theory lies in performing the sums over m quantum numbers and in expanding the result into a sum of spherical harmonics. The proportionality coefficient required to complete the cross-section formula will be introduced at the end of the calculation.

We begin by working out the dependence of the transition-matrix element [Eq. (2)] upon the quantum numbers m. Invariance of collision processes under space rotations suggests that (2) be expanded into components corresponding to different magnitudes of the total angular momentum

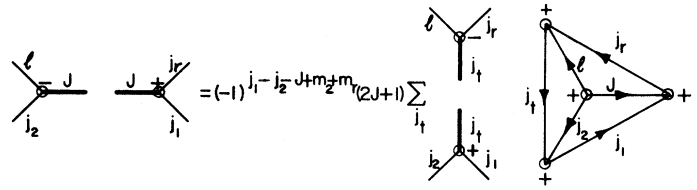
$$\vec{J}=\vec{j}_1+\vec{j}_r=\vec{j}_2+\vec{l}\ . \qquad (4)$$

Thus we write, in terms of Wigner coefficients,

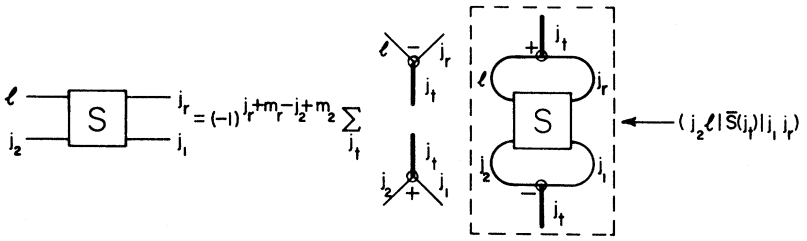
$$(j_2 m_2,\ lm\,|S|\,j_1 m_1,\ j_r m_r)=\sum_J (j_2 m_2,\ lm\,|\,JM)$$



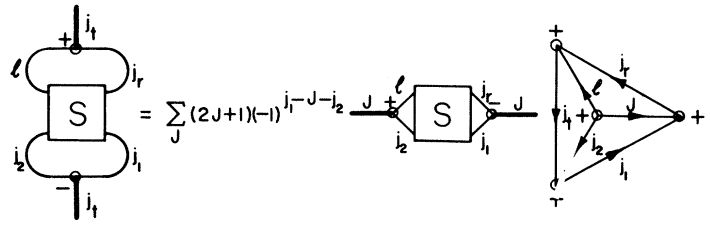
(a)



(b)



(c)



(d)

FIG. 1(a). Diagrammatic representation of Eq. (5). See Eq. (4.11) and (4.14) of Ref. 4. The minus (or plus) sign on the nodes of the diagrams indicates that the momenta corresponding to the lines emerging from the nodes are coupled in clockwise (or counterclockwise) order. (b) Representation of Eq. (7); 6-j symbol as in (4.20) of Ref. 4. (c) Representation of Eq. (8). (d) Representation of $(j_2 l|\bar{S}(j_t)|j_1 j_r)$, Eq. (9).

$$\times(j_2 l \mid S(J) \mid j_1 j_r)(JM \mid j_1 m_1, j_r m_r)\,\delta_{m_2+m,\,m_1+m_r}\,, \tag{5}$$

where $(j_2 l \mid S(J) \mid j_1 j_r)$ indicates a submatrix of S which is rotationally invariant and hence independent of M. Figure 1(a) illustrates this expansion using the representation of Briggs.[4]

Consider now the angular momentum transferred to the unobserved reactants (to molecular rotation in our example):

$$\vec{j}_t = \vec{j}_2 - \vec{j}_1 = \vec{j}_r - \vec{l}\,. \tag{6}$$

This momentum can be introduced in (5) by a recoupling transformation with the intent of joining together on the one hand the angular momenta $\vec{j}_1$ and $\vec{j}_2$ whose orientation is not observed and on the other hand the momenta $\vec{j}_r$ and $\vec{l}$ which pertain to observed variables. The transformation is represented analytically by[5]

$$\begin{aligned}(j_2 m_2,\ lm \mid JM)(JM \mid j_1 m_1,\ j_r m_r) &= (-1)^{j_1 - j_2 - J + m_2 + m_r}(2J+1) \\ &\times \sum_{j_t}(j_2 - m_2, j_1 m_1 \mid j_t\, m_1 - m_2) \\ &\times (j_t\, m_2 - m_1 \mid l - m,\ j_r m_r) \begin{Bmatrix} l & j_2 & J \\ j_1 & j_r & j_t \end{Bmatrix}\,, \end{aligned} \tag{7}$$

and diagrammatically by Fig. 1(b). Substitution into (5) gives

$$(j_2 m_2,\ lm \mid S \mid j_1 m_1, j_r m_r)$$

$$= (-1)^{j_r+m_r-j_2+m_2}$$
$$\times \sum_{j_t} (j_2 - m_2, j_1 m_1 \,|\, j_t m_1 - m_2)$$
$$\times (j_t m_2 - m_1 \,|\, l - m, j_r m_r)$$
$$\times (j_2 l \,|\, \bar{S}(j_t) \,|\, j_1 j_r)\, \delta_{m_2+m, m_1+m_r}, \quad (8)$$

where [Fig. 1(c)]

$$(j_2 l \,|\, \bar{S}(j_t) \,|\, j_1 j_r)$$
$$= \sum_J (-1)^{j_1-J-j_r} (2J+1) \begin{Bmatrix} l & j_2 & J \\ j_1 & j_r & j_t \end{Bmatrix} (j_2 l \,|\, S(J) \,|\, j_1 j_r) \quad (9)$$

is a new type of invariant matrix represented in Fig. 1(d). This matrix will be discussed in Sec. II.

The square of the probability amplitude (3) contains products of the matrix elements of S and of its Hermitian conjugate

$$(j_2 m_2, lm \,|\, S \,|\, j_1 m_1, j_r m_r)$$
$$\times (j_1 m_1, j_r m_r \,|\, S^\dagger \,|\, j_2 m_2, l' m'). \quad (10)$$

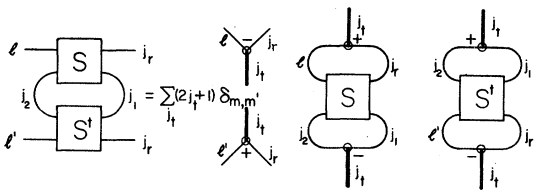


FIG. 2. Representation of Eq. (11). The sums over m_1 and m_2 are indicated by the joined j_1 and j_2 lines as in (4.15) of Ref. 4.

In this expression the primed indices allow for the occurrence of cross terms with different indices (lm) and $(l'm')$; cross terms with $(j_r' m_r') \neq (j_r m_r)$ are excluded from this paper for simplicity. Cross terms pertaining to unobserved variables are excluded by the problem formulation, that is, an essential step consists of setting $j_1 = j_1'$, etc., and summing over m_1 and m_2. It is here that the recoupling transformation (7) pays off, since the orthonormality of Wigner coefficients gives (Fig. 2)

$$\sum_{m_1} \sum_{m_2} (j_2 m_2, l m \,|\, S \,|\, j_1 m_1, j_r m_r)(j_1 m_1, j_r m_r \,|\, S^\dagger \,|\, j_2 m_2, l' m')$$
$$= \sum_{j_t} (j_t m_r - m \,|\, l - m, j_r m_r)(j_2 l \,|\, \bar{S}(j_t) \,|\, j_1 j_r)(j_1 j_r \,|\, \bar{S}^\dagger(j_t) \,|\, j_2 l')(l' - m', j_r m_r \,|\, j_t m_r - m')\, \delta_{m,m'}. \quad (11)$$

Had we used the expansion (5) of the scattering matrix, in terms of J, the result (11) would be represented by a double sum over J and J' with cross terms that are essential to the angular distribution, whereas we have in (11) a single "incoherent" sum over j_t. The sums over J and J' are actually hidden in the definitions of $\bar{S}(j_t)$ and $\bar{S}^\dagger(j_t)$. As anticipated, the sum over j_t may reduce to a single term owing to triangular relations, for example, when j_2 takes the largest or the smallest value consistent with conservation of angular momentum. On the other hand, the j_t expansion may be wasteful when a single value of J contributes to the transition, as for example in resonance phenomena.

To obtain the cross section expanded in spherical harmonics, we expand the products of harmonics $Y_{lm}(\theta, \phi) Y^*_{l'm'}(\theta, \phi)$ in the squared modulus of (3). However, we need only the formula for $m' = m$ owing to the coefficient $\delta_{m,m'}$ in (11). The expansion formula is[6]

$$Y_{lm}(\theta, \phi) Y^*_{l'm}(\theta, \phi) = (-1)^m \frac{(2l+1)^{1/2}(2l'+1)^{1/2}}{4\pi}$$
$$\times \sum_k (l0, l'0 \,|\, k0)\, P_k(\cos\theta)\, (k0 \,|\, lm, l' - m), \quad (12)$$

and is illustrated in Fig. 3. Upon substitution of this formula and of (11) in the squared modulus of (3), there remains a single sum over m quantum numbers which is carried out by a formula of Racah algebra,[7]

$$\sum_m (-1)^m (k0 \,|\, lm, l' - m)(j_t m_r - m \,|\, l - m, j_r m_r)$$
$$\times (l' - m, j_r m_r \,|\, j_t m_r - m) = (-1)^{j_t+l+l'+m_r} (2j_t + 1)$$
$$\times \begin{Bmatrix} j_r & j_r & k \\ l & l' & j_t \end{Bmatrix} (k0 \,|\, j_r m_r, j_r - m_r), \quad (13)$$

illustrated in Fig. 4.

Combination of the results (11)–(13) yields the desired differential cross section to within a proportionality coefficient. To determine this coefficient we must specify the normalization of the continuum states $|\, j_r m_r)$ and $(lm\,|$ which form the basis set for the scattering matrix. These states are assumed to satisfy the so-called "outgoing-" and "incoming-wave" boundary conditions, respectively, and to represent a current of one particle (or photon) per unit time incident on, or emitted by, the target. The cross section equals then[8]

$$\frac{d\sigma}{d\omega} = \pi(2j_r+1)\lambda\!\!\!^{-}_r{}^2(2j_1+1)^{-1}\sum_{m_1}\sum_{m_2}\left|\sum_{lm} Y_{lm}(\theta,\phi)\,(j_2 m_2, lm \mid S \mid j_1 m_1, j_r m_r)\right|^2$$

$$= \pi(2j_r+1)\lambda\!\!\!^{-}_r{}^2(2j_1+1)^{-1}\sum_l\sum_{l'}\sum_{j_t}(j_2 l \mid \bar{S}(j_t) \mid j_1 j_r)(j_1 j_r \mid \bar{S}(j_t)^\dagger \mid j_2 l')\,\Theta(j_t; j_r m_r, l l'; \theta)\,, \tag{14}$$

where $\lambda\!\!\!^{-}_r$ is the wavelength of the incident radiation (divided by 2π), and where we have introduced a function

$$\Theta(j_t; j_r m_r, l l'; \theta)$$

$$= (-1)^{j_t+m_r}\frac{(2l+1)^{1/2}(2l'+1)^{1/2}}{4\pi}(2j_t+1)$$

$$\times\sum_k \begin{Bmatrix} j_r & j_r & k \\ l & l' & j_t \end{Bmatrix}(l0, l'0 \mid k0)P_k(\cos\theta)$$

$$\times(k0 \mid j_r m_r, j_r - m_r)\,, \tag{15}$$

which depends only on j_t and on the quantum numbers of the observed radiations.

Note that the function $\Theta(\theta)$ is simple for the low values of quantum numbers which are often relevant, in spite of the complication of its general form (15). In the example of the reaction (1) for which $j_r = l = l' = 1$ and $m_r = 0$ (linear polarization along z), only three values of the quantum number j_t are compatible with triangular conditions, and we have

$$\Theta(0;\theta) = (1/12\pi)[1+2P_2(\cos\theta)] = (1/4\pi)\cos^2\theta\,,$$

$$\Theta(1;\theta) = (1/4\pi)[1-P_2(\cos\theta)]$$

$$= (3/8\pi)(1-\cos^2\theta)\,, \tag{16}$$

$$\Theta(2;\theta) = (5/12\pi)[1+\tfrac{1}{5}P_2(\cos\theta)]$$

$$= (3/8\pi)(1+\tfrac{1}{3}\cos^2\theta)\,.$$

Note finally that the angular distribution functions Θ include only finite sets of spherical harmonics P_k, limited by the triangular and symmetry conditions under which the Wigner coefficients are nonzero. These conditions imply the Yang theorems on angular distributions.[3] The Wigner coefficients in (15) depend only on quantum numbers of the incident and emitted radiations, not on those of the unobserved target. Nonzero values of the $6j$ coefficient in (15) imply additional triangular conditions which restrict the values of j_t.

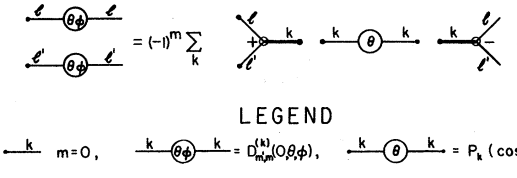


FIG. 3. Representation of Eq. (12); spherical harmonics are normalized here as Legendre polynomials.

II. TRANSITION MATRIX IN BODY FRAME

Molecules, as well as most nuclei, have structural properties which are brought out by treating their reactions in a body-fixed coordinate frame. For simplicity we consider here only collisions involving a diatomic molecule with initial and final rotational states $|j_1 m_1)$ and $(j_2 m_2|$. These states are also characterized by quantum numbers λ which indicate eigenvalues of the angular momentum about the internuclear axis. These quantum numbers are usually taken to be non-negative—in which case they correspond to the label Σ, Π, etc., —but they must then be supplemented by an additional quantum number (plus or minus) which identifies the parity under reflection on planes containing the internuclear axis. Here we regard λ as positive *or* negative; thereby we achieve a formal simplification but fail to establish selection rules based on parity, as will be explained further below.

The rotational part of the initial molecular wave function can be represented explicitly by writing

$$|\lambda_1 j_1 m_1) = |\lambda_1)\,D^{(j_1)}_{\lambda_1 m_1}(0,\theta,\phi)\,[(2j_1+1)/4\pi]^{1/2}\,, \tag{17}$$

where $|\lambda_1)$ indicates a state of the electronic and vibrational motion in the body frame, (θ,ϕ) are now coordinates of the internuclear axis, the third Euler angle χ of the body frame being set at zero. D indicates a wave function of the symmetric top which serves also as a matrix to transform ket states from the body frame to the lab frame. A formula analogous to (17) holds for the final state $(\lambda_2 j_2 m_2|$. The state $|j_r m_r)$ of the incident radiation can be expanded into body-frame states by a unitary transformation formula which is also analogous to (17) but includes a summation and has a different normalization

$$|j_r m_r) = \sum_{\lambda_r}|j_r \lambda_r)\,D^{(j_r)}_{\lambda_r m_r}(0,\theta,\phi)\,. \tag{18}$$

The state $(lm|$ of the collision product to be detected is represented by an expansion analogous to (18). [Note that the body-frame states $|\lambda_1)$ and $(\lambda_2|$ are treated as independent of j_1 and j_2, in accordance with the Born-Oppenheimer approximation, but that the quantum numbers j_r and l are relevant to interactions in the body frame.]

Substitution of (17), (18), and the analogous formulas into an expression of the scattering-matrix element (2) permits explicit integration over the

rotational coordinates (θ, ϕ). The lab-frame matrix element (2) is thus expressed in terms of a body-frame matrix element $(\lambda_2, l\lambda \mid S \mid \lambda_1, j_r\lambda_r)$ whose separate evaluation need not concern us,

$$(\lambda_2 j_2 m_2, lm \mid S \mid \lambda_1 j_1 m_1, j_r m_r) = \frac{[(2j_2+1)(2j_1+1)]^{1/2}}{4\pi} \int_0^{2\pi} d\phi \int_0^{\pi} \sin\theta\, d\theta\, [D^{(j_2)}_{\lambda_2 m_2}(0,\theta,\phi)]^* \times \sum_{\lambda,\lambda_r} [D^{(l)}_{\lambda m}(0,\theta,\phi)]^* (\lambda_2, l\lambda \mid S \mid \lambda_1, j_r\lambda_r) D^{(j_1)}_{\lambda_1 m_1}(0,\theta,\phi)\, D^{(j_r)}_{\lambda_r m_r}(0,\theta,\phi). \quad (19)$$

To carry out the integration over (θ, ϕ) one may sort out the product of four D functions into two products of two factors each, and then expand each product of two D into a superposition of single D factors. After this "reduction" procedure, the integral is given by the orthonormality of the D functions. The point of interest to us lies in the alternative ways of sorting out the four D factors of (19) into two pairs [Ref. 1(c)]. If $D^{(j_1)}$ and $D^{(j_r)}$ are paired, one obtains an expansion of

$$(\lambda_1 j_1 m_1, lm \mid S \mid \lambda_2 j_2 m_2, j_r m_r)$$

into contributions classified by the quantum number J of the combined system, as in (5); if $D^{(j_1)}$ and $(D^{(j_2)})^*$ are paired, one obtains an expansion into contributions with the quantum number j_t, as in (8). The two expansions are

$$(\lambda_2 j_2 m_2, lm \mid S \mid \lambda_1 j_1 m_1, j_r m_r) = \sum_J (j_2 m_2, lm \mid J\, m_2+m) \times \left[\sum_{\lambda,\lambda_r} (-1)^{2l-2j_r} (2j_2+1)^{1/2} (2j_1+1)^{1/2} \begin{pmatrix} j_2 & l & J \\ \lambda_2 & \lambda & -\lambda_2-\lambda \end{pmatrix} \begin{pmatrix} j_1 & j_r & J \\ \lambda_1 & \lambda_r & -\lambda_1-\lambda_r \end{pmatrix} \times (\lambda_2, l\lambda \mid S \mid \lambda_1, j_r\lambda_r)\delta_{\lambda_2+\lambda,\lambda_1+\lambda_r}\right] (J\, m_1+m_r \mid j_1 m_1, j_r m_r)\, \delta_{m_2+m, m_1+m_r}, \quad (20)$$

$$(\lambda_2 j_2 m_2, lm \mid S \mid \lambda_1 j_1 m_1, j_r m_r) = \sum_{j_t} (j_2 - m_2, j_1 m_1 \mid j_t\, m_1 - m_2) \times (j_t\, m - m_r \mid lm, j_r - m_r)\, \delta_{m_1-m_2, m-m_r} \left[\sum_{\lambda,\lambda_r} (-1)^{\lambda_r-\lambda_2+m_2-m_r+2j_1-2j_r} (2j_2+1)^{1/2} (2j_1+1)^{1/2} \times \begin{pmatrix} j_2 & j_1 & j_t \\ -\lambda_2 & \lambda_1 & \lambda_2-\lambda_1 \end{pmatrix} \begin{pmatrix} l & j_r & j_t \\ \lambda & -\lambda_r & \lambda_r-\lambda \end{pmatrix} (\lambda_2, l\lambda \mid S \mid \lambda_1, j_r\lambda_r)\delta_{\lambda-\lambda_r, \lambda_1-\lambda_2}\right]. \quad (21)$$

Comparison with (5) shows that the expression in the brackets of (20) represents $(\lambda_2 j_2 l \mid S(J) \mid \lambda_1 j_1 j_r)$; similarly comparison with (8) shows that the expression in the parenthesis of (21) coincides with $(\lambda_2 j_2 l \mid \bar{S}(j_t) \mid \lambda_1 j_1 j_r)$ to within a phase factor. Thus we find the initial result that the matrix of $\bar{S}(j_t)$, to be used in the cross-section formula (14), is obtained directly from the body-frame matrix S. This avoids the by pass through $S(J)$ which is required by the defining Eq. (9). More specifically, the matrix of $\bar{S}(j_t)$ is defined in terms of the body-frame matrix by

$$(\lambda_2 j_2 l \mid \bar{S}(j_t) \mid \lambda_1 j_1 j_r) = (-1)^{2j_1+j_2+j_r} (2j_2+1)^{1/2} (2j_1+1)^{1/2} \sum_{\lambda,\lambda_r} (-1)^{\lambda_r-\lambda_2} \times \begin{pmatrix} j_2 & j_1 & j_t \\ -\lambda_2 & \lambda_1 & \lambda_2-\lambda_1 \end{pmatrix} \begin{pmatrix} l & j_r & j_t \\ \lambda & -\lambda_r & \lambda_r-\lambda \end{pmatrix} \times (\lambda_2, l\lambda \mid S \mid \lambda_1, j_r\lambda_r)\, \delta_{\lambda-\lambda_r, \lambda_1-\lambda_2}. \quad (22)$$

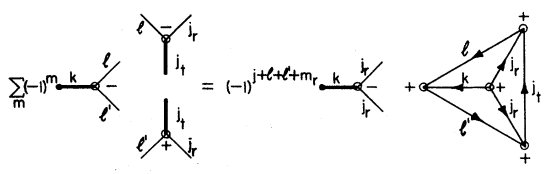


FIG. 4. Representation of Eq. (13).

The same formula can be obtained by extracting the matrix of $S(J)$ from (20) and substituting it in the definition (9) of $\bar{S}(j_t)$, after which the sum over J can be carried out analytically.

A more important result is that the dependence of (22) on the quantum numbers j_1 and j_2 of the molecule factors out. Thus the matrix of $\bar{S}(j_t)$ can be

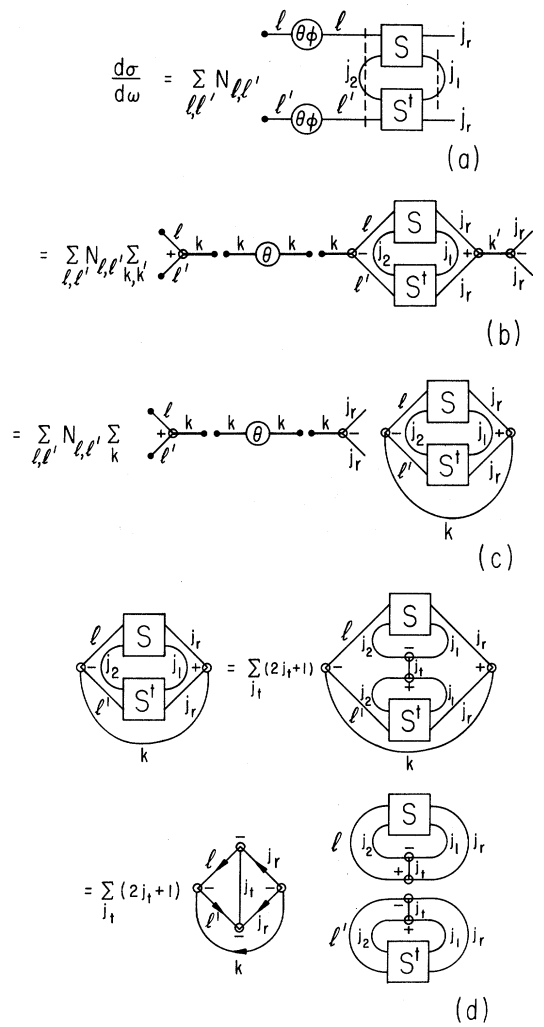


FIG. 5. Outline of complete diagrammatic calculation.

expressed in terms of an intrinsic body-frame operator $M(l j_r; j_t)$, which is independent of molecular rotation

$$(j_2 l \,|\, \bar{S}(j_t) \,|\, j_1 j_r) = (-1)^{2j_1+j_2-\lambda_2}(2j_2+1)^{1/2}(2j_1+1)^{1/2} \times \begin{pmatrix} j_2 & j_1 & j_t \\ -\lambda_2 & \lambda_1 & \lambda_2-\lambda_1 \end{pmatrix} (\lambda_2 | M(l, j_r; j_t) | \lambda_1), \quad (23)$$

where

$$(\lambda_2 | M(l, j_r; j_t) | \lambda_1) = \sum_{\lambda, \lambda_r} (-1)^{j_r+\lambda_r} \times \begin{pmatrix} l & j_r & j_t \\ \lambda & -\lambda_r & \lambda_r-\lambda \end{pmatrix} (\lambda_2, l\lambda \,|\, S \,|\, \lambda_1, j_r \lambda_r) \delta_{\lambda-\lambda_r, \lambda_1-\lambda_2}. \quad (24)$$

An example of this type of parameter occurs in the theory of rotational Raman scattering by molecules.[9] In this case we have $\lambda_2 = \lambda_1$ representing the electronic Λ (or Ω) quantum number of the molecule and $l = j_r = 1$ representing the dipole character of the incident and scattered light. Two values of j_t occur usually, namely, $j_t = 0$ and 2; the corresponding values of $|M|^2$ are proportional, respectively, to the scalar and quadrupole polarizabilities of the molecule. Equation (24) may be regarded as a generalization of the polarizability formulas to processes in which the incident and the emitted radiation may be different, with arbitrary multipolarity, and which need not be diagonal in λ_1 and λ_2.

Note finally that the factoring of the dependence of $\bar{S}(j_t)$ on j_1 and j_2 is responsible for a well-known sum rule, which gives a total angular distribution in terms of body-frame parameters only, independently of rotational quantum numbers. Substituting (23) into (14) we find that the cross section depends on the target momenta j_1 and j_2 only through the weighting factor[10]

$$(2j_2+1) \begin{pmatrix} j_2 & j_1 & j_t \\ -\lambda_2 & \lambda_1 & \lambda_2-\lambda_1 \end{pmatrix}^2, \quad (25)$$

which when summed over j_2 equals unity. Thus one obtains the cross section which is observed when one fails to resolve the contributions of the alternative final states of a "rotational band", namely.

$$\sum_{j_2} \frac{d\sigma}{d\omega} = \pi(2j_r+1)\lambda_r^2 \sum_{l, l', j_t} (\lambda_2 \,|\, M(l, j_r; j_t) \,|\, \lambda_1) \times (\lambda_1 \,|\, M(l', j_r; j_t)^\dagger \,|\, \lambda_2) \Theta(j_t; j_r m_r, l l'; \theta). \quad (26)$$

This is the result one obtains in a fixed-nuclei approximation.[11]

APPENDIX

The first equality in (14) constitutes a formal definition of $d\sigma/d\omega$ for the processes of interest, and the second equality gives the final form of $d\sigma/d\omega$, broken down into separate standard factors. Here we shall obtain the final form from the initial one, to within a phase factor, by the graphical procedure of Briggs.[4] The procedure consists of representing the initial definition of $d\sigma/d\omega$ by a diagram and then of breaking down this diagram into smaller ones by a sequence of standard operations. Since a complete procedure for keeping track of all phase factors graphically is not immediately available in our context we omit such factors and the corresponding graphical indications by arrows.

The initial formal definition $d\sigma/d\omega$ is represented by Fig. 5(a), which employs the spherical harmonics symbols introduced in Fig. 3 and the coefficient

$$N_{l,l'} = \pi(2j_r+1)\lambda_r^2(2j_1+1)^{-1}(2l+1)^{1/2}(2l'+1)^{1/2}/4\pi.$$

The summations over m quantum numbers are symbolized in Fig. 5(a) by the joining of full lines where

they cross the vertical dotted lines. The first step of the calculation consists of "pinching" together the pairs of lines (l, l') and (j_r, j_r), as (j_1, j_2) are pinched in Eq. (4.26) of Ref. 4. Note that the existence of open lines in Fig. 5(a) prevents application of (4.28) of Ref. 4, just as the existence of a dotted line does in (4.26) of Ref. 4. Figure 5(b) shows the result of the double pinching obtained using also our (12) and its representation in Fig. 3. Here and in the following we represent implicitly by the use of heavy lines some of the factors $[j]$ which appear explicitly in the Briggs equations.

At this point the central block of Fig. 5(b), which contains the scattering operators, is connected to the rest by two lines only. We can then apply (4.28) of Ref. 4 in which α represents our central block and α' all the rest; (k, k') correspond to (j_1, j_2). The result is represented by Fig. 5(c), where the closed diagram on the right has to be reduced further.

This final reduction is achieved in two steps as shown in Fig. 5(d). The first step consists of pinching the (j_1, j_2) lines into a single j_t line, applying (4.26) of Ref. 4. The second step consists of a twofold application of (4.30) of Ref. 4 in which one "pinches off" a set of three lines, namely, (j_r, j_t, l) in one case and (j_r, j_t, l') in the other where they intersect the dotted lines.

The resulting 6-j coefficient [e.g., (4.20) of Ref. 4] and $2j_t + 1$ factor are included in the function $\Theta(j_t; j_r m_r, l l'; \theta)$ with the other elements of the diagram. The two remaining blocks represent the $\bar{S}(j_t)$ and $\bar{S}^\dagger(j_t)$ invariant matrix elements on the right-hand side of (14).

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[1]See, e.g., (a) U. Fano, Nuovo Cimento 5, 1358 (1957); and Natl. Bur. Std. (U.S.) Tech. Note No. 83 (1960) (unpublished); (b) U. Fano and G. Racah, *Irreducible Tensorial Sets* (Academic, New York, 1959), p. 114; (c) E. S. Chang and A. Temkin, Phys. Rev. Letters 23, 399 (1969); J. Phys. Soc. Japan 29, 172 (1970). A quantum number j_t has also been introduced as a dummy variable without explicit physical meaning by (d) A. D. Buckingham, B. J. Orr, and J. M. Sichel, Phil. Trans. Roy. Soc. London A268, 147 (1970).

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[6]See, e.g., Ref. 5, Eq. (1.43).

[7]See, e.g., Ref. 5, Eq. (2.20).

[8]See, e.g., F. J. Blatt and V. Weisskopf, *Theoretical Nuclear Physics* (Wiley, New York, 1952), pp. 317–324 and 517–521. An incident-plane-wave state, representing the flux of one particle per second per unit area and expanded in spherical states $|j_r m_r\rangle$ with the values of m_r appropriate to its polarization, has expansion coefficients equal to $\pi^{1/2}(2j_r+1)^{1/2}\lambda_r$. That is, the ingoing flux of the j_rth component equals the flux $\pi(2j_r+1)\lambda_r^2$ which hits a ring target with outer radius $(j_r+1)\lambda_r$ and inner radius $j_r\lambda_r$.

[9]See, e.g., *Raman Spectroscopy, Theory and Practice*, edited by H. A. Szymanski (Plenum, New York, 1967), p. 10ff.

[10]The factors $Q(j \equiv j_t)$ given in Ref. 1(d) are specific examples of Eq. (25).

[11]For specific examples of the general result (26) see, e.g., Ref. 1(c) for the case of electron-molecule scattering, and also, e.g., J. C. Tully, R. S. Berry, and B. J. Dalton, Phys. Rev. 176, 95 (1968) for the case of molecular photoionization.

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Electron Excitation of the Sodium *D* Lines*

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The electron excitation of the sodium resonance lines (*D* lines) has been measured in the energy range from threshold to 1000 eV. The electron-beam full width at half-maximum was $\sim \frac{1}{3}$ eV, and the sodium-beam optical depth was small and varied. After correction for minor cascade contributions and the measured polarization, the excitation function has been normalized to the Born theory in a high-energy limit where the energy dependence converges to the theoretical behavior. The resulting normalized cross section and the polarization are in excellent agreement with recent close-coupling calculations for the energy region from threshold to 5 eV.

I. INTRODUCTION

Despite the relatively simple electronic structure of sodium, various theoretical calculations of the electron excitation cross section for the 3*s*-3*p* transition have differed considerably.[1] Pre-