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Angular Correlation of Gamma-Rays

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The calculation of the angular correlation of successive gamma-rays from an atomic nucleus, carried out by Hamilton, has been extended to take into account the effect of the magnetic field at the nucleus because of the extra-nuclear electrons. Results are given in a form valid for all multipole orders. In addition, it is shown that an external magnetic field may be used to reduce the effect of the extra-nuclear electrons on the angular correlation. A method of calculating the effect of an external magnetic field on the angular distribution of the successive gamma-rays is indicated.

INTRODUCTION

When an atomic nucleus emits two gamma-rays in succession, one might expect from general considerations of radiation theory that the direction of emission of the second gamma-ray is related in some way to the direction of propagation of the first. This correlation is expected to depend on the electric or magnetic multipole nature of the radiation and on the angular momentum of the nuclear levels involved. Some light may be shed on these factors by a comparison of experimentally determined angular distributions with the theoretical expectations.

The pioneer investigations of Hamilton[1] into the theory of the angular distribution of successive gamma-rays emitted by a nucleus were made with the restriction that torques due to fields external to the nucleus be sufficiently small to have a negligible effect on the angular correlation. Inasmuch as fields external to the nucleus, arising from the spin and orbital angular moments of the extra-nuclear electrons, may produce appreciable torques, it seems desirable to investigate the quantitative effect of fields external to the nucleus and to determine their influence on the angular distribution of the successive gamma-rays.

THEORY

Consider an atom which is visualized as existing in any one of three gross energy levels, A', B', and C'. Each of the gross levels consists of several sublevels which arise through the coupling of one of the nuclear energy levels A, B, and C, with the external electronic structure of the atom. That is, each gross level is composed of a set of hyperfine structure sublevels with separations very small by comparison with the separation of the gross levels.

The sublevels of the gross energy levels A' are indicated by α, α', α'', $\cdots$; those of B' by β, β', β'', $\cdots$; and those of C' by γ, γ', γ'', $\cdots$. If the sequence of transitions

1. from a level α to a level β with the emission of a gamma-ray ρ of frequency ν_ρ,
2. from the level β to a level γ with the emission of a gamma-ray σ of frequency ν_σ,

is assumed, the equations for the probability amplitudes become[2]

$$-i\hbar\dot{a}_\alpha=\sum_{\beta,\rho}(\alpha|H_\rho|\beta)b_{\beta,\rho}+h\nu_\alpha a_\alpha, \tag{1a}$$

$$-i\hbar\dot{b}_{\beta,\rho}=\sum_\alpha(\alpha|H_\rho|\beta)^*a_\alpha+\sum_{\gamma,\sigma}(\beta|H_\sigma|\gamma)c_{\gamma,\rho,\sigma}+h(\nu_\rho+\nu_\beta)b_{\beta,\rho}, \tag{1b}$$

$$-i\hbar\dot{c}_{\gamma,\rho,\sigma}=\sum_\beta(\beta|H_\sigma|\gamma)^*b_{\beta,\rho}+h(\nu_\rho+\nu_\sigma+\nu_\gamma)c_{\gamma,\rho,\sigma}, \tag{1c}$$

where $E_\alpha=h\nu_\alpha$, etc., and the zero of energy is chosen at any convenient value.

The following solutions are assumed, subject to determination of the constants involved by substitution into Eq. (1).

$$a_\alpha=\delta_\alpha\exp(-2\pi\Gamma_\alpha t), \tag{2a}$$

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[1] D. R. Hamilton, Phys. Rev. 58, 122 (1940).

[2] In setting up and solving Eq. (1), the procedure of reference 1 is followed closely. Cf. G. Breit, Rev. Mod. Phys. 5, 91 (1933).

$$b_{\beta,\rho}=\sum_\alpha \zeta_{\alpha,\beta,\rho}\{\exp(-2\pi\Gamma_\alpha t)-\exp(-2\pi\Gamma_{\beta,\rho}t)\},\quad (2b)$$

$$c_{\gamma,\rho,\sigma}=\sum_\alpha \epsilon_{\alpha,\gamma,\rho,\sigma}\{\exp(-2\pi\Gamma_\alpha t)-\exp(-2\pi\Gamma_{\gamma,\rho,\sigma}t)\}+\sum_\beta \eta_{\beta,\gamma,\rho,\sigma}\{\exp(-2\pi\Gamma_{\beta,\rho}t)-\exp(-2\pi\Gamma_{\gamma,\rho,\sigma}t)\}.\quad (2c)$$

Upon substitution of Eq. (2) into Eq. (1), one finds

$$\Gamma_\alpha=\gamma_A-i\nu_\alpha,\quad (3a)$$

$$\Gamma_{\beta,\rho}=\gamma_B-i(\nu_\rho+\nu_\beta),\quad (3b)$$

$$\Gamma_{\gamma,\rho,\sigma}=-i(\nu_\rho+\nu_\sigma+\nu_\gamma),\quad (3c)$$

where

$$h^2\gamma_A=\sum_{\beta,\rho}|(\alpha|H_\rho|\beta)|^2,\quad (4a)$$

$$h^2\gamma_B=\sum_{\gamma,\sigma}|(\beta|H_\sigma|\gamma)|^2.\quad (4b)$$

γ_A and γ_B are independent of the particular sublevels α or β of A' or B' from which they are computed. $\sum_{\beta,\rho}$ denotes the sum over all states β which can be reached by all perturbations H_ρ. One may deduce from Eqs. (2)–(4) that $4\pi\gamma_A$ is the total transition probability from any level α.

Also,

$$\lim_{t\to\infty} c_{\gamma,\rho,\sigma}=\sum_{\alpha,\beta}\frac{\delta_\alpha^*(\alpha|H_\rho|\beta)^*(\beta|H_\sigma|\gamma)^*\exp(i2\pi[\nu_\rho+\nu_\sigma-\nu_\alpha]t)}{(i\gamma_B+\nu_{\beta,\gamma}-\nu_\sigma)(i\gamma_A+\nu_{\alpha,\gamma}-\nu_\rho-\nu_\sigma)},\quad (5)$$

where $\nu_{\alpha,\beta}=\nu_\alpha-\nu_\beta$, etc. δ_α expresses the arbitrariness of the phases of the wave functions of the initial states, so that

$$\langle\delta_\alpha^*\delta_{\alpha'}\rangle_{\mathrm{Av}}=\delta_{\alpha,\alpha'},\quad (6)$$

where $\delta_{\alpha,\alpha'}$ is the Kronecker delta-function and the angular brackets denote an average over all nuclei.

With the aid of Eq. (5), the angular correlation of the successive quanta may be obtained as follows. w, defined by

$$w=|\lim_{t\to\infty} c_{\gamma,\rho,\sigma}|^2,\quad (7)$$

may be interpreted as the probability that a given nucleus will be found in a final state γ and that quanta ρ and σ will have been emitted with definite directions, frequencies, and polarizations. The average of w over all nuclei, all gamma-ray energies,[3] both directions of polarization of each quantum, and all finall states γ of the system, gives the desired directional correlation of the successive gamma-rays:

$$W=\sum_{P,N}\int_0^\infty d\nu_\rho\int_0^\infty d\nu_\sigma \sum_\gamma w,\quad (8)$$

where $\sum_{P,N}$ denotes an average over all nuclei and all polarizations of the emitted quanta.

Using Eqs. (5)–(8), one obtains

$$W(\mathbf{k}_\rho,\mathbf{k}_\sigma)=\sum\{\alpha,\beta,\beta',\gamma,P_\rho,P_\sigma\}\frac{(\alpha|H(\mathbf{k}_\rho,P_\rho)|\beta)^*(\beta|H(\mathbf{k}_\sigma,P_\sigma)|\gamma)^*(\alpha|H(\mathbf{k}_\rho,P_\rho)|\beta')(\beta'|H(\mathbf{k}_\sigma,P_\sigma)|\gamma)}{1-i\nu_{\beta,\beta'}/2\gamma_B}.\quad (9)$$

In Eq. (9), $\mathbf{k}_\rho$ and $\mathbf{k}_\sigma$ are vectors indicating the directions of propagation of the quanta, whereas P_ρ and P_σ indicate the polarizations (it is convenient in the following to use circular polarization, so that the quantum number P has the values 1 and -1 corresponding to left- or right-handed circular polarization).

Each sublevel, α, β, or γ, is completely characterized by appropriate values of the quantum numbers I, J, F, and m[4]; I is the nuclear angular momentum quantum number, J the electronic angular momentum quantum number, F the resultant of I and J, and m the projection of F on the Z axis. One may therefore write (it is assumed that the emitted gamma-ray does not alter the configuration of the electronic state, so that J remains unaltered)

$$(\alpha|H(\mathbf{k}_\rho,P_\rho)|\beta)=(aF_\alpha m_\alpha|H(\mathbf{k}_\rho,P_\rho)|bF_\beta m_\beta).\quad (10)$$

Here, b represents the quantum numbers that

[3] The matrix elements in Eq. (5) are assumed to be effectively independent of frequency over a range of frequencies greater than both the radiation breadth of the nuclear levels and the hyperfine structure separation of the sublevels.

[4] It is assumed in the present calculation that J is a good quantum number.

are the same for all sublevels of B'; that is, b represents I_B, J, and any other quantum numbers necessary to describe the nuclear level B. Also, $\sum_\beta$ means $\sum\{F_{\beta,} m_\beta\}$, etc.

By using the well-known methods of transformation theory, the matrix elements in Eq. (10) may be written, in the first approximation,

$$\sum_{l,\, m_J,\, n} (I_AJF_\alpha m_\alpha | I_AJlm_J)(alm_J | H(\mathbf{k}_\rho, P_\rho) | bnm_J) \times (I_BJnm_J | I_BJF_\beta m_\beta). \quad (11)$$

In Eq. (11), l is the component along the Z axis of I_A, n that of I_B, and m_J that of J. Similarly, in the following p will denote the component along the Z axis of I_C.

Since the matrix element

$$(alm_J | H(\mathbf{k}_\rho, P_\rho) | bnm_J)$$

is independent of m_J, this quantum number will be omitted when writing the matrix element. The $(IJFm | IJm_Im_J)$ are the transformation coefficients for vector addition[5] and form a real and unitary matrix, so that

$$\sum_{F,\, M} (IJm_Im_J | IJFm)(IJFm | IJm_I'm_J') = \delta_{m_I,\, m_I'}\delta_{m_J,\, m_J'}, \quad (12)$$

$$\sum_{m_I,\, m_J} (IJFm | IJm_Im_J)(IJm_Im_J | IJF'm') = \delta_{F,\, F'}\delta_{m,\, m'}. \quad (13)$$

It is convenient to remember that

$$(IJm_Im_J | IJFm) = (IJFm | IJm_Im_J) = 0 \quad \text{unless } m = m_I + m_J. \quad (14)$$

Upon substituting of Eq. (10) and (11) and a corresponding set of relations for $(\beta | H(\mathbf{k}_\sigma, P_\sigma) | \gamma)$ into Eq. (9), one obtains, with the aid of Eqs. (12)–(14) and (17), for either $\mathbf{k}_\rho$ or $\mathbf{k}_\sigma$ along the Z axis,

$$W(\mathbf{k}_\rho, \mathbf{k}_\sigma) = \sum\{l, n, n', p, P_\rho, P_\sigma\} | (al | H(\mathbf{k}_\rho, P_\rho) | bn) |^2 S_{n,\, n'} | (bn' | H(\mathbf{k}_\sigma, P_\sigma) | cp) |^2, \quad (15)$$

where

$$S_{n,\, n'} = \sum_{r,\, s,\, m,\, F,\, F',\, m'} \frac{(I_BJnr | I_BJFm)(I_BJFm | I_BJn's)(I_BJn's | I_BJF'm')(I_BJF'm' | I_BJnr)}{\{2J+1\}\{1+(\nu_{F,\, F'}/2\gamma_B)^2\}}. \quad (16)$$

The summation in Eq. (16) is over all values of r, s, m, F, and F' consistent with the definition of the transformation coefficients and the given values of I_B, J, n, and n'. $\nu_{F,\, F'}$ is the separation of the levels β and β' corresponding to F and F'. The factor of $2J+1$ in the denominator of Eq. (16) is merely inserted for convenience. The multiplication of W by any constant factor is permissible, since one is interested in the relative, not the absolute, value of W as a function of the angle between the successive quanta.

In the calculation of Eq. (15), use was made of the relation that, for $\mathbf{k}$ along the Z axis,

$$(al | H(\mathbf{k}, P) | bn)^*(al | H(\mathbf{k}, P) | bn') = 0 \quad \text{unless } n = n', \quad (17)$$

which follows from Eqs. (29)–(31), and (34) below.

Equations (15) and (16) are the formal results for the angular distribution of successive quanta emitted by a nucleus, including the effect of the magnetic moment of the extranuclear electrons. It is of interest to see how these reduce to Hamilton's[1] result. The criterion for this is that

$$(\nu_{F,\, F'}/2\gamma_B)^2 \ll 1, \quad (18)$$

so that, from Eq. (16),

$$S_{n,\, n'} = \delta_{n,\, n'}; \quad [(\nu_{F,\, F'}/2\gamma_B)^2 \ll 1]. \quad (19)$$

If Eq. (19) is substituted in Eq. (15), the following is obtained.

$$W(\mathbf{k}_\rho, \mathbf{k}_\sigma) = \sum\{l, n, p, P_\rho, P_\sigma\} \times | (al | H(\mathbf{k}_\rho, P_\rho) | bn) |^2 \times | (bn | H(\mathbf{k}_\sigma, P_\sigma) | cp) |^2. \quad (20)$$

This is precisely the result of reference 1.

The criterion (18) is in agreement with the conclusion of Hamilton, who stated that the radiation width of the levels β must be much greater than the splitting of B' because of the magnetic field of the atomic electrons.

[5] E. U. Condon and G. H. Shortley, *Theory of Atomic Spectra* (Cambridge University Press, Cambridge, 1935), especially pages 73–78. See also reference 7.

Equation (18) may be put in a somewhat more instructive form. Writing

$$S_{n,n'}=\delta_{n,n'}-\alpha_{n,n'}, \tag{21}$$

one has from Eq. (16)

$$\alpha_{n,n'}=\sum_{F,F'}\left(\frac{[\nu_{F,F'}/2\gamma_B]^2}{1+[\nu_{F,F'}/2\gamma_B]^2}\right)\sum_{r,s,m,m'}\frac{(nr|Fm)(Fm|n's)(n's|F'm')(F'm'/nr)}{2J+1}, \tag{22}$$

$$|\alpha_{n,n'}|\leqslant(\Delta\nu/2\gamma_B)^2,$$

where $\Delta\nu$ is the largest value, for all F and F', of $\nu_{F,F'}$. If one assumes as the criterion that Eq. (20) be valid the relation $|\alpha_{n,n'}|\leqslant 0.01$, a sufficient condition for the validity of Eq. (20) is

$$|\Delta\nu/2\gamma_B|\leqslant 0.1. \tag{23}$$

In a slightly different form, one has

$$\tau_B=\frac{1}{4\pi\gamma_B}\leqslant\frac{1}{20\pi\Delta\nu} \tag{24}$$

as an upper limit to the lifetime of the intermediate nuclear state for the validity of the calculation of the angular correlation with the nelect of the interaction of the nuclear magnetic moment with that of the electrons. For a hyperfine structure splitting of 1 cm^{-1}, Eq. (24) yields as the upper limit to the lifetime of the intermediate state 0.5×10^{-12} sec. This estimate seems conservative.

The matrix elements in Eq. (15) may be evaluated in a general manner applicable to transitions of any multipole order by employing the expansion of the potential of a plane electromagnetic wave in terms of the spherical wave solutions of the radiation equations. A convenient form of this expansion is derived in Appendix 1 by group theoretical methods.

The matrix elements in Eq. (15) correspond to the emission from a nucleus of a plane wave in a given direction and with a specified polarization. A possible set of potentials to describe a plane electromagnetic wave is

$$\mathbf{A}(\mathbf{k},P)=2^{-\frac{1}{2}}(\mathbf{u}_1+iP\mathbf{u}_2)\exp i(\mathbf{k}\cdot\mathbf{r}-kct)$$
$$+2^{-\frac{1}{2}}(\mathbf{u}_1-iP\mathbf{u}_2)\exp i(-\mathbf{k}\cdot\mathbf{r}+kct), \tag{25}$$
$$\varphi(\mathbf{k},P)=0,$$

where

$$\mathbf{k}\cdot\mathbf{k}=k^2;\quad \mathbf{k}=k\mathbf{u}_3;\quad P=\pm1,$$
$$\mathbf{u}_i\cdot\mathbf{u}_j=\delta_{i,j},\quad i,j=1,2,3;$$
$$\mathbf{u}_1\times\mathbf{u}_2=\mathbf{u}_3.$$

Equation (25) describes an electromagnetic plane wave of frequency $kc/2\pi$ propagated in direction $\mathbf{u}_3$. This wave is left- or right-handed circularly polarized, depending on whether P is 1 or -1, as may be verified by using

$$\mathbf{H}=\nabla\times\mathbf{A};\quad \mathbf{E}=-\frac{1}{c}\frac{\partial\mathbf{A}}{\partial t}$$

TABLE I. Spherical wave potentials for the electromagnetic field.* In the gauge where the scalar potential is zero, a complete set of potentials for the radiation field is given by the following electric and magnetic multipoles. The multipole order corresponding to a given L is 2^L, and there are $2(2L+1)$ solutions for each L, since M can take on any of the $2L+1$-values $-L$, $-(L-1)$, $\cdots$, $(L-1)$, L, and one can have either electric or magnetic radiation. All potentials are normalized to $(\pi^2\hbar k)^{-1}$ quanta/sec.

Magnetic multipoles, $\mathbf{A}_0(LM)$

$$2^{-\frac{1}{2}}(A_{0x}+iA_{0y})=B_1f_L(kr)Y_L^{M+1}\exp -ikct$$
$$A_{0z}=B_0f_L(kr)Y_L^M\exp -ikct$$
$$-2^{-\frac{1}{2}}(A_{0x}-iA_{0y})=B_{-1}f_L(kr)Y_L^{M-1}\exp -ikct,$$

where

$$B_\sigma=(L\,1LM|L\,1M+\sigma\,-\sigma),$$
$$\sigma=\pm1,\,0.$$

Electric multipoles, $\mathbf{A}_e(LM)$

$$2^{-\frac{1}{2}}(A_{ex}+iA_{ey})=\{C_1f_{L+1}(kr)Y_{L+1}^{M+1}+D_1f_{L-1}(kr)Y_{L-1}^{M+1}\}\exp -ikct$$
$$A_{ez}=\{C_0f_{L+1}(kr)Y_{L+1}^M+D_0f_{L-1}(kr)Y_{L-1}^M\}\exp -ikct$$
$$-2^{-\frac{1}{2}}(A_{ex}-iA_{ey})=\{C_{-1}f_{L+1}(kr)Y_{L+1}^{M-1}+D_{-1}f_{L-1}(kr)Y_{L-1}^{M-1}\}\exp -ikct,$$

where

$$C_\sigma=-\left(\frac{L}{2L+1}\right)^{\frac{1}{2}}(L+1\;1LM|L+1\;1\;M+\sigma\;-\sigma)$$
$$D_\sigma=+\left(\frac{L+1}{2L+1}\right)^{\frac{1}{2}}(L-1\;1LM|L-1\;1\;M+\sigma\;-\sigma)$$
$$\sigma=\pm1,\,0.$$

* See references 6 and 16 in text.

to obtain

$$\mathbf{E}\cdot\mathbf{u}_1=-2^{\frac{1}{2}}k\sin(\mathbf{k}\cdot\mathbf{r}-kct),$$
$$\mathbf{E}\cdot\mathbf{u}_2=P2^{\frac{1}{2}}k\cos(\mathbf{k}\cdot\mathbf{r}-kct),$$
$$\mathbf{E}\cdot\mathbf{u}_3=0,$$
$$\mathbf{H}\cdot\mathbf{u}_1=-P2^{\frac{1}{2}}k\cos(\mathbf{k}\cdot\mathbf{r}-kct),$$
$$\mathbf{H}\cdot\mathbf{u}_2=-2^{\frac{1}{2}}k\sin(\mathbf{k}\cdot\mathbf{r}-kct),$$
$$\mathbf{H}\cdot\mathbf{u}_3=0.$$

Only the term in Eq. (25) containing the factor $\exp(-ickt)$ contributes to the emission of radiation, so that only this term need be considered further. The following expansion is derived in Appendix (1):

$$2^{-\frac{1}{2}}(\mathbf{u}_1+iP\mathbf{u}_2)\exp i(\mathbf{k}\cdot\mathbf{r}-kct)$$
$$=\pi\sum_{L=1}^{\infty}\sum_{M=-L}^{L}i^L(2L+1)^{\frac{1}{2}}\mathbf{D}^{(L)}(\varphi,\vartheta,0)_{M,P}\times[\mathbf{A}_0(LM)+iP\mathbf{A}_e(LM)].\quad(26)$$

Here, $\mathbf{A}_e(LM)$ and $\mathbf{A}_0(LM)$ are the various spherical eigenwave solutions of Maxwell's equations, as derived by Heitler[6] and as listed in Table I. The $\mathbf{D}^{(L)}(\varphi,\vartheta,0)_{M,P}$ are known functions of ϑ and φ, some of whose properties are briefly discussed in Appendix (2).[7] ϑ and φ are the polar coordinates specifying the direction of the propogation vector $\mathbf{k}$, so that

$$k_x=k\sin\vartheta\cos\varphi;\quad k_y=k\sin\vartheta\sin\varphi;$$
$$k_z=k\cos\vartheta.$$

Since, in the gauge where $\varphi=0$,

$$H(\mathbf{k},P)=\mathbf{p}\cdot\mathbf{A}(\mathbf{k},P)\quad\text{(non-relativistic)}\quad(27a)$$
$$H(\mathbf{k},P)=\boldsymbol{\alpha}\cdot\mathbf{A}(\mathbf{k},P)\quad\text{(relativistic)}\quad(27b)$$

($\mathbf{p}$ is the linear momentum operator of the radiating particle and $\boldsymbol{\alpha}$ is the Dirac vector matrix), one finds with the aid of Eq. (25) and (26)

$$(bn|H(\mathbf{k},P)|cp)=\pi\sum_{L,M}i^L(2L+1)^{\frac{1}{2}}\mathbf{D}^{(L)}(\varphi,\vartheta,0)_{M,P}\times(bn|{\textstyle\frac{\mathbf{p}}{\boldsymbol{\alpha}}}\cdot[\mathbf{A}_0(LM)+iP\mathbf{A}_e(LM)]|cp).\quad(28)$$

[6] W. Heitler, Proc. Camb. Phil. Soc. **32**, 112 (1936). An independent solution has been given by W. W. Hansen, Phys. Rev. **47**, 139 (1935).

[7] For a more complete discussion, see Eugen Wigner, *Gruppentheorie* (Friedrich Vieweg und Sohn Akt.-Ges., Braunschweig, 1931), Chapters 14 and 15.

The assumption is now made that the nuclear transition corresponds to the emission of light of some definite multipole order.[8] Then, all the matrix elements in Eq. (28) vanish except those corresponding to, say, 2^L-pole radiation, so that

$$(bn|H(\mathbf{k},P)|cp)$$
$$=\pi i^L(2L+1)^{\frac{1}{2}}\textstyle\sum_M\mathbf{D}^{(L)}(\varphi,\vartheta,0)_{M,P}\times(bn|{\textstyle\frac{\mathbf{p}}{\boldsymbol{\alpha}}}\cdot[\mathbf{A}_0(LM)+iP\mathbf{A}_e(LM)]|cp).\quad(29)$$

From Appendix (3),

$$(bn|{\textstyle\frac{\mathbf{p}}{\boldsymbol{\alpha}}}\cdot[\mathbf{A}_0(LM)+iP\mathbf{A}_e(LM)]|cp)$$
$$=C(I_Bn|Y_L{}^M|I_Cp)=C'(I_CLpM|I_CLI_Bn),\quad(30)$$

where C and C' are independent of n, p, or M. Also,

$$(In|Y_L{}^M|I'p)=0\quad\text{unless } n=M+p\quad(31)$$

(cf. Eq. (14)). Upon substituting Eq. (30) and (29) into Eq. (15) and using Eq. (31) (ϑ is written for the angle between $\mathbf{k}_\rho$ and $\mathbf{k}_\sigma$), one finds

$$W(\vartheta)=\sum_{M,M',n,n'}|(I_An+M'|Y_{L'}{}^{M'}|I_Bn)|^2\times S_{n,n'}|(I_Bn'|Y_L{}^M|I_Cn'-M)|^2f_{LM}(\vartheta),\quad(32)$$
$$M'=\pm1,$$

where $S_{n,n'}$ is defined by Eq. (16) and

$$f_{LM}(\vartheta)=\sum_{P=\pm1}|\mathbf{D}^{(L)}(\varphi,\vartheta,0)_{M,P}|^2.\quad(33)$$

In obtaining Eq. (32), $\mathbf{k}_\rho$ was taken along the Z axis and use was made of the result from Appendix (2) that

$$\mathbf{D}^{(L)}(0,0,0)_{M',P}=\delta_{M',P}.\quad(34)$$

If $\mathbf{k}_\sigma$ is taken along the Z axis rather than $\mathbf{k}_\rho$, the

[8] Magnetic and electric multipole radiation of the same order cannot occur in the same transition, since one or the other will be forbidden by the parity selection rules. However, a mixing of radiations of different multipole orders, such as magnetic dipole and electric quadrupole, might occur. (Cf. A. C. Helmholz, Phys. Rev. **60**, 415 (1941) and S. M. Dancoff and P. Morrison, Phys. Rev. **55**, 122 (1939)). In this case, it is necessary, to determine the angular correlation, to know the relative values of the constant C in Eq. (30) for the two types of radiation. Our knowledge of nuclear structure is insufficient to enable the determination of the complex constant C.

following apparently different result is found

$$W(\vartheta)=\sum_{M,M',n,n'}|(I_An+M'|Y_{L'}^{M'}|I_Bn)|^2$$
$$\times S_{n,n'}|(I_Bn'|Y_L^M|I_Cn'-M)|^2 f_{L'M'}(\vartheta),\quad (32')$$
$$M=\pm 1.$$

Since the angular correlation is independent of the choice of coordinate axis, Eqs. (32) and (32′) should give the same result for $W(\vartheta)$. The appropriate transformation of coordinates, when applied to Eq. (32), can be shown to convert Eq. (32) into Eq. (32′). This equivalence of Eqs. (32) and (32′) is a useful check in specific calculations.

Equation (32) may be transformed so as to verify a conjecture of Hamilton.[1] $W(\vartheta)$ is a polynomial of degree k in $\cos^2\vartheta$, where k is an integer such that[9]

$$k\leqslant L;\quad k\leqslant L'.$$

Hamilton conjectured that also

$$k\leqslant I_B.$$

To show this, note that

$$\sum_M|(I_Bn'|Y_L^M|I_Cn'-M)|^2 f_{LM}(\vartheta)$$
$$=\sum_{p,\,(P=\pm 1)}(I_Bn'|\sum_M \mathbf{D}^{(L)}(R)_{M,P}Y_L^M|I_Cp)$$
$$\times(I_Bn'|\sum_{M'}\mathbf{D}^{(L)}(R)_{M',P}Y_L^{M'}|I_Cp)^*,\quad (35)$$

where R denotes the rotation of coordinates that would bring the original Z axis (along $\mathbf{k}_\rho$) into coincidence with $\mathbf{k}_\sigma$. Upon applying the rotation of coordinates R to Eq. (35) and using the results of Appendix (2), one obtains

$$W(\vartheta)=\sum_{M,M',n,n',n''}|(I_An+M'|Y_{L'}^{M'}|I_Bn)|^2$$
$$\times S_{n,n'}|\mathbf{D}^{(I_B)}(R^{-1})_{n'',n'}|^2$$
$$\times|(I_Bn''|Y_L^M|I_Cn''-M)|^2,\quad (32'')$$
$$M=\pm 1;\quad M'=\pm 1.$$

From Eq. (32″) it is seen that the highest power of $\cos\vartheta$ that appears in $W(\vartheta)$ is not greater than $2I_B$.[10] Hence, $k\leqslant I_B$.

[9] One may verify from Eq. (33) and reference 7, page 180, Eq. (27), that $f_{LM}(\theta)$ is a polynomial of degree L in $\cos^2\vartheta$. From Eq. (32) one sees that $k\leqslant L$. Likewise, from Eq. (32′), $k\leqslant L'$.

[10] This again follows from reference 7, page 180, Eq. (27).

TABLE II. The following values are obtained by the use of Eq. (33) in conjunction with Eq. (A19).

L	$\|M\|$	$2f_{LM}(\theta)$
1	0	$2-2\cos^2\vartheta$
1	1	$1+\cos^2\vartheta$
2	0	$6\cos^2\vartheta-6\cos^4\vartheta$
2	1	$1-3\cos^2\vartheta+4\cos^4\vartheta$
2	2	$1-\cos^4\vartheta$

Values of the matrix elements $(In|Y_L^M|I'p)$ are given in many places,[11] while the $f_{LM}(\vartheta)$ are listed in Table II. A comparison of Eq. (32) with the corresponding result of reference 1 shows that whenever Eq. (19) is satisfied, the present results are identical with those of reference 1 for dipole and quadrupole radiation. Higher multipoles are not considered in detail in reference 1.

To summarize, the angular correlation of gamma-rays emitted in successive nuclear transitions is given by either Eq. (32) or Eq. (32′) in conjunction with Eqs. (16) and (33). If the lifetime of the nucleus in its intermediate state satisfies Eq. (18), then $S_{n,n'}$ may be replaced by $\delta_{n,n'}$. This case may be indicated by saying that the probability of reorientation of the nuclear angular momentum while the nucleus is in its intermediate state is negligible. In fact, it is clear from Eq. (15) that $S_{n,n'}$ may be interpreted as the probability that, if the nucleus has a component n of angular momentum along the Z axis immediately after the first gamma-ray is emitted, it will have a component n' just before the second gamma-ray is emitted.

EFFECT OF AN EXTERNAL MAGNETIC FIELD

Inasmuch as the projection along the field of the angular momentum of a system in a steady magnetic field is a constant of the motion, one might expect that the reorientation probability could be reduced by the application of a sufficiently strong field along the Z axis. (Note that the direction of the Z axis has been previously defined by the statement that one of the emitted gamma-rays is collected along the Z axis. Hence, the magnetic field must be applied so that it coincides in direction with the line from the radioactive source to one of the gamma-ray detectors.) That this expectation is justified is

[11] For example, references 5 and 7 (use is made of Eq. (30)).

shown below and an estimate of the strength of the necessary magnetic field is obtained.

To calculate the angular correlation of the successive gamma-rays in the presence of an external magnetic field in the direction specified above, the derivation of Eqs. (15) and (16) is repeated, taking into account the presence of the external field. This field alters the zero-order wave functions to be used in describing the states of the system.

In the derivation of Eqs. (15) and (16) the wave functions of the radiating system were taken in the form

$$\psi(IJFm)=\sum\{m_I,\, m_J\}\psi_N(Im_I)\times\psi_E(Jm_J)(IJm_Im_J\,|\,IJFm),$$

where the transformation coefficients are so defined that the wave functions $\psi(IJFm)$ diagonalized the hyperfine structure operator

$$H_0=f(r)\mathbf{I}\cdot\mathbf{J}. \tag{36}$$

If a magnetic field is present, the operator defined in Eq. (36) is replaced by

$$H_1=f(r)\mathbf{I}\cdot\mathbf{J}+H_z(\mu_I I_z+\mu_J J_z), \tag{37}$$

where H_z is the applied field. The eigenfunctions which diagonalize H_1 may be written as

$$\psi(IJM_IM_J)=\sum\{m_I,\, m_J\}\psi_N(Im_I)\times\psi_E(Jm_J)(m_Im_J\,|\,M_IM_J), \tag{38}$$

where M_I and M_J represent the eigenvalues of a complete set of operators which characterize the levels of the system when the effect of H_1 is taken into account and which, in the limit of vanishing hyperfine structure separation or large external field, become identical with m_I and m_J.

It is assumed for simplicity that J remains a good quantum number for all magnetic fields considered. This assumption will not affect the validity of the conclusions to be obtained, which depend primarily on the strength of the applied field being sufficiently great to decouple the nuclear spin from all other angular momenta that may be present.

In terms of the wave functions of Eq. (38), one obtains in place of Eqs. (15) and (16) an equation identical to Eq. (15) and the following in place of Eq. (16):

$$S_{n,n'}(H_z)=\sum\{r,\, s,\, M_I,\, M_J,\, M_I',\, M_J'\}\frac{(nr\,|\,M_IM_J)(M_IM_J\,|\,n's)(n's\,|\,M_I'M_J')(M_I'M_J'\,|\,nr)}{(2J+1)\{1+[(\nu_{M_IM_J}-\nu_{M_I'M_J'})/2\gamma_B]^2\}}. \tag{39}$$

It is apparent from the foregoing that Eq. (39) will reduce to Eq. (16) (although the use of the quantum numbers M_I and M_J is not convenient in this limit) for $H_z=0$.

The transformation coefficients $(m_Im_J\,|\,M_IM_J)$ may be evaluated by the usual perturbation theory for a degenerate state. From the condition that $\psi(IJM_IM_J)$ diagonalizes H_1, one obtains

$$\sum\{m_I,\, m_J\}(m_I'm_J'\,|\,H_1\,|\,m_Im_J)(m_Im_J\,|\,M_IM_J)=E_{M_IM_J}(m_I'm_J'\,|\,M_IM_J), \tag{40}$$

from which

$$|(m_I'm_J'\,|\,H_1\,|\,m_Im_J)-\lambda\delta_{m_I,\,m_I'}\delta_{m_J,\,m_J'}|=0, \tag{41}$$

where λ denotes any $E_{M_IM_J}$

Now, from Eq. (37) using transformation theory and the results of the theory of hyperfine structure, one obtains

$$\begin{aligned}(m_I'm_J'\,|\,H_1\,|\,m_Im_J)&=H_z(\mu_Im_I+\mu_Jm_J)\delta_{m_I,\,m_I'}\delta_{m_J,\,m_J'}\\&\quad+\sum_{F,F',m,m'}(m_I'm_J'\,|\,F'm')(F'm'\,|\,f(r)\mathbf{I}\cdot\mathbf{J}\,|\,Fm)(Fm\,|\,m_Im_J)\\&=\{H_z(\mu_Im_I+\mu_Jm_J)\delta_{m_I,\,m_I'}+\sum_{F,m}(m_I'm_J'\,|\,Fm)h\nu_F(Fm\,|\,m_Im_J)\}\delta_{m_I+m_J,\,m_I'+m_J'},\end{aligned} \tag{42}$$

where the ν_F are defined by

$$h\nu_F=C_{IJ}[F(F+1)-I(I+1)-J(J+1)] \tag{43}$$

and C_{IJ} is independent of F and m. That is, ν_F is the displacement from some zero position of the various levels in the hyperfine structure multiplet.

One may now substitute Eq. (42) into Eq. (41) to find $E_{M_IM_J}$ and then use Eq. (40) to determine the $(m_I m_J|M_I M_J)$. But such a specific result is not required. Noting that

$$|\sum_{F,m}(m_I'm_J'|Fm)\nu_F(Fm|m_Im_J)|\leqslant\tfrac{1}{2}\Delta\nu, \quad (44)$$

where $\Delta\nu$ is the greatest value (for all F and F') of $\nu_F-\nu_{F'}$, and assuming that

$$h\Delta\nu \ll H_z\mu_J, \quad (45)$$

one finds

$$E_{M_IM_J}=(m_Im_J|H_1|m_Im_J)$$
$$+\text{terms of order } H_z\mu_J(h\Delta\nu/H_z\mu_J)^2, \quad (46)$$

with $M_I=m_I$; $M_J=m_J$; also,

$$(m_Im_J|M_IM_J)=\delta_{m_I+m_J\ M_I+M_J}$$
$$\times\{\delta_{m_I,M_I}+\text{terms of order } (h\Delta\nu/H_z\mu_J)\}. \quad (47)$$

Substituting Eq. (47) into Eq. (39), one finds

$$S_{n,n'}(H_z)=\delta_{n,n'}$$
$$-\text{terms of order } (h\Delta\nu/H_z\mu_J)^2. \quad (48)$$

Thus, the probability of reorientation is negligible if the Zeeman splitting of the electronic levels because of an external magnetic field is several times as great as the hyperfine structure splitting of the same levels.

It has been shown that with magnetic fields obtainable in the laboratory it is possible to reduce the reorientation probability to a negligible amount, provided the magnetic field is oriented in a direction defined by the gamma-ray emitter and one of the detectors. The field need merely be large enough to give rise to a Zeeman effect of the atomic levels several times larger than the expected hyperfine structure splitting of the intermediate state.[12]

An interesting experimental possibility might be to measure the angular correlation as a function of the strength of the magnetic field. If one can infer the spins and multipole nature of the radiations for the nuclear transitions involved from the available evidence as to angular correlation with a strong magnetic field and from measured internal conversion coefficients, one might then use these values to deduce from the preceding theory the angular correlation at intermediate fields. A comparison between theory and experiment would check the assignment of spin values and radiation multipolarity and might also enable an estimate of the hyperfine structure splitting of the intermediate state.

[12] The criterion of Eq. (45) may be written

$$H_z^2 \gg (2\times10^4\Delta\nu)^2$$

where H_z is in gauss and $\Delta\nu$ in cm^{-1}. For bismuth ($\Delta\nu\sim3$ cm^{-1}), the Paschen-Back effect of the hyperfine structure is almost complete at 43,000 gauss. For most atoms, $\Delta\nu$ will be much less, so that smaller magnetic fields will suffice.

SUMMARY

It has been demonstrated above that the angular correlation of successive nuclear gamma-rays emitted by an isolated atomic system is given by either Eq. (32) or Eq. (32′), with $S_{n,n'}$ and $f_{LM}(\vartheta)$ defined by Eqs. (16) and (33), respectively In Eqs. (16), (32), (32′), and (33) the following notation is used:

ϑ	the angle between the successive gamma-rays,
$W(\vartheta)$	the relative probability of a given value of ϑ,
L'	the first nuclear transition gives rise to a $2^{L'}$-pole quantum,
L	the second nuclear transition gives rise to a 2^L-pole quantum,
M, M'	summation indices such that $-L\leqslant M\leqslant L$; $-L'\leqslant M'\leqslant L'$, unless otherwise noted,
n, n'	summation indices such that. $-I_B\leqslant n,\ n'\leqslant I_B$,
I_A, I_B, I_C	spins of the initial, intermediate, and final states of the nucleus, respectively,
J	angular momentum of extra-nuclear electrons,
r, s	summation indices such that $-J\leqslant r,\ s\leqslant J$,
F, F'	summation indices such that $\lvert I_B-J\rvert\leqslant F,\ F'\leqslant I_B+J$,
m, m'	summation indices such that $-F'\leqslant m'\leqslant F'$; $-F\leqslant m\leqslant F$,
$1/(4\pi\gamma_B)$	lifetime of intermediate nuclear state,
$\nu_{FF'}$	hyperfine structure splitting of sublevels F and F' of the intermediate state,
$(I_BJnr\lvert I_BJFm)$	transformation amplitude for vector addition (cf. reference 5),
$(I_An+M'\lvert Y_{L'}^{M'}\rvert I_Bn)$	matrix element of the spherical harmonics (cf. reference 11),
$\mathbf{D}^{(L)}(\varphi,\vartheta,0)_{M,\pm1}$	$2L+1$ dimensional representation of the three-dimensional rotation group (cf. Appendix 2, especially Eq. (A19), and reference 7).

If the intermediate state is sufficiently short lived, one may write

$$S_{n,n'}=\delta_{n,n'}. \quad (19)$$

The criterion for this is given by either Eq. (18) or Eq. (23). When Eq. (19) is used in place of

TABLE III. $S_{n,n'}$ for $J=\frac{1}{2}$ (cf. Eq. (16)).

$$S_{n,n}=1-2T\frac{I_B(I_B+1)-n^2}{(2I_B+1)^2}$$

$$S_{n,n\pm1}=T\frac{I_B(I_B+1)-n(n\pm1)}{(2I_B+1)^2},$$

where

$$T=(\nu_B/2\gamma_B)^2/[1+(\nu_B/2\gamma_B)^2]$$

and ν_B is the hyperfine structure separation of the two sublevels of the intermediate state.

Eq. (16) one obtains Hamilton's results for the angular correlations.

Even when the hyperfine structure separation is appreciable, the validity of the approximate Eq. (19) may be secured by the application of an external magnetic field in the direction of propagation of one of the collected gamma-rays. For an indication of the necessary field strength, see the discussion following Eq. (48).

DISCUSSION

It is clear from Eqs. (15) and (32) and from the subsequent discussion that the hyperfine structure interaction of nuclear and electronic magnetic moments will reduce the angular correlation between the successive quanta over that which would obtain for an isolated nucleus. The shorter the lifetime of the intermediate state, the less important is the effect of the external electrons. As an example, one may consider a specific application of the formula. For $J=1/2$, $I_A=I_B=I_C=I$, both transition dipole ($L=L'=1$), one finds for the angular correlation

$$W(\theta)=1+A\cos^2\theta,$$

where

$$A=\frac{(2I-1)(2I+3)-6T(2I-1)(2I+3)/(2I+1)^2}{12I^2+12I+1+2T(2I-1)(2I+3)/(2I+1)^2},$$

and (see Table III)

$$T=(\nu_B/2\gamma_B)^2/[1+(\nu_B/2\gamma_B)^2].$$

ν_B is the hyperfine structure separation of the two levels of the intermediate state. For $I=\frac{1}{2}$, the distribution is isotropic and independent of the hyperfine structure separation ν_B. For $I>\frac{1}{2}$, A is a monotonically decreasing function of ν_B, as is to be expected.

The above discussed decrease in predicted angular correlation may, as was previously pointed out,[13] be in part responsible for the small angular correlations observed experimentally.

It is clear that the preceding calculations will not apply in the event that there exist reorienting torques on the atom other than those arising from a uniform magnetic field. Thus, in the case of a solid, the electric fields of adjoining atoms will probably result in torques which reorient the atoms, so that the angular correlation of successive gamma-rays is not correctly given by the foregoing formalism unless the lifetime in the intermediate state of the nucleus is so short that reorientation may be neglected. On the other hand, there is no doubt as to the applicability of the above results to a monatomic gas or vapor at sufficiently low pressure.

Another limitation on the applicability of this work is the possibility that the nuclear transition gives rise to mixed radiations.[8] In such a case, the angular correlations calculated above will not apply.

In conclusion, I wish to express my gratitude to Professor I. S. Lowen for suggesting this problem and for continued valuable suggestions and discussions throughout the work. In addition, I thank the Radio Receptor Company wholeheartedly for the grant of a research fellowship which made this work possible.

APPENDIX 1. EXPANSION OF A PLANE ELECTROMAGNETIC WAVE IN TERMS OF SPHERICAL WAVES[14]

One may expand the potential

$$\mathbf{A}(\mathbf{k},P)=2^{-\frac{1}{2}}(\mathbf{u}_1+iP\mathbf{u}_2)\exp i(\mathbf{k}\cdot\mathbf{r}-kct) \tag{A1}$$

in terms of the spherical potentials of Table I. The symbols in Eq. (A1) have the meanings given them after Eq. (25).

[13] A preliminary report on some aspects of the present work has been previously published (G. Goertzel and I. S. Lowen, Phys. Rev. 69, 533 (1946)).

[14] See also Hansen, reference 7.

The following expansion is given in many places:[15]

$$\exp i\mathbf{k}\cdot\mathbf{r}=\pi 2^{\frac{1}{2}}\sum_{l=0}^{\infty} i^l(2l+1)^{\frac{1}{2}}f_l(kr)\,Y_l^0(\vartheta,\,\varphi) \tag{A2}$$

where $\cos\vartheta=\mathbf{u}_3\cdot\mathbf{r}/r$. Substituting Eq. (A2) in Eq. (A1), one obtains

$$\mathbf{A}(\mathbf{k},\,P)=\pi(\mathbf{u}_1+iPu_2)\exp -ikct\sum_{l=0}^{\infty} i^l(2l+1)^{\frac{1}{2}}f_l(kr)\,Y_l^0(\vartheta,\,\varphi). \tag{A3}$$

Equation (A3) is the expansion of a plane wave in terms of spherical waves in a coordinate system in which the plane wave is propagated along the $\mathbf{u}_3$ axis. It is desirable to obtain this expansion in terms of any arbitrary axis system.

A new axis system defined by unit vectors $\mathbf{v}_1$, $\mathbf{v}_2$, $\mathbf{v}_3$ is set up relative to the $\mathbf{u}_1$, $\mathbf{u}_2$, $\mathbf{u}_3$ system by the transformation:

$$\begin{aligned}
\mathbf{u}_1&=(\cos\alpha\cos\beta\cos\gamma-\sin\alpha\sin\gamma)\mathbf{v}_1+(\sin\alpha\cos\beta\cos\gamma+\cos\alpha\sin\gamma)\mathbf{v}_2-\sin\beta\cos\gamma\;\mathbf{v}_3;\\
\mathbf{u}_2&=-(\cos\alpha\cos\beta\sin\gamma+\sin\alpha\cos\gamma)\mathbf{v}_1-(\sin\alpha\cos\beta\sin\gamma-\cos\alpha\cos\gamma)\mathbf{v}_2+\sin\beta\sin\gamma\;\mathbf{v}_3;\\
\mathbf{u}_3&=\cos\alpha\sin\beta\;\mathbf{v}_1+\sin\alpha\sin\beta\;\mathbf{v}_2+\cos\beta\;\mathbf{v}_3.
\end{aligned} \tag{A4}$$

Define r, ϑ, φ as the spherical coordinates of a point relative to the $\mathbf{u}_1$, $\mathbf{u}_2$, $\mathbf{u}_3$, system and r, Θ, and Φ as the coordinates of the same point relative to the $\mathbf{v}_1$, $\mathbf{v}_2$, $\mathbf{v}_3$ coordinate system.

The factors $(\mathbf{u}_1+iP\mathbf{u}_2)$ and $Y_l^0(\vartheta,\,\varphi)$ on the right side of Eq. (A3) may be expressed in the $\mathbf{v}_1$, $\mathbf{v}_2$, $\mathbf{v}_3$ coordinate system as

$$-2^{-\frac{1}{2}}P(\mathbf{u}_1+iP\mathbf{u}_2)=\sum_{\sigma=0,\,\pm1}\mathbf{D}^{(1)}(\alpha,\,\beta,\,\gamma)_{\sigma,\,P}\mathbf{w}_\sigma, \tag{A5}$$

$$Y_l^0(\vartheta,\,\varphi)=\textstyle\sum_m \mathbf{D}^{(l)}(\alpha,\,\beta,\,\gamma)_{m,\,0}Y_l^m(\Theta,\,\Phi), \tag{A6}$$

where

$$\mathbf{w}_{-1}=2^{-\frac{1}{2}}(\mathbf{v}_1-i\mathbf{v}_2);\quad \mathbf{w}_0=\mathbf{v}_3;\quad \mathbf{w}_1=-2^{-\frac{1}{2}}(\mathbf{v}_1+i\mathbf{v}_2)$$

and the $\mathbf{D}^{(L)}$ are the $2L+1$ dimensioned representations of the three-dimensional rotation group. Substituting Eqs. (A5) and (A6) in Eq. (A3) and using the formula

$$\mathbf{D}^{(1)}(\alpha,\,\beta,\,\gamma)_{\rho,\,P}\mathbf{D}^{(l)}(\alpha,\,\beta,\,\gamma)_{m,\,0}=\sum_{L=l-1}^{l+1}(l1m\rho|l1Lm+\rho)\mathbf{D}^{(L)}(\alpha,\,\beta,\,\gamma)_{m+\rho,\,P}(l1LP|l10P), \tag{A7}$$

which is a special case of Eq. (A18) below, one obtains

$$\begin{aligned}\mathbf{A}(\mathbf{k},\,P)=-2^{\frac{1}{2}}\pi P\sum_{\rho,\,m,\,l,\,L} & i^l(2l+1)^{\frac{1}{2}}(l1m\rho|lL1m+\rho)\mathbf{D}^{(L)}(\alpha,\,\beta,\,\gamma)_{m+\rho,\,P}\\
&\times(l1LP|l10P)f_l(kr)\,Y_l^m(\Theta,\,\Phi)\mathbf{w}_\rho\exp -ikct.\end{aligned} \tag{A8}$$

But, for $P=\pm1$

$$\begin{aligned}
2^{\frac{1}{2}}(2l+1)^{\frac{1}{2}}(l1LP|l10P)&=(2L+1)^{\frac{1}{2}}\left(\frac{L+1}{2L+1}\right)^{\frac{1}{2}}\quad\text{for } L=l+1,\\
&=(2L+1)^{\frac{1}{2}}\left(\frac{L}{2L+1}\right)^{\frac{1}{2}}\quad\text{for } L=l-1,\\
&=-(2L+1)^{\frac{1}{2}}P\quad\text{for } L=l,
\end{aligned} \tag{A9}$$

[15] For example, E. T. Whittaker and G. N. Watson, *Modern Analysis* (The Macmillan Company, New York, 1944), American edition, p. 401, exercise 9.

so that

$$\mathbf{A}(\mathbf{k}, P)=\pi \sum_{L, M, \rho} i^L(2L+1)^{\frac{1}{2}}\mathbf{D}^{(L)}(\alpha, \beta, \gamma)_{M, P}\mathbf{w}_{-\rho} \exp -ikct$$
$$\times\Bigg\{-iP\Bigg[(L+1\ 1M+\rho\ -\rho|L+1\ 1LM)\left(\frac{L}{2L+1}\right)^{\frac{1}{2}}Y_{L+1}{}^{M+\rho}(\Theta, \Phi)f_{L+1}(kr)$$
$$-(L-1\ 1M+\rho\ -\rho|L-1\ 1LM)\left(\frac{L+1}{2L+1}\right)^{\frac{1}{2}}Y_{L-1}{}^{M+\rho}(\Theta, \Phi)f_{L-1}(kr)\Bigg]$$
$$+(L1\ M+\rho\ -\rho|L1LM)\,Y_L{}^{M+\rho}(\Theta, \Phi)f_L(kr)\Bigg\}. \quad \text{(A10)}$$

Noting that

$$\mathbf{A}=2^{-\frac{1}{2}}(A_x+iA_y)\mathbf{w}_{-1}-2^{-\frac{1}{2}}(A_x-iA_y)\mathbf{w}_1+A_z\mathbf{w}_0 \quad \text{(A11)}$$

and comparing Eq. (A10) with Table I, one obtains

$$\mathbf{A}(\mathbf{k}, P)=\pi \sum_{L=1}^{\infty} \sum_{M=-L}^{L} i^L(2L+1)^{\frac{1}{2}}\mathbf{D}^{(L)}(\alpha, \beta, \gamma)_{M, P}\{\mathbf{A}_0(LM)+iP\mathbf{A}_e(LM)\} \quad \text{(A12)}$$

where $\mathbf{A}_0$ and $\mathbf{A}_e$ are given in Table I,

$$\mathbf{k}=k\mathbf{u}_3=k(\mathbf{v}_1 \cos \alpha \sin \beta+\mathbf{v}_2 \sin \alpha \sin \beta+\mathbf{v}_3 \cos \beta),$$

and the $\mathbf{A}(LM)$ are evaluated in the $\mathbf{v}_1$, $\mathbf{v}_2$, $\mathbf{v}_3$ coordinate system.

APPENDIX 2. ROTATION OF COORDINATES AND SOME PROPERTIES OF THE REPRESENTATION OF THE THREE-DIMENSIONAL ROTATION GROUP.[7]

Consider two sets of coordinate axes specified by the unit vectors $\mathbf{u}_1$, $\mathbf{u}_2$, $\mathbf{u}_3$ and $\mathbf{v}_1$, $\mathbf{v}_2$, $\mathbf{v}_3$, respectively, where the relation between the $\mathbf{u}_1$, $\mathbf{u}_2$, $\mathbf{u}_3$, and the $\mathbf{v}_1$, $\mathbf{v}_2$, $\mathbf{v}_3$ is given by Eq. (A4). Let x_1, x_2, x_3 and r, ϑ, φ be rectangular and polar coordinates of an arbitrary point in the $\mathbf{u}_1$, $\mathbf{u}_2$, $\mathbf{u}_3$ set of coordinates and let X_1, X_2, X_3 and r, Θ, Φ be the coordinates of the same point in the v_1, v_2, v_3 system. It is a well-known principle of quantum theory that any wave function $\psi_{Jm}(x)$ of a system in free space may be expressed as a linear combination of the wave functions $\psi_{JM}(X)$ for the same system. Here, J is the total angular momentum of the system and m, M are the components of J along $\mathbf{u}_3$ and $\mathbf{v}_3$, respectively. Therefore, one may write

$$\psi_{Jm}(x)=\sum_M \psi_{JM}(X)\mathbf{D}^{(J)}(\alpha, \beta, \gamma)_{M, m}, \quad \text{(A13)}$$

which specifies the $\mathbf{D}^{(J)}$ completely, if the phases of the ψ's are specified. A specialization of Eq. (A13) of particular interest is[16]

$$Y_L{}^m(\vartheta, \varphi)=\sum_M Y_L{}^M(\Theta, \Phi)\mathbf{D}^{(L)}(\alpha, \beta, \gamma)_{M, m}. \quad \text{(A14)}$$

From Eq. (A13) it is immediately apparent that

$$\mathbf{D}^{(J)}(0, 0, 0)_{M, m}=\delta_{M, m}. \quad \text{(A15)}$$

Also, a comparison of Eq. (A14) with the spherical harmonic addition theorem

$$Y_L{}^0(\vartheta, \varphi)=\left(\frac{4\pi}{2L+1}\right)^{\frac{1}{2}} \sum_M Y_L{}^M(\beta, \alpha)^* Y_L{}^M(\Theta, \Phi)$$

[16] We use Condon and Shortley's (reference 5, page 52) choice of phases for the associated Legendre polynomials.

shows that

$$\mathbf{D}^{(L)}(\alpha,\beta,\gamma)_{M,0}=\left(\frac{4\pi}{2L+1}\right)^{\frac{1}{2}}Y_L{}^M(\beta,\alpha)^*. \tag{A16}$$

Since

$$(4\pi/3)^{\frac{1}{2}}rY_1{}^1(\vartheta,\varphi)=-2^{\frac{1}{2}}(x_1+ix_2);\quad (4\pi/3)^{\frac{1}{2}}rY_1{}^0(\vartheta,\varphi)=x_3;\quad (4\pi/3)^{\frac{1}{2}}rY_1{}^{-1}(\vartheta,\varphi)=2^{\frac{1}{2}}(x_1-ix_2),$$

it is apparent that $-2^{-\frac{1}{2}}(x_1+ix_2)$, x_3, $2^{-\frac{1}{2}}(x_1-ix_2)$ and $-2^{-\frac{1}{2}}(\mathbf{u}_1+i\mathbf{u}_2)$, $\mathbf{u}_3$, $2^{-\frac{1}{2}}(\mathbf{u}_1-i\mathbf{u}_2)$ transform as $Y_1{}^1(\vartheta,\varphi)$, $Y_1{}^0(\vartheta,\varphi)$, $Y_1{}^{-1}(\vartheta,\varphi)$. Hence, one finds with the aid of Eq. (A4) the following values of $\mathbf{D}^{(1)}(\alpha,\beta,\gamma)_{i,j}$.[17]

$i \backslash j$	-1	0	1
-1	$e^{i\alpha}\frac{1+\cos\beta}{2}e^{i\gamma}$	$e^{i\alpha}\frac{\sin\beta}{\sqrt{2}}$	$e^{i\alpha}\frac{1-\cos\beta}{2}e^{-i\gamma}$
0	$-\frac{\sin\beta}{\sqrt{2}}e^{i\gamma}$	$\cos\beta.$	$\frac{\sin\beta}{\sqrt{2}}e^{-i\gamma}$
1	$e^{-i\alpha}\frac{1-\cos\beta}{2}e^{i\gamma}$	$-e^{-i\alpha}\frac{\sin\beta}{\sqrt{2}}$	$e^{-i\alpha}\frac{1+\cos\beta}{2}e^{-i\gamma}$

(A17)

A relation of particular interest to us is derived in group theory:[18]

$$\mathbf{D}^{(J)}(\alpha,\beta,\gamma)_{M,N}\mathbf{D}^{(j)}(\alpha,\beta,\gamma)_{m,n}=\sum_{F=|J-j|}^{J+j}(jJmM|jJF\,m+M)\mathbf{D}^{(F)}(\alpha,\beta,\gamma)_{M+m,N+n}(jJF\,n+N|jJnN). \tag{A18}$$

Upon setting $N=0$, $j=1$ in Eq. (A18) and using Eqs. (A16) and (A17), one obtains

$$\begin{aligned}\mathbf{D}^{(L)}(\alpha,\beta,\gamma)_{M,1}=-\left[\left(\frac{4\pi}{2L+1}\right)\frac{1}{L(L+1)}\right]^{\frac{1}{2}}\Big\{&MY_L{}^M(\beta,\alpha)^*\sin\beta e^{-i\gamma}\\ &+(L+1+M)^{\frac{1}{2}}(L-M)^{\frac{1}{2}}Y_L{}^{M+1}(\beta,\alpha)^*e^{i\alpha}\frac{1-\cos\beta}{2}e^{-i\gamma}\\ &-(L+1-M)^{\frac{1}{2}}(L+M)^{\frac{1}{2}}Y_L{}^{M-1}(\beta,\alpha)^*e^{-i\alpha}\frac{1+\cos\beta}{2}e^{-i\gamma}\Big\}.\end{aligned} \tag{A19}$$

Also,

$$\mathbf{D}^{(L)}(\alpha,\beta,\gamma)_{M,-1}=(-)^{M+1}\mathbf{D}^{(L)}(\alpha,\beta,\gamma)_{-M,1}{}^*.$$

APPENDIX 3. EVALUATION OF A CLASS OF MATRIX ELEMENTS

It may be shown by group theoretical methods that if an operator transforms under change of coordinates according to the relation

$$H(L,M;\vartheta,\varphi)=\textstyle\sum_{M'}H(L,M';\Theta,\Phi)\mathbf{D}^{(L)}(\alpha,\beta,\gamma)_{M',M}, \tag{A20}$$

the matrix elements of the operator are given by

$$(Jm|H(LM)|J'm')=C(J'LJm|J'Lm'M)=C'(Jm|Y_L{}^M|J'm'), \tag{A21}$$

where C and C' are independent of m, m', and M.[19]

[17] See also reference 7, page 182.
[18] Reference 7, page 204, Eq. (16b).
[19] Reference 7, page 264, Eq. (19).

The operators

$$H_1(LM)=\mathbf{p}\cdot\mathbf{A}_0(LM);\quad H_2(LM)=\mathbf{p}\cdot\mathbf{A}_e(LM), \tag{A22}$$

where $\mathbf{p}$ is any vector and $\mathbf{A}_0$ and $\mathbf{A}_e$ are the potentials of a spherical electromagnetic wave as given in Table I, satisfy Eq. (A20). To show this, one proceeds as follows.

Let

$$p_1=-2^{-\frac{1}{2}}(p_x+ip_y);\quad p_0=p_z;\quad p_{-1}=2^{-\frac{1}{2}}(p_x-ip_y),$$
$$A_1=2^{-\frac{1}{2}}(A_x+iA_y);\quad A_0=A_z;\quad A_{-1}=-2^{-\frac{1}{2}}(A_x-iA_y).$$

Then

$$\begin{aligned}
H_1(L,M;\vartheta,\varphi)&=\sum_{\sigma=\pm1,0} p_\sigma(\vartheta,\varphi)A_{0,-\sigma}(L,M;\vartheta,\varphi)\\
&=\sum_{\sigma=\pm1,0} p_\sigma(\vartheta,\varphi)(L1LM|L1\ M-\sigma\ \sigma)f_L(kr)Y_L^{M-\sigma}(\vartheta,\varphi)\exp -ikct\\
&=\sum_{\sigma,\sigma',M'}\mathbf{D}^{(1)}(\alpha,\beta,\gamma)_{\sigma',\sigma}\mathbf{D}^{(L)}(\alpha,\beta,\gamma)_{M',M-\sigma}p_{\sigma'}(\Theta,\Phi)(L1LM|L1\ M-\sigma\ \sigma)\\
&\qquad\times f_L(kr)Y_L^{M'}(\Theta,\Phi)\exp -ikct\\
&=\sum_{\sigma,\sigma',M',L'}(L1M'\sigma'|L1L'\ M'+\sigma')\mathbf{D}^{(L')}(\alpha,\beta,\gamma)_{M'+\sigma',M}(L1L'M|L1\ M-\sigma\ \sigma)\\
&\qquad\times p_{\sigma'}(\Theta,\Phi)(L1LM|L1\ M-\sigma\ \sigma)f_L(kr)Y_L^{M'}(\Theta,\Phi)\exp -ikct\\
&=\sum_{\sigma,M'}(L1LM'|L1\ M'-\sigma\ \sigma)\mathbf{D}^{(L)}(\alpha,\beta,\gamma)_{M',M}f_L(kr)Y_L^{M'-\sigma}(\Theta,\Phi)p_\sigma(\Theta,\Phi)\exp -ikct\\
&=\textstyle\sum_{M'}\mathbf{D}^{(L)}(\alpha,\beta,\gamma)_{M',M}H_1(L,M;\Theta,\Phi).
\end{aligned}$$

A similar demonstration gives the corresponding result for $H_2(LM)$. It has therefore been shown that the matrix elements of the operators of Eq. (A22) are given by Eq. (A21).

Errata: A New Method of Measuring the Electric Dipole Moment and Moment of Inertia of Diatomic Polar Molecules

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THERE are typographical errors in Eqs. (1) and (4). These should read as follows:

$$f=\frac{6\mu^2IE^2}{20(9)10^4h\hbar^2}=4.529\times10^{75}\mu^2IE^2. \tag{1}$$

$$I=(\beta/\alpha^2)(1.97\times10^{-28}),\ \text{g cm}^2. \tag{4}$$