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On Directional Correlation of Successive Quanta

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A theoretical investigation shows that there should be a correlation between the directions of propagation of the quanta emitted in two successive transitions of a single radiating system. This correlation is described by a function $W(\theta)$ which gives the relative probability that the second quantum will be emitted at an angle θ with the first; W is determined by the angular momenta of the three levels involved in the two transitions and by the multipole order of the radiation emitted in these transitions. The explicit forms of W for all angular momenta and for dipole and quadrupole radiation are given; experimental determination of W in any given case should limit these factors to a small number of possibilities. This has particular interest as a means of investigating the nuclear energy levels involved in γ-radiation; here W should be observable by measuring the variation with θ of gamma-gamma coincidence counting rates.

INTRODUCTION

IT has been suggested by Dunworth[1] that there might be some correlation between the directions of emission of two successive gamma-quanta emitted by a nucleus when this nucleus passes from an excited level A to the ground level C, by way of a definite intermediate level B. This suggestion was pointed out to the author by Dr. I. A. Getting in connection with the latter's search for such a correlation by means of gamma-gamma coincidence experiments. The present paper is a theoretical discussion of the question.

The problem of resonance radiation[2] is basically similar to the present one, since both are concerned with the radiation from an excited level in which a system finds itself as the result of an anisotropic process. This process is, in the first case, an absorption from a unidirectional (and usually polarized) beam of light; in the second, an emission of a quantum in one particular direction.

An explicit formulation involves the transitions between the $(2J+1)$ m-states of each level. ($\mathbf{J}$ is total angular momentum of a level, the m are the eigenvalues of J_z.) We designate the states of the nucleus as A_l, B_n, C_p; subscripts are values of m. For a given multipole order of the transition AB (or BC), the angular distribution of quanta emitted in a transition A_lB_n (or B_nC_p) depends only on $|\Delta m|$, where $\Delta m \equiv (p-n)$ or $(n-l)$; hence we write these distributions as $\varphi_{|n-l|}(\theta)$ and $f_{|p-n|}(\theta)$. The relative probabilities of the various transitions A_lB_n and B_nC_p are denoted by g_{ln} and G_{np}, respectively. Now suppose that the nuclei are initially oriented at random—i.e., all states A_l equally populated for any arbitrary axis of quantization. In the transition AB, the sum of the probabilities of all components A_lB_n with a given Δm is independent of Δm; hence the probability that a quantum emitted at an angle θ_1 with the axis has been emitted in a transition with given Δm is proportional to $\varphi_{|\Delta m|}(\theta_1)$. The relative populations of the B_n are then $\sum_l g_{ln}\varphi_{|n-l|}(\theta_1)$ and the angular distribution of radiation from the decay of state B_n alone is $\sum_p G_{np}f_{|p-n|}(\theta_2)$.

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[1] J. V. Dunworth, Rev. Sci. Inst. **11**, 167 (1940).
[2] V. F. Weisskopf, Ann. d. Physik **9**, 27 (1931).

Thus one might at first sight take as the total angular distribution of the second quantum the weighted sum of the angular distributions from each state B_n, given by

$$W(\theta_2)=\sum_n[\sum_l g_{ln}\varphi_{|n-l|}(\theta_1)] \times[\sum_p G_{np}f_{|p-n|}(\theta_2)]. \quad (1)$$

However, Eq. (1) seems incorrect since it has $W(\theta_2)$ depending not on the angle between the quanta, but on their directions with respect to the axis. This deeply offends one's sense of spectroscopic stability, according to which the choice of axis should affect the ease of a calculation but not the final result. As will be shown later, the point overlooked in obtaining Eq. (1) is the fact that if the first quantum is emitted in a definite direction then the phases of the states B_n are related; hence these states do not radiate independently of each other and the last step in obtaining Eq. (1) is based on a fallacy.

The same difficulty with phases beset the older quantum-theory calculations of the polarization of resonance radiation. There it was found that a physically reasonable result was obtained for one particular choice of axis (parallel to the electric vector of incident plane polarized light, or parallel to the direction of propagation of a circularly polarized beam); this was later justified quantum mechanically. Analogously, it will be shown that in our case the "naive" theory gives the correct result if one takes Oz, the axis of quantization, along the direction of propagation of the first quantum, since in this case the phases of the B_n are random. Furthermore, with this choice of axis the observed quanta are emitted only in transitions for which $\Delta m=\pm 1$. (Proposition 2a, later.) Hence, on setting $\theta_1=0$ and $\theta_2=\theta$, and dropping the constant factor $\varphi_1(0)$, the relative probability that the second quantum will be emitted at an angle θ with the first is

$$W(\theta)=\sum_n(g_{n+1,n}+g_{n-1,n})(\sum_p G_{np}f_{|p-n|}(\theta)). \quad (2)$$

We have discussed the naive treatment because, although incorrect, it suggests a simplified form (Eq. (2)) of the result of the exact treatment (Eq. (10)) which reduces by at least one order of magnitude the labor required for explicit calculation of $W(\theta)$, as well as providing additional physical insight.

THEORY

The exact treatment is based on the time dependent Schrödinger equation (Eq. (4)) which deals with the interdependence of probability amplitudes rather than of their absolute squares.

Emission or absorption of a quantum constitutes a transition between two states of the system nucleus-plus-quantized-radiation-field, and is caused by the small matter-radiation coupling, H, in the Hamiltonian of the whole system. Since the velocities of the heavy particles in the nucleus are small compared with the velocity of light we take

$$H=-\int \mathbf{i}\cdot\mathbf{A}dv=-c^{-1}\int \rho\dot{\mathbf{r}}\cdot\mathbf{A}dv,$$

where ρ and $\mathbf{i}$ are the charge and current densities associated with the given transition of the nucleus. The radiation field vector potential, $\mathbf{A}$, is taken in the gauge in which the scalar potential is zero.

This radiation field is quantized by quantizing the amplitudes of its normal modes. These modes might, for example, be plane or spherical waves; the particular form is a matter of convenience and depends on arbitrarily chosen boundary conditions. Running plane waves are the most straightforward in dealing with directions of propagation. The radiation field, then, is represented by an assembly of simple harmonic oscillators, each of which corresponds to a running plane wave of frequency ν, direction of propagation $\boldsymbol{\kappa}_0$, and polarization $\mathbf{e}$. (The latter are both unit vectors.) The vector potential of such an oscillator is given by $\mathbf{A}=q\mathbf{e}\exp(i\boldsymbol{\kappa}\cdot\mathbf{r})$ where $\boldsymbol{\kappa}=\kappa\boldsymbol{\kappa}_0=2\pi\nu c^{-1}\boldsymbol{\kappa}_0$. The time dependent amplitude of the oscillator, q, is independent of $\mathbf{r}$. The matrix elements of H for a given transition are calculated from

$$H=-qc^{-1}\mathbf{e}\cdot\int \rho\dot{\mathbf{r}}\exp(i\boldsymbol{\kappa}\cdot\mathbf{r})dv, \quad (3)$$

where $\boldsymbol{\kappa}_0$ and $\mathbf{e}$ refer to the quantum emitted in the transition.

(The procedure now followed, from Eq. (4) to Eq. (7) inclusive, is very similar to that used in reference 2; we therefore omit many details.)

Let n_i be the occupation number of the ith field oscillator. Then if the first and second quanta are emitted into the ρth and σth field oscillators, respectively, the necessary states of the system may be specified, and their probability amplitudes written, as follows:

State	Probability amp.
$(A_l, n_1\cdots n_\rho\cdots n_\sigma\cdots)$	a_l
$(B_n, n_1\cdots n_\rho+1\cdots n_\sigma\cdots)$	$b_{n\rho}$
$(C_p, n_1\cdots n_\rho+1\cdots n_\sigma+1\cdots)$	$c_{p\rho\sigma}$.

The time dependence of the probability amplitudes is given by

$$-i\hbar\dot{a}_j=\sum_k H_{jk}a_k+E_ja_j, \tag{4}$$

where a_j and a_k are any of the above a_l, $b_{n\rho}$, or $c_{p\rho\sigma}$. Writing $(A_l\cdots n_\rho\cdots|H|B_n\cdots n_\rho+1\cdots)$ $\equiv(A_l|H_\rho|B_n)$, etc., Eq. (4) becomes

$$-i\hbar\dot{a}_l=\sum_{n,\rho}(A_l|H_\rho|B_n)b_{n\rho}, \tag{5a}$$

$$-i\hbar\dot{b}_{n\rho}=\sum_l(A_l|H_\rho|B_n)^*a_l+\sum_{\sigma,p}(B_n|H_\sigma|C_p)c_{p\rho\sigma}+h(\nu_\rho-\nu_{AB})b_{n\rho}, \tag{5b}$$

$$-i\hbar\dot{c}_{p\rho\sigma}=\sum_n(B_n|H_\sigma|C_p)^*b_{n\rho}+h(\nu_\rho+\nu_\sigma-\nu_{AC})c_{p\rho\sigma}. \tag{5c}$$

Here $h\nu_{AB}=E_A-E_B$, etc., and E_A is taken as zero. The following solutions to (5) are then assumed, subject to determination of the unknown quantities therein so that (5) is satisfied:

$$a_l=\alpha_l\exp[-2\pi\Gamma(A_l)t],\quad b_{n\rho}=\sum_l\beta_{ln\rho}(\exp[-2\pi\Gamma(A_l)t]-\exp[-2\pi\Gamma_\rho(B_n)t]), \tag{6a, b}$$

$$c_{p\rho\sigma}=\sum_l\epsilon_{l\rho\sigma}(\exp[-2\pi\Gamma(A_l)t]-\exp[-2\pi\Gamma_{\rho\sigma}(C_p)t])$$
$$+\sum_n\delta_{n\rho\sigma}(\exp[-2\pi\Gamma_\rho(B_n)t]-\exp[-2\pi\Gamma_{\rho\sigma}(C_p)t]). \tag{6c}$$

Substitution of Eq. (6) into Eq. (5) (most conveniently carried out by working upwards from Eqs. (5c) and (6c)) gives the following results:

$$\begin{aligned}
&\Gamma(A_l)=\gamma(A_l),\quad \Gamma_\rho(B_n)=i(\nu_{AB}-\nu_\rho)+\gamma(B_n),\quad \Gamma_{\rho\sigma}(C_p)=i(\nu_{AC}-\nu_\rho-\nu_\sigma),\\
&h\beta_{ln\rho}=\alpha_l(A_l|H_\rho|B_n)^*/(i[\Gamma(A_l)-\gamma(B_n)]+\nu_{AB}-\nu_\rho)\\
&h\epsilon_{l\rho\sigma}=\sum_n\beta_{ln\rho}(B_n|H_\sigma|C_p)^*/[\nu_{AC}-\nu_\rho-\nu_\sigma+i\Gamma(A_l)],\\
&h\delta_{n\rho\sigma}=-(B_n|H_\sigma|C_p)^*\sum_l\beta_{ln\rho}/[\nu_{AC}-\nu_\rho-\nu_\sigma+i\Gamma_\rho(B_n)].
\end{aligned} \tag{7}$$

The α_l are entirely undetermined, corresponding to the fact that their values constitute the initial conditions of the problem. The $\gamma(A_l)$ are given by $h^2\gamma(A_l)=\sum_n\pi F_{ln}(\nu_{AB})$ where $F_{ln}(\nu)d\nu$ $=\sum_\rho|(A_l|H_\rho|B_n)|^2$. ($\sum_\rho$ indicates summation from $\nu_\rho=\nu$ to $\nu+d\nu$, and over all directions and polarizations.) Hence $4\pi\gamma(A_l)$ is the probability of radiative transition from the state A_l. An exactly analogous statement holds for $\gamma(B_n)$. The $\gamma(A_l)$ and $\gamma(B_n)$ are independent of l and n, as would be expected from symmetry considerations.

From Eq. (3),

$$(A_l|H_\rho|B_n)=-c^{-1}(n_\rho|q|n_\rho+1)(A_l|H(\mathbf{\kappa}_0{}^\rho,\mathbf{e}^\rho)|B_n)$$

where

$$(A_l|H(\mathbf{\kappa}_0{}^\rho,\mathbf{e}^\rho)|B_n)\equiv\mathbf{e}^\rho\cdot(A_l|\dot{\mathbf{r}}\exp(i\mathbf{\kappa}^\rho\cdot\mathbf{r})|B_n). \tag{8}$$

(Similarly for $(B_n|H_\sigma|C_p)$.) H_ρ and $H(\mathbf{\kappa}_0\rho,\mathbf{e}\rho)$ are effectively independent of ν_ρ over the line breadth range of frequencies. Remembering this and calculating a_l, $b_{n\rho}$, and $c_{p\rho\sigma}$ by substitution of Eq. (7) into Eq. (6), we find that each probability amplitude is the product of two factors. The first depends on t and some or all of ν_ρ, ν_σ, $\gamma(B_n)$ and $\gamma(A_l)$. The second depends on the initial conditions and on the matrix elements of $H(\mathbf{\kappa}_0, \mathbf{e})$. As a result $W_{\rho\sigma}$, the probability of emission of quanta into the oscillators σ and ρ, will be the product of two factors of the same type. ($W_{\rho\sigma}$ is given by limit $(t\rightarrow\infty)\sum_p\langle|c_{p\rho\sigma}|^2\rangle_{\text{Av}}$.) In calculating $W(\mathbf{\kappa}_0', \mathbf{e}', \mathbf{\kappa}_0'', \mathbf{e}'')$, the probability of emission of two quanta characterized, respectively, by $\mathbf{\kappa}_0'$, $\mathbf{e}'$ and $\mathbf{\kappa}_0''$, $\mathbf{e}''$, one integrates $W_{\rho\sigma}$ over ν_ρ and ν_σ. The second factor in $W_{\rho\sigma}$ is unaltered by this, and the integration of the first factor introduces no new dependence on $\mathbf{\kappa}_0'$, $\mathbf{e}'$, $\mathbf{\kappa}_0''$, $\mathbf{e}''$, since the frequency distribution of oscillators is independent of $\mathbf{\kappa}_0$ and $\mathbf{e}$. Hence the first factor in $W_{\rho\sigma}$ and its parents, the first factors in the probability amplitudes, are useless since we are interested only in the *relative* variation of $W(\mathbf{\kappa}_0', \mathbf{e}', \mathbf{\kappa}_0'', \mathbf{e}'')$ with $\mathbf{\kappa}_0'$, $\mathbf{e}'$, $\mathbf{\kappa}_0''$, $\mathbf{e}''$. We therefore drop these factors and write the abbreviated probability amplitudes as a_l, b_n, c_p:

$$a_l=\alpha_l, \quad b_n=\sum_l \alpha_l(A_l|H(\mathbf{\kappa}_0', \mathbf{e}')|B_n)^*, \tag{9a, b}$$

$$c_p=\sum_n b_n(B_n|H(\mathbf{\kappa}_0'', \mathbf{e}'')|C_p)^*=\sum_l \alpha_l[\sum_n(A_l|H(\mathbf{\kappa}_0', \mathbf{e}')|B_n)^*(B_n|H(\mathbf{\kappa}_0'', \mathbf{e}'')|C_p)^*]. \quad \text{(9c}') \text{ (9c}'')$$

The α_l specify the initial distribution of nuclei among the states A_l. Normally this will be completely random. This is equivalent to saying that $\alpha_l=\exp(i\delta_l)$ (omitting normalization) where the δ_l are random—that is, where $\langle\exp[i(\delta_{l_1}-\delta_{l_2})]\rangle_{\text{Av}}=\delta_{l_1l_2}$. ($\langle\ \rangle_{\text{Av}}$ hereafter indicates an averaging over all nuclei, i.e. over all values of the δ_l.) Any result is to be averaged over the δ_l. Furthermore, to each $\mathbf{\kappa}_0$ there correspond two arbitrary orthogonal $\mathbf{e}$'s; and since a γ-ray counter cannot differentiate between these two polarizations any final result must be summed over $\mathbf{e}'$ and $\mathbf{e}''$.

Thus at any given time the relative probability of two quanta having been emitted in the directions $\mathbf{\kappa}_0'$, $\mathbf{\kappa}_0''$, in the solid angles $d\omega'$, $d\omega''$, is $Wd\omega'd\omega''$ where

$$W=\sum_{p,e',e''}\langle|c_p|^2\rangle_{\text{Av}}=\sum_{l,p,e',e''}|\sum_n(A_l|H(\kappa_0', e')|B_n)^*(B_n|H(\kappa_0'', e'')|C_p)^*|^2 \tag{10}$$

by Eq. (9c''). Since W is not an absolute probability, we shall occasionally drop constant factors from it without further comment.

The interference of probabilities mentioned in the Introduction is evident in Eq. (10). A nucleus may pass from the state A_l to C_p by way of several intermediate states B_n; but the probability of the transition A_lC_p is not the sum of the individual transition probabilities for the several routes $A_lB_nC_p$. The interference arises because the contributions to c_p from these different routes are summed before squaring rather than afterwards. If W is written down using Eq. (9c') instead of Eq. (9c'') it will be seen that the cross terms vanish when $\sum_{e'}\langle b_nb_k^*\rangle_{\text{Av}}=0$ for $n\neq k$, i.e. when the phases of the b_n are random.

We now state two propositions (previously referred to) which enable us to reduce Eq. (10) to Eq. (2), and which hold for all multipole orders (proofs in Appendix I):

1. The value of W is independent of the direction which we choose for the axis of quantization, hence we may choose this direction to suit our convenience.

2. If we take the axis along the direction of propagation of the first quantum, then: (2a) $l-n=\pm1$; (2b) $\sum_{e'}\langle b_nb_k^*\rangle_{\text{Av}}=0$ for $n\neq k$.

Proposition (1) involves simply a slight generalization of the principle of spectroscopic stability. (2a) is useful in proving (2b) as well as in calculating W; its physical significance was mentioned in the introduction. It might be noted that while (2b) makes the phases of the b_n random when we do not observe the polarization of the first quantum, they are also random if the first quantum is circularly polarized.

These propositions enable us to put $\mathbf{\kappa}_0=\mathbf{k}$ and write

$$W=\sum_{nlpe'e''}|(A_l|H(\mathbf{k}, \mathbf{e}')|B_n)|^2|(B_n|H(\mathbf{\kappa}_0'', \mathbf{e}'')|C_p)|^2, \quad l=n\pm1. \tag{11}$$

The interference has been removed. (In a sense one may say that the degeneracy of level B has been removed, since if B_n were nondegenerate there would have been no interference in the first

TABLE I. R/Q, *for both transitions dipole.*

	$\Delta J=-1$	$\Delta J=0$	$\Delta J=1$
$\Delta j=-1$	$\frac{1}{13}$	$\frac{-(2J-1)}{(14J+13)}$	$\frac{J(2J-1)}{(26J^2+67J+40)}$
$\Delta j=0$	$\frac{-(2J+3)}{(14J+1)}$	$\frac{(2J-1)(2J+3)}{(12J^2+12J+1)}$	
$\Delta j=1$	$\frac{(J+1)(2J+3)}{(26J^2-15J-1)}$		

TABLE II. R/Q, *for first transition quadrupole, second dipole.*

	$\Delta J=1$	$\Delta J=0$	$\Delta J=-1$
$\Delta j=2$	$\frac{-3}{29}$	$\frac{3(2J+3)}{(26J-3)}$	$\frac{-3(J+1)(2J+3)}{(58J^2-23J+3)}$
$\Delta j=1$	$\frac{3(J-5)}{(55J+61)}$	$\frac{-3(2J+3)(J-5)}{(58J^2+49J-15)}$	$\frac{3(2J+3)(J-5)}{(110J^2-49J+15)}$
$\Delta j=0$	$\frac{(2J-3)(2J+5)}{(36J^2+92J+61)}$	$\frac{-(2J-3)(2J+5)}{5(4J^2+4J-1)}$	$\frac{(2J-3)(2J+5)}{(36J^2-20J+5)}$
$\Delta j=-1$	$\frac{3(2J-1)(J+6)}{(110J^2+269J+174)}$	$\frac{-3(2J-1)(J+6)}{(58J^2+67J-6)}$	$\frac{3(J+6)}{(55J-6)}$
$\Delta j=-2$	$\frac{-3J(2J-1)}{(58J^2+139J+84)}$	$\frac{3(2J-1)}{(26J+29)}$	$\frac{-3}{29}$

place.) The quantities $\sum_{e'}|(A_l|H(\boldsymbol{\kappa}_0'\mathbf{e}')|B_n)|^2$ and $\sum_{e''}|(B_n|H(\boldsymbol{\kappa}_0''\mathbf{e}'')|C_p)|^2$ contain in their dependence on $\boldsymbol{\kappa}_0'$ and $\boldsymbol{\kappa}_0''$ the angular distributions of quanta emitted in the transitions A_lB_n and B_nC_p, and when integrated over all directions of emission are proportional to the total probabilities of these transitions. Hence

$$\sum_{e'}|(A_l|H(\boldsymbol{\kappa}_0', \mathbf{e}')|B_n)|^2 \cong g_{ln}\,\varphi_{|n-l|}(\theta_1), \quad \sum_{e''}|(B_n|H(\boldsymbol{\kappa}_0'', \mathbf{e}'')|C_p)|^2 \cong G_{np}\,f_{|p-n|}(\theta_2). \tag{12}$$

(φ and f normalized to any convenient constant.) Putting Eq. (12) into Eq. (11), setting $\theta_1=0$ and $\theta_2=\theta$, and dropping the constant $\varphi_1(0)$, we obtain Eq. (2) as was promised.

The g_{ln}, G_{np}, and $f_{|p-n|}(\theta)$ depend on the multipolarity of the transition to which they refer, and this multipolarity is in turn determined by the predominating term in the expansion of exp $(i\boldsymbol{\kappa}\cdot\mathbf{r})$ in $H(\boldsymbol{\kappa}_0, \mathbf{e})$. In Appendix II the expanded form of $H(\boldsymbol{\kappa}_0, \mathbf{e})$ is given and the method of calculating the $f_{|p-n|}(\theta)$ is outlined. The resulting angular distributions (of course identical with those obtained by various other methods[3]) are as follows:

Dipole radiation: $f_1(\theta)=1+\cos^2\theta$, $f_0(\theta)=2(1-\cos^2\theta)$.
Quadrupole radiation: $f_2(\theta)=1-\cos^4\theta$, $f_1(\theta)=1-3\cos^2\theta+4\cos^4\theta$, $f_0(\theta)=6(\cos^2\theta-\cos^4\theta)$.

(Note that in accordance with proposition (2a), $f_0(0)=f_2(0)=0$.) The g_{ln} and G_{np} for dipole or quadrupole transitions may be obtained from the matrix elements in reference 3, pp. 63 or 95.

CALCULATIONS

We designate the angular momenta of the levels A, B and C as $J-\Delta j$, J, and $J+\Delta J$, respectively, so that the angular momentum changes in the first and second transitions are Δj and ΔJ.

The g_{ln} are usually given as functions of the initial quantum numbers $J-\Delta j$ and l but are more convenient for use here as functions of J and n. Making this change and defining

$$d_n=g_{n+1,\,n}+g_{n-1,\,n},\quad D_n=G_{n,\,n+1}+G_{n,\,n-1},\quad E_n=G_{n,\,n+2}+G_{n,\,n-2},$$

we have

$$W=f_0(\theta)\sum_n d_nG_{nn}+f_1(\theta)\sum_n d_nD_n+f_2(\theta)\sum_n d_nE_n.$$

[3] E. U. Condon and G. H. Shortley, *Theory of Atomic Spectra* (Cambridge University Press, 1935), for example, Chap. IV.

TABLE III. R/Q, *for both transitions quadrupole.*

	$\Delta J=0$	$\Delta J=-1$	$\Delta J=-2$
$\Delta j=-2$	$\frac{-(2J-3)(2J+1)}{(2J+3)(6J+5)}$	$\frac{(J+3)}{(17J+15)}$	$\frac{1}{8}$
$\Delta j=-1$	$\frac{(5J-2)(2J-3)(2J+5)}{(20J^3+52J^2+41J+6)}$	$\frac{-(17J^2+17J-30)}{(35J^2+35J+6)}$	$\frac{(J-2)}{(17J+2)}$
$\Delta j=0$	$\frac{-(2J-3)(4J^2+4J-7)(2J+5)}{(2J-1)(2J+3)(4J^2+4J-1)}$	$\frac{(5J+7)(2J-3)(2J+5)}{(20J^3+8J^2-3J+3)}$	$\frac{-(2J+1)(2J+5)}{(2J-1)(6J+1)}$
$\Delta j=1$		$\frac{-(2J+3)(17J^3+69J^2-77J-105)}{(70J^4-9J^3-73J^2-27J-9)}$	$\frac{(2J+3)(J^2+18J+5)}{(34J^3-57J^2+8J+3)}$
$\Delta j=2$			$\frac{(J+1)(2J+3)(2J^2-9J+1)}{(2J-1)(16J^3-42J^2+29J+3)}$

	$\Delta J=2$	$\Delta J=1$
$\Delta j=-2$	$\frac{J(2J-1)(2J^2+13J+12)}{(2J+3)(16J^3+90J^2+161J+84)}$	$\frac{(2J-1)(J^2-16J-12)}{(34J^3+159J^2+224J+96)}$
$\Delta j=-1$		$\frac{-(2J-1)(17J^3-18J^2-164J-24)}{(70J^4+289J^3+374J^2+188J+24)}$

TABLE IV. S/Q, *for both transitions quadrupole.*

	$\Delta J=0$	$\Delta J=-1$	$\Delta J=-2$
$\Delta j=-2$	$\frac{4(J-1)(2J-3)}{3(2J+3)(6J+5)}$	$\frac{-4(2J-3)}{3(17J+15)}$	$\frac{1}{24}$
$\Delta j=-1$	$\frac{-16(J-1)(2J-3)(2J+5)}{3(20J^3+52J^2+41J+6)}$	$\frac{16(2J-3)(2J+5)}{3(35J^2+35J+6)}$	$\frac{-4(2J+5)}{3(17J+2)}$
$\Delta j=0$	$\frac{16(J-1)(J+2)(2J-3)(2J+5)}{3(2J-1)(2J+3)(4J^2+4J-1)}$	$\frac{-16(J+2)(2J-3)(2J+5)}{3(20J^3+8J^2-3J+3)}$	$\frac{4(J+2)(2J+5)}{3(2J-1)(6J+1)}$
$\Delta j=1$		$\frac{16(2J-3)(2J+3)(J+2)(2J+5)}{3(70J^4-9J^3-73J^2-27J-9)}$	$\frac{-4(2J+3)(J+2)(2J+5)}{3(34J^3-57J^2+8J+3)}$
$\Delta j=2$			$\frac{(J+1)(2J+3)(J+2)(2J+5)}{3(2J-1)(16J^3-42J^2+29J+3)}$

	$\Delta J=2$	$\Delta J=1$
$\Delta j=-2$	$\frac{J(2J-1)(J-1)(2J-3)}{3(2J+3)(16J^3+90J^2+161J+84)}$	$\frac{-4(2J-1)(J-1)(2J-3)}{3(34J^3+159J^2+224J+96)}$
$\Delta j=-1$		$\frac{16(2J-1)(J-1)(2J-3)(2J+5)}{3(70J^4+289J^3+374J^2+188J+24)}$

If the second transition is dipole,

$$W=\sum_n d_n(D_n+2G_{nn})+\cos^2\theta\sum_n d_n(D_n-2G_{nn})\equiv Q+R\cos^2\theta, \tag{13a}$$

while if it is quadrupole,

$$W=\sum_n d_n(D_n+E_n)+\cos^2\theta\sum_n d_n(2G_{nn}-3D_n)+\cos^4\theta\sum_n d_n(-2G_{nn}+4D_n-E_n)\equiv Q+R\cos^2\theta+S\cos^4\theta. \tag{13b}$$

Dropping a constant factor, our correlation function for calculation is

$$\mathbf{W}(\theta)=1+(\mathbf{R}/\mathbf{Q})\cos^2\theta+(\mathbf{S}/\mathbf{Q})\cos^4\theta.$$

The g_{ln}, G_{np}, d_n, D_n and E_n are linear functions of n^2 for dipole transitions and quadratic functions of n^2 for quadrupole transitions, so that the summations indicated may involve $\sum_n 1, n^2, n^4, n^6, n^8$ where $n=-J, \cdots J$. These sums (for n integral or half-integral) are given by[4]

[4] The author is indebted to Dr. J. R. Stehn for a derivation of the formulae for $\sum n^6$ and $\sum n^8$.

$$\sum 1=2J+1,\quad 3\sum n^2=J(J+1)(2J+1),\quad 15\sum n^4=J(J+1)(2J+1)(3J^2+3J-1),$$
$$21\sum n^6=J(J+1)(2J+1)(3J^4+6J^3-3J+1),$$
$$45\sum n^8=J(J+1)(2J+1)(5J^6+15J^5+5J^4-15J^3-J^2+9J-3).$$

In Q, R, and S, each of these sums occurs multiplied by a polynomial in J. When the summations are completed for the most complicated case (both transitions quadrupole), Q, R and S are nearly always of the form $J(J+1)(2J+1)$ times a sixth-degree polynomial in J. These polynomials are found to contain a number of linear factors such that R/Q and S/Q are ratios of much smaller polynomials.

Since the coupling term is Hermitian, it is obvious from Eq. (10) that if two pairs of transitions are each other's inverses, i.e., differ by an interchange of initial and final angular momenta and radiation processes, and have the same intermediate angular momentum, then their correlation functions are identical. However, the processes of calculating W for a sequence and for its inverse are distinctly different; this provides a valuable check on the calculations. $1\to2\to0$ (dipole, quadrupole) and $0\to2\to1$ (quadrupole, dipole), for example, are two such sequences. For the second of these, (13a) shows that there is no $\cos^4\theta$ term in W; for the first, this may be shown only by proving explicitly that $S=0$.

The functions R/Q and S/Q are given in Tables I to IV. Any sequence not shown is to be obtained from its inverse. These functions are shown graphically in Figs. 1 to 5. Each curve begins at the lowest value of J permitted by the selection rules. (See Appendix II.) Beyond $J=7$, each curve is connected by an oblique line to its asymptote.

Discussion

Some generalizations to higher multipole orders may be made. Electric and magnetic multipoles of the same order will not be distinguishable, since the g, G, f and φ are the same for each. $W(\theta)$ will in general be a polynomial in $\cos^2\theta$ of degree l, where the lowest multipole present is a 2^l-pole. It will be noted that for dipole and quadrupole radiation $W(\theta)$ is a constant for $J=0,\ \frac{1}{2}$, and is linear in $\cos^2\theta$ for $J=1,\ 3/2$; one suspects that for all multipoles $\cos^{2k}\theta$ may appear only when $J\geqslant k$.

Our procedure is changed only formally, and the final results not at all, if one or both transitions are absorptions rather than emissions. Thus, for example, W should give the angular distribution of resonance radiation excited by a unidirectional unpolarized beam of light.

We have tacitly assumed that during τ, the lifetime of level B, J_κ is constant. But, for example, the atomic electrons produce at the nucleus a field $\mathbf{H}$ oriented at random with respect to $\boldsymbol{\kappa}$; $\mathbf{J}$ precesses about $\mathbf{H}$ with the Larmor precession frequency $\nu=\mu H/Jh$. Roughly, J_κ will change by $\pm\hbar$ for half the nuclei when $\mathbf{J}$ has precessed through an angle $(3\pi/4J)$, i.e., in time $t=(3/8J\nu)=(3h/8\mu H)$ sec. The hyperfine structure splitting of an atomic level is $\Delta\nu\sim(3\mu H/hc)\sim1/tc$ cm^{-1}; for $\Delta\nu\leq1$ cm^{-1} (which is usually true), $t\geqslant3\times10^{-11}$ sec. For three levels

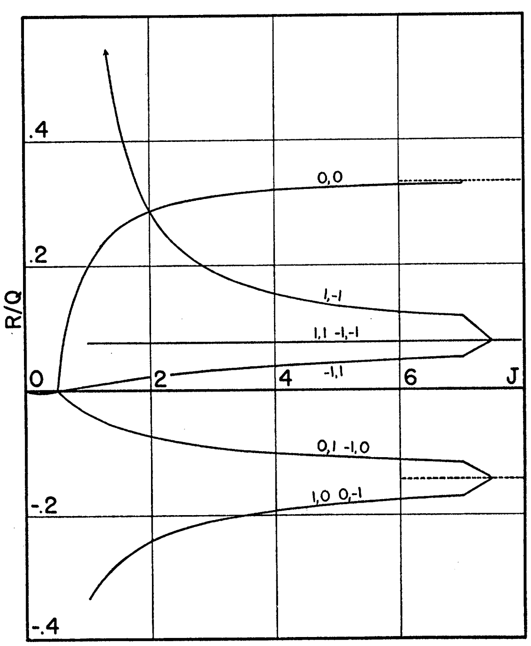

FIG. 1. R/Q as function of J; both transitions dipole. Curves labeled Δj, ΔJ. Point not shown: on $(1,-1)$ curve, $J=1$, $R/Q=1$.

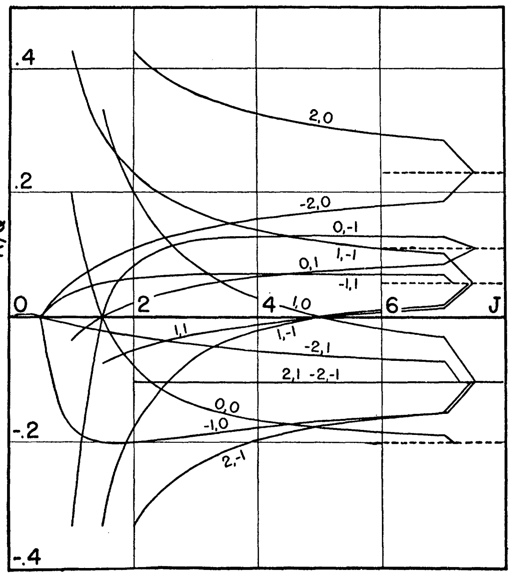

FIG. 2. R/Q as function of J; first transition quadrupole, second dipole. Curves labeled Δj, ΔJ.

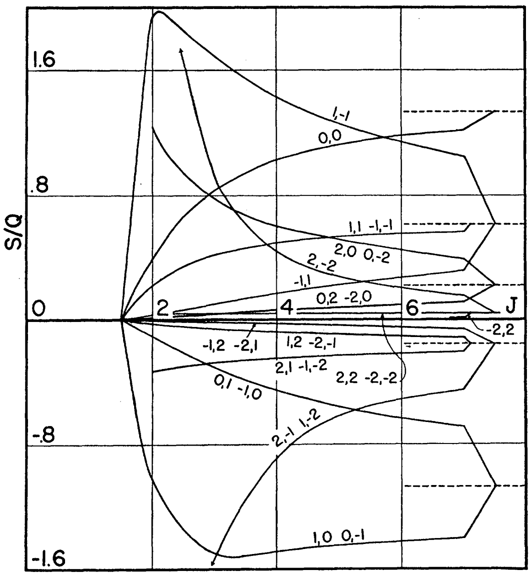

FIG. 4. S/Q as function of J; both transitions quadrupole. Curves labeled Δj, ΔJ. Points not shown: on $(2, -2)$ curve, $J=2$, $S/Q=4$; on $(2, -1)$, $(1, -2)$ curve, $J=2$, $5/2$, $S/Q=-16/3$, $-80/33$.

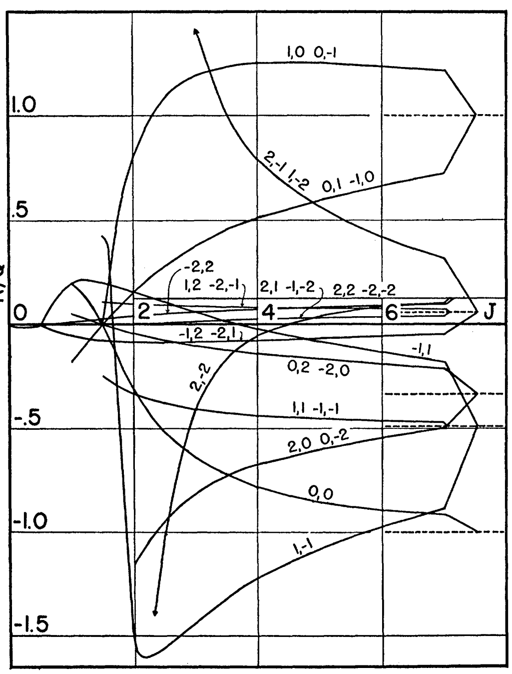

FIG. 3. R/Q as function of J; both transitions quadrupole. Curves labeled Δj, ΔJ. Points not shown: on $(2, -2)$ curve, $J=2$, $R/Q=-3$; on $(2, -1)$, $(1, -2)$ curve, $J=2$, $5/2$, $R/Q=5$, $25/11$.

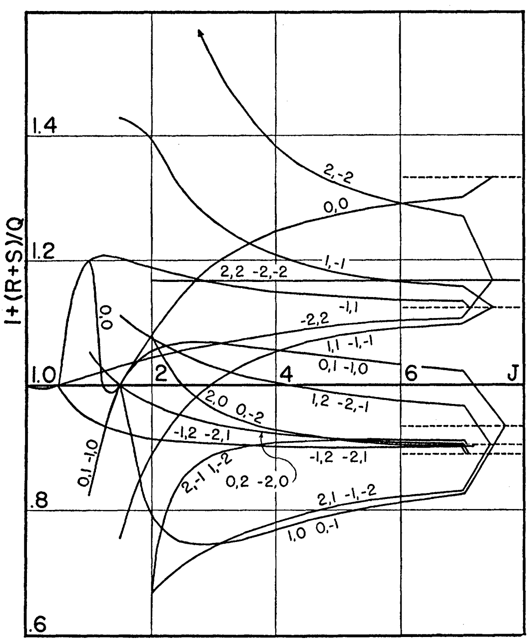

FIG. 5. $1+(R+S)/Q=W(0 \text{ or } \pi)/W(\pi/2)$, as function of J; both transitions quadrupole. Curves labeled Δj, ΔJ. Point not shown: on $(2, -2)$ curve, $J=2$, $1+(R+S)/Q=2$.

of RaC′ and ThC′ for which τ is known to be of the order of 5×10^{-13} sec.,[5] it is then probably true that $t \geqslant 60\tau$, which is satisfactory. However, a nucleus of mass 200 emitting a 1-Mev quantum recoils with 2.5 volts energy and if unimpeded

[5] H. A. Bethe, Rev. Mod. Phys. 9, 69 (1937).

would travel 8×10^{-8} cm, or two atomic diameters, in the above τ. It is hard to judge the effect of this motion on the time average **H**. As regards light nuclei, the γ-lifetimes of five excited states formed by proton capture[5] are much shorter and recoil energies much higher (by factors 10^{-2} to 10^{-4} and 10^2 to 10^4, respectively) than in the above cases; the distances traveled in τ range from 0.08 to 4.5×10^{-8} cm. Reorientation is very improbable here unless the fields encountered in the recoil motion are larger, by 10^3 to 10^5, than the static field indicated by hyperfine structure.

The condition $t\gg\tau$ (criterion that J_κ=constant, as discussed above) is identical with the condition that the splitting of level B under the influence of **H** be much less than the radiation width of B.

As already mentioned, an attempt is being made by Getting to observe the angular correlation effect, using the γ-rays M and X of ThPb.[6] The counting rates involved are low because of the inefficiency of γ-ray counters and because each counter subtends a comparatively small solid angle at the source; during the necessarily long runs the characteristics of the counters change so that no final results are yet available.

The author is greatly indebted to Professor J. H. Van Vleck for several discussions of important aspects of the questions here treated.

Appendix I

Proposition (1)

a_l, b_n, c_p are probability amplitudes of eigenstates with a fixed axis of quantization; let the corresponding probability amplitudes for some arbitrary axis be α_l, β_n, γ_p. (α, β, γ are not the quantities previously designated by these letters!) We first indicate the proof that $\sum_p|c_p|^2=\sum_p|\gamma_p|^2$ if the α_l which enter into the calculation of the γ_p are determined by a unitary transformation from the initially given a_l. The usual form of the principle of spectroscopic stability (reference 3, Eq. (2²25)), applied to our case, only shows that the total transition probability is independent of the axis of quantization when the states of the initial level have equal amplitudes and random phases; thus it only tells us that $\langle\sum_n|b_n|^2\rangle_{\text{Av}}=\langle\sum_n|\beta_n|^2\rangle_{\text{Av}}$. For the transition BC we need a generalized form for arbitrary initial states. Following Condon and Shortley[3] it is easily shown that

$$\sum_p|\gamma_p|^2=\sum_{n,k,p} b_n^*b_k(B_n|H|C_p)(B_k|H|C_p)^*=\sum_p|c_p|^2,$$

which may be written in their notation by putting $b_n=(\Gamma_b'|)$, $\beta_n=(\Delta_b'|)$, $c_p=(\Gamma_c'|)$, $(B_n|H|C_p)=(\Gamma_b'|H|\Gamma_c')$, etc.

As concerns the unitary transformation from the a_l to the α_l, this is trivial for our particular initial conditions; it is physically obvious, or may be shown by making the transformation, that if $\langle a^*_{l_1}a_{l_2}\rangle_{\text{Av}}=\delta_{l_1l_2}$ then $\langle\alpha^*_{l_1}\alpha_{l_2}\rangle_{\text{Av}}=\delta_{l_1l_2}$. Hence the α_l also are equal in amplitude and random in phase and averages of any quantity over the phases of the a_l or of the α_l are equivalent processes. Thus $W=\sum\langle|c_p|^2\rangle_{\text{Av}}=\sum\langle|\gamma_p|^2\rangle_{\text{Av}}$ where the γ_p are calculated from the α_l in the same way as were the c_p from the a_l.

Proposition (2a)

Physically, $l-n=\pm1$ means that whenever a system is observed to emit a quantum in a particular direction, the projection of **J** on this direction has changed by $\mp\hbar$; therefore the projection of the angular momentum of the quantum on its direction of propagation is $\pm\hbar$. The latter result follows from Heitler's treatment of the eigenwaves of the radiation field in a spherical hohlraum.[7] Heitler chooses the set of eigenwaves which, when quantized, have definite values of the total angular momentum and its projection on Oz; these eigenwaves correspond to the various multipole radiation fields emitted in transitions of radiating matter between eigenstates of $\mathbf{J}^2$ and J_z. The angular dependence of the field strengths of these eigenwaves shows that the flow of energy along the axis of quantization Oz vanishes for all eigenwaves except those for which the projection of the angular momentum on the axis is $\mp\hbar$, and which therefore were emitted in a transition $\Delta J_z=\Delta m=\pm\hbar$.

[6] F. Oppenheimer, Proc. Camb. Phil. Soc. **32**, 328 (1936).
[7] W. Heitler, Proc. Camb. Phil. Soc. **32**, 112 (1936).

The proposition may also be proved easily for the explicit form of H given in Eq. (8):

$$(A_l|H(\boldsymbol{\kappa}_0', \mathbf{e}')|B_n)=\mathbf{e}'\cdot(A_l|\dot{\mathbf{r}}\exp(i\boldsymbol{\kappa}'\cdot\mathbf{r})|B_n)=\mathbf{e}'\cdot\sum_{D,s}(A_l|\dot{\mathbf{r}}|D_s)(D_s|\exp(i\boldsymbol{\kappa}'\cdot\mathbf{r})|B_n).$$

(D_s indicates all states except those of levels A and B.) As is permitted by proposition (1), we make Oz coincide with $\boldsymbol{\kappa}_0'$. Then $i\boldsymbol{\kappa}\cdot\mathbf{r}=i\kappa z$; z, and hence $\exp(i\kappa z)$, is diagonal in J_z, therefore $(D_s|\exp(i\kappa z)|B_n)=0$ unless $s=n$. Except when $l-n=\pm1$, $\mathbf{e}\cdot(A_l|\dot{\mathbf{r}}|D_n)$ is zero since $\mathbf{e}$ lies in the xy plane. Hence $(A_l|H(\boldsymbol{\kappa}_0', \mathbf{e}')|B_n)=0$ unless $l-n=\pm1$.

Proposition (2b)

By Eqs. (8) and (9b),

$$\sum_{e'}\langle b_n{}^*b_k\rangle_{\text{Av}}=\sum_l(A_l|\dot{\mathbf{r}}\exp(i\boldsymbol{\kappa}'\cdot\mathbf{r})|B_n)\cdot(\mathbf{e}_1'^*\mathbf{e}_1'+\mathbf{e}_2'^*\mathbf{e}_2')\cdot(A_l|\dot{\mathbf{r}}\exp(i\boldsymbol{\kappa}'\cdot\mathbf{r})|B_k)^*,$$

which is to be evaluated for $\boldsymbol{\kappa}_0'=\mathbf{k}$. The vectors $\mathbf{e}_1'$ and $\mathbf{e}_2'$ are the two arbitrary unit orthogonal (in general complex) polarization vectors associated with $\boldsymbol{\kappa}_0'$. $\mathbf{e}_1'$ and $\mathbf{e}_2'$ lie in the xy plane; it may easily be shown that $\mathbf{e}_1'^*\mathbf{e}_1'+\mathbf{e}_2'^*\mathbf{e}_2'\equiv\mathbf{ii}+\mathbf{jj}$. Since $(A_l|H(\boldsymbol{\kappa}_0', \mathbf{e}')|B_n)=0$ for $\boldsymbol{\kappa}_0'=\mathbf{k}$ unless $n=l\pm1$, we have $\langle b_n{}^*b_k\rangle_{\text{Av}}=0$ for $n\neq k$ unless $k=n\pm2$. The only quantity to be investigated, then, is

$$\begin{aligned}\sum_{e'}\langle b_{l+1}{}^*b_{l-1}\rangle_{\text{Av}}&=(A_l|\dot{\mathbf{r}}\exp(i\kappa z)|B_{l+1})\cdot(\mathbf{ii}+\mathbf{jj})\cdot(A_l|\dot{\mathbf{r}}\exp(i\kappa z)|B_{l-1})^*\\&=\{\textstyle\sum_D(A_l|\dot{x}|D_{l+1})(D_{l+1}|\exp(i\kappa z)|B_{l+1})\}\{\textstyle\sum_D(A_l|\dot{x}|D_{l-1})^*(D_{l-1}|\exp(i\kappa z)|B_{l-1})^*\}\\&\quad+\{\textstyle\sum_D(A_l|\dot{y}|D_{l+1})(D_{l+1}|\exp(i\kappa z)|B_{l+1})\}\\&\qquad\times\{\textstyle\sum_D(A_l|\dot{y}|D_{l-1})^*(D_{l-1}|\exp(i\kappa z)|B_{l-1})^*\}.\end{aligned}\tag{14}$$

Now $(A_l|\dot{\mathbf{r}}|D_{l\pm1})=2\pi i\nu_{AD}(A_l|\mathbf{r}|D_{l\pm1})$. From the matrices of x and y (e.g. reference 3, p. 63) one may verify that if $(A_l|x|D_{l+1})\equiv\alpha(l)$ then $(A_l|y|D_{l+1})=i\alpha(l)$; $(A_l|x|D_{l-1})^*=-\alpha^*(-l)$; and $(A_l|y|D_{l-1})^*=i\alpha^*(-l)$. From this it follows that the terms in x in Eq. (14) exactly cancel those in y; hence $\sum_{e'}\langle b^*{}_{l+1}b_{l-1}\rangle_{\text{Av}}=0$ as was to be shown.

Appendix II

Expanding,

$$\begin{aligned}H(\boldsymbol{\kappa}_0, \mathbf{e})&=\mathbf{e}\cdot\dot{\mathbf{r}}\exp(i\boldsymbol{\kappa}\cdot\mathbf{r})=\mathbf{e}\cdot\dot{\mathbf{r}}+2\pi i\nu c^{-1}\mathbf{e}\cdot\dot{\mathbf{r}}\mathbf{r}\cdot\boldsymbol{\kappa}_0+\cdots\\&=\mathbf{e}\cdot\dot{\mathbf{r}}+\pi i\nu c^{-1}[\mathbf{e}\cdot(\dot{\mathbf{r}}\mathbf{r}+\mathbf{r}\dot{\mathbf{r}})\cdot\boldsymbol{\kappa}_0+\mathbf{e}\cdot(\dot{\mathbf{r}}\mathbf{r}-\mathbf{r}\dot{\mathbf{r}})\cdot\boldsymbol{\kappa}_0]+\cdots.\end{aligned}$$

Since $(N'|\dot{\mathbf{r}}|N'')=2\pi i\nu_{N'N''}(N'|\mathbf{r}|N'')$, N' and N'' any two states of the nucleus, it follows easily that $(N'|\dot{\mathbf{r}}\mathbf{r}+\mathbf{r}\dot{\mathbf{r}}|N'')=2\pi i\nu_{N'N''}(N'|\mathbf{rr}|N'')$. By classical or quantum-vector analysis,

$$(N'|\dot{\mathbf{r}}\mathbf{r}-\mathbf{r}\dot{\mathbf{r}}|N'')\cdot\kappa_0=(N'|\mathbf{r}\times\dot{\mathbf{r}}|N'')\times\boldsymbol{\kappa}_0.$$

The multipole moments of a system with charge and current densities ρ and $\mathbf{i}$ are defined thus:

electric dipole moment $=\mathbf{P}=\int\rho\mathbf{r}dv$,
electric quadrupole moment $=\mathfrak{N}=\int\rho\mathbf{rr}dv$,
magnetic dipole moment $=\mathbf{M}=(1/2c)\int\rho\mathbf{r}\times\dot{\mathbf{r}}dv=(\frac{1}{2})\int\mathbf{r}\times\mathbf{i}dv$

and in terms of these, $H(\boldsymbol{\kappa}_0, \mathbf{e})\sim\mathbf{e}\cdot(\mathbf{P}+\mathbf{M}\times\boldsymbol{\kappa}_0+i\pi\nu c^{-1}\mathfrak{N}\cdot\boldsymbol{\kappa}_0+\cdots)$.

The vector part of the matrix elements of $\mathbf{P}$ and $\mathbf{M}$ (reference 3, p. 63) is one of three orthonormal vectors $\mathbf{T}(\Delta m)$. ($\mathbf{T}(i)\cdot\mathbf{T}(j)=\delta_{ij}$.) The J selection rule is $\Delta J=0$, ±1 with $0\rightarrow0$ forbidden. Similarly the matrix elements of $\mathfrak{N}$ (reference 3, p. 95) are proportional to one of five "orthonormal" dyadics $\mathfrak{K}(\Delta m)$ (i.e. $\mathfrak{K}(i):\mathfrak{K}(j)=\delta_{ij}$); and the selection rule on J is here $\Delta J=0$, ±1, ±2, with $0\rightarrow0$, $\frac{1}{2}\rightarrow\frac{1}{2}$, $1\rightleftharpoons0$ forbidden. The angular distributions of radiation emitted by these multipoles depend only on Δm and are proportional to $\sum_e|\mathbf{e}\cdot\mathbf{T}(\Delta m)|^2$, $\sum_e|\mathbf{e}\cdot\mathbf{T}(\Delta m)\times\boldsymbol{\kappa}_0|^2$ and $\sum_e|\mathbf{e}\cdot\mathfrak{K}(\Delta m)\cdot\boldsymbol{\kappa}_0|^2$. Since $\mathbf{e}_1{}^*\mathbf{e}_1+\mathbf{e}_2{}^*\mathbf{e}_2\equiv\mathbf{I}-\boldsymbol{\kappa}_0\boldsymbol{\kappa}_0$, ($\mathbf{I}\equiv\mathbf{ii}+\mathbf{jj}+\mathbf{kk}$, the identity dyadic), these functions may be written $\mathbf{T}^*(\Delta m)\cdot(\mathbf{I}-\boldsymbol{\kappa}_0\boldsymbol{\kappa}_0)\cdot\mathbf{T}(\Delta m)$, etc. Use of the explicit forms for $\mathbf{T}(\Delta m)$ and $\mathfrak{K}(\Delta m)$ gives the results in the text.