sections. The elastic collision cross section for hydrogen used in this theory is probably correct within 10 percent. Calculations of the theory indicated that this will not introduce more than 2 or 3 percent error in electric fields. The excitation and ionization efficiencies are very difficult to measure and the experimental error in the best measurements in hydrogen may be as high as 20 percent. These introduce an error of approximately 14 percent in the theoretical electric fields. These effects combine to give a possible error of 16 percent in theoretical fields and indicate a need for more precise collision cross-section measurements. The maximum error in the experimental electric fields in the 10-cm wavelength region is 5 percent and in pressure is 1 percent. The derivation of the equation for the distribution function implicitly assumed that each electron dropped back to zero energy after an inelastic collision. Since excitation takes place over a certain range of energy, this is not exactly correct, but the error which it introduces is small.

Equation (29), calculated from kinetic theory and using no gas discharge data other than collision crosssection measurements and involving no adjustable constants, predicts breakdown electric fields well within the limits of accuracy over a large range of pressure, container size and frequency.

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# Interference Effects in Gamma-Gamma Angular Correlations

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The theory of the directional correlation of successive nuclear gamma-rays is extended to include transitions in which mixtures of multipoles are present. For such cases interference effects can radically modify the angular correlation from what is predicted in the usual theory assuming pure multipole transitions. Correlation functions are tabulated for all possible cascade emissions in which one of the transitions is a mixture of magnetic dipole and electric quadrupole and the other either dipole or quadrupole. It is shown that the experimental data on Sr88 which had previously seemed anomalous can be consistently interpreted with the mixture theory developed here, but the agreement with the observed angular correlation in Pd<sup>106</sup> is not possible if the highest gamma-multipole order is assumed to be quadrupole.

#### I. INTRODUCTION

HE theory of the directional correlation of successive nuclear  $\gamma$ -rays has been treated in detail by Hamilton<sup>1</sup> and Goertzel.<sup>2</sup> Hamilton has given the basic quantum mechanical theory and has put the results of his calculations in a form which can be compared with experiment whenever the multipole orders of the radiation are dipole or quadrupole. (The distinction between the electric or magnetic character of the multipole radiation can be made in an angular correlation experiment only if the polarization of one or both of the  $\gamma$ -rays is specified.)<sup>3, 4</sup> The conditions which must be fulfilled in order that Hamilton's theory and tables can be validly applied are:

(1) That the natural line width of the intermediate nuclear state be much larger than the hyperfine splitting of that state, and (2) that the respective  $\gamma$ -transitions each correspond to pure

multipole radiation.

Goertzel has extended the theory to the case when (1) is not satisfied due to the presence of internal atomic fields or an externally applied magnetic field. However, he still retains assumption (2).

In view of the absence of any detailed knowledge of the wave functions and hence charge and current distributions for nuclear states, assumption (2) proves to be particularly convenient for angular correlation calculations since it can then be shown that the correlation function  $W(\theta)$  is independent of the intensities of the respective  $\gamma$ -rays. In fact, it is then possible (as is done in reference 1) to tabulate  $W(\theta)$  wholly in terms of "rotational information" such as the spins of the nuclear states involved and the known angular distributions of the energy radiated for the given multipole orders of the  $\gamma$ -rays. However, comparison of theory with experiment<sup>5-7</sup> shows good agreement in several cases, but poor agreement in others, notably for Pd<sup>106</sup> and Sr<sup>88</sup>. In these experiments condition (1) is satisfied

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 <sup>&</sup>lt;sup>1</sup> D. R. Hamilton, Phys. Rev. **58**, 122 (1940).
 <sup>2</sup> G. Goertzel, Phys. Rev. **70**, 897 (1946).
 <sup>3</sup> D. L. Falkoff, Phys. Rev. **73**, 518 (1948).
 <sup>4</sup> D. R. Hamilton, Phys. Rev. **74**, 782 (1948).

<sup>&</sup>lt;sup>5</sup> E. L. Brady and M. Deutsch, Phys. Rev. 72, 870 (1947); 74, 1541 (1948).

<sup>&</sup>lt;sup>6</sup> M. Deutsch and F. Metzger, Phys. Rev. 74, 1542 (1948).

<sup>&</sup>lt;sup>7</sup> M. Wiedenbeck, private communication.

TABLE I. Multipole vector potentials in the gauge  $\varphi = 0$ . The multipole order corresponding to given L is  $2^L$ . There are 2L+1 linearly independent "submultipoles"  $(L, M), M = -L, -(L-1), \dots, L-1, L$  for each electric or magnetic  $2^L$  pole. All potentials are normalized to  $1/\pi^2\hbar k$  quanta/sec.

$$Magnetic multipoles, A_{m}(L, M)$$

$$A_{z} = \frac{M}{(L(L+1))^{\frac{1}{2}}} R_{L}(kr) Y_{L}^{M}(\theta, \varphi) e^{-ickt}$$

$$A_{x} \pm iA_{y} = -\left(\frac{(L \mp M)(L \pm M + 1)}{L(L + 1)}\right)^{\frac{1}{2}} R_{L}(kr) Y_{L}^{M \pm 1}(\theta, \varphi) e^{-ickt}$$
Electric multipoles,  $A_{e}(L, M)$ 

$$A_{z} = -\left\{\left(\frac{L(L + M + 1)(L - M + 1)}{(L + 1)(2L + 1)(2L + 3)}\right)^{\frac{1}{2}} R_{L+1}(kr) Y_{L+1}^{M}(\theta, \varphi) - \left(\frac{(L + 1)(L - M)(L + M)}{L(2L - 1)(2L + 1)}\right)^{\frac{1}{2}} R_{L-1}(kr) Y_{L-1}^{M}(\theta, \varphi)\right\} e^{-ickt}$$

$$A_{z} \pm iA_{y} = \mp \left\{\left(\frac{L(L \pm M + 1)(L \pm M + 2)}{(L + 1)(2L + 1)(2L + 3)}\right)^{\frac{1}{2}} R_{L+1}(kr) Y_{L+1}^{M \pm 1}(\theta, \varphi) - \left(\frac{(L + 1)(L \mp M - 1)(L \mp M)}{L(2L + 1)(2L - 1)}\right)^{\frac{1}{2}} R_{L-1}(kr) Y_{L-1}^{M \pm 1}(\theta, \varphi)\right\} e^{-ickt}$$
with  $R_{L}(kr) = \frac{i}{(kr)^{\frac{1}{2}}} H_{L+\frac{1}{2}}^{(1)}(kr) \rightarrow (-i)^{L} \left(\frac{2}{\pi}\right)^{\frac{1}{2}} \frac{e^{ikr}}{kr}$  for  $kr \gg 1$ .

as was tested by observing no change in the angular correlation with strong applied magnetic field. One is therefore led to examine the modifications in the theory which result when assumption (2) is not valid.

The assumption of pure multipole transitions, while convenient, is otherwise quite arbitrary, for although the angular momentum and parity selection rules do limit the possible multipole radiations between two quantum states, the simultaneous occurrence of two or more multipole radiations is often consistent with these selection rules. As an example of such a mixed transition, magnetic dipole and electric quadrupole radiation are both possible between states differing by  $0, \pm 1$  units of angular momentum and having the same parity. Moreover, in the case of nuclear  $\gamma$ -rays, unlike atomic optical spectra, electric dipole radiation is often wholly absent on account of the symmetry of the nuclear charge distribution, and higher multipole radiations such as electric quadrupole and magnetic dipole can occur with comparable intensities.<sup>8-12</sup> In this paper we shall treat the theory of the directional correlation of successive nuclear  $\gamma$ -rays when one of the transitions is mixed. In particular, the tables of reference 1 will be extended to include all possible  $W(\theta)$  for the case where one of the transitions is a mixture of electric quadrupole and magnetic dipole radiation, and the other is either dipole or quadrupole. It will then be shown that the experimental data<sup>5</sup> on Sr<sup>88</sup> which had previously seemed anomalous, can be consistently interpreted with the theory developed here.

# **II. ON MIXED TRANSITIONS**

On first consideration, it might be thought that the correlation function for successive nuclear  $\gamma$ -emissions in which the first transition was a mixture of magnetic dipole and electric quadrupole, say, with respective intensities  $|\alpha|^2$  and  $|\beta|^2$ , and the second  $\gamma$ -ray any pure  $2^{L}$ -pole, should be simply the weighted sum  $|\alpha|^2 W(\theta)_{D-L} + |\beta|^2 W(\theta)_{Q-L}$  where  $W(\theta)_{D-L}$  is the correlation function for a dipole- $2^{L}$ -pole cascade emission and  $W(\theta)_{Q-L}$  that for successive quadrupole and  $2^{L}$ -pole  $\gamma$ -rays. However, in general this is not the case: Interference contributions arise from the mixing of the electric quadrupole and magnetic dipole fields. Such interference effects have in fact been observed<sup>13</sup> for a single transition in the Zeeman effect for forbidden lines in the atomic spectra of PbI, and the necessary theory given by Gerjouy.<sup>14</sup> In this section we give a resume of the theory of the interference due to mixtures of multipoles in a single transition which will at the same time serve to introduce the necessary notions and notations for treating angular correlations with mixtures in Section III.

# (A) Angular Distributions for Multipole Fields

Consider first the classical electromagnetic problem of finding the angular distribution, i.e., magnitude of the Poynting vector as a function of angle, for the various multipole radiation fields. In general, for any electromagnetic field with vector potential  $Ae^{-ickt}$  one obtains for the magnitude of the Poynting vector at large distances from all sources of the field in terms of the asymptotic form of A:

$$\mathbf{n} \cdot \mathbf{S} = \frac{c \kappa^2}{8\pi} [\mathbf{A} \cdot \mathbf{A}^* - (\mathbf{n} \cdot \mathbf{A})(\mathbf{n} \cdot \mathbf{A}^*)]$$
(1)

where **n** is a unit vector in the direction **S**. For the electric and magnetic multipoles, the well-known<sup>15</sup> spherical

<sup>&</sup>lt;sup>8</sup> H. A. Bethe, Rev. Mod. Phys. 9, 222 (1937).

<sup>&</sup>lt;sup>9</sup> Hulme, Mott, Oppenheimer, and Taylor, Proc. Roy. Soc. A155, 315 (1936).

<sup>&</sup>lt;sup>10</sup> A. C. Helmholtz, Phys. Rev. 60, 415 (1941).

<sup>&</sup>lt;sup>11</sup> M. H. Hebb and E. Nelson, Phys. Rev. **58**, 486 (1940). <sup>12</sup> E. Segrè and A. C. Helmholtz, Rev. Mod. Phys. **21**, 271 (1949).

<sup>&</sup>lt;sup>13</sup> F. A. Jenkins and S. Mrozowski, Phys. Rev. 60, 225 (1941). <sup>14</sup> E. Gerjouy, Phys. Rev. 60, 233 (1941). See also Shortley,

 <sup>&</sup>lt;sup>15</sup> W. Heitler, Proc. Camb. Phil. Soc. 32, 112 (1936); W. W. Hansen, Phys. Rev. 47, 139 (1935); H. A. Kramers, Physica 10, 261 (1943); G. Goertzel, see reference 2; O. Laporte, Am. J. Phys. 16, 206 (1948).

eigenwave solutions of Maxwell's equations,  $A_e(L, M)$ and  $A_m(L, M)$  respectively, are listed in Table I. Inserting these in (1), one obtains for both electric and magnetic (LM) pole:

$$\mathbf{n} \cdot \mathbf{S}_{L}{}^{M} = \frac{c}{2\pi r^{2}} \frac{1}{L(L+1)} [2M^{2}|Y_{L}{}^{M}|^{2} + (L-M)(L+M+1)|Y_{L}{}^{M+1}|^{2} + (L+M)(L-M+1)|Y_{L}{}^{M-1}|^{2}].$$
(2)

It is convenient for later references to define the angular distribution associated with an (LM) pole by:

$$F_L{}^M(\theta) = \frac{8\pi^3 r^2}{c} \mathbf{n} \cdot \mathbf{S}_L{}^M.$$
(3)

It is then evident that  $F_L^M(\theta)$  is a polynomial of degree L in  $\cos^2\theta$  satisfying:

$$F_L{}^M(\theta) = F_L{}^{-M}(\theta), \qquad (3a)$$

$$\int F_L^M(\theta) d\Omega = 8\pi, \text{ independent of } M, \qquad (3b)$$

$$\sum_{M=-L}^{L} F_{L}^{M}(\theta), \qquad \text{independent of } \theta, \qquad (3c)$$

and

$$F_L^M(0) = 0 \qquad \text{unless } M = \pm 1. \tag{3d}$$

In particular (3) yields for electric or magnetic dipole radiation:

$$F_1^{0}(\theta) = 3(1 - \cos^2\theta),$$
  

$$F_1^{\pm 1}(\theta) = 3/2(1 + \cos^2\theta),$$
(4)

and for electric or magnetic quadrupole:

$$F_{2}^{0}(\theta) = (5/2)(6\cos^{2}\theta - 6\cos^{4}\theta)$$

$$F_{2}^{\pm 1}(\theta) = (5/2)(1 - 3\cos^{2}\theta + 4\cos^{4}\theta)$$

$$F_{2}^{\pm 2}(\theta) = (5/2)(1 - \cos^{4}\theta).$$
(5)

Consider now the radiation field from a charge distribution which gives rise to two multipole fields, say A(L, M) and A(L', M'), with respective complex amplitudes  $\alpha$  and  $\beta$ . Inserting  $\mathbf{A} = \alpha \mathbf{A}(L, M) + \beta \mathbf{A}(L', M')$ in (1), the resultant angular distribution is found to be:

$$\mathbf{n} \cdot \mathbf{S} = |\alpha|^2 \mathbf{n} \cdot \mathbf{S}_L{}^M + |\beta|^2 \mathbf{n} \cdot \mathbf{S}_{L'}{}^{M'} + [\alpha \beta^* \mathbf{n} \cdot \mathbf{S}_{LL'}{}^{MM'} + c.c.] \quad (6)$$

where  $\mathbf{n} \cdot \mathbf{S}_{LL'}{}^{MM'}$  is the interference term due to the mixing of the two multipoles:

$$\mathbf{n} \cdot \mathbf{S}_{LL'}{}^{MM'} = (ck^2/8\pi)\mathbf{A}(L, M) \cdot \mathbf{A}^*(L', M').$$
(7)

Without recourse to the explicit functional form of the A(L, M) in Table I, it is possible to state that

$$\int \mathbf{n} \cdot \mathbf{S}_{LL'}{}^{MM'} d\Omega = 0, \qquad (8)$$

if  $L \neq L'$  or  $M \neq M'$  or also if L = L' but one multipole is electric, the other magnetic. The physical interpretation of this is that the interference in the radiation pattern is just a redistribution of the energy radiated; the total energy radiated is still the same, namely, the sum of that from each multipole individually. The proof of (8) follows from the fact that the A(L, M) transform irreducibly under the three-dimensional rotation group<sup>16</sup> and the group theoretic theorem<sup>17</sup> that two functions transforming according to different irreducible representations of a group are orthogonal.

For the case of most interest in nuclear transitions, a mixture of an (L, M) electric  $2^{L}$  pole and (L-1, M)magnetic  $2^{L-1}$  pole (the *M*'s must be the same because of the magnetic quantum number selection rule), (7) and (3) yield for the interference contribution to the angular distribution:

$$F_{L,L-1}^{M}(\theta) = -4\pi \left( \frac{(2L+1)}{(2L-1)} \frac{(L+M)(L-M)}{L^{2}(L^{2}-1)} \right)^{\frac{1}{2}} \\ \times \left[ 2M | Y_{L-1}^{M} |^{2} + (L-M-1) | Y_{L-1}^{M+1} |^{2} \\ - (L+M-1) | Y_{L-1}^{M-1} |^{2} \right].$$
(9)

This interference term is a polynomial of degree (L-1) in  $\cos^2\theta$  having the properties:

$$\int F_{L, L-1}{}^{M}(\theta)d\Omega = 0, \qquad (9a)$$

$$F_{L, L-1}{}^{M}(\theta) = -F_{L, L-1}{}^{-M}(\theta),$$
 (9b)

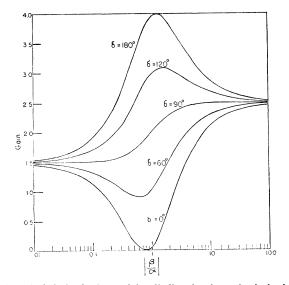


FIG. 1. Gain in the forward  $(\theta = 0)$  direction for a classical mixture of magnetic dipole and electric quadrupole radiation as a function of the mixture amplitude ratio  $|\beta/\alpha|$  and phase difference, δ.

<sup>16</sup> See H. A. Kramers, reference 15, and G. Goertzel, reference 2. Also H. C. Brinkman, Zur Quantenmechanik der Multipolstrah-lung, 1932. <sup>17</sup> See E. Wigner, Gruppentheorie (1931), p. 124.

whence

$$F_{L,L-1}^{0}(\theta) \equiv 0, \qquad (9c)$$

and

$$\sum_{M=-(L-1)}^{L-1} F_{L, L-1}{}^{M}(\theta) = 0.$$
 (9d)

In particular for a mixture of electric quadrupole and magnetic dipole, the only non-zero interference angular distributions will be:

$$F_{2,1}^{\pm 1}(\theta) = \pm (15^{\frac{1}{2}}/2)(3\cos^2\theta - 1) \tag{10}$$

and the total angular distributions (6) for such a mixture, with  $M = \pm 1$  take the form:

$$F^{\pm_{1}}(\theta) = 3/2 |\alpha|^{2} (1 + \cos^{2}\theta) + 5/2 |\beta|^{2} (1 - 3 \cos^{2}\theta + 4 \cos^{4}\theta) \pm R(\alpha\beta^{*}) (15)^{\frac{1}{2}} (3 \cos^{2}\theta - 1) \quad (11)$$

where  $\alpha$  and  $\beta$  are the amplitudes of the magnetic dipole and electric quadrupole fields respectively.

To show the effect of the interference term in this purely classical angular distribution, we have plotted in Fig. 1 the gain in the  $\theta = 0$  direction as a function of the magnitude  $|\beta/\alpha|$  and phase  $\delta = \arg \beta/\alpha$  of the mixture amplitude ratio for the angular distribution (11) with M = -1. (We define the gain<sup>18</sup> in a given direction as the ratio of the intensity in that direction to the average intensity over all directions). It is evident from Fig. 1 that interference can radically change the directional pattern for the radiating system from what one would have for pure dipole, pure quadrupole, or for an arbitrarily weighted mixture with no interference:  $\delta = 90^{\circ}$ . Indeed for pure dipole or quadrupole fields, the gain would be 3/2 or 5/2 respectively, but when these are superposed with different phases and amplitudes, the gain can vary continuously between 0 and 4.

# (B) Ouantum Mechanical Intensities

The quantum mechanical intensities for radiative transitions are most easily obtained by the correspondence theory method<sup>19</sup> of replacing the classical field amplitudes by appropriate quantum mechanical matrix elements. Thus the probability for emission of an (LM) pole  $\gamma$ -quantum with direction  $\mathbf{k}(\theta, \varphi)$ in a transition between magnetic sublevels m and m'of two degenerate nuclear states denoted by their quantum numbers  $\sigma Jm$  and  $\sigma' J'm'$  (where  $\sigma$ ,  $\sigma'$  denote all other quantum numbers than those for the total angular momentum J and its z-component m) can be gotten from the classical formula (1) provided one takes as the vector potential:

$$(\sigma Jm | LM | \sigma' J'm') \mathbf{A}(L, M)$$
(12)

where  $(\sigma Jm | LM | \sigma' J'm')$  is the matrix element for the transition. This matrix element can always be written in the form:<sup>20</sup>

$$(\sigma Jm | LM | \sigma'J'm') = (\sigma J | L | \sigma'J')(Jm | LM | J'm').$$
(13)

The matrix element  $(Jm | LM | J'm') \equiv (JLJ'm' | JLmM)$ is the (real) transformation coefficient<sup>21</sup> for the vector addition of angular momenta:  $\mathbf{J} + \mathbf{L} = \mathbf{J}', \ m + M = m',$ and depends only on the quantum numbers J, J', L, m, m'M. The matrix element  $(\sigma J | L | \sigma' J')$  is independent of m and m' and hence the same for each component of a line. In general it will depend on the explicit form of the unknown nuclear wave functions.

From (12) and (1) one gets that the intensity of the radiation in the direction  $\theta$  for an (LM) pole  $\gamma$ -emission between states  $\sigma Jm$  and  $\sigma' J'm'$  is proportional to:

$$|(\sigma J | L | \sigma' J')|^2 (Jm | LM | J'm')^2 F_L^M(\varphi), \quad (14)$$

so that the relative intensities of the components of the line which differ in initial and final sublevels m and m'are determined by:

$$(Jm|LM|J'm')^{2}F_{L}^{M}(\vartheta).$$
(15)

On account of the degeneracy of the initial and final states, it is necessary to sum (14) over all initial and final magnetic sublevels, m and m', consistent with the selection rule m' = m + M, to get the total intensity in the direction  $\theta$ . Using the group theoretic relation:<sup>22</sup>

$$\sum_{m} (Jm | LM | J'm') (Jm | L'M | J'm') = \frac{2J'+1}{2L+1} \delta_{LL'}, \quad (16)$$

and (3c), one verifies that the intensity in any direction due to the sum of all components of the line is independent of  $\theta$ . Another isotropy requirement, following from (14), (16), and (3a) is that the probability for a transition terminating in any sublevel m' of the final state is the same for each m' provided one averages over-all directions of emission of the  $\gamma$ -ray. If, however, one does not average over  $\theta$  but rather specifies the direction of emission, then even though the initial sublevels m are assumed equally populated, the final sublevels m' will not in general have equal probabilities to be occupied.

Consider now a transition between the same two degenerate states with emission of a mixture of  $2^{L}(L, M)$ 

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<sup>&</sup>lt;sup>18</sup> See J. C. Slater, *Microwave Transmission* (McGraw-Hill Book Company, Inc., New York, 1942), p. 215. We depart from conventional usage in evaluating the gain for  $\theta=0$  even though this is not in general the direction of maximum intensity when mixtures are involved. However, this is more convenient for <sup>19</sup> E. U. Condon and G. H. Shortley, *Theory of Atomic Spectra* 

<sup>(</sup>Cambridge University Press, London, 1935), Ch. IV.

<sup>&</sup>lt;sup>20</sup> See C. Eckart, Rev. Mod. Phys. 2, 305 (1930). Also E. Wigner, reference 17, p. 264. <sup>21</sup> See Condon and Shortley, reference 19, pp. 73–78, or Wigner,

<sup>&</sup>lt;sup>22</sup> This theorem has been used by H. B. Casimir, Archives du Musée Teyler, Series III, VIII, 274 (1936), to show that the total probability for internal conversion in a field which is a superposition of two multipole fields is the sum of the probabilities due to each separately. For a formal proof of (16) see G. Breit and B. T. Darling, Phys. Rev. 71, 405 (1947).

$\Delta J = 1$	$\Delta j = 1$	$Q = 13J  \alpha ^{2} + (5/21)(55J - 6)  \beta ^{2} + (2/3)(15J(J + 2))^{\frac{1}{2}} Re(\alpha\beta^{*})$ $R = J  \alpha ^{2} + (5/7)(J + 6)  \beta ^{2} - 2(15J(J + 2))^{\frac{1}{2}} Re(\alpha\beta^{*})$
	$\Delta j = 0$	$\begin{split} &Q = J(14J+13) \left  \alpha \right ^2 + (5/21)(58J^2+67J-6) \left  \beta \right ^2 - (2/3)(15J(J+2))^{\frac{1}{2}}(2J-1)Re(\alpha\beta^*) \\ &R = -J(2J-1) \left  \alpha \right ^2 - (5/7)(J+6)(2J-1) \left  \beta \right ^2 + 2(15J(J+2))^{\frac{1}{2}}(2J-1)Re(\alpha\beta^*) \end{split}$
	$\Delta j = -1$	$\begin{split} &Q = (26J^2 + 67J + 40)    \alpha  ^2 + (5/21)(110J^2 + 269J + 174)    \beta  ^2 + (2/3)(15J(J+2))^{\frac{1}{2}}(2J-1)Re(\alpha\beta^*) \\ &R = J(2J-1)    \alpha  ^2 + (5/7)(J+6)(2J-1)    \beta  ^2 - 2(15J(J+2))^{\frac{1}{2}}(2J-1)Re(\alpha\beta^*) \end{split}$
$\Delta J = 0$	$\Delta j = 1$	$\begin{aligned} Q &= (2J-1)(14J+1)  \alpha ^2 + (5/7)(36J^2 - 20J+5)  \beta ^2 - 2(5(2J-1)(2J+3))^{\frac{1}{2}} Re(\alpha\beta^*) \\ R &= -(2J-1)(2J+3)  \alpha ^2 + (5/7)(2J+5)(2J-3)  \beta ^2 + 6(5(2J-1)(2J+3))^{\frac{1}{2}} Re(\alpha\beta^*) \end{aligned}$
	$\Delta j = 0$	$\begin{split} &Q = (12J^2 + 12J + 1)    \alpha  ^2 + (5/7)(20J^2 + 20J - 5)    \beta  ^2 + 2(5(2J - 1)(2J + 3))^{\frac{1}{2}} Re(\alpha \beta^*) \\ &R = (2J - 1)(2J + 3)    \alpha  ^2 - (5/7)(2J - 3)(2J + 5)    \beta  ^2 - 6(5(2J - 1)(2J + 3))^{\frac{1}{2}} Re(\alpha \beta^*) \end{split}$
	$\Delta j = -1$	$\begin{aligned} Q &= (2J+3)(14J+13)  \alpha ^2 + (5/7)(36J^2+92J+61)  \beta ^2 - 2(5(2J-1)(2J+3))^{\frac{1}{2}} Re(\alpha\beta^*) \\ R &= -(2J-1)(2J+3)  \alpha ^2 + (5/7)(2J-3)(2J+5)  \beta ^2 + 6(5(2J-1)(2J+3))^{\frac{1}{2}} Re(\alpha\beta^*) \end{aligned}$
	$\Delta j = 1$	$\begin{aligned} Q &= (26J^2 - 15J - 1)  \alpha ^2 + (5/21)(110J^2 - 49J + 15)  \beta ^2 - (2/3)(15(J^2 - 1))^{\frac{1}{2}}(2J + 3)Re(\alpha\beta^*) \\ R &= (J+1)(2J+3)  \alpha ^2 + (5/7)(2J+3)(J-5)  \beta ^2 + 2(15(J^2 - 1))^{\frac{1}{2}}(2J+3)Re(\alpha\beta^*) \end{aligned}$
$\Delta J = -1$	$\Delta j = 0$	$\begin{split} &Q = (J+1)(14J+1)    \alpha  ^{2} + (5/21)(58J^{2} + 49J - 15)    \beta  ^{2} + (2/3)(15(J^{2} - 1))^{\frac{1}{2}}(2J+3)Re(\alpha\beta^{*}) \\ &R = -(J+1)(2J+3)    \alpha  ^{2} - (5/7)(2J+3)(J-5)    \beta  ^{2} - 2(15(J^{2} - 1))^{\frac{1}{2}}(2J+3)Re(\alpha\beta^{*}) \end{split}$
	$\Delta j = -1$	$Q = 13(J+1)  \alpha ^2 + (5/21)(55J+61)  \beta ^2 - (2/3)(15(J^2-1))^{\frac{1}{2}} Re(\alpha\beta^*)$ $R = (J+1)  \alpha ^2 + (5/7)(J-5)  \beta ^2 + 2(15(J^2-1))^{\frac{1}{2}} Re(\alpha\beta^*)$

# TABLE II. Q and R for dipole-mixture correlation.

electric multipole and  $2^{L-1}(L-1, M)$  magnetic multipole radiation. The appropriate vector potential is now:

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$$\mathbf{A} = (\sigma J | L | \sigma' J') (Jm | LM | J'm') \mathbf{A}_{e}(L, M) + (\sigma J | L-1 | \sigma' J') (Jm | L-1, M | J'm') \mathbf{A}_{m}(L-1, M).$$

Denoting the unknown nuclear matrix elements  $(\sigma J | L | \sigma' J')$  and  $(\sigma J | L - 1 | \sigma' J')$  by  $\alpha$  and  $\beta$  respectively,<sup>23</sup> the quantum mechanical analogue of (6) for the intensity of the component  $m \rightarrow m' = m + M$  in the direction  $\theta$  is of the same form as the classical expression (6):

$$\mathbf{n} \cdot \mathbf{S}_{m \to m'} = |\alpha|^{2} (Jm | LM | J'm')^{2} F_{L}{}^{M}(\theta) + |\beta|^{2} (Jm | L-1, M | J'm')^{2} F_{L-1}{}^{M}(\theta) + (\alpha \beta^{*} + \alpha^{*} \beta) (Jm | LM | J'm') \times (Jm | L-1, M | J'm') F_{L, L-1}{}^{M}(\theta),$$
(17)

where  $F_L{}^M(\theta)$  and  $F_{L, L-1}{}^M(\theta)$  are given by (3) and (9), respectively. Hence, here too the interference term will vanish when averaged over  $\theta$ . But in addition, even for specified  $\theta$ , the mixture contribution will vanish when summed over all initial and final sublevels, m and m', in virtue of (16)! How then is the interference observable?

If one considers only those transitions  $m \rightarrow m'$  characterized by fixed M = m' - m, then the interference term need not vanish. When a strong magnetic field is present so that those components with different Mdiffer in frequency, as in the atomic Zeeman effect, the interference contributions to the angular intensities are detectable as reported in references 13 and 14.

Interference effects are also detectable in angular correlations. In this case the individual components of a line are distinguished not by their frequencies, which are all the same, but by different statistical weights.

<sup>&</sup>lt;sup>23</sup> Strictly speaking,  $\alpha$  and  $\beta$  are not the usual multipole matrix elements but are implicitly defined by Eq. (17). Summing over-all initial and final degenerate sublevels *m* and *m'*, and over-all directions of emission, one gets for the total intensity of the line (using (3b), (9a) and (16)):  $8\pi(2J'+1)[|\alpha|^2+|\beta|^2]$ . Hence  $|\alpha|^2$  and  $|\beta|^2$  determine the absolute intensities of the two multipole radiations and must, except for a common proportionality factor, be the same as the Einstein coefficients for spontaneous emission for the respective multipoles. One may, therefore, interpret  $\alpha$  and  $\beta$  as phenomenologically renormalized matrix elements whose absolute value squared are the respective Einstein probabilities. The phases of  $\alpha$  and  $\beta$  are defined to be the same as those of the associated nuclear matrix elements. Experimentally one can take  $|\alpha|^2$  and  $|\beta|^2$  as the number of quanta emitted by the respective multipoles; actually only their ratio is needed here.

TABLE III. Q, R, and S for quadrupole-mixture correlation.

Print to send of the second second		
$\Delta J = 1$	$\Delta j = 2$	$Q = 87J  \alpha ^2 + 5(17J+2)  \beta ^2 - 6(15J(J+2))^{\frac{1}{2}}R(\alpha\beta^*)$ $R = -9J  \alpha ^2 + 5(J-2)  \beta ^2 + 18(15J(J+2))^{\frac{1}{2}}R(\alpha\beta^*)$ $S = -(20/3)(2J+5)  \beta ^2$
	$\Delta j = 1$	$\begin{split} &Q = (3/2)J(55J+61)  \alpha ^2 + (5/2)(35J^2 + 35J+6)  \beta ^2 + 3(15J(J+2))^{\frac{1}{2}}(J-5)R(\alpha\beta^*) \\ &R = (9/2)J(J-5)  \alpha ^2 - (5/2)(17J^2 + 17J - 30)  \beta ^2 - 9(15J(J+2))^{\frac{1}{2}}(J-5)R(\alpha\beta^*) \\ &S = (40/3)(2J-3)(2J+5)  \beta ^2 \end{split}$
	$\Delta j = 0$	$\begin{aligned} Q &= 3J(36J^2 + 92J + 61)  \alpha ^2 + 5(20J^3 + 52J^2 + 41J + 6)  \beta ^2 + 2(15J(J+2))^{\frac{1}{2}}(2J-3)(2J+5)R(\alpha\beta^*) \\ R &= 3J(2J-3)(2J+5)  \alpha ^2 + 5(2J-3)(2J+5)(5J-2)  \beta ^2 - 6(15J(J+2))^{\frac{1}{2}}(2J-3)(2J+5)R(\alpha\beta^*) \\ S &= -(80/3)(2J-3)(2J+5)(J-1)  \beta ^2 \end{aligned}$
		$Q = (3/2)(J+2)(110J^{2}+269J+174)  \alpha ^{2} + (5/2)(70J^{4}+289J^{3}+374J^{2}+188J+24)  \beta ^{2} + 3(15J(J+2))^{\frac{1}{2}}(J+2)(2J-1)(J+6)R(\alpha\beta^{*})$
	$\Delta j = -1$	$R = (9/2)J(J+2)(J+6)(2J-1) \alpha ^{2} - (5/2)(2J-1)(17J^{3} - 18J^{2} - 164J - 24) \beta ^{2} - 9(15J(J+2))^{3}(J+2)(2J-1)(J+6)R(\alpha\beta^{*})$
		$S = (40/3)(J-1)(2J-3)(2J+5)(2J-1) \beta ^2$
		$Q = (3/2)(J+2)(58J^2+139J+84) \alpha ^2 + (5/2)(34J^3+159J^2+224J+96) \beta ^2 - 3(15J(J+2))^{\frac{1}{2}}(J+2)(2J-1)R\alpha(\beta^*)$
	$\Delta j = -2$	$R = -(9/2)J(J+2)(2J-1) \alpha ^{2} + (5/2)(2J-1)(J^{2}-16J-12) \beta ^{2} + 9(15J(+2))^{\frac{1}{2}}(J+2)(2J-1)R(\alpha\beta^{*})$ $S = -(10/3)(J-1)(2J-1)(2J-3) \beta ^{2}$
$\Delta J = 0$	$\Delta j = 2$	$\begin{split} &Q = (3/2)(2J-1)(26J-3)  \alpha ^2 + (15/2)(2J-1)(6J+1)  \beta ^2 + 9(5(2J-1)(2J+3))^{\frac{1}{2}} R(\alpha\beta^*) \\ &R = (9/2)(2J-1)(2J+3)  \alpha ^2 - (15/2)(2J+1)(2J+5)  \beta ^2 - 27(5(2J-1)(2J+3))^{\frac{1}{2}} R(\alpha\beta^*) \\ &S = 10(J+2)(2J+5)  \beta ^2 \end{split}$
		$Q = (3/2)(2J-1)(58J^2+49J-15) \alpha ^2 + (15/2)(20J^3+8J^2-3J+3) \beta ^2 - 9(5(2J-1)(2J+3))^{\frac{1}{2}}(J-5)R(\alpha\beta^*)$
	$\Delta j = 1$	$R = -(9/2)(2J-1)(2J+3)(J-5) \alpha ^{2} + (15/2)(2J-3)(2J+5)(5J+7) \beta ^{2} + 27(5(2J-1)(2J+3))^{\frac{1}{2}}(J-5)R(\alpha\beta^{*})$
		$S = -40(J+2)(2J-3)(2J+5) \beta ^2$
		$Q = 15(2J-1)(2J+3)(4J^2+4J-1) \alpha ^2 + 15(2J-1)(2J+3)(4J^2+4J-1) \beta ^2 - 6(5(2J-1)(2J+3))^{\frac{1}{2}}(2J-3)(2J+5)R(\alpha\beta^*)$
	$\Delta j = 0$	$ \begin{array}{c} R = -3(2J-1)(2J+3)(2J-3)(2J+5)     \alpha   ^2 - 15(2J-3)(2J+5)(4J^2+4J-7)     \beta   ^2 \\ \qquad $
		$S = 80(2J-3)(2J+5)(J-1)(J+2) \beta ^2$
	$\Delta j = -1$	$Q = (3/2)(2J+3)(58J^2+67J-6) \alpha ^2 + (15/2)(20J^3+52J^2+41J+6) \beta ^2 -9(5(2J-1)(2J+3))^{\frac{1}{2}}(J+6)R(\alpha\beta^*)$
		$R = -(9/2)(2J+3)(2J-1)(J+6)  \alpha ^2 + (15/2)(2J-3)(2J+5)(5J-2)  \beta ^2 + 27(5(2J-1)(2J+3))^{\frac{1}{2}}(J+6)R(\alpha\beta^*)$
		$S = -40(2J-3)(2J+5)(J-1) \beta ^2$
	$\Delta j = -2$	$\begin{aligned} Q &= (3/2)(2J+3)(26J+29)  \alpha ^2 + (15/2)(2J+3)(6J+5)  \beta ^2 + 9(5(2J-1)(2J+3))^{\frac{1}{2}}R(\alpha\beta^*) \\ R &= (9/2)(2J+3)(2J-1)  \alpha ^2 - (15/2)(2J-3)(2J+1)  \beta ^2 - 27(5(2J-1)(2J+3))^{\frac{1}{2}}R(\alpha\beta^*) \\ S &= 10(J-1)(2J-3)  \beta ^2 \end{aligned}$

Namely, if, say, the second transition is mixed, then by specifying the direction of emission of the first quantum, the end states for this first transition which are the initial sublevels for the second transition are unequally populated. Hence, Eq. (16) is no longer applicable, each m for the initial sublevels of the mixed transition being weighted differently, and the interference cross terms in (17) need not vanish when aver-

	$\Delta j = 2$	$Q = (3/2)(J-1)(58J^2-23J+3) \alpha ^2 + (5/2)(34J^3-57J^2+8J+3) \beta ^2 + 3(15(J^2-1))^{\frac{1}{2}}(J-1)(2J+3)R(\alpha\beta^*)$ $R = -(9/2)(J-1)(2J+3)(J+1) \alpha ^2 + (5/2)(2J+3)(J^2+18J+5) \beta ^2 - 9(15(J^2-1))^{\frac{1}{2}}(J-1)(2J+3)R(\alpha\beta)^*$
		$S = -(10/3)(2J+3)(J+2)(2J+5) \beta ^2$
		$ Q = (3/2)(J+1)(J-1)(110J^2 - 49J + 15)  \alpha ^2 + (5/2)(70J^4 - 9J^3 - 73J^2 - 27J - 9)  \beta ^2 - 3(15(J^2 - 1))^{\frac{1}{2}}(2J+3)(J-5)(J-1)R(\alpha\beta^*) $
	$\Delta j = 1$	$ \begin{split} R = (9/2)(J+1)(J-1)(2J+3)(J-5)     \alpha   ^2 - (5/2)(2J+3)(17J^3 + 69J^2 - 77J - 105)     \beta   ^2 \\ + 9(15(J^2-1))^{\frac{1}{2}}(2J+3)(J-5)(J-1)R(\alpha\beta^*) \end{split} $
$\Delta J = -1$		$S = (40/3)(2J+3)(2J-3)(2J+5)(J+2) \beta ^2$
		$Q = 3(J+1)(36J^2 - 20J+5)  \alpha ^2 + 5(20J^3 + 8J^2 - 3J+3)  \beta ^2 - 2(15(J^2 - 1))^{\frac{1}{2}}(2J-3)(2J+5)R(\alpha\beta^*)$
	$\Delta j = 0$	$R = 3(J+1)(2J-3)(2J+5) \alpha ^{2} + 5(2J-3)(2J+5)(5J+7) \beta ^{2} + 6(15(J^{2}-1))^{\frac{1}{2}}(2J-3)(2J+5)R(\alpha\beta^{*})$ $S = -(80/3)(J+2)(2J-3)(2J+5) \beta ^{2}$
		$Q = (3/2)(J+1)(55J-6)  \alpha ^2 + (5/2)(35J^2 + 35J+6)  \beta ^2 - 3(15(J^2-1))^{\frac{1}{2}}(J+6)R(\alpha\beta^*)$
	$\Delta j = -1$	$R = (9/2)(J+1)(J+6) \alpha ^2 - (5/2)(17J^2 + 17J - 30) \beta ^2 + 9(15(J^2 - 1))^{\frac{1}{2}}(J+6)R(\alpha\beta^*)$
		$S = (40/3)(2J-3)(2J+5) \beta ^2$
		$Q = 87(J+1)  \alpha ^2 + 5(17J+15)  \beta ^2 + 6(15(J^2-1))^{\frac{1}{2}}R(\alpha\beta^*)$
	$\Delta j = -2$	$R = -9(J+1)  \alpha ^2 + 5(J+3)  \beta ^2 - 18(15(J^2-1))^{\frac{1}{2}} R(\alpha\beta^*)$
	C	$S = -(20/3)(2J-3) \beta ^2$

TABLE III.-Continued.

aged over all initial and final states (see Eqs. (18a) or (18b)).

# **III. ANGULAR CORRELATION WITH MIXTURES**

A rigorous quantum mechanical derivation of the angular correlation function  $W(\theta)$  requires a secondorder time dependent perturbation calculation beginning with an initial system of excited nucleus plus quantised radiation field. Hamilton<sup>1</sup> has given such a derivation in a form sufficiently general to apply to mixed transitions as well. If the successive nuclear states involved have spins J'', J and J' with z-components m'', m and m', and if the first emission is a pure  $2^{L'}$  multipole and the second a mixture of  $2^{L}$ electric and  $2^{L-1}$  magnetic multipoles, then the angular correlation function  $W(\theta)$  for the relative probability that the second  $\gamma$ -ray be emitted at an angle  $\theta$  with respect to the first  $\gamma$ -ray may be written in either of the equivalent forms:

$$W(\theta) = \sum_{m', m, m''} \left[ (J''m'' | L'M' | Jm)^2 F_{L'}^{M'}(0) \right] \\ \times \mathbf{n} \cdot \mathbf{S}_{mm'}(\theta) \quad (18a)$$
$$W(\theta) = \sum_{m', m, m''} \left[ (J''m'' | L'M' | Jm)^2 F_{L'}^{M'}(\theta) \right] \\ \times \left[ \mathbf{n} \cdot \mathbf{S}_{mm'}(0) \right] \quad (18b)$$

according as the direction of emission of the first or second  $\gamma$ -ray is taken as the z-axis,  $\theta = 0$ . Here  $\mathbf{n} \cdot \mathbf{S}_{mm'}(\theta)$ is the relative probability for the  $\gamma$ -ray in the second mixed transition to be emitted in the direction  $\theta$  in the component  $m \rightarrow m'$  as given by (17). On expanding (18a or b),  $W(\theta)$  takes the form:

$$W(\theta) = |\alpha|^2 W_{L', L}(\theta) + |\beta|^2 W_{L', L-1}(\theta) + 2R(\alpha \beta^*) W_I(\theta) \quad (19)$$

where  $W_{L'L}(\theta)$  is the angular correlation due to successive  $2^{L'}$  and  $2^{L}$  multipoles (and similarly for  $W_{L'L-1}$ ) while  $W_I(\theta)$ , the coefficient of  $\alpha\beta^* + \alpha^*\beta = 2R(\alpha\beta^*)$  is the interference contribution due to the mixed multipoles in the second transition.

The explicit evaluation of  $W(\theta)$  thus requires knowing  $|\alpha|^2$ ,  $|\beta|^2$ ,  $R(\alpha\beta^*)$ ,  $F_{L^M}(\theta)$ ,  $F_{L'L-1}{}^M(\theta)$  and the matrix elements (Jm|LM|J'm'). When both transitions are pure multipoles, either  $\alpha$  or  $\beta$  is zero, say  $\beta$ . Then it is not necessary to know  $\alpha$  since  $W(\theta)$  is a relative probability and the common factor  $|\alpha|^2$  can be dropped. For the mixture case both  $\alpha$  and  $\beta$  are nonzero. On dividing (19) by  $|\alpha|^2$ , only the ratio  $\beta/\alpha$ enters in  $W(\theta)$ . Since this ratio is a complex number in general, one has to specify both its modulus and argument. Physically this means that both the *relative intensities* of the mixed multipoles and the *relative phases*<sup>24</sup> of their matrix elements must be given. The angular distributions  $F_L M(\theta)$  and  $F_{L, L-1} M(\theta)$  are given in a form easy to evaluate for any L by Eqs. (3)

<sup>&</sup>lt;sup>24</sup> For the atomic Zeeman effect considered in reference 14,  $\alpha$  and  $\beta$  are simply matrix elements involving radial wave functions and hence both real. For the nuclear case  $\alpha$  and  $\beta$  could be evaluated theoretically if one chose some particular model for the nucleus, such as liquid drop.

and (9). General formulas for the normalized transformation coefficients are available<sup>25</sup> but these have been tabulated<sup>26</sup> only for L=1 and 2.<sup>27</sup> Hence the correlation functions have been worked out in detail<sup>1</sup> only for dipole and quadrupole  $\gamma$ -rays, and accordingly, we have confined our explicit evaluation of mixture correlations to the cases when the pure multipole transition is either dipole or quadrupole and the mixture multipoles are electric quadrupole and magnetic dipole.

It should be noted, that although for L or L'=1 or 2, the  $W_{L'L}(\theta)$  must be equivalent to those obtained by Hamilton, it is not correct to insert Hamilton's results directly into (18), for his correlation functions are not properly normalized when mixtures are taken into account. This is shown in the Appendix. Except for this normalization, the actual procedure used in obtaining the  $W_{L'L}(\theta)$  completely parallels the method outlined by Hamilton. The interference contribution (19) can be reduced, using (3a, d) and (10), to the form:

$$W_{I}(\theta) = (15^{\frac{1}{2}}/2)(3 \cos^{2}\theta - 1) \cdot$$

$$\sum_{m=-J}^{J} \{ [(J''m-1|L', 1|Jm)^{2} + (J''m+1|L', -1|Jm)^{2}] \\ \times [(Jm|1, 1|J'm+1)(Jm|2, 1|J'm+1) \\ - (Jm|1, -1|J'm-1)(Jm|2, -1|J'm-1)] \}.$$

The evaluation of the indicated sums of products of normalized transformation coefficients presents the only new computational task for the preparation of Tables II and III which are for dipole-(mixture) and quadrupole-(mixture) correlations, respectively. The notation of Hamilton is used throughout. Successive nuclear states have spins  $J-\Delta j$ , J, and  $J+\Delta J$ . The correlation function has the form:

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

Factors common to Q, R, and S are dropped in these tables. When  $\alpha = 0$  or  $\beta = 0$ , each entry in the tables reduce to the corresponding entry in Hamilton's tables for pure multipole transitions which provided a check on our calculation. The same tables may be used to obtain the correlation functions when the first transition is mixed and the second pure dipole or quadrupole. This is illustrated in the following discussion.

### IV. DISCUSSION AND APPLICATION TO EXPERIMENT

We now apply the preceding theory to the experimental  $\gamma - \gamma$  angular correlations is Sr<sup>88</sup> and Pd<sup>106</sup> which were measured by Deutsch<sup>5</sup> and which are not in satisfactory agreement with the theory as given by Hamilton.

The measured correlation function for Sr<sup>88</sup> can be adequately represented by  $W(\varphi) = 1 + R/Q \cos^2\theta$  with  $R/Q = W(\pi) - W(\pi/2) = -0.08$ . From Hamilton's tables, this value of R/Q can be obtained only if the spins of the ground, intermediate and initial states are, respectively, 2, 2, 2, or 3, 2, 2, or 2, 2, 3. However, Sr<sup>88</sup> being an even-even nucleus, its ground state would be expected to have spin 0. In fact, Peacock,<sup>28</sup> on the basis of internal conversion measurements proposes a disintegration scheme for Sr<sup>88</sup> in which the spins of ground, intermediate, and initial states are respectively 0, 1, 2. Which this choice of spins if both transitions are dipole theory gives R/Q = +0.077, while if the first transition is quadrupole, the second dipole, R/Q = +0.429. These are the only possible combinations of pure dipole and quadrupole radiations consistent with Peacock's assignment of nuclear spins and the observed  $\cos^2\theta$  dependence of  $W(\theta)$ , and neither of these R/Q values agrees with experiment. One is, therefore, led to examine the possibility that one of the transitions may be mixed.

Since quadrupole radiation is forbidden for the second  $(1\rightarrow 0)$  transition, it must be pure (electric or magnetic) dipole and we take the first  $(2\rightarrow 1)$  transition as the mixture of magnetic dipole and electric quadrupole radiation. Tables II and III are prepared for the case when the first transition is pure and the second mixed. However, on account of the Hermiticity of all matrix elements in  $W(\theta)$ , the correlation function must be the same for any direct process as its inverse. Hence the angular correlation for the proposed decay scheme in which the first  $\gamma$ -transition is mixed, the second pure with  $\Delta i = \Delta J = -1$ , J = 1 is the same as that tabulated for the case when the first transition is pure, the second mixed with  $\Delta j = \Delta J = +1$ , J = 1. From Table II, one has from the  $\Delta j = \Delta J = 1$  entry after division by  $|\alpha|^2$ :

$$R/Q = \frac{1+5|\beta/\alpha|^2 - 13.4R(\beta/\alpha)}{13+11.7|\beta/\alpha|^2 + 4.46R(\beta/\alpha)}$$

This function, R/Q is plotted in Fig. 2 for various values of the magnitude and phase  $\delta$  of the mixture amplitude ratio  $\beta/\alpha$ . The intersection of these curves with the line R/Q = -0.08 gives the locus of values of  $\beta/\alpha$  which yield agreement with experiment. It is seen that for any  $0 \le \delta \le 58^\circ$ , there are two admissible values of  $\beta/\alpha$ . For example for  $\delta = 0$ , i.e.,  $\beta/\alpha$  real, the suitable values of  $\beta/\alpha$  are 0.18 and 2.0 corresponding to a quadrupole to dipole intensity ratio of 0.032 or 4.0 respectively. The arbitrariness in the choice of the relative phases and magnitudes of  $\alpha$  and  $\beta$  which can satisfactorily explain the observed  $\gamma - \gamma$  angular correlation could be removed if the same mixed  $\gamma$ -transition also gave rise to internal conversion electrons. Then the same  $\alpha$  and  $\beta$  would enter in the internal-conversion—

<sup>&</sup>lt;sup>25</sup> See reference 17, p. 206.

<sup>&</sup>lt;sup>26</sup> See reference 19, pp. 76, 77. <sup>27</sup> A table for L=3 has been prepared but not yet published by one of us (D. L. F.).

<sup>&</sup>lt;sup>28</sup> W. C. Peacock, unpublished, quoted by Deutsch, reference 5.

 $\gamma$ -angular correlation and afford a consistency check on the assignment of the complex parameter  $\beta/\alpha$ .

It is instructive to consider in more detail the influence of the interference term on the angular correlation as shown in Fig. 2. The curve for  $\delta = 90^{\circ}$  gives the values which R/O would have if there were no interference present; in this case R and O for the correlation with mixed transition are simply the weighted average of the expressions for pure transitions. It is clear from the figure that the correlation can be radically affected both in magnitude and sign by even a small mixture ratio. Roughly, the relative change in the angular correlation due to interference is of the same order of magnitude as the ratio of the multipole amplitudes, rather than their intensities, in the mixed transition, for the ratio of the interference term to the intensity of the dominant multipole will be of order  $|\alpha\beta|/|\alpha|^2$  $\sim |\beta/\alpha|$  if  $|\alpha|^2 > |\beta|^2$ . Although Fig. 2 is drawn for the special choice of nuclear quantum numbers in the proposed decay scheme for Sr<sup>88</sup>, it is evident from its similarity with Fig. 1 that one may expect the same general behavior due to interference whatever the particular nuclear states involved. Indeed one might say that although the existence of an angular correlation is itself purely a quantum mechanical effect, the interference effects in the angular correlation have a purely classical origin in the interference contributions occurring in the classical angular distribution (6) or (11) due to a mixture of multipoles. In fact, the summands  $\mathbf{n} \cdot \mathbf{S}_{mm'}(0)$  for  $m' = m \pm 1$  occurring in the correlation function (18b) are to within a proportionality factor (if one absorbs the transformation coefficients into the definition of  $\alpha$  and  $\beta$  just the gain in the  $\theta = 0$  direction, as defined in Section IIA and plotted in Fig. 1, for the angular distribution (17) resulting from the m-m' component of the mixed transition.

Consider now the case of Pd<sup>106</sup>. We have shown elsewhere<sup>29</sup> that if one assumes the highest multipole radiation present is quadrupole and the ground state of this even-even nucleus has spin 0, then the measured<sup>5,7</sup> correlation function forces the spin of the intermediate state to be 2 and that of the initial state, 1, 2, or 3. The most general set of transitions consistent with these requirements is one in which the first transition is a dipole quadrupole mixture and the second is pure quadrupole radiation. We now show that even the use of mixtures cannot explain the experimental  $\gamma - \gamma$ -angular correlation, and therefore one of the  $\gamma$ -rays in the Pd<sup>106</sup> transitions must be at least octupole.

The observed  $W(\theta)$  has the form  $W(\theta) = 1 + R/Q \cos^2 \theta$ + $S/Q \cos^4 \theta$  with R/Q = -1.66 and S/Q = 2.16. If the initial nuclear spin is 1, then  $\Delta j = 1$ ,  $\Delta J = -2$ , J = 2. Since the order of the mixed and pure transition is the reverse of that for which the tables are calculated, one must use the  $\Delta j = 2$ ,  $\Delta J = -1$  entry in Table III with J=2 which gives:

$$R/Q = \frac{-94.5 + 787.5 |\beta/\alpha|^2 - 423R(\beta/\alpha)}{283.5 + 157.5 |\beta/\alpha|^2 + 141R(\beta/\alpha)},$$

$$S/Q = \frac{-840 |\beta/\alpha|^2}{283.5 + 157.5 |\beta/\alpha|^2 + 141R(\beta/\alpha)}.$$
(20)

Equating these to the observed values one obtains two simultaneous linear equations for the two real unknowns  $|\beta/\alpha|^2$  and  $R(\beta/\alpha)$ . These have a unique solution, but it is not physically admissible since one gets  $|\beta/\alpha|^2 = -0.425$  while it should always be non-negative. (More directly, in this particular case, one can easily see that no matter what value of  $\beta/\alpha$  is used in S/Q in Eq. (20), S/Q will always be negative, in contrast to the experimental value of  $\pm 2.16$ .) This disagreement is well outside the experimental error. Similar inconsistencies occur if one assumes the initial state to have spin 2 or 3. Thus the observed angular correlation in Pd<sup>106</sup> cannot be explained either with or without mixtures when the highest  $\gamma$ -multipole order is assumed to be quadrupole.

This negative result shows also that the presence of two additional parameters in mixture angular correlations is by no means a guarantee that a suitable mixture can be found to explain any correlation which eludes the pure multipole tables. On the contrary, a fit may not be possible at all, as with Pd<sup>106</sup>, or only for restricted values of  $\beta/\alpha$ , as for Sr<sup>88</sup>. The essential differ-

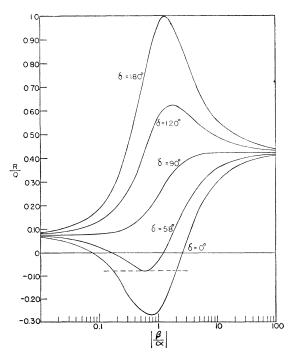


FIG. 2. R/Q as a function of  $|\beta/\alpha|$  and  $\delta$  for a mixture-dipole angular correlation with  $\Delta j = \Delta J = -1$ , J = 1. The dotted line represents the experimental value -0.08 for Sr<sup>88</sup>.

<sup>&</sup>lt;sup>29</sup> D. S. Ling, Jr. and D. L. Falkoff, Phys. Rev. 76, 431 (1949).

with

ence between these two cases is that for Sr<sup>88</sup> the observed correlation requires that the non-mixed transition be pure dipole. Hence only one number, R/Q, need be given to determine  $W(\theta)$ . Any point of intersection of the line R/Q = -0.08 with the curves of Fig. 2 yields admissible values for  $|\beta/\alpha|$  and  $\delta = \arg\beta/\alpha$ . However, for Pd<sup>106</sup>, the observed correlation having a  $\cos^4\theta$  as well as  $\cos^2\theta$  dependence requires the specification of two constants, R/Q and S/Q. For such correlations, as shown above, there is no reason to expect that the solution of the resulting simultaneous equations should be physically admissible. If, however, a consistent solution exists it will also be unique.

We conclude with a remark on mixtures in other nuclear processes. Evidence for mixtures of multipole radiation in internal conversion has often been cited.<sup>9-12</sup> As remarked by Casmir<sup>22</sup> interference effects from such mixtures would not ordinarily be detectable in a single transition. However, they would be detectable in internal conversion—internal conversion or internal conversion— $\gamma$ -angular correlations.<sup>30</sup> Mixtures of matrix elements also arise quite naturally in the theory of forbidden  $\beta$ -decay<sup>31</sup> and interference effects of the type discussed here will, therefore, also occur in the  $\beta - \gamma$ angular correlations. The theory of the  $\beta$ - $\gamma$ -correlation has been treated by one of us<sup>32</sup> and will be discussed elsewhere.

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#### APPENDIX

We indicate why it is not correct to merely substitute Hamilton's tabulated correlation functions for  $W_{LL'}(\theta)$ , L, L'=1 or 2, in Eq. (19). For a dipole—(magnetic) dipole correlation,  $W(\theta)$  has the form: (a)  $W_{11}(\theta) = Q_{11} + R_{11} \cos^2\theta$ . Since this is a relative probability, it can equally well be written as (b)  $W_{11}(\theta) = 1 + R_{11}/Q_{11} \cos^2\theta$  which is the form tabulated by Hamilton. Similarly a dipole—(electric) quadrupole correlation could be written as either:  $(a')W_{12}(\theta) = Q_{12} + R_{12} \cos^2\theta$  or  $(b')W_{12}(\theta) = 1 + R_{12}/Q_{12} \cos^2\theta$ . When the second transition is mixed, and if for simplicity we assume that  $R(\alpha\beta^*) = 0$  so that the interference term vanishes, then (19) yields for the correlation with mixtures:

$$W(\theta) = |\alpha|^2 W_{11}(\theta) + |\beta|^2 W_{12}(\theta) = \lceil |\alpha|^2 Q_{11} + |\beta|^2 Q_{12} \rceil + \lceil |\alpha|^2 R_{11} + |\beta|^2 R_{12} \rceil \cos^2 \theta$$

which is the weighted average of  $W_{11}(\theta)$  and  $W_{12}(\theta)$  when taken in the form (a) and (a'). This can now be written in the form:

 $W(\varphi) = 1 + R'/Q \cos^2\theta$  $R/Q = \frac{|\alpha|^2 R_{11} + |\beta|^2 R_{12}}{|\alpha|^2 Q_{11} + |\beta|^2 Q_{12}}$ 

which is correct but clearly not the same as one would get by substituting the forms (b) and (b') of reference 1 into (19). The argument is the same when interference is taken into account, only one get additional terms with coefficients  $R(\alpha\beta^*)$  in R and Q.

Another respect in which the calculation of correlations for mixtures differs from that for pure multipoles is the necessity to retain the normalizing factors in the transformation coefficients Only those factors common to both multipoles can be dropped.

<sup>&</sup>lt;sup>30</sup> The theory of angular correlations involving internal conversion electrons has been treated by D. S. Ling, Jr., thesis, University of Michigan, April 1948, and D. S. Ling, Jr. and G. E. Uhlenbeck, (to be published). See also V. B. Berestetski, J. Exp. Theor. Phys. U.S.S.R. 18, 1057 (1948), (*In Russian*).

<sup>&</sup>lt;sup>a1</sup> E. J. Konopinski and G. E. Uhlenbeck, Phys. Rev. **60**, 308 (1941).

<sup>&</sup>lt;sup>32</sup> See D. L. Falkoff, thesis, University of Michigan, April, 1948; D. L. Falkoff and G. E. Uhlenbeck, Phys. Rev. 73, 649A (1948).