

## Angular Correlation Perturbed by an Anisotropic Hyperfine Interaction\*

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A theoretical investigation is made of the effect on a gamma-gamma angular correlation of the extra-nuclear perturbations which arise from the coupling between the magnetic dipole and electric quadrupole moments of the nucleus and the electromagnetic field of the electron shell in a paramagnetic crystal of axial symmetry. The hyperfine interaction is assumed to be time independent. Results are obtained for both single-crystal and polycrystalline sources. For a single-crystal source, the effect on the correlation of an external magnetic field parallel to the crystalline axis is investigated. The behavior of the correlation over the whole range of partially to fully decoupled nuclear and electronic spins is calculated. It is shown that under certain conditions an "angular correlation resonance experiment" is possible, i.e., an experiment in which the coincidence counting rate, for fixed positions of the detectors, exhibits sharp interference peaks as a function of the magnetic field. For a powder source, the attenuation factors are computed as a function of the hyperfine constants for arbitrary nuclear spin values. New possibilities for investigation of nuclear moments of excited states are discussed.

### I. INTRODUCTION

THE gamma-gamma angular correlation of a nucleus which is surrounded by a paramagnetic electron shell is in general strongly perturbed by the hyperfine interaction, i.e., the interaction between the electromagnetic field created by the electrons and the magnetic dipole and electric quadrupole moments of the nucleus. Moreover, if the paramagnetic ion is embedded in a crystal, the hyperfine interaction is appreciably altered by the action of the static electric field produced by the neighbor ions (the crystalline field). The crystalline field interacts with the orbital motion of the electrons, completely or partially removing the degeneracy in the (ground) state of the free ion. Each electronic state in the crystalline field is characterized by a certain configuration of the electrons which reflects the symmetry of the crystal. In a crystalline field of noncubic symmetry the hyperfine interaction assumes an anisotropic form. In a field of axial symmetry this interaction is given by the following "spin-Hamiltonian"<sup>1,2</sup>:

$$K = AJ_z I_z + B(J_z I_x + J_y I_y) + P[I_z^2 - \frac{1}{3}I(I+1)], \quad (1)$$

where  $\mathbf{J}$  is an effective electronic angular momentum, and  $\mathbf{I}$  is the spin of the nucleus. The first two terms in (1) represent the magnetic dipole interaction, while the third term describes the quadrupole interaction. The quantities  $A$ ,  $B$ , and  $P$  are constants for a particular electronic state.

In an angular correlation experiment carried out under normal conditions, the perturbation of the correlation is further complicated by the interaction of

the ion with the phonons of the crystal and with the spins of the paramagnetic neighbors (spin-relaxation processes). As a consequence of these interactions each electronic state of the ion has a finite lifetime, and the observed perturbation of the correlation is the result of an average over many electronic states, each of which is associated with a different set of hyperfine constants  $A$ ,  $B$ , and  $P$ . In general, the effect of the spin-relaxation processes (characterized by spin-relaxation times) is a decrease in the over-all perturbation. For extremely short relaxation times (high temperatures) the effect can be an averaging to zero of the magnetic field near the nucleus and a return of the correlation to its unperturbed form.

The problem of computing the angular correlation for a general case in terms of the hyperfine constants and the relaxation times would be mathematically too complicated, even if a precise knowledge of these parameters for real crystals did exist. A great simplification occurs, however, if the relaxation times are such that during the lifetime of the intermediate nuclear state, the ion is almost exclusively in one electronic state. In this case the perturbation of the correlation can be treated as a static problem, and is independent of relaxation processes. A static environment may possibly be realized at low temperatures. The perturbation of the correlation in a static case depends only on the hyperfine constants,  $A$ ,  $B$ , and  $P$ , associated with the electronic state in question. If a theoretical calculation of the correlation function were available for such a case, a comparison with the measured correlation could provide a determination of these constants. From such a determination and a knowledge of the wave function of the electronic state, which is available through paramagnetic resonance absorption measurements for certain crystals, information about nuclear moments of excited states might be extracted.<sup>3</sup>

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<sup>1</sup> A. Abragam and M. H. L. Pryce, Proc. Roy. Soc. (London) **A205**, 135 (1951).

<sup>2</sup> R. J. Elliott and K. W. H. Stevens, Proc. Roy. Soc. (London) **A218**, 553 (1953).

<sup>3</sup> The calculations presented in this paper have been performed to provide the theoretical basis for an experiment in progress at this laboratory.

In this paper, a theoretical study is made of the perturbation of a gamma-gamma angular correlation caused by a static hyperfine interaction of the form (1). To this Hamiltonian a term is added which represents the interaction between an applied magnetic field along the crystalline axis and the electronic shell of the ion. The magnetic field is introduced in order that the coupling between the nucleus and the electron shell can be influenced in a controlled way. Since the angular correlation function of a gamma-gamma cascade for which the initial gamma emission is replaced by an absorption is identical with the angular distribution function of resonance fluorescence scattering, the results obtained here are valid for both processes.<sup>4</sup> The calculations are carried out for the important case of an electronic doublet state. Results are obtained for an arbitrary nuclear spin  $I$  of the intermediate state. Only the time-integrated perturbation of the correlation is considered, which means that the resolving time of the coincidence system is assumed long compared to the lifetime of the intermediate nuclear state.

Studies of angular correlations perturbed by an isotropic magnetic hyperfine interaction ( $A=B$ ) have been made by several authors.<sup>5-7</sup> Explicit results for an anisotropic hyperfine interaction have been obtained by Abragam and Pound<sup>8</sup> for the special case in which  $B$  is zero.

The general theory of perturbed angular correlations has been treated in detail in the literature.<sup>7</sup> The first step in the calculation of a perturbed correlation consists in solving the eigenvalue problem associated with the Hamiltonian which describes the interaction between the nucleus and the electromagnetic field of its surroundings. This problem is treated in Sec. II for the Hamiltonian described above. In Sec. III the angular correlation function is expressed in the form given by the general theory and the perturbation of the correlation is computed in terms of the eigenvalues and eigenvectors obtained in Sec. II. Section IV treats the angular correlation of a single-crystal source. The correlation function is investigated under the three separate conditions of maximum magnetic anisotropy, long intermediate state lifetime, and emission along the crystalline axis. Section V deals with the correlation of a crystalline powder source. A general formula is given for the attenuation factors. In Sec. VI the exact conditions under which the calculations apply are summarized and possible methods for measuring hyperfine coupling constants are discussed.

<sup>4</sup> The energy spread of the incident beam in the resonance fluorescence experiment should be larger than the energy splitting in the initial state, and further, the beam should be unpolarized.

<sup>5</sup> G. Goertzel, Phys. Rev. **70**, 897 (1946).

<sup>6</sup> K. Alder, Helv. Phys. Acta **25**, 235 (1952).

<sup>7</sup> For the most recent review article on the theory of angular correlations see: S. Devons and L. J. B. Goldfarb, in *Encyclopedia of Physics*, edited by S. Flügge (Springer-Verlag, Berlin, 1957), Vol. 42.

<sup>8</sup> A. Abragam and R. V. Pound, Phys. Rev. **92**, 943 (1953).

## II. HYPERFINE INTERACTION (HAMILTONIAN AND EIGENSTATES)

The hyperfine interaction in paramagnetic ions has been extensively investigated, mainly in connection with electronic paramagnetic resonance experiments and their interpretation. It has been shown by Abragam and Pryce,<sup>1</sup> for the iron group, and by Elliott and Stevens,<sup>2</sup> for the rare-earth group, that the hyperfine interaction associated with a great majority of the electronic states in crystals of axial symmetry is expressed by the following spin-Hamiltonian:

$$K = AJ_z I_z + B(J_x I_x + J_y I_y) + P[I_z^2 - \frac{1}{3}I(I+1)] + GJ_z, \quad (2)$$

with  $J = \frac{1}{2}$ .

Here, the  $z$  axis is identical with the symmetry axis of the crystal.  $\mathbf{I}$  is the nuclear spin operator and  $\mathbf{J}$  is, in general, an effective electronic spin operator. The magnitude of  $J$  is defined such that the multiplicity of the electronic state energy levels in the absence of the hyperfine interaction is  $(2J+1)$ . The restriction to the case  $J = \frac{1}{2}$  implies that the ionic states considered are doublets. The anisotropic magnetic hyperfine interaction is represented by the first two terms in (2), with  $A$  and  $B$  proportional to the magnetic dipole moment of the nuclear state. The term involving the quantity  $P$  describes the interaction between the nuclear quadrupole moment and the electron shell,  $P$  being proportional to the quadrupole moment. The last term in (2) represents the interaction between an applied magnetic field  $H_z$  (along the  $z$  axis) and the magnetic moment of the electron shell. The parameter  $G$  is defined by the relation

$$G = g_{\parallel} \mu_B H_z, \quad (3)$$

where  $g_{\parallel}$  is the electronic  $g$  factor parallel to the crystalline axis and  $\mu_B$  is the Bohr magneton. The magnitude of the applied field is assumed to be sufficiently small so that its direct interaction with the nuclear magnetic moment is negligible compared to all the terms included in Eq. (2).<sup>9</sup> It is further assumed that the direct interaction between the gradient of the crystalline field and the quadrupole moment of the nucleus can be neglected. This assumption is valid for the paramagnetic ions considered here. In references 1 and 2 and Baker and Bleaney<sup>10</sup> crystalline symmetries for which the spin Hamiltonian (2) is valid are discussed, and the quantities  $A$ ,  $B$ ,  $P$ , and  $g_{\parallel}$  are expressed in terms of the properties of the electronic states.

The remainder of this section is devoted to the solution of the eigenvalue problem defined by the Hamiltonian (2). It is known from the general theory, which is reviewed in the following section, that the dependence of the correlation on extra nuclear effects

<sup>9</sup> This is generally a valid assumption for magnetic fields as large as several thousand gauss.

<sup>10</sup> J. M. Baker and B. Bleaney, Proc. Roy. Soc. (London) **A245**, 156 (1958).

is completely determined by the eigenvalues and eigenvectors of the system in the intermediate state. The angular momentum properties of the system are naturally expressed by an expansion of the eigenvectors  $|b\rangle$  of  $K$  in terms of the complete set of angular momentum eigenstates  $|mn\rangle$ . The quantum numbers  $m$  and  $n$  are associated, respectively, with the possible projections,  $I_z$  and  $J_z$ , of the nuclear and (effective) electronic angular momenta along the  $z$  axis, for given quantum numbers  $I$  and  $J$ . We write

$$|b\rangle = \sum_{mn} |mn\rangle \langle mn|b\rangle, \quad (4)$$

where

$$K|b\rangle = E_b|b\rangle, \quad (5)$$

and  $n = \pm \frac{1}{2}$ . By substitution of the expansion (4) in (5) and use of the orthogonality of the states  $|mn\rangle$ , we obtain the eigenvalue equation

$$\sum_{mn} [\langle m'n'|K|mn\rangle - E_b \delta_{m'm} \delta_{n'n}] \langle mn|b\rangle = 0. \quad (6)$$

The quantities  $\mathbf{I}$  and  $\mathbf{J}$  are coupled quantum mechanically to form the resultant  $\mathbf{F}$ . The operator  $\mathbf{F}^2$  does not, in general, commute with the Hamiltonian  $K$  (except in the special cases  $A=B$  and  $B=0$ ). We note as a basic property of  $K$ , however, that it commutes with the operator  $F_z$  defined by

$$F_z = I_z + J_z. \quad (7)$$

The eigenstates of  $K$  can therefore be constructed to be simultaneous eigenstates of  $F_z$ . This is explicitly indicated by including in the "ket"  $|b\rangle$  the label  $M$  which specifies the eigenvalues of  $F_z$  according to the relation

$$F_z|(M)b\rangle = M|(M)b\rangle. \quad (8)$$

It follows that the coefficients in expansion (4) are nonzero only if  $m+n=M$ . Further, since  $J$  has the value  $\frac{1}{2}$ ,  $n$  has only the values  $\pm \frac{1}{2}$  and there are, for a given eigenstate, at most two nonzero coefficients  $\langle mn|b\rangle$  in expansion (4). Equations (6) thus reduce for a given  $M$  to a set of two equations. The associated secular equation of second order determines in general two distinct eigenvalues and eigenvectors for each  $M$ . With the following notation the eigenvalues and eigenvectors of  $K$  can be expressed in a convenient general form. We designate the eigenvalues and eigenvectors by  $E_{M(\pm)}$  and  $|M\pm\rangle$ , respectively, with the stipulation that, where  $M$  has its extreme values  $(I+\frac{1}{2})$  and  $-(I+\frac{1}{2})$ , we use the simpler notation  $E_M$  and  $|M\rangle$ . The former notation  $E_b$  and  $|(M)b\rangle$  (or simply  $|b\rangle$ ) is used whenever we refer to the eigenvalues and eigenvectors in general. The eigenvalues expressed in terms of  $A$ ,  $B$ ,  $P$ , and  $G$  are<sup>11</sup>

$$E_{M(\pm)} = -\frac{1}{4}A + [M^2 + \frac{1}{4} - \frac{1}{3}I(I+1)]P \pm \frac{1}{2}D_M, \\ \text{for } M = -I + \frac{1}{2}, -I + \frac{3}{2}, \dots, I - \frac{1}{2}; \quad (9)$$

$$E_{\pm(I+\frac{1}{2})} = \frac{1}{2}IA + \frac{1}{3}I(2I-1)P \pm \frac{1}{2}G, \\ \text{for } M = \pm(I + \frac{1}{2}),$$

where

$$D_M = \{[M(A-2P)+G]^2 + B^2[(I+\frac{1}{2})^2 - M^2]\}^{1/2}. \quad (10)$$

From Eqs. (6) and the unitary condition on the matrix of coefficients  $\langle mn|b\rangle$  we obtain for the nonzero eigenvector coefficients  $\langle mn|(M)b\rangle$ :

$$\langle M \pm \frac{1}{2}, \mp \frac{1}{2}|M\pm\rangle = (1+a_M^2)^{-1/2}, \\ \langle M \mp \frac{1}{2}, \pm \frac{1}{2}|M\pm\rangle = \pm a_M(1+a_M^2)^{-1/2}, \\ \text{for } M = -I + \frac{1}{2}, \dots, I - \frac{1}{2}; \quad (11)$$

$$\langle \pm I, \pm \frac{1}{2}|\pm(I+\frac{1}{2})\rangle = 1, \text{ for } M = \pm(I+\frac{1}{2});$$

where

$$a_M = \frac{D_M + M(A-2P) + G}{B[(I+\frac{1}{2})^2 - M^2]^{1/2}}. \quad (12)$$

The signs in Eqs. (9) and (11) are coherent in the sense that one must choose consistently either the upper or the lower sign.

### III. GENERAL FORM OF THE CORRELATION FUNCTION

The process under consideration is the successive emission of two gamma rays from a nucleus which is coupled to the electron shell via the electric and magnetic fields. The interaction between the nucleus and the shell is described by the Hamiltonian  $K$ . The initial, intermediate, and final (gross) states of the total system are characterized by sets of eigenstates of  $K$ , termed substates  $|a\rangle$ ,  $|b\rangle$ , and  $|c\rangle$ , respectively. The assembly of radioactive source atoms or ions is assumed to have equally populated initial substates  $|a\rangle$ .

The direction-direction correlation function is defined as the relative probability for observation of the second gamma radiation of the cascade transition in a direction  $\Omega_2$  if the first radiation is observed in the direction  $\Omega_1$ . The correlation function  $W$  is thus determined by the transition probability  $W_{ac}(t)$  for the emission of two gamma rays with definite directions and polarizations, the first emitted at time  $t=0$  and the second at time  $t$ , in the cascade transition between a given pair of states  $|a\rangle$  and  $|c\rangle$ <sup>8</sup>:

$$W = \sum_{1,2} \sum_{ac} \int_0^\infty \frac{dt}{\tau} e^{-t/\tau} W_{ac}(t), \quad (13)$$

where

$$W_{ac}(t) = \sum_{bb'} \langle a|H_1|b\rangle \langle b|H_2|c\rangle \langle c|H_2|b'\rangle \\ \times \langle b'|H_1|a\rangle e^{i(E_b - E_{b'})t/\hbar}.$$

$H_1$  and  $H_2$  are the Hamiltonians responsible for the emission of the first and second radiation, and  $\mathbf{S}_{1,2}$

<sup>11</sup> These eigenvalues are also given by Bleaney [see B. Bleaney, *Phil. Mag.* **42**, 441 (1951)]. Since in the correlation function only energy differences occur, we will drop in what follows the unimportant constant  $-\frac{1}{4}A + [\frac{1}{4} - \frac{1}{3}I(I+1)]P$ .

denotes a sum over the unobserved properties of the radiations. The time integral in Eq. (13) represents a mean over many decay processes, and  $\tau$  is the average lifetime of the intermediate state. The integration over  $t$  can be carried out and gives

$$W = \mathbf{S} \sum_{1,2} \sum_{abb'c} \frac{\langle a | H_1 | b \rangle \langle b | H_2 | c \rangle \langle c | H_2 | b' \rangle \langle b' | H_1 | a \rangle}{1 - (i\tau/\hbar)(E_b - E_{b'})}. \quad (14)$$

The terms in the summand of (14) which involve two distinct intermediate substates  $|b\rangle$  and  $|b'\rangle$  represent interference effects. These effects arise from the fact that the intermediate state through which a cascade transition between  $|a\rangle$  and  $|c\rangle$  proceeds is a superposition of all those substates  $|b\rangle$  for which both transition matrix elements  $\langle a | H_1 | b \rangle$  and  $\langle b | H_2 | c \rangle$  are nonzero. If there is only one substate  $|b\rangle$  with this property no interference effects can occur and the cascade transition can be said to proceed through a well-defined set of eigenstates  $|a\rangle$ ,  $|b\rangle$ , and  $|c\rangle$ .

In the special case in which  $I_z$  and  $J_z$  commute with the Hamiltonian  $K$ , the presence of interference terms is directly related to the perturbation of the correlation. Since  $I_z$  and  $J_z$  commute with  $K$ , the energy eigenstates in this case are simultaneously eigenstates of the nuclear and electronic angular momentum operators. The nuclear and electronic spin are thus effectively uncoupled, and since the electron shell is unaltered during the emission of the radiation, the intermediate eigenstates are essentially identical with those which occur in the unperturbed correlation. The total effect of the perturbation  $K$  in this case is the removal of the degeneracy in the eigenvalues of the intermediate state which gives rise to the energy denominator  $1 - (i\tau/\hbar)(E_b - E_{b'})$  in Eq. (14). In the absence of interference effects, no terms with  $E_b \neq E_{b'}$  occur and hence the perturbation of the intermediate state has no effect on the angular correlation. This property is referred to in subsequent sections.

The general direction-direction correlation function (14) can be reduced to the form<sup>6,8</sup>

$$W(\Omega_1\Omega_2) = \sum_{k_1 k_2 \mu_1 \mu_2} \frac{I(k_1)}{(2k_1+1)^{1/2}} \frac{II(k_2)}{(2k_2+1)^{1/2}} \times \text{III}(k_1 k_2 \mu_1 \mu_2) Y_{k_1 \mu_1}(\Omega_1) Y_{k_2 \mu_2}^*(\Omega_2), \quad (15)$$

where the spherical harmonic functions  $Y_{k\mu}(\Omega)$  express the dependence on the angles  $\Omega_1 = (\theta_1, \phi_1)$  and  $\Omega_2 = (\theta_2, \phi_2)$  which specify the emission directions of the first and second radiation, respectively. The summation indices  $k_1$  and  $k_2$  are even integers restricted by the conditions

$$0 \leq k_1, \quad k_2 \leq 2I, \quad 0 \leq k_1 \leq 2L_1, \quad 0 \leq k_2 \leq 2L_2, \quad (16)$$

where  $I$  is the nuclear spin of the intermediate state and  $L_1$  and  $L_2$  are, respectively, the highest orders in the multipole expansions of the successive radiations. The factors  $I(k_1)$  and  $II(k_2)$  are dependent on the

reduced matrix elements associated with the nuclear transitions and on the character of the radiations. These factors are defined in reference 6.

For pure multipole transitions<sup>12</sup>

$$\frac{I(k_1)}{(2k_1+1)^{1/2}} = F_{k_1}(L_1 I_A I); \quad \frac{II(k_2)}{(2k_2+1)^{1/2}} = F_{k_2}(L_2 I_C I), \quad (17)$$

where the functions  $F_k$  are tabulated in the review article of Biedenharn and Rose.<sup>13</sup> In the case of mixed transitions the quantities  $I(k_1)$  and  $II(k_2)$  can be obtained in terms of the mixing ratios by use of the articles by Rose<sup>14</sup> and Ferentz or Rosenzweig.<sup>15</sup> The total effect of a perturbation in the intermediate state is contained in the factor  $\text{III}(k_1 k_2 \mu_1 \mu_2)$ :

$$\text{III}(k_1 k_2 \mu_1 \mu_2) = \sum_{bb'} [1 - (i\tau/\hbar)(E_b - E_{b'})]^{-1} \times \mathbf{S}_{bb'}(k_1 \mu_1) \mathbf{S}_{bb'}(k_2 \mu_2), \quad (18)$$

where the quantity  $\mathbf{S}_{bb'}$  is determined by the coefficients in the expansion (4) for the eigenstates of  $K$ :

$$\mathbf{S}_{bb'}(k\mu) = \sum_{mm'n} \langle I m' k \mu | I m \rangle \langle m n | b \rangle \langle b' | m' n \rangle. \quad (19)$$

The bracket  $( | )$  denotes a Clebsch-Gordan coefficient.

If the perturbation  $K$  vanishes, the intermediate state energy levels are degenerate and the factor  $[1 - (i\tau/\hbar)(E_b - E_{b'})]^{-1}$  in Eq. (2) is identically one. It follows from the unitarity condition on the coefficients  $\langle m n | b \rangle$  that  $\text{III}$  becomes in this case

$$\text{III}(k_1 k_2 \mu_1 \mu_2) = [2(2I+1)/2k_1+1] \delta_{k_1 k_2} \delta_{\mu_1 \mu_2}, \quad (20)$$

and the correlation function reduces to the form<sup>16</sup>

$$W(\Theta) = \sum_k A_k P_k(\cos\Theta), \quad (21)$$

where  $\Theta$  measures the angle between the emission directions of the radiations, and  $A_k$  is defined by the relation

$$A_k = I(k)II(k)/(2k+1). \quad (22)$$

In the following, we compute the quantity  $\text{III}$  for the Hamiltonian  $K$  of Eq. (2). Using the commutability of  $K$  with  $F_z$  and the simplifications which arise from the special value  $J = \frac{1}{2}$ , we are able to carry out the sums over the magnetic quantum numbers. Since  $M$  is a good quantum number the summand in (19) is nonzero only for

$$m+n = M; \quad m'+n = M', \quad (23)$$

<sup>12</sup> Here  $I_A$ ,  $I$ , and  $I_C$  denote the nuclear spins of the initial, intermediate, and final states in the cascade process.

<sup>13</sup> L. C. Biedenharn and M. E. Rose, Rev. Mod. Phys. **25**, 729 (1953).

<sup>14</sup> M. E. Rose, Phys. Rev. **93**, 477 (1954).

<sup>15</sup> M. Ferentz and N. Rosenzweig, Atomic Energy Commission Report ANL-5324, 1955 (unpublished).

<sup>16</sup> We consistently omit over-all multiplicative factors in the correlation function.

where  $M$  and  $M'$  are the eigenvalues of  $F_z$  for the states  $|b\rangle$  and  $|b'\rangle$ , respectively. Using (23) together with the properties of the Clebsch-Gordan coefficients in the terms  $\mathbf{S}_{bb'}$ , we conclude that  $\text{III}(k_1 k_2 \mu_1 \mu_2)$  is zero unless

$$\mu_1 = \mu_2 = \mu = M - M'. \quad (24)$$

From the equality of  $\mu_1$  and  $\mu_2$  it follows that the correlation function (15) depends only on the angles  $\theta_1, \theta_2$  and  $\Phi = \phi_1 - \phi_2$ , reflecting the rotational symmetry of the Hamiltonian with respect to the polar axis  $Oz$ . By use of relation (23) the summations over  $m$  and  $m'$  can be eliminated in (19). Moreover, since  $n$  has only the two values  $\pm \frac{1}{2}$ , the sum over  $n$  in (19) gives rise to but two terms:

$$\begin{aligned} \mathbf{S}_{bb'}(k\mu) = & (IM' - \frac{1}{2}k\mu | IM - \frac{1}{2}) \langle M - \frac{1}{2}, \frac{1}{2} | b \rangle \langle b' | M' - \frac{1}{2}, \frac{1}{2} \rangle \\ & + (IM' + \frac{1}{2}k\mu | IM + \frac{1}{2}) \langle M + \frac{1}{2}, -\frac{1}{2} | b \rangle \\ & \times \langle b' | M' + \frac{1}{2}, -\frac{1}{2} \rangle. \end{aligned} \quad (25)$$

We note the property<sup>17</sup>

$$\mathbf{S}_{bb'}(k\mu) = (-1)^\mu \mathbf{S}_{b'b}(k - \mu). \quad (26)$$

It is convenient to write  $\text{III}(k_1 k_2 \mu \mu) = \text{III}(k_1 k_2 \mu)$  as a sum of two parts, the first of which contains all the terms in the sum over  $b$  and  $b'$  for which  $\mu$  is zero:

$$\begin{aligned} \text{III}(k_1 k_2 \mu) = & \text{III}(k_1 k_2 0) \delta_{\mu 0} \\ & + \sum_{\substack{bb' \\ M \neq M'}} [1 - (i\tau/\hbar)(E_b - E_{b'})]^{-1} \\ & \times \mathbf{S}_{bb'}(k_1 \mu) \mathbf{S}_{bb'}(k_2 \mu) \delta_{\mu, M - M'}. \end{aligned} \quad (27)$$

$\text{III}(k_1 k_2 0)$  contains the terms in which  $|b'\rangle = |b\rangle$  together with the interference terms between the pairs of states with the same quantum number  $M$ . By means of a short calculation  $\text{III}(k_1 k_2 0)$  can be expressed in terms of the quantities  $a_M$  [defined by (12) and (10)] in the form<sup>18</sup>

$$\begin{aligned} \text{III}(k_1 k_2 0) = & \frac{2(2I+1)}{2k_1+1} \delta_{k_1 k_2} - 2 \sum_{M=-I+\frac{1}{2}}^{I-\frac{1}{2}} \frac{a_M^2}{(1+a_M^2)^2} T_M \\ & \times [(IM - \frac{1}{2}k_1 0 | IM - \frac{1}{2}) - (IM + \frac{1}{2}k_1 0 | IM + \frac{1}{2})] \\ & \times [(IM - \frac{1}{2}k_2 0 | IM - \frac{1}{2}) - (IM + \frac{1}{2}k_2 0 | IM + \frac{1}{2})], \end{aligned} \quad (28)$$

where

$$T_M = \frac{(\tau/\hbar)^2 D_M^2}{1 + (\tau/\hbar)^2 D_M^2}, \quad (29)$$

and  $D_M$  is the energy difference between the states  $|M+\rangle$  and  $|M-\rangle$ .

With the aid of expression (27) we can carry out the sum over  $\mu$  in the correlation function (15) and obtain the final result.

$$\begin{aligned} W(\theta_1 \theta_2 \Phi) = & \sum_{k_1 k_2} \text{I}(k_1) \text{II}(k_2) \text{III}(k_1 k_2 0) P_{k_1}(\cos \theta_1) P_{k_2}(\cos \theta_2) \\ & + 2 \sum_{\substack{bb' \\ M > M'}} \sum_{k_1 k_2} \frac{\text{I}(k_1)}{(2k_1+1)^{1/2}} \frac{\text{II}(k_2)}{(2k_2+1)^{1/2}} \mathbf{S}_{bb'}(k_1, M - M') \\ & \times \mathbf{S}_{bb'}(k_2, M - M') a_{k_1}^{M - M'}(\theta_1) a_{k_2}^{M - M'}(\theta_2) \\ & \times \frac{\cos[(M - M')\Phi] - (\tau/\hbar)(E_b - E_{b'}) \sin[(M - M')\Phi]}{1 + (\tau/\hbar)^2 (E_b - E_{b'})^2}, \end{aligned} \quad (30)$$

where the real quantities  $a_k^\mu(\theta)$  are defined through the relation

$$Y_{k^\mu}(\theta \phi) = a_{k^\mu}(\theta) e^{i\mu \phi}. \quad (31)$$

$\text{III}(k_1 k_2 0)$  and  $\mathbf{S}_{bb'}(k\mu)$  are given by Eqs. (28) and (25), respectively. The coefficients  $\mathbf{S}_{bb'}$  describe the interference effects of pairs of states  $|b\rangle$  and  $|b'\rangle$  with  $M \neq M'$  in terms of the eigenvector coefficients given by Eqs. (11). The  $b, b'$  summation in (30) is over all pairs of such states.

By use of formula (30) the correlation function can be obtained for arbitrary nuclear spins and mixing ratios.

#### IV. ANGULAR CORRELATION FOR A SINGLE-CRYSTAL SOURCE

The correlation function developed in the last section describes a gamma-ray cascade for a single-crystal source with a well-defined symmetry axis  $Oz$ , along which an external magnetic field may be applied. This correlation function, in general, has a rather complicated analytical form. We restrict consideration to three cases which should exhibit the important properties of a correlation with a single-crystal source. The results are obtained in a fairly simple analytical form. In the first case either one of the radiations is assumed to be emitted along the crystalline axis. The angular correlation function can formally be written here in the same way as in the case of a powder source. In the second case the hyperfine interaction is assumed to have a maximum anisotropy, such that  $B=0$ . This condition leads to a considerable simplification in the eigenvalue problem itself. In the third case, the lifetime of the intermediate state is assumed to be such that the width of the state is small compared to the mean hyperfine splitting. Under certain conditions interesting resonance effects can occur here.

##### A. Radiation Along Symmetry Axis

The correlation function (30) is considerably simplified in the case that either radiation is observed along the symmetry axis. If the first gamma ray is emitted along  $Cz$ ,  $\theta_1$  is zero and it follows from the relation

$$Y_{k^\mu}(0, \phi) = a_{k^\mu}(0) e^{i\mu \phi} = \left( \frac{2k+1}{4\pi} \right)^{1/2} \delta_{\mu 0} \quad (32)$$

<sup>17</sup> The coefficients  $\langle mm | (M)b \rangle$  are real in our problem.

<sup>18</sup> We note that  $\text{III}(k00) = \text{III}(0k0) = 2(2I+1)\delta_{k0}$ .

that the correlation function contains only the first term in (30) and becomes

$$W(\theta_2) = \sum_{k_1 k_2} \text{I}(k_1) \text{II}(k_2) \text{III}(k_1 k_2 0) P_{k_2}(\cos\theta_2), \quad (33)$$

with  $\text{III}(k_1 k_2 0)$  given by Eq. (28). In general  $\text{I}(k)$  is different from  $\text{II}(k)$  and hence the correlation function is changed if the roles of the first and the second radiation are interchanged.

The absence of the second term of (30) in (33) means that there is no interference between states of different  $M$ . This can be understood in terms of the conservation of angular momentum.<sup>19</sup> Since the component of total angular momentum along the direction of emission of the first radiation is conserved, the intermediate state in this case has a definite  $F_z$ , and interference terms can arise only between those substates which have the same  $M$  value.

The perturbation of the correlation function (33) can be expressed through "attenuation factors"  $\bar{G}_k$  defined by rewriting (33) in the form<sup>20</sup>

$$W(\theta_2) = \sum_k \bar{G}_k^{(2)} A_k P_k(\cos\theta_2), \quad (34)$$

where  $A_k$  is given by (22), and

$$\begin{aligned} \bar{G}_k^{(2)} = & 1 - \frac{2k+1}{2I+1} \sum_{k' \neq 0} \frac{\text{I}(k')}{\text{I}(k)} \sum_{M=-I+\frac{1}{2}}^{I-\frac{1}{2}} \frac{a_M^2}{(1+a_M^2)^2} T_M \\ & \times [ (IM - \frac{1}{2}k'0 | IM - \frac{1}{2}) - (IM + \frac{1}{2}k'0 | IM + \frac{1}{2}) ] \\ & \times [ (IM - \frac{1}{2}k0 | IM - \frac{1}{2}) - (IM + \frac{1}{2}k0 | IM + \frac{1}{2}) ]. \quad (35) \end{aligned}$$

The quantity  $T_M$  defined by Eq. (29), contains the dependence of the correlation function on the lifetime  $\tau$  of the intermediate state. If the lifetime is sufficiently

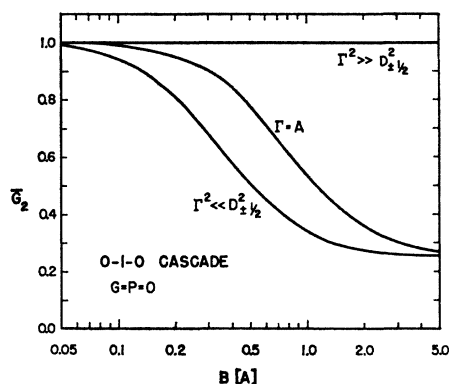


FIG. 1. Attenuation factor  $\bar{G}_2$  for a cascade with spin sequence 0-1-0 plotted as a function of the magnetic anisotropy  $B/A$  for different values of the width of the intermediate state.

<sup>19</sup> Although  $F_z$  is not the true angular momentum of the system the conservation law can nevertheless be formally applied.

<sup>20</sup> If the second radiation is emitted along the crystalline axis, Eq. (34) contains  $\theta_1$  instead of  $\theta_2$  and  $\bar{G}_k^{(2)}$  is replaced by  $\bar{G}_k^{(1)}$ . The latter is defined by (35) with  $\text{I}(k)$  replaced by  $\text{II}(k)$ .

long such that the width  $\Gamma = \hbar/\tau$  of the intermediate state is much smaller than the hyperfine splitting between the states, the correlation is strongly perturbed and  $T_M$  can be approximated by unity. From Eq. (29) the "strong perturbation" condition is seen to be

$$D_M^2 \gg \Gamma^2, \text{ for all } M \text{ with } |M| \neq I + \frac{1}{2}, \quad (36)$$

where  $D_M$  is the energy difference between the states  $|M+\rangle$  and  $|M-\rangle$ . If the lifetime is very short, such that  $D_M^2 \ll \Gamma^2$ , the quantities  $T_M$  approach zero and  $\bar{G}_k$  becomes one. It follows in this case that the correlation is unperturbed as expected.

Formula (35) can be used to compute the correlation function for a given set of parameters  $A$ ,  $B$ ,  $P$ , and  $G$ . It is of interest to study the correlation of a particular gamma-ray cascade as a function of the magnetic anisotropy  $B/A$ . We choose, as an example,  $G=P=0$  and a cascade with spin sequence 0-1-0. The complete correlation in this case is given by  $W(\theta) = 1 + \bar{G}_2 A_2 \times P_2(\cos\theta)$ . Figure (1) shows the attenuation factor  $\bar{G}_2$  as a function of  $B$  (in units of  $A$ ) for a fixed value of  $A$ . The different curves correspond to different values for the lifetime of the intermediate state. For  $B$  equal to zero (see also the following subsection),  $\bar{G}_2$  has the value unity for all lifetimes. This means that the correlation  $W(\theta)$  is unperturbed in the case of a maximum magnetic anisotropy. If we choose as a representative value of  $A$ ,  $10^{-2} \text{ cm}^{-1}$ , the curve  $\Gamma = A$  corresponds to a nuclear lifetime  $\tau$  equal to approximately  $5 \times 10^{-10} \text{ sec}$ .

It is particularly interesting to discuss the correlation  $W(\theta)$  as a function of the strength of the external magnetic field applied along the crystalline axis. For a strong magnetic field, the  $G$  term in the Hamiltonian  $K$  dominates the hyperfine coupling terms ( $G \gg A, B, P$ ). According to Eq. (12) the factor  $a_M^2/(1+a_M^2)^2$  in (35) becomes very small in this case and the attenuation factor  $\bar{G}_K$  approaches the value one. The correlation in a strong magnetic field is therefore unperturbed. This result has been noted before in the special case of an isotropic hyperfine interaction.<sup>5,6</sup> It can be understood on the basis of the general discussion in Sec. III. We apply a perturbation treatment to the eigenvalue problem associated with the Hamiltonian  $K$ , where the (unperturbed) Hamiltonian  $K_0$  is defined as the  $G$  term. Since  $I_z$  and  $J_z$  commute with  $K_0$ , the electronic spin is, in first approximation, decoupled from the nuclear spin; and it follows that a perturbation of the correlation can arise only through interference terms. With one radiation emitted along the crystalline axis, however, a cascade transition between a particular set of initial and final substates can proceed only through the two intermediate substates  $|M+\rangle$  and  $|M-\rangle$  with well-defined total angular momentum projection  $M$ . Moreover, since the electron shell, during the emission of the first gamma ray, remains in the initial state characterized by a definite projection,  $n_a$ , of the electronic spin, there is but *one* intermediate substate,

namely, that with  $m=M-n_a$ , which can be reached in the cascade, and interference terms cannot occur. The correlation is, therefore, unperturbed.

With the aid of formula (35) the correlation can be computed for arbitrary magnetic fields, that is, over the whole range of partly decoupled nuclear and electronic spins. As an example, we consider again the cascade with spin sequence 0-1-0, and choose  $P=0$  and  $B=4A$ . In Fig. 2 the attenuation factor  $\bar{G}_2$  for this case is plotted for different lifetimes as a function of the strength  $G$  of the external magnetic field. In the strong perturbation limit ( $\Gamma \ll A$ ) a magnetic field of about 5000 G is needed to reach a decoupling of  $\bar{G}_2 = 0.99$ , when the values  $A \approx 10^{-2} \text{ cm}^{-1}$  and  $g_{11} \approx 2$  are assumed.

### B. Maximum Anisotropy ( $B=0$ )

The hyperfine interaction which occurs in certain ionic states in axially symmetric crystals is described to a good approximation by the Hamiltonian (2) with  $B$  equal to zero.<sup>10</sup> Since  $K$  in this case commutes with  $I_z$  and  $J_z$ , the energy eigenstates  $|b\rangle$  are simultaneously eigenstates of the projections of the nuclear and electronic angular momenta. The eigenvalues and eigenvectors, characterized by  $m$  and  $n$ , can be written as

$$E_b = E_{mn} = mnA + [m^2 - \frac{1}{3}I(I+1)]P + nG; \quad (37)$$

$$|b\rangle = |mn\rangle.$$

It follows from Eqs. (18) and (19) that the factor  $\text{III}(k_1 k_2 \mu \mu)$  in this case has the form

$$\text{III}(k_1 k_2 \mu) = \sum_{mm'} \sum_n [1 - (i\tau/\hbar)(E_{mn} - E_{m'n})]^{-1} \\ \times (Im'k_1\mu | Im)(Im'k_2\mu | Im). \quad (38)$$

Introducing the eigenvalues (37), we obtain

$$\text{III}(k_1 k_2 \mu) = \sum_{mm'} (Im'k_1\mu | Im)(Im'k_2\mu | Im) \\ \times \left[ \left\{ 1 - (i\tau/\hbar) \left[ (m^2 - m'^2)P - \frac{1}{2}\mu A \right] \right\}^{-1} \right. \\ \left. + \left\{ 1 - (i\tau/\hbar) \left[ (m^2 - m'^2)P + \frac{1}{2}\mu A \right] \right\}^{-1} \right]. \quad (39)$$

It is interesting to note that  $\text{III}(k_1 k_2 \mu)$  is independent of  $G$ , which means that the correlation is not affected by the magnetic field along the crystalline axis.<sup>21</sup> If the quadrupole interaction can be neglected ( $P=0$ ), Eq. (39) is reduced to the form

$$\text{III}(k_1 k_2 \mu) = \delta_{k_1 k_2} \frac{2I+1}{2k_1+1} \left[ \frac{1}{1 + (i\tau/2\hbar)\mu A} \right. \\ \left. + \frac{1}{1 - (i\tau/2\hbar)\mu A} \right]. \quad (40)$$

<sup>21</sup> This statement is not restricted to the case of a magnetic field parallel to the axis, but is valid for an arbitrary magnetic field direction, since the component of the electronic  $g$  factor perpendicular to the axis is usually zero for ions with  $B=0$ .

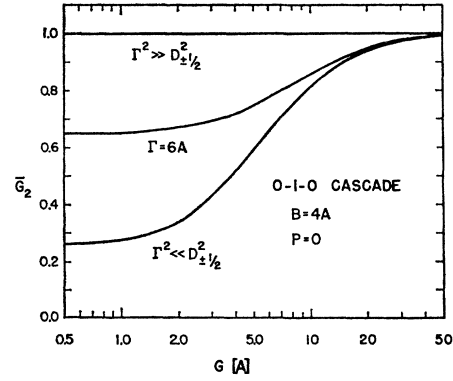


FIG. 2. Attenuation factor  $\bar{G}_2$  for a cascade with spin sequence 0-1-0 plotted as a function of the external magnetic field for different lifetimes of the intermediate state.

We consider here two special cases, one where the radiation is emitted along the crystalline axis, and the other where the crystalline axis is perpendicular to the plane defined by the source and the two detectors (subsequently referred to as the detector plane). In the first, only the terms in the correlation function with  $\mu=0$  are nonvanishing (see subsection A). It follows that (39) in this case is identical with (20), and hence the correlation is restored to the unperturbed form. This result, again, can be viewed as a consequence of the absence of interference terms.

In the case where the crystalline axis is perpendicular to the detector plane the correlation can, in general, be expressed as<sup>8</sup>

$$W(\Phi) = \sum_{\mu} B_{\mu} e^{i\mu\Phi}; \quad \Phi = \phi_1 - \phi_2, \quad (41)$$

with

$$B_{\mu} = \sum_{k_1 k_2} \frac{I(k_1)}{(2k_1+1)^{1/2}} \frac{\text{II}(k_2)}{(2k_2+1)^{1/2}} \\ \times \text{III}(k_1 k_2 \mu) a_{k_1}^{\mu}(\pi/2) a_{k_2}^{\mu}(\pi/2), \quad (42)$$

and the functions  $a_{k}^{\mu}(\pi/2)$  defined by relation (31).

If  $P$  here is zero, the perturbation can be expressed by attenuation factors  $G_{\mu}$ , which depend only on the Hamiltonian  $K$  and are independent of the properties of the radiations. These factors are defined by rewriting (41) in the form

$$W(\Phi) = \sum_{\mu} G_{\mu} b_{\mu} e^{i\mu\Phi}, \quad (43)$$

where  $b_{\mu}$  is the value of the coefficient  $B_{\mu}$  in (41) for an unperturbed correlation. Using (42), (20), and (40) we find

$$G_{\mu} = \frac{1}{2} \left[ \frac{1}{1 + (i\tau/2\hbar)\mu A} + \frac{1}{1 - (i\tau/2\hbar)\mu A} \right]. \quad (44)$$

A comparison of this result with the attenuation factor obtained by Alder<sup>6</sup> for a correlation perturbed by the

interaction between the nucleus and an external magnetic field perpendicular to the detector plane reveals the close connection between Alder's case and the case considered here ( $B=0$ ). The attenuation factor given by (44) can be interpreted as the mean value of two attenuation factors which arise from effective internal magnetic fields  $H_{\text{eff}}$  along the positive and negative  $z$  directions, respectively. The fields are produced by the electron shell and correspond to two possible orientations of the (effective) electronic spin, with

$$H_{\text{eff}} = \frac{1}{2}A/g_N\mu_N. \quad (45)$$

Here  $g_N$  is the nuclear  $g$  factor and  $\mu_N$  the nuclear magneton.

A close connection between the case  $B=0$  and cases previously treated in the literature exists even where there is a nonvanishing quadrupole interaction. The factor  $\text{III}(k_1k_2\mu)$ , given by Eq. (39) can be interpreted as the mean value of the factors  $\text{III}(k_1k_2\mu)$  and  $\text{III}(k_1k_2-\mu)$  associated with the problem of a correlation perturbed by an external magnetic field (acting on the nucleus) and a quadrupole interaction caused by the crystalline field.<sup>8,22,23</sup> [Compare, e.g., Eq. (39) with Eq. (77) of reference 8.] The relation between the hyperfine coupling constant  $A$  and the external magnetic field is given by expression (45). By means of this relationship, the known results for the correlation problem mentioned can be directly applied to the case treated here.

### C. Strong Perturbation

Under certain conditions the interference terms in the correlation function (14), i.e., the terms which involve distinct intermediate states  $|b\rangle$  and  $|b'\rangle$ , become relatively small and can be neglected. This occurs when the lifetime of the intermediate state is much longer than the quantity  $\hbar/(E_b - E_{b'})$ . In this limit of "strong perturbation" the time integral of the cross terms in Eq. (13) becomes vanishingly small due to the rapidly oscillating exponential factor in  $W_{ac}(t)$ . Since the time integral represents an average over many decay processes, the disappearance of the interference terms in the strong perturbation limit can be viewed as a consequence of the random distribution of the time dependent phase factors of the states  $b$  and  $b'$ , resulting from the randomness in the time of emission of the second gamma ray. Alternatively, one can say that the interference between states  $b$  and  $b'$  vanishes whenever the energy separation between the states is much greater than the natural width  $\hbar/\tau$  of the intermediate state. In this case the system can be said to pass through a well-defined intermediate energy state in making the transition between the initial and the final substate.

<sup>22</sup> K. Alder, H. Albers-Schonberg, E. Heer, and T. B. Novey, *Helv. Phys. Acta.* **26**, 761 (1953).

<sup>23</sup> K. Alder, E. Matthias, W. Schneider, and R. M. Steffen (to be published).

In discussing the strong perturbation limit it is convenient to distinguish two cases according to whether or not degeneracies occur in the eigenstates of the Hamiltonian  $K$ .

#### (1) Eigenstates of $K$ Nondegenerate

In general, the eigenstates of the Hamiltonian  $K$  in the intermediate state are nondegenerate and the strong perturbation condition can be expressed by the relation

$$|E_b - E_{b'}| \gg \Gamma, \quad (46)$$

where  $\Gamma = \hbar/\tau$  is the width of the intermediate state. If relation (46) is fulfilled for all pairs of states, the cross terms in the correlation function (14) can be neglected and Eq. (30) assumes the form

$$W_0(\theta_1\theta_2) = \sum_{k_1k_2} \text{I}(k_1)\text{II}(k_2)\text{III}_0(k_1k_20) \times P_{k_1}(\cos\theta_1)P_{k_2}(\cos\theta_2), \quad (47)$$

where  $\text{III}_0(k_1k_20)$  is the "strong perturbation part" of  $\text{III}(k_1k_20)$ , i.e., expression (28) with  $T_M=1$ . Formula (47) is valid for arbitrary emission directions of the two gamma rays. The disappearance of the  $\Phi$  dependence in this equation should be noted. It means, in particular, that the correlation for a single-crystal source with axis of symmetry perpendicular to the detector plane is completely isotropic. This result follows directly from the disappearance of the interference terms. In this case the second gamma ray is always emitted from a state with definite  $M$ . Hence the second radiation, connecting two states with definite angular momentum  $F_z$ , has also a well-defined angular momentum projection along  $O_z$  and the conjugate angular coordinate is, therefore, completely undetermined. It follows that the correlation function is independent of the angle  $\Phi$ .

For a nuclear spin  $I$  equal to or less than two, we obtain for the nonzero coefficients  $\text{III}_0(k_1k_20)$  the results:

$$\begin{aligned} \text{III}_0(000) &= 2(2I+1), \\ I=1: \quad \text{III}_0(220) &= (3/5)(2-3X_{1/2}), \\ I=3/2: \quad \text{III}_0(220) &= (8/5)(1-X_1), \\ I=2: \quad \text{III}_0(220) &= (1/7)(14-X_{1/2}-9X_{3/2}), \\ \text{III}_0(240) &= \text{III}_0(420) = (5/21)(2X_{1/2}-3X_{3/2}), \\ \text{III}_0(440) &= (5/63)(14-20X_{1/2}-5X_{3/2}), \end{aligned} \quad (48)$$

with

$$X_M = \frac{a_M^2}{(1+a_M^2)^2} + \frac{a_{-M}^2}{(1+a_{-M}^2)^2}.$$

#### (2) Degeneracies in Eigenstates of $K$ (Interference Effects)

If the parameters  $A$ ,  $B$ ,  $P$ , and  $G$  in the Hamiltonian of Eq. (2) have values such that certain of the eigen-



states are degenerate (or nearly degenerate), relation (46) is not satisfied for these states. The correlation function in this case is a sum of  $W_0(\theta_1, \theta_2)$ , Eq. (47), and the interference terms of Eq. (30) which involve the degenerate (or nearly degenerate) states. This means that whenever the energy separation between two intermediate substates is smaller than or of the order of the width  $\Gamma$ , it is impossible to determine the substate through which a cascade transition has passed and the substates interfere. In certain cases, however, interference terms which arise in this manner vanish as a result of the selection rules to be discussed below.

In the following a discussion of possible degeneracies in the eigenstates of the Hamiltonian (2) is given.

(a) An important case of degeneracy occurs for an isotropic magnetic interaction ( $A=B$ ) with  $G=P=0$ . The eigenvalue spectrum contains only two different energies corresponding to the total spin quantum numbers  $F=I+\frac{1}{2}$  and  $F=I-\frac{1}{2}$ , respectively. The high degree of degeneracy gives rise to a considerable number of interference terms which, in general, do not cancel. The effect of this degeneracy on the correlation function of a powder source is treated in Sec. V. An example of an ( $A=B$ ) degeneracy with  $G$  and  $P$  different from zero is discussed in this section (see Fig. 7).

(b) A quadrupole interaction of appropriate magnitude ( $P=\frac{1}{2}A$ ) compensates the  $A$  term of the magnetic interaction and causes a degeneracy of the pairs of states with quantum numbers  $M$  and  $-M$ . The corresponding interference terms are, in general, nonvanishing.

(c) In the absence of the external magnetic field ( $G=0$ ), each state  $|(M)b\rangle$  is degenerate with a state  $|(-M)b'\rangle$ , and the Hamiltonian  $K$  commutes not only with  $F_z$  but also with the time reversal operator. Each of the two degenerate states is essentially the time reverse of the other. It can be proved that the quantity  $\mathbf{S}_{bb'}$  of Eq. (19) is zero for these states unless  $(-1)^{2M}=1$ , i.e., unless  $M$  is an integer. Therefore, the interference terms between the degenerate states are zero whenever  $I$  has an integral value. If  $I$  is a half integer there are in general nonvanishing interference effects.

(d) A study of the eigenvalues of  $K$  in Eqs. (9) for given values of  $A$ ,  $B$ , and  $P$  shows that degeneracies can occur at certain discrete values of the external field (values of  $G$ ) other than zero.<sup>24</sup> The interference terms between the degenerate states at these critical values  $G_c$  cause the strongly perturbed correlation to assume a resonant behavior as a function of  $G$ . This behavior makes possible an "angular correlation reso-

nance experiment" which consists in a measurement of the coincidence counting rate for a fixed position of the two detectors as a function of the applied magnetic field.<sup>25</sup> The width of the "resonance line" in such an experiment depends essentially on the lifetime of the intermediate state and can become arbitrarily narrow for a sufficiently long lifetime. The resonance phenomenon is not present if one radiation is measured along the crystalline axis, since then only states with the same  $M$  can interfere, and the crossing of the corresponding levels is prohibited by the general rule which states that levels with the same symmetry never cross. A convenient arrangement for an angular correlation resonance experiment is one where the crystalline axis is perpendicular to the detector plane, i.e.,  $\theta_1=\theta_2=\pi/2$ . From Eq. (30) it follows in this case that the correlation function in the vicinity of two crossing levels  $E_b$  and  $E_{b'}$  (with distinct  $M$  values) is given by

$$W(\Phi) = W_0(\pi/2, \pi/2) + W_R(\Phi),$$

where

$$W_R(\Phi) = 2 \sum_{k_1 k_2} \frac{I(k_1)}{(2k_1+1)^{1/2}} \frac{\Pi(k_2)}{(2k_2+1)^{1/2}} \mathbf{S}_{bb'}(k_1, M-M') \\ \times \mathbf{S}_{bb'}(k_2, M-M') a_{k_1}^{M-M'}(\pi/2) a_{k_2}^{M-M'}(\pi/2) \\ \times \frac{\cos[(M-M')\Phi] - [(E_b - E_{b'})/\Gamma] \sin[(M-M')\Phi]}{1 + (E_b - E_{b'})^2/\Gamma^2}. \quad (49)$$

The resonant behavior of  $W(\Phi)$  as a function of the external magnetic field for a fixed angle  $\Phi$  results essentially from the magnetic field dependence of  $(E_b - E_{b'})$  in  $W_R$ . The quantities  $W_0$  and  $\mathbf{S}_{bb'}$  vary slowly with the magnetic field in the vicinity of the critical field.

From the form of  $W_R(\Phi)$  in (49) it follows that certain "selection rules" have to be fulfilled in order that the resonant term be nonvanishing. From the properties of the functions  $a_{k,\mu}(\theta)$ , defined through the spherical harmonics, it can be seen that interference effects can be observed only between those pairs of levels for which

$$|M-M'| \leq \min\{(k_1)\max, (k_2)\max\}, \quad (50)$$

and (since  $\theta=\pi/2$ )

$$|M-M'| = \text{even integer}. \quad (51)$$

At the critical magnetic field value for which the levels  $E_b$  and  $E_{b'}$  exactly cross, the angular dependence of the resonant term  $W_R$  in the correlation function (49) is expressed through the factor  $\cos[(M-M')\Phi]$ .

<sup>24</sup> In the present discussion it is assumed that  $B \neq 0$ . In the case of maximum anisotropy ( $B=0$ ), level crossings at particular values of  $G$  also occur. The corresponding interference terms in this case are bound to be zero, however, since they involve always pairs of states with different  $n$  values, and only levels with  $n=n_a$  can be reached in a cascade transition originating from  $|a\rangle$ .

<sup>25</sup> It has come to our attention recently that similar phenomena have been investigated in connection with resonance fluorescence experiments in the field of atomic spectroscopy: F. D. Colegrove, P. A. Franken, R. R. Lewis, and R. H. Sands, Phys. Rev. Letters **3**, 420 (1959); P. A. Franken, Phys. Rev. **121**, 508 (1961); M. E. Rose and R. L. Carovillano, *ibid.* **122**, 1185 (1961). Rose and Carovillano point out that resonance effects should occur in angular correlations.

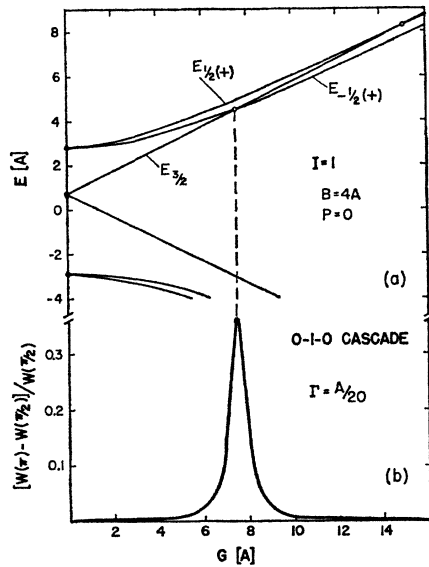


FIG. 3. (a) Eigenvalue spectrum for  $I=1$  as a function of the external magnetic field. (b) Anisotropy of the correlation for a 0-1-0 cascade plotted as a function of the external magnetic field. The interference peak corresponds to the crossing of the levels  $E_{3/2}$  and  $E_{-1/2}(+)$ .

$W_R$  has therefore extreme values  $\Phi_n = n\pi / |M - M'|$ ;  $n=1, 2, 3, \dots$ . The resonance effect in an experiment can be maximized by making measurements at two angles for which the resonant term has opposite sign. For example, if the interference is between levels for which  $|M - M'| = 2$ , the maximum effect is obtained by measuring as a function of the magnetic field the quantity  $[W(\pi) - W(\pi/2)] / W(\pi/2)$  termed the "anisotropy function."

The theoretical variation of the anisotropy function with external field strength in the region of an energy level crossing is shown graphically in Figs. 3(b), 6, and 7(b) for three example correlations. Figure 3 refers to a cascade with spin sequence 0-1-0. The energy spectrum of the hyperfine interaction in the intermediate state as a function of the magnetic field parameter  $G$  is shown in Fig. 3(a). The hyperfine constants are assumed here to have the values  $B=4A$  and  $P=0$ . The spectrum shows five level crossings. The three that occur at  $G=0$  do not give rise to a resonance effect since  $I$  has an integral value. [See the discussion of the ( $G=0$ ) degeneracy given above.] Moreover, the resonant term arising from the crossing of the  $E_{3/2}$  and  $E_{1/2}(+)$  levels vanishes due to the chosen geometry according to rule (51). The effect on the anisotropy function of the level crossing  $(\frac{3}{2}; -\frac{1}{2}+)$ <sup>26</sup> is shown in Fig. 3(b) for an assumed level width  $\Gamma = (1/20)A$ . We like to emphasize that the existence of a sharp interference peak presumes a sufficiently small width of the intermediate state, such that

<sup>26</sup> We denote the crossing of two levels with the subscripts of the corresponding energies.

condition (46) is fulfilled for all pairs of states (with nonzero interference term) except for the pair  $(\frac{3}{2}; -\frac{1}{2}+)$  in the vicinity of the critical field.

For arbitrary  $A$  and  $B$  (and  $P=0$ ) the critical magnetic field  $H_c$  at the crossing  $(\frac{3}{2}; -\frac{1}{2}+)$  has the value

$$H_c = \frac{1}{2} \left( \frac{B^2}{A^2} - 1 \right) \frac{A}{g_{11}\mu\beta}. \quad (52)$$

The width of the resonance line at half-maximum is given by the approximate relation

$$\Delta H \approx 2 \left( 1 + \frac{1}{2} \frac{B^2}{A^2} \right) \frac{\Gamma}{g_{11}\mu\beta}, \quad (53)$$

where  $A$  and  $B$  appear only through their ratio. For a correlation with known spins and mixing ratios the height of the resonance peak at the critical field  $H_c$  is, in general, a function of the parameters  $A$ ,  $B$ , and  $P$ . In Fig. 4, the maximum of the resonance peak associated with the crossing  $(\frac{3}{2}; -\frac{1}{2}+)$  is plotted as a function of the magnetic anisotropy  $B/A$  with  $P$  equal to zero. The plot shows that the maximum of the interference peak is large if  $B$  is much larger than  $A$ , and has a value of 36% for  $B=4A$ . Assuming the representative values  $A \approx 10^{-2}$  cm<sup>-1</sup>, and  $g_{11} \approx 2$ , the values of  $\tau$  and  $H_c$  for the example of Fig. 3 are:  $\tau \approx 10^{-8}$  sec,  $H_c \approx 800$  G.

In Figs. 5 and 6, plots similar to those of Fig. 3 are made for a cascade with spin sequence  $\frac{1}{2} - \frac{3}{2} - \frac{1}{2}$  with various lifetimes for the intermediate state. The energy spectrum, Fig. 5, exhibits five level crossings, only two of which, occurring at  $G_c=0$ , satisfy the selection rules (50) and (51). The interference terms between  $M$  and  $-M$  states do not vanish in this case since  $I$  has a half-integral value. The width of the resonance line at  $G=0$  is given by the formula

$$\Delta H \approx 2 \left( 1 + 3 \frac{B^2}{A^2} \right)^{1/2} \frac{\Gamma}{g_{11}\mu\beta}. \quad (54)$$

Again, the width depends on  $A$  and  $B$  only through their ratio.

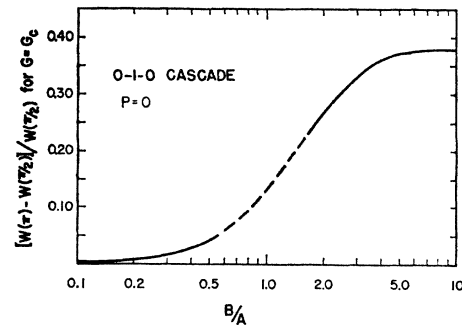


FIG. 4. Anisotropy of the 0-1-0 correlation at the critical magnetic field plotted as a function of the magnetic anisotropy  $B/A$ .

Figure 7 refers to the example of the 0-1-0 cascade in the case of an isotropic magnetic interaction ( $A=B$ ). Figures 7(a) and 7(b) show the eigenvalue spectrum and anisotropy function plotted against the external field for  $P=A$ . The critical field strength of the resonance is given for arbitrary  $A$  and  $P$  by the expression

$$H_c = P/g_{11}\mu_B. \quad (55)$$

In the limit of vanishing quadrupole interaction,  $P=0$ , the position of the level crossing ( $\frac{3}{2}; -\frac{1}{2}+$ ) coincides with that of the crossing ( $-\frac{3}{2}; \frac{1}{2}+$ ) and a resonance line equal to the sum of the two shown occurs at  $G=0$ . The width is

$$\Delta H \approx 3\Gamma/g_{11}\mu_B. \quad (56)$$

This situation is realized in a gaseous source and in a crystalline source of cubic symmetry.

### V. ANGULAR CORRELATION FOR A POWDER SOURCE

A powder source represents an assembly of single crystals with their symmetry axes oriented at random. The correlation for such a source without external field

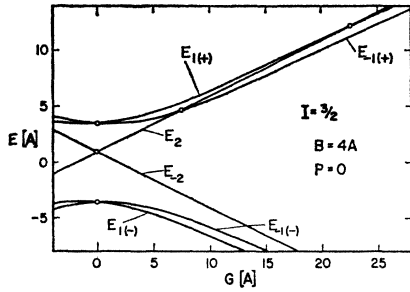


FIG. 5. Eigenvalue spectrum for  $I=3/2$  as a function of the external magnetic field.

( $G=0$ ) can be obtained by an averaging of the correlation function for a single-crystal source over all orientations of the frame of reference with fixed emission directions for the radiations. Replacing the product of spherical harmonics in the correlation function (15) by the average value of the product,  $(1/4\pi)P_{k_1}(\cos\Theta) \times \delta_{k_1 k_2} \delta_{\mu_1 \mu_2}$ , the powder correlation function is obtained in the form<sup>8</sup>

$$W(\Theta) = \sum_k G_k A_k P_k(\cos\Theta), \quad (57)$$

where the attenuation factors  $G_k$  are given by

$$G_k = \frac{1}{2(2I+1)} \sum_{\mu} \text{III}(kk\mu\mu). \quad (58)$$

Using Eq. (18) we obtain for  $G_k$  the formula

$$G_k = \frac{1}{2(2I+1)} \sum_{bb'} [1 - (i\tau/\hbar)(E_b - E_{b'})]^{-1} \times [\mathbf{S}_{bb'}(k, M-M')]^2, \quad (59)$$

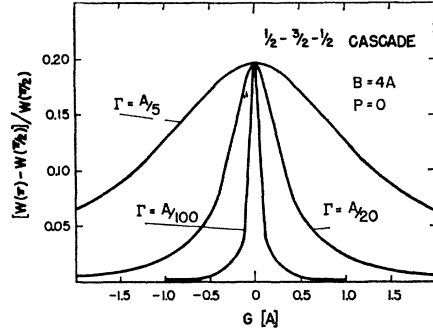


FIG. 6. Anisotropy of a  $\frac{1}{2}-\frac{3}{2}-\frac{1}{2}$  correlation plotted as a function of the external magnetic field for different lifetimes of the intermediate state.

with  $\mathbf{S}_{bb'}(k, M-M')$  given by expression (25). Formula (59) can be considerably simplified in the strong perturbation limit, where  $|E_b - E_{b'}|$  is much larger than  $\Gamma$  for all pairs of distinct states except those with  $M' = -M$  (which are degenerate since  $G=0$ ). The result is

$$G_k^0 = \frac{1}{2k+1} - \frac{1}{2I+1} \sum_{M=-I+\frac{1}{2}}^{I-\frac{1}{2}} \frac{a_M^2}{(1+a_M^2)^2} \times \{ [(IM - \frac{1}{2}k0 | IM - \frac{1}{2}) - (IM + \frac{1}{2}k0 | IM + \frac{1}{2})]^2 - [1 + (-1)^{2M}]^2 (IM + \frac{1}{2}k, -2M | I, -M + \frac{1}{2})^2 \}. \quad (60)$$

The last term in the above formula arises from the interference between the states  $|(M)b\rangle$  and  $|(-M)b'\rangle$  and is nonzero only if  $I$  is a half-integer. Equation (60) is not valid if there are degeneracies with non-

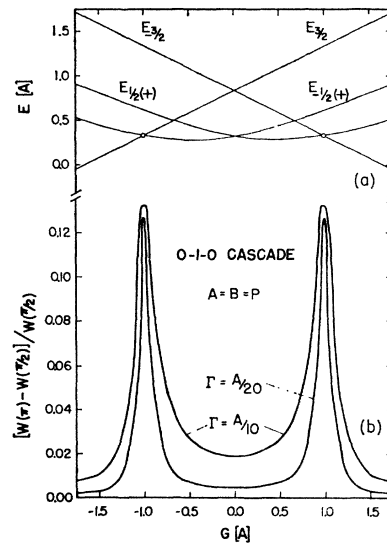


FIG. 7. (a) Eigenvalue spectrum for  $I=1$  and  $A=B=P$  as a function of the external magnetic field. (b) Anisotropy of the 0-1-0 correlation with  $A=B=P$  plotted as a function of the external magnetic field.

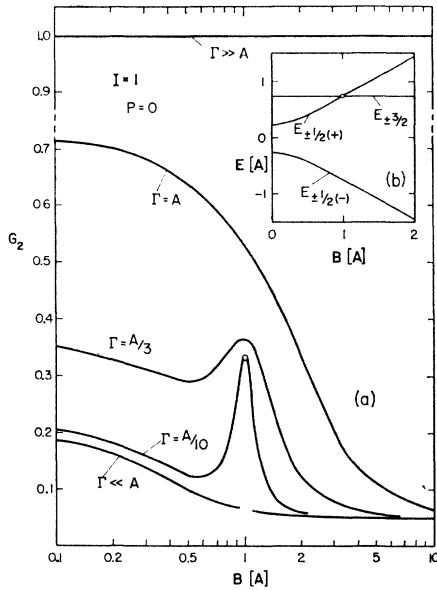


FIG. 8. (a) Attenuation factor for  $I=1$  plotted as a function of the magnetic anisotropy  $B/A$  for different lifetimes of the intermediate state. (b) Eigenvalue spectrum as a function of  $B/A$ .

vanishing interference effects other than those which arise from the fact that  $G$  is zero.

We state the result obtained from the exact formula (59) for the model case of a cascade with  $I=1$ . The complete correlation in this case is given by the expression<sup>27</sup>

$$W(\theta) = 1 + G_2 A_2 P_2(\cos\theta),$$

where

$$G_2 = \frac{1}{5} \left\{ 1 - 3 \frac{a_{1/2}^2}{(1+a_{1/2}^2)^2} + \left[ 1 + 3 \frac{a_{1/2}^2}{(1+a_{1/2}^2)^2} \right] \frac{\Gamma^2}{\Gamma^2 + D_{1/2}^2} + \frac{1 + 2a_{1/2}^2}{1 + a_{1/2}^2} \frac{\Gamma^2}{\Gamma^2 + \frac{1}{4}[\frac{3}{2}A + P + D_{1/2}]^2} + \frac{2 + a_{1/2}^2}{1 + a_{1/2}^2} \frac{\Gamma^2}{\Gamma^2 + \frac{1}{4}[\frac{3}{2}A + P - D_{1/2}]^2} \right\}. \quad (61)$$

Figure 8 shows the attenuation factor  $G_2$ , given by (61) plotted as a function of  $B$  for a fixed value of  $A$  ( $P=0$ ) and different lifetimes. A large peak in  $G_2$  occurs at  $A=B$ . The behavior of the attenuation factor can be qualitatively understood on the basis of the energy level scheme (see Fig. 8). We consider first the case where  $\Gamma$  is much smaller than  $A$ . For an isotropic hyperfine interaction ( $B=A$ ) the attenuation factor is given in the limit of small  $\Gamma$  by the "hard-core value"<sup>6</sup>

$$(G_k)_{\min} = 1 - k(k+1)/(2I+1)^2.$$

On the other hand, the attenuation factor is given by

<sup>27</sup> The attenuation factors for  $I=2$  have also been computed.

the strong perturbation value  $G_2^0$  in the region of  $B$  where  $|E_{\pm 3/2} - E_{\pm 1/2+}| \gg \Gamma$ . If  $B$  approaches  $A$ , the interference effects due to the crossing ( $\pm 1/2+$ ;  $\pm 3/2$ ) become important and give rise to the sharp peak at  $B=A$ . It is obvious that in this case a very small deviation from the isotropy of the magnetic interaction can result in a large change in the attenuation factor.

In the case of a shorter lifetime, for which  $\Gamma \gtrsim A$ , interference effects involving the states  $|\pm 1/2-\rangle$  have to be included in  $G_2$ . In the limit  $\Gamma \gg A$  the attenuation factor is identically one and the correlation is unperturbed.

## VI. CONCLUSIONS

Before discussing possible applications connected with the results of the present paper, it is useful to summarize the assumptions made in treating the correlation problem. The assumptions can be stated as follows:

(a) The energy substates in the initial state are assumed to be equally populated. This condition is fulfilled for temperatures as low as the temperatures in the liquid-helium range if the system is in thermal equilibrium with its surroundings.

(b) The interaction between the nucleus and the electron shell in the intermediate state is assumed to be time independent. This implies that, during the lifetime of the intermediate state, there are no major rearrangements in the electron shell which result in appreciable changes in the electric and magnetic fields near the nucleus.<sup>28</sup>

(c) The angular correlation is assumed to be unaffected by the recoil motion associated with the emission of the first radiation.<sup>29</sup>

The investigation of nuclear moments of excited states by study of angular correlations perturbed by the hyperfine interaction has to proceed in two steps. In the first step the measured perturbation of the correlation is compared with the theoretically computed perturbation which involves the hyperfine constants as parameters. The comparison between experiment and theory should provide a determination of the hyperfine constants. In the second step the nuclear moment of the intermediate state is deduced from the hyperfine constants. In order that the latter step be possible, sufficient knowledge of the wave function of the electronic state must be available. At present this knowledge is available to a large extent for paramagnetic crystals of the rare-earth group, which have

<sup>28</sup> Such effects are to be expected if the initial state of the gamma-ray cascade is excited by an electron capture process. Certainly, in a resonance fluorescence experiment, no such effects are possible.

<sup>29</sup> There are no known experimental results for gamma-gamma angular correlation or resonance fluorescence experiments which indicate that the correlations are affected by the recoil motion.

been investigated mainly by means of paramagnetic resonance experiments.<sup>2,30,31</sup>

The results of the present paper offer interesting possibilities for determination of the hyperfine constants (step one). In cases where paramagnetic resonance data exist for a particular rare-earth crystal, direct experimental values of  $g_{11}$  and  $A/B$  are usually known for the ionic ground state. (The  $P$  term is generally small.) If for such a crystal an angular correlation experiment or a resonance fluorescence experiment can be carried out with the ion in the ground state, the known values of  $g_{11}$  and  $A/B$  greatly facilitate the determination of a magnetic hyperfine constant. The value of  $A$  can, in fact, be deduced directly for known  $A/B$  and  $g_{11}$  from an angular correlation resonance experiment in which the critical magnetic field is measured [see, e.g., Eq. (52)]. This method requires neither the knowledge of the unperturbed correlation nor the value of the intermediate state lifetime.<sup>32</sup>

An alternative determination of a magnetic hyperfine constant can be based on the decoupling experiment (Sec. IV-A). From the knowledge of the ratio  $A/B$  and the lifetime of the intermediate state, the attenuation factor  $\bar{G}_K$  can be plotted as a function of  $G$ , in the manner of Fig. 2. A value of  $\bar{G}_K$  measured for a known decoupling field strength  $H$  corresponds to a value  $G/A = g_{11}\mu_B H/A$ , from which the hyperfine constant  $A$  can be extracted. In the strong perturbation limit the knowledge of the lifetime is not required.

It is interesting to note that the occurrence of sharp resonance lines is not restricted to the case of an anisotropic magnetic hyperfine interaction. The result in the final paragraph of Sec. IV C shows that an

isotropic hyperfine interaction which is realized for instance in a gaseous source can produce interference peaks in the angular correlation. Thus, a correlation experiment with a gaseous source would provide an alternative to a correlation experiment using single-crystal sources and low temperatures. The former experiment could be used to obtain information about nuclear moments for paramagnetic atoms for which the electronic wave functions are sufficiently well known. In this paper only electronic doublet states have been considered and the results are, therefore, valid only for an electronic spin of  $\frac{1}{2}$ . The calculation of a perturbed angular correlation for larger electronic spin values would be very complicated. However, if one is interested only in the critical magnetic fields for an angular correlation resonance experiment with a gaseous source, it is sufficient to inspect the energy spectrum of the hyperfine interaction and to compute the critical field values for which the energy levels of interfering states cross. The critical fields are only functions of two hyperfine constants  $A$  and  $P$ . (The electronic  $g$  factor is usually known.) Thus, from a measurement of the critical magnetic fields the hyperfine constants could be extracted. With the external field chosen parallel to the detector plane, the interference term has the energy and angular dependence of  $W_R(\Phi)$  in Eq. (49), and the selection rules derived from Eq. (49) are valid for arbitrary electronic spin  $J$ .

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<sup>30</sup> K. D. Bowers and J. Owen, *Reports on Progress in Physics* (The Physical Society, London, 1955), Vol. 18, p. 304.

<sup>31</sup> I. Lindgren, *Nucl. Phys.* **32**, 151 (1961).

<sup>32</sup> The lifetime could be obtained from the measured width of the resonance peak. [See Eqs. (53), (54), and (56).]