Influence of a Combined Magnetic Dipole and Electric Quadrupole Interaction on Angular Correlations*†

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The general theory of perturbed angular correlations has been applied to the special case of a static combined magnetic dipole and electric quadrupole interaction of comparable strength. Particular emphasis is given to the important case of an external magnetic field applied to a source with randomly oriented electrostatic gradients (polycrystalline source). Expressions for the differential and integral attenuation factors are derived as a function of the ratio $y=\omega_H/\omega_E$ of the magnetic (ω_H) and the electric interaction frequencies (ω_E), and for several values of the electric interaction parameter $x=\omega_E t$ and $x=\omega_E\tau$, respectively. Numerical values for the attenuation factors are computed for the spins 1, 3/2, 2, and 5/2 for 30 values of y and 24 values of the parameter x. The results are displayed in form of representative curves. As an example of the application of the general results, angular correlation functions are calculated for the two cases of a magnetic field perpendicular to the detector plane and parallel to the emission direction of one of the observed nuclear radiations.

I. INTRODUCTION

ANGULAR correlations of successively emitted radiations involving an intermediate nuclear state of lifetime τ may be influenced by extranuclear fields. The theory of angular correlations perturbed by an interaction of the nuclear moments with such fields has been treated by several authors.[1-3] The results of the theoretical and experimental investigations obtained so far are described in some review articles[4-7] which present a good survey of the present situation concerning perturbed angular correlations.

The theory of the influence of an external magnetic field on angular correlations has been treated by Alder.[1] The perturbation of a correlation by virtue of a static electric quadrupole interaction has been calculated by Alder *et al.*[2] for a polycrystalline source and for single crystals with different orientations relative to the plane of the detectors. In this paper, the authors also treated the special case of a combined magnetic and electric field directed parallel to each other. Abragam and Pound[3] have given a more general reformulation of the perturbation problem in angular correlations and presented a more detailed discussion of the electric quadrupole interaction in liquids and in polycrystalline sources. These authors also estimated the influence of an applied magnetic field in the presence of an internal static electric quadrupole interaction in a polycrystalline source for the two limiting cases $\omega_H \gg \omega_E$ and $\omega_E \gg \omega_H$, where ω_H and ω_E are the magnetic and electric interaction frequencies, respectively. The general case of a static combined electric and magnetic interaction of comparable strength, however, has not been treated as yet.

The magnetic dipole interaction has been used in many experiments for the determination of a large number of magnetic moments of excited nuclear states. With the application of refined measuring techniques, this method has become very powerful.[8] In all those cases where the mean life of the excited state is sufficiently long to influence the angular correlation by an external magnetic field, also a perturbation due to the electric quadrupole interaction must be expected. In most cases, the presence of the quadrupole interaction interferes with the determination of magnetic moments and the quantitative knowledge of the quadrupole interaction is required to obtain accurate values of the magnetic moments. In general, this involves a separate measurement of the electric quadrupole interaction to apply the necessary corrections. Quantitative investigations of the electric quadrupole moment have been done in a few cases only. This is mainly due to the fact that only the product $Q\partial^2 V/\partial z^2$ of the nuclear quadrupole moment Q and of the gradient $\partial^2 V/\partial z^2$ can be

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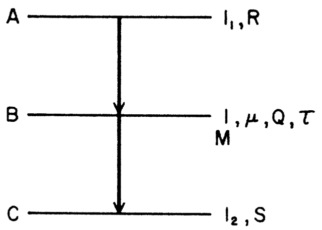


FIG. 1. Cascade of two nuclear radiations.

determined. The field gradient $\partial^2 V/\partial z^2$ is very difficult to compute and, in general, a precise determination of Q is almost impossible. On the other hand, there exists for some nuclei the possibility to eliminate the computation of the field gradient by measuring the ratio of the quadrupole moments of two different levels, assuming the gradient to be the same in both cases.

The effect of the quadrupole interaction can be kept small in many cases by the use of liquid sources, where the random motion of the field-producing ions causes a "smearing out" of the electric field gradients at the nucleus. The disadvantage of liquid sources, however, are their relatively small specific activities and the need for sometimes complicated chemical procedures. Higher specific activities can be obtained with powder sources. This may be of importance in measurements of magnetic moments of excited states, since smaller sized sources would allow to employ smaller pole gaps of the electromagnets which are used to produce the magnetic field at the nucleus to be investigated. Smaller pole gaps facilitate the production of higher magnetic fields. The use of solid polycrystalline sources is also anticipated in connection with small superconducting magnets.

Polycrystalline sources were rarely used for magnetic moment measurements for the simple reason that no theoretical calculations were available which would permit an interpretation of the experimental results. It is the purpose of this paper to present the theory and numerical results for the general case of a static combined magnetic dipole and electric quadrupole interaction of comparable strength and with randomly oriented axes of the electrostatic field gradients.[9] The results suggest a simultaneous measurement of the magnetic and the electric interaction frequencies by using polycrystalline sources in an external magnetic field. The theory includes the case of an electrostatic gradient axis at an arbitrary constant angle β with respect to an applied magnetic field (single crystal source). In view of space limitations, however, the very extensive numerical results for this single-crystal case are not included.

The results for the "polycrystalline" case are presented in such a way that there is no restriction concerning the direction of the magnetic field with respect to the detector plane. By chosing the angles θ and φ in the spherical harmonics corresponding to the geometrical arrangement the perturbed angular correlation can be calculated for an arbitrary direction of the magnetic field with respect to the detector plane. As an example, the correlation functions are calculated for the two most important cases, (1) a magnetic field perpendicular to the detector plane, and (2) a magnetic field parallel to the emission direction of a detected radiation.

Numerical results for the attenuation factors are given in form of curves for the nuclear spin values $I=1$, $3/2$, 2, and $5/2$ as a function of the interaction ratio $y=\omega_H/\omega_E$ and for various values of the electric interaction strength. Detailed tables with the numerical values will be made available in an AEC report.[10] Calculations of the attenuation factors for higher spin values are in progress.

II. THEORY

1. General

In the following, a cascade of nuclear radiations R_1 and R_2 is considered, involving the states A, B, C as the initial, intermediate, and final state with the spin and magnetic quantum numbers I_1R, IM, and I_2S, respectively (Fig. 1). The states A and C are assumed to be isotropic. The emission of the cascade radiations R_1 and R_2 is described by the Hamiltonians H_1 and H_2, respectively. If no external interactions are present during the lifetime τ of the intermediate state B, the unperturbed angular correlation can be represented in the form

$$W(\mathbf{\Omega}_1,\mathbf{\Omega}_2)=\sum_{RSMM'}(A_R|H_1|B_M)(B_M|H_2|C_S)\times(A_R|H_1|B_{M'})^*(B_{M'}|H_2|C_S)^*. \quad (1)$$

The state vectors $|B_M)$ form a complete and orthonormal set in the intermediate state B. If the states $|B_M(0))$ change by virtue of an interaction of the nuclear moments μ and Q with extranuclear fields during the time t ($t=0$ is defined by the emission time of R_1) to $|B_M(t))$,

$$|B_M(t=0))\xrightarrow{\text{interaction}}|B_M(t)),$$

then the perturbed angular correlation of R_1 and R_2 (emitted after the time t) is given by

$$W(\mathbf{\Omega}_1,\mathbf{\Omega}_2)=\sum_{RSMM'}(A_R|H_1|B_M(0))(B_M(t)|H_2|C_S)\times(A_R|H_1|B_{M'}(0))(B_{M'}(t)|H_2|C_S)^*. \quad (2)$$

In the case of static interactions, the evolution of the time-dependent state vectors $|B_M(t))$ can be repre-

[9] The authors were informed that the same problem is under investigation by H. Paul and W. Brunner, Ann. Physik 9, 316 and 323 (1962).

[10] R. M. Steffen, E. Matthias, and W. Schneider, Atomic Energy Commission, Division of Technical Information Reports No. 17089, Part 1 ($I=1$ and 2) and No. 17089, Part 2 ($I=3/2$ and 5/2) (unpublished).

sented as a linear combination of the state vectors $|B_{M''}(0))$ at time $t=0$.[3]

$$|B_M(t))=\sum_{M''}\Lambda_{MM''}(t)|B_{M''}(0)). \tag{3}$$

As long as classical fields are considered, the indices M and M'' in (3) are the projection quantum numbers of the nuclear spin I along a certain fixed axis Oz. The time dependence is described by the coefficients $\Lambda_{MM''}(t)$ which are the matrix elements of the evolution operator

$$\Lambda(t)=\exp[-(i/\hbar)\mathbf{H}t], \tag{4}$$

where $\mathbf{H}$ is the interaction Hamiltonian of the *static perturbation*. Combining (2) and (3) one gets for the angular correlation

$$W(\mathbf{\Omega}_1,\mathbf{\Omega}_2)=\sum_{\substack{RSMM''\\M'M'''}}(A_R|H_1|B_M)\Lambda_{MM''}^*(B_{M''}|H_2|C_S)$$
$$\times(A_R|H_1|B_{M'})^*\Lambda_{M'M'''}(B_{M'''}|H_2|C_S)^*, \tag{5}$$

with

$$\Lambda_{ik}=(B_i|\Lambda|B_k). \tag{6}$$

The matrix elements $\Lambda_{M'M'''}$ and $\Lambda_{MM''}^*$ can now be expressed in terms of the eigenvectors and the eigenvalues of the interaction Hamiltonian $\mathbf{H}$: The unitary matrix, which diagonalizes $\mathbf{H}$, is denoted by U,

$$U\mathbf{H}U^{-1}=E, \tag{7}$$

where E stands for the (diagonal) energy matrix. It can be shown by expansion of the e function that

$$U\exp[-(i/\hbar)\mathbf{H}t]U^{-1}=\exp[-(i/\hbar)Et]. \tag{8}$$

It follows from Eq. (4) that

$$\Lambda=U^{-1}\exp[-(i/\hbar)Et]U. \tag{9}$$

The matrix elements of Λ are given by

$$\begin{aligned}\Lambda_{M'M'''}&=\sum_{n'}u_{n'M'}^*e^{-(i/\hbar)E_{n'}t}u_{n'M'''},\\ \Lambda_{MM''}^*&=\sum_{n}u_{nM}e^{(i/\hbar)E_nt}u_{nM''}^*.\end{aligned} \tag{10}$$

The u_{ik} and the E_n are elements of the unitary matrix U and the diagonal matrix E, respectively.

By application of the methods of Racah algebra,[1] Eq. (5) can be written in the following way:

$$W(\mathbf{\Omega}_1,\mathbf{\Omega}_2)=\sum_{\substack{k_1k_2\\\mu_1\mu_2}}A_{k_1}A_{k_2}$$
$$\times\mathrm{III}_{k_1k_2}{}^{\mu_1\mu_2}Y_{k_1}{}^{\mu_1}(\mathbf{\Omega}_1)Y_{k_2}{}^{\mu_2*}(\mathbf{\Omega}_2), \tag{11}$$

where the average over the two polarization directions of the γ quanta has been taken. The factors A_{k_1} and A_{k_2} describe the radiations R_1 and R_2, respectively, and are defined by Alder.[1] For pure γ-multipole radiation with multipolarity L_1 and L_2, they are given by the F coefficients

$$A_{k_1}=F_{k_1}(L_1L_1I_1I),$$
$$A_{k_2}=F_{k_2}(L_2L_2I_2I),$$

which are tabulated by Ferentz and Rosenzweig.[11] If one or both of the radiations are mixtures of different multipoles L_ν and L_ν', one has to substitute for A_{k_ν}

$$A_{k_\nu}=[1/(1+\delta_\nu^2)]\{F_{k_\nu}(L_\nu L_\nu I_\nu I)+2\delta_\nu F_{k_\nu}(L_\nu L_\nu'I_\nu I)+\delta_\nu^2F_{k_\nu}(L_\nu'L_\nu'I_\nu I)\}.$$

The influence of a perturbing interaction between the nuclear moments and extranuclear fields is described by the factor

$$\mathrm{III}_{k_1k_2}{}^{\mu_1\mu_2}=\sum_{MM''}(Ik_1M'\mu_1|IM)(Ik_2M'''\mu_2|IM'')\times\Lambda_{MM''}^*\Lambda_{M'M'''}. \tag{12}$$

In view of the properties of the Clebsch-Gordan coefficients, the indices M' and M''' are related to M and M'', respectively, by $M=M'+\mu_1$ and $M''=M'''+\mu_2$, i.e., the summation over M' and M''' can be omitted.

If there is no perturbation present in the intermediate state, the evolution matrix reduces to the unit matrix $\Lambda(0)=1$, and consequently, $M=M''$ and $M'=M'''$ which gives $\mu_1=\mu_2=\mu$. By virtue of the properties of the Clebsch-Gordan coefficients, one obtains $k_1=k_2=k$ and the factor $\mathrm{III}_{k_1k_2}{}^{\mu_1\mu_2}$ takes the form

$$\mathrm{III}_{k_1k_2}{}^{\mu\mu}=[(2I+1)/(2k_1+1)]\delta_{k_1k_2}. \tag{13}$$

Introducing this result into Eq. (11) and using the addition theorem of spherical harmonics, one gets the expression for an unperturbed angular correlation

$$W(\mathbf{\Omega}_1,\mathbf{\Omega}_2)=W(\theta)=\sum_{k\text{ even}}A_k(R_1)A_k(R_2)P_k[\cos(\mathbf{\Omega}_1,\mathbf{\Omega}_2)]. \tag{14}$$

For a nonvanishing perturbation, the general expression for the attenuation factor $\mathrm{III}_{k_1k_2}{}^{\mu_1\mu_2}$ is obtained by combining Eqs. (10) and (12)

$$\begin{aligned}\mathrm{III}_{k_1k_2}{}^{\mu_1\mu_2}(\beta\gamma t)&=\sum_{\substack{MM''\\nn'}}(Ik_1M'\mu_1|IM)(Ik_2M'''\mu_2|IM'')\\&\quad\times u_{nM}(\beta\gamma)u_{nM''}^*(\beta\gamma)u_{n'M'''}(\beta\gamma)\\&\quad\times u_{n'M'}^*(\beta\gamma)e^{(i/\hbar)(E_n-E_{n'})t}.\end{aligned} \tag{15}$$

This formula is identical with Eq. (14) of Abragam and Pound[3] and is valid for all types of static perturbations. In order to investigate the influence of a particular perturbing interaction on the angular correlation, it is necessary to calculate the eigenvalues E_n and the eigenvectors u_{ik} of the interaction Hamiltonian.

[11] M. Ferentz and N. Rosenzweig, Atomic Energy Commission Report ANL-5324, 1955 (unpublished).

2. Simultaneous Static Electric Quadrupole and Magnetic Dipole Interaction

The electric field gradients are assumed to be of static nature and randomly oriented in space, while the magnetic field is considered to point in a fixed direction. Experimentally, this case can be realized by using a polycrystalline source placed in an external magnetic field for the angular correlation measurements.

The direction of the magnetic field is chosen parallel to the z axis of a fixed coordinate system S, i.e., the magnetic part of the interaction Hamiltonian is diagonal in S. Furthermore, let us define a coordinate system S' fixed to the microcrystals, which can be transformed to S by the rotation group $D(0,\beta,\gamma)$ (cf. Fig. 2. The angles in this figure should be labeled $2\pi-\beta$ and $2\pi-\gamma$.). Assuming axial symmetry of the electrostatic field gradients with respect to the z' axis of the S' systems, the components of the field gradient in one microcrystal are completely determined by $\partial^2V'/\partial z'^2$. In addition, it is assumed that the total interaction Hamiltonian can be represented in the form

$$\mathbf{H}=\mathbf{H}_{\text{magn}}+\mathbf{H}_{\text{el}}, \tag{16}$$

i.e., the following calculations are only valid for those cases where the presence of the magnetic field does not alter the electric field gradient.

As shown in detail in a previous paper,[12] the matrix elements of the total interaction Hamiltonian for this problem are given by

$$\mathbf{H}_{m,m'}=\hbar\omega_E\Bigg\{-ym\delta_{mm'}+(\pi/5)^{1/2}(-1)^{I-m} \times[(2I+3)(2I+2)(2I+1)2I(2I-1)]^{1/2} \times\begin{pmatrix} I & 2 & I \\ -m & m-m' & m' \end{pmatrix} Y_2^{m'-m}(\beta,\gamma)\Bigg\}. \tag{17}$$

The quadrupole interaction frequency ω_E is here defined as[13]

$$\omega_E=\frac{1}{\hbar}eQ\frac{\partial^2V'}{\partial z'^2}\frac{1}{4I(2I-1)}, \tag{18}$$

where $\partial^2V'/\partial z'^2$ describes the axially symmetric electrostatic gradient in an individual microcrystal. The interaction ratio y is defined as[14]

$$y=\omega_H/\omega_E, \tag{19}$$

where ω_H is the magnetic interaction frequency (Larmor frequency)

$$\omega_H=\mu H_0/\hbar I. \tag{20}$$

As was discussed elsewhere,[12] the assumption of axial symmetry leads to the conclusion that the eigenvalues of $\mathbf{H}(\beta,\gamma)$ are independent of γ. By virtue of the fact that the angle γ occurs in the matrix elements only in the form $e^{i\gamma(m'-m)}$, $\mathbf{H}(\beta,\gamma)$ and $\mathbf{H}(\beta,0)$ are connected through a unitary transformation $S(\gamma)$,

$$S(\gamma)\mathbf{H}(\beta,\gamma)S^{-1}(\gamma)=\mathbf{H}(\beta,0),$$

where

$$S(\gamma)=\begin{bmatrix} e^{iI\gamma} & & & & 0 \\ & \cdot & & & \\ & & e^{iM\gamma} & & \\ & & & \cdot & \\ 0 & & & & e^{-iI\gamma} \end{bmatrix}. \tag{21}$$

The unitary matrix $U(\beta,\gamma)$ which diagonalizes $\mathbf{H}(\beta,\gamma)$ is then given by

$$U(\beta,\gamma)=U(\beta,0)S(\gamma). \tag{22}$$

It follows that

$$U(\beta,\gamma)\mathbf{H}(\beta,\gamma)U^{-1}(\beta,\gamma)=U(\beta,0)\mathbf{H}(\beta,0)U^{-1}(\beta,0),$$

which shows that the eigenvalues $E(\beta)$ of $\mathbf{H}(\beta,\gamma)$ are independent of γ. Furthermore, since $\mathbf{H}(\beta,0)$ is a real matrix, the unitary matrix $U(\beta,0)$ is also real. The eigenvectors of $\mathbf{H}(\beta,0)$ are denoted by $u_n(\beta)$.

It is convenient to introduce the matrix

$$K(\beta,\gamma)=(1/\omega_E\hbar)\mathbf{H}(\beta,\gamma), \tag{23}$$

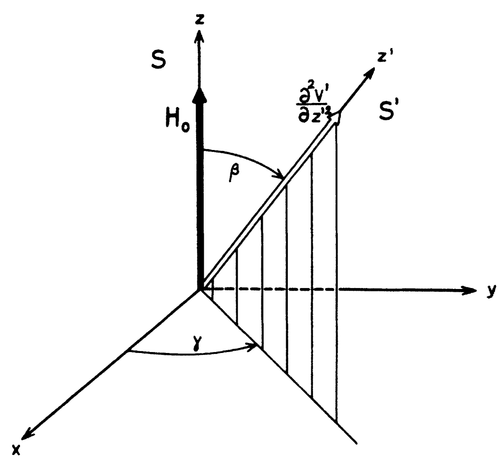


FIG. 2. Extranuclear fields in one microcrystal.

[12] E. Matthias, W. Schneider, and R. M. Steffen, Phys. Rev. **125**, 261 (1962). (Note the misprint on p. 265 of this paper. Sentence after Eq. (29) should read: Since the magnetic field is axially symmetric, the angle γ occurs in $H_{m,m'}$ in the form $\exp[i\gamma(m'-m)]$.)

[13] Note that our definition of ω_E is somewhat different from the conventional definition of the quadrupole interaction frequency ω_Q. Our ω_E is 1/3 of the conventional ω_Q for integer I, and 1/6 of the conventional ω_Q for half-integer I. The definition [Eq. (18)] was adopted because it is independent of the character of I.

[14] Equal strength of magnetic and electric interaction in the conventional definition of the electric quadrupole interaction frequency is represented in our notation by $y=3$ for integer I and by $y=6$ for half-integer I.

which depends on the interaction ratio y and β and γ only. The eigenvalues of $K(\beta,\gamma)$ are $D(\beta)=E(\beta)/\omega_E\hbar$, and the eigenvectors of $K(\beta,\gamma)$ are the same as those of $\mathbf{H}(\beta,\gamma)$, namely, according to Eqs. (21) and (22)

$$u_{nM}(\beta,\gamma)=u_{nM}(\beta)e^{iM\gamma}.$$

Substituting this expression into Eq. (10), we obtain for the time evolution matrix

$$\Lambda_{M'M'''}=\sum_{n'} u_{n'M'}^*(\beta)e^{-iM'\gamma}e^{-iD_{n'}(\beta)x}u_{n'M'''}(\beta)e^{iM'''\gamma},$$

$$\Lambda_{MM''}^*=\sum_{n} u_{nM}(\beta)e^{iM\gamma}e^{iD_n(\beta)x}u_{nM''}^*(\beta)e^{-iM''\gamma} \tag{24}$$

where we have introduced the interaction parameter

$$x=\omega_E t. \tag{25}$$

The time t is the time interval during which the nuclear state is exposed to the interacting fields. Inserting (24) into the general form of the attenuation factor (15), one obtains

$$\begin{aligned}\mathrm{III}_{k_1k_2}{}^{\mu_1\mu_2}(\beta,\gamma) &= \sum_{\substack{MM''\\nn'}} (Ik_1M'\mu_1|IM)(Ik_2M'''\mu_2|IM'')\\ &\quad\times u_{nM}(\beta)u_{nM''}^*(\beta)u_{n'M'''}(\beta)u_{n'M'}^*(\beta)\\ &\quad\times e^{i(D_n-D_{n'})x}e^{i\gamma(M-M'-M''+M''')}.\end{aligned} \tag{26}$$

Because we are interested in the perturbation in polycrystalline sources, it is necessary to average over the Euler angles β and γ. If one averages (26) over γ from 0 to 2π only those terms give a contribution for which $M-M'=M''-M'''$; this implies that only attenuation factors with $\mu_1=\mu_2=\mu$ occur.

$$\begin{aligned}\langle\mathrm{III}_{k_1k_2}{}^{\mu_1\mu_2}(\beta)\rangle_\gamma &= \sum_{\substack{MM''\\nn'}} (Ik_1M'\mu|IM)(Ik_2M'''\mu|IM'')\\ &\quad\times u_{nM}(\beta)u_{nM''}^*(\beta)u_{n'M'''}(\beta)u_{n'M'}^*(\beta)\\ &\quad\times e^{i(D_n-D_{n'})\cdot x}.\end{aligned} \tag{27}$$

Because of the reality of the eigenvectors the asterisks in (27) can be dropped.[15]

It follows from Eq. (26) that a combined magnetic and electric perturbation causes the occurrence of interference terms with $k_1\neq k_2$. The interference terms $\mathrm{III}_{k_1k_2}{}^{\mu_1\mu_2}(\beta,\gamma)(k_1\neq k_2)$ have the following symmetry properties. By exchanging all the indices k, μ, and M it follows directly from Eq. (26) that

$$\mathrm{III}_{k_1k_2}{}^{\mu_1\mu_2}(\beta,-\gamma)=\mathrm{III}_{k_2k_1}{}^{\mu_2\mu_1}(\beta,\gamma),$$

which implies that

$$\langle\mathrm{III}_{k_1k_2}{}^{\mu_1\mu_2}(\beta)\rangle_\gamma=\langle\mathrm{III}_{k_2k_1}{}^{\mu_2\mu_1}(\beta)\rangle_\gamma. \tag{28}$$

Furthermore, the interference terms vanish if one of the k's (either k_1 or k_2) is zero. This can be proved in the following way:

Due to the property of the Clebsch-Gordan coefficients

$$(I0M'0|IM)=\delta_{MM'},$$

the attenuation factor in (26) has the form

$$\begin{aligned}\mathrm{III}_{0k_2}{}^{0\mu_2}(\beta,\gamma) &= \sum_{M''nn'} (Ik_2M'''\mu_2|IM'')u_{n'M'''}(\beta)u_{nM''}(\beta)\\ &\quad\times e^{i(D_n-D_{n'})\cdot x}e^{i\gamma(M'''-M'')}\sum_M u_{nM}(\beta)u_{n'M}(\beta).\end{aligned} \tag{29}$$

The unitarity of the matrix U gives

$$\textstyle\sum_M u_{nM}u_{n'M}=\delta_{nn'}. \tag{30}$$

Using this argument twice and remembering that also $Ue^{i\gamma M}$ is an unitary matrix if U is unitary, one obtains $n=n'$, $M''=M'''$ and thus

$$\mathrm{III}_{0k_2}{}^{00}(\beta,\gamma)=\sum_{M''} (Ik_2M''0|IM''). \tag{31}$$

From the properties of the Clebsch-Gordan coefficients the relation

$$\sum_{M''} (Ik_2M''0|IM'')(I0M''0|IM'')=\frac{2I+1}{(2k_2+1)^{1/2}}\delta_{k_20} \tag{32}$$

can be verified, which gives

$$\mathrm{III}_{0k_2}{}^{00}(\beta,\gamma)=\frac{2I+1}{(2k_2+1)^{1/2}}\delta_{k_20}. \tag{33}$$

This means that all interference terms with $k_1=0$, $k_2\neq 0$ or $k_1\neq 0$, $k_2=0$ vanish.

In an analogous way, it can be proved that the interference terms vanish for the unperturbed case. Then Eq. (26) gets the form

$$\begin{aligned}\mathrm{III}_{k_1k_2}{}^{\mu_1\mu_2}(\beta,\gamma,0) &=\textstyle\sum_{MM''}(Ik_1M'\mu_1|IM)(Ik_2M'''\mu_2|IM'')\\ &\quad\times e^{i\gamma(M-M'-M''+M''')}\textstyle\sum_n u_{nM''}(\beta)u_{nM}(\beta)\\ &\quad\times\textstyle\sum_{n'} u_{n'M'}(\beta)u_{n'M'''}(\beta)\\ &=\textstyle\sum_M(Ik_1M'\mu|IM)(Ik_2M'\mu|IM),\end{aligned}$$

which leads to

$$\mathrm{III}_{k_1k_2}{}^{\mu_1\mu_2}(\beta,\gamma,0)=\frac{2I+1}{(2k_1+1)^{1/2}(2k_2+1)^{1/2}}\delta_{k_1k_2}. \tag{34}$$

From the Eqs. (33) or (34) the normalization of the attenuation factor for $k_1=k_2=0$ can be found

$$\mathrm{III}_{00}{}^{00}=2I+1, \tag{35}$$

which also follows directly from Eq. (13).

[15] The common factor 2π arising from the integration over α and γ has been dropped.

Splitting up the attenuation factor (Eq. (26)) into a real and an imaginary part and integrating over β and γ one obtains

$$\langle \mathrm{III}_{k_1k_2}{}^{\mu_1\mu_2}(t)\rangle_{\beta,\gamma} = \int_0^\pi c_{k_1k_2}{}^{\mu(r)}(\beta,t)\sin\beta d\beta + i\int_0^\pi c_{k_1k_2}{}^{\mu(i)}(\beta,t)\sin\beta d\beta = a_{k_1k_2}{}^{\mu}(t)+ib_{k_1k_2}{}^{\mu}(t), \quad (36)$$

where (r) and (i) denote the real and imaginary part, respectively. In this formula the following abbreviations are introduced:

$$c_{k_1k_2}{}^{\mu(r)}(\beta,t)=\sum_{nn'} U_{k_1k_2}{}^{\mu nn'}(\beta) \times\cos\{[D_n(\beta)-D_{n'}(\beta)]\cdot x\},$$

$$c_{k_1k_2}{}^{\mu(i)}(\beta,t)=\sum_{nn'} U_{k_1k_2}{}^{\mu nn'}(\beta) \times\sin\{[D_n(\beta)-D_{n'}(\beta)]\cdot x\}, \quad (37)$$

$$U_{k_1k_2}{}^{\mu nn'}(\beta)=\sum_{MM''}(Ik_1M'\mu|IM)(Ik_2M'''\mu|IM'') \times u_{nM}(\beta)u_{nM''}(\beta)u_{n'M'}(\beta)u_{n'M'''}(\beta).$$

From Eqs. (36) and (37) the perturbed *differential* angular correlation can be obtained. To get the perturbation for the *integral* case, the expression

$$\langle \mathrm{III}_{k_1k_2}{}^{\mu_1\mu_2}\rangle_{\beta,\gamma,t}=\frac{1}{\tau}\int_0^\infty e^{-t/\tau}[a_{k_1k_2}{}^{\mu}(t)+ib_{k_1k_2}{}^{\mu}(t)]dt \quad (38)$$

must be evaluated, where τ is the mean life of the intermediate state B. This yields

$$\langle \mathrm{III}_{k_1k_2}{}^{\mu_1\mu_2}\rangle_{\beta,\gamma,t} = \int_0^\pi \hat{c}_{k_1k_2}{}^{\mu(r)}(\beta)\sin\beta d\beta + i\int_0^\pi \hat{c}_{k_1k_2}{}^{\mu(i)}(\beta)\sin\beta d\beta = \hat{a}_{k_1k_2}{}^{\mu}+i\hat{b}_{k_1k_2}{}^{\mu}, \quad (39)$$

with

$$\hat{c}_{k_1k_2}{}^{\mu(r)}(\beta)=\sum_{nn'} U_{k_1k_2}{}^{\mu nn'}(\beta)\frac{1}{1+\{[D_n(\beta)-D_{n'}(\beta)]\cdot x\}^2},$$

$$\hat{c}_{k_1k_2}{}^{\mu(i)}(\beta)=\sum_{nn'} U_{k_1k_2}{}^{\mu nn'}(\beta)\frac{[D_n(\beta)-D_{n'}(\beta)]\cdot x}{1+\{[D_n(\beta)-D_{n'}(\beta)]\cdot x\}^2}. \quad (40)$$

In view of Eqs. (35), (36), and (39), one finds for both the integral and the differential attenuation factors

$$\int_0^\pi \mathrm{III}_{00}{}^{00}\sin\beta d\beta=2(2I+1). \quad (41)$$

Also, with this normalization, the attenuation factors are for vanishing interaction ($\omega_E=0$ and $\omega_H=0$, or $t=0$ and $\tau=0$, respectively) given by [cf. Eq. (34)]

$$\langle \mathrm{III}_{k_1k_2}{}^{\mu_1\mu_2}(0)\rangle_{\beta,\gamma}=\langle \mathrm{III}_{k_1k_2}{}^{\mu_1\mu_2}(0)\rangle_{\beta,\gamma,t} =2(2I+1)/(2k+1), \quad (42)$$

where $k_1=k_2=k$.

According to Eqs. (11), (36), and (39) the perturbed *differential* correlation function may now be written in the final form

$$W(\mathbf{\Omega}_1,\mathbf{\Omega}_2)=\sum_{\substack{k_1k_2\\ \mu}} A_{k_1}(R_1)A_{k_2}(R_2) \times[a_{k_1k_2}{}^{\mu}(x,y)+ib_{k_1k_2}{}^{\mu}(x,y)] \times Y_{k_1}{}^{\mu}(\theta_1,\varphi_1)Y_{k_2}{}^{\mu*}(\theta_2,\varphi_2), \quad (43)$$

where $x=\omega_E t$. For the *time-integrated* angular correlation one has to replace the perturbation factors in (43) by $\hat{a}_{k_1k_2}{}^{\mu}(x,y)$ and $\hat{b}_{k_1k_2}{}^{\mu}(x,y)$, respectively, with $x=\omega_E\tau$.

The angular correlation function (43) refers to an arbitrary geometrical arrangement in which the external magnetic field may be chosen in any direction with respect to the plane of the detectors. The arguments θ and φ of the spherical harmonics specify the directions $\mathbf{\Omega}_1$ and $\mathbf{\Omega}_2$ in which the nuclear radiations are observed (cf. Fig. 14).

It can be proved that Eq. (43) reduces to the usual expression for a polycrystalline source if no magnetic interaction is present. For $y=0$, the interaction Hamiltonian (17) can be diagonalized by a unitary transformation with the rotation matrix $D^{(I)}(\boldsymbol{\omega})$:

$$(D^{(I)}(\boldsymbol{\omega})\mathbf{H}(y=0)D^{(I)-1}(\boldsymbol{\omega}))_{kl} =\sum_{mm'} D_{km}{}^{(I)}(\boldsymbol{\omega})\mathbf{H}_{mm'}(y=0)D_{lm'}{}^{(I)*}(\boldsymbol{\omega}). \quad (44)$$

Thus, the attenuation factor in (43) can be written as

$$a_{k_1k_2}{}^{\mu}(x,0)+ib_{k_1k_2}{}^{\mu}(x,0) = \sum_{\substack{nn'M\\ M'M''M'''}}(Ik_1M'\mu|IM)(Ik_2M'''\mu|IM'') \times D_{nM}{}^{(I)}(\boldsymbol{\omega})D_{n'M'}{}^{(I)*}(\boldsymbol{\omega})D_{n'M'''}{}^{(I)}(\boldsymbol{\omega}) \times D_{nM''}{}^{(I)*}(\boldsymbol{\omega})e^{(i/\hbar)(E_n-E_{n'})\cdot t}. \quad (45)$$

Because of the properties of the rotation groups,[16] one can reduce the product of four matrix elements to a product of two. Applying the orthogonality relations of the rotation group and of the 3-j symbols, Eq. (45) becomes

$$a_{kk}{}^{\mu}(x,0)+ib_{kk}{}^{\mu}(x,0) =\frac{2(2I+1)}{2k+1}\sum_{nn'}\begin{pmatrix} I & I & k\\ n' & -n & \kappa\end{pmatrix}^2 e^{(i/\hbar)(E_n-E_{n'})\cdot t}, \quad (46)$$

[16] A. R. Edmonds, *Angular Momentum in Quantum Mechanics* (Princeton University Press, Princeton, New Jersey, 1960).

TABLE I. Properties of the attenuation coefficients.

	$a_{k_1k_2}{}^{\mu}=a_{k_1k_2}{}^{-\mu}$	$b_{k_1k_2}{}^{\mu}=-b_{k_1k_2}{}^{-\mu}$	$a_{k_1k_2}{}^{\mu}=a_{k_1k_2}{}^{-\mu}$	$b_{k_1k_2}{}^{\mu}=-b_{k_1k_2}{}^{-\mu}$
$k_1=k_2=\mu=0$	$2(2I+1)$	0	$2(2I+1)$	0
Vanishing perturbation or $t=\tau=0$ for any $k_1=k_2=k$ and μ	$\dfrac{2(2I+1)}{2k+1}$	0	$\dfrac{2(2I+1)}{2k+1}$	0
Interference terms $k_1\neq k_2$ for vanishing perturbation or $t=\tau=0$	0	0	0	0

where $k=k_1=k_2$. The perturbation factor is no longer μ dependent and the summation over μ makes it possible to apply the addition theorem of spherical harmonics which gives

$$W(\mathbf{\Omega}_1,\mathbf{\Omega}_2)=\sum_k A_k(R_1)A_k(R_2)g_k(x)\times P_k[\cos(\mathbf{\Omega}_1,\mathbf{\Omega}_2)], \quad (47)$$

with

$$g_k(x)=2(2I+1)\sum_{nn'}\begin{pmatrix} I & I & k \\ n' & -n & \kappa \end{pmatrix}^2 e^{(i/\hbar)(E_n-E_{n'})\cdot t}. \quad (48)$$

This expression for $g_k(x)$ is identical with the attenuation factor for polycrystalline sources, given by Abragam and Pound[3] [formula (20′)] apart from the different normalization factor $2(2I+1)$.

It should be noted that the formalism used in this paper requires a derivation of the attenuation factor for the pure polycrystalline quadrupole interaction which differs from the usual way of treating this problem. There the electric field gradient axis chosen parallel to the quantization axis is kept fixed in space and one averages over all directions of $\mathbf{\Omega}_1$ and $\mathbf{\Omega}_2$.

The electric quadrupole interaction is degenerate with respect to its sign which implies that the imaginary part vanishes for $y=0$, and the perturbation factor is given by the $a_{kk}{}^{\mu}(x)$ only. In order to get the conventionally used attenuation factors $G_k(x)$ normalized to $G_k(x=0)=G_0=1$ from the $a_{kk}{}^{\mu}$, the following relation must be applied:

$$G_k(x)=[(2k+1)/2(2I+1)]a_{kk}{}^{0}(x). \quad (49)$$

The integral attenuation factor $\hat{G}_k(x)$ is obtained by taking the corresponding factors $\hat{a}_{kk}{}^{0}(x)$.

III. NUMERICAL COMPUTATIONS AND RESULTS

For the computation of the factors given in (36), (37), (39), and (40) a general program has been set up in the FORTRAN automatic coding system II. In one single run of this program, these factors are calculated for a specific nuclear spin value I and for a series of values of the parameters x and y. The computations include three main parts, namely,

(1) the diagonalization of K [Eq. (23)],
(2) the calculation of the factors given in (37) and (40),
(3) the calculation of the attenuation factors given in (36) and (39).

After a detailed examination of the different ordinary diagonalization methods, a modified version of the Jacobi method turned out to be the most suitable for the present case. For the calculations presented in this paper, the eigenvalues are computed to better than 10^{-6} and the corresponding eigenvectors to better than 10^{-4}.[12] A general subroutine for the calculation of Clebsch-Gordan coefficients for all spin values has been incorporated in the program. The computations were carried to a maximum value for k_1 and k_2 of 4. In view of the symmetry properties of the factor $\langle \mathrm{III}_{k_1k_2}{}^{\mu\mu}\rangle$ the range of the values k_1, k_2, μ, y, x, and β can be restricted as follows:

(a) Three combinations of k_1 and k_2 must be considered [cf. Eqs. (28) and (33)]:

$$k_1=k_2=2;\quad k_1=k_2=4;\quad k_1=2,\, k_2=4.$$

(b) From the properties of the Clebsch-Gordan coefficients it is obvious that the index μ in (27) is determined by

$$-\min(k_1,k_2)\leq\mu\leq\min(k_1,k_2).$$

However, one easily verifies that only terms with $\mu\geq 0$ have to be computed. Starting from Eq. (26) the following property of the attenuation factor can be derived:

$$\begin{aligned}\langle \mathrm{III}_{k_1k_2}{}^{\mu\mu}(t)\rangle_{\beta,\gamma}{}^*&=\langle \mathrm{III}_{k_1k_2}{}^{-\mu-\mu}(t)\rangle_{\beta,\gamma}\\ \langle \mathrm{III}_{k_1k_2}{}^{\mu\mu}\rangle_{\beta,\gamma,t}{}^*&=\langle \mathrm{III}_{k_1k_2}{}^{-\mu-\mu}\rangle_{\beta,\gamma,t}.\end{aligned} \quad (50)$$

This implies that the real and imaginary parts in Eq. (43) for $+\mu$ and $-\mu$ are related by

$$a_{k_1k_2}{}^{\mu}=a_{k_1k_2}{}^{-\mu},\quad b_{k_1k_2}{}^{\mu}=-b_{k_1k_2}{}^{-\mu}, \quad (51)$$

with the corresponding relations for the integral factors. Consequently the imaginary parts vanish for $\mu=0$. Table I summarizes some of the properties of the attenuation coefficients.

(c) Only positive values of x and y have to be considered. For the electric interaction the sign degeneracy of the sublevels m in $\mathbf{H}_{\mathrm{el}}$ makes the attenuation factors independent of the sign of ω_E. Thus, from investigations of a combined perturbation only the magnitude of the electric quadrupole interaction can be obtained. A

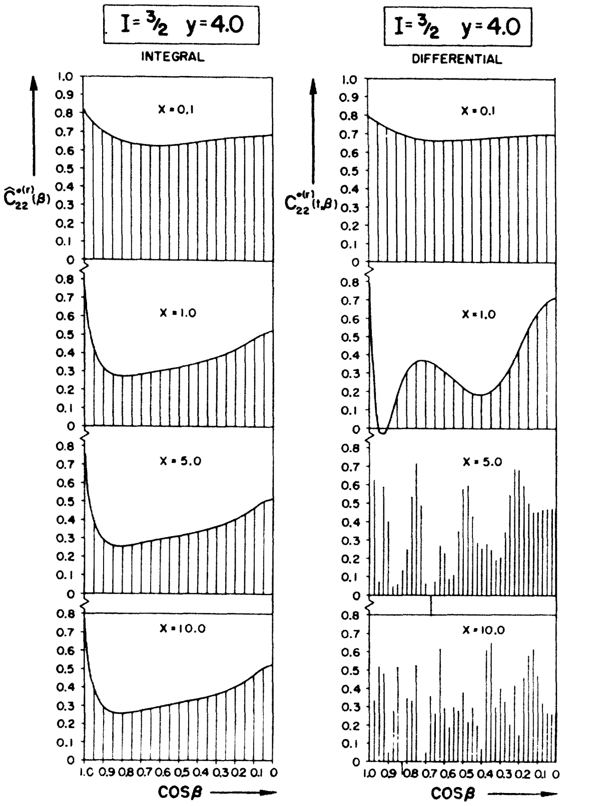


FIG. 3. The real parts $\hat{c}_{22}^{0(r)}(\beta)$ and $c_{22}^{0(r)}(t,\beta)$ of the attenuation factors for a single-crystal source oriented in various directions β relative to the magnetic field axis for some interaction parameters x. The vertical lines indicate the integration steps.

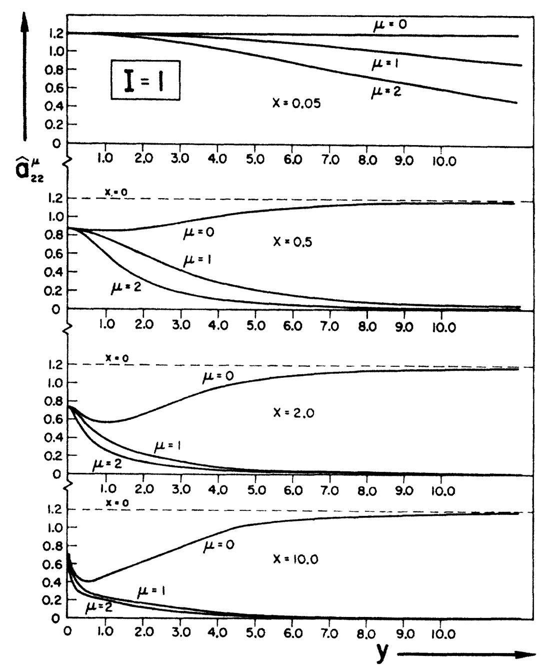


FIG. 4. The real part $\hat{a}_{22}^{\mu}(x,y)$ of the attenuation factors for the integral correlation with $I=1$, plotted as a function of $y=\omega_H/\omega_E$ for some selected values of the parameter $x=\omega_E\tau$.

change of the sign of ω_H, however, changes the sign of the imaginary parts $b_{k_1k_2}{}^{\mu}$, whereas the real parts $a_{k_1k_2}{}^{\mu}$ remain unaffected.

(d) As can be seen from formulas (17) and (23), the angular dependence of the matrix elements $K_{mm'}$ is determined by $\cos\beta$. Therefore, $\cos\beta$ is chosen as the

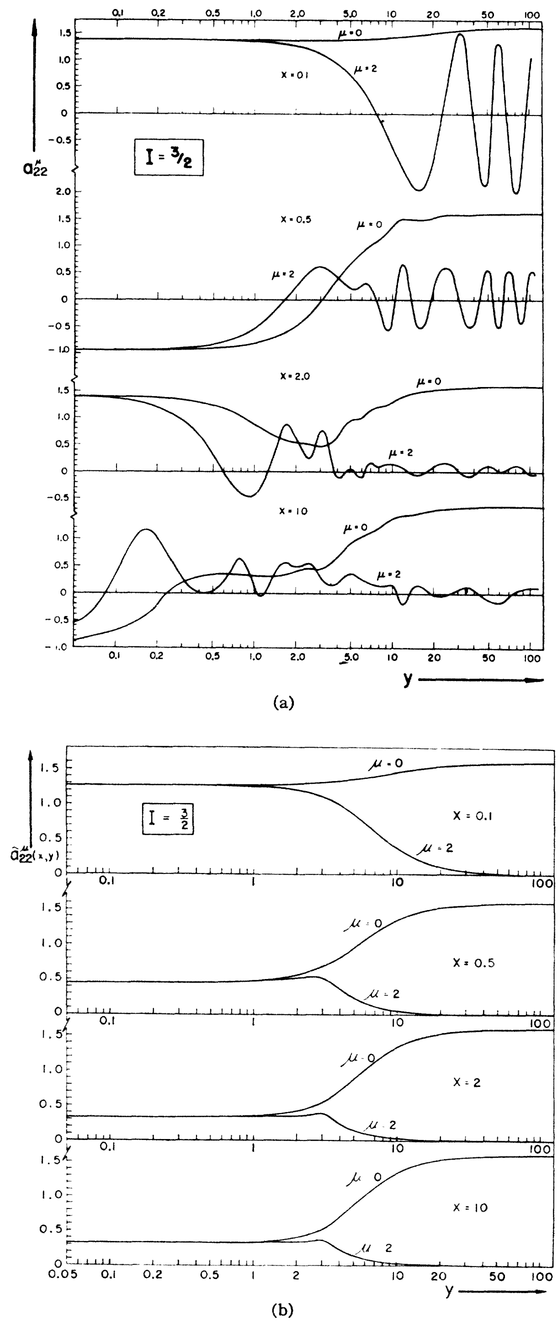


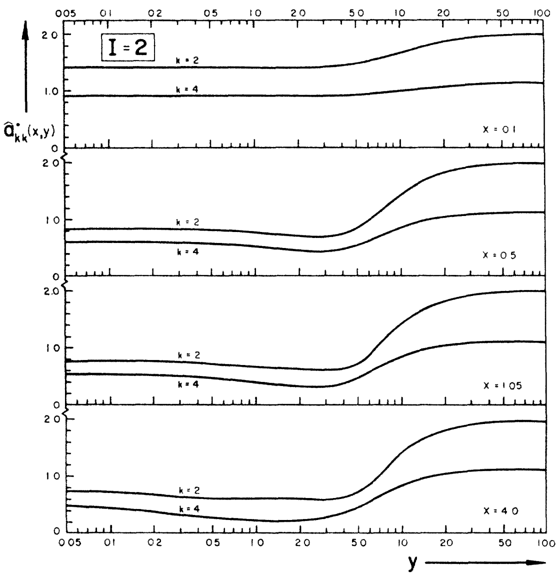
FIG. 5. (a) The real part $a_{22}^{\mu}(x,y)$ of the attenuation factors for the differential correlation with $I=3/2$, plotted as a function of $y=\omega_H/\omega_E$ for some selected values of the parameter $x=\omega_E t$. (b) The real part $\hat{a}_{22}^{\mu}(x,y)$ of the attenuation factors for the integral correlation with $I=3/2$, plotted as a function of $y=\omega_H/\omega_E$ for some selected values of the parameter $x=\omega_E\tau$.

variable for the integration over β. It can easily be verified that

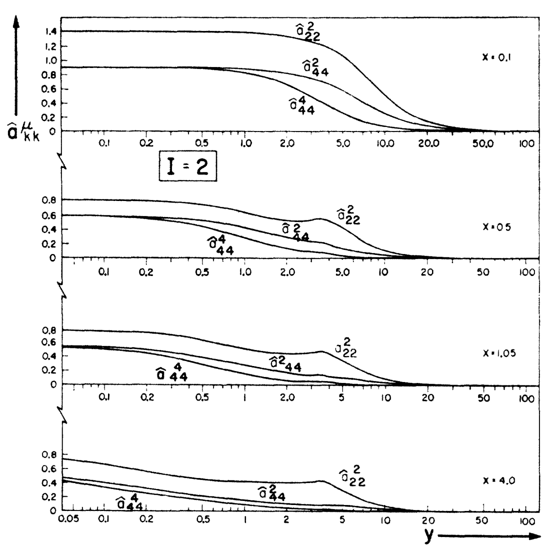
$$\int_0^{\pi} \langle \mathrm{III}_{k_1k_2}{}^{\mu\mu}(\beta)\rangle_\gamma d(\cos\beta) = 2\int_0^{\pi/2} \langle \mathrm{III}_{k_1k_2}{}^{\mu\mu}(\beta)\rangle_\gamma d(\cos\beta),$$

which reduces the required machine time by a factor of 2.

To cover a wide range of electric and magnetic interaction strengths, the attenuation factors have been computed for 30 y values between 0 and 100, and 24 x values covering the range from 0 to 5. From the experimental point of view it seems to be sufficient to tabulate the results only for x values up to 5. Tables of the attenuation factors will be available for the spin values 1, 3/2, 2, 5/2, and 7/2.[10] If special experimental conditions require numerical values which are not covered

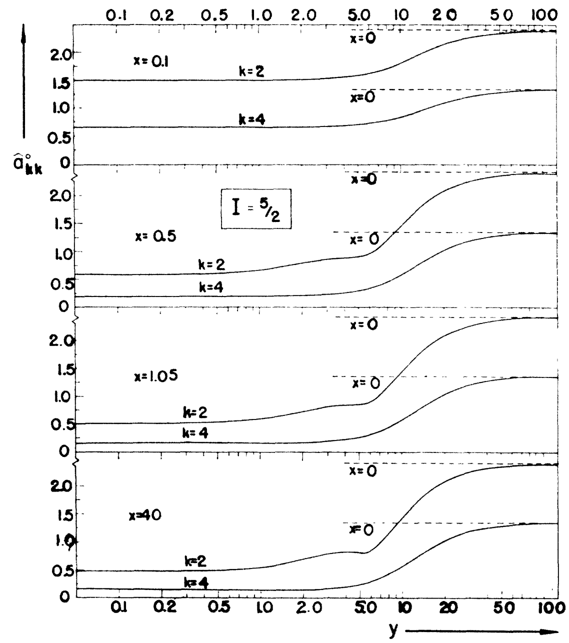


(a)

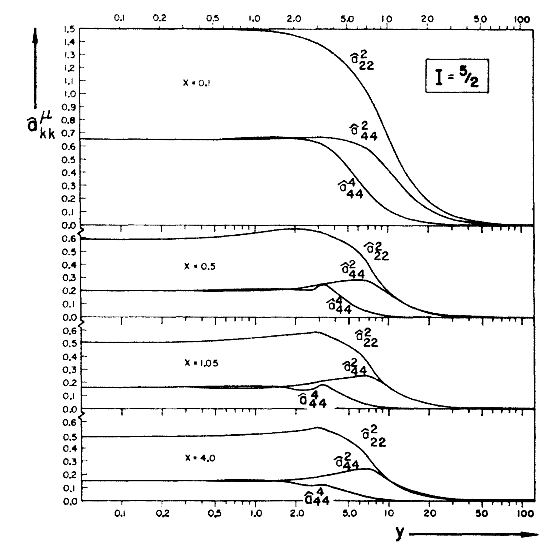


(b)

FIG. 6. (a) The real part $\hat{a}_{kk}{}^0(x,y)$ of the attenuation factors for the integral correlation with $I=2$, plotted as a function of $y=\omega_H/\omega_E$ for some selected values of the parameter $x=\omega_E\tau$. (b) The real part $\hat{a}_{kk}{}^\mu(x,y)$ of the attenuation factors for the integral correlation with $I=2$, plotted as a function of $y=\omega_H/\omega_E$ for some selected values of the parameter $x=\omega_E\tau$.



(a)



(b)

FIG. 7. (a) The real part $\hat{a}_{kk}{}^0(x,y)$ of the attenuation factors for the integral correlation with $I=5/2$, plotted as a function of $y=\omega_H/\omega_E$ for some selected values of the parameter $x=\omega_E\tau$. (b) The real part $\hat{a}_{kk}{}^\mu(x,y)$ of the attenuation factors for the integral correlation with $I=5/2$, plotted as a function of $y=\omega_H/\omega_E$ for some selected values of the parameter $x=\omega_E\tau$.

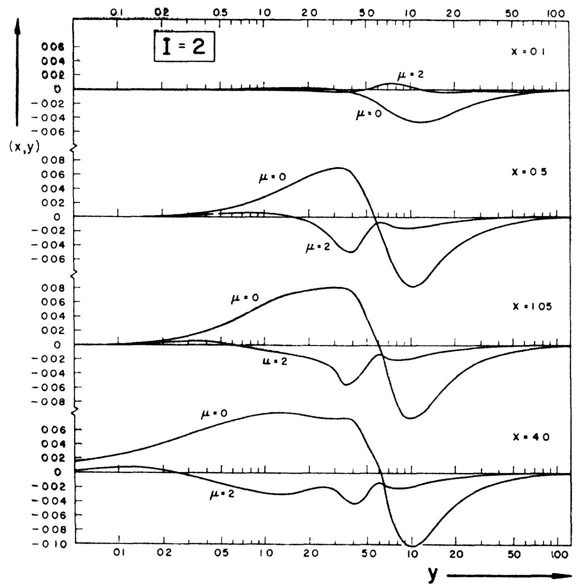


FIG. 8. The real part $\hat{a}_{24}{}^{\mu}(x,y)$ of the interference terms for the integral correlation with $I=2$, plotted as a function of $y=\omega_H/\omega_E$ for some selected values of the parameter $x=\omega_E\tau$.

by the tables, the FORTRAN program may be requested from the authors.

The accuracy of the tabulated values is mainly limited by the numerical integration process. The integration has been performed with the method of Simpson in steps of 0.025 between $\cos\beta=1$ and 0. In order to estimate the accuracy of the computed attenuation factors they have been recomputed for some values of x and y with much smaller integration intervals. A comparison shows that the errors reach a maximum at about $y=3$ and increase, in general, for large x values. In addition, the accuracy decreases slightly with increasing spin values. The accuracy of the computed *integral* attenuation factors is in all cases better than 2%. The differential attenuation factors are considerably less accurate. The reason for this is the oscillating behavior of the attenuation factors for single crystals which is shown in Fig. 3. The number of oscillations increases for increasing values of x.

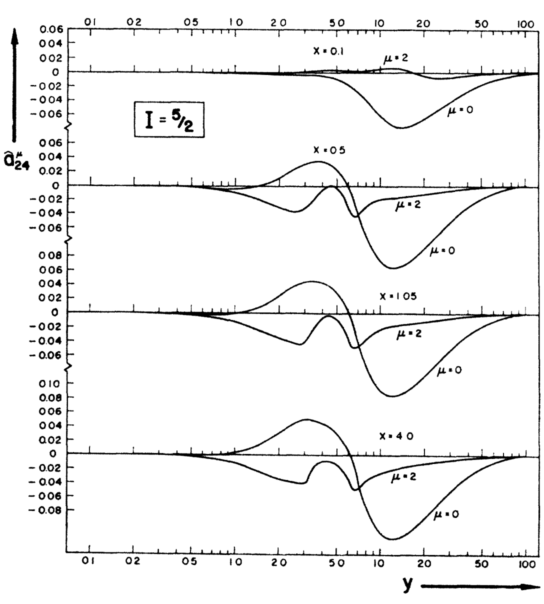


FIG. 9. The real part $\hat{a}_{24}{}^{\mu}(x,y)$ of the interference terms for the integral correlation with $I=5/2$, plotted as a function of $y=\omega_H/\omega_E$ for some selected values of the parameter $x=\omega_E\tau$.

It should be noted that the accuracy of the single-crystal values is better than 0.1%. Measurements with single-crystal sources would provide a sensitive check of the assumptions made for the derivation of the interaction Hamiltonian (17). The single-crystal results are

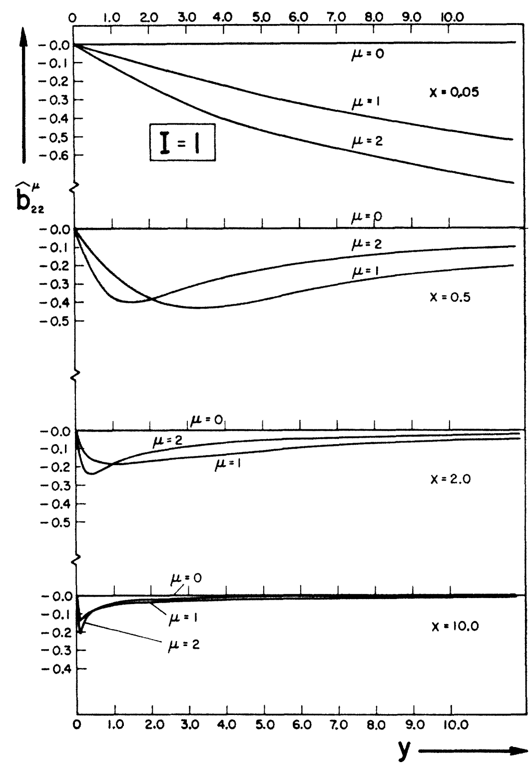


FIG. 10. The imaginary part $\hat{b}_{22}{}^{\mu}(x,y)$ of the attenuation factors for the integral correlation with $I=1$, plotted as a function of $y=\omega_H/\omega_E$ for some selected values of the parameter $x=\omega_E\tau$.

not given here. The tremendous bulk of data exceeds the scope of a publication. The single crystal results for the x and y values mentioned above may be obtained from the authors.

The real parts $a_{kk}{}^{\mu}$ and $\hat{a}_{kk}{}^{\mu}$ of the attenuation factors for polycrystalline sources are plotted in Figs. 4 to 7 as a function of y for some selected parameter values x. In the integral case, two trends in the curves are obvious. For strong magnetic fields, the different μ components are clearly separated by the dominant magnetic interaction. Furthermore, the form of the curves becomes more complex the stronger the electric interaction is. It should be noticed that a_{kk}^0 and $\hat{a}_{kk}^0$ approach the unperturbed asymptotic value for high values of y. This looks like a decoupling behavior which in the semiclassical picture of the vector model means that the spin is precessing around the magnetic field axis and the interaction Hamiltonian is essentially diagonal. In the differential case, the same principal trends as in the integral case can be observed. The time dependence, however, introduces more or less periodic oscillations. Obviously, the measurement of the differential angular correlation is more sensitive for the determination of the interaction strengths because of the more complex structure of the curves [see Fig. 5(a)].

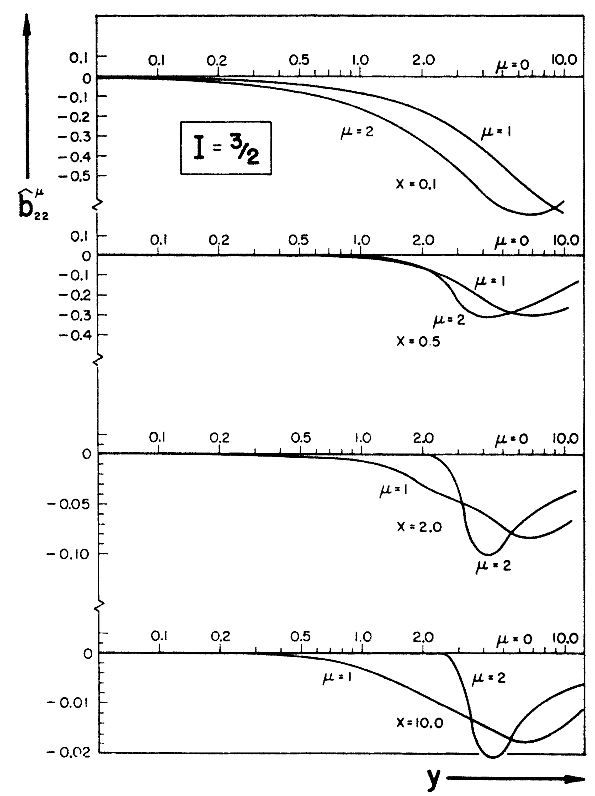


FIG. 11. The imaginary part $\hat{b}_{22}^{\mu}(x,y)$ of the attenuation factors for the integral correlation with $I=3/2$, plotted as a function of $y=\omega_H/\omega_E$ for some selected values of the parameter $x=\omega_E\tau$.

In Figs. 8 and 9 the real parts $\hat{a}_{24}^{\mu}$ of the interference terms of the attenuation factors are shown. These terms give an appreciable contribution only in the region where ω_E and ω_H are of comparable strength, and vanish in the asymptotic regions where $\omega_E \gg \omega_H$ or $\omega_E \ll \omega_H$. The absolute values of even the largest interference terms, however, are an order of magnitude smaller as compared to the $\hat{a}_{kk}^{\mu}$ terms. Since $(k_\nu)_{\max} \leq 2I$, interference terms are only present if $I \geq 2$.

The imaginary parts $\hat{b}_{k_1k_2}^{\mu}$ of the attenuation factors and of the interference terms are plotted in Figs. 10 to 13.

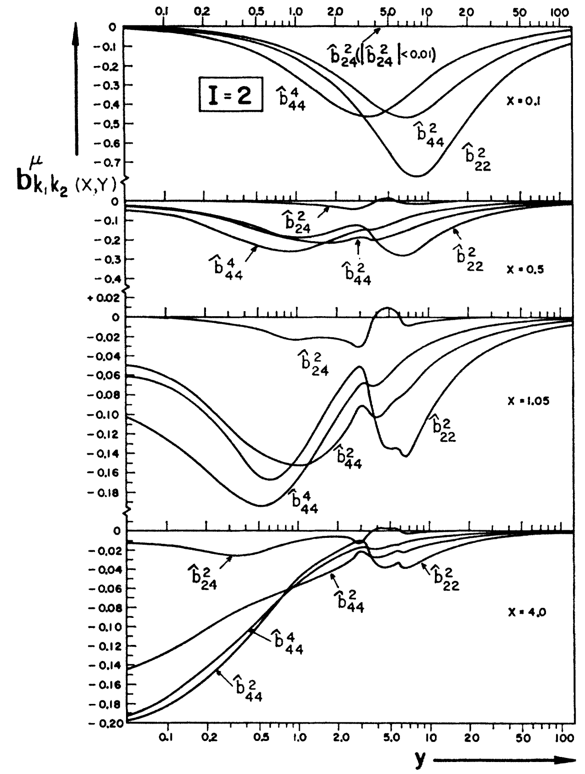


FIG. 12. The imaginary part $\hat{b}_{k_1k_2}^{\mu}(x,y)$ of the attenuation factors and of the interference terms for the integral correlation with $I=2$, plotted as a function of $y=\omega_H/\omega_E$ for some selected values of the parameter $x=\omega_E\tau$.

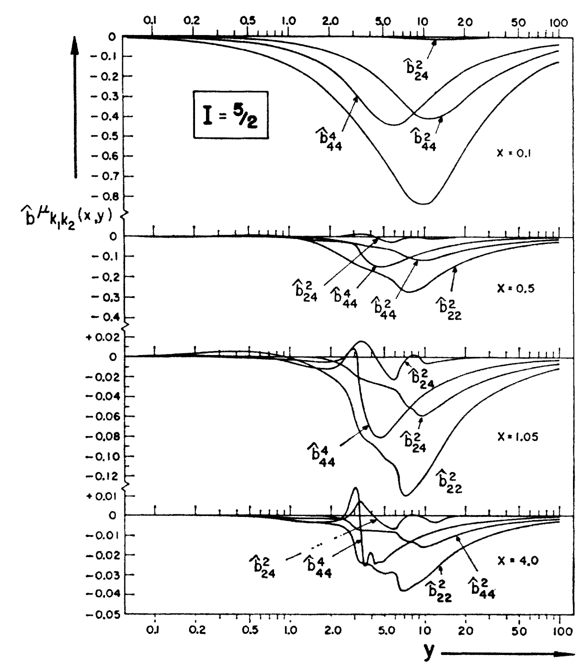


FIG. 13. The imaginary part $\hat{b}_{k_1k_2}^{\mu}(x,y)$ of the attenuation factors and of the interference terms for the integral correlation with $I=5/2$, plotted as a function of $y=\omega_H/\omega_E$ for some selected values of the parameter $x=\omega_E\tau$.

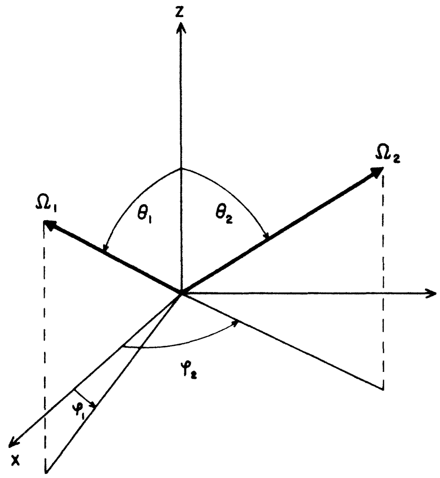


FIG. 14. The propagation directions $\mathbf{\Omega}_1$ and $\mathbf{\Omega}_2$ of the two cascade radiations.

IV. EXPERIMENTAL APPLICATIONS

As mentioned above, the formula (43) can be used to calculate the angular correlation function for a randomly oriented electric quadrupole interaction and a magnetic dipole interaction with an arbitrary direction relative to the detector plane. From the experimental point of view, the anisotropy and the rotation of the angular correlation in a magnetic field are the most interesting quantities; therefore, we find it worthwhile to give expressions for these quantities as an example of the application of the theoretical results. This will be done for the two most important cases: (1) a magnetic field applied perpendicular to the detector plane, and (2) a magnetic field parallel to the emission direction of one of the detected radiations.

1. Magnetic Field Perpendicular to the Detector Plane

As can be seen from Fig. 14, the arguments θ and φ of the spherical harmonics define the emission direction $\mathbf{\Omega}_1$ and $\mathbf{\Omega}_2$ of the two γ rays. If the direction of the magnetic field is chosen parallel to the z axis of the system S, the detectors are placed in the x-y plane. Thus, $\theta_1=\theta_2=\pi/2$ and $\varphi_1-\varphi_2=\Theta$ (see Fig. 14), and Eq. (43) takes the form

$$W_\perp(\Theta)=\sum_{\substack{k_1k_2\\ \mu}} A_{k_1k_2}(a_{k_1k_2}{}^\mu+ib_{k_1k_2}{}^\mu)N_{k_1k_2}{}^\mu\cdot e^{i\mu\Theta}, \tag{52}$$

where

$$A_{k_\nu k_\lambda}=A_{k_\nu}(R_1)\cdot A_{k_\lambda}(R_2),$$

and

$$N_{k_1k_2}{}^\mu=(-1)^\mu Y_{k_1}{}^\mu(\pi/2,0)Y_{k_2}{}^{-\mu}(\pi/2,0)$$

$$=\frac{(-1)^{1/2(k_1+k_2)+\mu}[(2k_1+1)(2k_2+1)(k_1-\mu)!(k_2-\mu)!(k_1+\mu)!(k_2+\mu)!]^{1/2}}{4\pi(k_1-\mu)!!(k_2-\mu)!!(k_1+\mu)!!(k_2+\mu)!!}\quad \text{for}\quad \mu=\text{even},^{17} \tag{53}$$

$$=0\quad \text{for}\quad \mu=\text{odd}.$$

Thus, in the case of a magnetic field perpendicular to the detector plane, only even values of μ must be considered.

The normalization factor in (52) satisfies the same conditions as the attenuation factor does [cf. Eqs. (28) and (50)], namely,

$$N_{k_1k_2}{}^\mu=N_{k_1k_2}{}^{-\mu}\quad\text{and}\quad N_{k_1k_2}{}^\mu=N_{k_2k_1}{}^\mu.$$

The real part of the correlation function (52) can be written in the form:

$$W_\perp(\Theta)=\sum_{\substack{k_1k_2\\ \mu}} A_{k_1k_2}N_{k_1k_2}{}^\mu \times(a_{k_1k_2}{}^\mu\cos\mu\Theta-b_{k_1k_2}{}^\mu\sin\mu\Theta). \tag{54}$$

The sum over μ extends over all values $-\min(k_1,k_2)\leq\mu\leq\min(k_1,k_2)$.

[17] The definition of $(k+\mu)!!$ is $1\times3\times5\times7\cdots$ for odd and $2\times4\times6\times8\cdots$ for even numbers $(k+\mu)$.

Expression (54) may be written in a somewhat different form which displays the rotation of the angular correlation pattern about the magnetic field axis

$$W_\perp(\Theta)=\sum_{\substack{k_1k_2\\ \mu}} A_{k_1k_2}N_{k_1k_2}{}^\mu[(a_{k_1k_2}{}^\mu)^2+(b_{k_1k_2}{}^\mu)^2]^{1/2} \times\cos[\mu(\Theta-\Delta\Theta_{k_1k_2}{}^\mu)], \tag{55}$$

where

$$\tan[\mu\Delta\Theta_{k_1k_2}{}^\mu]=b_{k_1k_2}{}^\mu/a_{k_1k_2}{}^\mu. \tag{56}$$

For the integral correlation the corresponding expressions hold. In Figs. 15(a) and 15(b) the displacement angle $\Delta\Theta_{22}{}^{(2)}$ is shown for the differential and integral correlation for the case of $k_{\max}=2$.

From Eqs. (54) or (55) the general expression for the anisotropy

$$\mathbf{A}=[W_\perp(\pi)-W_\perp(\pi/2)]/W_\perp(\pi/2)$$

can be calculated

$$\mathbf{A}(x,y)$$
$$=\left[60A_{22}a_{22}{}^{(2)}+30\times3^{1/2}(A_{24}+A_{42})a_{24}{}^{(2)}+45A_{44}a_{44}{}^{(2)}\right]$$
$$\times\Bigg[16(2I+1)+10A_{22}(a_{22}{}^{(0)}-3a_{22}{}^{(2)})$$
$$\times\frac{9}{2}A_{44}\left(\frac{9}{4}a_{44}{}^{(0)}-5a_{44}{}^{(2)}+\frac{35}{4}a_{44}{}^{(4)}\right)$$
$$-\tfrac{3}{2}(A_{24}+A_{42})(3\times5^{1/2}a_{24}{}^{(0)}-10\times3^{1/2}a_{24}{}^{(2)})\Bigg]^{-1}. \quad (57)$$

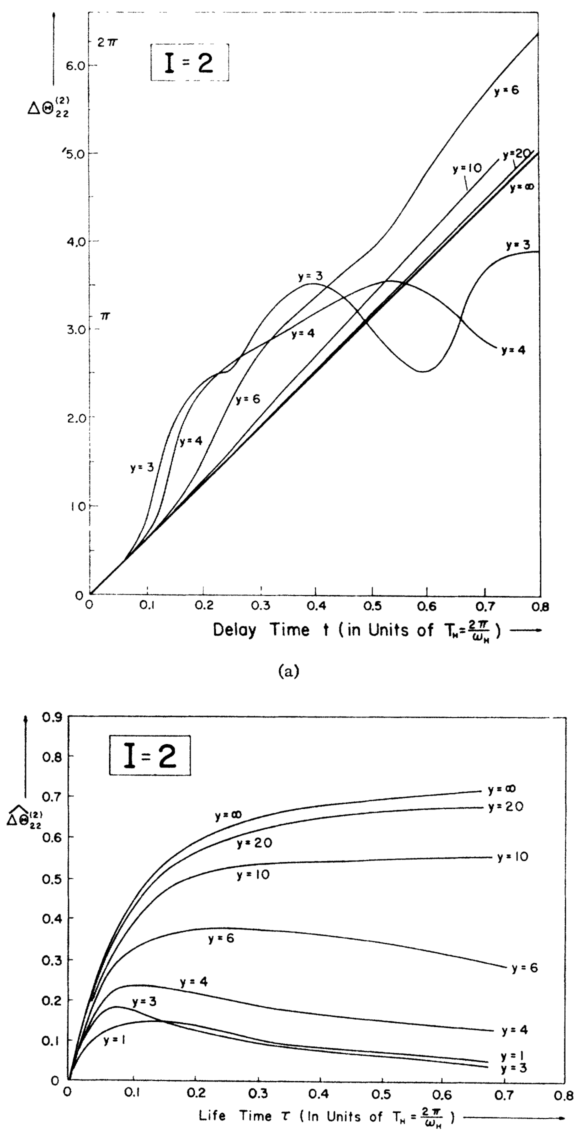


FIG. 15. (a) Displacement angle $\Delta\Theta_{22}{}^{(2)}(x,y)$ for the *differential* angular correlation with $I=2$ as function of y and x. (b) Displacement angle $\Delta\Theta_{22}{}^{(2)}$ for the *integral* angular correlation with $I=2$ as function of y and x.

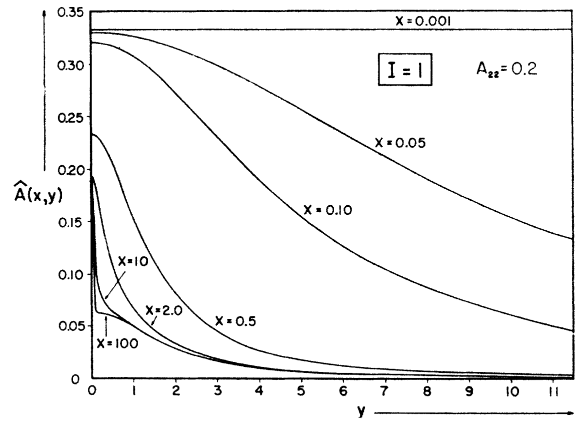


FIG. 16. The anisotropy $\mathbf{A}=[W_\perp(\pi)-W_\perp(\pi/2)]/W_\perp(\pi/2)$ of the integral correlation with magnetic field perpendicular to detector plane for $I=1$.

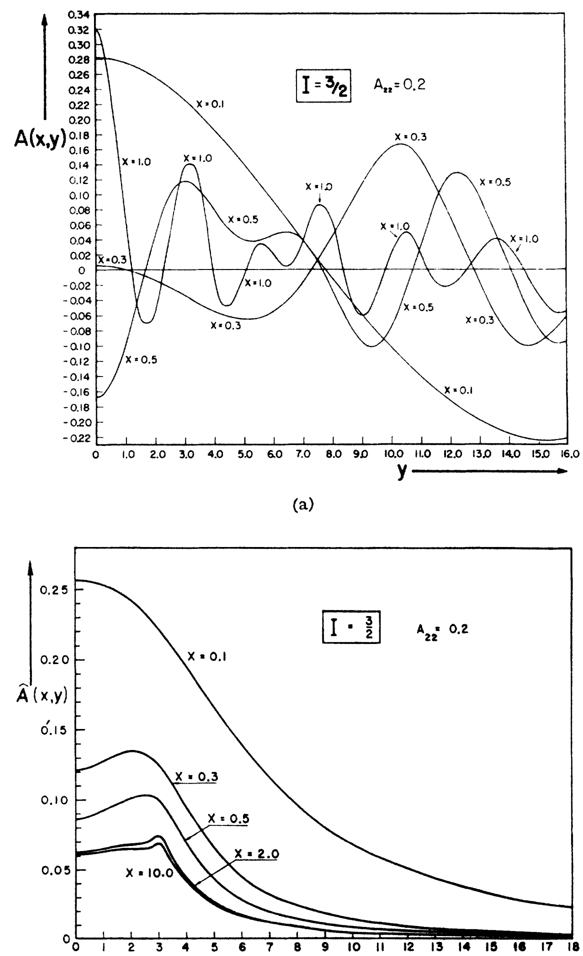


FIG. 17. (a) The anisotropy $\mathbf{A}=[W_\perp(\pi)-W_\perp(\pi/2)]/W_\perp(\pi/2)$ of the *differential* angular correlation with magnetic field perpendicular to detector plane for $I=3/2$. (b) The anisotropy $\mathbf{A}=[W_\perp(\pi)-W_\perp(\pi/2)]/W_\perp(\pi/2)$ of the *integral* correlation with magnetic field perpendicular to the detector plane for $I=3/2$.

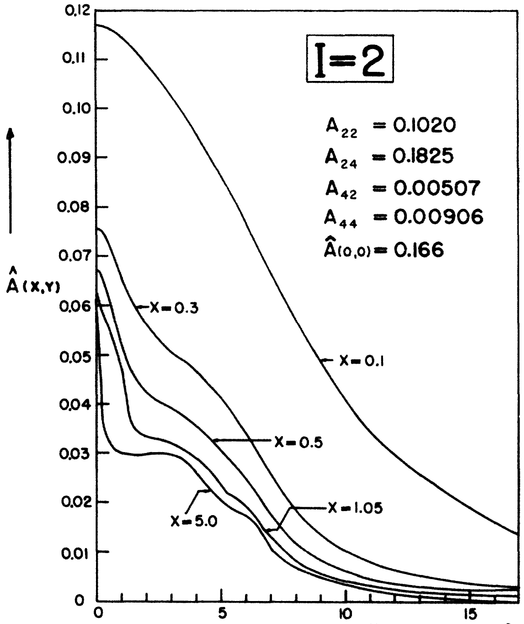


FIG. 18. The anisotropy $\mathsf{A}=[W_\perp(\pi)-W_\perp(\pi/2)]/W_\perp(\pi/2)$ of the integral angular correlation with magnetic field perpendicular to detector plane for $I=2$.

Inserting the factors $a_{k_1k_2}{}^\mu$ for the unperturbed case from Table I the expression for the anisotropy reduces to

$$\mathsf{A}(0)=(12A_{22}+5A_{44})/(8-4A_{22}+3A_{44}). \quad (58)$$

As can be seen from Eq. (57), the anisotropy of the perturbed correlation is determined by the real parts $a_{k_1k_2}{}^\mu$ of the attenuation factors only. This is expected, because the anisotropy is independent of the sign of the azimuthal shift of the angular correlation.

In order to show the behavior of the anisotropy as a function of the interaction strengths, it has been computed for $I=1$ and $I=3/2$ for a typical value of the coefficient $A_{22}=0.20$. The results are shown in Figs. 16 and 17 as a function of y for some selected x values. Figure 18 displays the anisotropy for a $4\rightarrow2\rightarrow0$ gamma-gamma cascade involving quadrupole radiation ($A_{22}=0.1020, A_{24}=0.1825, A_{42}=0.00507, A_{44}=0.00906$). The anisotropy of a pure quadrupole-quadrupole gamma cascade between nuclear states $3/2\rightarrow5/2\rightarrow1/2$ ($A_{22}=0.1020$, $A_{24}=0.1178$, $A_{42}=-0.3770$, $A_{44}=-0.4354$) is shown in Fig. 19. The different behavior of the integral anisotropy for integer and half-integer spin values is remarkable. The reason for this effect is probably the different sign degeneracy of the m sublevels (no degeneracy of the $m=0$ state).

In our discussion so far, it was assumed that the experimental arrangement is such that the detectors can distinguish between the two radiations involved in the angular correlation measurement. If the two detectors respond to each of the radiations with equal efficiency, the angular correlation is given by [cf. Eq. (52)]

$$\bar{W}_\perp(\Theta)=\tfrac{1}{2}\sum_{\substack{k_1k_2\\ \mu}} A_{k_1k_2}(a_{k_1k_2}{}^\mu+ib_{k_1k_2}{}^\mu) \times N_{k_1k_2}{}^\mu(e^{i\mu\Theta}+e^{-i\mu\Theta}) \quad (59)$$

or using Eq. (51)

$$\bar{W}_\perp(\Theta)=\sum_{\substack{k_1k_2\\ \mu}} A_{k_1k_2}N_{k_1k_2}{}^\mu a_{k_1k_2}{}^\mu \cos\mu\Theta. \quad (60)$$

The expression for the anisotropy in this case is, of course, the same as before [cf. Eq. (57)].

2. Magnetic Field Parallel to the Propagation Direction of one of the Radiations

In this case, the arguments of the spherical harmonics are $\theta_1=\varphi_1=0, \theta_2=\Theta$ (see Fig. 20). Since $Y_k{}^\mu(0,0)=[(2k+1)/4\pi]^{1/2}\delta_{\mu0}$ and $Y_k{}^0(\theta_2,\varphi_2)=[(2k+1)/4\pi)^{1/2}\times P_k(\cos\theta_2)$ the general expression [Eq. (43)] reduces to[18]

$$W_{\parallel}(\Theta)=\sum_k A_k(R_1)A_k(R_2)(2k+1)a_{kk}{}^0P_k(\cos\Theta). \quad (61)$$

Thus, in the parallel field case, the influence of the perturbation is represented by the factors $a_{kk}{}^0(x,y)$ or $\hat{a}_{kk}{}^0(x,y)$ only.

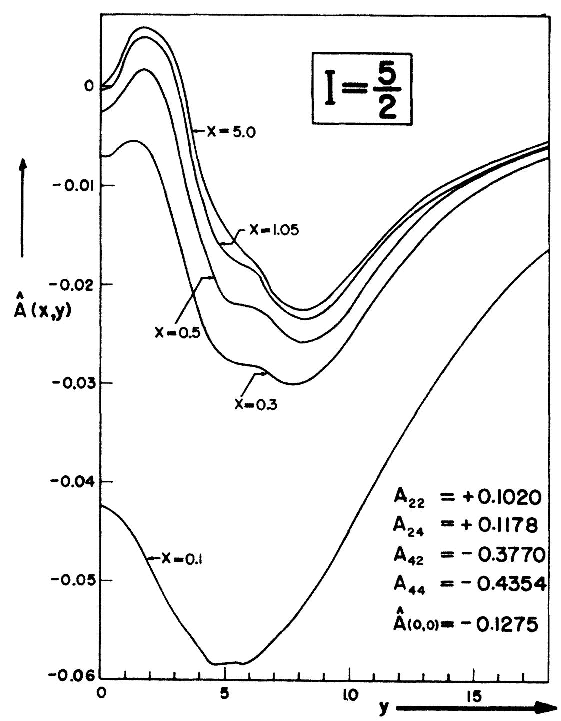


FIG. 19. The anisotropy $\mathsf{A}=[W_\perp(\pi)-W_\perp(\pi/2)]/W_\perp(\pi/2)$ of the integral angular correlation with magnetic field perpendicular to detector plane for $I=5/2$.

[18] The constant factor $1/4\pi$ has been dropped.

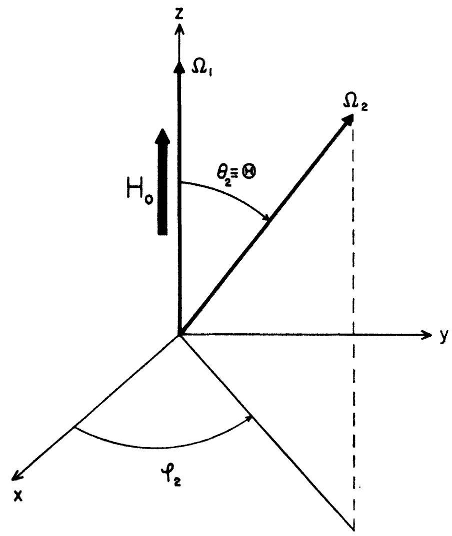


FIG. 20. Magnetic field parallel to the propagation direction of one of the radiations in a nuclear cascade.

From Figs. 4 to 7 it can be seen that for large values of y, i.e., large magnetic interaction the $a_{kk}{}^0$ and $\hat{a}_{kk}{}^0$ reach the unperturbed values asymptotically. This effect is due to some kind of "decoupling" phenomenon[19] which may be interpreted semiclassically. For large magnetic interaction, the precession about the magnetic field axis is much faster as compared to the "quadrupole" precessions, and the influence of the quadrupole interaction is smeared out. Since the magnetic field is parallel to one of the propagation directions, which can be chosen as the quantization axis for the angular correlation problem, the population of the m substates with respect to this axis is essentially stationary if the magnetic interaction is much larger than the quadrupole interaction in the individual microcrystals.

The curves of Figs. 4 to 7 show clearly that the expected effects are large for both the differential and the integral correlation. Thus, it seems possible that such "decoupling" experiments may become a valuable tool for the simultaneous determination of both the magnetic and electric interaction frequencies.

[19] R. Stiening and M. Deutsch, Phys. Rev. **121**, 1484 (1961).

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