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The Measurement of Nuclear Moments of Excited States by Angular Correlation Methods. I

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Summary. It is shown that angular correlation measurements are well suited for the determination of the magnetic moment and the electric quadrupole coupling in short lived excited states. Formulae and graphs are given for various experimental arrangements and spin values between 1 and 7/2. The influence of finite resolving time and delayed coincidence measurements is discussed. Sample calculations are made for the well known case of Cd^{111}.

Introduction.

For the study of nuclear structure and to aid in decision between different nuclear models it is of importance to obtain information about the different nuclear moments. These — the angular momentum, the magnetic moment and the electric quadrupole moment — can be obtained for stable nuclei by well known methods such as optical spectroscopy and nuclear resonance. These methods are applicable with limitations for long lived isomers but are inadequate for short lived excited states. The spin of such excited levels can be determined by the methods of nuclear spectroscopy (nuclear reactions, β and γ spectroscopy). Angular correlation measurement is an especially helpful tool provided that it is not influenced by extra nuclear fields. The investigation of such an influence gives on the other hand information concerning the nuclear moments. DEUTSCH[10]) first pointed out that the magnetic moment of an excited state can be determined by measuring the angular correlation as a function

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of an external magnetic field. This experiment has been carried out by the Zürich group[2)3)] in the case of the first excited state of Cd^{111}. Recently the same group has shown[4)6)13)] that the electric quadrupole interaction can be investigated by analogous experiments. By embedding the active atoms in crystals they measured the quadrupole coupling for the same level of Cd^{111}.

The theoretical base of the experiments was given in an earlier paper[8)]. The there used formalism is very general and an explicite formula is given only for one special case. Therefore it was considered to be worth while to collect the formulae for various experimental arrangements for the determination of magnetic and electric nuclear moments. The sensitivity of different proposed methods will be especially discussed.

When the angular correlation is measured between two nuclear radiations which are emitted successively, the correlation function $W(\vec{k}_1, \vec{k}_2)$ is defined as the relative probability for the emission of the first radiation in the direction $\vec{k}_1$ and the second radiation in the direction $\vec{k}_2$. Any undisturbed correlation function can be expressed as a series in even Legendre polynomials:

$$W(\Theta) = \sum_k A_{kk} P_k(\cos \Theta) \tag{1}$$

where Θ is the angle between $\vec{k}_1$ and $\vec{k}_2$. The coefficients A_{kk} depend on the type of the two radiations and on the spins (and parities) of the three levels involved. They are tabulated for nearly all interesting cases[9)12)15)16)].

In this paper we will restrict ourselves to the important case of unpolarized $\gamma-\gamma$ correlation. In the last section some other possible correlation experiments will be discussed. While no other restrictions are made on the formulae, numerical tables will be given for cases involving no higher multipole orders than quadrupole.

Formula (1) for the correlation function may be completely changed if extra nuclear fields act during the lifetime τ of the intermediate state. To have a measurable influence the interaction energy ΔE must be of the order $\hbar/\tau$. The available field strengths restrict the measurement to cases where τ is longer than about 10^{-9} sec. In the next section we will derive a general formula for the influenced correlation function. This influence depends on the geometry of the experimental arrangement and on attenuation factors G which describe the mechanism of the interaction. We will restrict ourselves to fields with axial symmetry. The magnetic

moment may be determined from its interaction with any magnetic field. The simplest experimental procedure is to apply an external magnetic field and to measure the correlation as a function of the field strength. For an analogous measurement of the electric quadrupole moment no sufficient field gradients are available. One may however use the highly inhomogeneous fields in crystals. According to the previously mentioned restriction our theory will be applicable only for crystals with axial symmetry*).

General Theory.

For the base of the theory we refer to an earlier paper by one of us[8]). The here used notation is in general the same. By I_1, I, I_2 we denote the spins of the three levels involved and by L_1, L_1' and L_2, L_2' the multipole orders of the (in general mixed) first and second radiation.

For the correlation function, i.e. the relative probability for emission of the two quanta in the direction $\vec{k}_1$, $\vec{k}_2$ we write:

$$W(\vec{k}_1\vec{k}_2) = \sum_{l\,p} \Big|\sum_{m} (A_l|H_1(\vec{k}_1)|\,B_m)\,(B_m(t)\,|H_2(\vec{k}_2)|C_p)\Big|^2$$

$$= \sum_{l\,p\,m\,m'} (A_l|H_1|B_m)\,(B_m(t)|H_2|\,C_p)\,(A_l|H_1|B_{m'})^*(B_{m'}(t)|H_2|C_p)^*. \quad (2)$$

Because of the finite lifetime τ of the intermediate state we write for the wavefunction $B_m(\vec{x}, t)$:

$$B_m(\vec{x}, t) = B_m(\vec{x})\, e^{i\,\omega_m t}\, e^{-t/2\tau};\ \ \frac{E_m}{\hbar} = \omega_m. \quad (3)$$

Introducing this expression in the preceding formula, we get:

$$W(\vec{k}_1, \vec{k}_2, t) = \sum_{l\,p\,m\,m'} (A_l|H_1|B_m)\,(B_m|H_2|C_p)\,(A_l|H_1|B_{m'})^*(B_{m'}|H_2|C_p)^*$$

$$\times\, e^{-[1/\tau - i\,(\omega_m - \omega_{m'})]\,t}. \quad (4)$$

$W(\vec{k}_1, \vec{k}_2, t)$ is then the correlation between the radiations when only particles with a fixed delay t between the first and the second

*) Axial symmetry of the field in this connection means that the tensor ellipsoid of $\partial^2 V/\partial x_i\,\partial x_k$ is rotational symmetric.

particle are detected. By integrating over t from 0 to ∞ we get GOERTZEL's formula[11]):

$$W(\vec{k}_1, \vec{k}_2) = \frac{1}{\tau}\int_0^\infty W(\vec{k}_1, \vec{k}_2, t)\, dt$$

$$= \sum_{l\,p\,m\,m'} (A_l|H_1|B_m)\,(B_m|H_2|C_p)\,(A_l|H_1|B_{m'})^*\,(B_{m'}|H_2|C_p)^* \times \frac{1}{1-i(\omega_m-\omega_{m'})\,\tau}\,. \tag{5}$$

In practise one may cut off quanta with too short or too long a delay. We can describe this cut off by a function $\zeta(t)$ which gives the probability for the detection of the two particles with a delay t. We write the integral:

$$\frac{\int_0^\infty \zeta(t)\, e^{-[1/\tau - i(\omega_m-\omega_{m'})]\,t}\, dt}{\int_0^\infty \zeta(t)\, e^{-t/\tau}\, dt} = \frac{H_\zeta[(\omega_m-\omega_{m'})\,\tau]}{1-i(\omega_m-\omega_{m'})\,\tau}\,. \tag{6}$$

The function $H_\zeta(x_{mm'})$ is calculated in a later section of this paper for a number of interesting cases. By introducing the abbreviation $x_{m,m'} = (\omega_m - \omega_{m'})\,\tau$ we have the generalized formula of GOERTZEL in the following form:

$$W(\vec{k}_1, \vec{k}_2) = \sum_{l\,p\,m\,m'} (A_l|H_1|B_m)\,(B_m|H_2|C_p)\,(A_l|H_1|B_{m'})^*\,(B_{m'}|H_2|C_p)^* \times \frac{H_\zeta(x_{mm'})}{1-i\,x_{mm'}}\,. \tag{7}$$

The method of the RACAH algebra can now be applied. For an unpolarized arbitrary correlation we get then:

$$W(\vec{k}_1, \vec{k}_2) = \sum_{\substack{k_1\,k_2\\ \mu}} I^{k_1}\, II^{k_2}\, III_\mu^{k_1 k_2}\, \frac{1}{\sqrt{2\,k_1+1}\,\sqrt{2\,k_2+1}}\, Y_{k_1}^{\mu}(\vec{k}_1)\cdot Y^{-\mu}_{k_2}(\vec{k}_2). \tag{8}$$

The abbreviations I, II, III have the same meaning as in reference[8]):

$$I^{k_1} = \sqrt{2\,k_1+1}\sum_{L_1\,L_1'} \alpha_{L_1}\,\alpha^*_{L_1'}(-1)^{I_1-I}\, W(I\,I_1\,k_1\,L_1'/L_1\,I) \times \sum_M \{C^{k_1\,0}_{L_1\,M\,L_1'-M}\, F^{MM}_{L_1 L_1'}(0)\,(-1)^{L_1-M}\},$$

$$II^{k} = \sqrt{2\,k_2+1}\sum_{L_2\,L_2'} \beta_{L_2}\,\beta^*_{L_2'}(-1)^{I_2-I}\, W(I\,I_2\,k_2\,L_2'/L_2\,I) \times \sum_M \{C^{k_2\,0}_{L_2\,M\,L_2'-M}\, F^{MM}_{L_2 L_2'}(0)\,(-1),^{L_2-M}\},$$

$$III_\mu^{k_1 k_2} = \frac{2\,I+1}{\sqrt{2\,k_1+1}\,\sqrt{2\,k_2+1}}\, \mathcal{G}_\mu^{k_1 k_2} = \sum_{mm'} \frac{C^{I\,m'}_{I\,m\,k_1\,\mu}\, C^{I\,m'}_{I\,m\,k_2\,\mu}}{1-i\,x_{mm'}}\, H_\zeta(x_{mm'})\,. \tag{9}$$

We introduce the coefficients:

$$A_{k_1 k_2} = I^{k_1} II^{k_2} \frac{1}{\sqrt{2\,k_1+1}\sqrt{2\,k_2+1}} \tag{10}$$

and we write the principle formula neglecting the constant factor $2\,I+1$,

$$W(\vec{k}_1 \vec{k}_2) = \sum_{\substack{k_1 k_2 \\ \mu}} A_{k_1 k_2}\, \mathcal{G}_\mu^{k_1 k_2} \frac{1}{\sqrt{2\,k_1+1}\sqrt{2\,k_2+1}}\, Y_{k_1}^{\mu}(\vec{k}_1)\; Y_{k_2}^{-\mu}(\vec{k}_2)\,. \tag{11}$$

For a general arrangement, illustrated in fig. 1a, we rewrite formula (11) by introducing the angles ϑ, φ, Θ. By a simple calculation we get from the spherical triangles in fig. 1b the following relations:

$$\begin{aligned} &\sphericalangle\,(\vec{c}\,\vec{k}_1) = \alpha && \cos\alpha = \cos\vartheta\cos\varphi \\ &\sphericalangle\,(\vec{c}\,\vec{k}_2) = \beta && \cos\beta = \cos\vartheta\cos(\Theta-\varphi) \\ &\sphericalangle\,(\vec{c}\,\vec{k}_1)\,(c\,\vec{k}_2) = \psi && \cos\psi = \frac{\cos\Theta - \cos\alpha\cos\beta}{\sin\alpha\sin\beta}\,. \end{aligned} \tag{12}$$

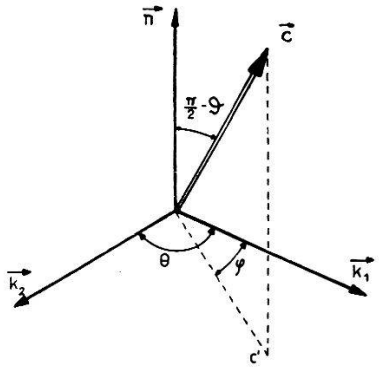

Fig. 1a.

General arrangement for the case of an axial symmetric field (in the direction $\vec{c}$). $\vec{k}_1$ and $\vec{k}_2$ are the directions of the first and the second ray, $\vec{n}$ the normal to the plane defined by $\vec{k}_1$ and $\vec{k}_2$.

For axial symmetric fields we get:

$$\begin{aligned} W(\vartheta, \varphi, \Theta) = Re\Big\{ &\varepsilon_1(1)\cdot\varepsilon_2(2) \sum_{k_1 k_2 \mu} A_{k_1 k_2}\, \mathcal{G}_\mu^{k_1 k_2}\, N_\mu^{k_1 k_2}\, P_\mu^{k_1}(\cos\vartheta\cos\varphi) \\ &\times P_\mu^{k_2}\big(\cos\vartheta\cos(\Theta-\varphi)\big)\, e^{i\mu\psi} \\ &+ \varepsilon_1(2)\,\varepsilon_2(1) \sum_{k_1 k_2 \mu} A_{k_2 k_1}\, \mathcal{G}_\mu^{k_2 k_1}\, N_\mu^{k_2 k_1}\, P_\mu^{k_2}(\cos\vartheta\cos\varphi) \\ &\times P_\mu^{k_1}\big(\cos\vartheta\cos(\Theta-\varphi)\big)\, e^{-i\mu\psi}\Big\} \end{aligned} \tag{13}$$

$$N_\mu^{k_1 k_2} = \sqrt{\frac{(k_1-\mu)!\,(k_2-\mu)!}{(k_1+\mu)!\,(k_2+\mu)!}}\,.$$

$N_\mu^{k_1 k_2}$ is the normalization factor of the Legendre polynomials and $\varepsilon_i(k)$ is the sensitivity of the counter i for the radiation k.

We can simplify formula (13) if we make the two following restrictions:

1. We assume $\varepsilon_1(1) \cdot \varepsilon_2(2) = \varepsilon_1(2) \cdot \varepsilon_2(1)$ (this means in practise that the counters are equally sensitive) —or—we assume the interaction to be pure electric.

2. We assume the interaction to be pure (electric or magnetic) —or— we take for one of the angles ϑ and φ only the special values 0 and $\pi/2$.

If these two conditions are fulfilled only the real part G of the attenuation factors $\mathcal{G}$ enters in the formula and we get:

$$W(\vartheta, \varphi, \Theta) = \sum_{k_1 k_2 \mu} A_{k_1 k_2} G_{\mu}^{k_1 k_2} N_{\mu}^{k_1 k_2} P_{\mu}^{k_1}(\cos\vartheta \cos\varphi) \times P_{\mu}^{k_2}\big(\cos\vartheta \cos(\Theta - \varphi)\big) \cos\mu\psi \,. \tag{14}$$

For the evaluation of (14) one has to sum over μ (taking the terms where $\mu \neq 0$ twice) and over both k_1 and k_2. Furthermore one has $G_0^{k_1 k_2} = \delta_{k_1 k_2}$ whereas all the other attenuation factors are given in a later section.

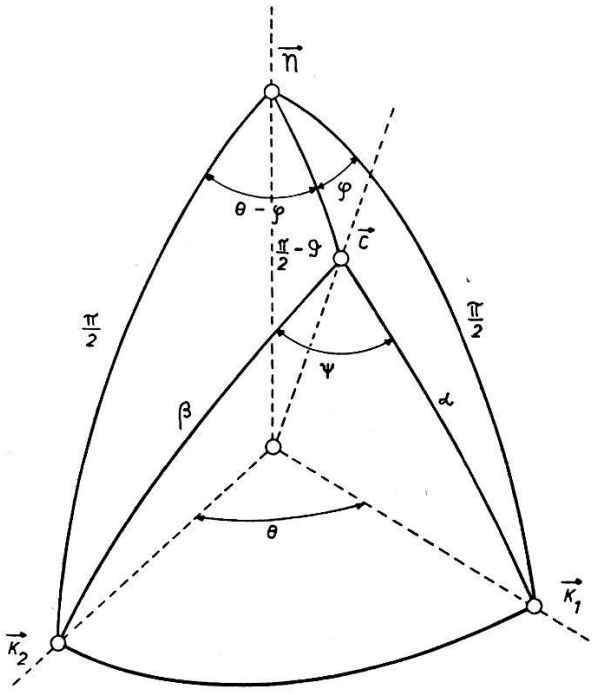

Fig. 1b.
The spherical triangle from which the relations (15) are derived.

Since formula (14) is rather complicated to be used for comparison with experiments we give here explicite formulae for two special cases for Θ, namely $W(\vartheta\, \varphi\, \pi)$ and $W(\vartheta\, \varphi\, \pi/2)$. The anisotropy

$$\varepsilon = W(\vartheta\, \varphi\, \pi)/W(\vartheta\, \varphi\, \pi/2) - 1$$

may be obtained from these functions.

For γ-radiation with no higher multipole order than quadrupole we have then:

$$W(\vartheta\varphi\pi) = a_0 + a_2\cos^2\vartheta\cos^2\varphi + a_4\cos^4\vartheta\cos^4\varphi + a_6\cos^6\vartheta\cos^6\varphi + a_8\cos^8\vartheta\cos^8\varphi \tag{15}$$

$$W(\vartheta\,\varphi\,\pi/2) = b_0 + b_2\cos^2\vartheta + b_4\cos^4\vartheta + b_8\cos^4\vartheta\cos^2\varphi\sin^2\varphi + b_{10}\cos^6\vartheta\cos^2\varphi\sin^2\varphi + b_{16}\cos^8\vartheta\cos^4\varphi\sin^4\varphi \tag{16}$$

where the coefficients a_n and b_n are tabulated in table 1 and table 2. These formulae will be used in the next section to discuss some special arrangements.

Table 1.

$$W(\vartheta\varphi\pi) = a_0 + a_2\cos^2\vartheta\cos^2\varphi + a_4\cos^4\vartheta\cos^4\varphi + a_6\cos^6\vartheta\cos^6\varphi + a_8\cos^8\vartheta\cos^8\varphi$$

$$\begin{aligned}
a_0 &= 1 + A_{22}\,(1/4 + 3/4\,G_2^{22}) + (A_{24}+A_{42})\,(-1/8\cdot\sqrt{15})\,G_2^{24} \\
&\quad + A_{44}\,(9/64 + 5/16\,G_2^{44} + 35/64\,G_4^{44}) \\
a_2 &= A_{22}\,(-3/2 + 3\,G_1^{22} - 3/2\,G_2^{22}) + (A_{24}+A_{42})\,(-9/2\sqrt{5/6}\,G_1^{24} + 9/8\sqrt{15}\,G_2^{24}) \\
&\quad + A_{44}\,(-45/16 + 45/8\,G_1^{44} - 5\,G_2^{44} + 35/8\,G_3^{44} - 35/16\,G_4^{44}) \\
a_4 &= A_{22}\,(9/4 - 3\,G_1^{22} + 3/4\,G_2^{22}) + (A_{24}+A_{42})\,(+30/2\sqrt{5/6}\,G_1^{24} - 15/8\sqrt{15}\,G_2^{24}) \\
&\quad + A_{44}\,(555/32 - 255/8\,G_1^{44} + 195/8\,G_2^{44} - 105/8\,G_3^{44} + 105/32\,G_4^{44}) \\
a_6 &= (A_{24}+A_{42})\,(-21/2\sqrt{5/6}\,G_1^{24} + 7/8\sqrt{15}\,G_2^{24}) \\
&\quad + A_{44}\,(-525/16 + 455/8\,G_1^{44} - 35\,G_2^{44} + 105/8\,G_3^{44} - 35/16\,G_4^{44}) \\
a_8 &= A_{44}\,(1225/64 - 245/8\,G_1^{44} + 245/16\,G_2^{44} - 35/8\,G_3^{44} + 35/64\,G_4^{44})
\end{aligned}$$

Table 2.

$$W(\vartheta\,\varphi\,\pi/2) = b_0 + b_2\cos^2\vartheta + b_4\cos^4\vartheta + b_8\cos^4\vartheta\cos^2\varphi\sin^2\varphi + b_{10}\cos^6\vartheta\cos^2\varphi\sin^2\varphi + b_{16}\cos^8\vartheta\cos^4\varphi\sin^4\varphi$$

$$\begin{aligned}
b_0 &= 1 + A_{22}\,(1/4 - 3/4\,G_2^{22}) + (A_{24}+A_{42})\,1/8\sqrt{15}\,G_2^{24} \\
&\quad + A_{44}\,(9/64 - 5/16\,G_2^{44} + 35/64\,G_4^{44}) \\
b_2 &= A_{22}\,(-3/4 + 3/4\,G_2^{22}) + (A_{24}+A_{42})\,\{-9/16\sqrt{15}\,G_2^{24}\} \\
&\quad + A_{44}\,(-45/32 + 5/2\,G_2^{44} - 35/32\,G_4^{44}) \\
b_4 &= (A_{24}+A_{42})\,7/16\sqrt{15}\,G_2^{24} + A_{44}\,(105/64 - 35/16\,G_2^{44} + 35/64\,G_4^{44}) \\
b_8 &= A_{22}\,(9/4 - 3\,G_1^{22} + 3/4\,G_2^{22}) + (A_{24}+A_{42})\,(+9/2\sqrt{5/6}\,G_1^{24} - 1/8\sqrt{15}\,G_2^{44}) \\
&\quad + A_{44}\,(+345/32 - 45/8\,G_1^{44} - 15\,G_2^{44} + 105/8\,G_3^{44} - 105/32\,G_4^{44}) \\
b_{10} &= (A_{24}+A_{42})\,(-21/4\sqrt{5/6}\,G_1^{24} + 7/16\sqrt{15}\,G_2^{24}) \\
&\quad + A_{44}\,(-525/32 + 105/8\,G_1^{44} + 105/8\,G_2^{44} - 105/8\,G_3^{44} + 105/32\,G_4^{44}) \\
b_{16} &= A_{44}\,(1225/64 - 245/8\,G_1^{44} + 245/16\,G_2^{44} - 35/8\,G_3^{44} + 35/64\,G_4^{44})
\end{aligned}$$

Besides the case illustrated in figure 1 where the $\vec{c}$ axis has the same direction for all the atoms, the case where the field axes are statistically distributed is also of importance. We get then the correlation as the average over all φ, ϑ:

$$W(\Theta) = \int W(\vartheta\, \varphi\, \Theta)\, d\Omega\,. \tag{17}$$

Physically this corresponds to the case of a crystalline powder. It can easily be proved that the result of the integration is:

$$N_{\mu}^{k_1 k_2} \int P_{k_1}^{\mu}[\cos\vartheta \cos\varphi]\, P_{k_2}^{\mu}[\cos\vartheta \cos(\Theta-\varphi)] \cos\mu\psi\, d\Omega = \frac{1}{2k_1+1}\, \delta_{k_1 k_2}\, P_{k_1}(\cos\Theta)\,. \tag{18}$$

We can therefore write in this case the perturbed correlation simply in terms of attenuation factors G^{kk}

$$W(\Theta) = \sum_k A_{kk}\, G^{kk}\, P_k(\cos\Theta)$$

where

$$G^{kk} = \frac{1}{2k+1}\left[1 + 2\sum_{\mu=1}^{k} G_{\mu}^{kk}\right]. \tag{19}$$

For vanishing interaction the attenuation factors $G_{\mu}^{k_1 k_2}$ are just $\delta_{k_1 k_2}$, so that the unperturbed correlation is of the well known form:

$$W(\Theta) = \sum_k A_{kk}\, P_k(\cos\Theta). \tag{20}$$

For our case we are only interested in γ—γ correlation. Then we can get the $A_{k_1 k_2}$ easily from the tables of BIEDENHARN and ROSE[9]). For pure multipoles we write:

$$A_{k_1 k_2} = F_{k1}(L_1 I_1 I)\, F_{k_2}(L_2\, I_2\, I). \tag{21}$$

If one of the two radiations is a mixture of two multipole orders L_1 and L_1' with the intensity ratio δ^2 we get the $A_{k_1 k_2}$ from the table of BIEDENHARN and ROSE:

$$A_{k_1 k_2} = [F_{k_1}(L_1\, I_1\, I) + \delta^2 F_{k_1}(L_1'\, I_1\, I) - 2\,\delta\,\sqrt{(2\,I+1)\,(2\,L_1+1)\,(2\,L_1'+1)}\, G_{k_1}(L_1\, L_1'\, I_1\, I)] \times F_{k_2}(L_2\, I_2\, I) \tag{22}$$

and similar if the other γ ray or both are mixed*).

*) Note that δ^2 is the intensity ratio in the notation of LING and FALKOFF[15]) and different from [9]).

Experimental Arrangements.

Formula (14) of the last section gives the angular correlation for the case in which external axial symmetric fields are present. The correlation depends strongly on the orientation of the symmetry axis with respect to the counter geometry. In this section we will specialize the formula for several experimentally realizable arrangements. The attenuation factors G which involve the mechanism and the strength of interaction will be given in the next section. The formulae of this section are valid for correlations involving no higher multipole order than quadrupole and for axially symmetric electric and magnetic and combined fields. If the interaction is not pure electric, then the sensitivities of the counters are assumed to be the same $(\varepsilon_1(1) \cdot \varepsilon_2(2) = \varepsilon_1(2) \cdot \varepsilon_2(1))$.

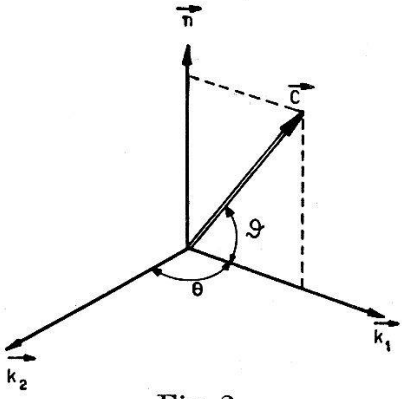

Fig. 2.

Diagram of arrangement 1 where the field axis $\vec{c}$ lies in the plane $(\vec{k}_1, \vec{n})$.

Arrangement 1. In this arrangement (fig. 2) the field axis $\vec{c}$ lies in the plane $(\vec{k}_1\, \vec{n})$. The formula for this case can be obtained from (15) and (16) for $\varphi = 0$. One obtains:

$$\varepsilon = \frac{W(\pi) - W(\pi/2)}{W(\pi/2)} = \frac{\Sigma\, d_n \cos^n \vartheta}{\Sigma\, e_n \cos^n \vartheta} \tag{23}$$

where the coefficients d_n and e_n are tabulated in table 3.

Arrangement 2 (fig. 3). Here the $\vec{c}$ axis lies in the plane $(\vec{k}_1\, \vec{k}_2)$. The formula for this case can be obtained from (15) and (16) for $\vartheta = 0$. One obtains:

$$\varepsilon = \frac{W(\pi) - W(\pi/2)}{W(\pi/2)} = \frac{\Sigma\, f_n \cos^n \varphi}{\Sigma\, g_n \cos^n \varphi} \tag{24}$$

where the coefficients f_n and g_n are tabulated in table 4.

Table 3.

Arrangement 1. $W(\pi)/W(\pi/2)-1 = \Sigma\, d_n \cos^n\vartheta / \Sigma\, e_n \cos^n\vartheta$

$$d_0 = A_{22}\,(3/2\,G_2^{22}) + (A_{24}+A_{42})\,(-1/4\sqrt{15}\,G_2^{24}) + A_{44}\,(5/8\,G_2^{44})$$

$$d_2 = A_{22}\,(-3/4+3\,G_1^{22}-9/4\,G_2^{22}) + (A_{24}+A_{42})\,(-9/2\sqrt{5/6}\,G_1^{24}+27/16\sqrt{15}\,G_2^{24})$$
$$+A_{44}\,(-45/32+45/8\,G_1^{44}-15/2\,G_2^{44}+35/8\,G_3^{44}-35/32\,G_4^{44})$$

$$d_4 = A_{22}\,(9/4-3\,G_1^{22}+3/4\,G_2^{22}) + (A_{24}+A_{42})\,(30/2\sqrt{5/6}\,G_1^{24}-27/16\sqrt{15}\,G_2^{24})$$
$$+A_{44}\,(+1005/64-255/8\,G_1^{44}+425/16\,G_2^{44}-105/8\,G_3^{44}+175/64\,G_4^{44})$$

$$d_6 = (A_{24}+A_{42})\,(-21/2\sqrt{5/6}\,G_1^{24}+7/8\sqrt{15}\,G_2^{24})$$
$$+A_{44}\,(-525/16+455/8\,G_1^{44}-35\,G_2^{44}+105/8\,G_3^{44}-35/16\,G_4^{44})$$

$$d_8 = A_{44}\,(1225/64-245/8\,G_1^{44}+245/16\,G_2^{44}-35/8\,G_3^{44}+35/64\,G_4^{44})$$

$$e_0 = 1+A_{22}\,(1/4-3/4\,G_2^{22}) + (A_{24}+A_{42})\,1/8\sqrt{15}\,G_2^{24}$$
$$+A_{44}\,(9/64-5/16\,G_2^{44}+35/64\,G_4^{44})$$

$$e_2 = A_{22}\,(-3/4+3/4\,G_2^{22}) + (A_{24}+A_{42})\,\{-9/16\sqrt{15}\,G_2^{24}\}$$
$$+A_{44}\,(-45/32+5/2\,G_2^{44}-35/32\,G_4^{44})$$

$$e_4 = (A_{24}+A_{42})\,7/16\sqrt{15}\,G_2^{24} + A_{44}\,(105/64-35/16\,G_2^{44}+35/64\,G_4^{44})$$

Table 4.

Arrangement 2. $W(\pi)/W(\pi/2)-1 = \Sigma\, f_n \cos^n\varphi / \Sigma\, g_n \cos^n\varphi$

$$f_0 = A_{22}\,(3/4+3/4\,G_2^{22}) + (A_{24}+A_{42})\,(-1/8\sqrt{15}\,G_2^{24})$$
$$+A_{44}\,(-15/64+5/16\,G_2^{44}+35/64\,G_4^{44})$$

$$f_2 = A_{22}\,(-15/4+6\,G_1^{22}-9/4\,G_2^{22}) + (A_{24}+A_{42})\,(-15/4\sqrt{5/6}\,G_1^{24}+13/16\sqrt{15}\,G_2^{24})$$
$$+A_{44}\,(45/16-15/8\,G_1^{44}-25/8\,G_2^{44}+35/8\,G_3^{44}-35/16\,G_4^{44})$$

$$f_4 = A_{22}\,(9/2-6\,G_1^{22}+3/2\,G_2^{22}) + (A_{24}+A_{42})\,(57/4\sqrt{5/6}\,G_1^{24}-25/16\sqrt{15}\,G_2^{24})$$
$$+A_{44}\,(-475/64+50/8\,G_1^{44}+115/16\,G_2^{44}-70/8\,G_3^{44}+175/64\,G_4^{44})$$

$$f_6 = (A_{24}+A_{42})\,(-42/4\sqrt{5/6}\,G_1^{24}+14/16\sqrt{15}\,G_2^{24})$$
$$+A_{44}\,(+175/32-35/8\,G_1^{44}-35/8\,G_2^{44}+35/8\,G_3^{44}-35/32\,G_4^{44})$$

$$g_0 = 1-1/2\,A_{22}+3/8\,A_{44}$$

$$g_2 = A_{22}\,(9/4-3\,G_1^{22}+3/4\,G_2^{22}) + (A_{24}+A_{42})\,(-3/4\sqrt{5/6}\,G_1^{24}+5/16\sqrt{15}\,G_2^{24})$$
$$+A_{44}\,(-45/8+15/2\,G_1^{44}-5/8\,G_2^{44})$$

$$g_4 = A_{22}\,(-9/4+3\,G_1^{22}-3/4\,G_2^{22}) + (A_{24}+A_{42})\,(3/4\sqrt{5/6}\,G_1^{24}-5/16\sqrt{15}\,G_2^{24})$$
$$+A_{44}\,(1585/64-305/8\,G_1^{44}+275/16\,G_2^{44}-35/8\,G_3^{44}+35/64\,G_4^{44})$$

$$g_6 = A_{44}\,(-1225/32+245/4\,G_1^{44}-245/8\,G_2^{44}+35/4\,G_3^{44}-35/32\,G_4^{44})$$

$$g_8 = A_{44}\,(1225/64-245/8\,G_1^{44}+245/16\,G_2^{44}-35/8\,G_3^{44}+35/64\,G_4^{44})$$

Arrangement 3. In this arrangement the $\vec{c}$ axis is parallel to the normal to the plane of the two counters ($\vartheta = \pi/2$). For this case we give not only the anisotropy but the whole correlation function. We write it in the form:

$$W(\Theta) = 1 + B_2 \cos 2\Theta + B_4 \cos 4\Theta \tag{25}$$

where

$$B_2 = \frac{1}{1+1/4\, A_{22}+9/64\, A_{44}} \{3/4\, G_2^{22} A_{22} + 5/16\, G_2^{44} A_{44} - \sqrt{15}/8\, G_2^{24}(A_{24}+A_{42})\}$$

$$B_4 = \frac{1}{1+1/4\, A_{22}+9/64\, A_{44}} \cdot 35/64\, G_4^{44} A_{44}\,. \tag{26}$$

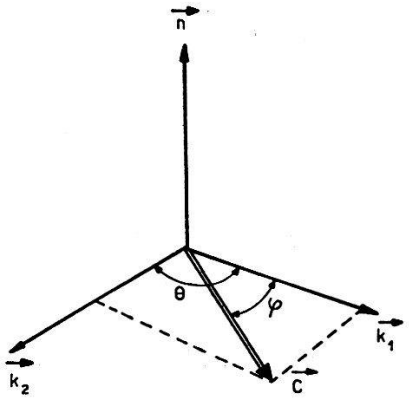

Fig. 3.
Diagram of arrangement 2 where the field axis $\vec{c}$ lies in the plane of the two radiations ($\vec{k_1}$, $\vec{k_2}$).

Arrangement 4. In this arrangement we measure only coincidences between quanta emitted in opposite directions ($\Theta = \pi$). For the coincidence rate $W_\pi(\vartheta)$ one gets an expecially simple expression depending on the angle ϑ:

$$W_\pi(\vartheta) = \sum a_n \cos^n \vartheta \tag{27}$$

where the coefficients a_n are tabulated in table 1.

Arrangement 5. Here we treat the case where the orientations of the $\vec{c}$ axis in the source are statistically distributed. As an example we mention angular correlation measurements in crystalline powder. If the unperturbed correlation is given by

$$W(\Theta) = \sum_k A_{kk} P_k(\cos\Theta) \tag{28}$$

one obtains the influenced correlation by multiplying each term with an attenuation factor:

$$W(\Theta) = \sum_k A_{kk} G^{kk} P_k(\cos\Theta)\,. \tag{29}$$

Attenuation Factors.

The formulae of the last section were given in terms of the attenuation factors G. These factors depend on the type of interaction, the strength of the interaction, the spin of the excitet state and on the coincidence measurement method. The attenuation factors are defined by formula (9) and (19).

$$G_{\mu}^{k_1 k_2} = Re\ \mathcal{G}_{\mu}^{k_1 k_2} = \frac{\sqrt{2k_1+1}\sqrt{2k_2+1}}{2I+1} \cdot \sum_{mm'} \frac{C_{I\,m\,k_1\,\mu}^{I\,m'} C_{I\,m\,k_2\,\mu}^{I\,m'}}{1+x_{mm'}^2} H_{\zeta}(x_{mm'}) \tag{9'}$$

$$G^{kk} = \frac{1}{2k+1}\left[1 + 2\sum_{\mu=1}^{k} G_{\mu}^{kk}\right]. \tag{19'}$$

For magnetic and electric fields with axial symmetry the hyperfine splitting (hfs) is given by

$$\Delta E_m = -\frac{eQ}{2}\frac{\partial E_z}{\partial z}\frac{3m^2 - I(I+1)}{(2I-1)I} + g\mu_k I H_m. \tag{30}$$

The m are magnetic quantum numbers. We introduce x and y which are quantities measuring the strength of interaction and are defined in the following way:

$$\left.\begin{aligned} x &= -\frac{eQ}{2\hbar}\frac{\partial E_z}{\partial z}\frac{3}{(2I-1)I}\tau \quad I \text{ integer} \\ x &= -\frac{eQ}{2\hbar}\frac{\partial E_z}{\partial z}\frac{6}{(2I-1)I}\tau \quad I \text{ half integer} \end{aligned}\right\} \text{for quadrupole interaction} \tag{31}$$

$$y = \frac{g\mu_k H}{\hbar}\tau \qquad \text{for magnetic interaction}$$

Q: electric quadrupole moment of the excited level,
$g\mu_k I = \mu$: magnetic moment of the excited level,
$\partial E_z/\partial z$: electric field gradient with respect to the symmetry axis,
H: magnetic field,
τ: mean life of the excited state.

We can write the attenuation factors in the following general manner:

$$G_{\mu}^{k_1 k_2} = \frac{1}{2}\sum_{m} S_{m\mu}^{k_1 k_2}\left[\frac{1}{1+(mx+\mu y)^2} H(mx+\mu y, \xi, \eta) + \frac{1}{1+(mx-\mu y)^2} H(mx-\mu y, \xi, \eta)\right]. \tag{32}$$

For negligible interaction the following relation holds:

$$G_{\mu}^{k_1 k_2} = \delta_{k_1 k_2}.$$

Formula (32) is valid for $\vec{c}_H = \vec{c}_Q$, i.e. the axis of the magnetic field is parallel to the symmetry axis of the crystal. For the special case that only one interaction (H or Q) is present one gets the attenuation factors simply by setting $x = 0$ or $y = 0$.

For pure magnetic interaction formula (32) simplifies to:

$$G_{\mu}^{k_1 k_2} = \delta_{k_1 k_2} \frac{H(\mu y, \xi, \eta)}{1+(\mu y)^2}. \tag{32a}$$

For powder sources (arrangement 5) when only a magnetic or an electric field is present*), the attenuation factors G^{kk} are given by:

$$G^{kk} = \sum S_m^{kk} \frac{1}{1+(m x)^2} H(mx, \xi, \eta). \tag{33}$$

(For magnetic interaction replace x by y).

The coefficients S are sums over Clebsch-Gordan coefficients (9) and are tabulated for the spin values 1, 3/2, 2, 5/2, 3 and 7/2 and all interesting cases in table 5.

The function H defined by formula (6) describes the influence of the coincidence measurement method and depends on the finite resolving time τ_R of the coincidence circuit, on the delay τ_D and on the nuclear lifetime τ of the intermediate state. As variables we chose the ratios

$$\xi = \tau_R/\tau \quad \text{and} \quad \eta = \tau_D/\tau.$$

For the most important cases the function H is given as follows:

A. Without delay and with infinite resolving time.

$\eta = 0, \quad \xi = 0$

$H \equiv 1$ (34)

*) The attenuation factors for powder sources where in addition an axial magnetic field is present is not given in this paper. This experiment which allows the determination of μ and Q in one experiment without creating a single crystal is of certain importance and will be treated and published later.

Table 5: $S_{m\mu}^{k_1 k_2}$ and S_m^{kk}.

The attenuation factors $G_\mu^{k_1 k_2}$ and G^{kk} for magnetic and electric interaction may be obtained from this table by introducing the here given coefficients $S_{m\mu}^{k_1 k_2}$ and S_m^{kk} in formulae (32) and (33).

	m / 1	0	1	2	3	4	5	6	7	8	9
S_{m1}^{22}	1		1								
	$\frac{3}{2}$		1								
	2		$\frac{1}{7}$		$\frac{6}{7}$						
	$\frac{5}{2}$		$\frac{2}{7}$	$\frac{5}{7}$							
	3		$\frac{2}{42}$		$\frac{15}{42}$		$\frac{25}{42}$				
	$\frac{7}{2}$		$\frac{5}{42}$	$\frac{16}{42}$	$\frac{21}{42}$						
S_{m2}^{22}	1	1									
	$\frac{3}{2}$		1								
	2	$\frac{3}{7}$				$\frac{4}{7}$					
	$\frac{5}{2}$		$\frac{9}{14}$		$\frac{5}{14}$						
	3	$\frac{6}{21}$				$\frac{10}{21}$				$\frac{5}{21}$	
	$\frac{7}{2}$		$\frac{20}{42}$		$\frac{15}{42}$		$\frac{7}{42}$				
S_{m1}^{44}	2		$\frac{6}{7}$		$\frac{1}{7}$						
	$\frac{5}{2}$		$\frac{5}{7}$	$\frac{2}{7}$							
	3		$\frac{15}{77}$		$\frac{32}{77}$		$\frac{30}{77}$				
	$\frac{7}{2}$		$\frac{27}{77}$	$\frac{15}{77}$	$\frac{35}{77}$						
S_{m2}^{44}	2	$\frac{4}{7}$				$\frac{3}{7}$					
	$\frac{5}{2}$		$\frac{5}{14}$		$\frac{9}{14}$						
	3	$\frac{20}{77}$				$\frac{3}{77}$				$\frac{54}{77}$	
	$\frac{7}{2}$		$\frac{48}{154}$		$\frac{1}{154}$		$\frac{105}{154}$				
S_{m3}^{44}	2				1						
	$\frac{5}{2}$				1						
	3				$\frac{2}{11}$						$\frac{9}{11}$
	$\frac{7}{2}$				$\frac{4}{11}$			$\frac{7}{11}$			

Table 5 continued.

	I \ m	0	1	2	3	4	5	6	7	8	9
S^{44}_{m4}	2	1									
	$\frac{5}{2}$			1							
	3	$\frac{5}{11}$								$\frac{6}{11}$	
	$\frac{7}{2}$			$\frac{15}{22}$				$\frac{7}{22}$			
S^{24}_{m1}	2		$-\frac{\sqrt{6}}{5}$		$\frac{\sqrt{6}}{5}$						
	$\frac{5}{2}$		$-\frac{\sqrt{10}}{7}$	$\frac{\sqrt{10}}{7}$							
	3		$\frac{-1}{7}\sqrt{\frac{5}{11}}$		$\frac{-4}{7}\sqrt{\frac{5}{11}}$		$\frac{5}{7}\sqrt{\frac{5}{11}}$				
	$\frac{7}{2}$		$\frac{-3}{14}\sqrt{\frac{10}{11}}$	$\frac{-4}{14}\sqrt{\frac{10}{11}}$	$\frac{7}{14}\sqrt{\frac{10}{11}}$						
S^{24}_{m2}	2	$\frac{-2}{7}\sqrt{3}$				$\frac{2}{7}\sqrt{3}$					
	$\frac{5}{2}$		$\frac{-3}{14}\sqrt{5}$		$\frac{3}{14}\sqrt{5}$						
	3	$\frac{-2}{7}\sqrt{\frac{10}{11}}$				$\frac{-1}{7}\sqrt{\frac{5}{11}}$				$\frac{3}{7}\sqrt{\frac{5}{11}}$	
	$\frac{7}{2}$		$\frac{-7}{14}\sqrt{\frac{5}{11}}$		$\frac{-1}{14}\sqrt{\frac{5}{11}}$		$\frac{8}{14}\sqrt{\frac{5}{11}}$				
S^{22}_{m}	1	$\frac{3}{5}$	$\frac{2}{5}$								
	$\frac{3}{2}$	$\frac{1}{5}$	$\frac{4}{5}$								
	2	$\frac{13}{35}$	$\frac{2}{35}$		$\frac{12}{35}$	$\frac{8}{35}$					
	$\frac{5}{2}$	$\frac{7}{35}$	$\frac{13}{35}$	$\frac{10}{35}$	$\frac{5}{35}$						
	3	$\frac{33}{105}$	$\frac{2}{105}$		$\frac{15}{105}$	$\frac{20}{105}$	$\frac{25}{105}$			$\frac{10}{105}$	
	$\frac{7}{2}$	$\frac{42}{210}$	$\frac{50}{210}$	$\frac{32}{210}$	$\frac{72}{210}$		$\frac{14}{210}$				
S^{44}_{m}	2	$\frac{29}{63}$	$\frac{12}{63}$		$\frac{16}{63}$	$\frac{6}{63}$					
	$\frac{5}{2}$	$\frac{7}{63}$	$\frac{15}{63}$	$\frac{18}{63}$	$\frac{23}{63}$						
	3	$\frac{187}{693}$	$\frac{30}{693}$		$\frac{92}{693}$	$\frac{6}{693}$	$\frac{60}{693}$			$\frac{192}{693}$	$\frac{126}{693}$
	$\frac{7}{2}$	$\frac{77}{693}$	$\frac{102}{693}$	$\frac{135}{693}$	$\frac{127}{693}$		$\frac{105}{693}$	$\frac{147}{693}$			

B. Without delay and with finite resolving time.

$\eta = 0, \quad \xi \neq 0$

$$H = \frac{1 - e^{-\xi}(\cos z\,\xi - z \sin z\,\xi)}{1 - e^{-\xi}} \tag{35}$$

C. With delay and with finite resolving time. (The sensitivities of the counters are assumed to bee the same).

a) $\eta \leqslant \xi$

$$H = \frac{2 - e^{-(\xi+\eta)}(\cos z(\xi+\eta) - z \sin z(\xi+\eta)) - e^{-(\xi-\eta)}(\cos z\,(\xi-\eta) - z \sin z\,(\xi-\eta))}{2 - e^{-(\xi+\eta)} - e^{-(\xi-\eta)}} \tag{36}$$

b) $\eta \geqslant \xi$

$$H = \frac{(\cos z\,(\eta-\xi) - z \sin z\,(\eta-\xi)) - e^{-2\xi}(\cos z\,(\eta+\xi) - z \sin z\,(\eta+\xi))}{1 - e^{-2\xi}} \tag{37}$$

D. With delay and a very short resolving time.

$\xi \ll 1, \quad \xi \ll \eta$

$$H = (1 + z^2) \cos z\,\eta \tag{38}$$

In the formulae (34) to (38) the varable z stands for $(mx + \mu y)$ or $(mx - \mu y)$.

Discussion of experimental methods.

In the previous sections it was shown that angular correlation measurement is able to give information about nuclear moments of short lived excited states. From the measurement we get the product of the interaction energy and the nuclear lifetime. As in the region in question (10^{-9} sec to 10^{-5} sec) lifetimes can be measured with good accuracy, the interaction energy can be calculated. From this we get the moment when the interacting field strength is known. For the magnetic case magnetic fields can be measured accurately and it is therefore possible to obtain the g factor and therefore the magnetic moment with an error of only a few percent. As will be deduced in the appendix the measurement of the sign of the magnetic moment can also be carried out very easily. The electric case is much more difficult. The field strength that enters into the interaction energy is the gradient of the electric crystalline field which in general can be calculated only with great difficulty. It would therefore be interesting to measure the quadrupole coupling not in

crystals but in atoms or molecules for which rather good calculations are available. The quadrupole moment may however be obtained by comparison of the measured quadrupole coupling which that of a stable nucleus, the latter being obtained from a nuclear induction experiment. Any calculation or measurement of the electric field gradient will in general be complicated by the fact that the interaction is measured in a state following a radioactive decay. The

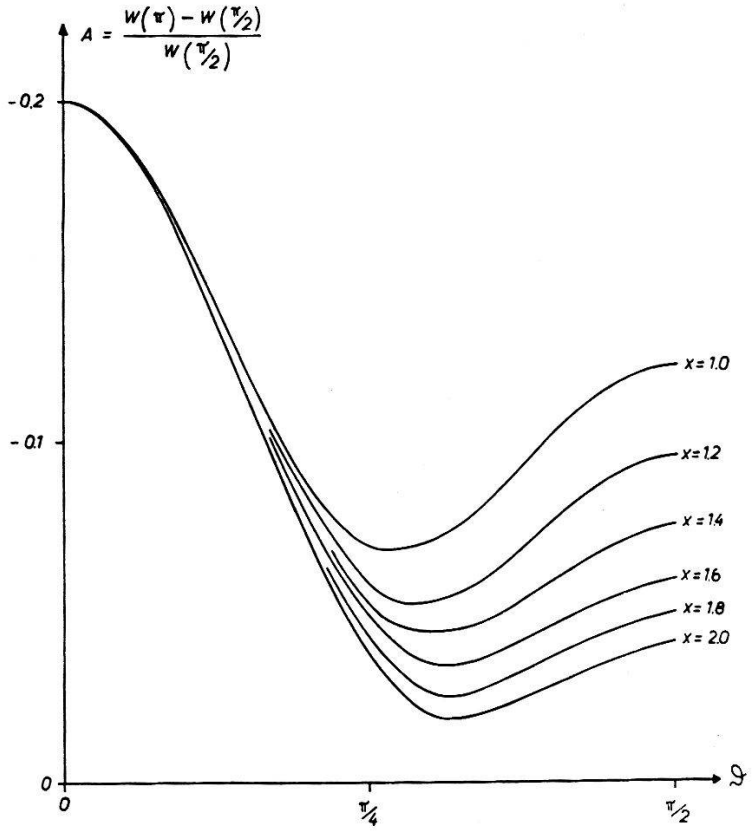

Fig. 4.

This figure illustrates the arrangement 1 in the case of Cd^{111}. The region of x has been chosen so that the curves may be compared with the recently reported experimental results [4)6]). The curves are corrected for finite resolving time ($\xi = 2{,}84$) but not for finite solid angle.

recoil energy may remove the active atom from a lattice position and radiations may disturb the electronic shell and may cause additional effects.

The applicability of the described method is limited in several directions. In principle the investigated state need not be the intermediate state of a $\gamma-\gamma$ cascade. The theory may be expanded to $\beta-\gamma$, $\alpha-\gamma$ and $e^-\!-\gamma$ correlation e.g., although the experiments are much more difficult. Furthermore the method may be applied to measurements of angular distributions in nuclear reactions.

*

Another restriction is that the lifetime of the intermediate state must lie between 10^{-9} sec and 10^{-5} sec. The lower limit is given by available field strengths whereas the upper limit will be determined by small disturbing effects present in any source.

For any accurate determination of the moments the source must fulfill some additional conditions. For the measurement of the magnetic moment there should be no quadrupole coupling present. As it was pointed out in a previous article[5]) cubic crystalline sources,

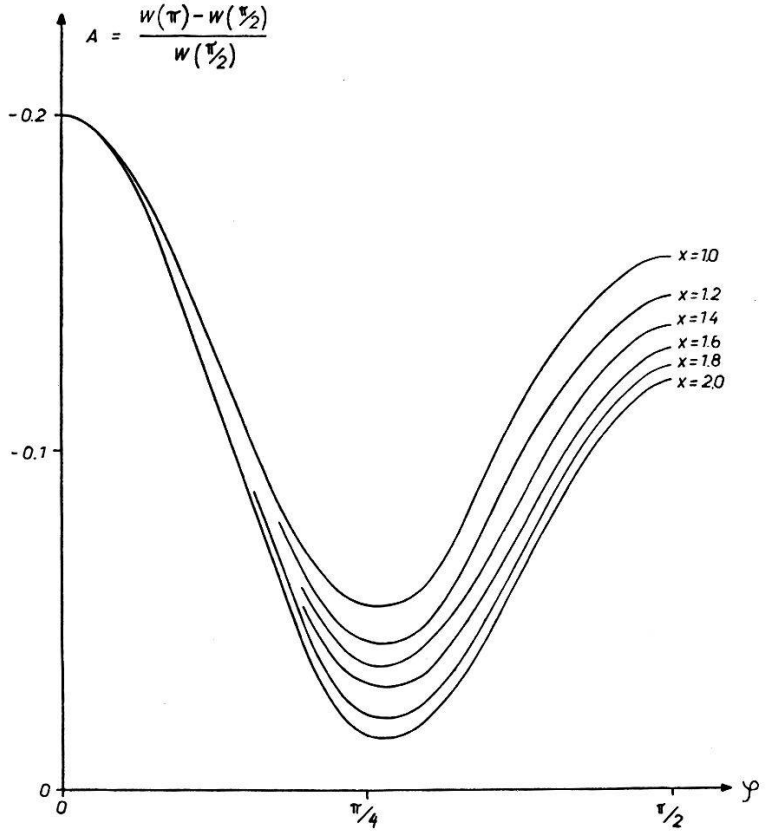

Fig. 5.

Same as fig. 4 but for arrangement 2.

solutions*) and melts are in general suited for this purpose. For the measurement of the electric quadrupole coupling one best uses a very pure single crystal of axial symmetry. The radioactive atoms must sit in regular lattice positions. For both cases diamagnetic compounds should be used to avoid disturbance by the (IJ) coupling.

We have then three methods for the determination of x (and y): 1. We keep the field, delay and resolving time constant and vary

*) See also: A. Abragam and R. V. Pound, to be. publ. and H. Albers-Schoenberg, E. Heer, F. Gimmi und T. B. Novey, Helv. Phys. Acta, **26**, 599 (1953).

the angles ϑ and φ. 2. We keep the field and the angles ϑ, φ constant and vary the delay or the resolving time; and 3. we keep ϑ, φ and the delay and resolving time constant and vary the field. For the determination of the magnetic moment possibilities 2. and 3. with the field axis perpendicular to the two gammas are best suited. Since the electric field gradient can not be varied easily one must vary ϑ and φ for the measurement of the quadrupole coupling. In

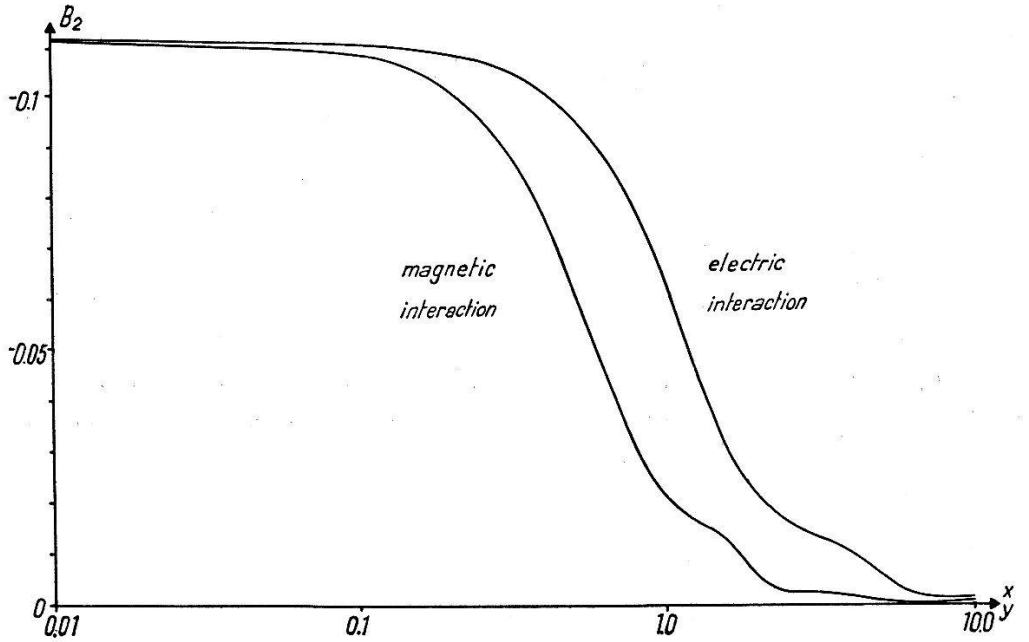

Fig. 6.

Illustration of arrangement 3 where the symmetry axis of the field is perpendicular to the plane of the two counters in the case of Cd^{111}. The disturbed a.c. is written as $W(\Theta) = 1 + B_2 \cos 2\Theta$. The field may either be pure electric or magnetic.

any case it is preferable to measure the attenuation as a function of one variable and to compare with the theory by the method of least square fit. In this way one can make sure that no other disturbing interaction is present.

With regard to the coincidence measurement method, the case is simplest without delay and with a finite resolving time. If the interaction is small it is however preferable to employ a delay of the order of the lifetime. One measures then only the nuclei that have been exposed to the disturbing field for a long time and the effects are larger (see fig. 8).

To complete this section we will illustrate some of the results of this paper for the well known case of Cd^{111}. The γ—γ cascade is a $7/2 \xrightarrow[E2]{M1} 5/2 \xrightarrow{E2} 1/2$ transition. The mixing ratio δ has been deter-

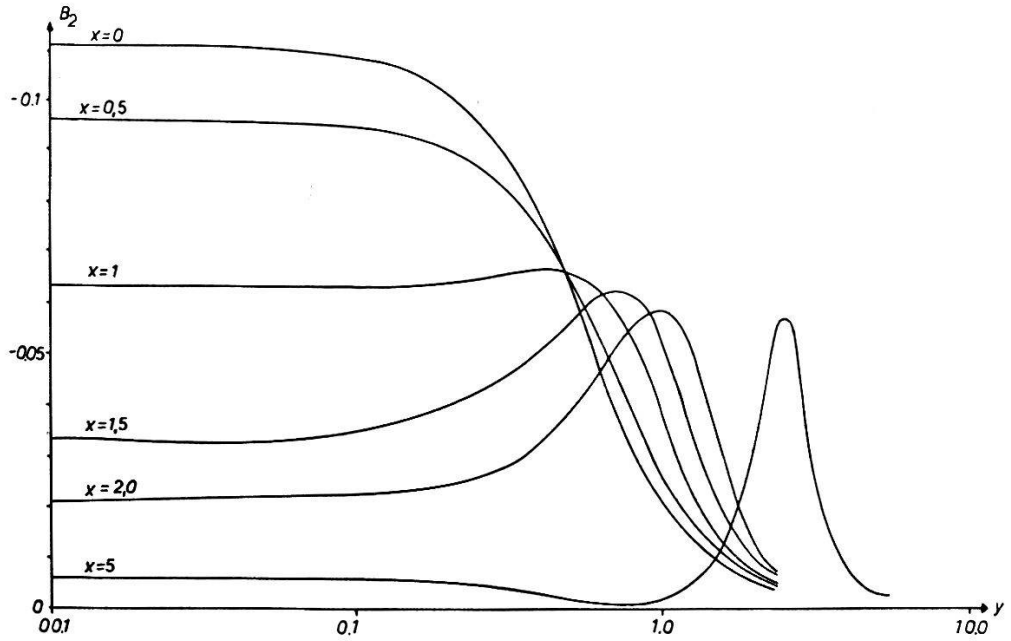

Fig. 7.

Same as fig. 6 but for combined parallel magnetic and electric field for several values of the electric interaction x. B_2 is given as a function of the magnetic interaction y.

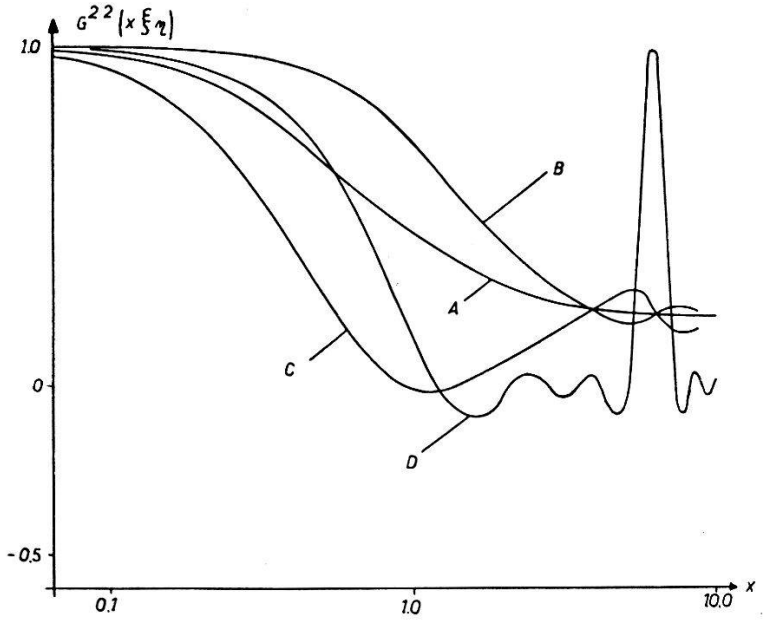

Fig. 8.

Attenuation factor G^{22} for polycrystalline powder sources for $I = 5/2$ as a function of the strength of interaction x. The curves A, B, C and D show the influence of finite resolving time and delay. The values used are: A: $\xi = \infty$, $\eta = 0$; B: $\xi = 1$, $\eta = 0$; C: $\xi = 1$, $\eta = 1$; D: $\xi \ll 1$, $\eta = 1$.

mined by angular correlation methods[1]) to be about $-0{\cdot}096$. From the table of BIEDENHARN and ROSE one gets for the coefficients $A_{22} = -0{\cdot}1429$, $A_{24} = -0{\cdot}1650$, $A_{42} = A_{44} = 0$ if we neglect terms in δ^2.

In figure 4 and 5 we have illustrated experimental arrangement 1 and 2. The recently published experimental results of the Zürich group[4])[2]) are in good agreement with these curves. Figure 6 and 7 show the important case where the symmetry axis is perpendicular to the two rays for pure magnetic, pure electric and combined magnetic and electric interaction. The rise of the anisotropy for small H (fig. 7) is characteristic for quadrupole interaction. This figure illustrates the possibility for a determination of μ and Q in one experiment[7]). Figures 4 to 7 are corrected for the finite resolving time ($\xi = 2{,}84$). In figure 8 the influence of the coincidence measurement method is illustrated. The attenuation factor G for a crystalline powder source is given as a function of x.

These curves show the sensitivity of the various methods of detecting and measuring the quadrupole interaction and allow one to select the best procedure to be used in any particular case.

We are grateful to Prof. SCHERRER for his continued encouragement and support of our work. We also thank O. BRAUN for help with many laborious calculations.

APPENDIX.

Some special theoretical problems.

1. *Symmetry behaviour.*

In the preceding sections the correlation function was partly written as a series of Legendre polynominals and partly as a series in $\cos 2n\Theta$. The form was chosen according to the symmetry of the problem. When the correlation function depends only on the angle Θ between the two counters and not on their orientation in space then the representation in Legendre polynominals is well suited. If on the other hand the correlation function is invariant on rotation of the counting system around a symmetry axis the adequate representation is that in $\cos 2n\Theta$. If there is no symmetry behaviour at all, the correlation function is very complicated and

is best expressed in spherical harmonics. For practical purpose it is then better not to calculate the whole correlation function but only the value for a fixed angle Θ ($\Theta = \pi$ or $\Theta = \pi/2$).

2. *Vector model.*

For magnetic interaction the formula for the influenced correlation can be obtained easily by semiclassical procedure. An applied magnetic field H is equivalent to a Larmor precession $\omega_L = g\,\mu_k H/\hbar$ of the magnetic moment of the nucleus around the field axis. By transforming to the rotating system we get

$$W = \int W(\vartheta_1,\, \varphi_1\,,\, \vartheta_2,\, \varphi_2 + \omega t)\; \zeta(t)\; e^{-t/\tau}\, dt$$

if $W(\vartheta_1 \varphi_1\; \vartheta_2 \varphi_2)$ is the undisturbed correlation. For quadrupole interaction it is more complicated since we have not only one but a whole spectrum of precession frequencies. For $I = 5/2$ e.g. we have the frequencies $\pm\,\omega$, $\pm\,2\,\omega$, $\pm\,3\,\omega$.

3. *Sign of the moment.*

The formulae of this paper are valid for equally sensitive counters. If the sensitivities of the two counters for the two γ-rays are not equal, the formulae for the magnetic interaction are more complicated. The attenuation factors are then given by the complex expression:

$$\mathcal{G}_{\mu}^{k_1 k_2} = \delta_{k_1 k_2}\left[\frac{1}{1+(\mu y)^2} + i\,\frac{\varepsilon_1(1)\,\varepsilon_2(2) - \varepsilon_1(2)\,\varepsilon_2(1)}{\varepsilon_1(1)\,\varepsilon_2(2) + \varepsilon_1(2)\,\varepsilon_2(1)}\,\frac{\mu y}{1+(\mu y)^2}\right]$$

where $\varepsilon_i(k)$ is the sensitivity of the counter i for the radiation k. Then the correlation $W(\vec{k}_1, \vec{k}_2)$ and $W(\vec{k}_1, -\vec{k}_2)$ are not the same and from their difference the sign of the magnetic moment can be obtained[3]).

In the case of the quadrupole interaction the attenuation factors are independent of the sensitivity of the counters and always real. Therefore the determination of the sign of the quadrupole interaction is not possible in the same way.

4. *Angular correlation in fields without symmetry axis*

This case includes crystals without symmetry axis, axial magnetic and axial electric fields which are not parallel and crystalline powder plus magnetic axial field. The angular correlation can even in this case be written in terms of attenuation factors $G^{k_1 k_2}_{\mu_1 \mu_2}$:

$$W(\vec{k}_1 \vec{k}_2) = \sum_{\substack{k_1 k_2 \\ \mu_1 \mu}} A_{k_1 k_2} G^{k_1 k_2}_{\mu_1 \mu_2} Y^{\mu_1}_{k_1}(\vec{k}_1) \; Y^{\mu_2}_{k_2}(\vec{k}_2) .$$

To calculate the attenuation factors one must know the *hfs* and the admixture of different magnetic quantum numbers. We hope that in a next paper more quantitative results can be discussed.

5. *(IJ) coupling.*

The interaction between the nucleus and the electronic shell of the active atom may in principle be electric or magnetic. For the attenuation factors G^{kk} one gets:

$$G^{kk} = \sum \frac{(2F+1)(2F'+1)[W(I\,J\,k\,F/F'I)]^2}{1+(\omega_{FF'}\tau)^2}$$

if the disturbed correlation is:

$$W(\Theta) = \sum_k A_{kk} G^{kk} P_k(\cos\Theta) .$$

In the magnetic case the *hfs* $\omega_{FF'}$ is given by

$$\omega_{FF'} = \frac{1}{2} A_s [F(F+1) - F'(F'+1)]$$

where A_s is a constant depending on the electron configuration and is tabulated e.g. in KOPFERMANN[14]). In the electric case the hfs $\omega_{FF'}$ is given by

$$\omega_{FF'} = \frac{1}{2} eQ \frac{\partial E_z}{\partial z} [\{F(F+1) - F'(F'+1)\}$$

$$\times \{F(F+1) + F'(F'+1) - 2I(I+1) - 2J(J+1) + 1\}] .$$

If both interactions are present $\omega_{FF'}$ is simply the sum of the two expressions.

Literature cited.

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A work of similar nature which comprises a natural complement to this paper and which treats especially the effect of Brownian motion in liquid sources has been completed by Abragam and Pound, to be published in the near future. We thank Prof. Pound for communicating this work to us.