Directional Correlation of Successive Nuclear Radiations

GIULIO RACAH* Institute for Advanced Study, Princeton, New Jersey (Received August 6, 1951)

Using the algebra of tensor operators a closed expression is obtained for the most general case of angular correlation. The structure of the correlation function is explained by means of semiclassical considerations.

`HE theory of the angular correlation of successive nuclear radiations has been treated recently by many authors, and rather formidable tables of results pertaining to various particular cases have been published.1-5

It is the purpose of this paper to show that the problem is much simpler than it seems, both from the algebraical and from the geometrical point of view. The most general case is here treated in a very natural and straightforward manner by the general methods of the algebra of tensor operators;⁶ using an appropriate expansion of the correlation function the final results are expressed in a simple closed form, and lead to a semiclassical geometrical interpretation.

The interaction hamiltonian for the emission of a particle along the direction of the z-axis may be expanded in the form,

$$H = \sum_{LM} \alpha_{LM}(\mathbf{A}_i) T_M^{(L)}(\mathbf{X}_i), \qquad (1)$$

where the $T_{M}^{(L)}(\mathbf{X}_{i})$ are the components of irreducible tensor operators of degree L which operate on the nucleus, and the $\alpha_{LM}(\mathbf{A}_i)$ are functions of the variables associated with the description of the emitted particles.7 If the emitted particle carries off a definite angular momentum (in particular if the emission corresponds to a definite multipole order), all $T_M^{(L)}$ vanish except for a given L; when mixtures of multipoles are considered, $T_M^{(L)}$ with different values of L will be different from zero.

The *M*-dependence of the α_{LM} is characteristic of the polarization properties of the emitted particles. Thus for α -particles only α_{L0} is different from zero, for γ -rays only $\alpha_{L,\pm 1}$ (and then the ratio $\alpha_{L1}/\alpha_{L,-1}$ depends on the polarization), for β -particles the α_{LM} depend on the energy of the electron, the direction of emission of the neutrino, etc.

For a particle emitted in a direction other than the z-axis we shall introduce a new system of axes $\xi \eta \zeta$, with the ζ -axis along the direction of emission, and come back

to the old axes using the fundamental group-theoretical property of the $T_M^{(L)}$ of transforming according to the (2L+1)-dimensional irreducible representation $D^{(L)}$ of the three-dimensional rotation group:

$$\bar{H} = \sum_{LM\mu} \alpha_{LM} T_{\mu}{}^{(L)} D^{(L)} (\alpha \beta \gamma)_{\mu M}.$$
⁽²⁾

The probability for the transition from a nuclear state of spin j_1 to a state of spin j with the emission of a particle in the direction of the z-axis and for a successive transition to a state of spin j_2 with the emission of a particle in the direction of the ζ -axis is given by⁸

$$W = \sum_{m_1mm'm_2} (j_1m_1 | H_1 | jm)^* (j_1m_1 | H_1 | jm') \times (jm | \bar{H}_2 | j_2m_2)^* (jm' | \bar{H}_2 | j_2m_2); \quad (3)$$

introducing (1) and (2) into (3) we obtain

$$W = \sum_{LMm\mu} \alpha_{L_{1}M_{1}}^{*} \alpha_{L_{1'M_{1'}}\alpha_{L_{2}M_{2}}}^{*} \alpha_{L_{2'}M_{2'}} \times (j_{1}m_{1} | T_{M_{1}}^{(L_{1})} | jm)^{*} (j_{1}m_{1} | T_{M_{1'}}^{(L_{1'})} | jm') \times (jm | T_{\mu}^{(L_{2})} | j_{2}m_{2})^{*} (jm' | T_{\mu'}^{(L_{2'})} | j_{2}m_{2}) \times D^{(L_{2})} (\alpha\beta\gamma)_{\mu M_{2}}^{*} D^{(L_{2'})} (\alpha\beta\gamma)_{\mu'M_{2'}}.$$
 (4)

The relation⁹

$$D^{(L_2)}(\alpha\beta\gamma)_{\mu M_2}^{*} = (-)^{M_2 - \mu} D^{(L_2)}(\alpha\beta\gamma)_{-\mu, -M_2} \quad (5)$$

and the fundamental formula¹⁰ for the reduction of the product of the two representations $D^{(L_2)}$ and $D^{(L_2')}$ vield

$$D^{(L_{2})}(\alpha\beta\gamma)_{\mu M_{2}} * D^{(L_{2}')}(\alpha\beta\gamma)_{\mu'M_{2}'} = \sum_{k\rho\sigma} (-)^{M_{2}-\mu} (L_{2} - \mu L_{2}'\mu' | L_{2}L_{2}'k\rho) \times D^{(k)}(\alpha\beta\gamma)_{\rho\sigma} (L_{2}L_{2}'k\sigma | L_{2} - M_{2}L_{2}'M_{2}'); \quad (6)$$

introducing it in (4) and using the expression II(29) for the matrix elements of $T_M^{(L)}$ and the expression II(16') for $(L_2 - \mu L_2' \mu' | L_2 L_2' k \rho)$, we get

$$W = \sum_{LMm\mu k\rho\sigma} \alpha_{L_1M_1} * \alpha_{L_1'M_1'} \alpha_{L_2M_2} * \alpha_{L_2'M_2'} \times (j_1 \| T^{(L_1)} \| j) * (j_1 \| T^{(L_1')} \| j) (j \| T^{(L_2)} \| j_2) * \\ \times (j \| T^{(L_{2'})} \| j_2) (-)^{m'-m+M_2-\mu+k+\rho} (2k+1)! \\ \times V(j_1jL_1; -m_1mM_1) V(j_1jL_1'; -m_1m'M_1') \\ \times V(jj_2L_2; -mm_2\mu) V(jj_2L_2'; -m'm_2\mu') \\ \times V(L_2L_2'k; -\mu\mu' -\rho) D^{(k)} (\alpha\beta\gamma)_{\rho\sigma} \\ \times (L_2L_2'k\sigma | L_2 - M_2L_2'M_2').$$
(7)

^{*} On sabbatical leave from the Hebrew University, Jerusalem, Israel.

¹D. R. Hamilton, Phys. Rev. 58, 122 (1940); and 74, 782 (1948). ²J. W. Gardner, Proc. Phys. Soc. (London) A62, 763 (1949); and A64, 238 (1951).

^a D. L. Falkoff and G. E. Uhlenbeck, Phys. Rev. **76**, 1639 (1949). ⁴ D. L. Falkoff and G. E. Uhlenbeck, Phys. Rev. **79**, 323 (1950).

I. Zines, Phys. Rev. 80, 386 (1950).
 G. Racah, Phys. Rev. 62, 438 (1942) §3; this paper will be

referred to as II. ⁷ As the z-axis has been chosen along the direction of emission the α_{LM} are no more tensors as in reference 4, Eq. (8).

⁸ Reference 1, Eq. (10); reference 5, Eq. (1). ⁹ This relation follows from E. Wigner, *Gruppentheorie* (Friedrich Vieweg and Sohn, Braunschweig, 1931), introducing in Eq.

⁽²⁷⁾ of p. 180 a new summation index $k' = k + \mu - \nu$. ¹⁰ Reference 9, p. 203, Eq. (16a).

The summation over the *m*'s and μ 's may be performed¹¹ by using the symmetry properties II(19) of the V's and applying twice II(41); we obtain, finally,

$$W(\alpha\beta\gamma) = \sum_{L_{1}L_{1}'L_{2}L_{2}'k_{\rho\sigma}} (j_{1}||T^{(L_{1})}||j)^{*}(j_{1}||T^{(L_{1}')}||j)$$

$$\times (j||T^{(L_{2})}||j_{2})^{*}(j||T^{(L_{2}')}||j_{2})(-)^{j_{1}+L_{1}-j_{2}-L_{2}+k}$$

$$\times c_{k\rho}(L_{1}'L_{1})^{*}c_{k\sigma}(L_{2}L_{2}')W(jL_{1}jL_{1}';j_{1}k)$$

$$\times W(jL_{2}jL_{2}';j_{2}k)D^{(k)}(\alpha\beta\gamma)_{\rho\sigma}, \quad (8)$$

where the W's are defined by II(36') and

$$c_{k\tau}(LL') = \sum_{MM'} (-)^{L-M} \alpha_{LM}^*(\mathbf{A}_i) \alpha_{L'M'}(\mathbf{A}_i) \\ \times (L - ML'M' | LL'k\tau). \quad (9)$$

If some of the characteristics of the emitted radiations (as polarization, direction of the neutrino, etc.) are not observed, the expressions of the $c_{k\tau}$ contain also an averaging over those A_i which are not observed.

Equation (8) seems to be still complicated, as there is a summation over seven variables; but this complication is only apparent, and is due to the fact that the equation is very general and holds for any kind of radiations with any kind of polarization and multipole mixture. If there is no multipole mixture, the summation over the L's reduces to one term; if a radiation is nonpolarized or circularly polarized, the corresponding $c_{k\tau}$ vanish for $\tau \neq 0$; if both radiations are nonpolarized or circularly polarized, only

$$D^{(k)}(\alpha\beta\gamma)_{00} = P_k(\cos\beta) \tag{9}$$

will appear in the expansion of $W(\beta)$. If one of the radiations is nonpolarized or linearly polarized, the coefficients of $D^{(k)}{}_{\rho\sigma}$ vanish for odd k; but if both the outgoing radiations are circularly or elliptically polarized, also the $D^{(k)}{}_{\rho\sigma}$ with odd k will appear in the correlation function.¹²

Equation (8) shows that the natural expansion of the correlation function is not in powers of the cosines of the angles, but in terms of $D^{(k)}{}_{\rho\sigma}$ (or legendre polynomials for nonpolarized particles), as the coefficients of this expansion break up in many independent factors. In order to understand the physical and geometrical meaning of these different factors, we shall consider the angular correlation of two γ -rays of given multipole order from a semiclassical point of view.¹³

If we assume as z-axis the direction of emission of the first quantum, and as xz-plane its polarization plane, the probability that the angular momentum L_1 carried off by the quantum has a direction $(\beta_1\alpha_1)$ will be a certain function $f_{L_1}(\beta_1\alpha_1)$; if the quantum is nonpolarized or circularly polarized, f_{L_1} will be independent of α_1 . To this emission corresponds a transition of the nucleus from a state of spin j_1 to a state of spin j; the directions of \mathbf{j}_1 and \mathbf{j} are not determined, but, owing to the conservation of the total angular momentum, the angle β_2 between \mathbf{L}_1 and \mathbf{j} is fixed by the values of L_1 , j_1 and j_1 .

Also for the second quantum, which we assume to be emitted along the ζ -axis and to be polarized in the ξ ;-plane, the angle β_3 between **j** and **L**₂ is known, and the probability that \mathbf{L}_2 has a direction $(\beta_4\gamma_4)$ with respect to the $\xi\eta\zeta$ -system will be given by $f_{L_2}(\beta_4\gamma_4)$.

We perform now the rotation $R(\alpha\beta\gamma)$ from the xyzsystem to the $\xi\eta\zeta$ -system in four steps, considering three intermediate systems with the polar axes along the directions of L_1 , j and L_2 ; then

$$R(\alpha\beta\gamma) = R(\alpha_1\beta_10)R(\alpha_2\beta_20)R(\alpha_3\beta_30)R(\alpha_4 - \beta_4 - \gamma_4), (10)$$

and

$$D^{(k)}(\alpha\beta\gamma)_{\rho\sigma} = \sum_{\lambda\mu\nu} D^{(k)}(\alpha_1\beta_10)_{\rho\lambda} D^{(k)}(\alpha_2\beta_20)_{\lambda\mu}$$
$$\times D^{(k)}(\alpha_3\beta_30)_{\mu\nu} D^{(k)}(\alpha_4 - \beta_4 - \gamma_4)_{\nu\sigma}. \quad (11)$$

We are interested in the coefficients of the expansion

$$W(\alpha\beta\gamma) = \sum_{k\rho\sigma} w_{k\rho\sigma} D^{(k)}(\alpha\beta\gamma)_{\rho\sigma}: \qquad (12)$$

as $w_{k\rho\sigma}$ is proportional to the mean value of $W(\alpha\beta\gamma)$ $\times D^{(k)}(\alpha\beta\gamma)_{\rho\sigma}^{*}$, and $W(\alpha\beta\gamma)$ is proportional to $f_{L_1}(\beta_1\alpha_1)$ $\times f_{L_2}(\beta_4 \gamma_4)$, we get from (11) that

$$\begin{split} w_{k\rho\sigma} \sim & \sum_{\lambda\mu\nu} [f_{L_1}(\beta_1\alpha_1)D^{(k)}(\alpha_1\beta_10)_{\rho\lambda}^*]_{Av} \\ \times [D^{(k)}(\alpha_2\beta_20)_{\lambda\mu}^*]_{Av} [D^{(k)}(\alpha_3\beta_30)_{\mu\nu}^*]_{Av} \\ \times [D^{(k)}(\alpha_4 - \beta_4 - \gamma_4)_{\nu\sigma}^* f_{L_2}(\beta_4\gamma_4)]_{Av}. \end{split}$$
(13)

As α_2 , α_3 , and α_4 are undetermined, the averaging over these angles will give nonvanishing results only for $\lambda = \mu = \nu = 0$, and owing to (9), and to the relations,

$$D^{(k)}(\alpha\beta\gamma)_{\rho 0} = \left[4\pi/(2k+1)\right]^{\frac{1}{2}} Y_{k}^{\rho}(\beta\alpha)^{*} D^{(k)}(\alpha -\beta -\gamma)_{0\sigma} = \left[4\pi/(2k+1)\right]^{\frac{1}{2}} Y_{k}^{\sigma}(\beta\gamma),$$
(14)

we get

$$w_{k\rho\sigma} \sim [f_{L_1}(\beta_1\alpha_1)Y_k{}^{\rho}(\beta_1\alpha_1)]_{\text{Av}}P_k(\cos\beta_2)P_k(\cos\beta_3) \\ \times [f_{L_2}(\beta_4\gamma_4)Y_k{}^{\sigma}(\beta_4\gamma_4)^*]_{\text{Av}}.$$
(15)

Comparing (15) with (8) we see that, apart from normalization and phase factors, $W(jL_ijL_i; j_ik)$ is the quantum-mechanical expression for $P_k(\cos \mathbf{j} \mathbf{L}_i)$, and the $c_{k\tau}$ are the quantum-mechanical expressions for the coefficients of the expansion in a series of spherical harmonics of the correlation function between the directions of the outgoing particle and the angular momentum carried off by it.

In conclusion, we wish to point out that our calculations do not "involve an unnecessary duplication of effort,"14 even for the cases which were already calculated. First of all a closed formula allows a deeper insight into the geometrical and physical reasons for the

¹¹ For a particular case see reference 2. ¹² Actually, for the essentially equivalent problem of the resonance radiation, D. R. Hamilton, Astrophys. J. 106, 457 (1947), obtained terms involving the first power of $\cos\theta$ in the expression of the correlation of the parameters measuring the circular polarization.

¹³ The author is indebted to Dr. U. Fano for many discussions on this point.

¹⁴ D. L. Falkoff, Phys. Rev. 82, 99 (1951).

angular correlations and the structure of the correlation function. Secondly, the use of Eq. (8) is simpler than the use of the formulas and tables previously published, as the expansion in powers of the cosines of the angles is not the natural one, and makes formulas and tables much more complicated than necessary. For the use of Eq. (8) it is sufficient to tabulate separately the values of $c_{k\tau}(LL')$ for the different kinds of radiations, and the values of $W(jLjL'; j_ik)$, which are independent of the kind of radiations, and to combine them according to necessity.¹⁵ Tables of this kind are being prepared for publication.¹⁶

The author wishes to express his gratitude to Professor J. R. Oppenheimer for the generous hospitality in the stimulating atmosphere of the Institute for Advanced Study, and to Professor M. Fierz who suggested that he deal with this problem.

¹⁵ Similar results have been obtained independently by S. P. Lloyd, Thesis, University of Illinois (1951); Phys. Rev. 80, 118 (1950).

¹⁶ J. M. Blatt and L. C. Biedenharn, Phys. Rev. 82, 123 (1951).

PHYSICAL REVIEW

VOLUME 84, NUMBER 5

DECEMBER 1, 1951

Neutron Scattering and Polarization by Ferromagnetic Materials

C. G. SHULL, E. O. WOLLAN, AND W. C. KOEHLER Oak Ridge National Laboratory, Oak Ridge, Tennessee (Received August 20, 1951)

Neutron diffraction studies are reported on a series of magnetized and unmagnetized ferromagnetic materials. The diffraction patterns for unmagnetized, polycrystalline samples of Fe and Co are found to possess both nuclear and magnetic components with the latter in agreement with the magnetic scattering theory with respect both to intensity of scattering and form factor angular variation. Studies on the magnetic structure of Fe₃O₄ are shown to strongly support Néel's proposed ferrimagnetic structure. Predictions of the theory regarding intensity effects upon sample magnetization are fully confirmed and the Schwinger-Halpern-Johnson formulation of the interaction function between the neutron's magnetic moment and the internal fields in a ferromagnet is substantiated. A pronounced variation of intensity around the Debye ring in the diffraction pattern for a magnetized sample is found. Neutron polarization effects in the Bragg scattered beams from magnetized crystals of Fe and Fe₃O₄ have been studied and it is shown that very highly polarized beams are obtained for certain reflections. This method of monochromatic beam polarization is found to compare very favorably with other methods with respect to polarization value, beam intensity, and ease of obtainment.

INTRODUCTION

HE theory of the scattering and polarization of neutrons by ferromagnetic substances has been given a very general treatment by Halpern and coworkers.1 Up to the present time, experimentation in this field has centered upon studies of the single and double transmission effect² and more recently upon studies of critical reflection from magnetized mirrors.³

The present report deals with studies of the intensity distribution and the polarization of the scattered neutron radiation from both unmagnetized and magnetized ferromagnetic substances. These studies give information on the form factor dependence of magnetic scattering, on the basic nature of the neutron's magnetic interaction and on the magnetic structure existing in certain ferromagnetics, viz., the spatial distribution of the various magnetic ions within the ferromagnetic lattice. A previous report⁴ has discussed neutron scattering by paramagnetic and antiferromagnetic lattices and brief reports of some aspects of the present work concerning the neutron's magnetic interaction and the polarization phenomena have been given in the literature.5

The general expression for the differential scattering cross section of a magnetic ion has been given by Halpern and Johnson¹ (H-J) as

$$F^2 = C^2 + 2CD\mathbf{q} \cdot \mathbf{\lambda} + D^2 q^2 \tag{1}$$

where C is the nuclear scattering amplitude, D the magnetic scattering amplitude, λ a unit vector describing the polarization state of the neutron being scattered,

$$\mathbf{q} = \mathbf{e}(\mathbf{e} \cdot \mathbf{\kappa}) - \mathbf{\kappa}$$
 and $q^2 = 1 - (\mathbf{e} \cdot \mathbf{\kappa})^2$ (2)

where \mathbf{e} is the unit scattering vector and $\mathbf{\kappa}$ a unit vector parallel to the magnetic moment vector of the magnetic ion. The magnetic scattering amplitude D is given by

$$D = (e^2/mc^2)\gamma Sf = 0.539 \times 10^{-12} Sf \text{ cm}$$
(3)

¹O. Halpern and M. H. Johnson, Phys. Rev. 55, 898 (1939). O. Halpern and T. Holstein, Phys. Rev. 59, 960 (1941). Halpern, Hamermesh, and Johnson, Phys. Rev. 59, 981 (1941)

² Hughes, Wallace, and Holtzman, Phys. Rev. **73**, 1277 (1948). Fleeman, Nicodemus, and Staub, Phys. Rev. **76**, 1774 (1949). Burgy, Hughes, Wallace, Heller, and Woolf, Phys. Rev. **80**, 953 (1950)

³ D. J. Hughes and M. T. Burgy, Phys. Rev. 81, 498 (1951).

⁴ Shull, Strauser, and Wollan, Phys. Rev. **83**, 333 (1951). ⁵ Shull, Wollan, and Strauser, Phys. Rev. **81**, 483 (1951). C. G. Shull, Phys. Rev. 81, 626 (1951).