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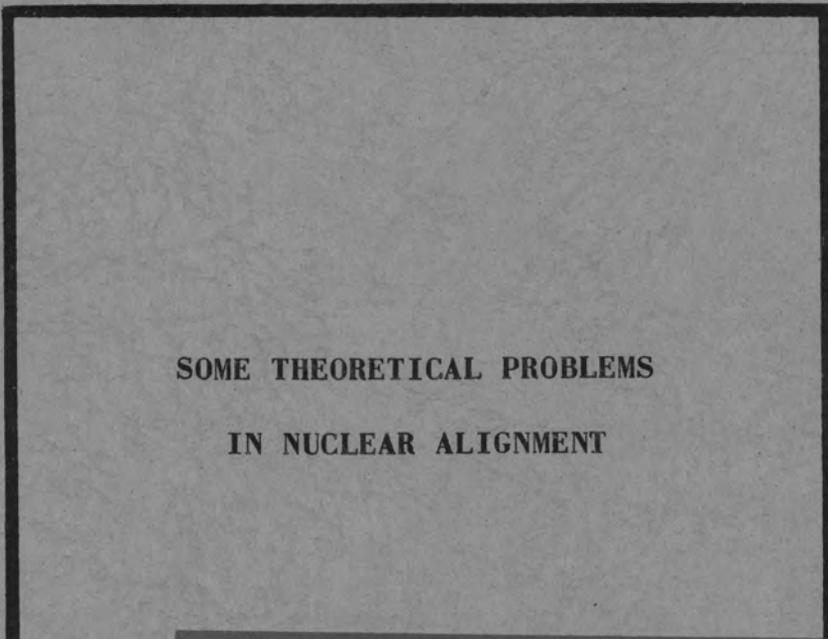


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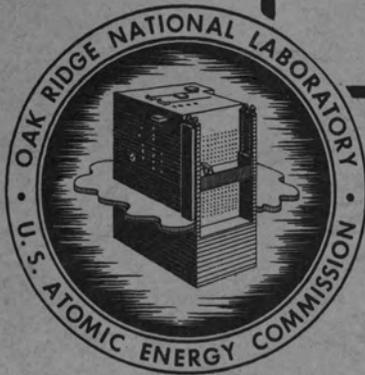
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IN NUCLEAR ALIGNMENT

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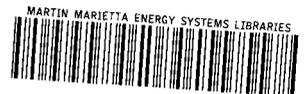
SOME THEORETICAL PROBLEMS IN NUCLEAR ALIGNMENT

J. M. Jauch
A. Simon

Date Issued: 1968

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SOME THEORETICAL PROBLEMS IN NUCLEAR ALIGNMENT

J. M. Jauch and A. Simon

I. INTRODUCTION

The direct method of nuclear alignment leads to an appreciable effect only if the magnetic field H and the temperature T are of such magnitude that

$$\frac{H}{T} \sim 10^7 \text{ gauss deg}^{-1} \quad (1.1)$$

It was pointed out by Rose and Gorter¹ that nuclear alignment could be obtained for nuclei of paramagnetic ions under much more favorable conditions if there exists a h.f. coupling between the ion and the nucleus.

A search for alignment in the case of the Mn nucleus has not yet given any positive results. We shall therefore examine once more the theoretical foundations of the effect.

In the course of this work we shall pay special attention to an objection to the theoretical prediction of nuclear alignment with h.f. coupling raised by Bleaney². Bleaney believes that the nuclear polarization (or alignment) requires magnetic fields so strong that the electronic splitting is large compared to the h.f. splitting. He also believes that the dilution of the salt must be so large as to reduce the line width of the magnetic levels to an amount less than the h.f. spread. Both these statements are wrong and lead to much too restrictive conditions on the experiment. Rose and Simon³ have recently answered these objections. We shall give a

slightly more rigorous derivation of the same result and develop a general method to calculate higher order effects in this type of problem. The general theory is then used to evaluate explicitly the order of magnitude of the correction Bleaney visualized in his appraisal of the method and it is found to be negligible.

A difficulty which will reduce the expected effect results from an entirely different quarter, however. This is due to the contribution to absorption (in the thermal neutron region) of the level with the other value of the resultant spin. The data available at present seems to indicate a possible reduction of the expected effect in a neutron absorption experiment (with Mn) by about a factor of two.

II. THE h.f. COUPLING THEORY

1. The Interaction Operator

Abraham and Pryce⁴ give the following general expression for the h.f. interaction energy.

$$W_{hf} = 2r\beta\beta_N \left\langle \frac{1}{r^3} \right\rangle \left\{ \underline{L} \cdot \underline{I} + (\sum L(L+1) - \kappa) \underline{S} \cdot \underline{I} - \frac{3}{2} \sum (\underline{L} \cdot \underline{S})(\underline{L} \cdot \underline{I}) - \frac{3}{2} \sum (\underline{L} \cdot \underline{I})(\underline{L} \cdot \underline{S}) \right\} \quad (2.1)$$

This is correct only for the submatrix of W referring to the $(2L+1)(2S+1)$ dimensional subspace of total orbital angular momentum L and spin S. L, S

are the vector operators for these quantities. This is still correct if there is spin-orbit interaction which splits the multiplets or a strong local electric field which quenches the orbital motion.

The various quantities entering in the expression (2.1) are:

μ the nuclear g-factor

$$\beta_N = \frac{e\hbar}{2Mc} \quad \text{nuclear magneton}$$

$$\beta = \frac{e\hbar}{2mc} \quad \text{Bohr magneton}$$

$$\left\langle \frac{1}{r^3} \right\rangle \quad \text{averaged for the 3d - electron shell}$$

$\frac{1}{2} \mu$ a numerical factor depending on the configuration and the term. For the normal states (i.e., the states of maximum S and L compatible with the exclusion principle) which are ground terms according to Hund's rule can be given by

$$\frac{1}{2} \mu = \frac{2l+1-4s}{5(2l-1)(2l+3)(2l-1)} \quad (l=2) \quad (2.2)$$

κ a numerical factor arising from the admixture of other configurations.

For the Mn^{++} - ion we have the term $6S$ for the ground state. For this term the operator L is zero and consequently W_{hf} reduces in this case to

$$W_{hf} = -2\mu\beta_N\beta \left\langle \frac{1}{r^3} \right\rangle \kappa \underline{S} \cdot \underline{I} \quad (2.3)$$

In order to obtain a clearer understanding of the origin of the terms in (2.1) and especially of the term (2.3) we shall give an elementary derivation starting from first principles.

The Hamiltonian for an n-electron system in a magnetic field $\underline{H} = \nabla \times \underline{A}$ is given by

$$\mathcal{H} = \sum_{v=1}^n \frac{1}{2m} \left(\underline{p}^{(v)} + \frac{e}{c} \underline{A}(\underline{x}^{(v)}) \right)^2 + 2\beta \sum_{v=1}^n \left(\underline{H}(\underline{x}^{(v)}) \cdot \underline{S}^{(v)} \right) + V \quad (2.4)$$

Here we have included in V all the terms arising from the Coulomb energy of the nuclear charge, the Coulomb interaction of the electrons, the spin-orbit and spin-spin interaction and the potential energy due to the crystalline electric field. We are only interested in the linear terms in \underline{A} and \underline{H} which arise from the magnetic moment of the nucleus, because these terms lead to the h.f. coupling term (2.1). The quadratic term in \underline{A} is entirely negligible as an order of magnitude estimate easily shows. The vector potential $\underline{A}(\underline{x})$ at the point \underline{x} due to a magnetic dipole $\underline{\mu} = \delta\beta_N \underline{I}$ located at the origin is given by

$$\underline{A}(\underline{x}) = \frac{1}{r^3} (\underline{\mu} \times \underline{x}) = -\underline{\mu} \times \nabla \left(\frac{1}{r} \right) \quad (2.5)$$

Thus

$$W_{hf} = \frac{e}{mc} \sum_{v=1}^n \underline{A}(\underline{x}^{(v)}) \cdot \underline{p}^{(v)} + 2\beta \sum_{v=1}^n \left(\nabla \times \underline{A}(\underline{x}^{(v)}) \cdot \underline{S}^{(v)} \right) \quad (2.6)$$

The first term of (2.6) is easily evaluated. Substituting (2.5) and using the vector identity

$$(\underline{a} \times \underline{b}) \cdot \underline{c} = (\underline{b} \times \underline{c}) \cdot \underline{a}$$

we obtain

$$\begin{aligned} \frac{e}{mc} \sum_{v=1}^n (\underline{A}(\underline{x}^{(v)}) \cdot \underline{p}^{(v)}) &= \frac{e}{mc} \sum_{v=1}^n \frac{1}{r_v^3} (\underline{\mu} \times \underline{x}^{(v)}) \cdot \underline{p}^{(v)} \\ &= \frac{e}{mc} \sum_{v=1}^n \frac{1}{r_v^3} (\underline{x}^{(v)} \times \underline{p}) \cdot \underline{\mu} \\ &= \frac{e\hbar}{mc} \sum_{v=1}^n \frac{1}{r_v^3} (\underline{L}^{(v)} \cdot \underline{\mu}) \end{aligned} \quad (2.7)$$

We now replace $\frac{1}{r_v^3}$ by some average over the d-shell (only d-electrons contribute to the sum since all the others are in closed shells). In this way we obtain finally

$$\mathcal{K}_1 = \frac{e}{mc} \sum_{v=1}^n (\underline{A}(\underline{x}^{(v)}) \cdot \underline{p}^{(v)}) = 2\hbar\beta_N\beta \langle \frac{1}{r^3} \rangle \underline{L} \cdot \underline{I} \quad (2.8)$$

with

$$\underline{L} = \sum_{v=1}^n \underline{L}^{(v)}$$

This accounts for the first term in (2.1).

The remaining terms in (2.1) arise all from the second term in (2.6) which we must examine now more carefully. The evaluation of this term is somewhat intricate on account of the singularity of the vector potential at

the origin. The singularity at the origin gives the singular contribution for S-electrons discovered by Fermi⁵ and contained in the κ -term in (2.1). If we disregard the singularity for a moment we obtain by differentiation

$$\mathcal{H}_2 = 2\delta\beta_N\beta \left\langle \frac{1}{r^3} \right\rangle \sum_{\nu=1}^n \left\{ \frac{3(\underline{x}^{(\nu)} \cdot \underline{s}^{(\nu)})(\underline{x}^{(\nu)} \cdot \underline{r})}{r_\nu^2} - (\underline{s}^{(\nu)} \cdot \underline{r}) \right\} \quad (2.9)$$

In order to exhibit the singularity we write $\underline{H}(\underline{x})$ in a Fourier representation. Since

$$\frac{1}{r} = \frac{1}{2\pi^2} \int \frac{1}{k^2} e^{i\underline{k} \cdot \underline{x}} d^3k \quad (2.10)$$

Thus

$$\begin{aligned} -\nabla \times (\underline{\mu} \times \nabla \left(\frac{1}{r} \right)) &= \frac{1}{2\pi^2} \int \frac{\underline{k} \times (\underline{\mu} \times \underline{k})}{k^2} e^{i\underline{k} \cdot \underline{x}} d^3k \\ &= \frac{1}{2\pi^2} \int \left(\underline{\mu} - \frac{1}{k^2} (\underline{k} \cdot \underline{\mu}) \underline{k} \right) e^{i\underline{k} \cdot \underline{x}} d^3k \end{aligned} \quad (2.11)$$

Since (2.11) is homogeneous of order zero in k the singularity at the origin is of the δ -type. We can thus determine it by evaluating the integral at $\underline{x} = 0$. After the angle integration we obtain for this singular part of the integrand

$$\underline{\mu} \left(1 - \frac{1}{3} \right) = \frac{2}{3} \underline{\mu}$$

Thus the singularity is

$$\frac{1}{2\pi^2} \frac{2}{3} \mu \int e^{i\mathbf{k} \cdot \mathbf{x}} d^3k = \frac{8\pi}{3} \mu \delta(\mathbf{x}) \quad (2.12)$$

$$\mathcal{H}_3 = 2\gamma\beta\beta_N \frac{8\pi}{3} \sum_{\nu=1}^n \delta(x^{(\nu)}) (\underline{S}^{(\nu)} \cdot \underline{I}) \quad (2.13)$$

The further simplification of these two terms which reduces them to the desired form can be carried through by an application of Eckarts theorem⁶.

We write for (2.9)

$$\mathcal{H}_2 = P \sum_{\nu, ik} d_{ik}^{(\nu)} S_i^{(\nu)} I_k \quad (2.14)$$

with

$$P = 2\gamma\beta\beta_N \left\langle \frac{1}{r^3} \right\rangle$$

$$d_{ik}^{(\nu)} = 3 \frac{\sum_i^{(\nu)} \sum_k^{(\nu)}}{\sum_i^{(\nu)} \sum_k^{(\nu)}} - \delta_{ik} \quad \left(\frac{\sum_i^{(\nu)}}{\sum_i^{(\nu)}} = \frac{x_i^{(\nu)}}{r_\nu} \right)$$

Eckarts theorem for this case says then that the matrix elements for $d_{ik}^{(\nu)} S_i^{(\nu)}$ in the subspace $(M_L M_S)$ of the term (L, S) are proportional to

$D_{ik} S_i$ where

$$D_{ik} = \frac{3}{2} (L_i L_k + L_k L_i) - \delta_{ik} L(L+1) \quad (2.15)$$

and
$$S_i = \sum_{v=1}^n S_i^{(v)}$$

Thus

$$d_{ik}^{(v)} S_i^{(v)} = c D_{ik} S_i \tag{2.16}$$

the proportionality factor c will depend on L, S and the configuration out of which the term L, S arises. In order to determine it, we need only to take one component say $i = k = 3$ and only one matrix element $M_L = L, M_S = S$, say.

Since

$$\begin{aligned} \langle D_{33} S_3 \rangle_{L,S} &= (3L^2 - L(L+1)) S \\ &= L(2L-1) S \end{aligned} \tag{2.17}$$

We have

$$c = \sum_{v=1}^n \frac{1}{S L(2L-1)} \langle (3L^2 S^{(v)} - 1) S_3^{(v)} \rangle_{L,S} \tag{2.18}$$

In order to evaluate this term we must use the expression for the electronic wave functions. It is sufficient to consider only the part which belongs to the unfilled shell. In our problem there are either 3d-electrons (transition element) or 4f-electron (rare earths).

Each electronic wave function may be characterized by a set of quantum numbers $m^{(v)} m_s^{(v)}$ ($v = 1, \dots, n$) where m is the z component of the orbital angular momentum and m_s the spin component. The whole wave function is written as the anti-symmetrical product (Slater determinant) of one body wave functions and symbolically written as a collection of the quantum numbers

$$\psi = \left(m^{(1)} m_s^{(1)}, m^{(2)} m_s^{(2)}, \dots, m^{(n)}, m_s^{(n)} \right) \tag{2.19}$$

We prove first that the expression $\langle (3\cos^2\vartheta^{(v)} - 1) S_3^{(v)} \rangle_{L,S}$ is independent of v and the same for the shell less than half filled as for the shell more than half filled with the number of holes in the second case equal to the number of electrons in the first. We shall limit our discussion to the case of the normal terms.

In this case a less than half filled shell, such as $3d^4$ would be given by

$$\psi_{LS} = (2^+ 1^+ 0^+ - 1^+)$$

and it represents the $M_L = L, M_S = S$ component of the term 5D . For the matrix element in question we obtain then

$$\begin{aligned} & (\psi_{LS}, (3\cos^2\vartheta^{(v)} - 1) S_3^{(v)} \psi_{LS}) \\ &= \frac{1}{2n} \sum_{m=\ell-(n-1)}^{\ell} \langle 3\cos^2\vartheta - 1 \rangle_m = \\ & \quad - \frac{1}{2n} \sum_{m=\ell-(n-1)}^{\ell} \frac{3m^2 - \ell(\ell+1)}{(2\ell-1)\ell} \frac{2\ell}{2\ell+3} \end{aligned} \tag{2.20}$$

For the corresponding matrix element for the more than half filled shell we obtain similarly

$$\psi'_{LS} = (2^+ 1^+ 0^+ - 1^+ - 2^+ 2^-)$$

$$\begin{aligned}
 (\psi'_{LS}, (3 \cos^2 \vartheta^{(v)} - 1) S_3^{(v)} \psi'_{LS}) &= -\frac{1}{2n'} \sum_{m=-l+n-1}^l \langle 3 \cos^2 \vartheta - 1 \rangle_m \\
 &= -\frac{1}{2n'} \sum_{m=l-(n-1)}^l \frac{3m^2 - l(l+1)}{(2l-1)l} \frac{2l}{2l+3}
 \end{aligned}$$

(2.21)

In the last relation we have made use of

$$\sum_{m=-l}^{+l} \langle 3 \cos^2 \vartheta - 1 \rangle_m = 0$$

The denominators n , respectively n' (= number of electrons in the shell) in (2.20) and (2.21) will just cancel in the summation over v . Thus the two expressions become equal as stated*.

It is then sufficient to evaluate the expression (2.20). The summation over m can at once be carried out to give

$$\begin{aligned}
 -\frac{1}{2} \sum_{m=l-(n-1)}^l \frac{3m^2 - l(l+1)}{(2l-1)l} \frac{2l}{2l+3} \\
 = -\frac{S [(2l+1) - 4S] [(2l+1) - 2S]}{(2l-1)(2l+3)}
 \end{aligned}$$

(2.22)

*NOTE: In the evaluation of this expression it was essential that we included the spin $S_3^{(v)}$. If instead we had taken only the quantity $(3 \cos^2 \vartheta^{(v)} - 1)$ we would have obtained a reversal in sign for the more than half filled shell. This error occurred in an earlier report (ORNL 813, page 37) and led to an erroneous interpretation of the h.f. structure in the Cu^{++} -ion.

$$\begin{aligned} \text{Since } L &= l + (l-1) + \dots + l - 2S + 1 \\ &= S [(2l+1) - 2S] \end{aligned}$$

(2.22) simplifies to

$$-L \frac{2l+1 - 4S}{(2l-1)(2l+3)}$$

and thus we find for C in (2.18)

$$C = - \frac{2l+1 - 4S}{S(2l-1)(2l+3)} = - \xi \quad (2.23)$$

This gives the expression (2.1) without the κ -term. The term involving

κ can be evaluated in the following way. It is obviously connected with the expression (2.13) for \mathcal{K}_3 . Now this expression involves a δ -function and therefore vanishes for any configuration which has only filled S-shells. In order to obtain a non-vanishing contribution for Mn^{++} it is thus necessary to have configurational mixing.

How such configurational mixing can occur may best be illustrated by the example of the Mn^{++} ions. The configuration which is mainly present is $\psi = 3d^5$ giving rise to the ground term $6S$. The nearest configuration belonging to the same term is obtained by the promotion of a 3s electron to a 4s level leading to $\psi' = 3s 3d^5 4s$.

Now there are two states of this type which in the notation of the previous section may be written as

$$\begin{aligned} \psi_a &= (0^+, 2^+, 1^+, 0^+, -1^+, -2^+, 0^-) \\ \psi_b &= (0^-, \underbrace{2^+ 1^+ 0^+ -1^+ -2^+}_{3d}, 0^+) \end{aligned} \quad (2.24)$$

$\begin{matrix} 3s & & 4s \end{matrix}$

We must select from these two functions the linear combination which belongs to the $6S$ -term since the admixture must belong to the same term system. A closer inspection shows that the only function which has this property is

$$\psi' = \frac{1}{\sqrt{2}} (\psi_a - \psi_b) \quad (2.25)$$

Thus we may write for the whole state function

$$\underline{\psi} = \psi + \alpha \psi'$$

where α is a numerical coefficient expressing the amount of admixture. It seems to be very difficult to calculate α directly. Even the sign of it is not easily calculated.

Instead of doing this we shall estimate the order of magnitude of from the amount of h.f. splitting in the Mn^{++} ion to which this example applies. For (2.13) we write again by applying Eckarts theorem

$$\begin{aligned} 2\gamma/\beta_N \frac{p_T}{3} \sum_v \delta(x^{(v)}) (\underline{S}^{(v)} \cdot \underline{I}) \\ = P_c(L, S) \underline{S} \cdot \underline{I} \end{aligned} \quad (2.26)$$

where

$$\kappa = c = \int \left\langle \frac{1}{r_3} \right\rangle \frac{8\pi}{3} \frac{1}{2} \sum_v (\Psi, \delta(x^{(v)}) \Psi) \quad (2.27)$$

To first order in α this is then

$$\kappa = \int \left\langle \frac{1}{r_3} \right\rangle \frac{8\pi}{3} \frac{1}{\sqrt{2}} \psi_{3s}(0) \psi_{4s}(0) \alpha \quad (2.28)$$

In the case of Mn^{++} the constant $A = PC$ is of order $A \sim 0.01 \text{ cm}^{-1}$ which corresponds to $\kappa \sim 0.60$.

In order to obtain from this value of κ we need to know $\psi_{3s}(0)$, $\psi_{4s}(0)$ and $\left\langle \frac{1}{r_3} \right\rangle$. We obtain an order of magnitude idea of the value of these if we take the values of these quantities from the case of neutral Fe^8 which is not too much different from $Mn^{++}(3d^5)$. Here we have

$$\psi_{3s}(0) \psi_{4s}(0) = \frac{180.7}{a_0^3}$$

$$\left\langle \frac{1}{r_3} \right\rangle = 3.3 \times 10^{24} \text{ cm}^{-3}$$

Thus

$$\frac{\psi_{3s}(0) \psi_{4s}(0)}{\left\langle \frac{1}{r_3} \right\rangle} \sim \frac{180.7}{4} \sim 45$$

$$\kappa = \alpha \frac{1}{5} \frac{8\pi}{3} \frac{1}{\sqrt{2}} 45$$

for the case of Mn^{++} $S = 5/2$

$$\kappa \sim 104 \alpha$$

Thus the amount of admixture needed in order to account for the h.f.-splitting Mn^{++} is

$$\alpha = \frac{0.6}{104} \sim \frac{1}{2} \% .$$

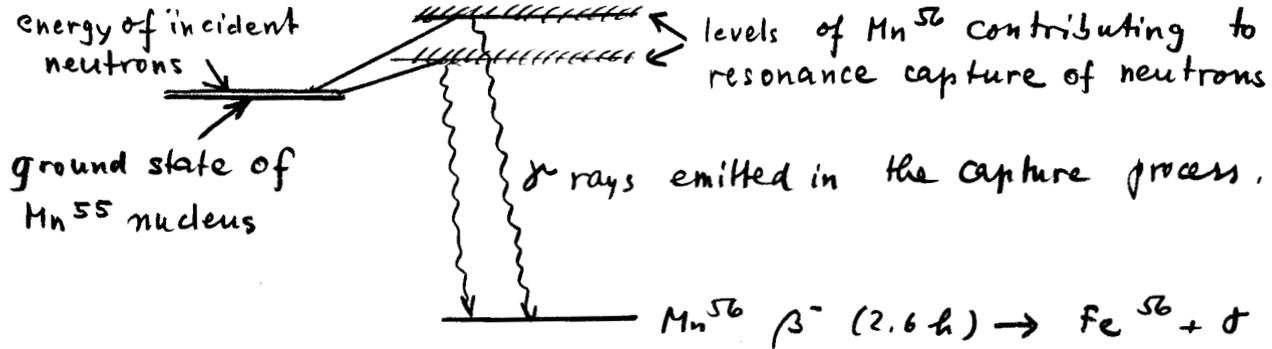
The existence of h.f. interaction in the Mn^{++} -ion is a brilliant confirmation of Fermi's interaction Hamiltonian (2.13). It should be emphasized that the old question as to which type of interaction exists between a neutron and an electron is thereby experimentally settled since it seems clear that the magnetic interaction between a neutron and an electron should in principle be the same as that between a proton or a nucleus and an electron. The interaction operator (2.13) is indeed equivalent with the operator derived by Schwinger⁹ for the neutron problem with the help of a relativistic discussion of the magnetic electron. This expression has now also been confirmed by direct neutron diffraction experiments¹⁰.

III. NEUTRON ABSORPTION BY ALIGNED NUCLEI

1. Absorption by a single nucleus

When a nucleus of spin i absorbs a slow neutron the resulting compound nucleus will have a spin either $j = i \pm \frac{1}{2}$. Thus, for instance, the slow neutron capture in Mn of spin $i = 5/2$ leads to an intermediate compound nucleus of spin either $j = 3$ or $j = 2$. This state then breaks up into a final nucleus and a γ -ray. The final state in this case is the β -radioactive Mn^{56} (2.60 h) which can be detected with standard experimental methods.

Let us denote the capture cross section with σ_j for states ψ_j of total angular momentum $j = i \pm \frac{1}{2}$. We ask



for the absorption cross section $\sigma(\psi)$ when the spin states of neutron and nucleus is given by a state vector ψ . We have then

$$\sigma(\psi) = \sum_j \sigma_j |c^j|^2 \quad (3.1)$$

where

$$\psi = \sum_j c^j \psi_j \quad (3.2)$$

Of particular interest is the case when the state ψ_{ms} is the simultaneous eigenstate of I_3 and S_3 . Let these eigenfunctions be ω_m, χ_s . Thus

$$\begin{aligned} I_3 \omega_m &= m \omega_m & (m = -i, -i+1, \dots, +i) \\ S_3 \chi_s &= s \chi_s & (s = \pm \frac{1}{2}) \end{aligned} \quad (3.3)$$

$$\psi_{ms} = \omega_m \chi_s \quad (3.4)$$

Now

$$\omega_m \chi_s = \sum_j c_{ms}^j \psi_j \quad (3.5)$$

where c_{ms}^j are the (normalized) Clebsch-Gordon coefficients. Thus we find

$$\sigma(ms) = \sum_j \sigma_j |c_{ms}^j|^2 \quad (3.6)$$

2. Absorption by a statistical ensemble

The actual experiment of neutron absorption is carried out with a large number of neutron-nucleus systems distributed in a certain way over all possible states. Thus the problem requires a statistical treatment for which the formalism with the density matrix is admirably suited.

We consider thus an assembly of N identical systems distinguished by an index α ($\alpha = 1, \dots, N$). Let $\psi^{(\alpha)}$ be the spin-state of the α th system (consisting of a neutron and nucleus). For such a state we may always write

$$\psi^{(\alpha)} = \sum_{ms} a_{(\alpha)}(ms) \psi_{ms} \quad (3.7)$$

For the cross section of an individual state α we obtain according to the foregoing

$$\sigma(\alpha) = \sum_{\substack{ms \\ m's'j}} \sigma_j a_{(\alpha)}^*(ms) a_{(\alpha)}(m's') c_{ms}^j c_{m's'}^j \quad (3.8)$$

and for the average cross section

$$\bar{\sigma} \equiv \frac{1}{N} \sum_{\alpha} \sigma(\alpha) = \sum_{\substack{m_s, \\ m's', j}} \sigma_j c_{m's'}^j (m's' | \rho | m_s) c_{m_s}^j \quad (3.9)$$

where we have introduced

$$(m's' | \rho | m_s) = \frac{1}{N} \sum_{\alpha} a_{(\alpha)}^*(m_s) a_{(\alpha)}(m's') \quad (3.10)$$

the density matrix of the statistical ensemble⁷. Since the neutron and nucleus are in this case statistically independent we may always write

$$(m's' | \rho | m_s) = p_s \delta_{ss'} (m' | \rho | m) \quad (3.11)$$

with

$$\sum_s p_s \equiv p_+ + p_- = 1$$

the quantity

$$f_n \equiv p_+ - p_- \quad (3.12)$$

is the neutron polarization. The problem is thus reduced essentially to the calculation of the density matrix for the nucleus.

3. Nuclear spin coupled with an ion

The state function $\psi^{(\alpha)}$ of one of the members of the statistical assembly may now be written

$$\psi^{(\alpha)} = \sum_j a_{(\alpha)}(M m_s) \phi_M \psi_j c_{m_s}^j \quad (3.13)$$

where ϕ_M is eigenstate of S_3 , the spin operator for the ions.

Going through the same arguments which led to equation (3.9) we obtain

$$\bar{\sigma} = \sum \sigma_j c_{m's}^j (m' | \rho_n | m) c_{ms}^j \rho_s \quad (3.14)$$

where now

$$\rho_s \delta_{ss'} (m' | \rho_n | m) = \frac{1}{N} \sum_{\alpha, M} a_{(\alpha)}^* (Mms) a_{(\alpha)} (Mm's') \quad (3.15)$$

or

$$(m' | \rho_n | m) = \frac{1}{N} \sum_{\alpha, M} a_{(\alpha)}^* (Mm) a_{(\alpha)} (Mm') \quad (3.16)$$

We see from this result, that the expression of the average absorption cross section depends only on the submatrix ρ_n for the nucleus which is obtained by taking the trace of the total ρ with respect to the spin variable of the ion. Thus we shall write

$$\rho_n = h c \rho \quad (3.17)$$

4. Ions coupled by magnetic or exchange interaction

The formulism can be generalized to the case where the ions strongly interact with each other through magnetic or exchange type of coupling. In this case it is no longer permissible to treat the individual systems of ions + nuclei as independent of each other. Instead the whole spin system

consisting of all ions together with their nuclei must be regarded as one individual system of an ensemble of a large number of such identical systems.

The general spin-state of one such system may then be given by

$$\psi^{(\alpha)} = \sum_{(M_1, M_2, \dots, m_1, m_2, \dots)} a_{(\alpha)}(M_1, M_2, \dots, m_1, m_2, \dots) \phi_{M_1, M_2, \dots} \omega_{m_1, m_2, \dots}$$

where

$$\phi_{M_1, M_2, \dots} = \phi_{M_1} \phi_{M_2} \dots$$

$$\omega_{m_1, m_2, \dots} = \omega_{m_1} \omega_{m_2} \dots$$

The cross section for the absorption by the nucleus v is then given by

$$\bar{\sigma} = \sum_j \sigma_j \sum_{\substack{m_v, s \\ m_v'}} c_{m_v', s}^j (m_v' | \rho | m_v) c_{m_v, s}^j p_s \quad (3.18)$$

where

$$(m_v' | \rho | m_v) = \frac{1}{N} \sum_{\substack{\alpha \\ M_1, M_2, \dots \\ m_1, m_2, \dots \\ m_1', m_2', \dots \\ \neq m_v, m_v'}} a_{(\alpha)}^* (M_1, M_2, \dots, m_1, m_2, \dots) a_{(\alpha)} (M_1, M_2, \dots, m_1', m_2', \dots) \times \prod_{\lambda \neq v} \delta_{m_\lambda, m_\lambda'} \quad (3.19)$$

5. Application to the case of a single ion coupled with a nucleus by H.F. interaction and in thermal equilibrium

We make the assumption that a hyperfine coupling exists of the form $\sum_{i=1}^3 A_i \mathbf{S}_i \cdot \mathbf{I}_i$ between nuclear spin \mathbf{I} and ionic spin \mathbf{S} , in an external magnetic field \mathbf{H} . The Hamiltonian for the system is then

$$\mathcal{H} = \sum_{i=1}^3 A_i S_i I_i + g\beta \underline{H} \cdot \underline{S} - g\beta_N \underline{H} \cdot \underline{I} \quad (3.20)$$

The last term can be neglected in comparison with the other terms in this expression. For thermal equilibrium corresponding to a temperature T the density matrix is given by

$$\rho = \frac{1}{Z} e^{-\mathcal{H}/kT} \quad (3.21)$$

where

$$Z = \text{tr} e^{-\mathcal{H}/kT} \quad (3.22)$$

is the partition function of the system.

We need only the submatrix ρ_n of ρ which refers to the nuclear spins defined by

$$\rho_n = \text{tr}_e \rho = \text{tr}_e \frac{1}{Z} e^{-\mathcal{H}/kT}$$

The density matrix contains all the statistical information of the ensemble. In particular it is then easy to calculate the amount of nuclear polarization (first moment of the nuclear spin)

$$\overline{I_3} = \text{tr} (I_3 \rho_n) \quad (3.23)$$

and similarly for the nuclear alignment (second moment of the nuclear spin)

$$\overline{I_3^2} = \text{tr} (I_3^2 \rho_n) \quad (3.24)$$

Thus one has the answer to all the problems if one can evaluate ρ_n .

The matrix elements of ρ_n are in general difficult to evaluate but it is possible to develop a perturbation technique with respect to the parameter $\frac{A}{kT}$.

IV. A PERTURBATION THEORY FOR THE DENSITY MATRIX

1. We wish to calculate the density matrix ρ and its submatrix for the nuclear spins for thermal equilibrium

$$\rho = \frac{1}{Z} e^{-\mathcal{H}/kT} \quad (Z = \text{tr} e^{-\mathcal{H}/kT}) \quad (4.1)$$

We assume the Hamiltonian to be of the form

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1 \quad (4.2)$$

$$\mathcal{H}_0 = \gamma \beta \underline{H} \cdot \underline{S} \quad (4.3)$$

$$\mathcal{H}_1 = F(S) + G(SI) \quad (4.4)$$

\mathcal{H}_1 contains the interaction energy of all the ionic spins (F) and the interaction of the ionic spins with the nuclear spins (G). In order not to

complicate the theory we assume the latter to be isotropic, thus

$$G = \sum G_\nu$$
$$G_\nu = A \left(\underline{S}^{(\nu)}, \underline{I}^{(\nu)} \right) \quad (4.5)$$

This special choice simplifies the theory a little but will not be essential in the following.

We notice that the problem of calculating (4.1) is equivalent to finding the solution of the time dependent Schrodinger equation

$$\dot{\Omega} = \mathcal{H} \Omega \quad (4.6)$$

Let the solution of this equation be

$$\Omega(\tau) = S(\tau) \Omega_0 \quad S(\tau) = e^{\tau \mathcal{H}} \quad (4.7)$$

with

$$S(0) = I \quad (\Omega(0) = \Omega_0) \quad (4.8)$$

Then putting $\tau = -\frac{1}{kT}$ we obtain

$$\rho = \frac{1}{Z} S\left(-\frac{1}{kT}\right) = \frac{1}{Z} e^{-\mathcal{H}/kT} \quad (4.9)$$

Thus the problem is shown to be equivalent to finding the "S-matrix" which belongs to the Hamiltonian \mathcal{H} .

In order to develop a perturbation theory we introduce the "interaction representation" by defining a new state vector

$$\omega(\tau) = e^{-\tau \mathcal{H}_0} \Omega(\tau)$$

$$\Omega(\tau) = e^{\tau \mathcal{H}_0} \omega(\tau) \quad (4.10)$$

Thus by differentiation:

$$\begin{aligned} \dot{\omega}(\tau) &= -\mathcal{H}_0 \omega(\tau) + e^{-\tau \mathcal{H}_0} \dot{\Omega}(\tau) \\ &= -\mathcal{H}_0 \omega(\tau) + e^{-\tau \mathcal{H}_0} (\mathcal{H}_0 + \mathcal{H}_1) e^{\tau \mathcal{H}_0} \omega(\tau) \end{aligned}$$

or

$$\dot{\omega}(\tau) = \mathcal{H}_1(\tau) \omega(\tau) \quad (4.11)$$

where we have defined the new time-dependent Hamiltonian

$$\mathcal{H}_1(\tau) = e^{-\tau \mathcal{H}_0} \mathcal{H}_1 e^{\tau \mathcal{H}_0} \quad (4.12)$$

Let the solution of (4.11) be written as

$$\omega(\tau) = T(\tau) \omega(0) \quad (4.13)$$

Then the connection $T(\tau)$ with the matrix $S(\tau)$ (4.7) is given by (4.10)

$$S(\tau) \Omega(0) = e^{\tau \mathcal{H}_0} T(\tau) \omega(0) \quad (4.14)$$

$$S(\tau) = e^{\tau \mathcal{H}_0} T(\tau)$$

since $\Omega(0) = \omega(0)$

We proceed now to solve Eq. (4.11) by the method of successive approximations treating $\mathcal{H}_1(\tau)$ as a small perturbation. This means that we find for $T(\tau)$ a series solution in the form

$$T(\tau) = \sum T_n(\tau) \quad (4.15)$$

where

$$\dot{T}_n(\tau) = \mathcal{H}_n(\tau) T_{n-1}(\tau) \quad (4.16)$$

and

$$T_n(0) = \begin{cases} 0 & \text{for } n \neq 0 \\ \underline{I} & \text{for } n = 0 \end{cases} \quad (4.17)$$

This set of operator equations (4.16) with the initial conditions (4.17) is by (4.13) equivalent to (4.11). The n^{th} equation (4.16) can be integrated

$$T_n(\tau) = \int_0^\tau d\tau_1 \mathcal{H}_n(\tau_1) T_{n-1}(\tau_1)$$

thus giving the n^{th} approximation in terms of the $(n-1)^{\text{st}}$. By iterating the process we obtain

$$T_n(\tau) = \int_0^\tau d\tau_1 \int_0^{\tau_1} d\tau_2 \dots \int_0^{\tau_{n-1}} d\tau_n \mathcal{H}_n(\tau_1) \mathcal{H}_n(\tau_2) \dots \mathcal{H}_n(\tau_n) \quad (4.18)$$

In order to progress further with this expression it is necessary to obtain an explicit representation of time dependent Hamiltonian $\mathcal{H}_n(\tau)$.

We write

$$\mathcal{H}_n(\tau) = \sum_{\substack{\nu\nu' \\ ik}} \delta_{\nu\nu'}^{ik} S_i^{(\nu)}(\tau) S_k^{(\nu')}(\tau) + A \sum_{\nu} (S^{(\nu)}(\tau) \cdot \underline{I}^{(\nu)}) \quad (4.19)$$

where $S_i^{(\nu)}(\tau)$ is the i^{th} component of the time dependent spin vector of ion (ν), defined by

$$S_i^{(\nu)}(\tau) = e^{-\tau \mathcal{H}_0} S_i^{(\nu)} e^{\tau \mathcal{H}_0} \quad (4.20)$$

It is possible to evaluate the τ -dependence of these operators rather easily by making use of the property of the spin operators to generate the infinitesimal rotation in the 3-dimensional space. This can be exhibited most concisely by choosing the direction of \underline{H} as the 3-direction and introducing the components

$$\begin{aligned} S_{\pm 1} &= \frac{1}{\sqrt{2}} (S_1 \pm i S_2) \\ S_0 &= S_3 \end{aligned} \quad (4.21)$$

(We suppress the index ν for the time being).

These components have the commutation rules

$$\begin{aligned} [S_0, S_{\pm}] &= S_{\pm} \\ [S_+, S_-] &= S_0 \end{aligned} \quad (4.22)$$

We find then with the help of the commutation rules

$$\begin{aligned} S_m(\tau) &= e^{-\theta S_3} S_m e^{\theta S_3} = e^{-m\theta} S_m \\ \theta &= \beta \mathcal{H} \tau \quad (m=0, \pm 1) \end{aligned} \quad (4.23)$$

We may now express (4.19) in terms of the components $S_m(\tau)$ (4.21) and obtain then

$$\mathcal{H}_i(\tau) = F(\tau) + G(\tau)$$

$$F(\tau) = \sum_{\substack{v v' \\ m m'}} \gamma_{v v'}^{m m'} e^{-(m+m')\theta} S_m^{(v)} S_{m'}^{(v')}$$

$$G(\tau) = A \sum_{v, n} (S_n^{(v)} \underline{I}^{(v)n}) e^{-n\theta} \quad (4.24)$$

Here the \underline{I}^n are defined in terms of the I_n by the rule $\underline{I}^n = I_{-n}$.

Then formulae (4.24) in conjunction with (4.18), (4.14), and (4.9) give a general expression for the density matrix which can be used to evaluate \int to any desired approximation.

2. A general theorem regarding the first order term in A

Rose and Simon stated the general result that the first order contribution in A to the nuclear alignment depends only on the total magnetization of the ions but not explicitly on the type of interaction between the ions.

The proof of this follows immediately from our general formalism.

We have the Hamiltonian

$$\mathcal{H} = \beta (H \cdot S) + F(S) + A (S^{(v)} \cdot \underline{I}^{(v)}) \quad (4.25)$$

$F(S)$ denotes the interaction between the ionic spins. We make no further assumption about this interaction. If in the expression $\rho_n = \frac{1}{Z} e^{-\mathcal{H}/kT}$ we retain only the terms independent of A and linear in A we can express it in the form

$$\rho_n = \frac{1}{Z} (P + A \underline{I}^{(v)} \cdot \underline{Q}^{(v)}) \quad (4.26)$$

where
$$P = \text{tr}_e e^{- [g\beta \underline{H} \cdot \underline{S} + F(S)] / kT}$$

$$\underline{Q}^{(v)} = \text{tr}_e \underline{S}^{(v)} e^{- [g\beta (\underline{H} \cdot \underline{S}) + F(S)] / kT} \quad (4.27)$$

The second equation of (4.27) is obtained in the following way: Write $\mathcal{H} = \mathcal{H}_0 + A \sum_{\nu} (\underline{S}^{(\nu)} \cdot \underline{I}^{(\nu)})$ and develop $e^{-\mathcal{H}/kT}$ in powers of A. The term which multiplies $A \underline{I}_i^{(\nu)}$ for instance is then

$$Q_i^{(\nu)} = \sum_{n=1}^{\infty} \frac{1}{n!} \text{tr}_e \sum_{p=0}^{n-1} \mathcal{H}_0^p S_i^{(\nu)} \mathcal{H}_0^{n-p-1} (kT)^{-n} \quad (4.28)$$

Now we make use of the invariance of the trace under cyclic permutations and find that in the summation over p all the terms are identical and thus just produce a factor n. In this way we obtain (4.27):

$$Q_i^{(\nu)} = \frac{1}{kT} \text{tr}_e \left(S_i^{(\nu)} \sum_{n=0}^{\infty} \frac{1}{n!} \left(\frac{\mathcal{H}_0}{kT} \right)^n \right) = \frac{1}{kT} \text{tr}_e S_i^{(\nu)} e^{-\mathcal{H}_0/kT} \quad (4.29)$$

The expression (4.26) can further be simplified by making use of the fact that for any average value of a quantity which is symmetrical in the nuclear spins we may replace all the $\underline{I}^{(\nu)}$ by any particular one of them and then omit the index ν . Thus we obtain now for the density matrix of one nuclear spin

$$\rho_n = \frac{1}{Z} \left(P + \frac{A}{kT} \underline{I} \cdot \underline{Q} \right) \quad (4.30)$$

$$P = k_e e^{-\mathcal{H}_0/kT} = Z_e, \quad Q_i = \frac{1}{N} k_e \sum_v S_i^{(v)} e^{-\mathcal{H}_0/kT} \quad (4.31)$$

N is the number of ions in the system. Neglecting terms of order A^2 we find

$$Z = k_e e^{-\mathcal{H}_0/kT} = (2i+1) k_e e^{-\mathcal{H}_0/kT} = (2i+1) Z_e$$

Z has no linear terms in A since the traces of the operators I_i all vanish.

Inspection of (4.31) shows that

$$\begin{aligned} Q_i &= \frac{1}{N} k_e S_i e^{-\mathcal{H}_0/kT} = -\frac{kT}{g\beta} \frac{1}{N} \frac{\partial}{\partial H_i} k_e e^{-\mathcal{H}_0/kT} \\ &= -\frac{kT}{g\beta} \frac{1}{N} \frac{\partial}{\partial H_i} Z_e \end{aligned} \quad (4.33)$$

Since the average magnetic moment of the electronic spins is given by

$$M_i = -kT \frac{1}{N} \frac{\partial}{\partial H_i} Z_e \quad (4.33)$$

we may write for Q_i

$$Q_i = \frac{Z_e}{g\beta} M_i \quad (4.34)$$

and finally for ρ_n

$$\rho_n = \frac{1}{2i+1} \left(1 + \frac{A}{kTg\beta} \frac{I \cdot M}{\quad} \right) \quad (4.35)$$

This formula is valid if $\frac{A}{kT} \ll 1$. No assumption is made as to the order of magnitude of $\frac{A}{g\beta H}$. The quantity

$$\frac{H}{g\beta A} = f_e \leq 1 \quad (4.36)$$

is the degree of electronic alignment.

It is worth emphasizing that the validity of (4.35) does not depend on the strength of the external magnetic field nor on the strength of the interaction between neighboring ions. Thus the nuclear alignment in first order in $\frac{A}{kT}$ is entirely a function of f_e , the degree of alignment of the electronic spins. With this we have answered both of Bleaney's objections against this method of producing nuclear alignment.

V. APPLICATIONS AND EXAMPLES

We shall now use the general theory of the last section to calculate some of the higher order effect in nuclear alignment with h.f. coupling. As a first example we calculate the second order term in $\frac{A}{kT}$ disregarding the interaction of the electron spins. That is, we put $F = 0$ in Eq. (4.4). Thus we can use Eq. (4.18) with $\mathcal{R}_2(\tau)$ replaced by $G(\tau)$ (Eq. (4.24)). The specialization to this case gives then the basic formulae

$$\begin{aligned} \rho_n &= k_e \rho & \rho &= \frac{1}{2} S(-\frac{1}{kT}) & Z &= \text{tr } S(-\frac{1}{kT}) \\ S(\tau) &= e^{\tau \mathcal{H}_0} T(\tau) \\ \mathcal{H}_0 &= g\beta H S_3 & T(\tau) &\approx I + T_1(\tau) + T_2(\tau) \\ T_2(\tau) &= A^2 \int_0^\tau d\tau_1 \int_0^{\tau_1} d\tau_2 e^{-(m_1 \theta_1 + m_2 \theta_2)} S_{m_1} S_{m_2} I^{m_1} I^{m_2} \\ \theta_i &= g\beta H \tau_i \end{aligned} \quad (5.1)$$

The last expression may also be written

$$T_2(\tau) = \left(\frac{A}{\int \beta H} \right)^2 F_2(m_1, m_2, \theta) S_{m_1} S_{m_2} I^{m_1} I^{m_2} \quad (5.2)$$

with

$$F_2(m_1, m_2, \theta) = \int_0^\theta d\theta_1 \int_0^{\theta_1} d\theta_2 e^{-(m_1 \theta_1 + m_2 \theta_2)} \quad (5.3)$$

The functions $F_2(m_1, m_2, \theta)$ may be evaluated and tabulated and are given below. Since we take afterwards the trace with respect to the electronic variables we need only the diagonal elements of (5.2) and this means that out of this term we need to consider only those for which $m_1 + m_2 = 0$. In the table below we have listed these quantities and also the corresponding

m_1	m_2	$F_2(m_1, m_2; \theta)$	$(M S_{m_1} S_{m_2} M)$
+1	-1	$\theta + e^{-\theta} - 1$	$1/2 [-]$
-1	+1	$-\theta + e^{\theta} - 1$	$1/2 [+]$
0	0	$1/2 \theta^2$	M^2

$[\pm] = S(S+1) - M(M \pm 1)$

diagonal matrix elements of $S_{m_1} S_{m_2}$. We obtain thus for $\rho_n^{(2)}$, the term in ρ_n which depends only on A^2 :

$$\begin{aligned} (m | \rho_n^{(2)} | m') &= \frac{1}{(2i+1)ze} \sum_M e^{-\theta M} \left\{ (\theta + e^{-\theta-1}) \frac{1}{4} [S(S+1) - M(M+1)] \right. \\ &\times [i(i+1) - m(m+1)] + (-\theta + e^{\theta-1}) \frac{1}{4} [S(S+1) - M(M+1)] [i(i+1) - m(m-1)] \\ &\left. + \frac{1}{2} \theta^2 M^2 m^2 - \alpha \right\} \delta_{mm'} \end{aligned} \quad (5.4)$$

where α is the correction term proportional to A^2 in the normalization factor

$$Z = (2i+1) Z_e (1 + \alpha) \quad (5.5)$$

α is so determined that

$$\text{tr } \rho_n^{(z)} \equiv \sum_m (m | \rho_n^{(z)} | m) = 0 \quad (5.6)$$

as it must if the normalization condition of the density matrix is to come out correct in the order A^2 :

$$\text{tr } \rho = 1 \quad (5.7)$$

It is now convenient to express this result in terms of the partition function $Z_e(\theta)$ of the electron spins only. Thus we have

$$Z(\theta) = \sum_M e^{-\theta M} = \frac{\cosh \frac{2S+1}{2} \theta}{\cosh \frac{\theta}{2}} \quad (5.8)$$

$$Z'(\theta) = - \sum M e^{-\theta M} \quad (5.9)$$

$$Z''(\theta) = \sum M^2 e^{-\theta M} \quad (5.10)$$

The final expression we may now express in the operator forms

$$\rho_n^{(z)} = \frac{1}{2i+1} \frac{1}{Z_e} \left(\frac{A}{kT} \right)^2 (a_0 - \alpha + a_1 I_3 + a_2 I_3^2) \frac{1}{\theta^2} \quad (5.11)$$

With

$$\begin{aligned}
 a_0 &= \frac{i(i+1)}{2} \left\{ [S(S+1) - Z''(\theta)] (\cosh \theta - 1) + Z'(\theta) (\sinh \theta - \theta) \right\} \\
 a_1 &= \frac{1}{2} Z'(\theta) (\cosh \theta - 1) + \frac{1}{2} (S(S+1) - Z''(\theta)) (\sinh \theta - \theta) \\
 a_2 &= \frac{1}{2} Z''(\theta) (\theta - \sinh \theta) + \frac{1}{2} [S(S+1) - Z''(\theta)] (1 - \cosh \theta) \\
 &\quad \left(\theta - \frac{g\beta H}{kT} \right) + \frac{1}{2} \theta^2 Z''(\theta)
 \end{aligned} \tag{5.12}$$

Since

$$\langle I_3 \rangle = 0 \quad \langle I_3^2 \rangle = (2i+1) \frac{i(i+1)}{3}$$

We find for α the value

$$\alpha = a_0 + \frac{i(i+1)}{3} a_2 \tag{5.13}$$

Thus we can express the result entirely in a_1, a_2

$$\rho_n^{(2)} = \frac{1}{2e} \left(\frac{A}{kT} \right)^2 \left(\frac{kT}{g\beta H} \right)^2 \left(-\frac{i(i+1)}{3} a_2 + a_1 I_3 + a_2 I_3^2 \right) \tag{5.14}$$

The correction to the nuclear alignment will be contained in the term with a_1 . It is of order of magnitude

$$O(\rho^{(2)}) \sim \frac{1}{2i+1} \left(\frac{A}{kT} \right)^2 (2S+1) \sim \left(\frac{A}{kT} \right)^2 \tag{5.15}$$

for all values of $\theta = \frac{g\beta H}{kT}$

Thus the contribution for Mn^{++} turns out to be at $kT \sim 0.1 \text{ cm}^{-1}$

$$\left(\frac{A}{0.1}\right)^2 \sim 10^{-2} = 1\% \quad (5.16)$$

The correction due to the interaction of the ions to the nuclear alignment calculated from the first term above will thus turn out to be

$$\left(\frac{A}{kT}\right)^2 \frac{\delta}{kT} \sim \frac{1}{100} \frac{\delta}{kT} \quad (5.17)$$

where δ is some mean of the coefficients in (4.19).

The Bleaney Alignment

In a recent letter Bleaney¹¹ proposed a method of nuclear alignment making use of the asymmetrical h.f. coupling which is known to exist in many crystals. The first order contribution to this effect, which is probably the only significant one, can be easily calculated.

The Hamiltonian may be taken as

$$\mathcal{H} = A S_0 I_0 + B (I_+ S_- + I_- S_+) \quad (5.18)$$

The density matrix is then given by

$$\rho_n = \frac{1}{Z} k_e e^{-\mathcal{H}/kT} \quad Z = \text{tr } \rho_n \quad (5.19)$$

We write it in the form

$$\rho_n = \rho^{(0)} + \rho^{(1)} + \rho^{(2)} + \dots \quad (5.20)$$

$$k \rho^{(v)} = 0 \quad v \neq 0$$

We obtain then for the successive approximations

$$\rho^{(0)} = \frac{1}{2I+1}$$

$$\rho^{(1)} = 0$$

$$\rho^{(2)} = \frac{1}{(kT)^2} \frac{1}{2!} \frac{1}{(2I+1)(2S+1)} \left\{ \text{tr}_e [A S_0 I_0 + B(S_+ I_- + S_- I_+)]^2 - \alpha \right\} \quad (5.21)$$

This leads to the following expression for $\rho^{(2)}$

$$\rho^{(2)} = \frac{1}{(kT)^2} \frac{1}{2!} \frac{1}{(2I+1)(2S+1)} \left[A^2 I_0^2 + B^2 (I_- I_+ + I_+ I_- + S_+ S_- + S_- S_+) - \alpha \right]$$

Now

$$k S_0^2 = \frac{1}{3} S(S+1)(2S+1)$$

$$k S_+ S_- = k S_- S_+ = \frac{1}{3} S(S+1)(2S+1)$$

Thus $\rho^{(2)}$ becomes

$$\rho^{(2)} = \frac{1}{2} \frac{1}{(kT)^2} \frac{S(S+1)}{3(2I+1)} \left\{ A^2 I_0^2 + B^2 [I(I+1) - I_0^2] - \alpha \right\}$$

The constant α is determined from the trace condition

$$k \rho^{(2)} = 0$$

or

$$\alpha = \frac{I(I+1)}{3} (2B^2 + A^2)$$

Thus finally

$$\rho^{(2)} = \frac{1}{2} \frac{1}{(kT)^2} \frac{S(S+1)}{3(2I+1)} (B^2 - A^2) \times \left(\frac{1}{3} I(I+1) - I_0^2 \right) \quad (5.22)$$

From this we see that the $\rho^{(2)}$ vanishes if $B^2 = A^2$, that is if the h.f. coupling is isotropic. We see furthermore that all the odd powers of I_0 have average values zero. Thus in particular

$$\text{tr } I_0 \rho = \overline{I_0} = 0 \quad (5.23)$$

Thus in this case there is no polarization of the nuclear spin. For the second power, on the other hand, we obtain

$$\overline{I_0^2} = \text{tr } I_0^2 \rho = \frac{I(I+1)}{3} + \Delta \quad (5.24)$$

is the difference of $\overline{I_0^2}$ from the value for the isotropic case

$\frac{1}{3} I(I+1)$ and represents thus a measure of the degree of nuclear alignment.

We obtain for Δ

$$\Delta = \text{tr } \rho^{(2)} I_0^2$$

Making use of the relations

$$\begin{aligned} \text{tr } I_0^2 &= \frac{1}{3} I(I+1)(2I+1) \\ \text{tr } I_0^4 &= \frac{1}{5} I^2(I+1)^2 \left[1 - \frac{1}{3I(I+1)} \right] (2I+1) \end{aligned} \quad (5.25)$$

We obtain finally

$$\Delta = \frac{A^2 - B^2}{(kT)^2} \frac{S(S+1)I(I+1)}{90} \left\{ \frac{4}{3} I(I+1) - 1 \right\} \quad (5.26)$$

The Pound Alignment

In Pound's method of nuclear alignment the quadrupole interaction of the nucleus with the crystal field is used to produce an uneven population of the states of the nucleus.

In case of cylindrical symmetry the relevant part of the Hamiltonian is then

$$\mathcal{H} = F \left(\frac{1}{3} I(I+1) - I_0^2 \right) \tag{5.27}$$

where

$$F = - \frac{3eq}{4I(2I-1)} \left(\frac{\partial^2 V}{\partial z^2} \right)_0 \tag{5.28}$$

Here q is the electric quadrupole moment of the nucleus and $-\frac{\partial^2 V}{\partial z^2}$ the gradient of the electric field in the direction of the symmetry axis.

Since \mathcal{H} is quadratic in the nuclear spin the density matrix has a term linear in $\frac{1}{kT}$ which is

$$S^{(1)} = \frac{1}{2I+1} \frac{F}{kT} \left(\frac{1}{3} I(I+1) - I_0^2 \right) \tag{5.29}$$

This is of the same form as (5.22) and will thus lead to the same result for the second moment with suitable change of constants.

The two effects become equal at a temperature T such that

$$|F| = \frac{1}{kT} \frac{S(S+1)}{6} |A^2 - B^2| \tag{5.30}$$

If $|A| \gg |B|$

$$kT \sim \frac{S(S+1)}{6} \left| \frac{A}{F} \right| |A| \quad (5.31)$$

Since

$$F \sim 10^{-3} \text{ cm}^{-1} \quad A \sim 10^{-2} \text{ cm}^{-1}$$

$$kT \sim \frac{S(S+1)}{60}$$

The two effects become thus comparable in the neighborhood of about 1° K. It should be noted that the two contributions add algebraically and may add as well as subtract, depending on the sign of F. We have thus a method of measuring the sign of F by measuring nuclear alignment.

VI. METHODS OF DETECTION OF NUCLEAR POLARIZATION AND ALIGNMENT

In order to detect and measure the nuclear polarization and alignment it is necessary to study physical effects which depend on the orientation of the nuclear spins. Possible effects of this sort are the absorption of slow polarized neutrons and the emission of γ -rays. We shall discuss these effects for the special case of the Mn nucleus since this seems to be so far the most suitable nucleus for producing polarization.

Detection with Neutron Absorption

The most recent data on the Mn nucleus were obtained by Harris, Hibdon and Muehlhaus¹³. According to these authors the lowest resonance levels of Mn are situated at

$$E_1 = 345 \text{ eV}$$

$$E_2 = 2400 \text{ eV}$$

They assign to them the following widths for n- and γ -emission

$$\Gamma_{\gamma_1} = 0.6 \text{ eV}$$

$$\Gamma_{n_1} = 21.6 \text{ eV}$$

$$\Gamma_{\gamma_2} \sim \text{order } 1 \text{ eV}$$

$$\Gamma_{n_2} = 304 \text{ eV}$$

(6.1)

The lower level corresponds to the resultant spin $J = 3$ and the upper to $J = 2$. Each of these two levels contributes a certain amount to the neutron absorption at low energy which can be calculated from the Breit-Wigner formula for non-interfering levels

$$\sigma^i = 4\pi \lambda^2 \kappa_i g_i \frac{\Gamma_{\gamma_i} \Gamma_{n_i}}{(\Gamma_{\gamma_i} + \Gamma_{n_i})^2 + (E - E_i)^2}$$

(6.2)

where g_i are the statistical weight factors for the two levels

$$g_i = \frac{1}{2} \left(1 \pm \frac{1}{2I+1} \right) \text{ for } J = I \pm \frac{1}{2}$$

(6.3)

In case of Mn we have

$$g_i = \begin{cases} \frac{7}{12} & J = 3 \\ \frac{5}{12} & J = 2 \end{cases}$$

(6.4)

For thermal neutrons we may develop (6.2) and write

$$\sigma^i \approx 4\pi \lambda^2 \kappa_i g_i \frac{\Gamma_{\gamma_i} \Gamma_{n_i}}{E_i^2}$$

(6.5)

The ratio for the two levels thus turns out to be independent of the neutron energy

$$\chi = \frac{\sigma_2}{\sigma_1} = \frac{\chi_2}{\chi_1} \frac{\Gamma_{r_2} \Gamma_{n_2} g_2}{\Gamma_{r_1} \Gamma_{n_1} g_1} \left(\frac{E_1}{E_2} \right)^2 \quad (6.6)$$

Since $\chi_i = \frac{\hbar M}{v_i} \sim E_i^{-1/2}$

$$\chi = \left(\frac{E_1}{E_2} \right)^{5/2} \frac{\Gamma_{n_2} g_2}{\Gamma_{n_1} g_1} \frac{\Gamma_{r_2}}{\Gamma_{r_1}} = 7.9 \times 10^{-2} \frac{\Gamma_{r_2}}{\Gamma_{r_1}} \quad (6.7)$$

It is seen that the ratio of the two absorption cross section depends only on the ratio of the γ -widths. Unfortunately the width of the upper level is not very well known. If we assume it to be the same as that of the lower level, then we obtain for

$$\chi = 7.9 \times 10^{-2} \quad \text{for } \Gamma_{r_2} = \Gamma_{r_1} \quad (6.8)$$

With χ we can calculate the effective absorption cross section for slow polarized neutrons by polarized nuclei

$$\begin{aligned} \bar{\sigma}_a &= \sigma_1 \left(1 + \frac{I}{I+1} f_n f_N \right) + \sigma_2 \left(1 - f_n f_N \right) \\ &= \sigma_1 \left[\left(1 + \frac{I}{I+1} f_n f_N \right) + \chi \left(1 - f_n f_N \right) \right] \\ &= \sigma_a \left[1 + \frac{I}{I+1} \frac{I - (I+1)\chi}{1+\chi} f_n f_N \right] \end{aligned} \quad (6.9)$$

where $\sigma_a = \sigma_1 + \sigma_2 = \sigma_1 (1+x)$

is the total absorption cross section for thermal neutrons. The polarization effect is thus always reduced by the contribution of the upper level to the slow neutron absorption. In order to obtain an estimate of this contribution we compare the total measured absorption cross section for thermal neutrons. ¹⁴⁾

$$\sigma_a = \sigma_1 + \sigma_2 = 12,1 \text{ b} \quad (6.10)$$

with the value obtained from the one level formula

$$\sigma_1 = 10,2 \text{ b} \quad (6.11)$$

The difference is then presumably due to the upper level which gives

$$\frac{\sigma_a}{\sigma_1} = 1+x = \frac{12,1}{10,2}, \quad x \sim 0,2$$

corresponding to a δ -width $\Gamma_2 \sim 1.52 \text{ ev}$. In the following table we give the polarization effect for three values of x .

x	$\lambda(x)$	% Polarization Effect	Γ_2
0	0.71	100	0
0.08	0.59	83	0.6
0.2	0.43	62	1.52
			ev

$$\lambda(x) = \frac{I - (I+1)x}{(I+1)(1+x)} \quad \text{for } I = 5/2$$

We see thus that a reduction of the polarization effect by 10 to 20% is very probable and perhaps as much as 40% is not impossible.

Detection with γ -emission

In order to make any statements regarding the angular distribution of the γ -rays emitted upon slow neutron capture by the Mn nucleus it is necessary to know something about the type of electromagnetic radiation (multipole order) to be expected from the intermediate compound nucleus. This is possible if we have some information on the spin and parity of the excited state and the ground state of Mn^{56} .

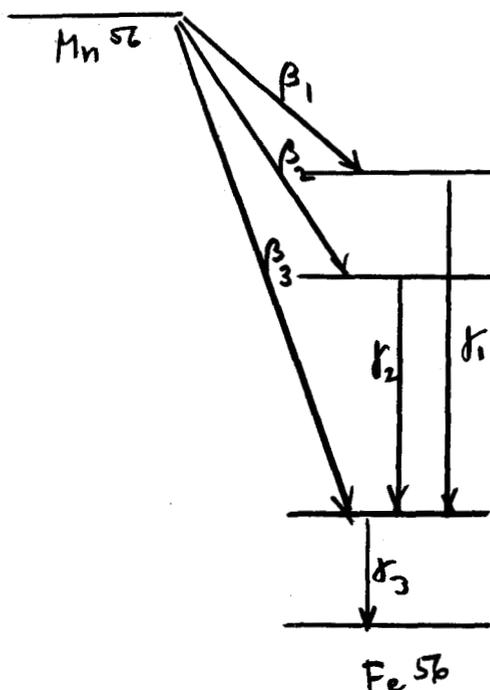
The spin of Mn^{55} is known to be $I = 5/2$. It is one of the few exceptions of the spin predictions of the shell model. The latter would have given $I = 7/2$ arising out of the configuration $f_{7/2}^5$. The actual spin and magnetic moment values indicate that the configuration is very likely $f_{7/2}^4 d_{5/2}$ leading to $i = 5/2$ and parity even.

The compound nucleus of the lower level (E_1) is formed by S-capture resulting thus in the excited state with spin $J = 3$ and parity even.

We have no direct information about the ground state of Mn^{56} . The shell model would predict parity odd since there are three $p_{3/2}$ neutrons to be filled in over the closed neutron shell of 28. As to the spin the shell model makes no definite prediction since Mn^{56} is an odd nucleus. The 5 known examples of odd odd nuclei with known spins show that the resultant

spin is obtained¹⁵ by combining the spin of the neutron core to a value which usually is neither the maximum nor the minimum but is perhaps closer to the maximum. In our case the resultant spins of the two cores are 5/2 for the protons and 3/2 for the neutrons thus the Mn^{56} ground state can have spins 1, 2, 3, 4 and parity odd with perhaps spin 3 most probable.

In order to fix the spin value of Mn^{56} with some degree of certainty we have to look for other evidence. This is obtained from the subsequent β -decay $Mn^{56} \rightarrow Fe^{56} + \beta$ (2.6 h). The investigations of Elliott and Deutsch¹⁶ and of Siegbahn¹⁷ have shown that the β -spectrum is complex and is accompanied by several γ -rays according to the following scheme:



It is notable that there is no direct transition to Fe^{56} . The various energies and lifetimes are given as

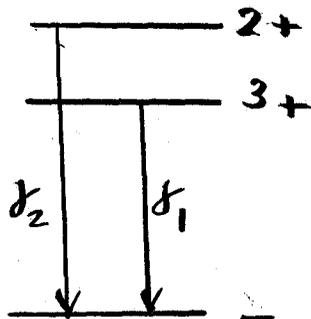
β -ray	E	t	ft
1	0.75	4.65	9.5
2	1.04	3.10	40
3	2.81	1.80	1320
	Mev	$\times 10^4$ sec	$\times 10^4$

This makes the transition β_1 allowed and β_2 , probably β_3 almost certainly first forbidden. This allows us to assign parity to the two middle levels of Fe^{56} since the parity change for first forbidden transitions is "yes". Since we have previously found the parity of Mn^{56} to be odd we must assign to these two levels even parity.

We know also that the spin of the ground level of Fe^{56} is 0 and parity even since it is an even even nucleus. Furthermore the first excited states of even-even nuclei have almost always spin two and even parity.

We are now in a position to rule out some of the spin values of the Mn^{56} nucleus. We have previously seen that possible values are 1, 2, 3, 4. Since there is no direct β -transition to the ground state Fe^{56} we must assume this transition to be at least second forbidden which (for Gamow-Teller selection rules) corresponds to a spin change $\Delta I \geq 3$. Thus the spin of Mn^{56} is either 3 or 4.

We look for further information on the Mn^{56} ground level by studying the radiation which can be emitted from its two excited states



Possible multipole radiation:

E	2^1	2^3	2^5	...
M		2^2	2^4	...

The two types of radiation E_3 and M_2 are the ones which have the right probability and which come into question. E_1 is usually much less probable than E_3 and may be excluded. Of the two possibilities E_3 and M_2 we would expect the former to be more probable for the energy involved here. Thus we conclude the capture γ -radiation is likely an electric 2^3 -pole radiation.

Of the two possible spin values 4, 3 for the ground state both are compatible with this radiation. It is, therefore, not possible on the basis of existing evidence to make any definite assignment of the ground level spin.

Assuming now that the capture occurs primarily through the lower level and that the radiation is electric 2^3 pole we obtain for the angular distribution of the μ -rays emitted from an assembly of nuclei with a probability distribution p_M for the M^{th} level¹⁸

$$F(\theta) = \sum_{m, M} p_M \left| C_{M-m, m}^{J J_F j} \right|^2 I_{jm}(\theta) \quad (6.12)$$

where $C_{M-m, m}^{J J_F j}$ are the Clebsch Gordon coefficients for the composition of angular momenta J, J_F to a resultant j and $I_{jm}(\theta)$ are the angular distribution of the electromagnetic spherical wave of angular momentum jm

$$I_{jm}(\theta) = \frac{1}{2} (j-m+1)(j+m) |P_j^{m-1}(\theta)|^2 + m^2 |P_j^m(\theta)|^2 + \frac{1}{2} (j+m+1)(j-m) |P_j^{m+1}(\theta)|^2 \quad (6.13)$$

For the special case of the Mn-nucleus we obtain the result for $J = 3$, $J_F = 3$ or 4 , and according to the foregoing $j = 3$ (electric 2^3 -pole radiation). In case electric dipole radiation is important we had instead to take $J = 3$, $J_F = 3$ or 4 and $j = 1$.

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