

21-minute  $Mn^{52}$  decays 99.95 percent by positron emission,<sup>6</sup> the electron capture to positron ratio for  $Fe^{52}$  was found to be  $1.6 \pm 0.4$ .

The  $Fe^{52}$  decay undoubtedly proceeds by an allowed transition ( $\log ft \sim 4$ ). One can therefore deduce<sup>7</sup> a maximum positron energy of  $640 \pm 40$  kev for  $Fe^{52}$  from the measured electron capture to positron ratio of  $1.6 \pm 0.4$ . Absorption measurements on sources of  $Fe^{52}$  in equilibrium with  $Mn^{52}$  gave an end point for the low energy  $\beta^+$  component compatible with this energy. The presence of the 2.7-Mev positrons of  $Mn^{52}$  makes an accurate determination of the  $Fe^{52}$  end point difficult.

Scintillation spectrometer measurements of an  $Fe^{52}$  sample in equilibrium with  $Mn^{52}$  showed the presence of annihilation radiation and of a  $\gamma$ -ray of about 1.5 Mev. Immediately after a manganese-iron separation, the  $Fe^{52}$  exhibited only annihilation radiation. The subsequent growth of the 1.5-Mev  $\gamma$ -ray was observed. It may be concluded that  $Fe^{52}$  emits no  $\gamma$ -rays of energy greater than 0.5 Mev in appreciable abundance.

For intensity reasons, the  $\gamma$ -ray measurements just described were made with  $Fe^{52}$  samples produced by the bombardment of chromium with 30-Mev helium ions according to the reaction  $Cr^{50}(\alpha, 2n)$ . However, the  $Fe^{52}$  so obtained could not be used for the determination of the electron capture to positron ratio because of the preponderance of  $Fe^{56}$  produced by the  $Cr^{52}(\alpha, n)$  reaction.

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### Yields of Iron Isotopes in High Energy Nuclear Reactions\*

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THE determination<sup>1</sup> of the electron capture to positron ratio in  $Fe^{52}$  has made it possible to obtain directly the relative disintegration rates of  $Fe^{52}$  and  $Fe^{55}$  in a sample by comparison of the 8-hour and long-lived Mn x-ray activities as measured with a proportional counter and pulse-height analyzer.<sup>2</sup> Such measurements have been carried out on iron samples separated from cobalt and copper targets bombarded with 370-Mev protons in the circulating beam of the Columbia University cyclotron at Nevis. With thin targets ( $\sim 5$ -Mev energy loss) the ratios of formation cross sections ( $\sigma_{55}/\sigma_{52}$ ) were  $136 \pm 20$  and  $168 \pm 25$  for cobalt and copper targets, respectively. With a thick copper target (energy loss  $\sim 60$  Mev) a ratio of  $206 \pm 30$  for ( $\sigma_{55}/\sigma_{52}$ ) was obtained. Although it is not certain that the difference between the results for the thick and thin copper targets is real, it is in the expected direction since the energy loss of the protons in the thick target would tend to reduce the  $Fe^{52}$  yield more than the  $Fe^{55}$  yield.<sup>3</sup>

The rather high cross section for the formation of  $Fe^{56}$  relative to that for  $Fe^{52}$  is of interest in connection with the results of Bartell *et al.*<sup>4</sup> They reported alternating high and low yields for successive atomic numbers among the products of the bombardment of copper with 190-Mev deuterons. Since they did not investigate the  $Fe^{55}$  yield, their observation that the yield of iron was low compared with that of manganese would be modified if the relative yields of  $Fe^{55}$  and  $Fe^{52}$  in their bombardment were similar to those found here. That this may be so is indicated by the fact

that the ratio of formation cross sections for  $Fe^{59}$  and  $Fe^{62}$  was similar in the two cases; Bartell *et al.* reported  $(\sigma_{59}/\sigma_{62}) = 20$  while, by absolute beta- and x-ray counting, we found  $(\sigma_{59}/\sigma_{62}) = 45 \pm 15$  in the bombardment of the thick copper target.

It is interesting to note that in our copper bombardment the yield of  $Fe^{55}$  is significantly higher than that of either  $Fe^{52}$  or  $Fe^{59}$ . This result is qualitatively consistent with the emission of nucleons by evaporation and "knock-on" processes.

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### Coupling among Nuclear Magnetic Dipoles in Molecules\*

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MULTIPLE nuclear magnetic resonance lines have been reported in several liquids, such as the Sb resonances<sup>1</sup> in aqueous  $NaSbF_6$ , and the  $P^{31}$  and  $F^{19}$  resonances<sup>2</sup> in  $POCl_2F$ ,  $POClF_2$ , and  $CH_3OPF_2$ . Suggested interpretations of these effects include hindrance to molecular rotation<sup>1-3</sup> and second-order magnetic dipolar interactions.<sup>4</sup> We feel that new measurements of ours, together with the previously published results,<sup>2</sup> exclude both of the above suggestions, in general, and we would like to propose the hypothesis that the splittings come from a second-order interaction between the nuclear magnetic moments and some magnetic field internal to the molecule.

Significant experimental facts seem to be as follows. (1) The splitting is associated with the nuclear magnetic moments since  $\delta H_A/\delta H_B = \mu_B/\mu_A$ , where  $\delta H$  is the multiplet separation.<sup>2</sup> (2) Nuclei of the same species do not interact when they are in chemically equivalent positions in a molecule.<sup>2</sup> As an additional example, in  $HPF_6$  we observe that P splits the F-resonance into two components and the F split the P into seven components, but there is no observable splitting of the F-resonances by the fluorines acting among themselves. (3) The relative intensities and number of components of a line *A* are determined by the statistical weights and the number of possible spin states of the *B* which cause the splitting.<sup>2</sup> Note that *B* have been in chemically equivalent positions in the twelve compounds we have observed thus far. (4) Our recent experiments show the splittings are independent of temperature and the strength of the static field  $H_0$ . The splittings remain the same in  $CH_3OPF_2$  and  $POCl_2F$  from room temperature until the narrow components diminish in intensity and are lost in a broadening background line at about  $-80^\circ C$ . (5) Although the splittings are several tenths of a gauss, we have found in almost all cases that the components are at least as narrow as the 0.01-gauss magnet inhomogeneity over the sample. (6) The  $P^{31}$ -resonances in  $PF_3$  and  $PH_3$  are quadruple with splittings of 0.82 and 0.10 gauss, respectively. The ratio of these splittings is 8.2 while the calculated ratio of the magnetic dipolar fields ( $\mu/r^3$ ) is about 0.7.

On the basis of these facts, we feel there are the following crucial objections to:

*Rotational hindrance:* In this case identical nuclei should interact, counter to observations. It is difficult also to reconcile any restriction of molecular tumbling with the observed narrow line widths or with the observed temperature independence of the splittings. Moreover, in the case of  $HPF_6$ , the phosphorus resonance has seven components, not nine as predicted by Andrew's analysis.<sup>5</sup> The evidence concerning  $NaSBF_6$ <sup>1</sup> is not conclusive al-

though the breadth of the Sb resonance lines tends to support Andrew's hypothesis for this particular compound.

**Second-order magnetic dipole interaction:** The second-order splitting is of order  $(\mu^2/r^6)/H_0$  gauss, where  $r$  is the internuclear distance. Predicted splittings are several milligauss, contrasting with observed  $H^1$ ,  $F^{19}$ , and  $P^{31}$  splittings in the range from 0.10 to 0.75 gauss. We find experimentally that the splittings are independent of field, disagreeing with the above estimate. Also, the observed ratio of the  $P^{31}$  splittings in  $PF_3$  and  $PH_3$  disagrees with the predicted ratio by a factor of ten. Lastly, the objections concerning chemically equivalent fluorines not interacting apply also to the second-order interaction.

In view of the arguments and data cited, both the rotational hindrance and direct second-order dipolar interaction mechanisms have serious flaws, and instead we wish to propose that the splittings arise from a second-order interaction between the nuclear magnetic moments and some magnetic field internal to the molecule. It seems to us that the most likely coupling is via the electrons by a mechanism analogous to the chemical shift. This view is supported to some extent by correlations found between observed values for the nuclear magnetic shielding and the splittings. Expressions similar to Ramsey's equations<sup>5</sup> have been derived, but the detailed calculations, as with Ramsey's equations, require a knowledge of molecular electronic wave functions. The expressions are similar also to Foley's equations<sup>6</sup> for the pseudo-quadrupole effect, except in our case we are concerned with coupling of different nuclei, rather than of a nucleus with itself. The order of magnitude in favorable cases appears to be similar to the fluorine effective field on itself; from Foley's paper one can calculate this field to be several tenths of a gauss.

The interaction that we propose is of the form  $A \mathbf{u}_1 \cdot \mathbf{u}_2$ , where  $A$  is a constant independent of temperature and  $H_0$ . Such a form can actually be obtained from arguments of rotational invariance alone, and thus would result from any form of coupling through molecular magnetic fields. In a private communication, E. L. Hahn has called our attention to the fact that, as a result of a selection rule, pairs of chemically equivalent nuclei will not give a split resonance from an interaction of the form we propose, whereas chemically nonequivalent nuclei will have a fine structure splitting in agreement with our observations. We have generalized Hahn's remark about pairs to the case of an arbitrary number of spins, and find the correct number, spacing, and intensities of lines. If the spin-spin interaction were proportional merely to the product of components of magnetic moment along  $H_0$ , there would be a splitting from interactions with chemically equivalent pairs. A more detailed account will appear in another journal.

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## Asymptotic Behavior of Angular Correlation Functions

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IN the standard theory of angular correlation,<sup>1</sup> one considers a "cascade process" consisting of a quick successive emission of two particles by a quantum-mechanical system at rest (e.g.,

an atomic nucleus emitting first an alpha-particle and, immediately after, a photon). If the direction of emission of, for example, the first emitted particle is chosen as the axis of quantization, then the relative probability  $W(\theta)$ , henceforth called "the correlation function," for the second particle to be emitted at an angle  $\theta$  with respect to this axis is given, according to the theory, by an expression of the form:

$$W(\theta) = \sum_{m_1} F_{J_1 m_1} \sum_m (J' m' | J_1 m_1 J m)^2 \Phi_m(\theta). \quad (1)$$

The symbols in Eq. (1) have the following meaning:  $J'$ ,  $m'$  and  $J$ ,  $m$  are the total angular momentum quantum numbers of the system before and after the emission of the first particle whose total angular quantum numbers are  $J_1$ ,  $m_1$ . The (real) transformation coefficient for the vector addition of these angular momenta is denoted by  $(J' m' | J_1 m_1 J m)$ . The  $F_{J_1 m_1}$ 's are numbers which depend on the nature and properties of the first particle as well as on  $J_1$ ,  $m_1$ . The  $\Phi_m(\theta)$ 's are functions of  $\theta$ , which depend solely on the characteristics of the second of the two transitions constituting the cascade process, i.e., on the angular momenta involved, and on the nature and properties of the second particle.

Let us now consider two cascade processes which are identical except for the nature and properties of the particle emitted in the first transition. In other words, all total angular momenta involved and the  $\Phi_m(\theta)$ 's are, respectively, equal in the two processes, but the set of the  $F_{J_1 m_1}$ 's for one process is different from that for the other. In general, the correlation functions for such two processes will, of course, be quite different. However, it will be shown below that, asymptotically, for  $J' \rightarrow \infty$  and  $J_1 \rightarrow \infty$ , they become identical. In other words: For large total angular momenta of the initial state of the system, and of the particle emitted in the first transition, the correlation function is always the same, no matter what the nature and properties of the particle are, provided all other characteristics of the cascade process are the same.<sup>2</sup>

In order to prove this proposition, it is sufficient to look into the asymptotic behavior of the transformation coefficients in Eq. (1). Starting from the well-known explicit expression for these coefficients (e.g., in the convenient form given by Van der Waerden<sup>3</sup>), one finds, by a simple calculation, the asymptotic equation:

$$(J' m' | J_1 m_1 J m) \sim J_1^J L(\lambda_1 J m), \quad (2a)$$

where

$$L(\lambda_1 J m) = [(J+m)!(J-m)!]^{-1} \sum_{\alpha} (-1)^{\alpha} \binom{J+m}{\lambda_1 - \alpha} \binom{J-m}{\alpha}. \quad (2b)$$

The parameter  $\lambda_1$  is defined by the equation  $J' = J_1 + J - \lambda_1$ . In the derivation of Eqs. (2),  $J_1$ ,  $m_1$ ,  $J$ ,  $m$ , and  $\lambda_1$  are kept constant, only  $J_1$  (and, hence,  $J'$  too) being allowed to increase indefinitely. Combining Eqs. (1) and (2), one obtains

$$W(\theta) \sim J_1^{2J} \{ \sum_{m_1} F_{J_1 m_1} \} \{ \sum_m [L(\lambda_1 J m)]^2 \Phi_m(\theta) \}. \quad (3)$$

Now, the correlation function being a relative probability, a  $\theta$ -independent factor does not change its physical meaning, and may be disregarded. One may thus drop both  $\theta$ -independent factors in Eq. (3). Hence, finally,

$$W(\theta) \sim \sum_m [L(\lambda_1 J m)]^2 \Phi_m(\theta). \quad (4)$$

This completes the proof of the proposition stated, for  $W(\theta)$ , as given by Eq. (4), is free from any reference to the nature and properties of the first particle.

Although the asymptotic form (4) of the correlation function does not depend on the total angular momentum quantum numbers of the initial state of the system and of the first particle, it does still depend, classically speaking, on the angle between the two angular momenta. For this is what the dependence of  $W(\theta)$  on  $\lambda_1$  in Eq. (4) means physically. The dependence on  $\lambda_1$  can be shown to disappear only if  $J \rightarrow \infty$  too. Of course, this mathematically possible case is very remote from any physically realizable situation. As an example, we may consider the case in which the particle emitted in the second transition is a photon whose total angular momentum (quantum number  $J_2$ ) is parallel to the total