Hyperfine field in ordered $Dy_{1-x}M_x Al_2$ (M = Y,Gd) compounds

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We report the results of nuclear-magnetic-resonance experiments on the ¹⁶³Dy nucleus in ferromagnetic alloys of $Dy_{1-x}Y_xAl_2$ and $Dy_{0.975}Gd_{0.025}Al_2$. The substitution of Dy by Y or Gd produces "satellite" lines in the ¹⁶³Dy NMR spectrum which we show are due to modification of the 4f hyperfine field. This modification is produced by a variation in the molecular field which modifies the angular momentum of the ground-state 4f wave function and hence the 4f moment. From the Gd substitution experiment we are able to estimate the molecular field appropriate to GdAl₂, which agrees with other experiments.

I. INTRODUCTION

Recently, there has been a considerable amount of work^{1,2} on the study of hyperfine fields in ferromagnetic Laves-phase compounds of the type GdM_2 (M = Al, Co, Fe, Rh, Ir, Zn) by NMR methods. There has been only one attempt other than the present work³ to study non-S-state ions in such ferromagnetic compounds and separate out the various contributions to the hyperfine field. The reason for the lack of study is partially experimental because of the necessity to have apparatus operating at high frequency (1.1 GHz for Dy), and partially because of the necessity to know accurately the dominant contribution to the hyperfine field which is that due to the orbital moment associated with the localized 4f electrons. With the present upsurge of interest in crystal fields⁴ it is possible to deduce accurately the groundstate magnetic moment of ions in ferromagnetic systems; thus, the orbital contribution to the hyperfine field can be separated out.

We have chosen to study the Laves-phase compound $DyAl_2$ because it is relatively simple to prepare and has a well-determined cubic structure involving 12 nearest neighbors of aluminum and four next-nearest neighbors of dysprosium. Furthermore, the NMR properties of ¹⁶³Dy are good. It is also fortunate that the crystal-field parameters and molecular field in $DyAl_2$ have been determined very accurately⁵ so that for this compound we can deduce the 4*f* electron wave functions for the ground state. In the following we will report the results of NMR measurements on 163 Dy in ferromagnetic compounds of DyAl₂, Dy_{1-x}Y_xAl₂, and Dy_{0.975}Gd_{0.025}Al₂. In the latter two cases the dilution with Y or Gd gives rise to satellite lines in the NMR spectrum. Contrary to current theory⁶ we shall show that these arise because dilution modifies the molecular field, which then gives rise to a modification of the ground-state moment of the Dy 4*f* electrons and consequently modifies the 4*f* field at the nucleus. For the case of Gd substitution of Dy we are able to predict accurately the molecular field in GdAl₂. Finally, we discuss various problems relating to the average molecular field as measured by, say, critical temperature, and its relevance in the NMR measurements.

II. EXPERIMENT

Samples were prepared by levitation melting of the appropriate quantities of rare earth and aluminum. The purity of the starting materials was 99.99% for the Al and 99.9% for the rare earth. Subsequent to melting the samples were crushed and sieved to obtain a maximum diameter of 80 μ m and then annealed for 60 h at 900°C in vacuum. Resonance lines obtained from unannealed samples were broad and poorly resolved.

The spin-echo NMR spectra of ¹⁶³Dy in $Dy_{1-x}Y_xAl_2$ (0 < x < 0.35) and $Dy_{0.975}Gd_{0.025}Al_2$ alloys were studied at 1.4 K in zero external field. For the sample containing no Y, a five-line spectrum was observed

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which could be fitted using the nuclear-spin (quadrupolar) Hamiltonian

$$H = A_0 I_z + 2P_0 [I_z^2 - \frac{1}{3}I(I+1)]$$

with A_0 equal to 1183.5 ± 0.2 MHz and $2P_0$ equal to 443.8 ± 0.4 MHz. The behavior of the central line $(\frac{1}{2} \leftrightarrow -\frac{1}{2}$ transition) as a function of Y concentration is shown in Fig. 1. As can be seen, when the Y concentration is increased, we see a broadening and small shift of the initially narrow line (L_0) and the appearance of satellite lines $(L_1, L_2...)$ whose amplitude increases to the detriment of the L_0 line.

We have also studied the $(\frac{3}{2} \leftrightarrow \frac{1}{2})$ transition $(\nu = 1627.3 \pm 0.2 \text{ MHz})$ for x = 0.1. This result is shown in Fig. 2, where one can see that the spacing between the satellite L_1 and L_0 depends upon the transition being studied. In the case of the $(\frac{1}{2} \leftrightarrow -\frac{1}{2})$ line we found $\nu(L_o) - \nu(L_1)$ equal to $5.5 \pm 0.5 \text{ MHz}$ and in the $(\frac{3}{2} \leftrightarrow \frac{1}{2})$ case, $8.5 \pm 0.5 \text{ MHz}$. This indicates that the quadrupolar term $2P_0$ is different for the satellite line L_1 , than for the central line L_0 .

Satellites have also been observed in the alloy $Dy_{0.975}Gd_{0.025}Al_2$, but in this case they occur at higher frequency than L_o rather than lower as for Y substitu-



FIG. 1. NMR spectra of the $(\frac{1}{2} \leftrightarrow -\frac{1}{2})$ transition of ¹⁶³Dy in Dy_{1-x}Y_xAl₂ alloys at 1.4 K showing the development of the satellite lines L_1, L_2, L_3 as a function of Y concentration.



FIG. 2. NMR spectrum of ¹⁶³Dy in Dy_{0.9}Y_{0.1}Al₂ for the $(\frac{1}{2} \leftrightarrow -\frac{1}{2})$ and $(\frac{3}{2} \leftrightarrow \frac{1}{2})$ transitions showing the central (L_0) and first satellite (L_1) lines at 1.4 K.



FIG. 3. NMR spectrum of ¹⁶³Dy in Dy_{0.975}Gd_{0.025}Al₂ for the $(\frac{1}{2} \leftrightarrow -\frac{1}{2})$ transition at 1.4 K.

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tion. The result is shown in Fig. 3. The spacing here $\nu(L_0) - \nu(L_1)$ is -4.0 ± 0.5 MHz.

The ferromagnetic ordering temperature T_c has also been measured in DyAl₂, Dy_{0.8}Y_{0.2}Al₂, and Dy_{0.6}Y_{0.4}Al₂ by low-field (~100 G) induction methods. The results indicate an approximate linear decrease in T_c extrapolating to zero for Dy_{0.2}Y_{0.8}Al₂ as has been found for other rare-earth Al₂ compounds.⁷

III. INTERPRETATION

We will consider the origin of the satellite lines to be related to the substitution of Dy atoms by Y or Gd which modifies the hyperfine field at a Dy nucleus through some physical mechanism. The intensity of a given satellite will thus be directly proportional to the statistical distribution of configurations giving rise to it (by configuration we mean a central Dy ion with 1, 2, 3, or 4 surrounding Dy ions replaced by Y or Gd). In Fig. 4 we show the calculated statistical "spectrum"; the frequency separation between satellites is assumed to be that found experimentally between L_0 and L_1 . From a comparison with the experimental results given in Fig. 1 it is evident that the physical process, whatever the origin, is correlated with the replacement



FIG. 4. Statistical "spectrum" based upon the probability distribution of the various configurations involving substitution of Dy nearest neighbors with Y.

of nearest-neighbor Dy ions. We proceed now to the physical origin of the variation in hyperfine field.

The hyperfine field at a rare-earth nucleus is assumed to be the sum of three contributions:

$$H = H_{4f} + H_{\rm sp} + H_{\rm nn}$$

where H_{4f} is the field created by the 4f electrons localized on the rare-earth site and is composed of parts due to orbit, spin, and core polarization.⁸ H_{sp} is the "self-polarization" field which arises due to polarization of the band electrons (say, s and d) by exchange with the 4f electron which subsequently is reflected in a conduction-electron contact (s) and a corepolarization (d) term:

$$H_{\rm sp} = A(z) \frac{J_s \chi_s \langle S_i \rangle}{g_e \mu_B} - \alpha_d \frac{J_d \chi_d \langle S_i \rangle}{g_e \mu_B} \quad . \tag{1}$$

 J_s and J_d are the exchange parameters for the s-f interaction and the d-f interaction. X_s and X_d are the sand d-band susceptibilities, and A(z) and α_d are the hyperfine-contact field per Bohr magneton⁹ and corepolarization field per Bohr magneton, respectively. The latter field is usually assumed to be about $\frac{1}{10}$ th of the contact field.¹⁰

Finally, the field produced by polarization of the conduction bands by neighboring ions (transferred hyperfine field) is given by H_{nn} . In the classic Rudermann, Kittel, Kasuya, and Yosida (RKKY) case¹¹ this has the form¹²

$$H_{nn} = (3n_s)^2 \frac{2\pi}{E_F} \frac{A(z)}{g_n \mu_n} J_s \sum_{m \neq n} F_s(2k_F R_{mn}) \langle S_m \rangle$$
$$- (3n_d)^2 \frac{2\pi}{E_F} \frac{\alpha_d}{g_n \mu_n}$$
$$\times J_d \sum_{m \neq n} F_d(2k_F R_{mn}) \langle S_m \rangle \quad , \qquad (2)$$

where F_s ($2k_F R_{mn}$) and F_d ($2k_F R_{mn}$) are RKKY functions, but we distinguish between them to allow for differences in the polarization and range of the s and d bands. Usually only polarization of the s band is considered. n_s and n_d are the numbers of s and d conduction electrons per atom, and $\langle S_m \rangle$ is the average value of the magnetic moment on the ion at site m, a distance R_{mn} away from the ion taken as origin.

Assuming that if we dilute $DyAl_2$ with Y or Gd we do not substantially modify the band structure¹³ we can assume that the self-polarization field [Eq. (1)] is independent of concentration. Variations in the hyperfine field that were measured must thus be due to variations in H_{4f} and H_{nn} . Traditionally, it is assumed that H_{4f} remains constant and this may be the case for ions with no orbital angular momentum; however, in the case where the ion has an orbital moment, e.g., Dy in $DyAl_2$, we will show that this may be erroneous.

According to Bleaney⁸ the 4f hyperfine field (including orbit, spin, and core polarization) may be approximated by

$$H_{4f} = A \langle J_z \rangle \quad , \tag{3}$$

where the definition of all parameters contained in A is given in Ref. 8.

A is known from measurements such as electron paramagnetic resonance in insulators⁸ and is found to be $152 \pm 3 \text{ MHz}/\mu_B$ for ¹⁶³Dy—we chose to quote the figure in MHz because our NMR measurements are made by sweeping frequency rather than field. It has been shown by Pelzl¹⁴ that the constants appearing in A vary little from the atomic to the insulator or metal case and we thus assume that the insulator result applies to the metallic case.

In the fully saturated weak-crystal-field case $\langle J_z \rangle$ equals $\frac{15}{2}$ for Dy; however, in our case the moment is not fully saturated and depends upon the crystal field and the internal molecular field. Using values found experimentally⁵ for $DyAl_2$, we have diagonalized the cubic crystal-field Hamiltonian including the molecular field and found $\langle J_z \rangle$ equal to 7.25. This yields a resonance frequency for Dy in DyAl₂ of 1105 ± 21 MHz. We have further calculated the variation of $\langle J_z \rangle$ with a molecular-field parameter (assuming constancy of crystal-field parameter). Using Eq. (4) and converting again to resonance frequency rather than field, we find the variation shown in Fig. 5. The initial slope of the frequency from the pure DyAl₂ value is found to be +0.247 MHz/kOe (equivalent to +1.24 kOe of H_{4f} per kOe of molecular field). It is now important to compare the variations in resonance frequency due to the 4f field (Δv_{4f}) and in the resonance frequency due to the transferred hyperfine field (Δv_{nn}) to see which is the most important and will create the experimentally observed satellites in dilution experiments. For simplicity we will work in the field and compare the variation of ΔH_{4f} (equal to +1.24 kOe per kOe of the molecular field) with ΔH_{nn} both within the framework of the RKKY model. The molecular field is given by

 $H_m = (3n_s)^2 \frac{2\pi}{E_F} \frac{J_s^2}{g_J \mu_B} \sum_{\substack{m \neq n}} F_s(2k_F R_{mn}) \langle S_m \rangle$ $+ (3n_d)^2 \frac{2\pi}{E_F} \frac{J_d^2}{g_J \mu_B} \sum_{\substack{m \neq n}} F_d(2k_F R_{mn}) \langle S_m \rangle \quad . \quad (4)$

Assuming that both for H_{nn} and H_m variations upon dilution of DyAl₂ with Y or Gd arise only from variations in the RKKY sums we can equate Eqs. (4) and (2) to give the ratio of ΔH_{4f} and ΔH_{nn} taking ΔH_{4f} equal to $1.24\Delta H_m$.



FIG. 5. Calculated variation of the 4*f* hyperfine field as a function of molecular field. Left-hand scale, resonance frequency in MHz; right-hand side, equivalent magnetic field (MOe) seen by the nucleus.

To proceed further we need to know the magnitudes of the various terms in Eqs. (2) and (4). Since we do not know a priori k_F nor the appropriate function $F_d(2k_F R_{mn})$ for a *d* band, we cannot at this point resolve the problem. From available NMR results on the transferred hyperfine field in various ordered Gdbased cubic compounds, the replacement of one nearest neighbor of Gd in GdAl₂ is found to produce a ΔH_{nn} of the order of 3.2 kOe. From Eq. (2) we expect the RKKY sums and their variations to be essentially the same in DyAl₂ as GdAl₂; we might thus expect the variation ΔH_{nn} for Dy to be of the order of 3 kOe. From experiment we find that the replacement of one Dy ion with Y causes a shift in resonance frequency of 5.5 ± 0.5 MHz corresponding to a field of 28 ± 2.8 kOe, which is an order of magnitude bigger than expected from the transferred hyperfine field. We thus conclude that the dominant factor in producing the satellites is the variation in H_{4f} provoked by local variation in the molecular field. Assuming therefore that the satellites are produced by variation of

 H_{4f} we conclude that one Dy ion produces a variation in H_{4f} through the molecular field of 28 ± 2.8 kOe. From the reasoning given previously, this is 1.24 times the variation in the molecular field which is thus 22 ± 2 kOe. Given that the molecular field⁵ in DvAl₂ is of the order of 252 kOe, this suggests that one nearest-neighbor Dy ion in DyAl₂ contributes (8.7 ± 0.8) % of the total molecular field. Within the framework of the RKKY model [Eq. (5)] the relative magnitude of the first-nearest-neighbor contribution to the total molecular field enables one to calculate k_F . However, again, in our case insufficient data is available for the various s and d terms to enable this to be done. If either s or d terms were dominant in the molecular field then one could argue from the RKKY sums for a value of k_F of the order of 1.2 Å⁻¹ which is somewhat smaller than usually assumed.¹⁵ However, it is possible that the two RKKY sums in Eq. (5) are of different signs so that cancellation effects occur in the sum of their variations and one cannot thus estimate k_F .

Variations in $H_{4f}(v_{4f})$ due to variations in molecular field can also be anticipated from knowledge of the variation of the ferromagnetic-ordering temperature T_c with concentration of nonmagnetic substituent. We have measured this behavior by the low-field magnetization measurement on various $Dy_{1-x}Y_xAl_2$ compounds. The results are presented in Fig. 6. As an example we study the variation of resonance frequency (v_{4f}) expected between the cases of $DyAl_2$ and $Dy_{0.2}Al_2$. From the variation of T_c we have deduced the variation of the molecular field and hence $\langle J_z \rangle$. We find

$$\langle J_z \rangle_{\mathrm{DyAl}_2} - \langle J_z \rangle_{\mathrm{Dy}_{0.8}\mathrm{Y}_{0.2}\mathrm{Al}_2} \approx 0.13,$$

which from Eq. (3) yields a variation of the resonance frequency of 20 ± 0.4 MHz. The experimental variation of the central resonance line of Dy (L_o) as a function of concentration is also included in Fig. 5. It can be seen that the measured variation in resonance frequency is more than one order of magnitude less than the calculated value. This strongly suggests that the local molecular field within a configuration of magnetic ions is not necessarily the average molecular field we infer from ordering-temperature measurement.

A. Quadrupolar splitting

In the presence of a quadrupolar field produced by the 4f electrons the resonance frequency is given by

$$\nu = A_0 + 2P_0(m_l - \frac{1}{2})$$

where A_0 is the resonance frequency in the absence of quadrupole terms. P_0 has the form $P'[3J_z^2 - J(J+1)]$ where $\langle J_z \rangle$ is the z component of the angular momen-



FIG. 6. Measured variation of critical temperature in $Dy_{1-x}Y_xAl_2$ alloys as a function of Y concentration, left-hand scale, temperature; right-hand scale, equivalent average molecular field. The dashed curve shows the variation of the molecular field as deduced from the frequency shift of the central resonance line (L_0) with Y concentration.

tum for the 4f electron and J is the total angular momentum $(\frac{15}{2}$ for Dy). P' contains the quadrupole moment and $\langle r_Q^{-3} \rangle$ for the 4f electrons.⁸ The spacing between the $\frac{1}{2} \leftrightarrow -\frac{1}{2}$ and $\frac{3}{2} \leftrightarrow \frac{1}{2}$ transition is thus given by 2P₀; hence, if J_z varies, the frequency separation between these two transitions will vary through variation in 2P₀. We have measured the splitting in Dy_{0.9}Y_{0.1}Al₂ between the $\frac{1}{2} \leftrightarrow -\frac{1}{2}$ and $\frac{3}{2} \leftrightarrow \frac{1}{2}$ lines both for the first satellite and the central line. The measured ratio of splittings is $2P_0(L_1)/2P_0(L_0) = 0.992 \pm 0.004$. For the satellites we determined from the resonance-frequency shift the value $\langle J_z \rangle$ equal to 7.216, whilst for the central line we have $\langle J_z \rangle$ equal to 7.25. Taking the ratio

$$\left(3J_z^2 - \frac{15\times17}{4}\right)_{\text{sat}} / \left(3J_z^2 - \frac{15\times17}{4}\right)_{\text{cent}}$$

we evaluate the variation in quadrupole term (P_0) to

be 0.990 ignoring variation in quadrupole moment and $\langle r_Q^{-3} \rangle$. Within the range of experimental accuracy the calculated and experimental results agree supporting our conclusions on the variation of $\langle J_z \rangle$ although some variation in $\langle r_Q^{-3} \rangle$ might be present due to distortion of the environment in replacement of Dy with Y.

B. Self-polarization field

The resonance frequency due solely to the 4f field was calculated to be 1105 ± 21 MHz or the equivalent of 5.53 ± 0.11 MOe. Subtracting the total estimated¹ transferred hyperfine field of the order of -30 kOe and the 4*f* field from the experimental value of 5.915 MOe leaves an equivalent self-polarization field of 409 ± 114 kOe. This value is much larger than the value of +190 kOe found¹ for GdAl₂. From Eq. (1) we would expect, assuming the exchange parameters for DyAl₂ and GdAl₂ are the same, the selfpolarization field to vary as $\langle S_i \rangle$. Given $\langle S_i \rangle$ of $\frac{7}{2}$ for Gd and $\frac{5}{2}$ for Dy leads to a predicted self-polarization field of +140 kOe for DyAl₂ which is a factor of 2.9 ± 1 less than the value found from experiment. Even if we were wrong in the value of $\langle J_z \rangle$ for the 4f field and we assumed the maximum possible value of $\frac{15}{2}$ this would still lead, after subtraction of 4f field and transferred hyperfine field, to a discrepancy of two between measured H_{sp} and that predicted from GdAl₂. The only other variable present is the exchange in the formula for H_{sp} but since nowhere in the literature is there evidence for a significant difference between the exchange for Gd and Dy ions we conclude that the origin of the discrepancy in the self-polarization fields must lie elsewhere.

IV. RESULTS FOR Dy 0.975Gd0.025Al2

Upon substitution of a Dy first-nearest neighbor with Gd we observe a satellite line at 4.0 ± 0.5 MHz on the high-frequency side of the central Dy resonance. Given that one Dy neighbor produces a 5.5 MHz shift (as deduced from Y substitution), we conclude that one nearest neighbor of Gd produces a total shift which is the sum, i.e., 9.5 ± 0.5 MHz. Using this result with the calculated conversion factor of 0.247 MHz per kOe of molecular field this is equivalent to a molecular field per Gd neighbor of 38 ± 2 kOe. In the case of Dy in DYAl₂ we found the first-neighbor field to be 8.7% of the total field; the same percentage applied to the Gd result enables us to extrapolate our result to the equivalent case of the molecular field seen by one Dy in pure GdAl₂. Taking 38 ± 2 kOe to be 8.7%, the total molecular field seen by a Dy ion in pure GdAl₂ would be 437 ± 24 kOe.

In a system containing two magnetic constituents, α and β , the Zeeman energy of the β ion is the molecu-

lar field H_m created by the exchange interaction with α ions is

$$g_J^{\beta} \mu_B H_m J^{\beta} = \kappa (g_J - 1)^{\alpha} (g_J - 1)^{\beta} \langle J^{\alpha} \rangle J^{\beta} \quad , \tag{5}$$

where κ is the Heisenberg exchange parameter.¹⁶

In our measurements we have measured the molecular field produced by a Gd ion (α) at a Dy ion (β). To deduce the appropriate molecular field produced by a Gd ion at another Gd ion (e.g., the case of pure GdAl₂), we must take account of the differences in g₁ in Eq. (5), i.e., H_m (as seen by a Gd ion in GdAl₂)/H_m (as seen by a Dy ion in GdAl₂) ≈ 2 .

Hence the molecular field in pure GdAl₂ (as seen by the Gd ions) is the value deduced from our extrapolation of the field seen by a Dy ion in GdAl₂ (i.e., 437 ± 24 kOe) times 2. Thus we deduce the molecular field in GdAl₂ to be 874 ± 48 kOe.

Noting that the ordering temperature is proportional to $g_J J (J+1) H_m / J_z$, and substituting the known values of 252 kOe for H_m in DyAl₂ and $\langle J_z \rangle$ of 7.25 together with the value of 874 ± 48 kOe deduced for H_m (GdAl₂), we find

$$T_c(\mathrm{GdAl}_2)/T_c(\mathrm{DyAl}_2) \approx 2.66 \pm 0.75$$

The experimental ratio is¹⁶ 2.63 which is very close to the value we are able to predict. Thus it seems that the molecular field in GdAl₂ can be accurately predicted from NMR experiments on alloys of $DyAl_2$ with Gd substitution for Dy.

V. CONCLUSION

The results presented represent the first case of the complete separation of the various contributions to the field at the nucleus of a non-S-state rare-earth ion in a metallic environment. We have been able to conclude from measurements with Y or Gd substitution for Dy that the origin of the satellite lines so produced is the variation in $\langle J_z \rangle$ due to perturbation of the molecular field. This contradicts the usually accepted view that variation in the transferred hyperfine field is predominant.⁶

A study of the concentration dependence of the resonance frequency for the Dy nucleus surrounded by four nearest-neighbor Dy ions (line denoted L_0), which is essentially the "normal" nearest-neighbor environment, suggests that the molecular field seen within such a configuration is not the average field deduced via measurement of the ordering temperature T_c . This suggests that, for an alloy containing various configurations of nearest-neighbor magnetic ions around a central ion, the molecular field is spatially variant. We are unable from the measurements, however, to determine how the statistical average of the various molecular fields should be taken so as to reproduce the average value found from T_c measurement.

The results for the self-polarization field (H_{sp}) in DyAl₂ and GdAl₂ are not consistent in that we would have expected, as shown earlier, H_{sp} for Dy to be smaller than for Gd, whereas it is considerably larger. Measurements on other rare-earth alloys are in pro-

gress to see if this discrepancy is systematic. The experiments involving substitution of Gd for Dy in DyAl₂ show that the molecular field in GdAl₂ can be accurately predicted from such measurements. This has potential application for systems such as PrFe₂ which are not stable under ordinary atmospheric conditions, but whose properties could be studied via dilution experiments on stable compounds such as DyFe₂ with Pr substituted for Dy.

Finally, it should be commented that satellite lines have also been observed³ in the NMR spectrum of the Ho nucleus in $Gd_xY_{1-x}Co_2$ upon dilution of Gd with Y. The crystal field and molecular field should, of course, play similar roles as in $Dy_{1-x}Y_xAl_2$. However, the system is much more complicated because the Co carries a moment which will produce a transferred hyperfine field at the Ho nucleus. Furthermore, the moment of the Co is itself induced¹⁷ by the presence of Gd so that substitution of Gd with Y will not only vary the transferred hyperfine field from the Gd ions. At the present time the simplest approach to understanding the hyperfine field at the nucleus of a non-Sstate ion in ferromagnetic compounds appears to be via measurements on systems containing only one magnetic constituent.

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- ¹E. Dormann, M. Huck, and K.H.J. Buschow, J. Magnetism Magn. Mat. <u>4</u>, 47 (1977).
- ²K. E. Eckrich, E. Dormann, A. Oppelt, and K.H.J. Buschow, Z. Phys. B <u>23</u>, (157 (1976).
- ³S. K. Arif, J. W. Ross, and M. A. H. McCausland, Physica (Utr.) <u>86-88 B and C</u>, 158 (1977).
- ⁴See, for example, Proceedings of the Second International Conference on Crystal Field Effects in Metals and Alloys, Zurich, 1976 (unpublished).
- ⁵B. Barbara, M. F. Rossignol, and P. Bak (unpublished).
- ⁶G. K. Shenoy, A. Hessel, G. M. Kalvius, W. Zinn, and W. Wiedmann, Conference on Perspectives for Hyperfine Interactions in Magnetically Ordered Systems by NMR and other Methods, L'Aquila, 1972 (unpublished).
- ⁷K. H. J. Buschow, J. F. Fast, A. M. van Diepen, and H. W. de Wijn, Phys. Status Solidi <u>24</u>, 715 (1967).

- ⁸B. Bleaney, in *Magnetic Properties of Rare-Earth Metals*, edited by R. J. Elliot (Plenum, New York, 1972), Chap. 8.
- ⁹I. A. Campbell, J. Phys. C <u>2</u>, 1338 (1969).
- ¹⁰R. E. Watson and A. J. Freeman, in *Hyperfine Interactions* edited by A. J. Freeman and R. B. Frankel, (Academic, New York, 1967), Chap. 2.
- ¹¹See, for example, K. N. R. Taylor and M. I. Darby, in *Physics of Rare-Earth Solids* (Chapman and Hall, London, 1972), Chap. 4.
- ¹²R. A. B. Devine and Y. Berthier, J. Phys. F 7, L53 (1977).
- ¹³A. C. Switendick, Proceedings of the Tenth Rare-Earth Research Conference, Colorado, 1973 (unpublished).
- ¹⁴J. Pelzl, Z. Phys. <u>251</u>, 15 (1972).
- ¹⁵E. Dormann, K. H. J. Buschow, K. N. R. Taylor, G. Brown, and M. A. A. Issa, J. Phys. F <u>3</u>, 220 (1973).
- ¹⁶W. E. Wallace, in *Rare-Earth Intermetallics* (Academic, New York, 1973), p. 35.
- ¹⁷R. Lemaire (private communication).