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Orientation of Nuclei in Ferromagnets*

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A theoretical discussion is given of the various orientation effects acting on nuclei in ferromagnets, and the results are compared to experimental measurements on the specific heat and the anisotropy in the emission of γ rays by radioactive Co^{60}. The orientation is due to an effective magnetic field which is, neglecting some small terms, the sum of one negative and four positive contributions all of which are roughly comparable. The uncertainty in the total field predicted is considerable but nevertheless rough agreement is obtained with the few experimental results at present available.

I. INTRODUCTION

RECENTLY several experiments on the orientation of nuclei in ferromagnets have been performed. Craer, Johnson, Scurlock, and Taylor[1] have observed the anisotropy in the γ-ray emission from radioactive orientated Co nuclei in Co metal; Johnson and Scurlock[2] have repeated these experiments on alloys of Co in Ni and Co in Fe; Heer and Erickson[3] have measured the nuclear specific heat of Co metal, and Kurti, Arp, and Peterson[4] have measured the nuclear specific heat of pure Co and of a Co-Ni alloy. The purpose of this paper is to analyze theoretically the various orientation effects so as to determine what may be learned from these experiments.

Because all the experiments performed so far have used Co nuclei, we shall be chiefly concerned with the orientation of these nuclei in pure Co and in Co alloys. The calculations are difficult to make accurately and there is a very considerable uncertainty in the final results. However, it is hoped that some more experimental work will give us a better understanding of some of the difficult problems involved. In Sec. II the various orientation effects are written down, and the fields which are not critically sensitive to the symmetry of the lattice are estimated. In Sec. III cubic alloys are considered in detail, and in Sec. IV hexagonal cobalt is discussed. In Sec. V the results obtained are discussed and compared to experiment. In an Appendix the use of an effective field is justified and formulas for the specific heat are derived.

II. EFFECTIVE MAGNETIC FIELD

The orientation of the nuclei may be regarded as due to an effective magnetic field H which may be written as

$$H=H_l+H_c+H_a, \tag{1}$$

where H_l is the local magnetic field at the position of the nucleus, H_c is the effective magnetic field which acts through the contact interaction with the 4*s* electrons and H_a is the effective field due to the interactions of the nucleus with the electrons on the same atom. This effective magnetic field gives rise to a nuclear specific heat

$$C_V/R=\tfrac{1}{3}I(I+1)(g_N\mu_N H/kT)^2+O(g_N\mu_N H/kT)^4+\cdots, \tag{2}$$

and, if the aligned nuclei are radioactive Co^{60}, to an anisotropy in the emission of γ rays given by

$$\epsilon=\frac{I(\pi/2)-I(0)}{I(\pi/2)}=\frac{39}{14}\left(\frac{g_N\mu_N H}{kT}\right)^2+O(g_N\mu_N H/kT)^4+\cdots. \tag{3}$$

Here $I(\theta)$ is the intensity of the emission of γ rays in a direction inclined at an angle θ to the aligned spins.

It is not obvious that these effects can be discussed in terms of an effective field as we have used here, and, in fact, Heer and Erickson give a formula for the specific heat which is incorrect for this particular problem. Therefore a discussion of this concept of an effective magnetic field is given in the Appendix where it is shown that apart from unimportant terms, (2) is correct. The essential idea is that in a ferromagnet at low temperatures the electron spins are completely "locked in" by the strong exchange forces. Therefore lowering the temperature still further does not affect the electron spin system, and so the electrons serve only the static role of providing an effective magnetic field to act on the nucleus.

The local magnetic field at the nucleus H_l is given by

$$H_l=H_e-DM+\tfrac{4}{3}\pi M+H', \tag{4}$$

where H_e is the external field, $-DM$ the demagnetizing field depending only on the shape of the specimen, $4\pi M/3$ the usual Lorentz field and H' the small residue of the Lorentz field which exists only for noncubic symmetry. We are interested in cubic ferromagnets and in hexagonal close-packed cobalt; for the former, H' is rigorously zero, and for the latter, H' is only about 10^{-3}

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[1] Craer, Johnson, Scurlock, and Taylor (to be published); Grace, Johnson, Kurti, Scurlock, and Taylor, *Conference de Physique des Basses Temperatures, 1955* (Centre National de la Recherche Scientifique and UNESCO, Paris, 1956).

[2] C. Johnson and R. Scurlock (private communication).

[3] C. V. Heer and R. A. Erickson, Phys. Rev. **108**, 896 (1957).

[4] Kurti, Arp, and Peterson (to be published).

of the Lorentz field.[5] We may, therefore, neglect H' completely.

For this problem there is no doubt that the Lorentz field $4\pi M/3$, rather than the Onsager field, should be used because we are concerned here with the fields produced by the large electron moments acting on the small nuclear moments. The reaction of the nuclear moments back onto the electron moments can be neglected. The values of the saturation magnetization M to be inserted into (4) are 1752 for iron, 1446 for cobalt, and 510 for nickel.[6]

The effective field H_c which is due to the contact interaction with the 4s electrons may be regarded as the sum of two parts. The first part is that proportional to the conduction electron polarization p and is

$$-(8\pi/3)\mu|\psi(0)|^2np,$$

where μ is the Bohr magneton, $|\psi(0)|^2$ is the average probability density of a conduction electron evaluated at a lattice point, n is the number of conduction electrons per atom, and p is their polarization. The second part is that due to the mixing of 4s functions into the 3d band. This mixing we expect to be reasonably large (several percent) although it is extremely difficult to make any kind of estimate for it. The sum of these two parts gives an effective field

$$H_c=-(8\pi/3)\mu|\psi(0)|^2n\tilde{p}, \tag{5}$$

where

$$\tilde{p}=(p+2\bar{S}a^2/n) \tag{6}$$

is the "effective polarization." Here $\bar{S}$ is the mean spin per atom and a^2 is an average of the amount of 4s wave function mixed into a mainly 3d wave function. Both p and $\bar{S}$ are negative because the g value for an electron is negative, so H_c is a positive field.

As usual we write

$$|\psi(0)|^2=\xi|\psi(0)|_A{}^2, \tag{7}$$

where $|\psi(0)|_A{}^2$ is the free atom value and ξ is a correction factor. We estimate $|\psi(0)|_A{}^2$ as follows. The hyperfine coupling constant $a(s)$, defined as

$$a(s)=(16\pi/3)g_N\mu_N\mu|\psi(0)|_A{}^2,$$

is known to be 0.190 cm^{-1} for copper. Using a mean value of the two copper isotopes for g_N, we find that for copper

$$|\psi(0)|_A{}^2=0.310\times10^{26}\ \text{cm}^{-3}, \quad [\text{for Cu}].$$

Ridley[7] has made estimates of these probability densities using extrapolation from Hartree calculations on Fe and Zn and finds a value of 0.277×10^{26} for copper and 0.233×10^{26} for cobalt. In view of the difficulty of these calculations the agreement between theory and experiment for copper is very satisfactory. We assume that the theoretical estimate for cobalt is in error by the same factor as that for copper, and so take

$$|\psi(0)|_A{}^2=0.267\times10^{26}\ \text{cm}^{-3}, \quad [\text{for Co}]. \tag{8}$$

This gives

$$H_c=-2.07\xi n\tilde{p}\times10^6\ \text{gauss}. \tag{9}$$

ξ will have a value somewhere between 0.5 and 1.0.

A rough estimate of the "true" polarization p and of the corresponding part of H_c can be made as follows. Suppose the interaction between a single conduction electron, normalized to a unit cell volume, and an atom be written as $-2J\mathbf{S}\cdot\mathbf{s}$, where $\mathbf{s}$ is the conduction electron spin and $\mathbf{S}$ is the atom spin. This interaction is equivalent to an effective magnetic field acting on the conduction electron of magnitude $J\bar{S}/\mu$. Now the Pauli paramagnetic susceptibility of a free electron gas is $3n\mu^2/2E_f$, where E_f is the Fermi energy. From this susceptibility and the effective magnetic field we find

$$p=3J\bar{S}/2E_f. \tag{10}$$

Assuming a free electron gas for the conduction electrons of cobalt, we have

$$E_f=0.93\times10^{-11}\ \text{erg}.$$

In the free atom J is roughly 2.4×10^{-13} erg. Substituting these values and setting $\bar{S}$ equal to -0.85, we get

$$p\approx-3.3\times10^{-2}, \quad [\text{for Co}] \tag{11}$$

and from (9) we get a value of roughly 4×10^4 gauss for the corresponding part of H_c. This must be regarded only as a very rough estimate because ξ is unknown and J may be expected to differ from the free atom value by a significant factor.

The effective field H_a can be estimated if we use the Van Vleck model of ferromagnetism. In this model we suppose that correlation effects are very important so that on any particular atom there will always be an integer number of 3d electrons coupled together according to Hund's rule. Thus, for example, in cobalt we suppose 70% of the atoms to have the $(3d)^8\,{}^3F$ configuration and 30% to have $(3d)^9\,{}^2D$. This gives the mean moment of 1.7 μ per atom. Of course the same configurations do not stay on the same atom but are continually changing position.

Using this model, we want to estimate the fields acting on the nucleus of an atom with a $(3d)^9$ and with a $(3d)^8$ configuration, and then average with the appropriate probabilities. The field for each configuration can be obtained from the usual spin Hamiltonian which is used to study paramagnetic salts. The relevant terms are

$$\mathcal{H}=AS_zI_z+B(S_xI_x+S_yI_y). \tag{12}$$

In the ferromagnet the electron spins are aligned by the strong exchange interactions, so we may replace S_x and S_y by zero, and S_z by the spin value S of the

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[5] L. W. McKeehan, Phys. Rev. **43**, 1025 (1933).

[6] C. Kittel, *Introduction to Solid State Physics* (John Wiley and Sons, Inc., New York, 1953), p. 407.

[7] C. Ridley (private communication).

configuration. Hence we obtain

$$H_a = -A\bar{S}/g_N\mu_N. \tag{13}$$

The full justification for this step is given in the Appendix. In the usual notation, the quantity A is given by

$$A = -P(\kappa + 3Jl_{zz} + 2\lambda\Lambda_{zz} - 3J\lambda u_{zz}), \tag{14}$$

where

$$P = 2g_N\mu_N\mu\langle 1/r^3\rangle, \quad J = \frac{5-4S}{21(2L-1)S}, \tag{15}$$

$$l_{\alpha\beta} = \tfrac{1}{2}\langle 0|L_\alpha L_\beta + L_\beta L_\alpha|0\rangle - \tfrac{1}{3}L(L+1)\delta_{\alpha\beta}, \tag{16}$$

$$\Lambda_{\alpha\beta} = \sum_n{}' \frac{\langle 0|L_\alpha|n\rangle\langle n|L_\beta|0\rangle}{(E_n - E_0)}, \tag{17}$$

$$u_{\alpha\beta} = -\tfrac{1}{2}i\epsilon_{\alpha\beta\varphi}\sum_n{}' \frac{\langle 0|L_\varphi|n\rangle\langle n|L_\beta L_\gamma + L_\gamma L_\beta|0\rangle}{(E_n - E_0)}. \tag{18}$$

In these expressions λ is the spin-orbit coupling constant, and the states $|n\rangle$ are the orbital states in the crystalline field. In writing (14) we have assumed that the ground state is quenched, so that there is no orbital contribution to the moment or to A, other than that introduced by spin-orbit coupling.

The first term of (14), $-P\kappa$, has been discussed extensively by Abragam, Horowitz, and Pryce[8] and arises through the contact interaction of the nucleus with the inner $3s$ and $2s$ electrons. These inner electrons see slightly different potentials, depending upon whether their spins are parallel or antiparallel to the spin of the $3d$ electrons. They therefore have slightly different amplitudes at the nucleus and, although this difference is only a small percentage, the effect is appreciable because the probability density of these electrons at the nucleus is very large. The magnitude of this effect has been estimated with only moderate success by Abragam, Horowitz, and Pryce,[8] by Heine,[9] and by Wood and Pratt.[10] Experimentally it is observed that the quantity $P\kappa/g_N$, which is independent of nuclear properties, is roughly constant for paramagnetic ions and neutral atoms. We might hope therefore that it is roughly constant for different metals. Abragam, Horowitz, and Pryce quote the values

$\kappa P/g_N = 3.8\times 10^{-3}$ cm^{-1} for neutral cobalt,

$\kappa P/g_N = 5.3\times 10^{-3}$ cm^{-1} for Co^{++}.

Using the neutral cobalt value and denoting the corresponding effective magnetic field by H_{a1}, we find

$$H_{a1} = -12.8\times 10^4 \text{ gauss}. \tag{19}$$

It seems quite likely that this represents the order of magnitude of this effect in the metal too, because the inner electron wave functions and the $3d$ electron wave functions should not differ drastically from the free atom. But possibly it may be that the effect is sensitive to the exact overlap of the wave functions and so H_{a1} may differ considerably from this value.

The second term of (14) represents the dipole-dipole magnetic interaction between the nuclear and electron spins averaged over the zero-order wave function. The third term is the magnetic interaction between the nucleus and the unquenched part of the orbital moment. The fourth term is a cross effect of the dipole-dipole nuclear-electron spin interaction and the spin-orbit coupling. These three terms depend upon the symmetry of the crystal, so we shall discuss separately the two cases we are particularly interested in, cubic alloys containing cobalt and pure hexagonal cobalt, in Sec. III and Sec. IV, respectively.

III. CUBIC ALLOYS

In alloys we may still regard cobalt as a mixture of two configurations, 70% of $(3d)^8\,{}^3F$ and 30% of $(3d)^9\,{}^2D$. In the cubic field which has the same sign as is usual in paramagnetic salts, the $(3d)^8\,{}^3F$ configuration splits into the representations $\Gamma_2+\Gamma_4+\Gamma_5$, with the Γ_2, i.e. a singlet orbital state, lowest. Hence for the $(3d)^8\,{}^3F$ configuration the ground state to be inserted in (16) is nondegenerate and, therefore, has the full cubic symmetry. Therefore,

$$l_{xx} = l_{yy} = l_{zz} = \tfrac{1}{3}l_{\alpha\alpha} = 0. \tag{20}$$

This is just the well known result that the dipole-dipole interactions average to zero in cubic symmetry. The $(3d)^9\,{}^2D$ configuration splits into the representations $\Gamma_3+\Gamma_5$, with the Γ_3, a doublet orbital state, lowest. For neither of these two orbital states separately does $l_{\alpha\beta}$ vanish (except for special choices of representation); but because the two states are degenerate, and in the metal they remain degenerate because there is no distortion to split them as there usually is in paramagnetic salts, we are interested only in the mean value of $l_{\alpha\beta}$ and this is zero. Hence the second term of (14) and the corresponding contribution H_{a2} to H_a are zero for cubic alloys.

This argument has presupposed that each cobalt atom in the alloy sees a cubic field, and this will be strictly true only in very dilute or in very concentrated alloys. For alloys of intermediate concentrations each cobalt atom will have some nearest neighbors of cobalt and some of a different atom so that the field seen by this cobalt atom will not be cubic. Only the average field seen by all the cobalt atoms will be cubic. This effect will, therefore, give no contribution to the mean field acting on the nucleus, but it will give a contribution to the mean square field and hence to the specific heat and γ-ray anisotropy. It does not seem possible to estimate this term; but in alloys between elements which are close in the periodic table, e.g., Co-Ni, we

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[8] Abragam, Horowitz, and Pryce, Proc. Roy. Soc. (London) **A230**, 169 (1955).

[9] V. Heine, Phys. Rev. **107**, 1002 (1957).

[10] J. H. Wood and G. W. Pratt, Jr., Phys. Rev. **107**, 995 (1957).

might hope the effect is small even for the 50% concentration alloy, especially as the effect of a noncubic field on the $(3d)^8$ configuration must be small, so that effectively only the 30% of the atoms which have the $(3d)^9$ configuration can give rise to this extra term.

The third term of (14) will always be roughly of the same order of magnitude and can be estimated as follows. Using (13) and (14), we find the corresponding part of H_a is

$$H_{a3}=S(2-g')2\mu(1/r^3). \tag{21}$$

Using $S=0.85$, a typical value of 1.9 for g', and a value of 5.2 a_0^{-3} for $1/r^3$, we find H_{a3} is about 6×10^4 gauss. However, for any particular case a more careful estimate of this term can easily be made, although in view of the uncertainties in the experimental value of g' and in the model we are using, there is considerable doubt if such an estimate is really justified. To illustrate the calculation, consider a small percentage of cobalt in nickel.

We assume that the cobalt sees the same cubic field as nickel. If we write this field as

$$V=C[x^4+y^4+z^4-\tfrac{3}{5}r^4], \tag{22}$$

we find, remembering that the easy direction of magnetization in Ni is the [111] direction so the subscript z in (14) really refers to this direction, not the crystallographic z axis,

$$\begin{aligned}\Lambda_{zz}&=21/C\langle r^4\rangle_{\text{Av}} \quad \text{for } (3d)^8\,{}^3F,\\ \Lambda_{zz}&=10.5/C\langle r^4\rangle_{\text{Av}} \quad \text{for } (3d)^9\,{}^2D.\end{aligned} \tag{23}$$

We calculate the coefficient C, using the known g' value of pure nickel, from the relation

$$(2-g')=-2\lambda_1 10.5/C\langle r^4\rangle_{\text{Av}}, \tag{24}$$

where λ_1 is the spin-orbit parameter for $\text{Ni}(3d)^9$. Using (13), (14), and (23) we find that, after averaging over configurations,

$$H_{a3}=\mu(2-g')(1/r^3)(0.3\lambda_2+2.8\lambda_3)/\lambda_1, \tag{25}$$

where λ_2 and λ_3 are the spin-orbit parameters for Co $(3d)^9$ and $\text{Co}(3d)^8$, respectively. Now theoretically

$$\lambda=\pm\xi/2S,$$

where ξ is a function only of the nuclear charge. Hence

$$\lambda_1=-\xi_{\text{Ni}}, \quad 2\lambda_3=\lambda_2=-\xi_{\text{Co}}. \tag{26}$$

Experimentally we find ξ_{Ni} and ξ_{Co} are 670 cm^{-1} and 540 cm^{-1}, respectively. Upon using these values with (26), Eq. (25) becomes

$$H_{a3}=1.4\mu(2-g')(1/r^3) \quad [\text{for Co in Ni}]. \tag{27}$$

If we use a value of 1.88 ± 0.04 for g' and 5.2 a_0^{-3} for $1/r^3$, this gives

$$H_{a3}=[5.4\pm1.8]\times10^4 \text{ gauss}. \tag{28}$$

The last term of (14) vanishes for this case because in cubic symmetry $u_{zz}=\frac{1}{3}u_{\alpha\alpha}$, which is zero by inspection of (18).

IV. HEXAGONAL COBALT

The crystalline field potential can be written as

$$V=a\{3z^2-r^2\}-b\{35z^4-30z^2r^2+3r^4\}. \tag{29}$$

The coefficient a is zero for close-packing so we shall neglect this first term, as the cobalt lattice is very nearly ideal close-packing. The coefficient b is positive. In this potential the $\text{Co}(3d)^9\,{}^2D$ configuration splits as follows:

Energy	States	Degeneracy
0	ψ_0	1×2
$-40b\langle r^4\rangle_{\text{Av}}/21$	$\psi_2;\psi_2$	2×2
$-80b\langle r^4\rangle_{\text{Av}}/21$	$\psi_1;\psi_{-1}$	2×2

and the $\text{Co}(3d)^8\,{}^3F$ configuration splits as follows:

Energy	States	Degeneracy
$104b\langle r^4\rangle_{\text{Av}}/21$	$\psi_2;\psi_{-2}$	2×3
$40b\langle r^4\rangle_{\text{Av}}/21$	$\psi_1;\psi_{-1}$	2×3
$24b\langle r^4\rangle_{\text{Av}}/21$	$\psi_3;\psi_{-3}$	2×3
0	ψ_0	1×3

Then

$$\begin{aligned}&\text{for}\quad \text{Co}(3d)^9\,{}^2D, \quad l_{zz}=-1, \quad J=2/21, \quad \bar{S}=-\tfrac{1}{2};\\ &\text{for}\quad \text{Co}(3d)^8\,{}^3F, \quad l_{zz}=-4, \quad J=1/105, \quad \bar{S}=-1.\end{aligned} \tag{30}$$

From (13), (14), and (30) we obtain, averaging over configurations,

$$H_{a2}=0.25\mu(1/r^3)=8.1\times10^4 \text{ gauss}. \tag{31}$$

The third term of (14) needs some care. It is easily verified that Λ_{zz} is zero both for $(3d)^9\,{}^2D$ and for $(3d)^8\,{}^3F$, but this does not mean that there is no interaction between the nucleus and the orbital moment because, in this case, in the first approximation the orbital moment of the $(3d)^9\,{}^2D$ configuration is not quenched. The quenching is done by the band-like motion of the $(3d)^9$ configurations in this case. The small orbital contribution which is left produces a field

$$H_{a3}=0.85(2-g')P/g_N\mu_N. \tag{32}$$

If we use a value of 1.85 for g', this is

$$H_{a3}=8.3\times10^4 \text{ gauss}. \tag{33}$$

The last term of (14) is difficult to calculate because the coefficient b in (29) is unknown. A reasonable estimate however shows the last term, H_{a4}, to be roughly only 1% of H_{a3}, so we may safely neglect it.

V. COMPARISON WITH EXPERIMENT

There are several experimental results on hexagonal cobalt available. Craer, Johnson, Scurlock, and Taylor[1] have observed the anisotropy in the γ-ray emission from Co^{60}, and from their results they deduce that, if the electron-nuclear interaction is written as AS^zI^z with $S^z=\pm\frac{1}{2}$, then for Co^{59}, A is 0.0145 ± 0.002 cm^{-1}. Since Co^{59} has a spin of $\frac{7}{2}$ and a moment of $4.64\,\mu_N$, we deduce

$$H=21.7\times10^4 \text{ gauss.} \qquad (34)$$

Heer and Erickson[3] have measured the specific heat of hexagonal Co^{59} and found

$$C_VT^2/R=4.0\times10^{-4}.$$

Hence, from (2), we have

$$H=18\times10^4 \text{ gauss.} \qquad (35)$$

Arp, Peterson, and Kurti[4] have also measured the specific heat and they found

$$C_VT^2/R=4.7\times10^{-4},$$

giving

$$H=19.5\times10^4 \text{ gauss.} \qquad (36)$$

We shall take the experimental value to be the mean of these three results, namely 19.7×10^4 gauss. Actually these experiments can measure only the magnitude and not the sign of H. We have assumed the sign to be positive in writing (34)–(36).

The total field which we have calculated for hexagonal cobalt is the sum of (9), (19), (31), and (33) and is

$$[1.45\xi(|p|+2.43a^2)\times10^6+3.6\times10^4] \text{ gauss.} \qquad (37)$$

Taking ξ to be 1.0 we find this consistent with the value 19.7×10^4 gauss, if we accept the estimate (11) for p and set a^2 equal to 0.032. This conclusion that the mainly $3d$ wave function contains about 3% of a $4s$ wave function seems very reasonable. From this result it would seem likely that the term H_{a1} does not differ too much from the free atom value, and is therefore not too sensitive to the exact overlap of the wave functions.

It would be very interesting to have experimental results on a high-concentration cubic Co alloy. Here we would expect all the fields to have roughly the same value, except for H_{a2}, which would become zero. We, therefore, expect a value of 11.5×10^4 gauss for H in this case. An experimental measurement of this quantity therefore ought to give information on the validity of the Van Vleck model which we have used. A complication is that on this model the quenching mechanism is different for hexagonal cobalt and cubic cobalt. We might therefore expect H_{a3} to change appreciably, too.

Experimental results on a high-cobalt-concentration cubic alloy are not yet available, but Arp and Peterson have reported measurements on an alloy of 60% Co, 40% Ni which give an effective field of 16.2×10^4 gauss. It is not clear how close we would expect the results for this alloy to come to the predicted result of 11.5×10^4 gauss for the high-Co-concentration alloy. It would appear from this result that the use of the Van Vleck model has exaggerated the size of the field H_{a2} by a factor of about two.

The total field which we have calculated for cobalt in nickel is

$$[1.45\xi(|p|+2.43a^2)\times10^6-7.4\times10^4] \text{ gauss.}$$

Assuming that the first term has the same value as in hexagonal cobalt, we get an effective field of 8.7×10^4 gauss. Hence the anisotropy in the emission of γ rays ought to be a fraction 0.19 of that for hexagonal cobalt; i.e., since Craer, Johnson, Scurlock, and Taylor observed an anisotropy of 5.7% at a temperature of 0.040°K for hexagonal cobalt, an anisotropy of 1.1% should be observed at the same temperature for a dilute alloy of cobalt in nickel. This is in rough agreement with some preliminary experiments of Johnson and Scurlock[2] on an alloy of 3% cobalt. They report that the anisotropy is very small.

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APPENDIX

In this Appendix we shall justify the use of an effective field to describe the nuclear spin orientations. For simplicity of notation we shall use the simple Heisenberg model of a ferromagnet with a spin S on every atom. We write the Hamiltonian for the system as

$$\mathcal{H}=\mathcal{H}_0+\mathcal{H}', \qquad (A.1)$$

where

$$\mathcal{H}_0=-2J\sum_{\langle i,j\rangle}\mathbf{S}_i\cdot\mathbf{S}_j+2\mu H_e\sum_i S_i^z, \qquad (A.2)$$

$$\mathcal{H}'=A\sum_i S_i^zI_i^z+B\sum_i(S_i^xI_i^x+S_i^yI_i^y)-2g_N\mu_NH_e\sum_i I_i^z. \qquad (A.3)$$

$\mathcal{H}_0$ is the Hamiltonian for the electron system alone, and $\mathcal{H}'$ is the nuclear Hamiltonian. Actually these expressions are not complete because the dipole-dipole interactions between the electron spins themselves, between the nuclei themselves, and between the electrons and nuclei have all been omitted (except for the dipole interaction between the nucleus and the electrons of the same atom which is included in the coefficients A and B). The neglect of the nuclear-nuclear dipole interactions is certainly justified and the neglect of the other terms will have as a consequence merely the replacement of the local field H_l, defined by (4), by the external field H_e.

From (A.3), with zero H_e and setting $S=\frac{1}{2}$, Heer and Erickson deduce the specific heat to be

$$C_V/R=I(I+1)(A^2+2B^2)/12k^2T^2. \tag{A.4}$$

This would be correct for a paramagnetic salt but, as we shall show, is incorrect for a ferromagnet. The correct expression, for zero H_e, is

$$C_V/R=I(I+1)A^2S^2/3k^2T^2. \tag{A.5}$$

Following Schafroth[11] we write the partition function, correct to second order in $\mathcal{H}'$, as

$$\begin{aligned}Z=Z_0-\beta\sum_n e^{-\beta E_n}\langle n|\mathcal{H}'|n\rangle\\ +\tfrac{1}{2}\beta\sum_{n,m}|\langle m|\mathcal{H}'|n\rangle|^2\frac{e^{-\beta E_n}-e^{-\beta E_m}}{(E_m-E_n)},\end{aligned} \tag{A.6}$$

where β is $1/kT$ and the states $|n\rangle$ are the eigenstates of $\mathcal{H}_0$. Z_0 is a partition function for the noninteracting system. The second term of (A.6) certainly vanishes because it is linear in $\mathcal{H}'$. To evaluate the second term we first diagonalize $\mathcal{H}_0$, using the familiar spin-wave operators. With the usual approximations, we have

$$\begin{aligned}S_i^z&=-S+a_i^*a_i,\\ S_i^+&=(2S)^{\frac{1}{2}}a_i^*(1-a_i^*a_i/2S)^{\frac{1}{2}}\simeq(2S)^{\frac{1}{2}}a_i^*\\ &=(2S/N)^{\frac{1}{2}}\textstyle\sum_{\mathbf{k}}(e^{i\mathbf{k}\cdot\mathbf{R}_i}a_{\mathbf{k}}^*),\\ S_i^-&=(2S)^{\frac{1}{2}}(1-a_i^*a_i/2S)^{\frac{1}{2}}a_i\simeq(2S)^{\frac{1}{2}}a_i\\ &=(2S/N)^{\frac{1}{2}}\textstyle\sum_{\mathbf{k}}(e^{-i\mathbf{k}\cdot\mathbf{R}_i}a_{\mathbf{k}}).\end{aligned} \tag{A.7}$$

Then

$$[a_i,a_j^*]=\delta_{ij};\quad [a_{\mathbf{k}},a_{\mathbf{k}'}^*]=\delta_{\mathbf{k},\mathbf{k}'}\ \text{etc.},$$

and, if we neglect all terms except those quadratic in the annihilation and creation operators, $\mathcal{H}_0$ is brought to the diagonal form

$$\mathcal{H}_0=C+\textstyle\sum_{\mathbf{k}}\epsilon_{\mathbf{k}}n_{\mathbf{k}}, \tag{A.8}$$

where

$$\begin{aligned}&n_{\mathbf{k}}=a_{\mathbf{k}}^*a_{\mathbf{k}},\quad \epsilon_{\mathbf{k}}=2\mu H+2SJ\gamma_{\mathbf{k}},\\ &\gamma_{\mathbf{k}}=\textstyle\sum_{\boldsymbol{\varrho}}\exp(i\mathbf{k}\cdot\boldsymbol{\varrho}),\end{aligned} \tag{A.9}$$

and the sum over $\boldsymbol{\varrho}$ goes over nearest neighbors. Then

$$Z_0=(2I+1)^Ne^{-\beta C}\textstyle\prod_{\mathbf{k}}[1-\exp(-\beta\epsilon_{\mathbf{k}})]^{-1}. \tag{A.10}$$

In terms of these variables, $\mathcal{H}'$ becomes

$$\begin{aligned}\mathcal{H}'=-\textstyle\sum_i I_i^z\{AS+2g_N\mu_NH-(A/N)\\ \times\textstyle\sum_{\mathbf{k},\mathbf{k}'}e^{i(\mathbf{k}-\mathbf{k})\cdot\mathbf{R}_i}a_{\mathbf{k}}^*a_{\mathbf{k}'}\}+B(S/2N)^{\frac{1}{2}}\textstyle\sum_i I_i^x\\ \times\textstyle\sum_{\mathbf{k}}\{e^{-i\mathbf{k}\cdot\mathbf{R}_i}a_{\mathbf{k}}+e^{i\mathbf{k}\cdot\mathbf{R}_i}a_{\mathbf{k}}^*\}+iB(S/2N)^{\frac{1}{2}}\textstyle\sum_i I_i^y\\ \times\textstyle\sum_{\mathbf{k}}\{e^{-i\mathbf{k}\cdot\mathbf{R}_i}a_{\mathbf{k}}-e^{i\mathbf{k}\cdot\mathbf{R}_i}a_{\mathbf{k}}^*\}.\end{aligned} \tag{A.11}$$

Then we find that the last term of (A.6) is the thermal average of

$$\begin{aligned}&\tfrac{1}{6}I(I+1)N\beta Z_0\beta[AS+2g_N\mu_NH-(A/N)\textstyle\sum_{\mathbf{k}}n_{\mathbf{k}}]^2\\ &+\frac{A^2}{N^2}\sum_{\mathbf{k}\neq\mathbf{k}'}(n_{\mathbf{k}}+1)n_{\mathbf{k}'}\frac{1-\exp[-\beta(\epsilon_{\mathbf{k}}-\epsilon_{\mathbf{k}'})]}{\epsilon_{\mathbf{k}}-\epsilon_{\mathbf{k}'}}\\ &+\frac{B^2S}{N}\sum_{\mathbf{k}}\frac{1}{\epsilon_{\mathbf{k}}}\{(n_{\mathbf{k}}+1)[1-\exp(-\beta\epsilon_{\mathbf{k}})]\\ &\qquad-n_{\mathbf{k}}[1-\exp(\beta\epsilon_{\mathbf{k}})]\}.\end{aligned} \tag{A.12}$$

Remembering that

$$\langle n_{\mathbf{k}}n_{\mathbf{k}'}\rangle_{\text{Av}}=\bar{n}_{\mathbf{k}}\bar{n}_{\mathbf{k}'}+\delta_{\mathbf{k},\mathbf{k}'}\bar{n}_{\mathbf{k}}(\bar{n}_{\mathbf{k}}+1), \tag{A.13}$$

and that

$$\bar{S}=-S+(1/N)\textstyle\sum_{\mathbf{k}}n_{\mathbf{k}}, \tag{A.14}$$

we get

$$\begin{aligned}Z/Z_0=1&+\tfrac{1}{6}\beta^2NI(I+1)(A\bar{S}-2Hg_N\mu_N)^2\\ &+\tfrac{1}{3}\beta NI(I+1)\frac{A^2}{N^2}\sum_{\mathbf{k},\mathbf{k}'}\left(\frac{1}{\epsilon_{\mathbf{k}}-\epsilon_{\mathbf{k}'}}\right)\left(\frac{1}{\exp(\beta\epsilon_{\mathbf{k}'})-1}\right)\\ &+\tfrac{1}{3}\beta NI(I+1)B^2\frac{S}{N}\sum_{\mathbf{k}}\frac{1}{\epsilon_{\mathbf{k}}}.\end{aligned} \tag{A.15}$$

From this we deduce, in zero field, that

$$\begin{aligned}C_V=C_V{}^0&+\frac{RI(I+1)}{3k^2T^2}A^2\left[\bar{S}^2+4\beta\bar{S}\frac{\partial\bar{S}}{\partial\beta}+\beta^2\frac{\partial}{\partial\beta}\left(\bar{S}\frac{\partial\bar{S}}{\partial\beta}\right)\right]\\ &+\frac{RI(I+1)}{3k^2T^2}\frac{A^2}{N^2}\frac{\partial^2}{\partial\beta^2}\left[\beta\sum_{\mathbf{k},\mathbf{k}'}\left(\frac{1}{\epsilon_{\mathbf{k}}-\epsilon_{\mathbf{k}'}}\right)\right.\\ &\qquad\left.\times\left(\frac{1}{\exp(\beta\epsilon_{\mathbf{k}'})-1}\right)\right].\end{aligned} \tag{A.16}$$

Now

$$\bar{S}=-S+c\beta^{-\frac{3}{2}}, \tag{A.17}$$

where, for a face-centered cubic lattice,

$$c=\frac{1}{8\pi^2}\left(\frac{1}{2SJ}\right)^{\frac{3}{2}}\int_0^\infty\frac{x^{\frac{1}{2}}dx}{e^x-1}=0.0294\left(\frac{1}{2SJ}\right)^{\frac{3}{2}}. \tag{A.18}$$

From (A.17) and (A.16) we find the leading terms in an expansion in inverse powers of β to be

$$\begin{aligned}C_V=C_V{}^0+\frac{RI(I+1)A^2}{3k^2T^2}\Big[&S^2+\tfrac{1}{4}Sc\beta^{-\frac{3}{2}}+c^2\beta^{-3}\\ &+\tfrac{3}{4}c\beta^{-\frac{5}{2}}\frac{v_0}{(2\pi)^3}\int d\mathbf{k}\frac{1}{\epsilon_{\mathbf{k}}}\Big].\end{aligned} \tag{A.19}$$

The last three terms in the brackets are negligible compared to the first term and $C_V{}^0$ is also negligible, so we obtain (A.5). Clearly the other effects which we have ignored in this simple model can be taken care of by replacing AS by $-g_N\mu_NH$, where H is given by (1).

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[11] M. R. Schafroth, Helv. Phys. Acta **24**, 645 (1951).