in the general picture of the process seems required except that the necessity for a threshold in the applied voltage is eliminated.

Electroluminescence with dc excitation of similar films has led to brightnesses of six hundred foot-lamberts, with efficiency rapidly increasing with brightness or current. These additional results, together with response times, transient, and polarity effects, will be described elsewhere.

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Ferromagnetic Resonance Linewidth in Cobalt-Substituted Ferrites*

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The effect of small concentrations of substitutional ions on the ferromagnetic resonance linewidth of a host ferrite is calculated. The resonance linewidth arises from magnon scattering induced by the variation from ion-to-ion of the spin-orbit interaction. This interaction is uniquely large for cobalt ions because of the orbital degeneracy of the ground state of the ion in a trigonally-symmetric crystalline field. The resultant contribution to the linewidth is found to be isotropic and of the order of 20–30 oersteds for each percent of cobalt in normal (i.e., noninverse) ferrites or in ferrous ferrite. In other ferrites the effect is diminished by the lifting of the ground-state orbital degeneracy at some of the cobalt sites; this diminution is calculated as a function of the degree of inversion.

I. INTRODUCTION

IN typical ferrites, containing any of a large number of substitutional ions, the intrinsic ferromagnetic resonance linewidths are of the order of 10 oersteds. However, the cobalt ion is unique in that even small admixtures of cobalt increase the linewidth drastically.

A contribution to the linewidth in typical ferrites (without cobalt) has been calculated by Callen and Pittelli[1] on the basis of magnon scattering induced by the random variations from ion to ion of the spin-orbit and intra-atomic spin-spin coupling parameters. The resulting linewidth contribution is anisotropic, and is of the observed order of magnitude.[2]

We consider specifically the contribution of small concentrations of cobalt ions to the linewidth. The effect of the spin-orbit coupling is enhanced in this ion because it has a degenerate orbital ground state when situated in a crystalline field of trigonal symmetry. This leads to three important differences in our results: (1) The linewidth contribution of a cobalt ion is considerably larger than that of a typical (nondegenerate) ion. (2) The dominant linewidth contribution of a cobalt ion is isotropic. (3) The linewidth contribution of a cobalt ion can be strongly influenced by the degree of inversion of the host ferrite, because of the destruction of the trigonal symmetry and lifting of the ground state degeneracy.

It is, of course, clear that variations from ion to ion of other parameters also lead to spin-wave scattering, and thence to linewidth. The strongest interaction at play is the exchange interaction, but because the exchange commutes with the total spin, variations of exchange cannot scatter magnons of zero wave number and hence cannot contribute to the linewidth. Another source of possible scattering is the variation in anisotropy. For cobalt ions the anisotropy is particularly large, of the order of 10^5 oersteds, again because of the degeneracy of the ground state, as shown by Slonczewski.[3] For one percent cobalt the mean square fluctuating anisotropy field is of the order of $0.01 \times (10^5)^2 = 10^8$ oersteds^2. However, Callen and Pittelli have shown that in the absence of short-range order it requires locally varying fields with mean square magnitudes of the order of 10^{10} oersteds^2 to produce a linewidth of 10 oersteds. Consequently, the linewidth resulting from variations in anisotropy field is negligible.

Another mechanism leading to magnon scattering in disordered ferrites is the variation from ion to ion of the magnitude of the spin. Callen and Pittelli did not calculate this effect because it is clearly isotropic and consequently is distinguishable from their anisotropic linewidth. However, as we shall see, the linewidth contribution of cobalt ions arising from spin-orbit coupling is also isotropic, and it therefore becomes

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† National Science Foundation Cooperative Graduate Fellow during part of this work.
[1] H. B. Callen and E. Pittelli, Phys. Rev. **119**, 1523 (1960).
[2] The first calculation of this kind was done by A. M. Clogston, H. Suhl, L. R. Walker, and P. W. Anderson, J. Phys. Chem. Solids **1**, 129 (1956), on the basis of assumed variations of the pseudodipolar interaction (which subsequently were found to be too small).
[3] J. C. Slonczewski, Phys. Rev. **110**, 1341 (1958).

necessary to examine the role of spin-magnitude fluctuations more carefully. This we do in the Appendix, where we show that fluctuations in spin magnitude do not make an appreciable contribution to the linewidth. We are left, therefore, with the variation of the spin-orbit interaction as the only mechanism of appreciable linewidth, and this we calculate.

The large linewidth contribution of the cobalt ions depends on the cobalt ions being situated in trigonally-symmetric crystalline fields, which is true only if all the ions on octahedral sites of the host ferrite are equivalent. If all ions on octahedral sites are not equivalent, some cobalt ions find themselves in non-trigonally symmetric environments. In certain configurations, this deviation from trigonal symmetry removes the ground-state degeneracy and thereby decreases the linewidth. In the case of a completely inversed ferrite $MeFe_2O_4$ we find that the linewidth contribution of additive cobalt ions is (for small concentrations) about $\frac{1}{5}$ of the contribution in normal (i.e., noninverse) ferrites.

Finally, we note that the linewidth is determined by the scattering of magnons of zero wave vector both into other magnons and into phonons. The scattering mechanism which we consider is a magnon-magnon scattering mechanism. The role of each type of scattering in determining the response of a ferromagnetic system to an applied field has been discussed by Callen,[4] and methods of experimentally distinguishing the various scattering mechanisms have been discussed by Fletcher, Le Craw, and Spencer[5] and by Seiden.[6] Because these methods have not been applied to ferrites, we cannot ascribe the observed linewidths to either predominant magnon-magnon or magnon-phonon scattering. However, there is some evidence, in addition to the rough agreement of our linewidth predictions with experiment, that the magnon-magnon scattering does dominate in the ferrites. This evidence arises from the contrast between the properties of the rare earth substituted garnets and the ferrites. Kittel[7] has postulated a rapid spin-lattice relaxation of the rare earth ions in the garnets and has shown that this leads to g values which correlate well with experiment; in the ferrites a similar mechanism would lead to g values which would be extremely sensitive to the degree of inversion, for which there does not appear to be any experimental evidence. In addition the unique and characteristic temperature dependence of both g values and linewidth, predicted and observed in the garnets[7,8] does not appear to apply to the ferrites. White,[9] in discussing the applicability of Kittel's direct spin-lattice relaxation to the ferrites has cited the observed correlation between linewidth and anisotropy; this correlation is, of course, also present in our spin-orbit scattering theory and does not provide a way to distinguish between spin-lattice and spin-spin mechanisms.

[4] H. B. Callen, J. Phys. Chem. Solids 4, 256 (1958).
[5] R. C. Fletcher, R. C. Le Craw, and E. G. Spencer, Phys. Rev. **117**, 955 (1960).
[6] P. E. Seiden (unpublished preprint, Institut Fourier, University of Grenoble).
[7] C. Kittel, Phys. Rev. **115**, 1587 (1959).
[8] P. G. de Gennes, C. Kittel, and A. M. Portis, Phys. Rev. **116**, 323 (1959).
[9] R. L. White, Phys. Rev. Letters **2**, 465 (1959).

II. ORIGIN OF SCATTERING TERM

An ion on an octahedral site of a spinel is surrounded by six nearest-neighbor oxygen ions and six next-nearest-neighbor metallic ions which also occupy octahedral sites.[10] If one neglects a slight displacement (i.e., a deviation of the "oxygen parameter" from its ideal value of 0.375), the oxygen ions are at the corners of a regular octahedron centered at the octahedral site. The six nearest octahedral sites exhibit trigonal symmetry about one of the body diagonals $[111]$, $[1\bar{1}\bar{1}]$, $[\bar{1}11]$, $[\bar{1}\bar{1}1]$ of the cubic crystal, depending upon the particular octahedral site being considered. If the metallic ions which occupy these neighboring octahedral sites are all the same, the octahedral site under consideration is in a trigonal environment.

In a ferrite with complete inversion, half the ions on the octahedral sites are ferric ions and half are divalent metal ions. Consequently, in such ferrites one generally has lower symmetry (i.e., nontrigonal) crystalline fields at the octahedral site. However, an interesting special case is presented by magnetite above the order-disorder transformation; here the ferrous and ferric ions rapidly interchange by electron transfer. From resonance experiments, this transfer time has been estimated[11] as 10^{-12} sec. A similar estimate is obtained by considering the conductivity of magnetite above the transition point. However, the period of precession of the spins is about 10^{-10} sec in a typical ferromagnetic resonance experiment. Therefore, in magnetite, resonance and loss processes are governed by the time-averaged charge distribution, which has trigonal symmetry.

We consider first a cobalt ion on an octahedral site of magnetite, or of any noninverse ferrite, and hence in the presence of cubic and trigonal crystalline fields. Abragam and Pryce[12] have discussed the effect of cubic and trigonal crystalline fields on the orbital states of the Co^{2+} ion.

In the Co^{2+} free ion, the lowest orbital state is a ^4F state with a ^4P state about 10^4 cm^{-1} higher. The cubic portion of the crystalline field (arising from the nearest neighbor oxygen ions) splits the sevenfold degeneracy of the ^4F state into two triplets and a singlet and in addition admixes some of the ^4P state. We consider only the lowest lying triplet and neglect all the higher states since they lie about 10^4 cm^{-1} above this triplet.

[10] See, for example, E. W. Gorter, Philips Research Repts. **9**, 295 (1954).
[11] W. A. Yager, J. K. Galt, and F. R. Merritt, Phys. Rev. **99**, 1203 (1955).
[12] A. Abragam and M. H. L. Pryce, Proc. Roy. Soc. (London) **A205**, 135 (1951); **A206**, 173 (1951).

The trigonal field splits this triplet into a lower lying doublet and a singlet which lies about 10^3 cm^{-1} above. The orbital angular momentum of the doublet state lies along the axis of trigonal symmetry; a body diagonal. In what follows we consider only this lowest lying doublet.

Abragam and Pryce find it convenient to introduce a fictitious orbital angular momentum operator $\mathbf{l}'$. If the z' axis is the body diagonal of trigonal symmetry and the x' and y' axes are perpendicular to the z' axis, the matrix representation of $\mathbf{l}'$ in a convenient and known representation of the orbital doublet is

$$l_{z'}'=\begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix},\quad l_{x'}'=l_{y'}'=\begin{pmatrix}0 & 0\\ 0 & 0\end{pmatrix}. \tag{1}$$

The real angular momentum is given by the relations $L_{z'}=-\alpha l_{z'}'$, $L_{x'}=0$, $L_{y'}=0$ where α is determined by the strength of the cubic crystalline field and the energy interval between the unperturbed ^4F and ^4P states.

Consider the effect of spin-orbit coupling, $\lambda\mathbf{L}\cdot\mathbf{S}$, on the doublet. One of the states is lowered by an amount $\alpha\lambda S_{z'}$ and the other state is raised by the same amount. Thus the final ground state—the lower state of the doublet—is acted on by an effective perturbing potential of the form, $-\alpha\lambda S_{jz'}$, where $S_{jz'}$ is the component of the spin along the trigonal symmetry axis of the jth site.

For simplicity we consider a simple cubic array of magnetic ions situated on four different kinds of sites. At each site we assume a local trigonal crystalline field with an axis of symmetry along one of the four body diagonals, the sites being otherwise equivalent.

H_0 is the average Hamiltonian, containing the exchange, Zeeman, and dipolar terms with the ionic parameters replaced by their average values.

As discussed in the Introduction and in the Appendix, the variations of the ionic parameters from the average values contribute appreciably to the resonance linewidth only in the case of the spin-orbit coupling parameters.

In the notation of Callen and Pittelli,[1] the effective Hamiltonian is then $H=H_0+V$ with

$$V=-\alpha\lambda\sum_{j,t}P_{jt}(q_j-c)(\boldsymbol{\varepsilon}_t\cdot\mathbf{S}_j), \tag{2}$$

where t is an index taking four values, indicating in turn the trigonal axis directions, [111], [1$\bar{1}\bar{1}$], [$\bar{1}$1$\bar{1}$], and [$\bar{1}\bar{1}$1]; $\boldsymbol{\varepsilon}_t$ is a unit vector along the tth axis; $P_{tj}=1$ if site j has a tth trigonal axis, =zero otherwise; $q_j=1$ if site j contains a Co^{2+} ion, =zero otherwise; and $c=\langle q_j\rangle_{\text{av}}$, the fraction of Co^{2+} ions present. The part of V involving c has been added so that H_0 is indeed the average Hamiltonian.

It is this effective perturbing potential, V, which produces magnon-magnon scattering. In terms of an effective perturbing field, $V/\gamma\hbar$, the perturbation has the magnitude $\approx|\alpha\lambda|/\gamma\hbar$. Slonczewski[3] finds $|\alpha\lambda|=132$ cm^{-1}, which corresponds to an effective field of 10^6 oersteds. Consequently, the mean square fluctuating field is, for 1% cobalt ions, $0.01\times(10^6)^2=10^{10}$ oersteds^2, which, according to the estimate given in the Introduction, should produce a linewidth of the order of 10 oersteds. Our actual calculation gives 20–30 oersteds for each percent of cobalt.

III. MAGNON SCATTERING

The spin operator $\mathbf{S}_j$ in Eq. (2) is conveniently resolved along x, y, z axes, the z axis being chosen along the direction of the magnetization $\mathbf{M}$:

$$\mathbf{S}_j=\boldsymbol{\alpha}''S_{jx}+\boldsymbol{\alpha}'S_{jy}+\boldsymbol{\alpha}S_{jz}. \tag{3}$$

The unit vector $\boldsymbol{\alpha}$ in the z direction has components α_1, α_2, α_3, which are the direction cosines of the magnetization with respect to the crystal axes ξ, η, ζ. Similar relations exist for $\boldsymbol{\alpha}'$ and $\boldsymbol{\alpha}''$, and

$$\begin{bmatrix}S_x\\ S_y\\ S_z\end{bmatrix}=\begin{bmatrix}\alpha_1'' & \alpha_2'' & \alpha_3''\\ \alpha_1' & \alpha_2' & \alpha_3'\\ \alpha_1 & \alpha_2 & \alpha_3\end{bmatrix}\begin{bmatrix}S_\xi\\ S_\eta\\ S_\zeta\end{bmatrix}. \tag{4}$$

Introducing the relations

$$S_j^{\pm}=S_{jx}\pm iS_{jy}, \tag{5}$$

we find that

$$(\boldsymbol{\varepsilon}_t\cdot\mathbf{S}_j)=(\boldsymbol{\varepsilon}_t\cdot\boldsymbol{\alpha})\left(S-\frac{S_j^-S_j^+}{2S}\right)+\tfrac{1}{2}[\boldsymbol{\varepsilon}_t\cdot(\boldsymbol{\alpha}''-i\boldsymbol{\alpha}')]S_j^+ +\tfrac{1}{2}[\boldsymbol{\varepsilon}_t\cdot(\boldsymbol{\alpha}''+i\boldsymbol{\alpha}')]S_j^-. \tag{6}$$

The spin-wave variables $a(\mathbf{k})$, $a^\dagger(\mathbf{k})$ are defined by

$$S_j^+=(2S/N)^{\frac{1}{2}}\sum_{\mathbf{k}}e^{-i\mathbf{k}\cdot\mathbf{r}_j}a^\dagger(\mathbf{k}), \tag{7}$$

$$S_j^-=(2S/N)^{\frac{1}{2}}\sum_{\mathbf{k}}e^{+i\mathbf{k}\cdot\mathbf{r}_j}a(\mathbf{k}). \tag{8}$$

In terms of these variables, V becomes

$$V=-\alpha\lambda\sum_{j,t}P_{tj}(q_j-c)\times\Bigg[(\boldsymbol{\varepsilon}_t\cdot\boldsymbol{\alpha})\left(S-\frac{1}{N}\sum_{\mathbf{k},\mathbf{k}'}a(\mathbf{k})a^\dagger(\mathbf{k}')e^{+i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}_j}\right) +\tfrac{1}{2}\boldsymbol{\varepsilon}_t\cdot(\boldsymbol{\alpha}''-i\boldsymbol{\alpha}')\left(\frac{2S}{N}\right)^{\frac{1}{2}}\sum_{\mathbf{k}}a^\dagger(\mathbf{k})e^{-i\mathbf{k}\cdot\mathbf{r}_j} +\tfrac{1}{2}\boldsymbol{\varepsilon}_t\cdot(\boldsymbol{\alpha}''+i\boldsymbol{\alpha}')\left(\frac{2S}{N}\right)^{\frac{1}{2}}\sum_{\mathbf{k}}a(\mathbf{k})e^{+i\mathbf{k}\cdot\mathbf{r}_j}\Bigg]. \tag{9}$$

The Holstein-Primakoff transformation,[13] which diagonalizes the unperturbed Hamiltonian, is

$$a^\dagger(\mathbf{k})=\cosh(\lambda_k/2)\sigma^\dagger(\mathbf{k})-\sinh(\lambda_k/2)e^{2i\phi_k}\sigma(-\mathbf{k}), \tag{10}$$

$$a(\mathbf{k})=\cosh(\lambda_k/2)\sigma(\mathbf{k})-\sinh(\lambda_k/2)e^{-2i\phi_k}\sigma^\dagger(-\mathbf{k}), \tag{11}$$

[13] T. Holstein and H. Primakoff, Phys. Rev. 58, 1098 (1940).

with

$$\omega(\mathbf{k})\cosh\lambda_k=\gamma[H-4\pi MN_z+H_e a^2k^2+2\pi M\sin^2\theta], \quad (12)$$

$$\omega(\mathbf{k})\sinh\lambda_k=\gamma[2\pi M\sin^2\theta], \quad (13)$$

$$\omega(\mathbf{k})=\gamma[(H-4\pi MN_z+H_e a^2k^2)(H-4\pi MN_z+H_e a^2k^2+4\pi M\sin^2\theta)]^{\frac{1}{2}}, \quad (14)$$

where H_e = exchange field, and a = interatomic distance. Then

$$V=-\alpha\lambda\sum_{j,t}P_{tj}(q_j-c)\Big\{(\boldsymbol{\varepsilon}_t\cdot\boldsymbol{\alpha})\Big[S-\frac{1}{N}\sum_{\mathbf{k},\mathbf{k}'}e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}_j}[\cosh(\lambda_k/2)\cosh(\lambda_{k'}/2)\sigma(\mathbf{k})\sigma^\dagger(\mathbf{k}')$$
$$+\sinh(\lambda_k/2)\sinh(\lambda_{k'}/2)e^{2i(\phi_{k'}-\phi_k)}\sigma^\dagger(-\mathbf{k})\sigma(-\mathbf{k}')-\cosh(\lambda_k/2)\sinh(\lambda_{k'}/2)e^{2i\phi_{k'}}\sigma(\mathbf{k})\sigma(-\mathbf{k}')$$
$$-\sinh(\lambda_k/2)\cosh(\lambda_{k'}/2)e^{-2i\phi_{k'}}\sigma^\dagger(-\mathbf{k})\sigma^\dagger(\mathbf{k}')]\Big]$$
$$+\tfrac{1}{2}\boldsymbol{\varepsilon}_t\cdot(\boldsymbol{\alpha}''-i\boldsymbol{\alpha}')\left(\frac{2S}{N}\right)^{\frac{1}{2}}\sum_{\mathbf{k}}e^{-i\mathbf{k}\cdot\mathbf{r}_j}[\cosh(\lambda_k/2)\sigma^\dagger(\mathbf{k})-\sinh(\lambda_k/2)e^{2i\phi_k}\sigma(-\mathbf{k})]$$
$$+\tfrac{1}{2}\boldsymbol{\varepsilon}_t\cdot(\boldsymbol{\alpha}''+i\boldsymbol{\alpha}')\left(\frac{2S}{N}\right)^{\frac{1}{2}}\sum_{\mathbf{k}}e^{i\mathbf{k}\cdot\mathbf{r}_j}[\cosh(\lambda_k/2)\sigma(\mathbf{k})-\sinh(\lambda_k/2)e^{-2i\phi_k}\sigma^\dagger(-\mathbf{k})]\Big\}. \quad (15)$$

The terms of zero order in the annihilation and creation operators are of course constant. The first-order terms do not contribute to scattering and can be eliminated by an appropriate transformation. The terms which can result in the destruction of one magnon and the creation of another are (neglecting an irrelevant change in notation because of this last transformation)

$$\frac{\alpha\lambda}{N}\sum_{j,t}\sum_{\mathbf{k},\mathbf{k}'}P_{tj}(q_j-c)(\boldsymbol{\varepsilon}_t\cdot\boldsymbol{\alpha})e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}_j}[\cosh(\lambda_k/2)\cosh(\lambda_{k'}/2)\sigma(\mathbf{k})\sigma^\dagger(\mathbf{k}')$$
$$+\sinh(\lambda_k/2)\sinh(\lambda_{k'}/2)e^{2i(\phi_{k'}-\phi_k)}\sigma^\dagger(-\mathbf{k})\sigma(-\mathbf{k}')]. \quad (16)$$

The matrix element between a $k=0$ spin-wave state and a $k\neq0$ spin-wave state is then

$$M_{0k}=\frac{\alpha\lambda}{N}\sum_{j,t}P_{tj}(q_j-c)(\boldsymbol{\varepsilon}_t\cdot\boldsymbol{\alpha})\cosh(\lambda_k/2)e^{i\mathbf{k}\cdot\mathbf{r}_j}. \quad (17)$$

If we take an ensemble average over all the distributions of the ions and assume that there is no correlation in the positioning of the cobalt ions, we find

$$\langle|M_{0k}|^2\rangle_{\mathrm{av}}=\frac{(\alpha\lambda)^2}{4N}c(1-c)\cosh^2(\lambda_k/2)\sum_t(\boldsymbol{\varepsilon}_t\cdot\boldsymbol{\alpha})^2. \quad (18)$$

It is easily shown that

$$\sum_t(\boldsymbol{\varepsilon}_t\cdot\boldsymbol{\alpha})^2=\tfrac{4}{3}, \quad (19)$$

so that

$$\langle|M_{0k}|^2\rangle_{\mathrm{av}}=\frac{(\alpha\lambda)^2}{3N}c(1-c)\cosh^2(\lambda_k/2). \quad (20)$$

Integrating over the final states degenerate with the homogeneous mode, the linewidth contribution is

$$\Delta H\approx\frac{\lambda_{0k}}{\gamma}=\frac{(\alpha\lambda)^2c(1-c)V}{12\pi^2\gamma\hbar^2N}\int_0^{2\pi}\int_0^{\pi}\left(\cosh^2(\lambda_k/2)k^2\frac{\partial k}{\partial\omega(k)}\right)_{\omega(\mathbf{k})=\omega(0)}\sin\theta_k d\theta_k d\phi_k, \quad (21)$$

where λ_{0k} is the total probability of scattering of the homogeneous spin-wave mode. This can be written as

$$\Delta H\approx\lambda_{0k}/\gamma=\tfrac{1}{4}(\alpha\lambda/\gamma\hbar)^2c(1-c)(4\pi M/H_e^3)^{\frac{1}{2}}$$
$$\times\Big\{3\left(\frac{H_e^3}{4\pi M}\right)^{\frac{1}{2}}\frac{2\gamma V}{15\pi N}\int_0^{\pi}\left[[\cosh^2(\lambda_k/2)+\tfrac{2}{3}\sinh^2(\lambda_k/2)]k^2\frac{\partial k}{\partial\omega(k)}\right]_{\omega(\mathbf{k})=\omega(0)}\sin\theta_k d\theta_k$$
$$+2\left(\frac{H_e^3}{4\pi M}\right)^{\frac{1}{2}}\frac{2\gamma V}{15\pi N}\int_0^{\pi}\left[k^2\frac{\partial k}{\partial\omega(k)}\right]_{\omega(\mathbf{k})=\omega(0)}\sin\theta_k d\theta_k\Big\}, \quad (22)$$

or in the notation of Callen and Pittelli[1]

$$\Delta H \approx \lambda_{0k}/\gamma = \tfrac{1}{4}(\alpha\lambda/\gamma\hbar)^2 c(1-c)(4\pi M/H_e^3)^{\frac{1}{2}}(3I_1+2I_2), \quad (23)$$

where the integral I_1 is $1/45\pi$ times the integral evaluated numerically by Clogston, Suhl, Walker, and Anderson[14] and the integral I_2 is an integral evaluated by Callen and Pittelli.[1,4]

We are interested in the contribution to the linewidth when small amounts of cobalt are present, that is when c is small. For small c, $c(1-c)\approx c$ and

$$\Delta H \approx \lambda_{0k}/\gamma = \tfrac{1}{4}(\alpha\lambda/\gamma\hbar)^2 c(4\pi M/H_e^3)^{\frac{1}{2}}(3I_1+2I_2). \quad (24)$$

Taking Slonczewski's[3] value of $|\alpha\lambda| = 2.63\times10^{-14}$ erg, and using $4\pi M = 5000$ gauss, $H_e = 10^6$ oersteds, and $\omega(0)/4\pi M\gamma = 0.7$ so that for a sphere $I_1 = 0.011$ and $I_2 = 0.008$, we find

$$\Delta H \approx \lambda_{0k}/\gamma = 1.8\times10^3 c \text{ oersteds}, \quad (25)$$

where we recall that c is the fraction of octahedral sites occupied by cobalt ions. Slonczewski's value of $|\alpha\lambda|$ was obtained by fitting his theory of the anisotropy. However, the value of λ for a free cobalt ion is 3.51×10^{-14} erg, and the theoretical lower limit for α is unity; hence, we would expect that $|\alpha\lambda| \gtrsim 3.51\times10^{-14}$ erg. Slonczewski attributes the smaller value of $|\alpha\lambda|$ found in his theory to covalency. In any case we note that if we were to adopt the theoretical minimum value for ionic bonding (i.e., $|\alpha\lambda| = 3.51\times10^{-14}$ erg) that

$$\Delta H \approx \lambda_{0k}/\gamma = 3.2\times10^3 c \text{ oersteds}. \quad (26)$$

Hence we conclude that for small concentrations each percent of cobalt should increase the linewidth of ferrous ferrite, or of any noninverse ferrite, by approximately 20–30 oersteds.

IV. LINEWIDTH CONTRIBUTION IN INVERSE FERRITES

We now consider ferrites in which two types of ions, in addition to the cobalt ions, occupy the octahedral sites. This can occur either as a result of the composition of the host ferrite or as a result of partial inversion of the ferrite.

As pointed out in Sec. II, the additional crystalline fields which are present when not all octahedral-site ions are equivalent may result in a splitting of the orbital doublet, and if this splitting is large compared to the spin-orbit coupling parameter there will be a marked decrease in the linewidth contribution. We shall assume that if the splitting of the orbital doublet of a particular cobalt ion is large compared to the spin-orbit coupling parameter, then the contribution of that ion to the linewidth is negligible. We begin by considering a particular configuration of A and B ions (having different ionic charges) on the six octahedral sites surrounding a cobalt ion. By a point-charge calculation we obtain an estimate of the magnitude of the nontrigonal crystalline field and determine if this field splits the orbital degeneracy by an amount exceeding $|\alpha\lambda|$.

The six octahedral sites surrounding a cobalt ion lie on two parallel planes which are perpendicular to the trigonal axis, as shown in Fig. 1. The coordinates of the sites relative to the Co^{2+} ion are

1: $\frac{1}{4}a(1,0,1)$; 4: $\frac{1}{4}a(-1,0,-1)$;
2: $\frac{1}{4}a(1,1,0)$; 5: $\frac{1}{4}a(-1,-1,0)$;
3: $\frac{1}{4}a(0,1,1)$; 6: $\frac{1}{4}a(0,-1,-1)$;

where a is the length of a side of the cubic cell and is approximately 8.4 A. Note that under a threefold rotation about the trigonal axis $1\rightarrow2\rightarrow3\rightarrow1$ and $4\rightarrow5\rightarrow6\rightarrow4$, and that under an inversion through the central site $1\rightarrow4$, $2\rightarrow5$ and $3\rightarrow6$.

Since two kinds of ions are placed on six sites, there are 64 different configurations. However, it is clear that any configuration generated from a given configuration by threefold rotation about the trigonal axis and by inversion through the origin has an equivalent effect on the orbital doublet of the cobalt ion. There are consequently thirteen nonequivalent classes of configurations, as listed in Table I.

By a direct perturbation calculation with a perturbing field of the proper symmetry and with the wave functions of the Co^{2+} orbital doublet, we find that of the thirteen distinct classes of configurations four do not remove the orbital degeneracy and that the remaining nine classes split the degeneracy by an amount large compared to the magnitude of the spin-orbit interaction. The classes of configurations which do not split the degeneracy are marked by an asterisk in Table I, and they represent a total of ten configurations.

It is possible to visualize which configurations do not split the degeneracy, as follows. Consider that the

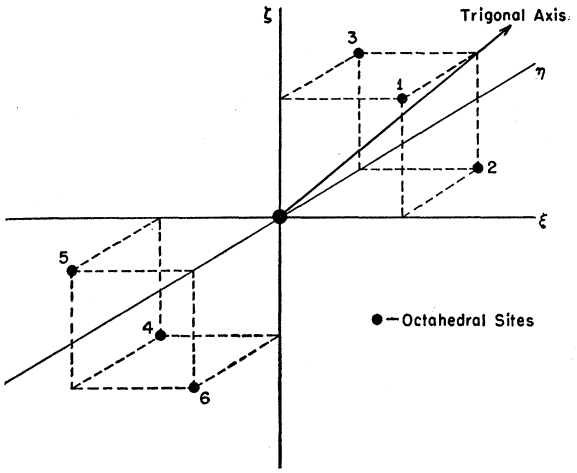

FIG. 1. Position of the six neighboring octahedral sites with respect to a cobalt ion.

[14] A. M. Clogston, H. Suhl, L. R. Walker, and P. W. Anderson, J. Phys. Chem. Solids **1**, 129 (1956).

crystalline field due to the six neighboring charges is expanded in powers of the spatial coordinates, in the vicinity of the cobalt ion. The linear terms do not split the effective P-state degeneracy. For consider the matrix element $\int\psi_1 V\psi_2 d\tau$, where ψ_1 and ψ_2 have the symmetry of real P states; i.e., $xf(r)$, $yf(r)$, or $zf(r)$, and where V is simply proportional to x, y, or z. In each case the integrand is odd in at least one coordinate and the matrix element vanishes. We therefore consider the quadratic terms in the expansion of the field; as we shall find that these can split the degeneracy, we shall ignore third and higher order terms as comparatively small. The quadratic form representing the second-order terms can be associated with a quadric surface, which must, in fact, be an ellipsoid. If this ellipsoid is an ellipsoid of revolution with a symmetry axis along the trigonal axis, it represents the type of trigonal symmetry which we have already discussed, its effect being to split off one of the three effective P states and to leave a remaining ground-state double degeneracy. This remaining degeneracy will be split only if the trace of the ellipsoid on a plane perpendicular to the trigonal axis is not circular. We consequently project all charges on this plane, and we consider the possibility of having an elliptic (i.e., noncircular) trace of the quadric surface on this plane.

Consider the configuration a listed in Table I; the corresponding projection of the charges is shown in Fig. 2(a). The resultant field clearly has circular symmetry in the plane, and we conclude that this configuration does not split the degeneracy—this result is, of course, obvious immediately. Consider now the configuration b listed in Table I, and shown in projection in Fig. 2(b). The quadric form can have the elliptic projection shown, and the resultant field will therefore split the degeneracy. The next three configurations in Table I are similarly shown in Figs. 2(c), 2(d), and 2(e), with the uniquely determined elliptic traces shown. Configuration f, shown in Fig. 2(f), is clearly circular and consequently does not split the degeneracy. Configuration g, shown in Fig. 2(g), is less obvious. Three hypothetical elliptical traces are shown on the figure, with their axes rotated by $\pi/3$. Inspection shows immediately, however, that each of these traces is equivalent; we therefore conclude that no unique axis exists for an elliptic trace, and that the trace must therefore be circular. Consequently, this configuration does not split the orbital ground-state degeneracy of the Co^{2+} ion. The remaining configurations can be analyzed similarly. The representation of configuration h is shown in Fig. 2(h). If the A and B ions are interchanged, the representation of configuration a becomes that of m, the representation of b that of l, and the representation of c, d, and e those of i, j, and k.

Having determined the effect of the various configurations on the orbital degeneracy, we now calculate the fraction of cobalt ions which contributes to the linewidth, as a function of the degree of inversion. We find the probability of the various configurations by making assumptions concerning the distribution of A and B ions and then weigh the configurations according to these probabilities.

TABLE I. Equivalent classes of configurations. Classes denoted by an asterisk do not split orbital degeneracy of Co^{2+} ion.

Class	Typical configuration of class (numbers indicate occupied sites)		Number of equivalent configuration in class	Probability of each configuration in class (random distribution)
*a	$A1, 2, 3, 4, 5, 6$		1	$(1-\eta)^6$
b	$A2, 3, 4, 5, 6$	$B1$	6	$\eta(1-\eta)^5$
c	$A2, 3, 5, 6$	$B1, 4$	3	
d	$A2, 4, 5, 6$	$B1, 3$	6	$\eta^2(1-\eta)^4$
e	$A2, 3, 4, 6$	$B1, 5$	6	
*f	$A1, 2, 3$	$B4, 5, 6$	2	
*g	$A1, 3, 5$	$B2, 4, 6$	6	$\eta^3(1-\eta)^3$
h	$A2, 5, 6$	$B1, 3, 4$	12	
i	$A1, 4$	$B2, 3, 5, 6$	3	
j	$A1, 3$	$B2, 4, 5, 6$	6	$\eta^4(1-\eta)^2$
k	$A1, 5$	$B2, 3, 4, 6$	6	
l	$A1$	$B2, 3, 4, 5, 6$	6	$\eta^5(1-\eta)$
*m		$B1, 2, 3, 4, 5, 6$	1	η^6

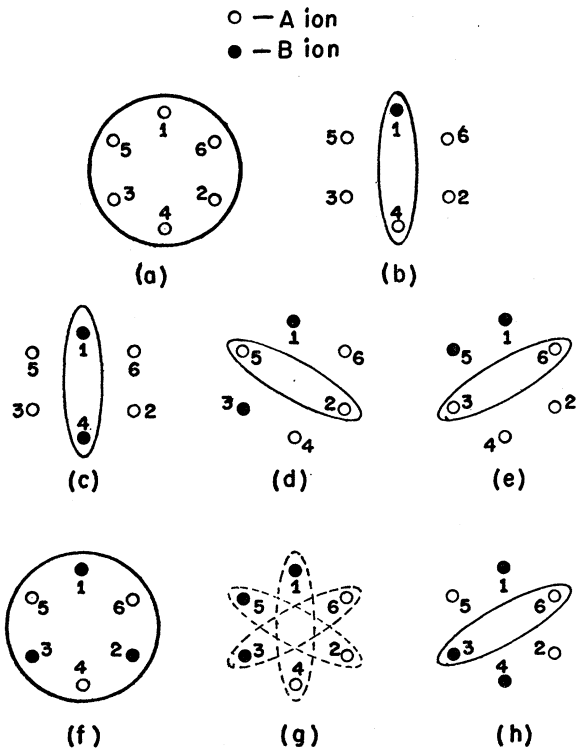

FIG. 2. Projection of charges onto plane perpendicular to trigonal axis.

For simplicity, we first assume that the distribution of A and B ions on the octahedral sites is completely random. If the fraction of A ions on octahedral sites is $(1-\eta)$ the probability of the configuration involving all B ions is η^6, whereas the probability of the configuration involving all A ions is $(1-\eta)^6$. Each of the eight configurations involving three A ions and three B ions has a probability of $\eta^3(1-\eta)^3$. Hence, the total probability that a given Co^{2+} ion will maintain its orbital degeneracy is

$$\eta^6+8\eta^3(1-\eta)^3+(1-\eta)^6. \quad (27)$$

In order to find the linewidth contribution, we must replace the fraction of cobalt ions c in Eq. (24) by the effective fraction $c[\eta^6+8\eta^3(1-\eta)^3+(1-\eta)^6]$. Thus in completely inverse $MeFe_2O_4$ ($\eta=\frac{1}{2}$), all 64 configurations have equal probability and only 10/64 or 0.16 of all cobalt ions contribute to the linewidth. The corresponding dependence of the linewidth on the amount of inversion of $MeFe_2O_4$ is shown in Fig. 3.

We have also calculated the fraction of cobalt ions which contributes to the linewidth as a function of the degree of inversion, first assuming that charge neutrality is maintained over a unit cell (four molecules) and then assuming that charge neutrality is maintained over half a unit cell (two molecules). For these two cases we calculated the probability of a given configuration which maintains charge neutrality over the specified region and divided by the total probability of finding all configurations which maintain charge neutrality over the same region. We are thus able to assign relative probabilities to the charge-neutral configurations and calculate the fraction of cobalt ions which contributes to the linewidth. These results are also shown in Fig. 3.

Fortunately, the different conditions placed on the distribution of the ions have little effect on the nature of the results. The curves of Fig. 3 may be broken up into two parts. As the Me ion concentration of the octahedral sites increases from 0 to 0.25, the fraction of cobalt ions which contributes to the linewidth decreases roughly linearly. As the Me ion concentration is increased from 0.25 to 0.50, the fraction of cobalt ions which contributes is roughly constant at approximately 0.15 to 0.20.

If we choose the values of M, H_e, and $\omega(0)$ given at the end of Sec. III, the linewidth of completely inversed (or more than half-inversed) $MeFe_2O_4$ should increase by 3-6 oersteds for each percent of cobalt. For a given ferrite the parameters would be known and the actual linewidth contribution could be evaluated from Eq. (24).

V. COMPARISON WITH EXPERIMENTAL RESULTS

According to the scattering model considered, small amounts of cobalt introduced into magnetite should result in an increase of the resonance linewidth by 20—30 oersteds for each percent of cobalt present. As ferrous ions on the octahedral site are replaced by other divalent metal ions, this contribution to the linewidth decreases and for the completely inverse spinel, $MeFe_2O_4$, the contribution is about $\frac{1}{5}$ of that found for magnetite (neglecting any change in $4\pi M$ or H_e).

Unfortunately, the theory cannot be checked easily by measurements on magnetite because the very itinerant electrons which average the charge distribution also contribute large damping.

Experimental data which can be compared directly with these predictions do not seem to exist. Various measurements have been made on polycrystalline samples, but in these samples the inhomogeneity broadening and surface scattering certainly dominate the intrinsic linewidth. Some single-crystal measurements have been made by Kaminow[15] in connection with a study of the effect of hydrostatic pressure on resonance properties. His measurements were made on nickel ferrite single crystals and on nickel-cobalt ferrite estimated to contain 5% cobalt on the basis of the melt composition. However, it is difficult to estimate the effect of the cobalt on the linewidth because of the large scatter in the linewidth measurements, and because the cobalt content was not measured in the

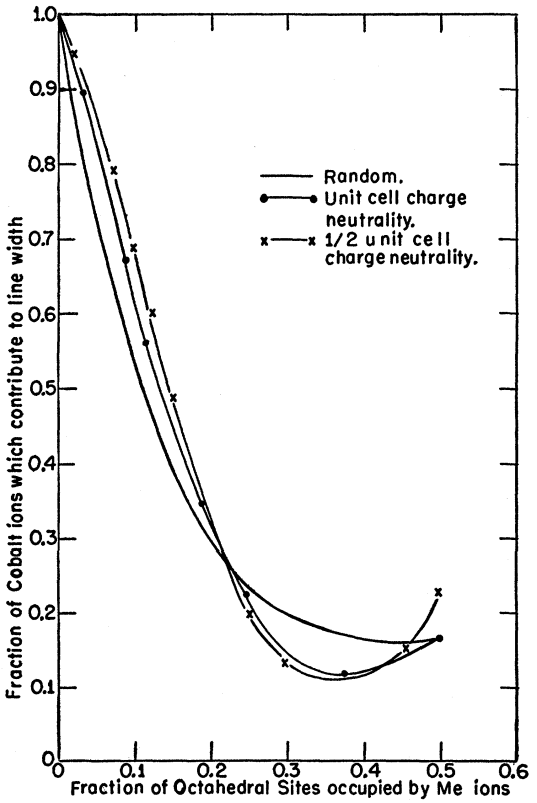

FIG. 3. Fraction of cobalt ions which contributes to linewidth as a function of degree of inversion of $MeFe_2O_4$.

[15] I. P. Kaminow, J. Appl. Phys. **31**, 220S (1960).

crystal itself. Furthermore, it should be noted from the experience of Schnitzler, Folen, and Rado[16] with lithium ferrite that extraordinary care must be taken with the polishing of the crystals in order to insure that surface scattering does not dominate the intrinsic linewidth.

Fortunately there do exist some data which can be compared with our conclusions, although somewhat less directly. Schlömann, Green, and Milano[17] have made high-power measurements on polycrystalline nickel ferrite containing small amounts of cobalt. As the power level of the rf field is raised sufficiently, the precession angle of the uniform mode approaches a maximum. This maximum precession angle, $\mu_{0\,\max}$, is related to the linewidth, ΔH_k, of the z-directed spin waves which are degenerate with the uniform mode by the expression[18]

$$\mu_{0\,\max}{}^2=\Delta H_k/(4\pi M). \tag{28}$$

Schlömann *et al.*[17] find that as the cobalt content of the octahedral sites is increased from 0 to 2.5%, $\mu_{0\,\max}{}^2$ increases approximately linearly from 0.5×10^{-2} to 1.0×10^{-2}. For nickel ferrite, $4\pi M=3400$ gauss and therefore ΔH_k increases 6.8 oersteds for each percent of cobalt present.

The wavelength of the z-directed spin modes degenerate with the uniform mode is about 200 interatomic distances. This is a long wavelength and the intrinsic scattering of these modes should be comparable to that of the uniform precessional mode. Furthermore, Le Craw and Spencer[19] have found that the scattering of such modes is not as surface sensitive as that of the uniform mode. Our calculation (making the correction for the change in $4\pi M$) would predict that the magnon-magnon scattering should result in a change in ΔH_k of about 3 oersteds for each percent of cobalt present.

APPENDIX. MAGNON SCATTERING CORRECTIONS DUE TO SPIN VARIATIONS

We show that the variation from ion to ion of the spin magnitude produces negligible magnon scattering. For this purpose it is analytically convenient to use the Dyson spin-wave formalism rather than the formalism employed in the body of the paper; although the two formulations are, of course, physically equivalent.

Consider a sample with two types of magnetic ions. On each, the spin commutation rules are

$$\mathbf{S}(\mathbf{R})\times\mathbf{S}(\mathbf{R})=i\mathbf{S}(\mathbf{R}), \tag{A.1}$$

which are formally independent of the magnitude of $\mathbf{S}(\mathbf{R})$. Then defining the flip-up and flip-down operation $S^{\pm}$ in the usual fashion, and letting

$$\mathbf{S}(\mathbf{k})=\frac{1}{\sqrt{N}}\sum_{\mathbf{R}}\mathbf{S}(\mathbf{R})e^{i\mathbf{k}\cdot\mathbf{R}}, \tag{A.2}$$

the commutation rules become

$$[S^{\pm}(\mathbf{k}),S_z(\mathbf{k}')]=\mp\frac{1}{\sqrt{N}}S^{\pm}(\mathbf{k}+\mathbf{k}'), \tag{A.3}$$

$$[S^{+}(\mathbf{k}),S^{-}(\mathbf{k}')]=\frac{2}{\sqrt{N}}S_z(\mathbf{k}+\mathbf{k}'). \tag{A.4}$$

All of this is identical to the situation in which all spins have the same magnitude.

Consider now the ground state, in which all spins are up. Then clearly

$$S_z(0)\,|\,g\rangle=\frac{1}{\sqrt{N}}\sum_{\mathbf{R}}S_z(\mathbf{R})\,|\,g\rangle=N^{\frac{1}{2}}\bar{S}\,|\,g\rangle, \tag{A.5}$$

where $\bar{S}$ is the average spin magnitude. Let the singly-occupied spin-wave state of wave vector $\mathbf{k}$ be

$$|\,\mathbf{k}\rangle=AS^{-}(\mathbf{k})\,|\,g\rangle, \tag{A.6}$$

where A is a normalization factor to be determined. Recalling that the adjoint of $S^{-}(\mathbf{k})$ is $S^{+}(-\mathbf{k})$, we have

$$\langle\mathbf{k}\,|\,\mathbf{k}\rangle=A^2\langle g\,|\,S^{+}(-\mathbf{k})S^{-}(\mathbf{k})\,|\,g\rangle \tag{A.7}$$

$$=A^2\left\langle g\left|\frac{2}{\sqrt{N}}S_z(0)+S^{-}(\mathbf{k})S^{+}(-\mathbf{k})\right|g\right\rangle \tag{A.8}$$

$$=2A^2\bar{S}, \tag{A.9}$$

where we have used Eq. (A.4) to derive (A.8), and Eqs. (A.5) and the flip-up property of S^{+} to derive (A.9). Thus $A=(2\bar{S})^{-\frac{1}{2}}$ and

$$|\,\mathbf{k}\rangle=(2\bar{S})^{-\frac{1}{2}}S^{-}(\mathbf{k})\,|\,g\rangle. \tag{A.10}$$

In both Eqs. (A.5) and (A.10) we note the natural introduction of the arithmetically averaged spin magnitude.

The spin-wave states defined by Eq. (A.10) are orthogonal to the ground state, for

$$\langle g\,|\,\mathbf{k}\rangle=(2\bar{S})^{-\frac{1}{2}}\langle g\,|\,S^{-}(\mathbf{k})\,|\,g\rangle \tag{A.11}$$

$$=(2\bar{S}N)^{-\frac{1}{2}}\textstyle\sum_{\mathbf{R}}e^{i\mathbf{k}\cdot\mathbf{R}}\langle g\,|\,S^{-}(\mathbf{R})\,|\,g\rangle=0. \tag{A.12}$$

However, the spin-wave states are not orthogonal to each other. For

$$\langle\mathbf{k}\,|\,\mathbf{k}'\rangle=(2\bar{S})^{-1}\langle g\,|\,S^{+}(-\mathbf{k})S^{-}(\mathbf{k}')\,|\,g\rangle \tag{A.13}$$

$$=(2\bar{S})^{-1}\left\langle g\left|\frac{2}{\sqrt{N}}S_z(\mathbf{k}'-\mathbf{k})+S^{-}(\mathbf{k}')S^{+}(-\mathbf{k})\right|g\right\rangle \tag{A.14}$$

[16] A. D. Schnitzler, V. J. Folen, and G. T. Rado, J. Appl. Phys. **31**, 348S (1960).
[17] E. Schlömann, J. J. Green, and U. Milano, J. Appl. Phys. **31**, 386S (1960).
[18] E. Schlömann, Technical Report No. R-48, Research Division, Raytheon Company, 1959 (unpublished).
[19] R. C. Le Craw and E. G. Spencer, J. Appl. Phys. **30**, 185S (1959).

$$= (\bar{S}N)^{-1} \sum_{\mathbf{R}} e^{i(\mathbf{k}'-\mathbf{k})\cdot\mathbf{R}} \langle g|S_z(\mathbf{R})|g\rangle \quad (A.15)$$

$$= \frac{1}{\bar{S}\sqrt{N}} S_z{}^g(\mathbf{k}-\mathbf{k}'), \quad (A.16)$$

where $S_z{}^g(\mathbf{k})$ is the kth Fourier component of $S_z(\mathbf{R})$ in the ground state. Of particular interest is the absolute square of the matrix element $|\langle\mathbf{k}|\mathbf{k}'\rangle|^2$, which by Eq. (A.15) is

$$|\langle\mathbf{k}|\mathbf{k}'\rangle|^2 = (\bar{S}N)^{-2} \sum_{\mathbf{R},\mathbf{R}'} e^{i(\mathbf{k}-\mathbf{k}')\cdot(\mathbf{R}-\mathbf{R}')} \times S_z{}^g(\mathbf{R}) S_z{}^g(\mathbf{R}'), \quad (A.17)$$

where $S_z{}^g(\mathbf{R})$ is the value of $S_z(\mathbf{R})$ in the ground state, equal to the spin magnitude of the ion at position $\mathbf{R}$. Taking the ensemble average over all distributions of the ions, we have

$$\langle|\langle\mathbf{k}|\mathbf{k}'\rangle|^2\rangle_{\mathrm{av}} = (\bar{S}N)^{-2} \sum_{\mathbf{R},\mathbf{R}'} e^{i(\mathbf{k}-\mathbf{k}')\cdot(\mathbf{R}-\mathbf{R}')} \times \langle S_z{}^g(\mathbf{R}) S_z{}^g(\mathbf{R}')\rangle_{\mathrm{av}}. \quad (A.18)$$

We now compute the correlation functions $\langle S_z{}^g(\mathbf{R}) S_z{}^g(\mathbf{R}')\rangle_{\mathrm{av}}$ assuming no short-range order. Let a fraction c of the ions have $S_z{}^g = s_1$ and a fraction $(1-c)$ have $S_z{}^g = s_2$. If $\mathbf{R}=\mathbf{R}'$, the correlation function clearly is the average squared spin magnitude $\langle S^2\rangle_{\mathrm{av}} = cs_1{}^2 + (1-c)s_2{}^2$. If $\mathbf{R}\neq\mathbf{R}'$ we first select the position $\mathbf{R}$; it may contain a spin s_1, with probability c, or it may contain a spin s_2, with probability $(1-c)$. If the site $\mathbf{R}$ has a spin s_1, the site $\mathbf{R}'$ has a probability $(Nc-1)/(N-1)$ of also having a spin s_1, and a probability $N(1-c)/(N-1)$ of having a spin s_2. If the site $\mathbf{R}$ has a spin s_2, the site $\mathbf{R}'$ has a probability $Nc/(N-1)$ of having a spin s_1, and a probability $[N(1-c)-1]/(N-1)$ of having a spin s_2. Thus, when $\mathbf{R}\neq\mathbf{R}'$ the correlation function is

$$\langle S_z{}^g(\mathbf{R}) S_z{}^g(\mathbf{R}')_{\mathbf{R}\neq\mathbf{R}'}\rangle_{\mathrm{av}} = cs_1\left[s_1\frac{Nc-1}{N-1} + s_2\frac{N(1-c)}{N-1}\right] + (1-c)s_2\left[s_1\frac{Nc}{N-1} + s_2\frac{N(1-c)-1}{N-1}\right] \quad (A.19)$$

$$= \left[\langle S^2\rangle_{\mathrm{av}}\frac{N}{N-1} - \frac{\langle S^2\rangle_{\mathrm{av}}}{N-1}\right], \quad (A.20)$$

and

$$\langle S_z{}^g(\mathbf{R}) S_z{}^g(\mathbf{R}')\rangle_{\mathrm{av}} = \left[\langle S^2\rangle_{\mathrm{av}}\frac{N}{N-1} - \frac{\langle S^2\rangle_{\mathrm{av}}}{N-1}\right] \times (1-\delta_{\mathbf{R},\mathbf{R}'}) + \langle S^2\rangle_{\mathrm{av}}\delta_{\mathbf{R},\mathbf{R}'}. \quad (A.21)$$

Inserting this correlation function in Eq. (A.18) gives

$$\langle|\langle\mathbf{k}|\mathbf{k}'\rangle|^2\rangle_{\mathrm{av}} = N^{-2}(N-1)^{-1}\left[N - \frac{\langle S^2\rangle_{\mathrm{av}}}{\langle S^2\rangle_{\mathrm{av}}}\right] \times \sum_{\mathbf{R}\neq\mathbf{R}'} e^{i(\mathbf{k}-\mathbf{k}')\cdot(\mathbf{R}-\mathbf{R}')} + \frac{\langle S^2\rangle_{\mathrm{av}}}{N\bar{S}^2}. \quad (A.22)$$

We add and subtract the N diagonal terms $\mathbf{R}=\mathbf{R}'$ to the summation. The double unrestricted sum is then $N^2\delta_{\mathbf{k},\mathbf{k}'}$, whereas the diagonal terms to be subtracted have the value N. Thus

$$\langle|\langle\mathbf{k}|\mathbf{k}'\rangle|^2\rangle_{\mathrm{av}} = (N-1)^{-1}\left[N - \frac{\langle S^2\rangle_{\mathrm{av}}}{\langle S^2\rangle_{\mathrm{av}}}\right] \times \left(\delta_{\mathbf{k},\mathbf{k}'} - \frac{1}{N}\right) + \frac{\langle S^2\rangle_{\mathrm{av}}}{N\langle S^2\rangle_{\mathrm{av}}} \quad (A.23)$$

$$= \delta_{\mathbf{k},\mathbf{k}'} + \frac{1}{N-1}\frac{\langle(\Delta S)^2\rangle_{\mathrm{av}}}{\langle S^2\rangle_{\mathrm{av}}}(1-\delta_{\mathbf{k},\mathbf{k}'}), \quad (A.24)$$

where $\langle(\Delta S)^2\rangle_{\mathrm{av}}$ is the mean square spin deviation, or $\langle S^2\rangle_{\mathrm{av}} - \langle S^2\rangle_{\mathrm{av}}$. It will be noted that $|\langle\mathbf{k}|\mathbf{k}'\rangle|^2$ is equal to unity if $\mathbf{k}=\mathbf{k}'$, as required by the normalization; furthermore it vanishes if $\mathbf{k}\neq\mathbf{k}'$ and if $s_1=s_2$, as required by the orthogonality of spin waves in single-component ferromagnets.

We now inquire as to the effect of the nonorthogonality of the spin-wave states on our scattering calculations. If a magnon of $\mathbf{k}=0$ is excited at any moment, a subsequent measurement will observe a nonzero population in other spin-wave states because of the nonorthogonality referred to above. The states of particular interest to us are those approximately degenerate with the $\mathbf{k}=0$ state; that is, those in an energy interval $\Delta E \approx \hbar/\tau$, where τ is the mean transition time for the scattering mechanism to be considered. The scattering mechanism itself will populate this group of states with a probability equal to $\frac{1}{2}$, in the time τ. Hence, we require that the sum of $|\langle 0|\mathbf{k}'\rangle|^2$ over all states in this degenerate band be much less than $\frac{1}{2}$ in order that the nonorthogonality can be neglected.

$$\sum_{\mathbf{k}' \text{ in } \Delta E\approx\hbar/\tau} \langle|\langle 0|\mathbf{k}'\rangle|^2\rangle_{\mathrm{av}} \ll \tfrac{1}{2}. \quad (A.25)$$

Because the matrix element is independent of $\mathbf{k}'$, we have simply

$$\frac{1}{N-1}\frac{\langle(\Delta S)^2\rangle_{\mathrm{av}}}{\langle S^2\rangle_{\mathrm{av}}}\rho(E)\frac{\hbar}{\tau} \ll \frac{1}{2}, \quad (A.26)$$

where $\rho(E)$ is the density-in-energy of spin-wave states degenerate with the $\mathbf{k}=0$ state. If we ignore the directional dependence of the spin-wave dispersion law

$$\rho(E) = \frac{N}{4\pi^2\gamma\hbar}\left(\frac{4\pi M_0}{3H_e{}^3}\right)^{\frac{1}{2}}. \quad (A.27)$$

$1/\tau$ is equal to λ_{0k}, the transition probability. Using the expression for the density-in-energy of final states given in (A.27),

$$\lambda_{0k}=\frac{N|M_{0k}|^2}{\pi\hbar^2\gamma}\left(\frac{4\pi M_0}{12H_e^3}\right)^{\frac{1}{2}}, \tag{A.28}$$

where M_{0k} is the matrix element of the scattering perturbation. If we represent the perturbation by a fluctuating effective magnetic field[1] $H(\mathbf{R})$, then

$$\lambda_{0k}=\frac{\gamma\langle S^2\rangle_{\rm av}|H(\mathbf{k})|^2}{(12)^{\frac{1}{2}}\pi}\left(\frac{4\pi M_0}{H_e^3}\right)^{\frac{1}{2}}, \tag{A.29}$$

where $H(\mathbf{k})$ is the $\mathbf{k}$th Fourier component of $H(\mathbf{R})$.

Substituting (A.27) and (A.29) into expression (A.26) results in the requirement

$$\frac{1}{N-1}\frac{\langle(\Delta S)^2\rangle_{\rm av}}{\langle S^2\rangle_{\rm av}}\frac{N}{2\pi^2\gamma}\left(\frac{4\pi M_0}{12H_e^3}\right)^{\frac{1}{2}} \times\frac{\gamma\langle S^2\rangle_{\rm av}|H(\mathbf{k})|^2}{\pi}\left(\frac{4\pi M_0}{12H_e^3}\right)^{\frac{1}{2}}\ll\frac{1}{2}, \tag{A.30}$$

or

$$\frac{N}{N-1}\frac{\langle(\Delta S)^2\rangle_{\rm av}}{6\pi^2}\frac{M_0}{H_e^3}|H(\mathbf{k})|^2\ll\tfrac{1}{2}, \tag{A.31}$$

where it is to be recalled that $|H(\mathbf{k})|\approx 10^6$. Inserting the values $4\pi M_0=5000$ gauss, $H_e=10^6$ oe, and setting $N/(N-1)=1$, we have

$$7\times10^{-6}\langle(\Delta S)^2\rangle_{\rm av}\ll\tfrac{1}{2}, \tag{A.32}$$

which is certainly satisfied. Thus the corrections to the magnon scattering due to the nonorthogonality of the spin waves in a disordered ferrite are totally negligible.

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Absolute Measurement of the Atomic Scattering Factors of Iron, Copper, and Aluminum

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The x-ray atomic scattering factors of iron, copper, and aluminum were carefully remeasured to obtain more reliable information on the outer electron charge densities in these elements. The scattering factors were obtained from measurements of the integrated Bragg intensities of powder samples using monochromatic Mo K_α radiation. The intensities were put on an absolute scale by direct measurements of the power in the primary x-ray beam. Extinction, surface roughness, and preferred orientation effects were shown to be negligible in the samples used. The ratios of the measured scattering factors of the three elements agree with those calculated from Hartree-Fock theory to within 1%. This substantiates the findings of Batterman, and, in contrast with the previous results of Weiss and DeMarco, indicates that there is no large discrepancy between the electronic structures of copper and iron. The absolute values of the measured scattering factors, however, lie about 4% below theory in the region of low $\sin\theta/\lambda$. It is pointed out that the high theoretical values for iron and copper could result from known differences in electronic structure between a free atom and one in the solid, but that present theory probably cannot account for the discrepancy in the case of aluminum.

INTRODUCTION

In the past few years, several experiments have been undertaken to measure the electron distribution in the iron transition metals. The ultimate aim of these experiments is to help in the basic understanding of unresolved problem of the magnetism of these metals.

In these experiments the intensities of x-ray Bragg reflections are experimentally determined and reduced to charge densities by comparing the experimental form factors obtained from these intensities with those calculated from theoretical wave functions. Briefly, the chronology of these experiments is as follows: Weiss and DeMarco[1] made absolute measurements on single crystals of several transition metals and concluded that body-centered cubic iron and chromium have a deficiency of $3d$ electrons compared with the free atom. Batterman[2] measured relative intensities of powders of copper, iron, and rock salt and reported form factors in good agreement with calculations for the free atom and consequently disagreed with the Weiss and DeMarco results. Weiss and DeMarco[1] then repeated

[1] R. J. Weiss and J. J. DeMarco, Revs. Modern Phys. **30**, 59 (1958); Phys. Rev. Letters **2**, 148 (1959).

[2] B. W. Batterman, Phys. Rev. Letters **2**, 47 (1959); Phys. Rev. **115**, 81 (1959).