in a region where the sublattices approach saturation. For the Gd sublattice this does not happen until T is appreciably below 100°K, but the Fe sublattices are over 70% saturated even at 300°K. This may explain why there is practically no contribution at all for the Fe ions, even though our molecular field treatment would predict a small but noticeable effect. (ii) Even in our best sample the theoretical saturation moment was not attained in the helium range. This makes the calculation of relative magnetization at higher temperatures somewhat uncertain, but the effect on the susceptibility is generally negligible. (iii) The discrepancy between the theoretical and experimental $(\sigma_s)_{T=0}$ value may have been due to small impurities of other rare earths, which might have the effect of increasing the magnetic anisotropy at the lowest temperatures (as in the experiments of Dillon and Nielsen[11]). Too large a magnetic hardness in the polycrystalline sample would lead to uncertainty in both the extrapolated values of σ_s and the deduced values of susceptibility. (iv) In estimating the Gd sublattice magnetization we assumed, with Pauthenet, that there is no "back action" by the Gd ions on the Fe sublattices. This cannot be true exactly, and in the regions in which σ_s varies rapidly with temperature the effect of this on the calculated susceptibility may be appreciable. Inclusion of this effect might very well account for the systematic discrepancy in the region of 100°K.

[11] J. F. Dillon and J. W. Nielsen, Phys. Rev. **120**, 105 (1960).

CONCLUSION

We have made measurements of the intrinsic susceptibility of a sample of polycrystalline gadolinium iron garnet between 2° and 300°K. The results are generally in reasonable agreement with earlier measurements by Pauthenet on a sample presumably less nearly stoichiometric. At high temperatures the susceptibility approximately follows Curie's law for free Gd^{3+} ions, but there are significant deviations which become very large at low temperatures. It is shown that these deviations are quantitatively consistent with the effects to be expected from paramagnetic saturation. Interactions between the rare-earth ions appear to be much less important than previously supposed.

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Bloch Wall Excitation. Application to Nuclear Resonance in a Bloch Wall*

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The excitation spectrum of an assembly of electronic spins in a Bloch wall structure is studied, assuming a uniaxial anisotropy. The spectrum may be divided into two branches; one is a specific wall excitation and does not spread outside the wall, the other one is similar to the spin-wave excitation spectrum in a uniform ferromagnet. These calculations are used to study the properties of the nuclear magnetic resonances in a Bloch wall. The relaxation times are evaluated, taking into account the damping of the motion of the electronic spins and are compared with experimental values. The spin-spin coupling and the variation of the magnetization across the wall is also estimated.

I. INTRODUCTION

Nuclear magnetic resonances in several ferromagnetic substances have been observed recently.[1] The large amplitude of the signals is explained by assuming that the observed nuclei are within the Bloch wall.

The relaxation times T_1 and T_2 have also been measured using spin-echo techniques.[2] The theoretical estimation of relaxation times and spin-spin couplings requires a detailed knowledge of the motion of the electronic spins within the Bloch wall.

A complete solution for this problem may be found by assuming a uniaxial anisotropy and using a simplified demagnetizing field, then the relaxation times are estimated, taking into account the damping of the wall motion. An indirect interaction is also estimated.

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[1] A. C. Gossard and A. M. Portis, J. Appl. Phys. **31**, 205S (1960); A. C. Gossard, A. M. Portis, and W. J. Sandle, J. Phys. Chem. Solids, **17**, 341 (1961); C. Robert and J. M. Winter, Compt. rend. **250**, 3831 (1960); J. Hervé and P. Veillet, Compt. rend. **252**, 99 (1961).

[2] M. Weger, E. L. Hahn, and A. M. Portis, J. Appl. Phys. **32**, 124S (1961); C. Robert and J. M. Winter, Nuovo cimento, (to be published).

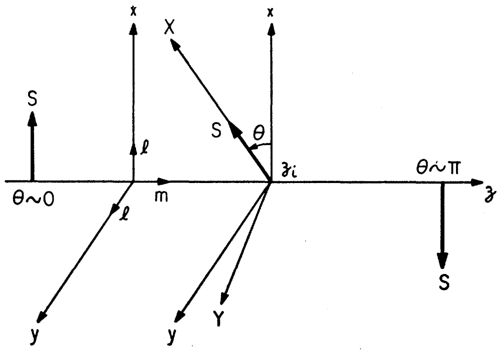

FIG. 1. Definition of axis.

Finally, the variation of the magnetization within the Bloch wall is calculated.

II. EQUATIONS OF MOTION

Let us assume a uniaxial anisotropy:

$$\mathcal{H}_A = K \sum_i [(S_i^y)^2 + (S_i^z)^2;$$

the Oz axis is perpendicular to the plane of the wall; the wall is a 180° wall, and the magnetization for $z=-\infty$ is parallel to Ox, for $z=+\infty$ antiparallel to Ox.

We assume a simple cubic lattice. The exchange part of the Hamiltonian may be written

$$\mathcal{H}_{\text{ex}} = -2J \sum_{\langle i,j\rangle} \mathbf{S}_i \cdot \mathbf{S}_j,$$

the summation being restricted to nearest neighbors. The angle between the static magnetization and Ox is called θ and θ is an unknown function of z. Let us choose a new system of axes OX, OY, Oz; OX is the spin direction, and Oz is not changed (Fig. 1). The directions of the axes vary from one atom to another. The Hamiltonian becomes

$$\begin{aligned}\mathcal{H} = & -2J \sum_{\langle i,j\rangle} \{S_z^i S_z^j\} - 2J \sum_{i,m} \cos(\theta_i - \theta_{i+m}) \\ & \times (S_X^i S_X^{i+m} + S_Y^i S_Y^{i+m}) \\ & -2J \sum_{i,m} \sin(\theta_i - \theta_{i+m})(S_X^i S_Y^{i+m} - S_Y^i S_X^{i+m}) \\ & -2J \sum_{i,l} (S_X^i S_X^{i+l} + S_Y^i S_Y^{i+l}) + K \sum_i [(S_X^i)^2 \sin^2\theta_i \\ & -2(S_X^i)(S_Y^i) \sin\theta_i \cos\theta_i + (S_Y^i)^2 \cos^2\theta_i + (S_z^i)^2].\end{aligned}$$

m is a unit vector of the cell along Oz and l is a unit vector perpendicular to Oz.

For the ground state we assume $S_X=S$, $S_Y=S_z=0$, and θ is determined by minimizing the energy. Thus we obtain the well-known result,[3]

$$J(\theta_i - \theta_{i+m})^2 = K \sin^2\theta_i,$$

or

$$d\theta_i/dz = (1/a)(K/J)^{\frac{1}{2}} \sin\theta_i, \tag{1}$$

$\lambda = a(J/K)^{\frac{1}{2}}$ being of the order of the wall thickness, where a is the lattice spacing. We deduce also the relation,

$$\sin\theta_i = 1/\cosh(z_i/\lambda). \tag{2}$$

Now keeping only terms of second order with respect to S_Y^i and S_z^i, the Hamiltonian becomes

$$\begin{aligned}\mathcal{H} = & -2J \sum_{\langle i,j\rangle} S_z^i S_z^j - 2J \sum_{i,m} \cos(\Delta\theta_i) S_Y^i S_Y^{i+m} \\ & -2J \sum_{i,l} S_Y^i S_Y^{i+l} + 2J \sum_i \cos\Delta\theta_i [(S_Y^i)^2 + (S_z^i)^2] \\ & +4J \sum_i [(S_Y^i)^2 + (S_z^i)^2] \\ & +K \sum_i [(S_Y^i)^2 (\cos^2\theta_i - \sin^2\theta_i) + (S_z^i)^2 \cos^2\theta_i]; \\ & \Delta\theta_i = \theta_i - \theta_{i+m}.\end{aligned} \tag{3}$$

(The linear terms disappear by taking into account the minimization condition.)

The equation of motion is

$$i\hbar d\mathbf{S}_i/dt = [\mathbf{S}_i, \mathcal{H}].$$

Then, assuming a slow spatial variation and using commutation relations, we get

$$\begin{aligned}\hbar\frac{dS_Y}{dt} &= -2JSa^2\nabla^2 S_z + 2KSS_z(\cos^2\theta - \sin^2\theta), \\ \hbar\frac{dS_z}{dt} &= 2Jsa^2\frac{d^2S_Y}{dz^2}\cos(\Delta\theta) + 2JSa^2\left(\frac{d^2S_Y}{dy^2} + \frac{d^2S_Y}{dx^2}\right) \\ &\quad -2KSS_Y(\cos^2\theta - \sin^2\theta).\end{aligned} \tag{4}$$

Now, if we look for an oscillatory time-varying solution [using (1) instead of $\cos\Delta\theta = 1 - \frac{1}{2}(K/J)\sin^2\theta$],

$$\begin{aligned}-iES_Y &= 2JSa^2\nabla^2 S_z - 2KSS_z(\cos^2\theta - \sin^2\theta), \\ iES_z &= 2JSa^2\nabla^2 S_Y - 2KSS_Y(\cos^2\theta - \sin^2\theta).\end{aligned} \tag{5}$$

For the particular value $E=0$, the solution is

$$S_Y = B\sin\theta, \quad S_z = C\sin\theta,$$

C and B being constants. The solution $S_Y = B\sin\theta$, $S_z=0$ corresponds to a small change of the angle between the magnetization and Ox, and $\Delta\theta = (B/S)\sin\theta$, giving rise to a translation Δz of the Bloch wall. Using Eq. (1),

$$\Delta z = \lambda(B/S).$$

Before looking for the general solution of these equations, we need to complete them for the following reasons.

1. With the Hamiltonian (3) the simplest excitation occurs exactly at zero energy, so any uniform perturbation, however small, may push the wall to infinity. It is known that the wall must have a stiffness coefficient α

[3] C. Kittel and J. K. Galt, *Solid-State Physics*, edited by F. Seitz and D. Turnbull (Academic Press, Inc., New York, 1956), Vol. 3.

related to the initial permeability. The energy required to push the wall on a distance z is $\alpha(z^2/2)$.

The simplest way to take into account the wall stiffness is to add to our Hamiltonian the term $K'\sum_i(S_Y{}^i)^2$. Let us show that this term gives a change in energy of $\alpha(\Delta z^2/2)$ for a translation of Δz. A Δz translation is equivalent to a rotation $\Delta\theta_i=(\Delta z/\lambda)\sin\theta_i$ for the spin i, and a transverse component appears, $S_Y{}^i=S(\Delta z/\lambda)\sin\theta_i$, giving a change of energy, $K'\sum_i(S_Y{}^i)^2=K'(\Delta z/\lambda)^2S^2\sum_i\sin^2\theta_i$. Then

$$\alpha=\frac{2}{s_0}\frac{K'S^2}{\lambda^2}\sum_i\sin^2\theta_i. \tag{6}$$

s_0 is the area of the wall (α is defined per unit area of the wall).

2. Döring[4] showed that demagnetizing effects give a wall mass. The evaluation of demagnetizing effects for an arbitrary excitation is a fairly complicated problem; but if we assume a slow spatial variation for the magnetization in the X and Y direction, we may use an approximate value.

In a magnetic substance the demagnetizing field is defined by the two relations,

$$\mathrm{div}\mathbf{H}_d=-4\pi\,\mathrm{div}\mathbf{M},\quad \mathrm{curl}\mathbf{H}_d=0. \tag{7}$$

As we shall see later, the solutions are plane waves in the X and Y directions, so for low values of K_X and K_Y the main term in the divergence is dM/dz (providing the spins are not too far from the center of the wall). Equations (7) are approximately satisfied by

$$H_d{}^z=-4\pi g\beta S_z{}^i,\quad H_d{}^X=H_d{}^Y=0,$$

providing $K_X\lambda\sin\theta_i\ll1$, and $K_Y\lambda\sin\theta_i\ll1$. Then we add to our Hamiltonian the term

$$\tfrac{1}{2}\mathbf{H}_d\cdot\mathbf{S}(g\beta)=-2\pi(g\beta)^2\sum_i(S_z{}^i)^2.$$

3. The experimental study of wall displacement shows that the motion is accompanied by energy losses. In metals the losses come mainly from the effects of eddy currents. In order to take these effects into account, we add to the equations of motion a damping term, which has the form first suggested by Landau and Lifshitz:

$$(d\mathbf{S}/dt)_{\mathrm{damping}}=-(\Gamma/S^2)\{\mathbf{S}_n\times[\mathbf{S}_n\times\mathbf{H}]\},$$

where $\mathbf{H}$ is the demagnetizing field.

Adding all the contributions, the equations of motion become

$$\begin{aligned}-iES_Y&=2JSa^2\nabla^2S_z\\&\quad-2KS(\cos^2\theta-\sin^2\theta)S_z+4\pi g\beta M_SS_z,\\ iES_z&=2JSa^2\nabla^2S_Y\\&\quad-2KS(\cos^2\theta-\sin^2\theta)S_Y+2K'SS_Y-\Gamma S_z.\end{aligned} \tag{8}$$

Now, if we insert the pure translational solution,

$$S_z=C\sin\theta,\quad S_y=B\sin\theta,$$

we get

$$\begin{aligned}-iEB&=4\pi g\beta M_SC,\\ iEC&=2K'SB-\Gamma C,\end{aligned} \tag{9}$$

and

$$E^2-iE\Gamma-(2K'S)(4\pi g\beta M_S)=0.$$

If $\Gamma\ll4\pi g\beta M_S$ and $2K'S\ll4\pi g\beta M_S$, the amplitude of S_z is much smaller than the amplitude of S_Y. The energy obtained for very small damping is the wall resonance energy; Eqs. (9) give the same result as the Döring formula using the wall mass, $\omega_0=(\alpha/m_w)^{\frac{1}{2}}$, with $m_w=[\hbar^2/4\pi(g\beta)^2](1/\lambda^2)(1/S_0)\sum_i\sin^2\theta_i$ (see reference 3), and using formula (6),

$$\hbar\omega_0=(2K'S)^{\frac{1}{2}}(4\pi g\beta M_S)^{\frac{1}{2}}.$$

In iron ω_0 is of the order of 500 Mc/sec.

III. GENERAL SOLUTION

S_Y and S_z may be written:

$$S_{Y\text{ or }z}=Z(z)\exp(iK_XX+iK_YY).$$

We are left with the set of equations for the z dependence:

$$\begin{aligned}-iES_Y&=2JSa^2(d^2S_z/dz^2)\\&\quad-2KS(\cos^2\theta-\sin^2\theta)S_z+4\pi g\beta M_SS_z,\\ iES_z&=2JSa^2(d^2S_Y/dz^2)\\&\quad-2KS(\cos^2\theta-\sin^2\theta)S_Y+2K'SS_Y-\Gamma S_z.\end{aligned} \tag{10}$$

The equation,

$$\epsilon Z=2SJSa^2(d^2Z/dz^2)-2KS(\cos^2\theta-\sin^2\theta)Z,$$

has the following solutions:

$$\begin{aligned}Z(z)=\exp\{&\pm[1-(\epsilon/2KS)]^{\frac{1}{2}}(z/\lambda)\}\\&\times\{\pm\tanh(z/\lambda)+[1-(\epsilon/2KS)]^{\frac{1}{2}}\}.\end{aligned}$$

The general solution is any combination of these two. The only well-behaved solution for $z\rightarrow\infty$ occurs when $\epsilon=0$ (translation) or when $\epsilon>2KS$ (spin-wave-like solution). The excitation spectrum may be divided into two branches:

1. A typical wall-type excitation is

$$\begin{aligned}S_Y&=[B/\cosh(z/\lambda)]\exp[i(K_XX+K_YY)],\\ S_z&=[C/\cosh(z/\lambda)]\exp[i(K_XX+K_YY)].\end{aligned}$$

C and B obey the equations,

$$\begin{aligned}iEB&=(2JSa^2k^2+4\pi g\beta M_S)C,\\ -iEC&=(2JSa^2k^2+2K'S)B-\Gamma C;\end{aligned} \tag{11}$$

then

$$\begin{aligned}E^2-iE\Gamma-(2K'S+2JSa^2k^2)&\\ \times(4\pi g\beta M_S+2JSa^2k^2)&=0,\end{aligned} \tag{12}$$

with $k^2=K_X{}^2+K_Y{}^2$. The wave vector has no component along z.

[4] W. Döring, Z. Naturforsch. **3a**, 373 (1948).

The wall excitation may be described as a translation in the z direction, the amplitude of the translation varying sinusoidally in x and y directions. The wall excitation branch is very important for the nuclear relaxation, because the bottom of the spectrum occurs at a relatively low energy. The S_Y motion is nonuniform in the z direction, explaining the occurrence of several relaxation times. This branch does not exist outside the wall.

2. There is a spin-wave-like excitation[5]:

$$S_Y = Be^{i\mathbf{K}\cdot\mathbf{R}}[\tanh(z/\lambda)+i\lambda k_z],$$
$$S_z = (C/B)S_Y,$$

with the equations (neglecting the damping term)

$$\begin{aligned} -iEB &= (2JSa^2k^2+4\pi g\beta M_S+2KS)C, \\ iEC &= (2JSa^2k^2+2K'S+2KS)B; \end{aligned} \tag{13}$$

then

$$E^2 = (2KS+2K'S+2JSa^2k^2)\times(2KS+4\pi g\beta M_S+2JSa^2k^2). \tag{14}$$

The lowest state occurs at an energy $(2KS)^{\frac{1}{2}}(4\pi g\beta M_S)^{\frac{1}{2}}$ higher than the bottom of the wall excitation branch, assuming $4\pi g\beta M_S > 2KS > 2K'S$.

We must remember that the calculation is valid only for small values of k and inside the Bloch wall. It is well known that for a uniform ferromagnet[6] there are several spin-wave branches depending on the angle between $\mathbf{k}$ and the magnetization. The bottom of the branches varies between $2KS$ and $(2KS)^{\frac{1}{2}}(4\pi g\beta M_S)^{\frac{1}{2}}$. In the wall for low k the faster variation of $\mathbf{S}$ is always in the z direction, and only the branch corresponding to excitation in a direction transverse to $\mathbf{S}$ is found.

The occurrence of the term $\tanh(z/\lambda)$ in the solution shows that for small k the amplitude of the motion inside the wall is reduced. For this reason the spin-wave-like solution is not very important for the relaxation in the wall.

IV. AMPLITUDE OF THE THERMAL MOTION

We need to calculate the amplitude of the motion for a wall excitation. The energy of the excitation is

$$\begin{aligned} E_{\text{ex}} = &-Ja^2\textstyle\sum_i \mathbf{S}_i\cdot\nabla^2\mathbf{S}_i \\ &+K\textstyle\sum_i[(S_i^Y)^2+(S_i^z)^2](\cos^2\theta_i-\sin^2\theta_i) \\ &+K'\textstyle\sum_i(S_i^Y)^2+2\pi g\beta\sum_i(S_i^z)^2, \end{aligned} \tag{15}$$

or

$$E_{\text{ex}} = [Ja^2k^2(C^2+B^2)+K'B^2+2\pi(g\beta)^2C^2]\textstyle\sum_i\sin^2\theta_i.$$

This energy is also $E_{\text{ex}} = n(E)E$, where $n(E)$ is the number of magnons at a given temperature (we shall call magnons the wall-type excitation as well as the spin-wave-type excitation).

Then we find

$$\begin{aligned} C^2 &= \frac{a^2S}{2S_0}\left(\frac{K}{J}\right)^{\frac{1}{2}} n(E)\left(\frac{2K'S+2JSa^2k^2}{4\pi g\beta M_S+2JSa^2k^2}\right)^{\frac{1}{2}}, \\ B^2 &= \frac{a^2S}{2S_0}\left(\frac{K}{J}\right)^{\frac{1}{2}} n(E)\left(\frac{4\pi g\beta M_S+2JSa^2k^2}{2K'S+2JSa^2k^2}\right)^{\frac{1}{2}}. \end{aligned} \tag{16}$$

The amplitude in the spin-wave branch may be calculated in the same way (we neglect K' here):

$$\begin{aligned} B^2 &= \frac{a^3}{V_0}\frac{n(E)S}{1+\lambda^2k_z^2}\left(\frac{2KS+4\pi g\beta M_S+2JSa^2k^2}{2KS+2JSa^2k^2}\right)^{\frac{1}{2}}, \\ C^2 &= \frac{a^3}{V_0}\frac{n(E)S}{1+\lambda^2k_z^2}\left(\frac{2KS+2JSa^2k^2}{2KS+4\pi g\beta M_S+2JSa^2k^2}\right)^{\frac{1}{2}}, \end{aligned} \tag{17}$$

where V_0 is the volume of the sample.

All these equations implicitly assume a small damping; but as far as the relaxation time is concerned, the damping plays a fundamental role and the formula (16) is not very useful. The quantities we are really interested in are the spectral density of S_Y and S_z for a frequency ω_0.[7]

Starting from Eqs. (10) and using the random-force technique,[8] $(S_Y^2)_\omega$ spectral density due to thermal excitation is given by

$$(S_Y^2)_\omega = \sin^2\theta_i\frac{a^2S}{S_0}\left(\frac{K}{J}\right)^{\frac{1}{2}}\frac{(K_BT)(\Gamma_K)}{\pi}\times\frac{4\pi g\beta M_S+2JSa^2k^2}{(\omega^2-E'^2)^2+\omega^2\Gamma_K^2}, \tag{18}$$

with

$$E'^2 = (2K'S+2JSa^2k^2)(4\pi g\beta M_S+2JSa^2k^2),$$

assuming $KT\gg\omega$ (classical approximation).

In the limit Γ_K going to zero, Eq. (18) becomes

$$(S_Y^2)_\omega = \sin^2\theta_i\frac{a^2S}{S_0}\left(\frac{K}{J}\right)^{\frac{1}{2}}K_BT\frac{(4\pi\beta M_S+2JSa^2k^2)}{E'}\times[\delta(\omega-E')+\delta(\omega+E')],$$

which is (16) for $K_BT\gg E'$.

From (11) we also derive

$$(S_z^2)_\omega = \frac{\omega^2}{(4\pi g\beta M_S+2JSa^2k^2)^2}(S_Y^2)_\omega;$$

then

$$(S_z^2)_\omega \ll (S_Y^2)_\omega.$$

[5] C. Kittel, *Introduction to Solid-State Physics* (John Wiley & Sons, Inc., New York, 1956), 2nd ed., Appendix O.

[6] C. Herring and C. Kittel, Phys. Rev. **81**, 869 (1951).

[7] A. Abragam, *Principles of Nuclear Magnetism* (Oxford University Press, New York, 1961).

[8] L. D. Landau and E. Lifschitz, *Statistical Physics* (Pergamon Press, New York, 1958), p. 387.

V. NUCLEAR RELAXATION TIME

The nuclear spins are coupled to the electronic spin by the hyperfine coupling,

$$\mathcal{H}_{\text{int}}=\sum_i A\mathbf{I}_i\cdot\mathbf{S}_i.$$

The nuclear resonance energy is obtained by replacing $\mathbf{S}$ by its mean value,

$$\langle\mathcal{H}_{\text{int}}\rangle=\sum_i I_X{}^i\langle S_X{}^i\rangle,$$

and then

$$\omega_0=A\langle S_X{}^i\rangle,$$

which is proportional to $AM(T)$, and at very low temperature $\omega_0=AS$.

For the relaxation time calculation we are interested in the fluctuations of the hyperfine interaction around its mean value,

$$\mathcal{H}_{\text{int}}-\langle\mathcal{H}_{\text{int}}\rangle=\sum_i [AI_X{}^i(S_X{}^i-\langle S_X{}^i\rangle) +A(I_Y{}^iS_Y{}^i+I_z{}^iS_z{}^i)].$$

The term $S_X-\langle S_X\rangle$ is proportional to $S_Y{}^2+S_z{}^2$ and is smaller than the transverse terms S_Y and S_z, and we shall neglect it for the moment. T_1 may be computed using the transition probability formula,

$$\left(\frac{1}{T_1}\right)_i=\frac{2\pi}{\hbar}\left(\frac{A^2}{4}\right)\{\sum_{E_i,E_f}[(\langle E_i|S_Y{}^i|E_F\rangle)^2 +(\langle E_i|S_z{}^i|E_F\rangle)^2]\rho(E_f)\delta(E_i-E_f), \quad (19)$$

where E_i is the energy in the initial state, E_f is the energy in the final state, and $S_Y{}^i$ and $S_z{}^i$ are considered as operators in the electronic system. In our semiclassical calculation $|\langle E_i|S_Y{}^i|E_F\rangle|^2\delta(E_i-E_f)$ may be replaced by $(S_Y{}^i)_{\omega_0}{}^2$; formula (19) becomes

$$\left(\frac{1}{T_1}\right)_i=\frac{1}{\hbar}\frac{\pi A^2}{2}\sum_k[(S_Y{}^i)_\omega{}^2+(S_z{}^i)_\omega{}^2]. \quad (20)$$

The term in S_z may be neglected.

Now for T_2 we note that the perturbation, being along the Y axis, only induces transition when the spin is in the z direction; as the spin is rotating around the X axis it spends only half its time in the z direction and the relaxation rate is half as fast. More generally, T_2 is found to be twice T_1 when the perturbation has no component along the direction of the magnetic field at the nuclei.

If the damping is neglected, by using (20) and (19) a zero relaxation rate is found if the following condition is fulfilled;

$$\omega_0<\Delta'=(2K'S)^{\frac{1}{2}}(4\pi g\beta M_S)^{\frac{1}{2}}. \quad (21)$$

This condition expresses the fact that the minimum energy of the wall excitation is larger than the nuclear Zeeman splitting. In such a condition the energy cannot be conserved in a transition involving the emission or destruction of a single magnon accompanied by a nuclear spin flip. The lifetime of the magnons being not infinite, there is a certain indeterminancy in their energy, and the nuclei may absorb or emit magnons which differ in energy from ω_0.

We shall first discuss T_1 when $\omega_0>\Delta'$ without damping and then take damping into account. The density of states per unit energy range $\rho(E)$ for the wall-type excitation is

$$\rho(E)=Es_0/2\pi(2JSa^2)[2K'S+4\pi g\beta M_S+4JSa^2k^2], \quad E>\Delta', \quad (22)$$
$$\rho(E)=0, \quad E<\Delta',$$

using the fact that the number of states with energy in dE at E is

$$(2\pi/(2\pi)^2)k(dk/dE)dES_0.$$

From (22) we deduce the useful relation,

$$(S_Y{}^2+S_z{}^2)_E{}^i\rho(E)=\sin^2\theta\, n(E)(1/8\pi J)(K/J)^{\frac{1}{2}}. \quad (23)$$

The relaxation time is

$$\left(\frac{1}{T_1}\right)_i=\frac{1}{\hbar}\frac{\omega_0{}^2}{16S^2}\sin^2\theta_i\, n(\omega_0)\frac{1}{J}\left(\frac{K}{J}\right)^{\frac{1}{2}}$$
$$=\sin^2\theta_i\frac{\omega_0}{16S}\left(\frac{K_BT}{JS}\right)\left(\frac{K}{J}\right)^{\frac{1}{2}}\frac{1}{\hbar}, \quad (24)$$

when $KT\gg\omega_0$ and $\omega_0>\Delta'$, and $(1/T_1)_i=0$ when $\omega_0<\Delta'$. Now, if we take the damping into account, (20) is written

$$\left(\frac{1}{T_1}\right)_i=\frac{A^2}{2\hbar}\frac{a^2S}{S_0}\left(\frac{K}{J}\right)^{\frac{1}{2}}\sin^2\theta_i\,(K_BT)(\Gamma) \times\sum_k\frac{4\pi g\beta M_S+2JSa^2k^2}{(\omega_0{}^2-E'^2)+\omega^2\Gamma^2},$$

assuming Γ independent of K. The summation in k is replaced by an integration and, taking E' as a new variable,

$$\left(\frac{1}{T_1}\right)_i=\frac{A^2}{2h}\sin^2\theta_i\left(\frac{K}{J}\right)^{\frac{1}{2}}S(\Gamma)(K_BT) \times\int_{\Delta'}^\infty\frac{E'}{4\pi JS}\frac{4\pi g\beta M_S+2JSa^2k^2}{2K'S+4\pi g\beta M_S+4JSa\,k^2} \times\frac{dE'}{(\omega_0{}^2-E'^2)^2+\omega_0{}^2\Gamma^2}.$$

The main contribution to the integral comes from values of E' such as $E'<\Gamma$ or $2JSa^2k^2<\Gamma^2/4\pi g\beta M_S$ or $\Gamma<4\pi g\beta M_S$. $2JSa^2k^2$ may be neglected compared to

$4\pi g\beta M_S$. The result is easily obtained:

$$\left(\frac{1}{T_1}\right)_i=\sin^2\theta_i\frac{\omega_0}{\hbar}\frac{K_BT}{16\pi JS}\left(\frac{K}{J}\right)^{\frac{1}{2}}\times\left[\frac{\pi}{2}-\tan^{-1}\left(\frac{\omega_0^2-\Delta'^2}{\omega_0\Gamma}\right)\right], \quad (25)$$

or

$$\sin^2\theta_i\frac{\omega_0}{\hbar}\frac{K_BT}{16\pi JS}\left(\frac{K}{J}\right)^{\frac{1}{2}}\tan^{-1}\left(\frac{\omega_0\Gamma}{\Delta'^2-\omega_0^2}\right).$$

If Γ is small compared to ω_0 and Δ, we find the same result as before for $\omega_0>\Delta'$, but for $\omega_0<\Delta'$,

$$\frac{1}{T_1}=\left(\frac{1}{T_1}\right)_{\omega_0>\Delta'}\frac{\omega_0\Gamma}{\Delta'^2-\omega_0^2}.$$

If Γ is very large, more precisely $\omega_0\Gamma>|\Delta'^2-\omega_0^2|$, then

$$1/T_1=\tfrac{1}{2}(1/T_1) \quad \text{for} \quad \Gamma=0, \quad \omega_0>\Delta.$$

T_1 becomes independent of Γ and Δ'.

Comparison with experiment is not very easy. The measurements in the three metals show a spread of relaxation times suggesting an important relaxation by wall-type excitations. But for comparison with theory, we want to know the shortest relaxation time (i.e., the relaxation time at the center of the wall), and the extrapolation is not easy. On the other hand, Γ and Δ' are not too well known. In metals the damping comes from the effects of eddy currents and depends on the size of the powder; Δ' is related to the stiffness parameter and is a structure-sensitive quantity. In iron Δ'/h is typically 500 Mc/sec and ω_0/h is 45 Mc/sec. The parameter Γ/h may very well vary between 1000 Mc/sec to 20 Mc/sec, according to the size of the powder.

At room temperature in iron $(K/J)^{\frac{1}{2}}=1.7\times10^{-2}$, and

$$K_BT/JS=1.9.$$

Equation (25) for a nuclear spin at $z=0$ may be written

$$1/T_1=0.65\times10^{-3}(\omega_0/\hbar)\tan^{-1}(\omega_0\Gamma/\Delta'^2).$$

Assuming $\Gamma/h=1000$ Mc/sec, we find

$$1/T_1=3\times10^4\ \text{sec}^{-1}.$$

Now, assuming $\Gamma/h=20$ Mc/sec,

$$1/T_1=0.6\times10^3\ \text{sec}^{-1}.$$

The experimental value is 1.1×10^3 sec^{-1}.

In cobalt the comparison is nearly impossible, because the value of K in the cubic phase is not known. In nickel the experimental result at room temperature $1/T_1=3\times10^3$ sec^{-1} is explained if we assume $\Delta'/h=500$ Mc/sec and $\Gamma/h=300$ Mc/sec. Taking into account the uncertainty in both theoretical and experimental results, the agreement is not too bad.

The parameters Γ and Δ' being temperature-dependent, the relaxation rate at the center of the wall does not vary with temperature as kT. The effect of the remaining term $S_X-\bar{S}_X$ must be estimated. This term gives a contribution only to T_2. In the quantum-mechanical description this term involves two magnon operators and the relaxation is produced by a Raman scattering of magnons. The relaxation rate is given by

$$(1/T_2)_i=(\pi/8)(A^2/\hbar S)\int_0^\infty\int_0^\infty[(S_Y{}^i)^2+(S_z{}^i)^2]\times n(E)[n(E')+1]\rho(E)\rho(E')\delta(E-E')dEdE'.$$

Using the high-temperature approximation, we obtain

$$(1/T_2)_i=\sin^4\theta_i(\omega_0^2/\hbar S^4)(1/64\pi)(K/J)(K_BT/J)^2(1/\Delta').$$

The numerical evaluation gives $1/T_2$ much smaller than 10^3 sec^{-1} at room temperature in the three metals, and this process may be neglected.

VI. INDIRECT INTERACTION

Several years ago Suhl showed that in a ferromagnet an indirect coupling exists between nuclear spins, involving a virtual excitation of a spin wave.[9] The same kind of interaction may be computed in the wall using a virtual excitation of a wall magnon. It is convenient to quantize the wall magnons. S_Y and S_z are written in the following way:

$$\begin{aligned}S_Y{}^i&=(S/2)^{\frac{1}{2}}[2(J/K)^{\frac{1}{2}}(S_0/a^2)]^{-\frac{1}{2}}\times\textstyle\sum_k[a_K{}^\dagger\exp(i\mathbf{K}\cdot\mathbf{R}_i)+a_K\exp(-i\mathbf{K}\cdot\mathbf{R}_i)]\times\sin\theta_i,\\ S_z{}^i&=-i(S/2)^{\frac{1}{2}}[2(J/K)^{\frac{1}{2}}(S_0/a^2)]^{-\frac{1}{2}}\times\textstyle\sum_k[a_K{}^\dagger\exp(i\mathbf{K}\cdot\mathbf{R}_i)-a_K\exp(-i\mathbf{K}\cdot\mathbf{R}_i)]\times\sin\theta_i.\end{aligned} \quad (26)$$

$a_K{}^\dagger$ and a_K are usual creation and destruction operators and the term before the summation is a normalizing factor. Using Eq. (15), the Hamiltonian becomes

$$\mathcal{H}=\textstyle\sum_k[(2JSa^2k^2+K'S+2\pi g\beta M_S)a_K{}^\dagger a_K+(K'S-2\pi g\beta M_S)(a_Ka_{-K}+a_K{}^\dagger a_{-K}{}^\dagger)]. \quad (27)$$

The Hamiltonian may be diagonalized by the usual canonical transformation,[10]

$$b_K=\mu_Ka_K+\nu_Ka_{-K}{}^\dagger, \quad \mu_K{}^2-\nu_K{}^2=1.$$

The transverse part of the hyperfine coupling is written in terms of the operators b_K and $b_K{}^\dagger$:

$$\mathcal{H}_{\text{int}}=\sum_{i,k}(A/2)(2S)^{\frac{1}{2}}[(K/J)^{\frac{1}{2}}(2a^2/S_0)]^{\frac{1}{2}}\times\sin\theta_i[I_+{}^i\exp(i\mathbf{K}\cdot\mathbf{R}_i)\times(\mu_Kb_K{}^\dagger-\nu_Kb_K)+\text{complex conjugate}], \quad (28)$$

$$I_+=I_Y+iI_z,$$

[9] H. Suhl, J. phys. radium **20**, 333 (1959).
[10] T. Holstein and H. Primakoff, Phys. Rev. **58**, 1098 (1940).

and the indirect interaction between the nuclear spins i and j is, keeping only the secular part,

$$\mathcal{H}_{ij}=\sum_k (A^2S/2)(K/J)^{\frac{1}{2}} \times (S_0/2a^2)\sin\theta_i \sin\theta_j \exp(i\mathbf{K}\cdot\mathbf{R}_{ij}) \times (I_+{}^iI_-{}^j+I_-{}^iI_+{}^i)\{(\mu_K{}^2+\nu_K{}^2) \times[2E/(E^2-\omega_0{}^2)]+2\omega_0/(E^2-\omega_0{}^2)\}. \quad (29)$$

For low k, μ_K and ν_K are very large compared to one, and expanding in the power of k, keeping only the terms in k^2, Eq. (29) becomes

$$\mathcal{H}_{ij}=(A^2/8\pi)(K/J)^{\frac{1}{2}}(1/2J) \times\int_0^\infty\int_0^\infty \exp(i\mathbf{K}\cdot\mathbf{R}_{ij})/(\beta^2+k^2)dK_XdK_Y, \quad (30)$$

$$\beta^2=\frac{1}{a^2}\frac{\Delta'^2-\omega_0{}^2}{4\pi g\beta M_S}\frac{1}{2JS},$$

if $\omega_0\ll\Delta'$, $\beta=(1/a)(K'/J)^{\frac{1}{2}}$. $1/\beta$ is a measure of the range of the interaction in the X and Y direction and is usually larger than the wall thickness λ. The integral over K is (assuming β real)[11]

$$\int\frac{\exp(i\mathbf{K}\cdot\mathbf{R})dk\, k\sin\theta d\theta}{\beta^2+k^2} =\frac{1}{2\beta R}[e^{-\beta R}\bar{E}_i(\beta R)-e^{\beta R}E_i(-\beta R)]=\tfrac{1}{2}I(\beta R).$$

The Van Vleck second moment for a nucleus at the center of the wall is given by the formula

$$h^2\Delta\nu^2=(A^2/16\pi J)^2(K/J)[I(I+1)/3]\sum_j \sin^2\theta_j I^2(\beta R_{ij}).$$

We obtain, after using an integration instead of a summation,

$$h\langle\Delta\nu^2\rangle^{\frac{1}{2}}=(A^2/4\pi J)(K/J)^{\frac{1}{4}}\left[\frac{I(I+1)}{3}\right]^{\frac{1}{2}} \times\left[\frac{(4\pi g\beta M_S)(2JS)}{\Delta'^2-\omega_0{}^2}\right]^{\frac{1}{2}},$$

and, if $\omega_0\ll\Delta'$,

$$h\langle\Delta\nu^2\rangle^{\frac{1}{2}}=(A^2/4\pi J)[I(I+1)/3]^{\frac{1}{2}}(K/K')^{\frac{1}{2}}(J/K)^{\frac{1}{4}}.$$

The second moment is larger by a factor $(K/K')^{\frac{1}{2}}$ than the second moment calculated by Suhl for a uniform ferromagnet.

If $\omega_0>\Delta$, the integration in k space may be done (taking the principal part of the integral), but the range becomes infinite giving an infinite second moment. In fact the range can never be infinite because of the finite lifetime of magnons and they cannot propagate the interaction over infinite distances.

[11] A. Erdelyi, W. Magnus, F. Oberhettinger, and F. G. Tricomi, *Tables of Integral Transform* (McGraw-Hill Book Company, Inc., New York, 1954), Vol. 1, p. 56.

VII. MAGNETIZATION IN THE WALL

The relative variation of S is given by $\Delta S_X/S=(S_Y{}^2+S_z{}^2)/2S$. The magnitude of the magnetization is no longer uniform, because S_Y and S_z vary along the wall. The nonuniformity of the magnetization gives a shift and a line width to the nuclear resonance line (and also an asymmetry).

The mean value of ΔS_X is easily evaluated for the wall-type excitation, using the relations (16) and (23),

$$[(S_Y{}^i)^2+(S_z{}^i)^2]/2S =\sin^2\theta_i(1/16J\pi S^2)(K/J)^{\frac{1}{2}}\int_{\Delta'}^\infty n(E)dE =\sin^2\theta_i(KT/16\pi JS^2)(K/J)^{\frac{1}{2}}\ln(K_BT/\Delta').$$

The effect is maximum at the center of the wall.

The contribution of the spin-wave branch is given by the equation,

$$\left(\frac{\Delta S_X}{S}\right)_{\text{S-W}}=\frac{1}{2S^2}\sum_k n(E)\frac{a^3}{V_0}\frac{S[\lambda^2k_z{}^2+\tanh(z/\lambda)]}{1+\lambda^2k_z{}^2} \times\frac{4KS+4\pi g\beta M_S+4JSa^2k^2}{E}.$$

We are especially interested in the difference between $\Delta S/S$ at the center of the wall and outside the wall,

$$\left(\frac{\Delta S}{S}\right)_{\text{edge}}-\left(\frac{\Delta S}{S}\right)_{\text{center}}=\frac{1}{2S^2}\sum_k n(E)\frac{a^3S}{V_0}\frac{1}{1+\lambda^2k_z{}^2} \times\frac{4KS+4\pi g\beta M_S+4JSa^2k^2}{E}.$$

The main contribution to the integral is obtained when $k_z\lambda<1$, and we always have $2JSk_z{}^2a^2\ll kT$, when $K_BT\gg K$. The integral is approximately

$$\frac{1}{2S}\frac{a^3}{8\pi^3}\int_0^\infty\frac{dk_z}{1+\lambda^2k_z{}^2}\int_0^\infty\frac{1}{\exp(E_0/K_BT)-1} \times\frac{4KS+4\pi g\beta M_S+4JSa^2k_\perp{}^2}{E_0}dK_XdK_Y,$$

with $E_0=E$ (for $k_z=0$), and $k^2=k_z{}^2+k_\perp{}^2$.

The integral in K_X and K_Y is very similar to the integral we use for calculating $\Delta S/S$ coming from the wall excitation branch except that Δ' has to be replaced by Δ,

$$\Delta=(2KS)^{\frac{1}{2}}(4\pi g\beta M_S)^{\frac{1}{2}}.$$

By adding the two contributions which act in opposite

directions, we get

$$\left(\frac{\Delta S}{S}\right)_{\text{center}}-\left(\frac{\Delta S}{S}\right)_{\text{edge}}=\frac{1}{8\pi S}\left(\frac{K}{J}\right)^{\frac{1}{2}}\left(\frac{KT}{2JS}\right)\ln\left(\frac{\Delta}{\Delta'}\right)=\frac{1}{16\pi S}\left(\frac{K}{J}\right)^{\frac{1}{2}}\left(\frac{KT}{2JS}\right)\ln\left(\frac{K}{K'}\right). \quad (31)$$

It may be useful to compare this result to the $\Delta S/S$ in a uniform ferromagnet:

$$\left(\frac{\Delta S}{S}\right)_{\text{center}}-\left(\frac{\Delta S}{S}\right)_{\text{edge}}=\left(\frac{\Delta S}{S}\right)_{\text{uniform}}0.33\left(\frac{K}{J}\right)^{\frac{1}{2}}\left(\frac{KT}{2JS}\right)\ln\left(\frac{K}{K'}\right).$$

The maximum relative variation of the resonance frequency is

$$\Delta\nu/\nu=\tfrac{1}{3}(2KS/KT)^{\frac{1}{2}}\ln(K/K')(\Delta M/M)_{\text{uniform}}. \quad (32)$$

The frequency is lower in the wall. The effect of the spin-wave branch has already been estimated by Suhl,[12] using a different method.

For iron at room temperature we obtain (assuming $K/K'=100$)

$$\Delta\nu=18 \text{ kc/sec}.$$

The effect is not negligible; the observed linewidths are of the same order of magnitude, but they show a very small temperature dependence. Here also the theory predicts the maximum value for the linewidth. The linewidth due to this effect may be smaller if the observed spins are not at the center of the wall.

[12] H. Suhl, Bull. Am. Phys. Soc. **5**, 175 (1960).

VIII. CUBIC ANISOTROPY, 90° WALL

The anisotropy energy is

$$\mathcal{H}_A=(K/S^2)[S_x^2(S_y^2+S_z^2)+S_y^2S_z^2], \quad (33)$$

which becomes, in the new frame (using only quadratic terms),

$$\mathcal{H}_A=(K/S^2)\textstyle\sum_i[(S_x^i)^4\sin^2\theta_i\cos^2\theta_i+(S_x^i)^2(S_z^i)^2+(S_x^i)^2(S_Y^i)^2(1-6\cos^2\theta_i\sin^2\theta_i)].$$

The minimization of energy gives the condition,

$$K\sin^2\theta_i\cos^2\theta_i=J\Delta\theta_i^2.$$

Then

$$d\theta/dz=(1/\lambda)(\sin2\theta/2),\quad \sin2\theta=1/\cosh(z/\lambda),\quad 0<\theta<\pi/2.$$

The equations of motion for the z dependence are

$$\begin{aligned} iES^Y&=-2JSa^2(d^2S_z/dz^2)+2KSS_z(1-3\sin^2\theta\cos^2\theta)-4\pi g\beta M_SS_z,\\ iES_z&=2JSa^2(d^2S_Y/dz^2)-2KSS_Y(1-8\sin^2\theta\cos^2\theta)+2K'SS_Y-\Gamma S_z. \end{aligned} \quad (34)$$

The exact solution is difficult to find, but if we remember that the largest term in the first equation is the demagnetizing term, S_z may be eliminated using $iES^Y=-4\pi g\beta M_SS_z$, and the equation of S_Y is the same as before.

The same considerations are true for low k_z in the spin-wave spectrum; and, as in all our calculations, the effects are important only when $S_Y>S_z$ and the results are not changed.

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