Nuclear Magnetic Resonance within an Antiferromagnetic Bloch Wall*

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In a previous paper by the author, the bound spin wave excitation spectrum existing within an antiferromagnetic Bloch wall was derived. We calculate, here, the effect of this energy excitation spectrum on the linewidth and relaxation times of the magnetic resonance of nuclei existing within the wall. A relaxation time T_1, caused by excitation of the transverse components of the electron spin by the resonance field, is shown to be smaller in antiferromagnets than in ferromagnets due to the larger minimum wall excitation energy. The linewidth due to indirect coupling of the nuclear spins through the virtual excitation of a spin wave, however, proves to be increased by the square root of the ratio of the anisotropy to the stiffness parameters compared to the uniform antiferromagnet. This may account for the rather large linewidths observed in nuclear magnetic resonance experiments in antiferromagnets. A second nuclear linewidth, resulting from the variation of the deviation of the longitudinal component of the electron spin from the center to the edge of the wall is also calculated. Numerical results are obtained and, where possible, compared with experiment.

I. INTRODUCTION

THE existence of antiferromagnetic domains has been experimentally demonstrated both by neutron and optical studies.[1] In a previous paper by the author[2] (hereafter referred to as DP I), explicit formulas were derived for the bound excitation spectrum of the electron spins within the wall as well as the free spin wave spectrum outside of the wall. In this paper we estimate the effect of the antiferromagnetic wall excitation spectrum on the resonance spread and relaxation times of nuclei in nuclear magnetic resonance experiments. The analogous ferromagnetic case has been considered by Winter[3] and by Boutron and deGennes.[4]

II. FORMULATION OF THE PROBLEM

Consider an infinite antiferromagnetic crystal possessing orthorhombic type magnetic spin symmetry, the spins being located on two interpenetrating sublattices A and B such as in $CuCl_2\cdot 2H_2O$. The crystal is divided into two domains by a 180° Bloch wall perpendicular to the z axis with the center of the wall located at z equals zero. In the absence of a perturbation (i.e., spin wave), the magnetization vectors outside of the wall are in the plus x direction on sublattice A and in the minus x direction on sublattice B for z less than zero and in the reverse directions for z greater than zero. As shown in DP I, the Bloch wall separating the domains has a finite thickness symbolized by the angle θ which measures the rotation away from the x axis in the plane of the Bloch wall of the static magnetization on sublattice A. Thus, as is illustrated in Fig. 1, we can choose a second system of axes, X, Y, and z where X is the spin direction for the static magnetization and X and Y vary from atom to atom while z is not changed.

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

$$\mathcal{H}=\sum_i A\mathbf{I}_i\cdot\mathbf{S}_i+\sum_j A\mathbf{I}_j\cdot\mathbf{s}_j, \tag{1}$$

where $\mathbf{S}_i$ is the spin of the ith atom on sublattice A and correspondingly $\mathbf{s}_j$ is the spin of the jth atom on sublattice B. If the frequency of the fluctuation of the electron spin around its mean value $\langle \mathbf{S}_X{}^i\rangle$ or $\langle \mathbf{s}_X{}^j\rangle$ is of the correct order of magnitude, then its transverse component will cause a relaxation of the nuclear spins. The fluctuation of the hyperfine interaction about its mean value is

$$\begin{aligned}\mathcal{H}_{\text{int}}-\langle\mathcal{H}_{\text{int}}\rangle=&\sum_i A[I_X{}^i(S_X{}^i-\langle S_X{}^i\rangle)+I_Y{}^iS_Y{}^i+I_z{}^iS_z{}^i]\\&+\sum_j A[I_X{}^j(s_X{}^j-\langle s_X{}^j\rangle)+I_Y{}^js_Y{}^j+I_z{}^js_z{}^j].\end{aligned} \tag{2}$$

The terms $S_X{}^i-\langle S_X{}^i\rangle$ and $s_X{}^j-\langle s_X{}^j\rangle$ are smaller than the transverse terms (as shown in Sec. III), and we shall neglect them.

In DP I, we showed that there exists two sets of solutions for the bound states of the electron spins.

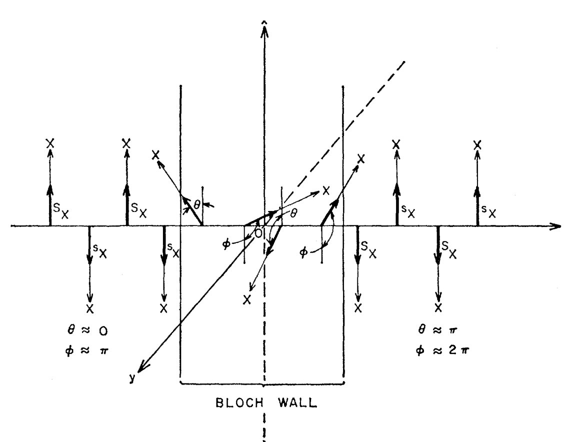

FIG. 1. Diagram showing fixed axis z perpendicular to the Bloch wall, angles of deviation θ and ϕ of the different sublattice spins inside the wall, and varying coordinate axis X.

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[1] W. Roth, J. Appl. Phys. 31, 2000 (1960).
[2] D. Paul, Phys. Rev. 126, 78 (1962).
[3] J. M. Winter, Phys. Rev. 124, 452 (1961).
[4] Mlle. F. Boutron and P. G. deGennes, Compt. rend. 253, 1662 (1961).

Let us consider the first set given by the relations

$$S_Y = s_Y = B\sin\theta\exp[i(k_x x + k_y y)] = \sigma_Y,$$

and

$$S_z = s_z = C\sin\theta\exp[i(k_x x + k_y y)] = \sigma_z, \quad (3)$$

where as in DP I, the direction of the coordinate Y of $\mathbf{s}$ is the negative of that of $\mathbf{S}$. Note, also, that σ as used here equals $(1/2)\sigma$ of DP I. Using Eq. (3), the nuclear relaxation time T_1 for this process is given by the formula

$$(1/T_1)_p = \pi A^2(2\hbar)^{-1}\sum_{E_i,E_f}[(\langle E_i|\sigma_Y{}^p|E_f\rangle)^2 + (\langle E_i|\sigma_z{}^p|E_f\rangle)^2]\rho(E_f)\delta(E_i - E_f), \quad (4)$$

where the index p represents any lattice site. Inasmuch as it is important to include the viscosity of the wall, we shall make a semiclassical calculation of T_1. Thus, if we use the spectral densities, $(\sigma_Y{}^2)_\omega$ and $(\sigma_z{}^2)_\omega$ of σ_Y and σ_z for a frequency ω, we may write

$$(1/T_1)_p = \pi A^2(2\hbar)^{-1}\sum_{E_f}[(\sigma_Y{}^p)_\omega{}^2 + (\sigma_z{}^p)_\omega{}^2]\rho(E_f). \quad (5)$$

The spectral densities are obtained from the equations of motion for $\mathbf{S}$ and $\mathbf{s}$ given in DP I, Eqs. (11). Using standard methods given by Landau and Lifschitz[5] and the notation of DP I and reference 3, we get

$$(\sigma_Y{}^2)_\omega = \frac{2a^2S}{\pi S_0}\left(\frac{K_1}{-J}\right)^{\frac{1}{2}}\frac{k_bT\sin^2\theta}{2K'S - 2JSa^2k^2} \times\frac{\Gamma_2(E'^2 + \Gamma_1\Gamma_2) + \omega^2\Gamma_1}{(\omega^2 - E'^2 - \Gamma_1\Gamma_2)^2 + \omega^2(\Gamma_1 + \Gamma_2)^2},$$

$$(\sigma_z{}^2)_\omega = \frac{2a^2S}{\pi S_0}\left(\frac{K_1}{-J}\right)^{\frac{1}{2}}\frac{k_bT\sin^2\theta}{-24JS + 2MS} \times\frac{\Gamma_1(E'^2 + \Gamma_1\Gamma_2) + \omega^2\Gamma_2}{(\omega^2 - E'^2 - \Gamma_1\Gamma_2)^2 + \omega^2(\Gamma_1 + \Gamma_2)^2}, \quad (6)$$

where E' is the energy of the spin wave excitation spectrum given in Eq. (18) of DP I, when the viscosity parameters Γ_1 and Γ_2 are set equal to zero, i.e.,

$$E'^2 = (-48JS + 4MS)(K'S - JSa^2k^2). \quad (7)$$

The quantities J, M, and K' are the exchange, inertia, and stiffness parameters, respectively, while K_1 is the anisotropy parameter in the xy plane. Further, S_0 is the area of the Bloch wall. We have placed the lattice parameters a_x and a_y equal to a (approximately true for such orthorhombic substances as $CuCl_2\cdot H_2O$), and k^2 represents the two-dimensional wave number $k_x{}^2 + k_y{}^2$. From Eqs. (6), we note that for $J \gg K'$, $(\sigma_z{}^2)_\omega \ll (\sigma_Y{}^2)_\omega$. Thus, for this case, the perturbation is along the Y axis and induces T_2-type transitions when the spin is in the z direction only. Inasmuch as the spin rotates around the X axis, the relaxation time T_2 is twice T_1. In the limits Γ_2 going to zero and Γ_1 proportional to Γ_2 we get

$$\lim(\sigma_Y{}^2)_\omega = \frac{k_bTa^2S/S_0}{2K'S - 2JSa^2k^2}\left(\frac{K_1}{-J}\right)^{\frac{1}{2}}\delta(\omega - E)\sin^2\theta,$$

$$\lim(\sigma_z{}^2)_\omega = \frac{k_bTa^2S/S_0}{-24JS + 2MS}\left(\frac{K_1}{-J}\right)^{\frac{1}{2}}\delta(\omega - E)\sin^2\theta. \quad (8)$$

The density of states per unit energy for the bound excitations is obtained from Eq. (7) and is

$$\rho(E) = ES_0/(-2\pi JSa^2)(-48JS + 4MS), \quad E > \Delta',$$
$$\rho(E) = 0, \quad E < \Delta', \quad (9)$$

where

$$\Delta' = [-48JK'S^2 + 4MK'S^2]^{\frac{1}{2}}. \quad (10)$$

Thus, when damping is neglected, there is a zero relaxation rate if the nuclear magnetic resonance frequency, ω_0, is less than Δ'. This is similar to the ferromagnetic case of Winter.[3] However, the minimum energy of the wall excitation may be quite large in the antiferromagnetic case inasmuch as it contains a term proportional to J, the exchange integral, while the ferromagnetic case does not. We shall consider numerical values in Sec. III.

III. CALCULATION OF T_1

We first calculate the nuclear relaxation time T_1 in the limit of zero damping. We note that the Bose statistics for the magnons has the high-temperature limit

$$n(E) = k_bT/E, \quad (11)$$

where $n(E)$ is the number of magnons with energy E at temperature T. Further, we define ω_0 as the nuclear resonance frequency equal to $A\langle\sigma_X\rangle$. Then, Eqs. (8) and (9), when substituted into Eq. (5), yield

$$(1/T_1)_p = 0, \quad \omega_0 < \Delta'$$

$$(1/T_1)_p = \sin^2\theta_p\frac{\omega_0{}^2 n(\omega_0)}{-8\hbar JS^2}\left(\frac{K_1}{-J}\right)^{\frac{1}{2}}\left[1 + \frac{K' - Ja^2k^2}{-12J + M}\right], \quad \omega_0 > \Delta'. \quad (12)$$

This result can be reproduced by the operator technique of Sec. IV since there is no viscosity. We note that the term in the bracket is approximately unity for large J.

If we include damping, then we must use Eqs. (6) rather than Eqs. (8) in Eq. (5) and sum over all the allowed energy states from Δ' to infinity. Neglecting $(\sigma_z{}^2)_\omega$ compared to $(\sigma_Y{}^2)_\omega$ and replacing the summation by an integration, we may write for T_1,

$$\left(\frac{1}{T_1}\right)_p = \sin^2\theta_p\frac{A^2k_bT}{-4\pi\hbar J}\left(\frac{K_1}{-J}\right)^{\frac{1}{2}} \times\int_{\Delta'}^{\infty}\frac{\Gamma_2E' + (\Gamma_1\Gamma_2{}^2/E') + (\omega_0{}^2\Gamma_1/E')}{(\omega_0{}^2 - E'^2 - \Gamma_1\Gamma_2)^2 + \omega_0{}^2(\Gamma_1 + \Gamma_2)^2}dE'. \quad (13)$$

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

Inasmuch as our semiclassical calculations in DP I for the energy excitation spectrum was valid for long wavelengths only, ($ka<1$), it is important to note that the major contributions to the integral come from values of E' such that $E'<\Gamma$. From the definition of E' given in Eq. (7), it is apparent that the shorter wavelengths will not affect the value of the integral. Our result is

$$\left(\frac{1}{T_1}\right)_p=\sin^2\theta_p\frac{A^2k_bT}{-4\pi J\hbar}\left\{\frac{(K_1/-J)^{\frac{1}{2}}}{2\omega_0(\Gamma_1+\Gamma_2)}\right.$$
$$\times\left[\Gamma_2+\frac{\Gamma_1(\omega_0^2+\Gamma_2^2)(\omega_0^2-\Gamma_1\Gamma_2)}{(\omega_0^2-\Gamma_1\Gamma_2)^2+\omega_0^2(\Gamma_1+\Gamma_2)^2}\right]$$
$$\times\tan^{-1}\left[\frac{\omega_0(\Gamma_1+\Gamma_2)}{\Delta'^2-\omega_0^2+\Gamma_1\Gamma_2}\right]$$
$$+\frac{\Gamma_1}{2}\frac{(\Gamma_2^2+\omega_0^2)(K_1/-J)^{\frac{1}{2}}}{(\omega_0^2-\Gamma_1\Gamma_2)^2+\omega_0^2(\Gamma_1+\Gamma_2)^2}$$
$$\times\ln\{[(\Delta'^2-\omega_0^2+\Gamma_1\Gamma_2)^2$$
$$\left.+\omega_0^2(\Gamma_1+\Gamma_2)^2]^{\frac{1}{2}}/\Delta'^2\}\right\}. \quad (14)$$

We consider the case, Γ_1 and Γ_2 small with respect to ω_0 and Δ'. If ω_0 is greater than Δ', we get the same result as Eq. (12) with $J>K'$. If, however, the nuclear resonance frequency ω_0 is less than the minimum wall excitation energy Δ', we find the finite relaxation time

$$\left(\frac{1}{T_1}\right)_{\omega_0<\Delta'}=\frac{\omega_0}{\pi}\frac{\Gamma_1+\Gamma_2}{\Delta'^2-\omega_0^2}\left(\frac{1}{T_1}\right)_{\omega_0>\Delta'}. \quad (15)$$

Thus, we see that the viscosity Γ acts as a drag on the bound electron spin wave excitation causing it to be excited even when the external excitation energy is less than the minimum wall excitation energy. However, for most values of viscosity, this function decreases very rapidly with increasing values of the wall excitation to nuclear resonance frequency ratio. This is illustrated in Fig. 2.

Experimental values for the parameters relating to the motion of the antiferromagnetic Bloch wall are scarce. Roth and Slack[6] have observed domain wall motion in antiferromagnetic NiO and estimate the viscosity, Γ, as approximately 10^{-19} erg/atom per cycle. A similar value is given for Γ in iron powder[3] but ferrites[7] such as Fe_3O_4 appear to have values larger by several orders of magnitude. We shall consider values of Γ between the range 5×10^{-17} and 10^{-20}. The stiffness parameter K' is structure sensitive. Values between 10^{-18} and 5×10^{-20} erg/atom do not appear incompatible

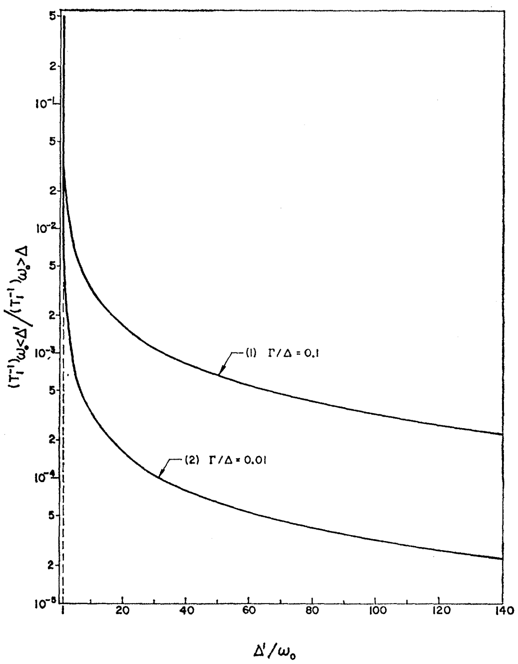

FIG. 2. Decay of spin-lattice relaxation rate as function of ratio of minimum wall excitation energy Δ' to nuclear resonance energy ω_0.

with data[3,6] for NiO or iron powder. For the inertia term, M, we use the value $\pi(g\beta)^2/a^3$ given in reference 7 or approximately 10^{-17}.

If we apply formula (15) to an antiferromagnetic substance with orthorhombic structure, exactly fitting our model, we must use $CuCl_2\cdot2H_2O$ which has a Curie temperature of 4.3°K. However, no nuclear magnetic resonance experiments have been performed using the magnetic copper ion and the proton resonance[8] has a rather weak hyperfine interaction. The nuclear magnetic resonance of the fluorine nuclei in NiF_2 and MnF_2 (both with Curie temperatures of approximately 70°K) have been done by Shulman[9] and Jaccarino and Shulman,[10,11] and exhibit a strong hyperfine interaction. These two substances have a rutile type crystal structure and a body-centered tetragonal type magnetic lattice. The spin of the body-centered magnetic ions are antiparallel to the corner ions, and, thus, magnetically, these two substances consist of two interpenetrating sublattices. Since our wall parameters are uncertain, and we are mainly interested in orders of magnitude, we shall

[6] W. L. Roth and G. A. Slack, J. Appl. Phys. **31**, 3525 (1960).
[7] C. Kittel and J. K. Galt in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1956), Vol. 3.

[8] N. J. Poulis and G. E. G. Hardeman, Physica **18**, 201 (1952).
[9] R. G. Shulman, Phys. Rev. **121**, 125 (1961).
[10] V. Jaccarino and R. G. Shulman, Phys. Rev. **107**, 1196 (1957).
[11] R. G. Shulman, J. Appl. Phys. Suppl. **32**, 1268 (1961) has experimentally observed the effect of the antiferromagnetic domain walls in nuclear magnetic resonance experiments on NiF_2 at 20.3°K, thus fitting in with our hypothesis. Saturation effects appeared for external perturbing fields of magnitude 0.002 G. (Normal resonance for NiF_2 at 20.3°K does not saturate even with fields equal to 0.1 G.) The linewidth at this temperature was estimated at 7.7×10^5 cps, where a Lorentzian line shape was assumed.

apply our model directly to these substances even though they resemble body-centered cubic rather than simple cubic lattices. Using values for the anisotropy and exchange given by Shulman,[9] Jaccarino and Shulman,[10] and Nagamiya *et al.*,[12] we find, for all three substances, that the maximum value of $1/T_1$ (at the center of the wall) varies between $4\times10^3 T$ (Γ large and K' small), to $2\times10^{-2}T$ (Γ small and K' large), for the ranges of wall viscosity and stiffness given above. As Shulman and Jaccarino point out, the resonances actually obtained in NiF_2 and MnF_2 exhibit an unusual line broadening which is Gaussian in shape and of approximately 5×10^4 cps at liquid helium temperatures for NiF_2 and 3×10^4 cps at temperatures between 1.3 and 20.4°K for MnF_2. They suggest that this is due to indirect coupling of nuclear spins, discussed in our Sec. IV.

The next substance we shall consider is $CrCl_3$ with a Curie temperature of approximately 20°K. Narath[13] has performed a nuclear magnetic resonance experiment on the magnetic Cr^{53} ion at liquid helium temperatures and reports anomalous intensity enhancement and large linewidths of approximately 100 kc/sec. This is similar to the results found in ferromagnetic cobalt by Portis and Gossard[14] who attributed them to the nuclei within the Bloch wall. However, $CrCl_3$ is composed of hexagonal ferromagnetic layers and the antiferromagnetic coupling is weak. Thus, it is not consistent with our model used above. Our results for $1/T_1$ range between 10^2T and $7\times10^{-4}T$ depending on the values of the wall viscosity and stiffness.

Finally, we consider the substance $Co^{59}F_2$ which has a Curie temperature of 50°K and a magnetic crystal structure similar to that of MnF_2 and NiF_2 discussed above. The nuclear magnetic resonance of the magnetic Co^{59} ion was performed by Jaccarino[15] who obtained very large resonance frequencies (165–195 Mc/sec) at 1.3°K as well as large Gaussian linewidths (5.8×10^5 to 9.4×10^5 cps). An indirect coupling of nuclear spins is indicated and will be discussed here also in Sec. IV. Calculations for $1/T_1$ give the range $0.5T$ to 10^5T due to the uncertainty in the wall parameters.

We now estimate the effect of the remaining terms $S_X{}^i-\langle S_X{}^i\rangle$ and $s_X{}^j-\langle s_X{}^j\rangle$ in Eq. (2). As pointed out by Winter,[3] these terms give a contribution to T_2 only—the relaxation being produced by a Raman scattering of the magnons. Expanding these terms, our additional Hamiltonian H' is

$$\mathcal{H}'=A(2S)^{-1}\{\textstyle\sum_i I_X{}^i[(S_Y{}^i)^2+(S_z{}^i)^2]+\sum_j I_X{}^j[(s_Y{}^j)^2+(s_z{}^j)^2]\},\quad(16)$$

and

$$1/T_2=2\pi\hbar^{-1}\int_{Ei}\int_{Ef}(\mathcal{H}')_p{}^2 n(E_i)[n(E_f)+1]\times\rho(E_i)\rho(E_f)\delta(E_i-E_f)dE_idE_f.\quad(17)$$

Using Eq. (3), the spectral density functions of Eq. (8), and the fact that J, the exchange constant is greater than K', the stiffness parameter, we may write

$$\frac{1}{T_2}=\frac{\pi A^2I(I+1)}{2\hbar S^2}\int_{\Delta'}^{\infty}(\sigma_Y{}^2)_\omega{}^2\rho^2(E')dE'\quad(18)$$

or

$$\frac{1}{T_2}=\frac{\omega_0{}^2I(I+1)}{32\pi\hbar S^4}\left(\frac{K_1}{-J}\right)\left(\frac{k_bT}{-J}\right)^2\frac{\sin^4\theta}{\Delta'}.\quad(19)$$

This term is many times smaller than $1/T_1$ and may be neglected.

IV. INDIRECT COUPLING OF NUCLEAR SPINS

In addition to the spin-lattice relaxation mechanism calculated in Sec. III, there is a linewidth produced by the indirect coupling of the nuclear spins through the virtual excitation of a spin wave. We shall calculate this effect, first proposed by Suhl,[16] for the bound wall excitations and shall obtain the second moment of the linewidth.

Substituting Eqs. (3) into the results of DP I, we may write for our bound spin-wave Hamiltonian

$$\mathcal{H}=\textstyle\sum_p[(Ja^2k^2-12J+M)(\sigma_z{}^p)^2+(K'-Ja^2k^2)(\sigma_Y{}^p)^2],\quad(20)$$

where the index p refers to all lattice sites. Following Winter,[3] we quantize the wall magnons as

$$\sigma_Y{}^p=a[(S/2S_0)(K_1/-J)^{\frac{1}{2}}]^{\frac{1}{2}}\sin\theta_p\times\textstyle\sum_k[a_k{}^\dagger e^{i\mathbf{k}\cdot\mathbf{r}_p}+a_ke^{-i\mathbf{k}\cdot\mathbf{r}_p}],$$
$$\sigma_z{}^p=ia[(S/2S_0)(K_1/-J)^{\frac{1}{2}}]^{\frac{1}{2}}\sin\theta_p\times\textstyle\sum_k[a_k{}^\dagger e^{i\mathbf{k}\cdot\mathbf{r}_p}-a_ke^{-i\mathbf{k}\cdot\mathbf{r}_p}].\quad(21)$$

Substituting Eqs. (21) and (22) into Eq. (20), we find that we can diagonalize the Hamiltonian by the canonical transformation[17]

$$a_k=\mu_k\alpha_k-\nu_k\alpha_{-k}{}^\dagger,$$

where

$$\mu_k{}^2-\nu_k{}^2=1,\qquad \mu_k{}^2+\nu_k{}^2=(K'S+MS-12JS)/E'.\quad(22)$$

Thus, we may write for the transverse part of the hyperfine coupling of Eq. (1),

$$\mathcal{H}=A[(Sa^2/2S_0)(K_1/-J)^{\frac{1}{2}}]^{\frac{1}{2}}\textstyle\sum_{p,k}\sin\theta_p\times[(\mu_k\alpha_k{}^\dagger-\nu_k\alpha_{-k})I_p{}^+e^{i\mathbf{k}\cdot\mathbf{r}_p}+(\mu_k\alpha_k-\nu_k\alpha_{-k}{}^\dagger)I_p{}^-e^{-i\mathbf{k}\cdot\mathbf{r}_p}].\quad(23)$$

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[12] T. Nagamiya, K. Yosida, and R. Kuba in *Advances in Physics*, edited by N. F. Mott (Taylor and Francis, Ltd., London, 1955), Vol. 4, p. 1.
[13] A. Narath, Phys. Rev. Letters **7**, 410 (1961).
[14] A. M. Portis and A. C. Gossard, J. Appl. Phys. **31**, 205S (1960).
[15] V. Jaccarino, Phys. Rev. Letters **2**, 163 (1959).
[16] H. Suhl, J. phys. radium **20**, 333 (1959).
[17] T. Holstein and H. Primakoff, Phys. Rev. **58**, 1098 (1940).

The second order perturbation of Eq. (23),

$$\Delta E=-\sum_k|\langle 0|\mathcal{H}|k\rangle|^2/E', \tag{24}$$

yields (keeping only the secular part)

$$\Delta E=\tfrac{1}{2}\sum_{p,q}B_{pq}[I_p^+I_q^-+I_p^-I_q^+], \tag{25}$$

where

$$B_{pq}=\sum_k A^2Sa^2(2S_0)^{-1}(K_1/-J)^{\frac{1}{2}}\sin\theta_p\sin\theta_q \times[(\nu_k^2+\mu_k^2)/E']\exp[i\mathbf{k}\cdot(\mathbf{r}_p-\mathbf{r}_q)]. \tag{26}$$

In Eq. (26) we have used the fact that $(\mu_k^2+\nu_k^2)\gg 1$.

As Nakamura[18] points out, a nuclear spin belonging to one of the sublattices is subjected to a local field which is different from that belonging to the other sublattice, and hence terms of the form $I_i^+I_j^-$ must oscillate with a high frequency. Accordingly, the $j(i)$th nuclear spin will not induce any important fluctuating field on the $i(j)$th nuclear spin in a Larmor cycle of the latter, yielding only subsidiary lines. Therefore, they should be excluded from the Hamiltonian in calculating the second moment of the linewidth. Our effective Hamiltonian reduces to

$$\Delta E=2\sum_{p>p'}B_{pp'}(\mathbf{I}_p\cdot\mathbf{I}_{p'}-I_p^zI_{p'}^z), \tag{27}$$

where the p'th lattice site is on the same sublattice as the pth lattice site. Then, the second moment formula of Van Vleck[19] for a nucleus within the Bloch wall is

$$\langle(\hbar\Delta\omega)^2\rangle_{\text{av}}=(4/3)I(I+1)\sum_{p'}B_{pp'}^2. \tag{28}$$

If we replace the summation over k by an integration, the term $\sum_{p'}B_{pp'}^2$ can be evaluated. We obtain

$$[\langle(\hbar\Delta\omega)^2\rangle_{\text{av}}]^{\frac{1}{2}}=\frac{A^2\sin\theta_p}{16(-\pi JK')^{\frac{1}{2}}}\left(\frac{K_1}{-J}\right)^{\frac{1}{4}}\left[\frac{I(I+1)}{3}\right]^{\frac{1}{2}} \times[1+K'/(-12J+M)] \tag{29}$$

for the second moment of the NMR of the magnetic ions in an antiferromagnet. This is larger by the square root of the ratio of the anisotropy to the wall stiffness parameters, i.e., $(K_1/K')^{\frac{1}{2}}$, compared to the corresponding formula for a uniform antiferromagnet obtained by Nakamura.[18]

Calculations of these linewidths and their application to the substances discussed in Sec. III have been done for the uniform antiferromagnet by the various authors mentioned in our references and have been shown to be within the range of applicability. We merely point out that the domain walls may even further enhance the linewidths due to this mechanism.

V. LINEWIDTH AND MAGNETIZATION VARIATION

There is still another mechanism for nuclear linewidth resulting from the spin-wave excitation spectrum within the Bloch wall. The quantity, $(\sigma-\sigma_X)/\sigma$ (which is the deviation of the longitudinal component of the electron spin), varies along the wall in the z direction causing a nonuniformity in the magnitude of the magnetization. This is due to the corresponding variation of σ_Y and σ_z from the center to the edge of the wall.

At a given temperature, the average value of $\Delta\sigma_X/\sigma$ for the bound excitations is

$$\Delta\sigma_X/\sigma=\frac{1}{2\sigma^2}\int_{\Delta'}^{\infty}[(\sigma_{Y^2})_\omega+(\sigma_z^2)_\omega]\rho(E)dE. \tag{30}$$

Substituting Eqs. (8) and (9) into Eq. (30), we get

$$\frac{\Delta\sigma_X}{\sigma}=\frac{\sin^2\theta_p}{-8\pi J\sigma^2}\left(\frac{K_1}{-J}\right)^{\frac{1}{2}}\int_{\Delta'}^{\infty}n(E)\left[1+\frac{K'-Ja^2k^2}{-12J+M}\right]dE. \tag{31}$$

If we replace the high-temperature approximation for $n(E)$ by the more accurate Bose relationship, $[\exp(E/k_bT)-1]^{-1}$, then we note that the main contribution to the integral is from long wavelengths only, and we can discard the term $-Ja^2k^2$. Our final result is

$$\frac{\Delta\sigma_X}{\sigma}=\frac{\sin^2\theta_p}{-8\pi JS^2}\left(\frac{K_1}{-J}\right)^{\frac{1}{2}}\left[1+\frac{K'}{-12J+M}\right]k_bT\ln\frac{k_bT}{\Delta'}. \tag{32}$$

This result is similar to that obtained in the ferromagnetic case by Winter,[3] the effect varying from a maximum of $\theta_i=\pi/2$ at the center of the wall to zero outside the wall.

For the free spin-wave branch, we have, from DP I, the formulas

$$E'^2=(-48JS^2+4MS^2)(-Ja^2k^2+K_1+K'), \tag{33}$$

and

$$\begin{aligned}\lim_{\Gamma\to 0}(\sigma_{Y}^2)_\omega&=\frac{\tanh^2(zh)+(k_z/h)^2}{-Ja^2k^2+K_1+K'}\,\frac{a^3k_bT}{1+(k_z/h)^2}\,\frac{\delta(\omega-E)}{V_0},\\ \lim_{\Gamma\to 0}(\sigma_z^2)_\omega&=\frac{\tanh^2(zh)+(k_z/h)^2}{-12J+M}\,\frac{a^3k_bT}{1+(k_z/h)^2}\,\frac{\delta(\omega-E)}{V_0},\end{aligned} \tag{34}$$

analogous to Eqs. (7) and (8) for the bound spin waves. Here, however, a^2k^2 represents $a_x^2k_x^2+a_y^2k_y^2+a_z^2k_z^2$ while a^3 equals a^2a_z. The difference between $\Delta\sigma_X/\sigma$ at the center and at the edge of the wall for the free spin-wave branch is

$$\left(\frac{\Delta\sigma}{\sigma}\right)_{\text{ctr}}-\left(\frac{\Delta\sigma}{\sigma}\right)_{\text{edge}}=-\frac{a^3}{V_0}\sum_k\frac{n(E)}{E'}\delta(\omega_0-E) \times\frac{-24J+2(M+K'+K_1)-2Ja^2k^2}{1+(k_z/h)^2}. \tag{35}$$

This summation is similar to the one obtained by

[18] T. Nakamura, Progr. Theoret. Phys. (Kyoto) **20**, 542 (1958).
[19] J. H. Van Vleck, Phys. Rev. **74**, 1168 (1948).

Winter[3] for the ferromagnetic case. The result is

$$\left(\frac{\Delta\sigma}{\sigma}\right)_{\text{ctr}}-\left(\frac{\Delta\sigma}{\sigma}\right)_{\text{edge}}=-\frac{[K_1/(-J)]^{\frac{1}{2}}}{-16\pi JS^2}\times\left(1+\frac{K_1+K'}{-12J+M}\right)k_bT\ln\left(\frac{k_bT}{\Delta}\right), \quad (36)$$

where

$$\Delta=(4MS^2-48JS^2)^{\frac{1}{2}}(K_1+K')^{\frac{1}{2}}. \quad (37)$$

Finally, adding Eqs. (32) and (36), the maximum relative variation of the resonance frequency is

$$\frac{\Delta\nu}{\nu}=\frac{[K_1/(-J)]^{\frac{1}{2}}}{-16\pi JS^2}k_bT\ln\left(\frac{k_bT}{\Delta'}\right). \quad (38)$$

This linewidth mechanism is logarithmically dependent on the value of the wall stiffness parameter K'. For $CuCl_2\cdot 2H_2O$, the maximum $\Delta\nu$ at the center of the wall varies from 5×10^3 cps (for small K') to 10^5 cps (for large K') at liquid helium temperatures, and is thus of the magnitude of experimental linewidths.[8] The substances, MnF_2 and NiF_2 both have linewidths which vary from 10^4 to 10^5 cps at $T=4$°K while the maximum value of CoF_2 at the center of the wall goes from 10^6 to 10^7 cps at 4°K, and of course drops for lower values of the temperature. Finally, the magnitude of the frequency variation of $CrCl_3$ due to the above mechanism is of the order of 10^5 cps at T equal 4°K and is thus comparable with the observed linewidths of Narath.[13] In all these calculations, however, we are limited by the uncertainty in the wall stiffness parameter. Further, our values are maximum values reached only by those nuclei at the very center of the wall where θ equals $\pi/2$.

VI. SECOND SET OF SOLUTIONS

In addition to the solutions given in Eq. (3), a second set of solutions for the bound and free spin-wave excitations were obtained in DP I. They are given by (for the bound excitations),

$$S_Y=-s_Y=D\sin\theta\exp[i(k_xx+k_yy)]=\gamma_Y,$$
$$S_z=-s_z=E\sin\theta\exp[i(k_xx+k_yy)]=\gamma_z. \quad (39)$$

Note that γ as used here equals $(1/2)\gamma$ of DP I. The spectral densities are obtained from Eqs. (12) of DP I, i.e.,

$$(\gamma_Y^2)_\omega=\frac{a^2}{\pi S_0}\left(\frac{K_1}{-J}\right)^{\frac{1}{2}}\frac{k_bT\sin^2\theta}{-12J+K'}\times\frac{\Gamma_2(E_2'^2+\Gamma_1\Gamma_2)+\omega^2\Gamma_1}{(\omega^2-E_2'^2-\Gamma_1\Gamma_2)^2+\omega^2(\Gamma_1+\Gamma_2)^2}, \quad (40)$$

$$(\gamma_z^2)_\omega=\frac{a^2}{\pi S_0}\left(\frac{K_1}{-J}\right)^{\frac{1}{2}}\frac{k_bT\sin^2\theta}{K_2-K_1+M-Ja^2k^2}\times\frac{\Gamma_1(E_2'^2+\Gamma_1\Gamma_2)+\omega^2\Gamma_2}{(\omega^2-E_2'^2-\Gamma_1\Gamma_2)^2+\omega^2(\Gamma_1+\Gamma_2)^2},$$

where now the energy of the bound spin-wave excitation spectrum with zero viscosity is

$$E_2'=(-48JS+4K'S^2)^{\frac{1}{2}}(K_2-K_1+M-Ja^2k^2)^{\frac{1}{2}}. \quad (41)$$

The quantity K_2 is the anisotropy parameter in the xz plane. The minimum excitation energy Δ_2' is given by placing k^2 equal to zero in this equation. Thus, for large J, we must substitute the quantity K_2-K_1+M instead of K' in the analogous Eq. (10) for the first set of solutions. Inasmuch as one would expect the sum of the anisotropy terms K_2-K_1 and inertia term M to be larger than the stiffness term K', the minimum energy of the wall excitation remains large compared to the ferromagnetic case. The expressions given in Eqs. (13) and (15) of Sec. III remain the same for the second set of solutions except for the substitution of Δ_2' for Δ'.

The indirect coupling of nuclear spins by a virtual electron spin wave discussed in Sec. IV is similarly modified for this second set of solutions. Our new bound spin-wave Hamiltonian, analogous to Eq. (20) is got from DP I and is

$$\mathcal{H}=\sum_p[(-12J+K'+Ja^2k^2)(\gamma_Y{}^p)^2+(K_2-K_1+M-Ja^2k^2)(\gamma_z{}^p)^2]. \quad (42)$$

Following the procedures used in Sec. IV, our result for the linewidth is the same as in Eq. (29) for large J, except that again we must substitute the quantity K_2-K_1+M for K'. Thus, for this second set of solutions, our linewidth is now smaller by the factor $(K_1/K_2-K_1+M)^{\frac{1}{2}}$ compared to the corresponding formula for a uniform antiferromagnet and thus physically less interesting.

Finally, the results for the linewidth caused by the relative variation of the magnetization along the Bloch wall discussed in Sec. V are, for large J, also similar to that given by Eq. (38) for the first set of solutions. Here again, one should substitute Δ_2' for Δ' but, inasmuch as this quantity appears in the logarithm, the difference between these results is not so marked.

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