Exchange Enhancement of Nuclear Spin-Lattice Relaxation in Antiferromagnets*

A. BROOKS HARRIS†
Department of Physics, University of Pennsylvania, Philadelphia, Pennsylvania 19104
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The nuclear spin-lattice relaxation rate in an antiferromagnetic insulator at low temperature is calculated to lowest order in $1/z$. The effects of anisotropy are included but correctly vanish for spin $\frac{1}{2}$. Instead of a divergence in the limit of zero anisotropy as found by Beeman and Pincus, we obtain results similar to those for a ferromagnet.

I. INTRODUCTION

RECENTLY, Pincus[1] and Beeman and Pincus[2] have discussed the enhancement in the nuclear magnetic relaxation rate, T_1^{-1}, in magnetic insulators at low temperatures due to the interactions between spin waves. These calculations are of particular interest to the theory of magnetism because the relaxation rate is proportional[3,4] to the density of spin-wave excitations at essentially zero frequency. In I, we showed that their calculations for a ferromagnet, ostensibly valid to lowest order in $1/S$, are in fact valid to all orders in $1/S$ to lowest order in kT/H_E, where H_E is the exchange energy, $2JzS$. Some differences between the two methods of calculation are of note. Pincus,[1] and Beeman and Pincus[2] used the Holstein-Primakoff[5] representation, whereas in I the Dyson-Maleev[6] boson representation for spin operators was employed. In the Holstein-Primakoff formalism the first-order exchange enhanced single-magnon[7] process dominates the three-magnon process. Thus one is led to ask whether repeated scatterings of spin waves give further large enhancement to T_1^{-1}. However, the treatment of repeated scattering within this formalism is rather cumbersome algebraically. Using the Dyson-Maleev transformation we were able to express T_1^{-1} in terms of the zero-temperature t matrix which describes the effect of repeated scatterings and which takes an especially simple form when the Dyson-Maleev transformation is used. From this study we concluded that the first-Born-approximation result of Refs. 1 and 2 for ferromagnets was exact to lowest order in kT/H_E. As a result, one is tempted to predict that a calculation of T_1^{-1} in an antiferromagnet based on the first Born approximation must also give qualitatively correct results.

In Ref. 2, there appears, however, a striking difference between the ferro- and antiferromagnetic relaxation rates. Whereas in the former case, the enhancement (in the Holstein-Primakoff formalism) due to spin-wave interactions is large but finite, it was found to be proportional to $\ln H_A$ for the antiferromagnet, where H_A is the anisotropy energy. This result, if true, would have very fundamental implications for the usual picture of the antiferromagnetic state, because it would imply a divergence in the density of spin-wave excitations at low energy in the limit of vanishing anisotropy. However, no treatment of the thermodynamics of the antiferromagnet[8,9] has shown any such anomaly.

Accordingly, we have reexamined the calculations of T_1^{-1} for the antiferromagnet, and find that the $1/z$ expansion proposed previously[9] can be used successfully here. Our calculation has the property, noted previously, that for spin $\frac{1}{2}$ there are no effects from the anisotropy term in conformity with the requirements of spin kinematics. Furthermore, corrections from repeated scattering of spin waves are expected to be smaller by a factor $1/z$, so that our results are probably qualitatively reliable. Upon examination of the calculations of Beeman and Pincus, we find that the spurious divergence in T_1^{-1} is the result of an algebraic error in the transformation of the Holstein-Primakoff Hamiltonian to normal coordinates. This point is discussed more fully in the Appendix.

II. CALCULATION

We will carry out the calculations using the Dyson-Maleev transformation

$$S_z = S - a^\dagger a, \tag{1a}$$

$$S_+ = (2S)^{1/2} a - (2S)^{-1/2} a^\dagger a a, \tag{1b}$$

$$S_- = (2S)^{1/2} a^\dagger \tag{1c}$$

for spins on the a or "up" sublattice and

$$S_z = -S + b^\dagger b, \tag{1d}$$

$$S_+ = (2S)^{1/2} b^\dagger - (2S)^{-1/2} b^\dagger b^\dagger b, \tag{1e}$$

$$S_- = (2S)^{1/2} b \tag{1f}$$

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† Alfred P. Sloan Postdoctoral Fellow.

[1] P. Pincus, Phys. Rev. Letters 16, 398 (1966).
[2] D. Beeman and P. Pincus, Phys. Rev. 166, 359 (1968).
[3] A. Abragam, *The Principles of Nuclear Magnetism* (Oxford University Press, Oxford, 1961), p. 310.
[4] A. B. Harris, J. Phys. C. 2, 463 (1969). We will refer to this paper as I.
[5] T. Holstein and H. Primakoff, Phys. Rev. 58, 1098 (1940).
[6] F. J. Dyson, Phys. Rev. 102, 1217 (1956); 102, 1230 (1956); S. V. Maleev, Zh. Eksperim. i Teor. Fiz. 33, 1010 (1957) [English transl.: Soviet Phys.—JETP 6, 776 (1958)].
[7] By a single-magnon process we mean the contribution to the relaxation rate from the correlation function $\langle a_R(t) a_R^\dagger(0)\rangle$, see Eq. (16b) of I. The single-magnon relaxation rate, therefore, depends on what transformation to bosons is used.
[8] T. Oguchi, Phys. Rev. 117, 117 (1960).
[9] A. B. Harris, Phys. Rev. Letters 21, 602 (1968).

for spins on the b or "down" sublattice. We treat the case of a bcc antiferromagnet with nearest-neighbor exchange coupling and uniaxial anisotropy governed by the Hamiltonian

$$H=2J\sum_{\langle ij\rangle}\mathbf{S}_i\cdot\mathbf{S}_j-D\sum_i(S_{iz})^2. \quad (2)$$

Using Eq. (1) we write this in terms of boson operators as

$$H=H_0+V+E_0, \quad (3)$$

with

$$E_0=-2JzS^2N-2NDS^2, \quad (4a)$$

$$H_0=(H_E+H_A)\sum_k\{a_k{}^\dagger a_k+b_k{}^\dagger b_k+\xi\gamma_k a_k{}^\dagger b_{-k}{}^\dagger+\xi\gamma_k a_k b_{-k}\}, \quad (4b)$$

$$V=-N^{-1}\sum_{k,1234}\delta_k(1+2-3-4)\times\{H_E/2S(2a_1{}^\dagger b_{-2}a_3b_{-4}{}^\dagger\gamma_{2-4}+\gamma_2a_1{}^\dagger b_{-2}a_3a_4+\gamma_{3+4-2}a_1{}^\dagger b_{-2}b_{-3}{}^\dagger b_{-4}{}^\dagger)+D(a_1{}^\dagger a_2{}^\dagger a_3a_4+b_1{}^\dagger b_2{}^\dagger b_3b_4)\} \quad (4c)$$

in the usual notation[8] with $H_A=D(2S-1)$ and $\xi=H_E/(H_E+H_A)$. In Eq. (4c), the Kronecker delta conserves momentum up to a reciprocal-lattice vector. We will carry the calculations to lowest order in $1/z$. In this connection we recall that the expansion in powers of $1/z$ is obtained by expanding all momentum-dependent quantities which do not appear as factors of occupation numbers n_k in powers of γ_k.[9] Here,

$$n_k\equiv n(\epsilon_k)=\{\exp[-\beta(H_E+H_A)\epsilon_k]-1\}^{-1}, \quad (5a)$$

$$\epsilon_k=(1-\xi^2\gamma_k{}^2)^{1/2}. \quad (5b)$$

We will need to use the transformation to normal coordinates

$$a_k{}^\dagger=l_k\alpha_k{}^\dagger+m_k\beta_{-k}, \quad (6a)$$

$$b_{-k}=m_k\alpha_k{}^\dagger+l_k\beta_{-k}, \quad (6b)$$

where

$$l_k=[(1+\epsilon_k)/2\epsilon_k]^{1/2}, \quad (7a)$$

$$m_k=-[(1-\epsilon_k)/2\epsilon_k]^{1/2}. \quad (7b)$$

Note that

$$N^{-1}\sum_k\gamma_k{}^2=1/z, \quad (8a)$$

$$N^{-1}\sum_k m_k{}^2\sim\xi^2/4z. \quad (8b)$$

Repeated exchange scatterings introduce further factors of γ_k as can be seen from Eq. (4c). Accordingly, repeated exchange scatterings introduce further factors of $1/z$, and henceforth, will be neglected. This was already noted by Wang and Callen[10] using a more complicated spin formalism. In contrast, repeated scatterings from the interactions due to the anisotropy term in V give contributions of the same order in $1/z$. Hence, to lowest order in $1/z$, we must sum over all such repeated scatterings if we wish to obtain results which explicitly display the spin kinematics which require that for spin $\frac{1}{2}$ the results should be independent of D.

Since we are only interested in a lowest order (in $1/z$) calculation, we will not introduce the more systematic, but also more esoteric, apparatus of many-body perturbation theory, but will follow the method of calculation used by Beeman and Pincus.[2] We wish to calculate the matrix element $\langle i|V_{SL}|f\rangle\langle f|V_{SL}|i\rangle$ between suitable initial and final states. Here V_{SL} is the interaction responsible for spin-lattice relaxation which we take to be

$$V_{SL}=A\mathbf{I}_i\cdot\mathbf{S}_i. \quad (9)$$

In particular, we are not interested in Raman processes involving the term $AI_{iz}S_{iz}$ which we omit henceforth. We may write V_{SL} in terms of bosons as

$$V_{SL}=A(\tfrac{1}{2}S)^{1/2}I_i{}^+[a_i-(2S)^{-1}a_i{}^\dagger a_ia_i]+A(\tfrac{1}{2}S)^{1/2}I_i{}^-a_i{}^\dagger \quad (10a)$$

$$\equiv V_{SL}{}^++V_{SL}{}^-. \quad (10b)$$

In terms of normal coordinates $V_{SL}{}^\pm$ becomes

$$V_{SL}{}^+=(S/2N)^{1/2}AI_i{}^+\{\sum_1 l_1(\alpha_1-x_1\beta_{-1}{}^\dagger)-(2NS)^{-1}\sum_{123}l_1l_2l_3(\alpha_1{}^\dagger-x_1\beta_{-1})\times(\alpha_2-x_2\beta_{-2}{}^\dagger)(\alpha_3-x_3\beta_{-3}{}^\dagger)\}, \quad (11a)$$

$$V_{SL}{}^-=(S/2N)^{1/2}AI_i{}^-\sum_1 l_1(\alpha_1{}^\dagger-x_1\beta_{-1}), \quad (11b)$$

where $x_i=-m_i/l_i=[(1-\epsilon_i)/(1+\epsilon_i)]^{1/2}$. Note that to lowest order in $1/z$ we may rearrange all creation operators to the left of the destruction operators in Eq. (11a) because of Eq. (8b). We will use this type of reasoning later without further comment. Following Ref. 2 we construct the effective interactions $V_{SL,eff}{}^\pm$ as

$$V_{SL,eff}{}^+=-(S/2N)^{1/2}A(H_A+H_E)^{-1}I_i{}^+\sum l_1\epsilon_1{}^{-1}\times[\alpha_1+x_1\beta_{-1}{}^\dagger,V]-(8N^3S)^{-1/2}AI_i{}^+\times\sum l_1l_2l_3(\alpha_1{}^\dagger-x_1\beta_{-1})(\alpha_2-x_2\beta_{-2}{}^\dagger)\times(\alpha_3-x_3\beta_{-3}{}^\dagger), \quad (12a)$$

$$V_{SL,eff}{}^-=-(S/2N)^{1/2}A(H_A+H_E)^{-1}I_i{}^-\times\sum_1 l_1\epsilon_1{}^{-1}[V,(\alpha_1{}^\dagger+x_1\beta_{-1})]. \quad (12b)$$

This procedure amounts to a first-Born-approximation calculation of the cross section for spin-wave interactions.

[10] Y. L. Wang and H. B. Callen, Phys. Rev. 148, 433 (1966).

We now need to express V in terms of normal coordinates. From Eqs. (4c) and (6), we find

$$V=-(H_E/4NS)\sum_{1234}\delta(1+2-3-4)l_1l_2l_3l_4$$
$$\times\{\alpha_1{}^\dagger\alpha_2{}^\dagger\alpha_3\alpha_4\Phi_{1234}{}^{(1)}+2\alpha_1{}^\dagger\beta_{-2}\alpha_3\alpha_4\Phi_{1234}{}^{(2)}$$
$$+2\alpha_1{}^\dagger\alpha_2{}^\dagger\alpha_3\beta_{-4}{}^\dagger\Phi_{1234}{}^{(3)}+4\alpha_1{}^\dagger\beta_{-2}\alpha_3\beta_{-4}{}^\dagger\Phi_{1234}{}^{(4)}$$
$$+2\beta_{-1}\beta_{-2}\alpha_3\beta_{-4}{}^\dagger\Phi_{1234}{}^{(5)}+2\alpha_1{}^\dagger\beta_{-2}\beta_{-3}{}^\dagger\beta_{-4}{}^\dagger\Phi_{1234}{}^{(6)}$$
$$+\alpha_1{}^\dagger\alpha_2{}^\dagger\beta_{-3}{}^\dagger\beta_{-4}{}^\dagger\Phi_{1234}{}^{(7)}+\beta_{-1}\beta_{-2}\alpha_3\alpha_4\Phi_{1234}{}^{(8)}$$
$$+\beta_{-1}\beta_{-2}\beta_{-3}{}^\dagger\beta_{-4}{}^\dagger\Phi_{1234}{}^{(9)}\}\,, \quad (13)$$

where

$$\Phi_{1234}{}^{(1)}=\Phi_{1234}{}^{(9)}=[\gamma_{1-4}x_1x_4+\gamma_{1-3}x_1x_3+\gamma_{2-4}x_2x_4+\gamma_{2-3}x_2x_3-\gamma_1x_2x_3x_4-\gamma_2x_1x_3x_4-\gamma_2x_2-\gamma_1x_1+2q+2qx_1x_2x_3x_4]\,, \quad (14a)$$

$$\Phi_{1234}{}^{(2)}=\Phi_{2134}{}^{(6)}=[-\gamma_{2-4}x_4-\gamma_{2-3}x_3-\gamma_{1-4}x_1x_2x_4-\gamma_{1-3}x_1x_2x_3+\gamma_1x_3x_4+\gamma_2x_1x_2x_3x_4+\gamma_2+\gamma_1x_1x_2-2qx_2-2qx_1x_3x_4]\,, \quad (14b)$$

$$\Phi_{1234}{}^{(3)}=\Phi_{1243}{}^{(5)}=[-\gamma_{2-4}x_2-\gamma_{1-4}x_1-\gamma_{2-4}x_1x_3x_4-\gamma_{2-3}x_2x_3x_4+\gamma_1x_2x_3+\gamma_2x_1x_3+\gamma_2x_2x_4+\gamma_1x_1x_4-2qx_4-2qx_1x_2x_3]\,, \quad (14c)$$

$$\Phi_{1234}{}^{(4)}=[\gamma_{2-4}+\gamma_{1-4}x_1x_2+\gamma_{1-4}x_3x_4+\gamma_{1-3}x_1x_2x_3x_4-\gamma_1x_3-\gamma_2x_1x_2x_3-\gamma_2x_4-\gamma_1x_1x_2x_4+2qx_1x_3+2qx_2x_4]\,, \quad (14d)$$

$$\Phi_{1234}{}^{(7)}=\Phi_{1234}{}^{(8)}=[\gamma_{2-4}x_2x_3+\gamma_{2-3}x_2x_4+\gamma_{2-3}x_1x_3+\gamma_{2-4}x_1x_4-\gamma_1x_1x_3x_4-\gamma_2x_2x_3x_4-\gamma_1x_2-\gamma_2x_1+2qx_3x_4+2qx_1x_2]\,, \quad (14e)$$

with

$$q=2DS/H_E\,. \quad (15)$$

We have neglected umklapp processes because the relaxation rate depends on processes for which all momenta are small.

Using Eqs. (12) and (13), we find

$$V_{\mathrm{SL,eff}}{}^+=(8N^3S)^{-1/2}AI_i{}^+\sum_{123}l_1l_2l_3\epsilon_4{}^{-2}[\Psi_{123}{}^{+(1)}\alpha_1\alpha_2\alpha_3{}^\dagger+\Psi_{123}{}^{+(2)}\beta_{-1}{}^\dagger\beta_{-2}{}^\dagger\beta_{-3}+2\Psi_{123}{}^{+(3)}\alpha_1\beta_{-2}{}^\dagger\alpha_3{}^\dagger+2\Psi_{123}{}^{+(4)}\beta_{-1}{}^\dagger\alpha_2\beta_{-3}]\,, \quad (16a)$$

$$V_{\mathrm{SL,eff}}{}^-=(8N^3S)^{-1/2}AI_i{}^-\sum_{123}l_1l_2l_3\epsilon_4{}^{-2}[\Psi_{123}{}^{-(1)}\alpha_1{}^\dagger\alpha_2{}^\dagger\alpha_3+\Psi_{123}{}^{-(2)}\beta_{-1}\beta_{-2}\beta_{-3}{}^\dagger+2\Psi_{123}{}^{-(3)}\alpha_1{}^\dagger\beta_{-2}\alpha_3+2\Psi_{123}{}^{-(4)}\beta_{-1}\alpha_2{}^\dagger\beta_{-3}{}^\dagger] \quad (16b)$$

apart from terms involving the simultaneous creation or absorption of three particles, which we shall not need. Here,

$$\Psi_{123}{}^{+(1)}=-\epsilon_4{}^2+\xi l_4{}^2\epsilon_4(\Phi_{3412}{}^{(1)}-x_4\Phi_{3412}{}^{(2)})\,, \quad (17a)$$
$$\Psi_{123}{}^{+(2)}=x_1x_2x_3\epsilon_4{}^2+\xi l_4{}^2\epsilon_4(\Phi_{4312}{}^{(6)}-x_4\Phi_{4312}{}^{(9)})\,, \quad (17b)$$
$$\Psi_{123}{}^{+(3)}=x_2\epsilon_4{}^2+\xi l_4{}^2\epsilon_4(\Phi_{3412}{}^{(3)}-x_4\Phi_{3412}{}^{(4)})\,, \quad (17c)$$
$$\Psi_{123}{}^{+(4)}=-x_1x_3\epsilon_4{}^2+\xi l_4{}^2\epsilon_4(\Phi_{4321}{}^{(4)}-x_4\Phi_{4321}{}^{(5)})\,, \quad (17d)$$
$$\Psi_{123}{}^{-(1)}=\xi l_4{}^2\epsilon_4(\Phi_{1234}{}^{(1)}-x_4\Phi_{1234}{}^{(3)})\,, \quad (17e)$$
$$\Psi_{123}{}^{-(2)}=\xi l_4{}^2\epsilon_4(\Phi_{1243}{}^{(5)}-x_4\Phi_{1243}{}^{(9)})\,, \quad (17f)$$
$$\Psi_{123}{}^{-(3)}=\xi l_4{}^2\epsilon_4(\Phi_{1234}{}^{(2)}-x_4\Phi_{1234}{}^{(4)})\,, \quad (17g)$$
$$\Psi_{123}{}^{-(4)}=\xi l_4{}^2\epsilon_4(\Phi_{2143}{}^{(4)}-x_4\Phi_{2143}{}^{(6)})\,, \quad (17h)$$

with $\mathbf{k}_4=\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3$. Then the relaxation rate is given by

$$T_1{}^{-1}=\frac{8\pi}{\hbar}\frac{A^2}{8N^3S}\sum_{123}n_1n_2(1+n_3)\delta(E_1+E_2-E_3)\times l_1{}^2l_2{}^2l_3{}^2\epsilon_4{}^{-4}\{\Psi_{123}{}^{+(1)}\Psi_{123}{}^{-(1)}+\Psi_{123}{}^{+(2)}\Psi_{123}{}^{-(2)}\}$$
$$+\frac{16\pi}{\hbar}\frac{A^2}{8N^3S}\sum_{123}(1+n_1)n_2n_3\delta(E_1-E_2-E_3)\times l_1{}^2l_2{}^2l_3{}^2\epsilon_4{}^{-4}\{\Psi_{123}{}^{+(3)}\Psi_{123}{}^{-(3)}+\Psi_{123}{}^{+(4)}\Psi_{123}{}^{-(4)}\}\,, \quad (18)$$

where $E_i=(H_E+H_A)\epsilon_i$. When the condition $E_3=E_1+E_2$ is fulfilled we find that[11]

$$\Psi_{123}{}^{\pm(1)}=\xi^2\gamma_{1-4}x_1\gamma_4+\xi^2\gamma_{2-4}x_2\gamma_4+\xi\gamma_{1-3}x_1x_3+\xi\gamma_{2-3}x_2x_3+\gamma_4\xi^2\rho^+[2qx_1x_2x_3-\gamma_1x_2x_3-\gamma_2x_1x_3]+\xi\rho^-[2q-x_2\gamma_2-x_1\gamma_1]\,, \quad (19a)$$

$$-\Psi_{123}{}^{\pm(2)}=\xi\gamma_{2-4}x_2+\xi\gamma_{1-4}x_1+\xi^2\gamma_{2-4}x_1x_3\gamma_4+\xi^2\gamma_{1-4}x_2x_3\gamma_4+\xi\rho^+[2qx_1x_2x_3-\gamma_1x_2x_3-\gamma_2x_1x_3]+\xi^2\gamma_4\rho^-[2q-\gamma_2x_2-\gamma_1x_1]\,. \quad (19b)$$

When the condition $E_1=E_2+E_3$ is fulfilled we find that

$$-\Psi_{123}{}^{\pm(3)}=\xi^2\gamma_{1-3}\gamma_4+\xi\gamma_{2-3}x_3+\xi^2\gamma_{2-3}x_1x_2\gamma_4+\xi\gamma_{1-3}x_1x_2x_3+\xi\rho^-[2qx_2-\gamma_2-\gamma_1x_1x_2]+\xi^2\gamma_4\rho^+[2qx_1x_3-\gamma_1x_3-\gamma_2x_1x_2x_3]\,, \quad (19c)$$

$$\Psi_{123}{}^{\pm(4)}=\xi\gamma_{1-3}+\xi\gamma_{2-3}x_1x_2+\xi^2\gamma_{2-3}x_3\gamma_4+\xi^2\gamma_{1-3}x_1x_2x_3\gamma_4+\xi^2\gamma_4\rho^-[2qx_2-\gamma_2-\gamma_1x_1x_2]+\xi\rho^+[2qx_1x_3-\gamma_1x_3-\gamma_2x_1x_2x_3]\,, \quad (19d)$$

where

$$\rho^\pm=[1\pm\tfrac{1}{2}\epsilon_3]/[1\pm\tfrac{1}{2}\epsilon_2-\xi q/2S]\,. \quad (20)$$

Actually, the above procedure would yield Eqs. (19) with $\rho^\pm$ everywhere replaced by unity. The presence of the factors $\rho^\pm$ is due to a renormalization as will be described in the next paragraph.

Up to now we have discussed only lowest-order perturbation-theory contributions to $V_{\mathrm{SL,eff}}{}^\pm$. In order

[11] The equality of the $\Psi_{123}{}^{+(i)}$ and $\Psi_{123}{}^{-(i)}$ is an interesting check on the correctness of our algebra.

to obtain all contributions to lowest order in $1/z$, we must include the effects of repeated scattering by the anisotropy interaction terms of V. To illustrate the analysis we will discuss a typical term arising from inserting the second term of Eq. (4c) into $[V,\alpha_k^+]$ as required by Eq. (12b). In other words, we are now discussing the term

$$-(H_E/2S)\gamma_2[a_1^+b_{-2}a_3a_4,\alpha_k^+], \tag{21}$$

which we need to evaluate when all the momenta are small. From our discussion we will see how to take account of repeated scatterings by the anisotropy through a simple renormalization procedure. We represent the term (21) by the schematic diagram of Fig. 1(a). When the transformation to normal coordinates, Eq. (6), is performed, this term becomes a sum of various terms involving α and β operators, some of which are represented in Figs. 1(b)–1(d). We will now renormalize the contribution from Fig. 1(b) by including anisotropy ladders as represented in Fig. 2(a). Similar renormalizations[12] of the terms represented in Figs. 1(c) and 1(d) are shown in Figs. 2(c) and 2(d). In doing this we must sum over the momenta of all internal lines. Consider the case where the anisotropy ladder consists of a single vertex and the internal lines have momenta k_5 and k_6. To lowest order in $1/z$, we have, from Eq. (8), that $m_5\approx m_6\approx 0$, $l_5\approx l_6\approx 1$, and $\epsilon_5\approx\epsilon_6\approx 1$. Accordingly, the contributions from Figs. 1(b) and 2(a) are, respectively,

$$-(H_E/S)\gamma_2x_2\alpha_1^\dagger\alpha_2^\dagger\alpha_3 \tag{22a}$$

and

$$-\frac{H_E}{NS}\gamma_2x_2\sum_{5,6}2D(H_E+H_A)^{-1}\times(\epsilon_5+\epsilon_6-\epsilon_3)^{-1}l_5{}^2l_6{}^2\alpha_1^\dagger\alpha_2^\dagger\alpha_3, \tag{22b}$$

which is then to lowest order in $1/z$

$$-(H_E/S)\gamma_2x_2[D(H_E+H_A)^{-1}(1-\tfrac{1}{2}\epsilon_3)^{-1}]\alpha_1^\dagger\alpha_2^\dagger\alpha_3. \tag{23}$$

Note that the expression (23) differs from (22a) by the factor in square brackets. Inclusion of more anisotropy interactions leads to further terms in a geometric series, so that repeated anisotropy scatterings lead to the renormalization of (22a) as

$$-(H_E/S)\rho^-\gamma_2x_2\alpha_1^\dagger\alpha_2^\dagger\alpha_3, \tag{24}$$

with ρ^- as given in Eq. (20). Note also that Fig. 2(b), which bears a superficial resemblance to Fig. 2(a), does not in fact contribute to lowest order in $1/z$. This is because the transformation to normal modes leads to the inclusion of a factor m_6 which, when summed over, gives a factor $1/z$. Renormalizations do not occur for all terms. For instance, the first term in Eq. (4c) is not renormalized at all because the expression analogous to Eq. (22b) would include a factor γ_5 or γ_6 which leads to an extra power of $1/z$. This discussion is actually equivalent to and motivated by the rules for calculating the self-energy $\sum_k(\omega)$ as defined in many-body perturbation theory.[13]

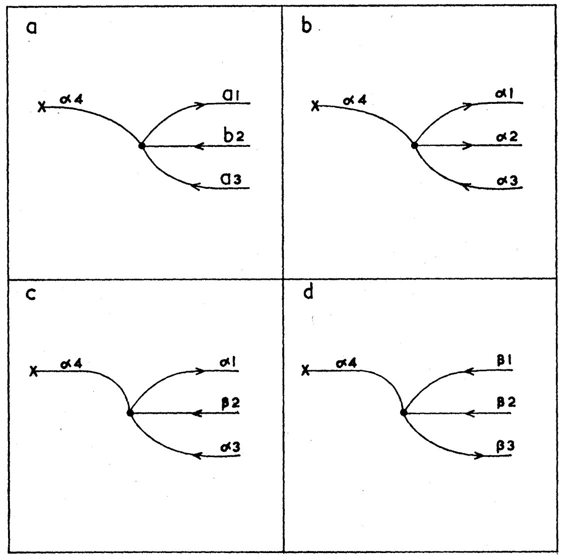

FIG. 1. Some unrenormalized vertices representing spin-wave interactions. Figures 1(b), 1(c), and 1(d) are typical terms resulting from Fig. 1(a) when normal coordinates are introduced. The lines are labeled according to the spin-wave branch, α or β, and with their momenta. Lines to the right are creation operators, those to the left are destruction operators.

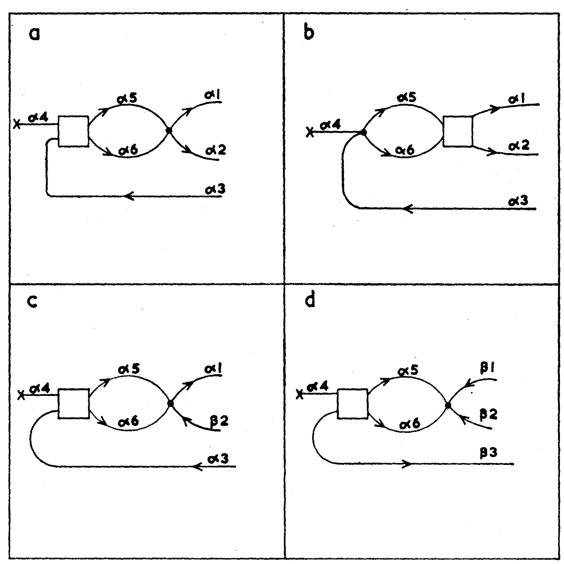

FIG. 2. Some renormalized vertices representing spin-wave interactions. The box represents repeated anistropy scatterings. Figures 2(a), 2(c), and 2(d) are renormalized versions of Figs. 1(b), 1(c), and 1(d), respectively. The labeling is as in Fig. 1.

[12] The energy denominator is obtained as the sum of energy of lines going forward (to the right) minus the energy of lines going backwards. Accordingly, Fig. 2(c) indicates a renormalization with ρ^-, whereas Fig. 2(d) is a renormalization with ρ^+.

[13] G. Baym and A. M. Sessler, Phys. Rev. **131**, 2345 (1965).

The relaxation rate can now be calculated by inserting Eq. (19) into Eq. (18). In Sec. III, we will discuss some properties and limiting cases of our calculation.

III. DISCUSSION

The expressions we have obtained for $\Psi_{123}{}^{\pm(i)}$ are interesting in that they display explicitly the property required by spin kinematics that for spin $\frac{1}{2}$ they are independent of D. We will demonstrate this for $\Psi_{123}{}^{\pm(1)}$. From the definition of ξ and Eqs. (15) and (20), one sees that

$$(2S)^{-1}\xi\rho^{\pm}q=(1\pm\tfrac{1}{2}\epsilon_3)(\rho^{\pm}-1). \tag{25}$$

This relation enables us to write $\Psi_{123}{}^{\pm(1)}$ as

$$\begin{aligned}\Psi_{123}{}^{\pm(1)}=&(2S-1)[(\rho^{-}-1)(2-\epsilon_3)+(\rho^{+}-1)\\&\times\xi x_1x_2x_3\gamma_4(2+\epsilon_3)]+\xi^2\gamma_{1-4}x_1\gamma_4\\&+\xi\gamma_{1-3}x_1x_3+\xi\gamma_{2-3}x_2x_3+\xi^2\gamma_{2-4}x_2\gamma_4\\&-(2+\epsilon_3)x_1x_2x_3\gamma_4\xi-(2-\epsilon_3).\end{aligned} \tag{26}$$

For spin $\frac{1}{2}$, $H_A=0$, so that $\xi=1$, and the expression is independent of D. To obtain this result it was necessary to include the renormalization factors $\rho^{\pm}$ which occur only if one sums all contributions of order $(1/z)^0$.

One could of course write down the rather cumbersome expression for T_1^{-1}. Since such an expression would be rather unenlightening we will content ourselves with an approximate evaluation in the two asymptotic regions:

$$T\ll T_{AE}\quad\text{(Region I)}, \tag{27a}$$

$$T\gg T_{AE}\quad\text{(Region II)}, \tag{27b}$$

where $kT_{AE}=(H_A+H_E)\epsilon_0$ with $\epsilon_0^2=1-\xi^2$, i.e., $kT_{AE}=(2H_AH_E+H_A^2)^{1/2}$ and we will assume for simplicity that $H_A\ll H_E$. In region I, we evaluate $\Psi_{123}{}^{\pm(1)}$ and $\Psi_{123}{}^{\pm(2)}$ for $k_1=k_2=0$ and $k_3^2=3k_{AE}^2$, where $k_{AE}=2\epsilon_0=2(2H_A/H_E)^{1/2}$, which are the smallest allowed values of momenta. Likewise, we evaluate $\Psi_{123}{}^{\pm(3)}$ and $\Psi_{123}{}^{\pm(4)}$ for $k_2=k_3=0$ and $k_1^2=3k_{AE}^2$. Then we find, to lowest order in H_A/H_E,

$$-\Psi_{123}{}^{\pm(2)}\approx\Psi_{123}{}^{\pm(1)}\approx-8H_A/H_E, \tag{28a}$$

$$-\Psi_{123}{}^{\pm(3)}\approx\Psi_{123}{}^{\pm(4)}\approx 4H_A/H_E. \tag{28b}$$

Note that scattering due to the anisotropy term in Eq. (4c) gives contributions which are of the same order as those due to exchange scattering. Inserting these approximations into Eq. (18), we find

$$T_1^{-1}=24(A^2/\hbar H_ES)(kT/\pi H_E)^5I_3(T_{AE}/T), \tag{29}$$

where $I_3(T_{AE}/T)$ is as defined in Ref. 2:

$$\begin{aligned}I_3(x_0)=&\int_{x_0}^{\infty}dx_1\int_{x_0}^{\infty}dx_2(x_1^2-x_0^2)^{1/2}(x_2^2-x_0^2)^{1/2}\\&\times(e^{x_1}-1)^{-1}[(x_1+x_2)^2-x_0^2]^{1/2}(e^{x_2}-1)^{-1}\\&\times(1-e^{-x_1-x_2})^{-1}.\end{aligned} \tag{30}$$

In contrast, the three-magnon[7] process gives[14]

$$T_1^{-1}=12(A^2/\hbar H_ES)(kT/\pi H_E)^5I_3(T_{AE}/T). \tag{31}$$

Also, had we neglected anisotropy scatterings, we would have found

$$-\Psi_{123}{}^{\pm(2)}\approx\Psi_{123}{}^{\pm(1)}\approx-12H_A/H_E, \tag{32a}$$

$$\Psi_{123}{}^{\pm(3)}\approx\Psi_{123}{}^{\pm(4)}\approx 0, \tag{32b}$$

which would have given T_1^{-1} larger than that of Eq. (29) by a factor $\frac{3}{2}$. Note that although our enhancement factor 2 is about the same as that found by Beeman and Pincus, our final result is smaller numerically by a factor of about 7 due to some arithmetic errors in their work.

Now let us look at region II. Here we must obtain qualitatively different results than Beeman and Pincus because our relaxation rate has no divergence for zero anisotropy. In region II, we neglect the anisotropy completely so that $q=0$, $\rho^{\pm}=1$, and $\xi=1$. Then we obtain at long wavelengths

$$\Psi_{123}{}^{\pm(1)}\approx-\Psi_{123}{}^{\pm(2)}\approx\tfrac{1}{2}\mathbf{k}_3\cdot\mathbf{k}_4, \tag{33a}$$

$$\Psi_{123}{}^{\pm(3)}\approx-\Psi_{123}{}^{\pm(4)}\approx-\tfrac{1}{2}\mathbf{k}_3\cdot\mathbf{k}_4. \tag{33b}$$

Inserting these approximations into Eq. (18), we find

$$\begin{aligned}T_1^{-1}=&\frac{8\pi A^2}{\hbar SH_E}(2\pi)^{-9}\int d\mathbf{k}_1\int d\mathbf{k}_2n_2\\&\times\int d\mathbf{k}_3\int d\mathbf{k}_4\delta(\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3-\mathbf{k}_4)\\&\times(k_1k_2k_3)^{-1}(\mathbf{k}_3\cdot\mathbf{k}_4/k_4^2)^2\\&\times\{\delta(\epsilon_1+\epsilon_2-\epsilon_3)n_1(1+n_3)\\&+2\delta(\epsilon_1-\epsilon_2-\epsilon_3)n_3(1+n_1)\}.\end{aligned} \tag{34}$$

The crucial difference between our results and those of Ref. 2 is that we have a weaker interaction which leads to a regular integrand, whereas in Ref. 2 $(\mathbf{k}_3\cdot\mathbf{k}_4/k_4^2)^2$ is replaced by a term $(\mathbf{k}_3\cdot\mathbf{k}_4-\frac{1}{2}k_1k_2)^2/k_4^4$ which then leads to a divergence in T_1^{-1}.

We perform the integration over $\mathbf{k}_1$ first. For notational convenience we replace $\mathbf{k}_2$, $\mathbf{k}_3$, and $\mathbf{k}_4$ by $-\boldsymbol{\lambda}$, $\boldsymbol{\mu}$, and $\boldsymbol{\sigma}$, respectively. Also for the angular integrations we use

$$d\Omega_\mu d\Omega_\lambda=d\varphi_\mu d\varphi_\lambda r\,dr\,ds(\lambda\mu\sigma)^{-1}, \tag{35}$$

where

$$r=|\boldsymbol{\lambda}+\boldsymbol{\mu}+\boldsymbol{\sigma}|, \tag{36a}$$

$$s=|\boldsymbol{\mu}+\boldsymbol{\sigma}|. \tag{36b}$$

The integrations over Ω_σ, φ_μ, and φ_λ are trivial. We are

[14] In going from Eq. (2.24) to (2.25) in Ref. 2, a factor of 8 was lost. The remaining factor of 6/5 which we obtain is due to differences in counting matrix elements in going from Eq. (2.23) to (2.24) of Ref. 2.

left with

$$T_1^{-1}=\frac{2A^2}{\hbar SH_E}(2\pi)^{-5}\int_0^\infty n_\lambda d\lambda\int_0^\infty d\mu\int_0^\infty d\sigma \times\int_{|\mu-\sigma|}^{\mu+\sigma}ds\int_{|\lambda-s|}^{\lambda+s}dr\sigma^{-3}(s^2-\mu^2-\sigma^2)^2 \times\{n_r(1+n_\mu)\delta(\epsilon_r+\epsilon_\lambda-\epsilon_\mu) +2n_\mu(1+n_r)\delta(\epsilon_r-\epsilon_\lambda-\epsilon_\mu)\}\,, \quad (37)$$

which we write as

$$T_1^{-1}=\frac{2A^2}{\hbar SH_E}(2\pi)^{-5}\int_0^\infty n_\lambda d\lambda\int_0^\infty d\mu \times\{(1+n_\mu)n_{\mu-\lambda}G_+(\lambda,\mu) +2n_\mu(1+n_{\mu+\lambda})G_-(\lambda,\mu)\}\,, \quad (38)$$

where

$$G_\pm(\lambda,\mu)=\frac{1}{2}\int_{-\infty}^{\infty}\sigma^{-3}d\sigma\int_{|\mu-\sigma|}^{|\mu+\sigma|}ds\,F_\pm(s)\{s^2-\mu^2-\sigma^2\}^2 \quad (39)$$

with

$$F_\pm(x)=\int_{|\lambda-x|}^{\lambda+x}dr\delta(\epsilon_r\pm\epsilon_\lambda-\epsilon_\mu)\,. \quad (40)$$

It is now rather easy to evaluate $G_\pm(\lambda,\mu)$ by two successive integrations by parts. The result is expressed in terms of derivatives of $F_\pm(x)$. In this way, we find

$$G_\pm(\lambda,\mu)=-\int_{-\infty}^{\infty}d\sigma\{(\tfrac{2}{3}\sigma^3+2\mu\sigma^2)\ln|\sigma|-(2/9)\sigma^3+2\mu\sigma^2\} \times\{\delta(\epsilon_{|\mu+\sigma|+\lambda}\pm\epsilon_\lambda-\epsilon_\mu)+\mathrm{sgn}(\lambda-|\mu+\sigma|) \times\delta(\epsilon_{\lambda-|\mu+\sigma|}\pm\epsilon_\lambda-\epsilon_\mu)\}\,, \quad (41)$$

where $\mathrm{sgn}(x)=x/|x|$ and $\epsilon_{-\lambda}=\epsilon_\lambda$. In the case of $G_+(\lambda,\mu)$ the δ functions may be satisfied near $\sigma=0$, $\sigma=-2\mu$, $\sigma=2\lambda-2\mu$, and $\sigma=-2\lambda$, whence

$$G_+(\lambda,\mu)=(8/3)\theta(\mu-\lambda) \times[2\mu\lambda(\mu-\lambda)-(\mu-\lambda)^2(\mu+2\lambda)\ln2(\mu-\lambda) -\lambda^2(3\mu-2\lambda)\ln2\lambda+\mu^3\ln2\mu]\,, \quad (42a)$$

where $\theta(x)=(x+|x|)/2x$. For $G_-(\lambda,\mu)$, the δ functions may be satisfied near $\sigma=0$, $\sigma=-2\mu$, $\sigma=-2\lambda-2\mu$, and $\sigma=2\lambda$, so that

$$G_-(\lambda,\mu)=(8/3)[2\mu\lambda(\mu+\lambda) +(\mu-2\lambda)(\mu+\lambda)^2\ln2(\mu+\lambda) +\lambda^2(2\lambda+3\mu)\ln2\lambda-\mu^3\ln2\mu]\,. \quad (42b)$$

Using this evaluation of $G_\pm(\lambda,\mu)$, we obtain T_1^{-1} after some manipulation as

$$T_1^{-1}=32(A^2/\hbar H_ES)(kT/\pi H_E)^5I_3(0)\,, \quad (43)$$

so that in this case exchange scattering leads to an enhancement of T_1^{-1} by a factor of 8/3. Thus, we find no anomalous behavior in T_1^{-1} for low anisotropy as in Ref. 2. For ^{19}F in MnF_2 our result is smaller than that of Beeman and Pincus by a factor of about 10 for reasonable values of the parameters. Note also that for a material with low anisotropy, such as[15] $KMnF_3$, it would be vital to use our formula.

To summarize: We have recalculated the relaxation rate due to the transverse terms in the hyperfine coupling for a Heisenberg antiferromagnet at low temperatures. We find that the relaxation rate can be expanded in powers of $1/z$, and hence, we expect that the leading term should be qualitatively correct. We have given detailed formulas for possible use and have illustrated the results of our calculations for some limiting cases. Our calculations have the desirable property that they explicitly satisfy the requirements of spin kinematics and that for spin $\frac{1}{2}$ the results should be independent of D. We find no divergence in the relaxation rate in the limit of vanishing anisotropy. For $T\ll T_{AE}$, we find that the anisotropy energy gives rise to interactions between spin waves which are comparable to those due to the exchange energy. For ^{19}F in MnF_2, we find that T_1^{-1} is smaller than that predicted by Beeman and Pincus by a factor of about 10 for $T\gg T_{AE}$ and of about 7 for $T\ll T_{AE}$. Our results in both regimes indicate that spin-wave scattering leads to an enhancement in T_1^{-1} by a factor of about 5 over the three-magnon process as calculated using the Holstein-Primakoff formalism.[7] In other words, the ferromagnet and the antiferromagnet behave quite similarly in this respect. As is discussed by Beeman and Pincus, the relaxation process we have treated can only be observed under very special conditions, since usually other processes will dominate. However, the effect has been observed in a ferromagnet,[16] so perhaps it may also be observed in an antiferromagnet. A point of theoretical interest is that this calculation is closely related to the calculation of the wave-vector–dependent transverse correlation function in the static limit. This point will be developed in detail in a forthcoming publication. Finally, in the Appendix we have given the approximate spin-wave Hamiltonian for long wavelengths.

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APPENDIX

We will give the long-wavelength limit of $\Phi_{1234}^{(i)}$ for both the Dyson-Maleev Hamiltonian and the Holstein-Primakoff Hamiltonian arising from Eq. (2). We denote the coefficients for the Holstein-Primakoff Hamiltonian by $\tilde{\Phi}_{1234}^{(i)}$. It is readily seen that the Holstein-Primakoff Hamiltonian $\tilde{\mathcal{H}}$ is related to the Dyson-Maleev Hamiltonian by

$$\tilde{\mathcal{H}}=\tfrac{1}{2}(\mathcal{H}+\mathcal{H}^{\dagger}). \quad \text{(A1)}$$

Hence,

$$\tilde{\Phi}_{1234}^{(9)}=\tilde{\Phi}_{1234}^{(1)}=\tfrac{1}{2}(\Phi_{1234}^{(1)}+\Phi_{3412}^{(1)}), \quad \text{(A2a)}$$

$$\tilde{\Phi}_{3412}^{(3)}=\tilde{\Phi}_{1234}^{(2)}=\tilde{\Phi}_{2143}^{(6)}=\tilde{\Phi}_{3421}^{(5)} =\tfrac{1}{2}(\Phi_{1234}^{(2)}+\Phi_{3412}^{(3)}), \quad \text{(A2b)}$$

$$\tilde{\Phi}_{1234}^{(4)}=\tfrac{1}{2}(\Phi_{1234}^{(4)}+\Phi_{3412}^{(4)}), \quad \text{(A2c)}$$

$$\tilde{\Phi}_{1234}^{(7)}=\tilde{\Phi}_{1234}^{(8)}=\tfrac{1}{2}(\Phi_{1234}^{(7)}+\Phi_{3412}^{(8)}). \quad \text{(A2d)}$$

Thus, it is only necessary to give expressions for the $\Phi_{1234}^{(i)}$. For no anisotropy these coefficients are proportional to two powers of momenta, whereas for $D\neq 0$ they approach a constant value. We will evaluate only these two terms in each case. We use

$$x_i=(1-\epsilon_i)^{1/2}(1+\epsilon_i)^{-1/2}=1-\epsilon_i+\tfrac{1}{2}\epsilon_i^2+\cdots, \quad \text{(A3a)}$$

$$\gamma_i=1-\tfrac{1}{8}k_i^2, \quad \text{(A3b)}$$

whence

$$\Phi_{1234}^{(1)}\sim 4q+(\tfrac{1}{2}k_3\cdot k_4-2\epsilon_3\epsilon_4), \quad \text{(A4a)}$$

$$\Phi_{1234}^{(2)}\sim -4q-(\tfrac{1}{2}k_3\cdot k_4-2\epsilon_3\epsilon_4), \quad \text{(A4b)}$$

$$\Phi_{1234}^{(3)}\sim -4q-(\tfrac{1}{2}k_3\cdot k_4+2\epsilon_3\epsilon_4), \quad \text{(A4c)}$$

$$\Phi_{1234}^{(4)}\sim 4q+(\tfrac{1}{2}k_3\cdot k_4+2\epsilon_3\epsilon_4), \quad \text{(A4d)}$$

$$\Phi_{1234}^{(7)}\sim 4q+(\tfrac{1}{2}k_3\cdot k_4-2\epsilon_3\epsilon_4). \quad \text{(A4e)}$$

These formulas differ from those of Ref. 2 by the terms in $\epsilon_3\epsilon_4$.

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Spin Fluctuations Associated with the Formation of Localized Magnetic Moments in Superconductors

K. H. Bennemann

Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627*

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A new theory of superconductors containing transition-metal impurities is presented, explaining the observed anomalous magnetic behavior of the impurities with quenched or nearly quenched magnetic moment resulting from the localized spin fluctuations associated with the formation of localized magnetic moments. From this theory, the superconducting transition temperatures of *Al*Mn, *Al*Cr, and *V*Fe are calculated. The puzzling "slowing down" of the decrease in T_c observed for higher concentrations of transition-metal impurities is shown to arise partly from the temperature dependence of the electron scattering by the localized spin fluctuations and also partly from exchange and Coulomb coupling among the transition-metal impurities, which damp the formation of the localized magnetic moments and the spin fluctuations.

I. INTRODUCTION

Electron scattering by localized spins strongly weakens superconductivity,[1,2] in sharp contrast to the small mean-free-path effects of nonmagnetic impurities.[3] Consequently, superconductors should reflect sensitively the quenching of localized magnetic moments.[4] The observed superconducting transition temperatures T_c of Al, Zn, In, Sn, and V containing transition-metal impurities like Cr, Mn, Fe and Ni, for example, confirm this expectation.[2] However, the measurements of T_c revealed an anomalous magnetic behavior of the transition-metal impurities with quenched or nearly quenched localized magnetic moment. The unexpectedly large suppression of T_c in such alloys is demonstrated in Fig. 1. This is very puzzling, since magnetic measurements and the absence of a resistivity minimum in *Al*Mn, *Al*Cr, and *V*Fe, for example, have shown that localized magnetic moments are absent or at least are very faint in these alloys.[2] Furthermore, the very puzzling and interesting observation is made that T_c decreases less rapidly for larger transition-metal impurity concentrations c. It is the purpose of this paper to explain this puzzling behavior of T_c resulting from localized spin excitations associated with the formation of localized magnetic moments.

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