[6] J. S. Smart, *Effective Field Theories of Magnetism* (W. B. Sanders, Philadelphia, Pa., 1966).

[7] R. K. Quinn and R. C. Knauer, Rev. Sci. Instr. 43, 1543 (1972).

[8] L. N. Mulay, *Magnetic Susceptibility* (Interscience, New York, 1963).

[9] W. Opechowski, Physica 4, 181 (1937); 6, 1112 (1938).

[10] B. Strieb, H. B. Callen, and G. Horwitz, Phys. Rev. 130, 1798 (1963).

PHYSICAL REVIEW B VOLUME 7, NUMBER 1 1 JANUARY 1973

Temperature Dependence of the Ferromagnetic-Resonance Linewidth*

Peter K. Leichner
Department of Physics and Astronomy, University of Kentucky, Lexington, Kentucky 40506

and

Peter M. Richards†
Department of Physics, University of Kansas, Lawrence, Kansas 66044
and Sandia Laboratories, Albuquerque, New Mexico 87115

(Received 16 June 1972)

A calculation of the temperature dependence of the magnetic-resonance linewidth in ferromagnetic crystals is presented. It is based on the Kubo–Tomita relaxation function and Van Vleck's method of moments, generalized to finite temperatures. Our work differs from previous general theories in that detailed calculations are performed including the applied field so that meaningful comparison between theory and experiment is possible over a wide range of temperatures including the Curie region. The temperature dependence of the moments is calculated in two steps. First, the four- and six-spin correlation functions which occur in the second and fourth moments, respectively, are decoupled into sums of products of two-spin correlation functions. These two-spin thermal averages are then computed in the constant-coupling approximation, since we show that nearest-neighbor static correlations are by far the most important. In this manner linewidth calculations are made for two ferromagnets, $K_2CuCl_4 \cdot 2H_2O$ and Ni, for temperatures ranging from $T = 0$ K to $T = 10T_C$. It is found that the method gives reasonable agreement with the experiments of Ford and Jeffries on $K_2CuCl_4 \cdot 2H_2O$ and of Bhagat and Chicklis and of Salamon on Ni.

I. INTRODUCTION

In this paper we investigate the behavior of the exchange-narrowed ferromagnetic-resonance linewidth as a function of temperature. In spite of much previous effort on the general theory of magnetic-resonance linewidth,[1,2] there appear to be no detailed calculations of the temperature dependence of the linewidth which can be directly applied to the full range of experimental parameters for a ferromagnet. Treatments at finite temperature have tended to be either phenomenological or limited to nonrealistic situations such as zero applied field.[3,4] It is particularly important that the external field be included, because it removes the divergence of the uniform susceptibility at the Curie point, and this can drastically alter the linewidth in the critical region. Since experiments have been performed over a wide range of temperatures above and below the transition point in finite fields, there is considerable interest in having general formulas for finite-temperature linewidths and performing the computations necessary for meaningful comparison between theory and experiment.

The linewidth is calculated in terms of the second and fourth moments as originally proposed by Van Vleck.[5] The stochastic theory of Anderson and Weiss[6] and the relaxation-function approach of Kubo and Tomita also utilize the moments and reproduce Van Vleck's results in the infinite-temperature limit. Richards[7] has shown that these methods may also be applied at finite temperature; so one essentially has only to compute the second and fourth moments at the temperature of interest in order to compute the linewidth within the framework of Kubo and Tomita's general theory.

Describing the linewidth by second and fourth moments can be risky for pathological line shapes; however, for a well-behaved line shape—Lorentzian near the center and more sharply cut off in the wings—this has been shown to produce reasonable results in the infinite-temperature limit. Also, as mentioned by Kawasaki,[3] use of moments may be more correct for large external fields, as encountered in this work.

Green's-function techniques have also been used[8] to calculate temperature dependence of the linewidth, but these entail decoupling of time correlation functions which may be more questionable than decoupling of static correlations functions, as employed here. Also, they apparently lead to results which are quite different from those presented here and not in agreement with experiment (see Ref. 8). Our philosophy has been first to see what the conventional moments treatment produces before trying techniques which are more difficult computationally and which involve approximations whose physical basis is often obscure. That, as we show, good agreement can be obtained with experiment may be *a posteriori* justification for the techniques employed here.

The actual computation of the moments is achieved by decoupling the four- and six-spin correlation functions, which occur in the second and fourth moments, respectively, into sums of products of two-spin correlation functions. The resulting two-spin thermal averages are calculated in the constant-coupling[9,10] approximation for reasons which we discuss later. In this manner explicit calculations of the linewidth are made for two ferromagnets, $K_2CuCl_4 \cdot 2H_2O$ and Ni, for temperatures ranging from $T = 0$ K to $T = 10T_C$.

A notable feature of our results is that the various secular and nonsecular components do not have an identical temperature dependence. Furthermore, the temperature dependence of the effective exchange frequency ω_e is not, in general, consistent with the often-used argument that ω_e decreases with onset of ordering due to slowing down of the rate of fluctuations.

The linewidth calculations are then compared with the measurements of Ford and Jeffries[11] for the potassium salt and with those of Bhagat and Chicklis[12] and Salamon[13] for Ni. We find that the constant-coupling approximation can give reasonably good agreement with experiment. Comparison between $K_2CuCl_4 \cdot 2H_2O$ and Ni is of interest because the former has $\hbar\omega_0/k_BT_C \sim 1$ while the latter has $\hbar\omega_0/k_BT_C \ll 1$, where $\hbar\omega_0/k_BT_C$ is the ratio of Zeeman energy to thermal energy at the Curie point. We find that the width in Ni has a temperature dependence given simply by the denominator $\langle M_x^2\rangle^{-1}$ in the general theory [see Eq. (1)]; but the numerator, which is related to dynamical spin correlations, plays a decisive role in $K_2CuCl_4 \cdot 2H_2O$.

II. GENERAL THEORY OF LINEWIDTH

In Ref. 7, hereafter referred to as I, one of us extended the methods of Kubo and Tomita to finite temperature. The notation used here is the same as in I, and the reader is referred to it for background and additional references.

The relaxation rate η is given by[7]

$$\eta = \sum_M \int_{-\infty}^{\infty} d\tau \times \frac{e^{iM\omega_0\tau}\langle \hat{g}_M^\dagger(\tau) g_M(0)\rangle + e^{-iM\omega_0\tau}\langle \hat{g}_M(\tau) g_M^\dagger(0)\rangle}{8\hbar^2\langle M_x^2\rangle}, \qquad (1)$$

where

$$\hat{g}_M(\tau) = e^{-i\mathcal{H}_{\text{ex}}\tau/\hbar}[G_M,\ M_+]\, e^{i\mathcal{H}_{\text{ex}}\tau/\hbar}, \qquad (2)$$

in which $\mathcal{H}_{\text{ex}}$ is the isotropic Heisenberg-exchange Hamiltonian, M_+ is the usual raising operator associated with the transverse components of total magnetization M_x and M_y, and G_M is that part of the perturbation $\mathcal{H}'$ which induces a change M in total Zeeman quantum number. For the purposes of this paper $\mathcal{H}'$ is either the classical or pseudodipolar coupling, and thus $M = \pm 2, \pm 1, 0$. The unperturbed angular resonance frequency is

$$\omega_0 = g\mu_B H_0/\hbar, \qquad (3)$$

where g is the spectroscopic splitting factor, μ_B the Bohr magneton, and H_0 the applied field. Triangular brackets indicate thermal average with respect to the unperturbed density matrix in which $\mathcal{H}'$ is neglected. A Lorentzian line shape is assumed except far from the center, so that the full linewidth at half-maximum ΔH is related to η by

$$\Delta H = 2\eta\hbar/g\mu_B. \qquad (4)$$

In I the relaxation rate was derived by considering the Fourier transform of the line-shape function $\chi''(\omega)\coth(\frac{1}{2}\beta\hbar\omega)$, where χ'' is the imaginary part of the susceptibility and $\beta = 1/k_BT$, with k_B Boltzmann's constant and T the absolute temperature. We show in Appendix A that the same result is obtained from the Fourier transform of $\chi''(\omega)/\omega$ if the perturbation $\mathcal{H}'$ is neglected in computation of thermal averages.

Frequency shifts can be important at finite temperature. It was shown in I that Eq. (1) above holds as long as the frequency shift is small compared to ω_0. As pointed out in I, the fact that the linewidth can be computed without having to know the exact location of the line center is a virtue of exchange-narrowed lines. [See Eq. (19) of I and the ensuing discussion.] In nonspherical samples, frequency shifts of the order of ω_0 can occur owing to demagnetizing fields. We show, however, in Appendix B that such shifts do not affect our calculations.

The quantity $\langle\hat{g}_M^\dagger(\tau) g_M(0)\rangle$ is assumed to decay in a characteristic time τ_c. For either of the limiting cases $\omega_0\tau_c \ll 1$ or $\omega_0\tau_c \gg 1$, it is not necessary to know the precise form of $\langle\hat{g}_M^\dagger(\tau)\hat{g}_M(0)\rangle$ in order to study the temperature dependence of the linewidth. This is because, as discussed in I, if $\omega_0\tau_c \ll 1$, then $e^{iM\omega_0\tau} \approx 1$ for all times of interest, while if $\omega_0\tau_c \gg 1$, the $M = 0$ term dominates. In either case one is left with $\int_{-\infty}^{\infty} f_M(\tau)\,d\tau$ to evaluate, where M is restricted to zero if $\omega_0\tau_c \gg 1$, and we have defined

$$f_M(\tau)=\langle\hat{g}^\dagger_M(\tau)g_M(0)\rangle+\langle\hat{g}_M(\tau)g^\dagger_M(0)\rangle . \tag{5}$$

The approximation

$$\int_{-\infty}^{\infty} f_M(\tau)\,d\tau = cf_M(0)/\omega_e^{(M)} , \tag{6a}$$

with

$$\omega_e^{(M)}=[\ddot{f}_M(0)/f_M(0)]^{1/2} , \tag{6b}$$

is then made, where c is assumed to be temperature independent and of the order of unity [$c=(2\pi)^{1/2}$ for a Gaussian decay].

The frequency $\omega_e^{(M)}$ in (6) is of the order of the decay rate τ_c^{-1} and is equivalent to the exchange frequency ω_e introduced by Anderson and Weiss. A superscript (M) is necessary since $\omega_e^{(M)}$ is different for different M's and shows different temperature dependences.

If neither of the limiting conditions is satisfied, then an analytic expression for the dependence of $\langle\hat{g}^\dagger_M(\tau)g_M(0)\rangle$ is required. If the usual Gaussian form is taken,

$$f_M(\tau)=f_M(0)\,e^{-[\omega_e^{(M)}]^2\tau^2/2} , \tag{7}$$

then the expression (1) reduces to

$$\eta=(2\pi)^{1/2}\sum_M \frac{f_M(0)\exp\{-\frac{1}{2}(M\omega_0)^2/[\omega_e^{(M)}]^2\}}{8\omega_e^{(M)}\hbar^2\langle M_x^2\rangle} \tag{8}$$

if it is also assumed that $\langle g^\dagger_M(\tau)g_M(0)\rangle$ is an even function of τ [see Eqs. (11)–(13) and the discussion below]. Identical expressions have been obtained by Tomita[14] and Henderson and Rogers.[15] The terms in Eq. (8) are directly related to the second and fourth moments of the resonance line as shown in I. For future reference the Mth contribution to the second moment M_2 is

$$M_2^{(M)}=f_M(0)/4\hbar^2\langle M_x^2\rangle \tag{9a}$$

and the contribution to the fourth moment M_4 is

$$M_4^{(M)}=\ddot{f}_M(0)/4\hbar^2\langle M_x^2\rangle , \tag{9b}$$

so that Eq. (6) becomes

$$\omega_e^{(M)}=[M_4^{(M)}/M_2^{(M)}]^{1/2} . \tag{10}$$

Of the two materials studied here, we have $M\omega_0 \ll \omega_e^{(M)}$ for Ni, so that the Gaussian form is not crucial. For $K_2CuCl_4\cdot 2H_2O$, however, ω_0 and $\omega_e^{(M)}$ are comparable, so that detailed comparison requires the explicit form (7).

A complication at finite temperature is that the assumed Gaussian form of Eq. (8) makes the Fourier frequency transform $\langle g^\dagger_M g_M\rangle_\omega$ an even function of ω, whereas we know from general principles that

$$\langle g^\dagger g\rangle_\omega = e^{-\beta\hbar\omega}\langle gg^\dagger\rangle_{-\omega} \tag{11}$$

must hold for the frequency dependence of any time correlation function $\langle g^\dagger(\tau)g(0)\rangle$. A somewhat more sophisticated approximation would then be to take

$$\langle g^\dagger_M g_M\rangle_\omega=\langle g^\dagger_M g_M\rangle_{\omega=0}\,e^{-\beta\hbar\omega/2}\,e^{-\omega^2/2\sigma^2} \tag{12}$$

as a convenient, but by no means unique, way of satisfying Eq. (11) and maintaining a Gaussian character. It may be shown that σ is related to ω_e as defined in Eq. (6) by

$$\omega_e^2=\sigma^2(1+\tfrac{1}{4}\beta^2\hbar^2\sigma^2) . \tag{13}$$

We have investigated how (12) and (13) alter the calculated linewidth and find that, in the regions of experimental interest for the compounds treated, there is a negligible effect. For simplicity we therefore do not further consider modifications such as are required by Eq. (11).

III. CALCULATION OF M_2 AND M_4

For $\mathcal{H}'$ given by the classical dipolar perturbation, the quantities G_M are of the form

$$G_M=\sum_{ij} F_{ij}^{(M)}A_{ij}^{(M)} , \tag{14}$$

where we have defined $F_{ii}^{(M)}=0$. The factors $F_{ij}^{(M)}$ and $A_{ij}^{(M)}$ are given by

$$F_{ij}^{(0)}=-\tfrac{3}{4}(g\mu_B)^2r_{ij}^{-3}(3\cos^2\theta_{ij}-1) , \tag{15a}$$

$$F_{ij}^{(\pm1)}=-\tfrac{3}{4}(g\mu_B)^2r_{ij}^{-3}\sin\theta_{ij}\cos\theta_{ij}\,e^{\mp i\varphi_{ij}} , \tag{15b}$$

$$F_{ij}^{(\pm2)}=-\tfrac{3}{8}(g\mu_B)^2r_{ij}^{-3}\sin^2\theta_{ij}\,e^{\mp2i\varphi_{ij}} \tag{15c}$$

and

$$A_{ij}^{(0)}=S_i^zS_j^z-\tfrac{1}{3}\vec{S}_i\cdot\vec{S}_j , \tag{16a}$$

$$A_{ij}^{(\pm1)}=S_i^zS_j^\pm+S_i^\pm S_j^z , \tag{16b}$$

$$A_{ij}^{(\pm2)}=S_i^\pm S_j^\pm . \tag{16c}$$

The commutators g_M, Eq. (2), then become

$$g_{-2}=-4g\mu_B\sum_{ij} F_{ij}^{(-2)}S_i^zS_j^- , \tag{17a}$$

$$g_{-1}=2g\mu_B\sum_{ij} F_{ij}^{(-1)}(S_i^+S_j^--2S_i^zS_j^z) , \tag{17b}$$

$$g_0=2g\mu_B\sum_{ij} F_{ij}^{(0)}S_i^zS_j^+ , \tag{17c}$$

$$g_1=2g\mu_B\sum_{ij} F_{ij}^{(1)}S_i^+S_j^+ , \tag{17d}$$

$$g_2=0 , \tag{17e}$$

where we have used $F_{ij}^{(M)}=F_{ji}^{(M)}$. Using these results, one obtains for the second moment the following expressions:

$$\hbar^2\langle S_\perp^2\rangle M_2^{(-2)}=16\sum_{ijkl}F_{ij}^{(-2)}F_{kl}^{(+2)}\langle S_i^zS_j^-S_k^+S_l^z+S_k^+S_l^zS_i^zS_j^-\rangle , \tag{18a}$$

$$\hbar^2\langle S_\perp^2\rangle M_2^{(-1)}=4\sum_{ijkl}F_{ij}^{(-1)}F_{kl}^{(+1)}\langle S_i^+S_j^-S_k^+S_l^-+S_k^+S_l^-S_i^+S_j^- + 8S_i^zS_j^zS_k^zS_l^z-4(S_i^zS_j^zS_k^+S_l^-+S_k^+S_l^-S_i^zS_j^z)\rangle , \tag{18b}$$

$$\hbar^2\langle S_\perp^2\rangle M_2^{(0)}=4\sum_{ijkl}F_{ij}^{(0)}F_{kl}^{(0)}\langle S_i^zS_j^+S_k^-S_l^z+S_k^-S_l^zS_i^zS_j^+\rangle , \tag{18c}$$

$$\hbar^2\langle S_\perp^2\rangle M_2^{(1)} = 4\sum_{ijkl} F_{ij}^{(+1)}F_{kl}^{(-1)}\langle S_i^+S_j^+S_k^-S_l^- + S_k^-S_l^-S_i^+S_j^+\rangle\,, \tag{18d}$$

where $S_\perp^2 = S_+S_- + S_-S_+$. The component $M_2^{(+2)}$ is identically equal to zero because the corresponding commutator g_{+2} is zero.

The components of the fourth moment are evaluated according to Eqs. (2) and (1). After some lengthy commutator algebra, one finds the following relations:

$$\hbar^4\langle S_\perp^2\rangle M_4^{(-2)} = 4\sum_{ijk}\sum_{lmn}[J_{ij}(F_{ij}^{(-2)} - F_{jk}^{(-2)}) + J_{jk}(F_{ik}^{(-2)} - F_{ij}^{(-2)})]\,[J_{lm}(F_{ln}^{(2)} - F_{mn}^{(2)}) + J_{mn}(F_{ln}^{(2)} - F_{lm}^{(2)})]$$
$$\times\langle[S_i^-S_j^+S_k^- + 2S_i^zS_j^-S_k^z\,,\; S_l^+S_m^-S_n^+ + 2S_l^zS_m^+S_n^z]_+\rangle\,, \tag{19}$$

$$\hbar^4\langle S_\perp^2\rangle M_4^{(-1)} = 8\sum_{ijk}\sum_{lmn}[J_{ij}(F_{jk}^{(-1)} - F_{ik}^{(-1)}) + J_{jk}(F_{ik}^{(-1)} - F_{ij}^{(-1)}) + 2J_{ik}(F_{jk}^{(-1)} - F_{ij}^{(-1)})]$$
$$\times[J_{lm}(F_{ln}^{(1)} - F_{mn}^{(1)}) + J_{mn}(F_{lm}^{(1)} - F_{ln}^{(1)}) + 2J_{ln}(F_{lm}^{(1)} - F_{mn}^{(1)})]\langle[S_i^+S_j^zS_k^- - S_i^-S_j^zS_k^+\,,\; S_l^+S_m^zS_n^- - S_l^-S_m^zS_n^+]_+\rangle\,, \tag{20}$$

$$\hbar^4\langle S_\perp^2\rangle M_4^{(1)} = 16\sum_{ijk}\sum_{lmn}[J_{ij}(F_{jk}^{(1)} - F_{ik}^{(1)}) + J_{jk}(F_{ij}^{(1)} - F_{ik}^{(1)})]$$
$$\times[J_{lm}(F_{mn}^{(-1)} - F_{ln}^{(-1)}) + J_{mn}(F_{lm}^{(-1)} - F_{ln}^{(-1)})]\langle[S_i^+S_j^zS_k^+\,,\; S_l^-S_m^zS_n^-]_+\rangle\,. \tag{21}$$

The exchange Hamiltonian has been taken as

$$\mathcal{H}_{\text{ex}} = -\sum_{i,j} J_{ij}\vec{S}_i\cdot\vec{S}_j \tag{22}$$

in arriving at Eqs. (19)–(21).

The term $M_4^{(0)}$ is identical with $M_4^{(-2)}$, except that the lattice factors $F_{ij}^{(\pm 2)}$ are replaced by $F_{ij}^{(0)}$, and the right-hand side of Eq. (19) is multiplied by a factor of $\frac{1}{4}$. The resulting secular component $M_4^{(0)}$ is identical with that given by Van Vleck.[5]

The second and fourth moments involve four- and six-spin correlation functions, respectively. To make progress we decouple these into sums of products of two-spin correlation function in analogy with Wick's theorem.[16] For example, we take

$$\langle S_i^zS_j^-S_k^zS_l^+\rangle = \langle S_i^zS_k^z\rangle\langle S_j^-S_l^+\rangle\,, \tag{23a}$$

$$\langle S_i^+S_j^-S_k^+S_l^-\rangle = \langle S_i^+S_j^-\rangle\langle S_k^+S_l^-\rangle + \langle S_i^+S_l^-\rangle\langle S_j^-S_k^+\rangle\,, \tag{23b}$$

$$\langle S_i^zS_j^zS_k^zS_l^z\rangle = \langle S_i^zS_j^z\rangle\langle S_k^zS_l^z\rangle + \langle S_i^zS_k^z\rangle\langle S_j^zS_l^z\rangle + \langle S_i^zS_l^z\rangle\,\langle S_j^zS_k^z\rangle \tag{23c}$$

if all indices are different. Other combinations, such as $\langle S_i^zS_j^+\rangle$, are equal to zero because the unperturbed density matrix, with respect to which the thermal averages are taken, is invariant under rotations about the z axis, which is the direction of applied field. Decoupling schemes such as (23) are commonly used in investigations of the temperature dependence of the Heisenberg ferromagnet. In particular, the Green's-function equations of motion are decoupled in the same manner as above (see, for example, Tahir-Kheli and ter Haar[17] and Callen[18]).

For simplicity we decouple according to (23) even for spins on the same lattice site; so, for example, the first term of Eq. (18a) is written as

$$\sum_{ijkl} F_{ij}^{(-2)}F_{kl}^{(2)}\langle S_i^zS_j^-S_k^+S_l^z\rangle \approx \sum_{ijkl} F_{ij}^{(-2)}F_{kl}^{(2)}\langle S_i^zS_l^z\rangle\langle S_j^-S_k^+\rangle\,. \tag{24}$$

It is shown in detail elsewhere[19] that for $S=\frac{1}{2}$ and cubic symmetry the above approximation holds to $O(1/Z^2)$, where Z is the number of nearest-neighbor ions, at finite temperatures. (It is exact at infinite temperature and at zero temperature.) This is fully consistent with the decoupling of the correlation functions which is expected to be valid to $O(1/Z)$.

Therefore, in the approximation (24) one obtains

$$\hbar^2\langle S_\perp^2\rangle M_2^{(-2)} = 16\sum_{ijkl} F_{ij}^{(-2)}F_{kl}^{(+2)}C_{il}^{zz}(C_{jk}^{+-} + C_{jk}^{-+})\,, \tag{25}$$

where the following short-hand notation has been used:

$$C_{il}^{zz} \equiv \langle S_i^zS_l^z\rangle\,, \quad C_{jk}^{+-} \equiv \langle S_j^+S_k^-\rangle\,. \tag{26}$$

Similarly,

$$\hbar^2\langle S_\perp^2\rangle M_2^{(-1)} = 8\sum_{ijkl} F_{ij}^{(-1)}F_{kl}^{(1)}(C_{jk}^{-+}C_{il}^{+-} + 8C_{ik}^{zz}C_{il}^{zz})\,, \tag{27}$$

$$\hbar^2\langle S_\perp^2\rangle M_2^{(0)} = 4\sum_{ijkl} F_{ij}^{(0)}F_{kl}^{(0)}C_{il}^{zz}(C_{jk}^{+-} + C_{jk}^{-+})\,, \tag{28}$$

$$\hbar^2\langle S_\perp^2\rangle M_2^{(1)} = 8\sum_{ijkl} F_{ij}^{(1)}F_{kl}^{(-1)}(C_{ik}^{+-}C_{jl}^{+-} + C_{ik}^{-+}C_{jl}^{-+})\,. \tag{29}$$

The decoupling of the fourth moment proceeds in the same manner, except that the six-spin correlation functions are first decoupled into sums of products of two- and four-spin correlation functions; the latter are subsequently reduced to two-spin thermal averages. Thus, the contributions to the fourth-moment sums are given by

$$\hbar^4\langle S_\perp^2\rangle M_4^{(-2)} = 4\sum_{ijk}\sum_{lmn}\{[J_{ij}(F_{ik}^{(-2)} - F_{jk}^{(-2)}) + J_{jk}(F_{ik}^{(-2)} - F_{ij}^{(-2)})][J_{lm}(F_{ln}^{(2)} - F_{mn}^{(2)}) + J_{mn}(F_{ln}^{(2)} - F_{lm}^{(2)})]$$

$$\times[(C_{il}^{+-} + C_{il}^{-+})C_{jm}^{-+}C_{kn}^{+-} + (C_{in}^{+-} + C_{in}^{-+})C_{jm}^{+-}C_{kl}^{-+} + 4(C_{jm}^{+-} + C_{jm}^{-+})(C_{il}^{zz}C_{kn}^{zz} + C_{in}^{zz}C_{kl}^{zz})]\}. \quad (30)$$

The $M_4^{(0)}$ part is obtained from this by changing the lattice factors from $F_{ij}^{(\pm 2)}$ to $F_{ij}^{(0)}$ and multiplying the right-hand side by a factor of $\frac{1}{4}$. The $M_4^{(\pm 1)}$ components are

$$\hbar^4\langle S_\perp^2\rangle M_4^{(-1)} = 8\sum_{ijk}\sum_{lmn}\{[J_{ij}(F_{jk}^{(-1)} - F_{ik}^{(-1)}) + J_{jk}(F_{jk}^{(-1)} - F_{ij}^{(-1)}) + 2J_{ik}(F_{jk}^{(-1)} - F_{ij}^{(-1)} - F_{ij}^{(-1)})]$$

$$\times[J_{lm}(F_{ln}^{(1)} - F_{mn}^{(1)}) + J_{mn}(F_{lm}^{(1)} - F_{ln}^{(1)}) + 2J_{ln}(F_{lm}^{(1)} - F_{mn}^{(1)})]C_{jm}^{zz}[C_{in}^{+-}C_{kl}^{-+} + C_{in}^{-+}C_{kl}^{+-} - (C_{il}^{-+}C_{kn}^{+-} + C_{il}^{+-}C_{kn}^{-+})]\} \quad (31)$$

and

$$\hbar^4\langle S_\perp^2\rangle M_4^{(1)} = 16\sum_{ijk}\sum_{lmn}\{[J_{ij}(F_{jk}^{(1)} - F_{ik}^{(1)}) + J_{jk}(F_{ij}^{(1)} - F_{ik}^{(1)})][J_{lm}(F_{mn}^{(-1)} - F_{ln}^{(-1)}) + J_{mn}(F_{lm}^{(-1)} - F_{ln}^{(-1)})]$$

$$\times C_{jm}^{zz}(C_{il}^{+-}C_{kn}^{+-} + C_{in}^{+-}C_{kl}^{+-} + C_{il}^{-+}C_{kn}^{-+} + C_{kl}^{-+}C_{in}^{-+})\}. \quad (32)$$

Hence, we now have finite-temperature expressions for the second and fourth moments of the resonance line, with the temperature dependence determined by the behavior of the two-spin correlation functions. The decoupled expressions above are exact in the infinite-temperature limit and agree with those given by Van Vleck[5] and Tomita.[14]

It remains to compute the static two-spin correlation functions. Since there are no exact solutions for the correlations of a Heisenberg ferromagnet in an applied field, one is forced to use approximate results. The choice of approximation technique is dictated by the types of correlations which are likely to be most important. In zero field the correlations are dominated by long-wavelength fluctuations in the critical region and it is essential to use a theory which accounts for these properly. The applied field, however, alters the picture considerably and makes it appear that only short-range correlations are important. This is illustrated by the following consideration.

The parameter of interest is the correlation length κ^{-1} in the presence of an applied field. It is defined by

$$\langle S_q^- S_q^+\rangle \propto (q^2 + \kappa^2)^{-1}$$

for small q, where $\langle S_q^- S_q^+\rangle$ is the spatial Fourier component of $\langle S_i^+ S_j^-\rangle$ at wave vector q. From the random-phase approximation (RPA) or, equivalently, high-density or ring-diagram approximations,[20,21] we have

$$\langle S_q^- S_q^+\rangle = R(e^{\beta[g\mu_B H_0 + (J_0 - J_q)R]} - 1)^{-1}, \quad (33)$$

where $R = 2\langle S_i^z\rangle$ is the relative magnetization for spin $\frac{1}{2}$, H_0 is the applied field, and J_q is the spatial Fourier component at wave vector q of the exchange interaction J_{ij}. For nearest-neighbor exchange interactions in a cubic lattice with nearest-neighbor distance a, we then obtain

$$\kappa^{-1} \approx a(k_B T_C R/3g\mu_B H_0)^{1/2} \quad (34)$$

upon expanding the exponential in Eq. (35) for small values of its argument and using the $S = \frac{1}{2}$ mean-field relation $J_0 = 2k_B T_C$ for the Curie temperature T_C. The relative magnetization R in finite fields is reasonably well described by molecular-field theory:

$$R = \tanh[\tfrac{1}{2}g\mu_B H_0/k_B T + R(T_C/T)] \quad (35)$$

for spin $\frac{1}{2}$. For $K_2CuCl_4 \cdot 2H_2O$, we have $T_C \approx 1$ K and $H_0 \approx 3$ kOe in the experiment of Ford and Jeffries, so that $g\mu_B H_0/k_B T_C \approx 0.4$ and $R \approx 0.8$ at the Curie point. Use of these numbers in Eq. (34) then yields $\kappa^{-1} \approx 0.8a$. Hence, we see that for the field used in $K_2CuCl_4 \cdot 2H_2O$ the correlation length at T_C is less than one nearest-neighbor spacing. Even for Ni, where $g\mu_B H_0/k_B T_C \approx 1.5 \times 10^{-3}$ is appropriate for the data to be considered, we find $R \approx 0.2$ and thus $\kappa^{-1} \approx 7a$ at the Curie point.

The above figures demonstrate how effectively the field required for a magnetic-resonance experiment eliminates the long-range correlations in the critical region. Similar observations were made by Kawasaki.[3] A further effect which tends to discriminate against long-range correlations is the angular dependence of the dipole factors. Consider, for example, the $M = 0$ part of the second moment:

$$M_2^{(0)} = 4(\hbar^2\langle S_\perp^2\rangle)^{-1}\sum_{ijkl} F_{ij}^{(0)} F_{kl}^{(0)} C_{il}^{zz}(C_{jk}^{+-} + C_{jk}^{-+}).$$

If we assume zero field for the moment, then $\kappa^{-1} \to \infty$ at T_C and the correlations decay approximately as $C_{ij} \propto r_{ij}^{-1}$.

For simplicity we take the case of pseudodipolar coupling, for which the factors are of the form

$$F_{ij}^{(0)} = -\tfrac{3}{4}P_{ij}(3\cos^2\theta_{ij} - 1),$$

with $P_{ij} = P$ for nearest neighbors and 0 otherwise. We thus treat the summation

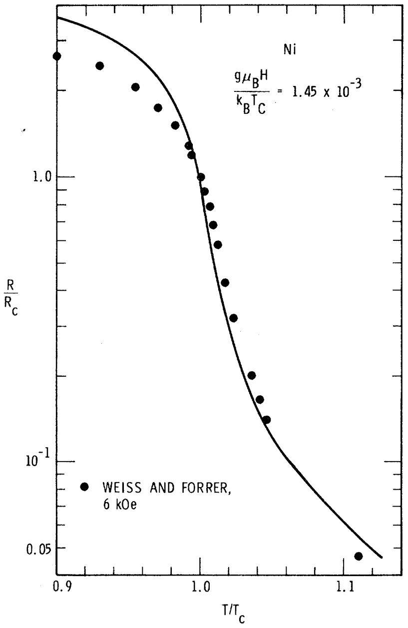

FIG. 1. Relative magnetization R in Ni normalized to its value R_c at the Curie point $T_C = 627$ K. Experimental points are data of Weiss and Forrer in a field of 6.015 kOe, for which $R_c = 0.234$. Curve is constant-coupling approximation at 6 kOe, for which $R_c = 0.141$. It is practically indistinguishable from the molecular-field-theory curve on this scale.

$$\sum_4^0 = \sum_{ijkl}{}' F_{ij}^{(0)} F_{kl}^{(0)} r_{il}^{-1} r_{jk}^{-1} ,$$

where the prime indicates that all indices are unequal. The dipole factors F_{ij} are restricted to nearest neighbors, but the distances r_{il} and r_{jk} are not restricted. With this constraint on F_{ij}, computer calculations were carried out in which both r_{il} and r_{jk} were permitted to take on values ranging from one to six nearest-neighbor distances. The results of these calculations show that for a fcc lattice 96% of the contributions to $\sum_4^0$ arise from terms in which both r_{il} and r_{jk} are equal to the nearest-neighbor distance. The presence of the angular factors in the lattice sums, and the resulting averaging which takes place, thus severly limit the importance of long-range correlations in the calculation of the linewidth even if the correlation length were to be infinite.

From the preceding discussion it is evident that, for the problem at hand, nearest-neighbor correlations are likely to be the most important by far. For this reason, we have chosen to use the constant-coupling approximation[9,10] which provides a reasonable picture of nearest-neighbor correlations which can be—and has been—readily extended to include an applied field.

The constant-coupling approximation is of the small-cluster type. It differs from other cluster results, such as those obtained by Oguchi[22] and the Bethe–Peierls–Weiss[23,24] method, in that it is internally self-consistent. Thus, it is considered to be the most satisfactory of the simple correlation theories. In the constant-coupling approximation we take

$$\langle S_i^\alpha S_j^\alpha \rangle = \langle S_i^\alpha \rangle \langle S_j^\alpha \rangle , \tag{36}$$

unless $i = j$ or i and j are nearest neighbors, where α is any component x, y, or z. For nearest neighbors we use the equations found in Refs. 9 and 10. The constant-coupling relative magnetization R_{cc} is also found in these references.

A measure of the accuracy of the constant-coupling approximation may be obtained from the recent work of Ritchie and Fisher.[25] They have estimated correlation functions of the Heisenberg ferromagnet from high-temperature series expansions and find $4\langle S_i^z S_j^z \rangle = 0.199$ and 0.187 for nearest neighbors at T_C in spin-$\frac{1}{2}$ bcc and fcc lattices, respectively. The constant-coupling result for this quantity is $1/(Z-1)$, where Z is the coordination number and thus is 0.143 and 0.091 for bcc and fcc, respectively. Hence, at worst, this approximation underestimates the correlation by about a factor of 2 at the Curie point in zero field. In finite fields the error may be considerably less because the fluctuations are greatly reduced, as discussed above.

In the calculations presented in the following section we have adopted the following procedure. The exchange coupling J is chosen to give the observed Curie temperature (627 K for Ni, 1.1 K for $K_2CuCl_4 \cdot 2H_2O$) in the constant-coupling approximation:

$$J = \tfrac{1}{2} k_B T_C \ln[Z/(Z-4)].$$

The denominator $\langle S_\perp^2 \rangle = \langle S_+ S_- + S_- S_+ \rangle$ appearing in the moments expression is calculated in terms of R from the identity

$$\langle S_\perp^2 \rangle = NR \coth(g\mu_B H_0 / 2k_B T) , \tag{37}$$

which is derived in Appendix C (N is the total number of spins). The relative magnetization R may be taken from the data of Ford and Jeffries for $K_2CuCl_4 \cdot 2H_2O$ and Weiss and Forrer[26] for Ni. For both systems we find that the temperature variation of R in the region of interest is adequately given either by mean-field theory [Eq. (35)] or by the constant-coupling value R_{cc}. Figure 1 illustrates the situation in Ni at 6 kOe in the critical region $0.9T_C \leq T \leq 1.1T_C$. Once again, we see that the finite field produces a smooth variation near T_C which is well described by the simple theories. Comparison between the measured and molecular-field values for R in the $K_2CuCl_4 \cdot 2H_2O$ may be found in Ref. 11.

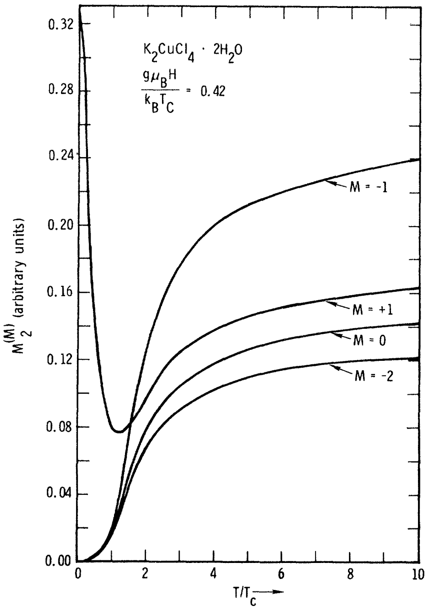

FIG. 2. Temperature variation of second moment $M_2^{(M)}$ for $K_2CuCl_4 \cdot 2H_2O$, with $H = 3.13$ kOe, $T_C = 1.1$ K.

IV. RESULTS OF NUMERICAL CALCULATIONS AND COMPARISON WITH EXPERIMENT

In this section we compare our calculations to linewidth measurements in two ferromagnetic substances, $K_2CuCl_4 \cdot 2H_2O$ and Ni.

A. $K_2CuCl_4 \cdot 2H_2O$

The potassium salt is regarded as an ideal Heisenberg spin-$\frac{1}{2}$ insulating ferromagnet with a Curie temperature of approximately 1.1 K. The crystal structure[27] is bcc with a 6% tetragonal distortion along the c axis, which we assume to be small enough to be ignored for the purpose of linewidth calculations. Miedema *et al.*[28] have used bcc lattice constants to compare specific-heat measurements with Dyson's[29] spin-wave theory. They find excellent agreement with theory for temperatures ranging from $T = 0.1T_C$ to $T = 0.5T_C$ by using nearest-neighbor exchange interactions only. The exchange constant determined in this manner has a value of $J/k_B = 0.282$ K. Ford and Jeffries[11] have performed linewidth measurements for temperatures in the range $0.17 \leq T \leq 4.2$ K, and it is primarily their data which we use for comparison with theory.

Thus, $K_2CuCl_4 \cdot 2H_2O$ is well suited for comparison of theory with experiment. It is a ferromagnetic insulator, so that the localized Hamiltonian on which our calculations are based would appear to be appropriate. Further, it is a spin-$\frac{1}{2}$ substance with predominantly nearest-neighbor exchange interactions and a cubic lattice structure, facts that greatly simplify numerical calculations. Finally, linewidth measurements are available for temperatures ranging from well below to well above T_C.

The measurements of Ford and Jeffries were performed in a field of approximately 3.13 kOe, so that, owing to the low Curie temperature of the potassium salt, the Zeeman and exchange energies are of the same order of magnitude. Consequently, Eqs. (7) and (8) were used to calculate the linewidth.

Figure 2 exhibits temperature dependence of the second moment, calculated in the constant-coupling approximation for H_0 along a cubic axis. There is a strong temperature dependence in the Curie region, and we note that the components do not display an identical temperature dependence.

Of particular interest is the behavior of the $M = +1$ component, which does not go to zero as $T \rightarrow 0$. This fact had previously been noted by Keffer.[30] It has the consequence of producing a finite linewidth of considerable magnitude at zero temperature, provided that the basic assumption of Eq. (6) is valid regarding moments and linewidth. We believe, however, that this width is greatly reduced if a more realistic model based in spin waves is invoked at low temperature (see Appendix D). In fact, the finite $M_2^{(1)}$ makes *no* contribution to ΔH at $T = 0$ for the conditions of applied field treated here. Short-wavelength spin waves can exist[31] even above T_C and thus may reduce the importance of the $M = +1$ term over a fairly broad temperature range. Because of this and because it is not possible to obtain reasonable agreement with experiment if the $M = +1$ component is retained in the form shown in Fig. 2, we have chosen to drop it altogether.

Thus, the linewidth is calculated including only the $M = 0$, -1, and -2 terms (the $M = +2$ term is identically zero). This will give a correct temperature dependence, provided that the $M = +1$ part, when properly modified for spin-wave effects, shows the same general trend as the other contributions.

The fact that $\sum_j F_{ij}^{(M)} = 0$ for cubic symmetry makes the $M = -2$ and $M = 0$ terms [Eqs. (25) and (28)] equal to zero at $T = 0$. This also makes the second term on the right-hand side of Eq. (27) equal to zero at zero temperature. The further relation $C_{jk}^{-+} = 0$ at $T = 0$ makes the first term in Eq. (27) zero at $T = 0$, so that the total $M = -1$ term also vanishes at $T = 0$.

The fourth moments $M_4^{(M)}$, as shown in Fig. 3, exhibit the same general dependence as the second moments. The exchange frequency $\omega_e^{(M)}$ is obtained from $M_4^{(M)}$ and $M_2^{(M)}$ by Eqs. (6), (9), and (10), and is given in Fig. 4. Shown are the $M = -1$ and $M = -2$ components; the temperature dependence of the $M = 0$ and $M = +1$ components is similar to that of the $M = -2$ curve. We note that ω_e decreases

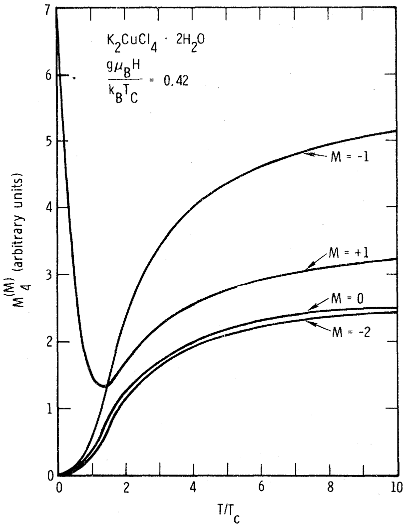

FIG. 3. Temperature variation of fourth moment $M_4^{(M)}$ for $K_2CuCl_4 \cdot 2H_2O$, $H = 3.13$ kOe, $T_C = 1.1$ K.

with decreasing temperature well above T_C. This is in agreement with the results of Goldsborough, Mandel, and Pake,[32] who have suggested that as the temperature is lowered, the effective exchange frequency is decreased. But the calculated exchange frequency increases sharply with decreasing temperature near T_C. This dependence agrees with the calculations of Richards,[33] who has examined ω_e in the special cases of very high applied fields and linear chain substances.

In Fig. 5 we compare the linewidth calculated from Eq. (8) (with the $M = +1$ term excluded) with the data of Ford and Jeffries at 4.2 K and below and with the datum of Kennedy, Choh, and Seidel[34] at 77 K. Two different procedures have been used. In the solid curve we have normalized to the total observed linewidth ΔH_{meas} at 4.2 K. In this way satisfactory agreement for the temperature dependence is obtained all the way from the high-temperature limit down to about $0.8T_C$. The following remarks are in order regarding the high-temperature point. The measurement of Kennedy *et al.* was taken with H_0 along the tetragonal c axis, where the g value is $g_{\parallel} = 2.06$, whereas the data of Ford and Jeffries are for H_0 perpendicular to the c axis and have $g_{\perp} = 2.22$. Since the dipolar linewidth in magnetic field units is proportional to g^3, we have multiplied the width of Kennedy *et al.*. by $(g_{\perp}/g_{\parallel})^3 = 1.28$ to bring it more into accord with a plausible extrapolation of the Ford–Jeffries data to high temperature. Room-temperature linewidths of between 115 and 140 Oe have been reported by Abe *et al.*,[35] Henderson and Rogers, and Kennedy *et al.* The large difference in ΔH between 77 K and room temperature is apparently due to a large change[34,36] in exchange constant J with temperature.

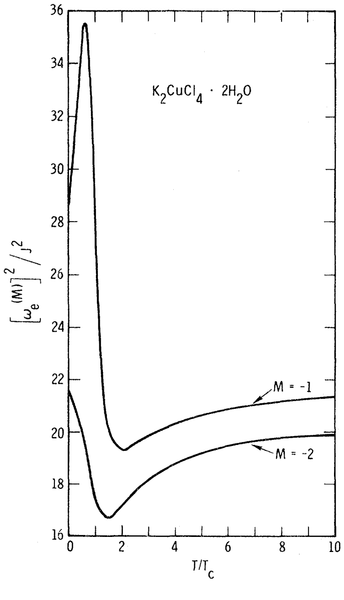

FIG. 4. Temperature variation of exchange frequency $\omega_e^{(M)}$ for $K_2CuCl_4 \cdot 2H_2O$. Curves are obtained from Figs. 2 and 3 through the relation $[\omega_e^{(M)}]^2 = M_4^{(M)}/M_2^{(M)}$. The curves for $M = 0$ and $M = +1$ are similar to the one for $M = -2$, and are not shown.

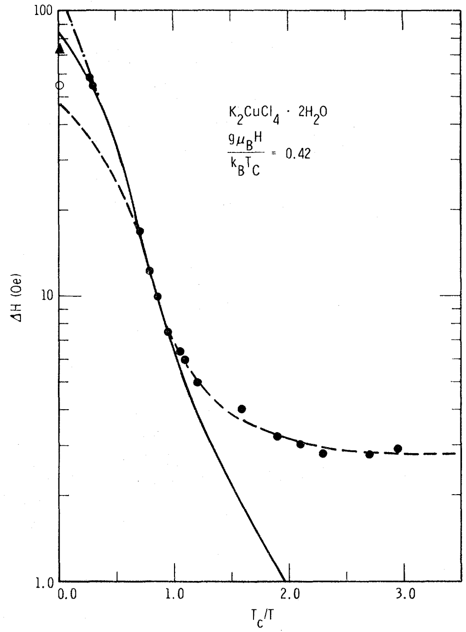

FIG. 5. Linewidth ΔH in $K_2CuCl_4 \cdot 2H_2O$. Solid circles are data of Ford and Jeffries at 9.5 GHz ($H \approx 3.13$ kOe). Open circle is 77-K point of Kennedy, Choh, and Seidel at about 9 GHz. Triangle is correction for different g factors, as explained in the text. Solid curve is theoretical ΔH normalized to data at 4.2 K ($T_C/T = 0.26$). Dashed curve is theoretical $\Delta H + \Delta H_0$ with the residual linewidth $\Delta H_0 = 2.8$ Oe normalized to data at 1.0 K ($T_C/T = 0.9$). Dash-dot curve for $T_C/T < 0.26$ is fit used by Ford and Jeffries, $\Delta H = \Delta H_0 + 145\, e^{-37/T}$. It is practically indistinguishable from the solid curve for $0.26 < T_C/T < 0.9$ and from the dashed curve for $T_C/T > 0.9$.

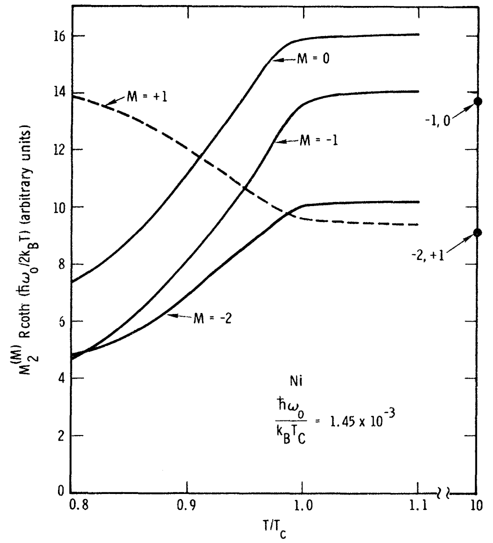

FIG. 6. Temperature variation of second-moment numerator $M_2^{(M)}$ R coth$(\hbar\omega_0/2k_BT)$ in Ni for a field of 6 kOe. Note break in scale above $1.1T_C$.

Thus we do not consider it meaningful to include room-temperature points on the graph. The exchange interaction is believed[34] to stay constant below 77 K.

The reason for the normalization procedure is that the moment calculations seem to underestimate the infinite-temperature linewidth by about a factor of 2, [15,37] so exact numerical agreement is not possible here. More sophisticated use of moments,[37] including M_6, can possibly account for the discrepancy. Also, at least for Cu^{++}, nondipolar interactions such as anisotropic exchange may contribute to line broadening. Our principal effort is directed toward temperature dependence, and this is independent of the normalization procedure.

In the dashed curve we emphasize the region below T_C by considering the temperature-dependent part of the linewidth,

$$\Delta H(T) = \Delta H_{\text{meas}} - \Delta H_0 ,$$

where $\Delta H_0 = 2.8$ Oe is the residual temperature-independent linewidth. At very low temperature, surface imperfections and other inhomogeneities[38] can give rise to a temperature-independent term which dominates the linewidth. These, of course, have not been accounted for in our calculations. As mentioned previously and explained in Appendix D, the $M = +1$ term should not contribute to ΔH_0, since the simple Gaussian approximations are not suitable for $T \rightarrow 0$. We have thus chosen simply to consider the temperature-dependent part by excluding the $M = +1$ component, so that the theoretical linewidth goes to zero at $T = 0$. The theoretical ΔH is normalized to the experimental $\Delta H(T)$ at T_C, and we thus find good agreement for the temperature dependence below T_C.

The other curve of Fig. 5 is $\Delta H(T) = 145e^{-3.7/T}$ Oe, which Ford and Jeffries used to obtain a satisfactory fit to the data. No theoretical justification was given by them for this empirical relation, and we see that below T_C it is essentially the same as our computed curve.

The temperature dependence of ΔH as given by Eq. (1) may be considered as arising from two sources: the denominator $\langle M_x^2 \rangle \propto \langle S_1^2 \rangle$ and the numerator which contains the dynamic correlation functions. Since we find that $\langle S_1^2 \rangle$ as given by Eq. (37) decreases by only a factor of 3.5 between T_C and $10T_C$, it is evident from Fig. 5 that a large part of the temperature dependence of ΔH comes from the numerator.

B. Ni

The second substance investigated in this paper is Ni, which has the fcc structure and is described in a localized model by spin $\frac{1}{2}$. Cooper and Keffer[39] have shown that the unusually large linewidth in Ni can be explained by using nearest-neighbor pseudodipolar rather than the dipolar coupling as the perturbing Hamiltonian. Even in an applied field of the order of 10 kOe, we have $\omega_e^{(M)} \gg \omega_0$, so that the Gaussian factors in Eq. (8) can be replaced by unity; i.e., we have the full "$\frac{10}{3}$ effect."[1,4]

In contrast to the situation for $K_2CuCl_4 \cdot 2H_2O$, the temperature dependence of the moments, and thus the linewidth, is completely dominated by the denominator $\langle M_x^2 \rangle$ of Eq. (1) in the region of interest. This is illustrated in Figs. 6, 7, and 8, which show, respectively, the second-moment numerator, exchange frequencies, and linewidth over a broad range of temperature. Calculations

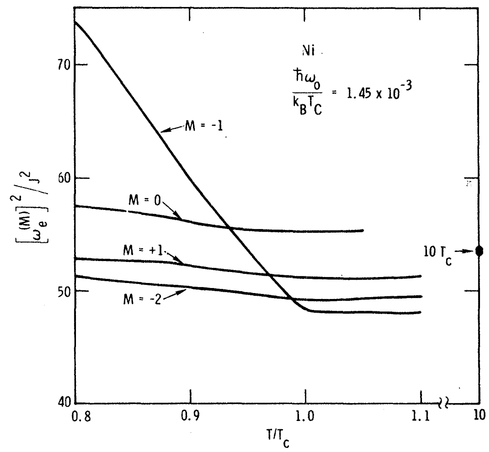

FIG. 7. Temperature variation of exchange frequency $\omega_e^{(M)}$ in Ni for a field of 6 kOe. Note break in scale above $1.1T_C$.

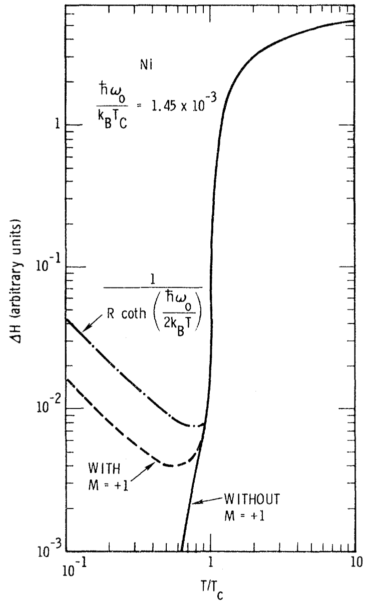

FIG. 8. Theoretical temperature dependence of linewidth ΔH in Ni for a field of 6 kOe. Solid curve: ΔH with $M=+1$ term excluded. Dashed curve: complete ΔH, including $M=+1$ term. Dot-dash curve: temperature dependence of $1/\langle M_x^2\rangle \propto 1/R\ \coth(\hbar\omega_0/2k_BT)$. All curves are normalized to same value at $10T_C$ and are practically indistinguishable above $0.9T_C$.

are for pseudodipolar coupling and H_0 along a cube axis. It is clear that above $0.9T_C$ the width is insensitive to the dynamic correlation numerator of Eq. (1).

In Fig. 9 the calculated linewidth is compared with the data of Bhagat and Chicklis[12] and of Salamon.[13] To display the temperature dependence we plot, similar to Fig. 5, $(\Delta H_{\text{meas}} - \Delta H_0)/\Delta H_{\text{meas}}(T_C)$ for the three sets of data, where ΔH_0 is the residual temperature-independent part and ΔH_{meas} is the total observed width. For the Salamon data, which do not extend below $0.99T_C$, we have taken $\Delta H_0 = 300$ Oe, the same value as found by Bhagat and Chicklis at a comparable frequency. The theoretical curve is normalized to the data at T_C. Once again, we have excluded the $M=+1$ term so that the theoretical ΔH is zero at $T=0$.

The calculations in Fig. 9 were made for a constant applied field of 6 kOe. The experimental fields for resonance were between 5 and 9.5 kOe, depending both on frequency and on temperature. Computations were also made at 8 kOe, and there was negligible difference between these results and the 6-kOe ones for the temperature dependence. We therefore do not expect the temperature- and frequency-dependent field required for resonance to be an important factor in comparing our calculations with experiment (see Appendix B also).

Between about $0.9T_C$ and $1.1T_C$ the data are adequately described by our calculations, which are indistinguishable from the simple dependence $\Delta H \propto 1/RT$ [since here $\coth(\frac{1}{2}\hbar\omega_0/k_BT) \approx 2k_BT/\hbar\omega_0$]. This relation had been noted by Salamon. Above $1.1T_C$, ΔH does not increase with temperature as rapidly as $1/RT$, and our theory is unable to account for this.

V. SUMMARY AND CONCLUSIONS

The principal purpose of this paper has been to investigate and make explicit calculations of the temperature dependence of the exchange-narrowed magnetic-resonance linewidth for realistic values of applied field. This has been done using the method of moments derived from the general theory of Kubo and Tomita as extended to finite temperatures in I.

The temperature dependence of the moments was calculated in essentially two steps. First, the four- and six-spin correlation functions which occur in the second and fourth moments, respectively, were decoupled into sums of products of two-spin correlation functions. These two-spin thermal

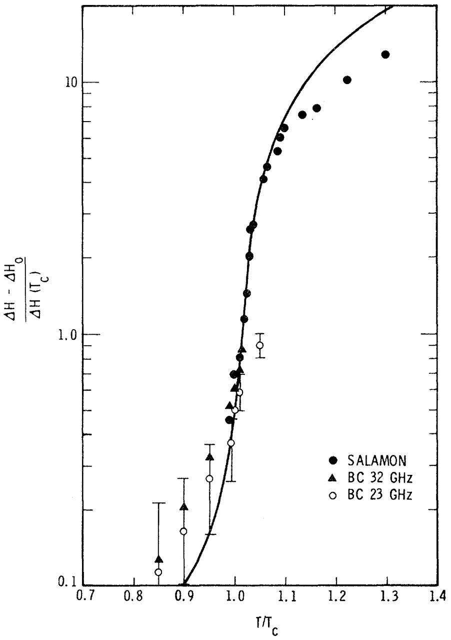

FIG. 9. Linewidth ΔH in Ni. Data are for temperature-dependent part $\Delta H - \Delta H_0$ normalized at T_C. Open circles, Bhagat and Chicklis (BC) data at 23 GHz, $\Delta H_0 = 300$ Oe, as determined from their data below $0.8T_C$. Triangles, BC data at 32 GHz, $\Delta H_0 = 450$ Oe, as determined from their data below $0.8T_C$. Solid circles, Salamon data at 23 GHz with $\Delta H_0 = 300$ Oe, as for BC at this frequency. Solid curve is same as in Fig. 8, normalized to $(\Delta H - \Delta H_0)/\Delta H(T_C) = 0.5$ at T_C. Error bars are placed on the BC 23-GHz data to indicate the approximately ± 50-Oe scatter in their data.

averages were then computed in the constant-coupling approximation.

We have found reasonable agreement for two ferromagnets, $K_2CuCl_4 \cdot 2H_2O$ and Ni, which satisfy the conditions $g\mu_B H_0/k_B T_C \sim 1$ and $g\mu_B H_0/k_B T_C \ll 1$, respectively.

The former case is the more significant test of our methods, since for $g\mu_B H_0/k_B T_C \sim 1$ we find a significant temperature dependence of the numerator in Eq. (1) which involves the specific dynamic correlations. Thus the semiquantitative agreement found with the Ford–Jeffries data is gratifying. To achieve this agreement, it was necessary to eliminate the $M=+1$ term. The deletion is, however, reasonable, since this component gives rise to a finite zero-temperature width in the Gaussian approximation, whereas a correct treatment (see Appendix D) shows that it cannot produce broadening at $T=0$ for the materials under consideration.

For $g\mu_B H_0/k_B T_C \ll 1$ as in Ni, our method of calculation gives a numerator in Eq. (1) which is essentially temperature independent in the critical region. Thus the temperature variation of ΔH is governed strictly by the denominator, proportional to RT (R is the relative magnetization). Experiment indicates this to be the case between about $0.9T_C$ and $1.1T_C$, so that we do have good agreement in this sense. Between $1.1T_C$ and $1.3T_C$ the numerator of Eq. (1) appears to be decreasing significantly with temperature, as pointed out originally by Salamon. It is doubtful that a more correct treatment of the localized Heisenberg Hamiltonian with temperature-independent parameters can account for this. The reason is that we should expect the numerator of Eq. (1) to have its strongest temperature dependence in the immediate vicinity of T_C. Since Salamon's data are consistent with its being temperature independent between T_C and $1.1T_C$, it is difficult to see how any such theory can have the numerator vary strongly between $1.1T_C$ and $1.3T_C$ without destroying the agreement between T_C and $1.1T_C$.

The reason for the success of the constant-coupling approximation has to do with the dominance of nearest-neighbor correlations. As explained in Sec. III, even in the case of Ni the finite field greatly reduces the correlation length so that only relatively short-range fluctuations exist at T_C. Thus, a theory which makes a reasonable estimate of nearest-neighbor correlations in the presence of an applied field is likely to be satisfactory. We also tried the RPA [Eq. (33) and Refs. 20 and 21] for calculating the correlations and obtained unreasonable answers—such as a negative fourth moment near T_C. This unphysical result is consistent with the observation by Liu and Siano[40] that the RPA leads to a negative specific heat at T_C for $S=\frac{1}{2}$.

Care was taken to display the temperature dependence of the secular and nonsecular components of the second and fourth moments. It has sometimes been assumed that the truncated and untruncated linewidths have the same temperature dependence. Our explicit calculations show that this is not the case.

Since decoupling of many-spin correlation functions and the constant-coupling approximation have been used, it is clear that our results do not have the correct critical exponents.[25] However, with the appreciable applied field H_0 required for magnetic resonance, the exact power law in the limit $H_0 \to 0$ is perhaps irrelevant, at least for the purpose of explaining existing data. Magnetization data for Ni shown in Fig. 1 support this contention.

Our general conclusion is that the calculated temperature-dependent linewidths do agree with experiment at least semiquantitatively in all cases considered. This gives support to the use of moments and the simple decoupling approximations employed, except at temperatures well below T_C where an obvious failure is the bogus contribution of the $M=+1$ term.

The linewidth formalism presented in this paper is general enough to be of use in future investigation. In particular, it can be extended to ferromagnetic substances with other than spin $\frac{1}{2}$ as well as to the antiferromagnetic case.

Finally, we emphasize the importance of including the applied field in calculations of the ferromagnetic-resonance linewidth. With $H_0=0$, theory predicts $\Delta H=0$ at T_C, since the denominator $\langle S_1^2\rangle$ diverges at the critical temperature. It is the explicit retention of H_0 in all the expressions which has enabled us to give what is to our knowledge the first treatment which can be applied to realistic situations. For antiferromagnets, however, H_0 is not likely to play so important a role, since the critical fluctuations are at zone-boundary wave vectors which do not couple directly to a uniform applied field.

ACKNOWLEDGMENTS

We are grateful to Professor N. C. Ford, Jr. for supplying a copy of the linewidth measurements in $K_2CuFl_4 \cdot 2H_2O$. The numerical calculations were carried out with facilities of the University of Kansas and University of Kentucky Computing Centers.

APPENDIX A: RELAXATION FUNCTIONS

In Ref. 7 the relaxation rate was calculated from consideration of the function

$$F(t) = \langle M_x [M_x(t) + M_x(-t)]\rangle / 2\langle M_x^2\rangle . \qquad (A1)$$

Kubo and Tomita,[1] on the other hand, based their theory on the relaxation function

$$\Phi(t)=\frac{\int_0^\beta d\lambda\,\langle e^{\lambda\mathcal{H}}M_xe^{-\lambda\mathcal{H}}[M_x(t)+M_x(-t)]\rangle}{2\int_0^\beta d\lambda\,\langle e^{\lambda\mathcal{H}}M_xe^{-\lambda\mathcal{H}}M_x\rangle}\quad , \qquad \text{(A2)}$$

which has a simple physical interpretation in that it describes decay of M_x after a steady field in the x direction is suddenly switched off.

In the infinite-temperature limit $F(t)$ and $\Phi(t)$ clearly are equal. We show here that they are equal at arbitrary temperatures provided that the perturbation $\mathcal{H}'$ is neglected in taking thermal averages. Since this approximation is nearly always made in linewidth calculations, it is useful to know that with it the integrals appearing in $\Phi(t)$ present no complication.

The proof is as follows. The unperturbed Hamiltonian is

$$\mathcal{H}_0=\mathcal{H}_{\text{ex}}-\hbar\omega_0S_z\ , \qquad \text{(A3)}$$

where S_z is the total z component of spin, $\hbar\omega_0$ is the Zeeman splitting, and $\mathcal{H}_{\text{ex}}$ is the isotropic-exchange Hamiltonian. We assume that even in the presence of $\mathcal{H}'$ there is symmetry between x and y, so that averages such as $\langle M\pm M\pm(t)\rangle$ are zero. Then (A1) and (A2) become

$$F(t)=\frac{\langle M_+[M_-(t)+M_-(-t)]+M_-[M_+(t)+M_+(-t)]\rangle}{2\langle M_+M_-+M_-M_+\rangle}\ , \qquad \text{(A4)}$$

$$\begin{aligned}\Phi(t)=\int_0^\beta d\lambda\,\langle e^{\lambda\mathcal{H}}M_+e^{-\lambda\mathcal{H}}[M_-(t)+M_-(-t)]\\ +e^{\lambda\mathcal{H}}M_-e^{-\lambda\mathcal{H}}[M_+(t)+M_+(-t)]\rangle\\ \times\Big[2\int_0^\beta d\lambda\langle e^{\lambda\mathcal{H}}M_+e^{-\lambda\mathcal{H}}M_-+e^{\lambda\mathcal{H}}M_-e^{-\lambda\mathcal{H}}M_+\rangle\Big]^{-1}.\end{aligned} \qquad \text{(A5)}$$

Now $\mathcal{H}_{\text{ex}}$ commutes with S_z and $M_\pm$, and we have

$$e^{-\lambda\hbar\omega_0S_z}M_\pm=e^{\pm\lambda\hbar\omega_0}M_\pm e^{-\lambda\hbar\omega_0S_z}\ . \qquad \text{(A6)}$$

Thus if $\mathcal{H}$ is replaced by $\mathcal{H}_0$ in the $e^{\pm\lambda\mathcal{H}}$ factors, the integrations over $d\lambda$ can readily be performed. This is of course legitimate if $\mathcal{H}'$ is neglected in the thermal averages, since $\lambda\leq\beta$. Furthermore, we have

$$\langle M_\pm\mathcal{O}\rangle=e^{\pm\beta\hbar\omega_0}\langle\mathcal{O}M_\pm\rangle \qquad \text{(A7)}$$

for any operator $\mathcal{O}$, since the unperturbed density matrix

$$\rho_0=e^{-\beta\mathcal{H}_0}/\text{Tr}\,\rho_0 \qquad \text{(A8)}$$

is used to calculate thermal averages.

With (A6) and (A7) it follows that

$$\begin{aligned}\int_0^\beta d\lambda\,\langle e^{\lambda\mathcal{H}}M_+e^{-\lambda\mathcal{H}}\mathcal{O}+e^{\lambda\mathcal{H}}M_-e^{-\lambda\mathcal{H}}\mathcal{O}\rangle\\ =[(1-e^{-\beta\hbar\omega_0})/\hbar\omega_0]\langle M_+\mathcal{O}+\mathcal{O}M_-\rangle\\ =[(e^{\beta\hbar\omega_0}-1)/\hbar\omega_0]\langle\mathcal{O}M_++M_-\mathcal{O}\rangle\ .\end{aligned} \qquad \text{(A9)}$$

Thus the factors resulting from integration can be made common to both the numerator and denominator of (A5), so that

$$\Phi=\frac{\langle M_+[M_-(t)+M_-(-t)]+[M_+(t)+M_+(-t)]M_-\rangle}{4\langle M_+M_-\rangle}\ . \qquad \text{(A10)}$$

Then, through use of (A7) together with the fact that

$$\langle A(t)B\rangle=\langle AB(-t)\rangle$$

for any operators A and B, we can put (A4) into the form (A10) as well, and hence produce the desired result

$$F(t)=\Phi(t)\ . \qquad \text{(A11)}$$

APPENDIX B: EFFECT OF SAMPLE SHAPE

Demagnetizing fields cause a shift in frequency at finite temperature for other than spherical samples. This shift is of the order of $4\pi\langle M_z\rangle g\mu_B/\hbar$ and is not necessarily small compared with $\omega_0=g\mu_BH_0/\hbar$. Our calculations have tacitly assumed an infinite medium or spherical sample, so that this shift does not occur. Since the experiments on Ni were performed on cylinders and disks, it is necessary to investigate the possible effects of sample shape on the linewidth. We show below that there is likely to be no effect, and thus our results can safely be applied to nonspherical samples.

For simplicity we consider samples with equal transverse demagnetizing factors. Then we have $\sum_j F_{ij}^{(M\neq 0)}=0$. However, $\sum_j F_{ij}^{(0)}$ is nonzero and given by

$$\sum_j F_{ij}^{(0)}=D\ . \qquad \text{(B1)}$$

This gives rise to a frequency shift which cannot be treated as a small perturbation. The quantity D is proportional to $N_z-\frac{1}{3}$, where N_z is the demagnetizing factor in the z direction and $N_z=\frac{1}{3}$ for a sphere. We redefine $\mathcal{H}_0$ as

$$\mathcal{H}_0=\mathcal{H}_{\text{ex}}-\hbar\omega_0S_z+2D\langle S_z\rangle S_z=\mathcal{H}_{\text{ex}}-h\tilde{\omega}_0S_z\ , \qquad \text{(B2)}$$

where $\tilde{\omega}_0=\omega_0-2D\langle S_z\rangle/\hbar$ is the new unperturbed resonance frequency and corresponds to what would be calculated using the Kittel ferromagnetic-resonance formula.[41]

The dipolar perturbation then becomes

$$\mathcal{H}'=\mathcal{H}_{\text{dip}}-2D\langle S_z\rangle S_z\ , \qquad \text{(B3)}$$

and correspondingly g_0 [Eqs. (2b) and (17c)] is redefined as

$$g_0'=\sum_{ij}F_{ij}^{(0)}(S_i^+S_j^z-N^{-1}\langle S_z\rangle S_+)\ , \qquad \text{(B4)}$$

where N is the total number of spins.

The quantities $g_{M\neq 0}$ are unaffected. The relaxation rate would then be calculated using (1) with ω_0 replaced by $\tilde{\omega}_0$ and g_0 replaced by g_0'. We now demonstrate that the second moment $\langle g_0'g_0'^\dagger\rangle$ is the same as $\langle g_0g_0^\dagger\rangle$.

We have for spin $\frac{1}{2}$

$$\langle g_0' g_0'^{\dagger} \rangle = \sum_{ijkl} F_{ij}^{(0)} F_{kl}^{(0)} \langle S_i^+ S_j^z S_k^z S_l^- \rangle + \tfrac{1}{4}(DR)^2 \langle S_+ S_- \rangle$$
$$- \tfrac{1}{2} DR \Bigl(\sum_{ij} F_{ij}^{(0)} (\langle S_i^+ S_j^z S_- \rangle + \langle S_+ S_i^z S_j^- \rangle) \Bigr), \qquad \text{(B5)}$$

where

$$R = 2N^{-1} \langle S_z \rangle \qquad \text{(B6)}$$

is the relative magnetization for spin $\frac{1}{2}$. When this is decoupled as in Sec. III, it reduces to

$$\langle g_0' g_0'^{\dagger} \rangle = \sum_{ijkl} F_{ij}^{(0)} F_{kl}^{(0)} \langle S_i^+ S_l^- \rangle \langle S_j^z S_k^z \rangle$$
$$- \tfrac{1}{4}(DR)^2 \langle S_+ S_- \rangle . \qquad \text{(B7)}$$

Let

$$h_{jk} = \langle S_j^z S_k^z \rangle - \langle S_j^z \rangle \langle S_k^z \rangle = \langle S_j^z S_k^z \rangle - \tfrac{1}{4} R^2 , \qquad \text{(B8)}$$

so that

$$\langle g_0' g_0'^{\dagger} \rangle = \sum_{ijk} F_{ij}^{(0)} F_{kl}^{(0)} \langle S_i^+ S_l^- \rangle h_{jk} . \qquad \text{(B9)}$$

In this equation both $\langle S_i^+ S_l^- \rangle$ and h_{jk} are nonvanishing only over distances which are small compared to the sample dimensions. That is, the correlation length κ^{-1} is much less than the macroscopic sample size. We therefore conclude that the summation in (B9) converges well before the sample boundaries are reached, since the summand goes essentially as r^{-6} owing to the restriction imposed by $\langle S_i^+ S_l^- \rangle h_{jk}$ of keeping r_{ij} and r_{kl} coupled together. This implies that $\langle g_0' g_0'^{\dagger} \rangle$ is independent of sample shape, and hence

$$\langle g_0' g_0'^{\dagger} \rangle = \langle g_0 g_0^{\dagger} \rangle , \qquad \text{(B10)}$$

the value for $D = 0$. A similar result is expected to hold for the fourth moment.

Thus, any effect of sample shape on linewidth can probably be accounted for by the trivial operations of replacing ω_0 by $\tilde{\omega}_0$ and using the Lorentz field $H_l = H_0 + 4\pi(\frac{1}{3} - N_z)\langle M_z \rangle$ for calculation of thermal averages, i.e., using (B2) in the unperturbed density matrix. Since experiments are performed at constant frequency, we may assume that H_l does not vary with temperature, so by calculating temperature dependence at constant field we have actually accounted for the effect.

In the above we have used the simplification discussed in Sec. III of decoupling for equal as well as unequal indices. For cubic symmetry, however, the result (B10) may be shown to hold equally well if decoupling is made only for unequal indices.

APPENDIX C: DERIVATION OF EQUATION (37)

Since S_+ and S_- are proportional to M_+ and M_-, respectively, we may use Eq. (A7) with $\Theta = S_-$ to show that

$$\langle S_+ S_- \rangle = e^{\beta \hbar \omega_0} \langle S_- S_+ \rangle . \qquad \text{(C1)}$$

From the commutation relation $[S_+, S_-] = 2S_z$, we also have

$$\langle S_+ S_- \rangle = \langle S_- S_+ \rangle + NR , \qquad \text{(C2)}$$

since $\langle S_z \rangle \equiv \frac{1}{2} NR$ for spin $\frac{1}{2}$.

By combining (C1) and (C2), the results

$$\langle S_+ S_- \rangle = \frac{NR}{1 - e^{-\beta \hbar \omega_0}} ,$$
$$\langle S_- S_+ \rangle = \frac{e^{-\beta \hbar \omega_0} NR}{1 - e^{-\beta \hbar \omega_0}} \qquad \text{(C3)}$$

are obtained, so that

$$\langle S_+ S_- \rangle + \langle S_- S_+ \rangle = NR \, \frac{1 + e^{-\beta \hbar \omega_0}}{1 - e^{-\beta \hbar \omega_0}}$$
$$= NR \coth \tfrac{1}{2} \beta \hbar \omega_0 , \qquad \text{(C4)}$$

which is equivalent to Eq. (37), since $\beta \hbar \omega_0 = g \mu_B H_0 / k_B T$.

APPENDIX D: $M = +1$ TERM AT LOW TEMPERATURE

Equation (18d) and Fig. 3 show that the second moment associated with the $M = +1$ change in the Zeeman quantum number is nonzero at $T = 0$. This would imply a sizable combination to the zero-temperature linewidth as originally suggested by Keffer.[30] We examine the question in detail here and show that this term really does not produce a $T = 0$ linewidth for conditions appropriate to the reported measurements in $K_2CuCl_4 \cdot 2H_2O$ and in Ni.

The correct expression for the relaxation rate η is given by Eq. (1), from which we have

$$\eta^{(1)} = (2\hbar \langle S_x^2 \rangle)^{-1} \sum_{ijkl} F_{ij}^{(1)} F_{kl}^{(-1)}$$
$$\times \int_{-\infty}^{\infty} d\tau [e^{i\omega_0 \tau} \langle S_i^-(-\tau) S_j^-(-\tau) S_k^+ S_l^- \rangle$$
$$+ e^{-\omega_0 \tau} \langle S_i^+(-\tau) S_j^+(-\tau) S_k^- S_l^- \rangle] \qquad \text{(D1)}$$

upon using Eq. (17d) in (1). Here $\eta^{(1)}$ is the $M = +1$ part of η and

$$S_i^+(\tau) = e^{i \mathcal{H}_{\mathrm{ex}} \tau / \hbar} S_i^+ e^{-i \mathcal{H}_{\mathrm{ex}} \tau / \hbar} . \qquad \text{(D2)}$$

At very low temperatures we replace the transverse components by magnon operators in the usual way[42]:

$$S_i^+ = \left(\frac{2S}{N} \right)^{1/2} \sum_k a_k e^{i \vec{k} \cdot \vec{r}_i} , \qquad \text{(D3)}$$

$$S_i^- = \left(\frac{2S}{N} \right)^{1/2} \sum_k a_k^{\dagger} e^{-i \vec{k} \cdot \vec{r}_i} , \qquad \text{(D4)}$$

where a_k and $a_k^{\dagger}$ are, respectively, annihilation and creation operators for a magnon of wave vector k. Thus,

$$\langle a_k^{\dagger} a_k \rangle = n_k , \quad \langle a_k a_k^{\dagger} \rangle = n_k + 1 ,$$

where n_k is the magnon occupation number. It is important to note at this point that the association of S_i^- with a magnon *creation* operator implies that the ground state has spins aligned in the $+z$ direction, and this is consistent with ω_0 being a positive quantity in Eq. (D1). [See also Eq. (B2), where we explicitly show the Zeeman ground state to have $\vec{S}$ in the $+z$ direction.]

The time dependence of the magnon operators is given by

$$a_k^\dagger(\tau) = a_k^\dagger e^{i\omega_k\tau} , \tag{D5}$$

$$a_k(\tau) = a_k e^{-i\omega_k\tau} , \tag{D6}$$

where ω_k is the spin-wave frequency and $\omega_k \geq 0$. In the low-temperature limit where $n_k \ll 1$, Eq. (D1) then reduces to [note that the argument of the time correlations in (D1) is $-\tau$]

$$\eta^{(1)} = \frac{8\pi S^2}{\hbar\langle S_x^2\rangle} \sum_k |F_k^{(1)}|^2 \delta(\omega_0 - 2\omega_k) . \tag{D7}$$

The energy-conservation δ function in Eq. (D7) states that there must be magnons of frequency $\omega_k = \frac{1}{2}\omega_0$ available to provide relaxation of the uniform-mode ($k=0$) magnon. This will be recognized as the condition for splitting of the $k=0$ magnon into magnons of wave vectors $\vec{k}$ and $-\vec{k}$. Since the $M=+1$ component arises from the $S_i^+ S_j^+$ terms of the dipolar Hamiltonian, the above result is entirely consistent with the fact that these terms give rise to three-magnon interactions,[43] of which only the splitting can contribute to relaxation at zero temperature. Examination of the spin-wave manifold shows that, for a sphere as used by Ford and Jeffries in the $K_2CuCl_4 \cdot 2H_2O$ experiment, we must have[43]

$$H_0 \leq \tfrac{8}{3}\pi M_0 \quad \text{(sphere)} \tag{D8}$$

in order for spin waves to exist with ω_k as low as $\frac{1}{2}\omega_0$. Since the saturation magnetization M_0 is only[11] 46.5 Oe in $K_2CuCl_4 \cdot 2H_2O$, it is clearly impossible to satisfy Eq. (D8) for the applied field of 3 kOe. The experiments on Ni were carried out in a disk magnetized along its axis,[13] for which it is not possible to have $\omega_k = \frac{1}{2}\omega_0$ for any values of H_0 or M_0, and in a cylinder magnetized along its axis,[12] for which the requirement is

$$H_0 \leq 2\pi M_0 \quad \text{(cylinder)} . \tag{D9}$$

For Ni we have $2\pi M_0 = 3.2$ kOe, so it is still not possible to have $\omega_k = \frac{1}{2}\omega_0$ in the fields of 5 kOe and upwards as used.

We conclude therefore that although the $M=+1$ second moment is finite at zero temperature, there can be no $M=+1$ contribution to relaxation for the values of H_0 and M_0 considered here, since it is not possible to conserve energy for the splitting of a $k=0$ magnon into magnons $\vec{k}$ and $-\vec{k}$.

---

*Work supported by the U. S. Atomic Energy Commission. A preliminary account was reported in Bull. Am. Phys. Soc. 14, 194 (1969).

†Present address: Sandia Laboratories, Albuquerque, N. M. 87115.

[1] R. Kubo and K. Tomita, J. Phys. Soc. Japan 9, 888 (1954).

[2] H. Mori and K. Kawasaki, Progr. Theoret. Phys. (Kyoto) 27, 529 (1962).

[3] K. Kawasaki, Progr. Theoret. Phys. (Kyoto) 39, 285 (1968).

[4] D. L. Huber, J. Phys. Chem. Solids 32, 2145 (1971).

[5] J. H. Van Vleck, Phys. Rev. 74, 1168 (1948).

[6] P. W. Anderson and P. R. Weiss, Rev. Mod. Phys. 25, 269 (1953).

[7] P. M. Richards, Phys. Rev. 142, 189 (1966), referred to as I. Equation (1) here follows directly from Eqs. (44) and (48) of I. It differs slightly from Eq. (51) of I, which was written incorrectly.

[8] M. Tanaka and K. Tomita, Progr. Theoret. Phys. (Kyoto) 29, 528 (1963); 29, 651 (1963). These authors show (see Fig. 4 on p. 660) a linewidth which does not decrease rapidly as T_C is approached from above, even in the limit of zero applied field. This is in marked contrast to the experimental results and calculations presented here (see our Figs. 5 and 9). The present authors do not understand why the strong temperature dependence near T_C, which is characteristic of most if not all other formulations, is absent in the work of Tanaka and Tomita.

[9] P. W. Kasteleijn and J. van Kranendonk, Physica 22, 317 (1956).

[10] B. Strieb, H. B. Callen, and G. Horwitz, Phys. Rev. 130, 1798 (1963).

[11] N. C. Ford, Jr. and C. D. Jeffries, Phys. Rev. 141, 381 (1966).

[12] S. M. Bhagat and S. M. Chicklis, Phys. Rev. 178, 828 (1966).

[13] M. B. Salamon, Phys. Rev. 155, 224 (1967).

[14] K. Tomita, in *Proceedings of the International Conference on Theoretical Physics, Kyoto*, 1953 (Science Council of Japan, Tokyo, 1954), p. 800.

[15] A. J. Henderson, Jr. and R. N. Rogers, Phys. Rev. 152, 218 (1966).

[16] G. C. Wick, Phys. Rev. 80, 268 (1950).

[17] R. A. Tahir-Kheli and D. ter Haar, Phys. Rev. 127, 88 (1962).

[18] H. B. Callen, Phys. Rev. 130, 890 (1963).

[19] P. K. Leichner, Ph.D. thesis (University of Kansas, 1969) (unpublished).

[20] R. Brout, *Phase Transitions* (Benjamin, New York, 1965); in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic, New York, 1965), Vol. II A.

[21] R. B. Stinchcombe, G. Horwitz, F. Englert, and R. Brout, Phys. Rev. 130, 155 (1963).

[22] T. Oguchi, Progr. Theoret. Phys. (Kyoto) 13, 148 (1955).

[23] R. Peierls, Proc. Cambridge Phil. Soc. 32, 477 (1936).

[24] P. R. Weiss, Phys. Rev. 74, 1493 (1948).

[25] D. S. Ritchie and M. E. Fisher, Phys. Rev. B 5, 2668 (1972).

[26] P. Weiss and R. Forrer, Ann. Phys. (Paris) 5, 153

(1926).

[27] S. B. Hendricks and R. G. Dickinson, J. Am. Chem. Soc. 49, 2149 (1927).

[28] A. R. Miedema, H. Van Kempen, and W. J. Huiskamp, Physica 29, 1266 (1963).

[29] F. J. Dyson, Phys. Rev. 102, 1217 (1956); 102, 1230 (1966).

[30] F. R. Keffer, Phys. Rev. 88, 686 (1952).

[31] P. A. Fleury, Phys. Rev. 180, 591 (1969).

[32] J. P. Goldsborough, M. Mandel, and G. E. Pake, Phys. Rev. Letters 4, 13 (1960).

[33] P. M. Richards, Phys. Rev. 142, 196 (1966).

[34] T. A. Kennedy, S. H. Choh, and G. Seidel, Phys. Rev. B 2, 3645 (1970).

[35] H. Abe, K. Ono, I. Hayashi, J. Shimada, and K. Iwanaga, J. Phys. Soc. Japan 9, 814 (1954).

[36] R. N. Rogers (private communication).

[37] J. E. Gulley, D. Hone, D. J. Scalapino, and B. G. Silbernagel, Phys. Rev. B 1, 1020 (1970).

[38] M. Sparks, R. Loudon, and C. Kittel, Phys. Rev. 122, 791 (1961).

[39] B. R. Cooper and F. Keffer, Phys. Rev. 125, 896 (1962).

[40] S. H. Liu and D. B. Siano, Phys. Rev. 164, (1967).

[41] C. Kittel, Phys. Rev. 73, 155 (1948).

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

[43] See, for example, M. Sparks, *Ferromagnetic-Relaxation Theory* (McGraw-Hill, New York, 1964).