

## Critical Spin Relaxation in Anisotropic Ferromagnets\*

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The decay times of the spin correlation functions have been calculated for uniaxial anisotropic ferromagnets which order in the plane perpendicular to the axis. The self-consistent theory when applied to the hydrodynamic region just above the critical temperature leads to coupled nonlinear integral equations, which have been solved numerically. In addition to obtaining a temperature dependence in agreement with the dynamic scaling laws, our calculation yields absolute values of the decay times that are one or two orders of magnitude larger than in the conventional theory, which predicts a temperature dependence inversely proportional to the susceptibility. The dynamic scaling-law behavior is followed over a temperature range of about  $0.1(T_y - T_z)/T_y$ , where  $T_y$  and  $T_z$  are the perpendicular and parallel paramagnetic Curie temperatures, respectively. We have also made a comparison with experimental values of the decay rates in terbium. The agreement, while not precise, is an order of magnitude better than with the conventional theory.

### I. INTRODUCTION

IN recent years the dynamical behavior of magnetic systems in the neighborhood of the ordering temperature has received considerable attention.<sup>1,2</sup> Particular emphasis has been placed on the study of time dependence of the spin correlations. Halperin and Hohenberg<sup>3</sup> suggested that near the critical temperature,  $T_c$ , the dynamical behavior fell into three categories (or regions): spin waves (or hydrodynamic below  $T_c$ ), transition, and hydrodynamic above  $T_c$ . They indicated that there might be connections between the characteristic frequencies associated with the three regions. In analogy with the static properties, these connections were designated dynamical scaling laws. The nature of the dynamical scaling laws has been explored in detail by Kawasaki in a recent series of papers.<sup>4-7</sup> He has indicated criteria that must be met if the scaling laws are to hold. In particular, he found that it is necessary that the dynamics of certain "critical dynamical variables" which exhibit long decay times be asymptotically closed within themselves.<sup>6</sup> These conditions are met, for example, in isotropic ferromagnets and antiferromagnets and in uniaxial anisotropic ferromagnets which order in the plane perpendicular to the axis.

The experimental situation is not completely clear. There is evidence from neutron scattering work on iron<sup>8</sup> and nickel<sup>9</sup> that the dynamics in the spin wave and

transition regions are connected in the manner predicted by the dynamic scaling laws. However, the scaling laws appear to break down when applied to the hydrodynamic region above  $T_c$ . It has been suggested that this breakdown may reflect the metallic nature of the targets.<sup>8</sup> How this comes about is still unclear.

For the most part the theoretical studies that have been made of the spin dynamics in the critical region have focused on the temperature and wavelength dependence of the characteristic frequencies. Less attention has been paid to the magnitude of the frequencies. This is unfortunate because the neutron studies provide such information and any complete (albeit approximate) theory should be able to account for the magnitude as well as the temperature dependence.

We have recently made a comparison between theoretical and experimental values of the spin-diffusion constant of iron<sup>10</sup> using a self-consistent theory for isotropic ferromagnets first developed by Bennett and Martin<sup>11</sup> and later elaborated by Kawasaki.<sup>12</sup> The agreement, while not precise, is still one to two orders of magnitude better than with previous theories. In the present paper we will extend the self-consistent theory to anisotropic ferromagnets having an easy plane of magnetization.<sup>13</sup> We calculate the magnitudes of the decay rate of spin fluctuations in the hydrodynamic region above  $T_c$  as a function of temperature for various degrees of anisotropy. Although a detailed comparison with experiment is not possible as in the case of iron, we do compare our results with recent neutron-scattering data on terbium which were obtained by Als-Nielsen *et al.*,<sup>14</sup> following the suggestion of these authors that Tb may be treated in the first approximation as a uniaxial ferromagnet which orders in the basal plane.

<sup>10</sup> D. A. Krueger and D. L. Huber, *Solid State Commun.* **6**, 869 (1968).

<sup>11</sup> H. S. Bennett and P. C. Martin, *Phys. Rev.* **138**, A608 (1965).

<sup>12</sup> K. Kawasaki, *J. Phys. Chem. Solids* **28**, 1277 (1967).

<sup>13</sup> A preliminary report of this work has been published [D. L. Huber, *Phys. Letters* **28A**, 644 (1969)]. There is an important typographical error in this article. Equation (6) should read:  $\Gamma_z(q) = A_3 q^2 (r^1_c)^{1/2}$ .

<sup>14</sup> J. Als-Nielsen, O. W. Dietrich, W. Marshall, and P. A. Lindgard, *Solid State Commun.* **5**, 607 (1967).

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<sup>1</sup> P. Heller, *Rept. Progr. Phys.* **30**, 731 (1967).

<sup>2</sup> W. Marshall and R. D. Lowde, *Rept. Progr. Phys.* **31**, 705 (1968).

<sup>3</sup> B. I. Halperin and P. C. Hohenberg, *Phys. Rev. Letters* **19**, 700 (1967); *Phys. Rev.* **177**, 952 (1969).

<sup>4</sup> K. Kawasaki, *Progr. Theoret. Phys. (Kyoto)* **39**, 1133 (1968).

<sup>5</sup> K. Kawasaki, *Progr. Theoret. Phys. (Kyoto)* **40**, 11 (1968).

<sup>6</sup> K. Kawasaki, *Progr. Theoret. Phys. (Kyoto)* **40**, 706 (1968).

<sup>7</sup> K. Kawasaki, *Progr. Theoret. Phys. (Kyoto)* **40**, 930 (1968).

<sup>8</sup> M. F. Collins, V. J. Minkiewicz, R. Nathans, L. Passell, and G. Shirane, *Phys. Rev.* **179**, 417 (1969).

<sup>9</sup> V. J. Minkiewicz, M. F. Collins, R. Nathans, and G. Shirane, *Phys. Rev.* **182**, 624 (1969).

A final word concerns the spirit of this work. We do not claim that the self-consistent theory we develop is exact. It is our hope that it incorporates enough of the features of an exact theory so that its results compare favorably with experimental data. As indicated in Sec. IV, there is evidence that this is the case. More experimental work on systems which come close to approximating the model Hamiltonian is certainly called for. Until such work is available, however, the usefulness of the theory is still open to question.

In Sec. II we develop the formal aspects of the theory, while in Sec. III we report on numerical studies. We discuss our results in Sec. IV where we make brief comparison with the Tb data. In the Appendix, we outline a Green's-function analysis of the system with emphasis on properties near and below the ordering temperature.

## II. FORMULATION

We consider a system of  $N$  localized spins governed by a Hamiltonian

$$H = -\sum_{\alpha,\beta} \{J_{\alpha\beta}(S_{\alpha^z}S_{\beta^z} + S_{\alpha^y}S_{\beta^y}) + K_{\alpha\beta}S_{\alpha^z}S_{\beta^z}\}, \quad (1)$$

$$H = -\frac{1}{N} \sum_{\mathbf{q}} \left\{ \frac{1}{2} J(\mathbf{q})(S_{\mathbf{q}^+}S_{-\mathbf{q}^-} + S_{\mathbf{q}^-}S_{-\mathbf{q}^+}) + K(\mathbf{q})S_{\mathbf{q}^z}S_{-\mathbf{q}^z} \right\}, \quad (2)$$

where

$$S_{\mathbf{q}^\mu} = \sum_{\alpha} e^{i\mathbf{q}\cdot\alpha} S_{\alpha^\mu} = (S_{-\mathbf{q}^{-\mu}})^*, \quad \mu = z, \pm \quad (3)$$

$$S_{\alpha^\pm} = S_{\alpha^\pm} \pm iS_{\alpha^y},$$

and

$$J(\mathbf{q}) = \sum_{\alpha} J_{\alpha\beta} e^{i\mathbf{q}\cdot(\alpha-\beta)}. \quad (4)$$

The relaxation time for a disturbance of wave vector  $\mathbf{q}$  may be obtained from the normalized relaxation function

$$\Xi_{\mu}(\mathbf{q}, t) = \{S_{\mathbf{q}^\mu}(t), S_{\mathbf{q}^\mu}(0)^*\} / \{S_{\mathbf{q}^\mu}(0), S_{\mathbf{q}^\mu}(0)^*\}, \quad (5)$$

where  $\{A, B\} = \int_0^\beta d\lambda \langle e^{\lambda H} A e^{-\lambda H} B \rangle - \beta \langle A \rangle \langle B \rangle$  and  $\langle C \rangle = \text{Tr} e^{-\beta H} C / \text{Tr} e^{-\beta H}$ . Mori<sup>15</sup> has shown quite generally that the Laplace transform is given by

$$\Xi_{\mu}(\mathbf{q}, z) = \int_0^\infty dt e^{-zt} \Xi_{\mu}(\mathbf{q}, t) = [z + \phi_{\mu}(\mathbf{q}, z)]^{-1}, \quad (6)$$

where

$$\phi_{\mu}(\mathbf{q}, z) = \int_0^\infty dt e^{-zt} \{ \partial_t \hat{S}_{\mathbf{q}^\mu}(t), \partial_t \hat{S}_{\mathbf{q}^\mu}(0)^* \} / \{ S_{\mathbf{q}^\mu}(0), S_{\mathbf{q}^\mu}(0)^* \}. \quad (7)$$

As discussed in Ref. 15  $\partial_t \hat{S}(t)$  is obtained from  $\partial_t S(t) = i[H, S(t)]$  by removing that part of  $S$  which corresponds to uniform precessional motion. Above the Curie point this distinction disappears. If  $\phi_{\mu}(\mathbf{q}, z)$  is a

slowly varying function of  $z$ , then the long time behavior of  $\Xi(t)$  is an exponential decay.

$$\Xi_{\mu}(t) \approx \exp\{-\phi_{\mu}(\mathbf{q}, 0)|t|\}. \quad (8)$$

Thus the characteristic time is  $\phi_{\mu}(\mathbf{q}, 0)^{-1}$ . To calculate this, we return to Eq. (7) and use  $\hat{S}_{\mathbf{q}^\mu} = i[H, S_{\mathbf{q}^\mu}]$  to obtain ( $\hbar = 1$ )

$$\begin{aligned} \phi_{+}(\mathbf{q}, z) &= 2(N^2\beta)^{-1} \sum'_{\mathbf{k}, \mathbf{p}} [K(\mathbf{q}-\mathbf{k}) - J(\mathbf{k})] \\ &\times [K(\mathbf{q}-\mathbf{p}) - J(\mathbf{p})] \int_0^\infty dt e^{-tz} \\ &\times \{S_{\mathbf{k}^+}(t)S_{\mathbf{q}-\mathbf{k}^z}(t), S_{\mathbf{q}-\mathbf{p}^z}S_{\mathbf{p}^+}^*\} / \{S_{\mathbf{q}^+}, S_{\mathbf{q}^+}^*\}. \quad (9) \end{aligned}$$

A similar equation holds for  $\phi_z(\mathbf{q}, z)$ . The primes on the sums are to indicate that only small momenta should be included. This reflects the assumed dominance of the long-wavelength fluctuations near the critical temperature.<sup>16</sup>

In the spirit of the self-consistent approximation developed for the isotropic interaction,<sup>16</sup> we make the approximation

$$\begin{aligned} \{S_{\mathbf{k}^+}(t)S_{\mathbf{q}-\mathbf{k}^z}(t), S_{\mathbf{q}-\mathbf{p}^z}S_{\mathbf{p}^+}^*\} \\ \approx \exp\{-|t|[\phi_{+}(\mathbf{k}) + \phi_z(\mathbf{q}-\mathbf{k})]\} \\ \times \{S_{\mathbf{k}^+}S_{\mathbf{q}-\mathbf{k}^z}, S_{\mathbf{q}-\mathbf{p}^z}S_{\mathbf{p}^+}^*\}. \quad (10) \end{aligned}$$

For isotropic systems Kawasaki has shown that applying the random phase approximation to the equal time correlation-functions yields the same temperature dependence as would be predicted from an analysis based on the static scaling laws as long as  $\mathbf{q}$ ,  $\mathbf{k}$ , and  $\mathbf{p}$  refer to the long-wavelength component in the sense implied in Eq. (9).<sup>4</sup> In view of this we will apply the random phase approximation to the right-hand side of (10), thus obtaining the result

$$\begin{aligned} \{S_{\mathbf{k}^+}(t)S_{\mathbf{q}-\mathbf{k}^z}(t), S_{\mathbf{q}-\mathbf{p}^z}S_{\mathbf{p}^+}^*\} \\ \approx 2\beta^{-1}\delta_{\mathbf{k}, \mathbf{p}} \exp\{-|t|[\phi_{+}(\mathbf{k}) + \phi_z(\mathbf{q}-\mathbf{k})]\} \\ \times \{S_{\mathbf{k}^y}, S_{\mathbf{k}^y}^*\} \{S_{\mathbf{q}-\mathbf{k}^z}, S_{\mathbf{q}-\mathbf{k}^z}^*\}. \quad (11) \end{aligned}$$

Equation (11) is the basic approximation of our calculation. By making such an approximation we are able to reduce the calculation of the decay constants to the solution of a coupled set of nonlinear integral equations, as shown below.

As a further approximation we employ the Ornstein-Zernike form for the static susceptibility,  $\chi^{\nu}(\mathbf{k})$ ,

$$\begin{aligned} N\chi^{\nu}(\mathbf{k})/g^2\mu_B^2 &= \{S_{\mathbf{k}^{\nu}}, S_{\mathbf{k}^{\nu}}^*\} \\ &= NC_{\nu}/(\mathbf{k}^2 + \kappa_{\nu}^2), \quad \nu = (y, z) \quad (12) \end{aligned}$$

where  $C_{\nu}$  is independent of temperature and  $\kappa_{\nu}$  denotes the correlation wave vector. It should be noted that more exact functional forms for  $\{S_{\mathbf{k}^{\nu}}, S_{\mathbf{k}^{\nu}}^*\}$  could be employed (i.e.,  $NC_{\nu}'/(k^2 + \kappa_{\nu}^2)^{-1+\eta/2}$ ). However, in light of the approximations already made, such refinements appear unnecessary. Combining (9)–(12) yields two

<sup>15</sup> H. Mori, Progr. Theoret. Phys. (Kyoto) **33**, 423 (1965).

<sup>16</sup> K. Kawasaki, Progr. Theoret. Phys. (Kyoto) **39**, 285 (1968).

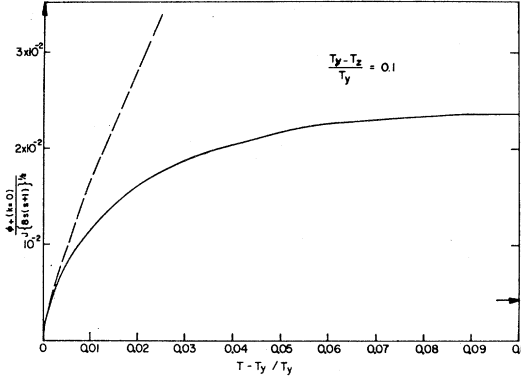


FIG. 1. Decay parameter for the component of spin in the easy plane of magnetization in the long-wavelength limit versus temperature. The solid curve was calculated from the self-consistent integral Eqs. (15) and (16). The broken curve is the extrapolation of the asymptotic forms given by Eqs. (25) and (26). An anisotropic exchange interaction has been used. The arrow indicates the value at infinite temperature obtained from a moment calculation, Eq. (32).

coupled, nonlinear integral equations for  $\phi_+(\mathbf{q}) = \phi_-(\mathbf{q})$  and  $\phi_z(\mathbf{q})$ :

$$\phi_+(\mathbf{q}) = \frac{4C_z}{N\beta} \times \sum_{\mathbf{k}}' \frac{[K(\mathbf{q}-\mathbf{k}) - J(\mathbf{k})]^2 (\kappa_y^2 + \mathbf{q}^2)}{[\phi_+(\mathbf{k}) + \phi_z(\mathbf{q}-\mathbf{k})] (\kappa_y^2 + \mathbf{k}^2) [\kappa_z^2 + (\mathbf{q}-\mathbf{k})^2]}, \quad (13)$$

$$\phi_z(\mathbf{q}) = \frac{4C_y^2}{C_z N \beta} \times \sum_{\mathbf{k}}' \frac{[J(\mathbf{q}-\mathbf{k}) - J(\mathbf{k})]^2 (\kappa_z^2 + \mathbf{q}^2)}{[\phi_+(\mathbf{k}) + \phi_+(\mathbf{q}-\mathbf{k})] (\kappa_y^2 + \mathbf{k}^2) [\kappa_y^2 + (\mathbf{q}-\mathbf{k})^2]}. \quad (14)$$

These equations determine  $\phi(\mathbf{q})$  if the static properties  $\kappa_y$ ,  $\kappa_z$ ,  $C_y$  and  $C_z$  are given. Since we are primarily interested in the time dependence, these static properties are assumed to be known. In the presence of anisotropy  $K(\mathbf{q}-\mathbf{k}) - J(\mathbf{k})$  does not vanish as  $\mathbf{q} \rightarrow 0$  so  $\phi_+(\mathbf{q}=0) \neq 0$ . However,  $J(\mathbf{q}-\mathbf{k}) - J(\mathbf{k})$  does vanish as  $\mathbf{q} \rightarrow 0$ , so  $\phi_z(\mathbf{q}=0) = 0$ . Thus  $\phi_z(\mathbf{q})$  exhibits kinematic slowing down (i.e.,  $\sim q^2$ ), whereas  $\phi_+(\mathbf{q})$  does not (i.e.,  $\sim \text{const}$ ). If, on the other hand, the anisotropy vanishes, then  $\phi_+(\mathbf{q})$  will have kinematic slowing down. We are interested in the long-wavelength fluctuations so we take  $\phi_z(\mathbf{q}) = \Lambda \mathbf{q}^2$ . Since the functional form of  $\phi_+(\mathbf{q})$  changes as the anisotropy parameter changes we retain the general functional form for  $\phi_+(\mathbf{q})$ . Thus we want to solve

$$\phi_+(\mathbf{q}) = \frac{4C_z}{N\beta} \times \sum_{\mathbf{k}}' \frac{[K(\mathbf{q}-\mathbf{k}) - J(\mathbf{k})]^2 (\kappa_y^2 + \mathbf{q}^2)}{[\phi_+(\mathbf{k}) + (\mathbf{q}-\mathbf{k})^2 \Lambda] (\kappa_y^2 + \mathbf{k}^2) [\kappa_z^2 + (\mathbf{q}-\mathbf{k})^2]}, \quad (15)$$

$$\Lambda = \lim_{\mathbf{q} \rightarrow 0} \left\{ \frac{4C_y^2 \kappa_z^2}{C_z \beta N} \sum_{\mathbf{k}}' \frac{[J(\mathbf{q}-\mathbf{k}) - J(\mathbf{k})]^2 \mathbf{q}^{-2}}{2\phi_+(\mathbf{k}) (\kappa_y^2 + \mathbf{k}^2)} \right\}. \quad (16)$$

Going to temperatures very close to the critical temperature (i.e.,  $\kappa_y \ll \kappa_z$ ) we can extract the temperature dependence by defining  $\phi_+(\mathbf{q}) = (a\kappa_y)^{3/2} \mathfrak{F}(q/\kappa_y)$  and  $\Lambda = (a\kappa_y)^{-1/2} \mathcal{G}$ . Substituting these into (15) and (16) and approximating the exchange integrals by their small  $k$  values yields

$$\mathfrak{F}(v) = \frac{4C_z V}{\beta \kappa_z^2 N} \int' \frac{d^3 u [K(0) - J(0)]^2 (1+v^2)}{(2\pi)^3 [\mathfrak{F}(u) + (\mathbf{v}-\mathbf{u})^2 \mathcal{G}] (1+u^2)}, \quad (17)$$

$$\mathcal{G} = \lim_{v \rightarrow 0} \left\{ \frac{C_y^2 \kappa_z^2 V}{C_z \beta N} \int' \frac{d^3 u [J(\mathbf{u}-\mathbf{v}) - J(\mathbf{u})]^2 v^{-2}}{(2\pi)^3 2\mathfrak{F}(u) (1+u^2)} \right\}, \quad (18)$$

where  $\mathbf{v} = \mathbf{q}/\kappa_y$  and  $\mathbf{u} = \mathbf{k}/\kappa_y$ . The integrals over  $u$  are for  $|\mathbf{u}| \leq m$  where  $m\kappa_y$  is the cutoff in momentum marking the boundary between the hydrodynamic and microscopic regions. Since  $\kappa_z$  is a constant we see that  $\mathfrak{F}(v)$  and  $\mathcal{G}$  are independent of temperature. Thus the temperature dependence of  $\phi_+(0)$  and  $\Lambda$  is given by  $\kappa_y^{3/2}$  and  $\kappa_y^{-1/2}$ , respectively. This we will see is in agreement with the dynamic scaling laws, provided the spin-wave frequencies vanish near the Curie point in the manner predicted by the Green's-function calculation outlined in the Appendix. It should also be noted that  $\phi_+(\mathbf{q})$  is of the form  $\kappa_y^a$  times a function of  $\mathbf{q}/\kappa_y$  in agreement with a basic assumption of the dynamic scaling laws.

### III. RESULTS

In the previous section we found the characteristic temperature dependence of the relaxation times for temperatures very close to the critical temperature. In the present section we present the results obtained from

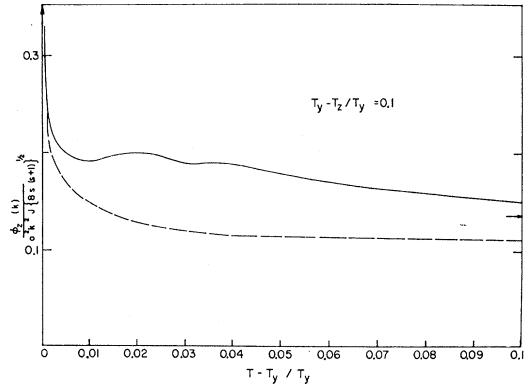


FIG. 2. Decay parameter for the component of spin perpendicular to the easy plane of magnetization in the long-wavelength limit versus temperature. The solid curve was calculated from the self-consistent integral Eqs. (15) and (16). The broken curve is the extrapolation of the asymptotic forms given by Eqs. (25) and (26). An anisotropic exchange interaction has been used. The arrow indicates the value at infinite temperature obtained from a moment calculation, Eq. (33).

a numerical analysis of Eqs. (13), (14), (17), and (18). We consider the specific case of a body centered cubic lattice with nearest neighbor interactions. The coefficients  $C_y$  ( $=C_x$ ),  $\kappa_y$ ,  $C_z$  and  $\kappa_z$  are obtained from Eqs. (A20) and (A21) of the Appendix in the limit  $\mathbf{k} \rightarrow 0$ . (Note that the coordinate system employed in the Appendix differs from that used here by a rotation through  $\frac{1}{2}\pi$  about the  $y$  axis.)  $(S_x, S_y, S_z)_{\text{Appendix}} \leftrightarrow (-S_z, S_y, S_x)_{\text{Text}}$ . Thus we have

$$C_y = (2Ja^2)^{-1}, \quad (19a)$$

$$(\kappa_y a)^2 = (2\chi_y J)^{-1}, \quad (19b)$$

$$C_z = (2Ka^2)^{-1}, \quad (20a)$$

$$(\kappa_z a)^2 = (2\chi_z K)^{-1}, \quad (20b)$$

where  $a$  is the lattice parameter and  $J$  and  $K$  denote the exchange integrals for a pair of ions. We make use of the molecular-field model for the susceptibilities  $\chi_y$  and  $\chi_z$  ( $\chi_{\perp}$  and  $\chi_{\parallel}$  in the notation of the Appendix).

$$\chi_y = \frac{S(S+1)}{3k_B(T-T_y)}, \quad (21)$$

$$\chi_z = \frac{S(S+1)}{3k_B(T-T_z)}, \quad (22)$$

where  $T_y$  ( $T_c$  in the notation of the Appendix) is given by

$$k_B T_y = \frac{2}{3} S(S+1) J(0) = 16S(S+1)J/3 \quad (23)$$

for the body-centered-cubic lattice. Likewise we have

$$k_B T_z = 16S(S+1)K/3 \quad (24)$$

for  $T_z$  ( $=T_{\parallel}$ ).

We first consider the numerical value of  $\phi_+$  and  $\phi_z$  near the Curie temperature (i.e.,  $\kappa_z \gg \kappa_y$ <sup>17</sup>). Defining  $f(u)$  and  $g$  by

$$\phi_+(\mathbf{k})(a\kappa_y)^{-3/2} = \mathfrak{F}(\mathbf{u}) = (K/\beta)^{1/2} \kappa_z a g f(\mathbf{u}), \quad (25)$$

$$\phi_z(\mathbf{k})(a\kappa_y)^{1/2} k^{-2} = \mathfrak{G} = (K/\beta)^{1/2} \kappa_z a^3 g \quad (26)$$

[where  $\mathfrak{F}$  and  $\mathfrak{G}$  are given by (17) and (18), and  $\mathbf{u} = \mathbf{k}/\kappa_y$ ], we find

$$g^2 = \frac{1}{3\pi^2} \int_0^m \frac{u^4 du}{f(u)(1+u^2)^2}, \quad (27)$$

$$f(\mathbf{v}) = \frac{1+v^2}{g^2 4\pi^2} \int_0^m \frac{u^2 du}{1+u^2} \int_{-1}^1 \frac{dx}{f(u)+u^2+v^2-2uvx}. \quad (28)$$

The functions  $f(u)$  and  $g$  are seen to be independent of temperature, the lattice constants, and exchange integrals but do depend on the lattice symmetry. Equations (27) and (28) were solved numerically by iteration.

<sup>17</sup> Note that this precludes taking the limit  $K \rightarrow J$  in Eqs. (19) and (20).

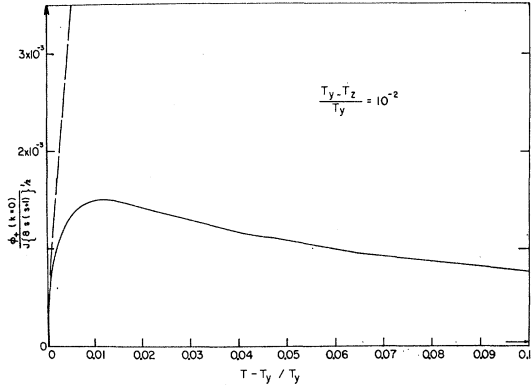


Fig. 3. Decay parameter for the component of spin in the easy plane of magnetization in the long-wavelength limit versus temperature. The solid curve was calculated from the self-consistent integral Eqs. (15) and (16). The broken curve is the extrapolation of the asymptotic forms given by Eqs. (25) and (26). An anisotropic exchange interaction has been used. The arrow indicates the value at infinite temperature obtained from a moment calculation, Eq. (32).

tion. We chose the cutoff  $m$  to be 3 corresponding to a value of  $3\kappa_y$  for the boundary between the hydrodynamic and microscopic regions. As noted below,  $\phi_+(0)$  is relatively insensitive to increases in  $m$  above 2.

We find

$$g = 0.102 \quad (29)$$

and the approximate expansions

$$f(u) = 1.47 + 0.938u^2 - 0.0488u^4, \quad (30a)$$

$$f(u) = 1.39 + 1.16u^2 - 0.130u^4, \quad (30b)$$

where (30a) is accurate to 6% for  $0 \leq u^2 \leq 9$  and (30b) is accurate to 1% for  $0 \leq u^2 \leq 1$ .

At higher temperatures we have solved (15) and (16) by iteration. Because of the assumed dominance of small momenta, we have approximated the exchange integrals by the first two terms of their power-series expansion. The results are shown in Figs. 1-4 for two values of the anisotropy which we take to be defined by  $(T_y - T_z)/T_y$ . We chose the value  $(T_y - T_z)/T_y = 0.01$  and 0.1. The solid curves are the solutions of the integral equations. The dotted curves are given by (25) and (26).

Also indicated are the infinite temperature values calculated by the method of moments assuming an exponential time dependence with a truncated Lorentzian for the spectral function.<sup>18</sup>

$$\phi = \frac{\pi}{2\sqrt{3}} \left\{ \frac{\langle \omega^2 \rangle^3}{\langle \omega^4 \rangle} \right\}^{1/2}. \quad (31)$$

For  $\phi_+(0)$  the moments at infinite temperature have been calculated by Van Vleck.<sup>19</sup> If  $(J-K) \ll J$  and the

<sup>18</sup> W. Marshall, Natl. Bur. Std. (U. S.) Misc. Publ. No. 273 (1965).

<sup>19</sup> J. H. Van Vleck, Phys. Rev. 74, 1168 (1948); see especially his Eqs. (10) and (21).

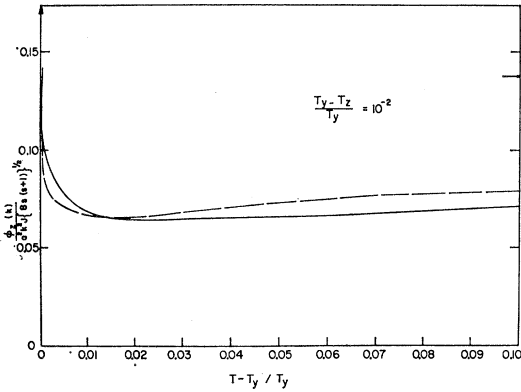


FIG. 4. Decay parameter for the component of spin perpendicular to the easy plane of magnetization in the long-wavelength limit versus temperature. The solid curve was calculated from the self-consistent integral Eqs. (15) and (16). The broken curve is the extrapolation of the asymptotic forms given by Eqs. (25) and (26). An anisotropic exchange interaction has been used. The arrow indicates the value at infinite temperature obtained from a moment calculation, Eq. (33).

coordination number,  $z \gg \frac{1}{2}$  one has

$$\frac{\phi_+(0)}{J[zS(S+1)]^{1/2}} = \frac{\pi}{3\sqrt{6}} \left\{ \frac{J-K}{J} \right\}^2. \quad (32)$$

For  $\phi_z$  the moments at infinite temperature have been calculated by Redfield and Yu.<sup>20</sup> If  $(J-K) \ll J$  and  $2zS(S+1) \gg 1$  we find

$$\frac{\phi_z(\mathbf{k})}{a^2 \mathbf{k}^2 J [zS(S+1)]^{1/2}} = \frac{\pi}{12} \left( \frac{2}{7} \right)^{1/2} \left\{ 1 + \frac{J-K}{2J} \right\}. \quad (33)$$

A number of comments about the numerical methods are in order. The iteration was stopped when the average change in  $\phi_+$  in an iteration was less than  $\frac{1}{2}\%$  per point calculated. This took about 10–15 iterations. One calculation was made requiring less than a  $10^{-6}\%$  change which resulted in a further change in  $\phi_+$  of less than  $1\%$ . A nine-point Gaussian quadrature scheme was used. Using a 27-point integration mesh changed  $\phi_+$  from that calculated using nine points by less than 1 and 10% for  $(T-T_y)/T_y = 10^{-6}$  and  $10^{-1}$ , respectively. The cutoff in momenta was varied from  $\kappa_y$  to  $5\kappa_y$  for  $(T_y - T_z)/T_y = 0.1$ . Increasing  $k_{\text{max}}$  from  $\kappa_y$  to  $2\kappa_y$  resulted in  $\phi_+(0)$  increasing by up to 50% and in  $\phi_z$  increasing by a factor of 3. Further increases in  $k_{\text{max}}$  up to  $5\kappa_y$  resulted in  $\phi_+(0)$  changing by less than about 15% and in  $\phi_z$  increasing by another factor of 3 at most. Thus  $\phi_+(0)$  is fairly insensitive to  $k_{\text{max}}$  if  $k_{\text{max}}$  is greater than  $2\kappa_y$ .

#### IV. DISCUSSION AND COMPARISON WITH TERBIUM DATA

In Sec. III we have reported the results obtained when the general theory developed in Sec. II is applied

<sup>20</sup> A. Redfield and W. Yu, Phys. Rev. 169, 443 (1968); see especially their Eqs. (22) and (23).

to the specific case of a body-centered-cubic lattice. We emphasize that we have used molecular-field model for the susceptibility. Because of this the temperature dependence of  $\phi_+$  and  $\phi_z$  in the vicinity of the Curie point may differ somewhat from the dependence that results from using more realistic values of the susceptibility.

First we note that according to (25) and (26) near the Curie point  $\phi_+(0)$  vanishes as  $\kappa_y^{3/2}$  while  $\phi_z$  diverges as  $\kappa_y^{-1/2}$ , in agreement with the general analysis of Sec. II. Such behavior was also found in a simplified calculation reported previously.<sup>13</sup> However the other results reported in Ref. 13 are not entirely supported by our detailed study. Namely, further from the Curie point it was previously found that  $\phi_+(0)$  decreased as  $\kappa_y^{1/2}$  and  $\phi_z$  as  $\kappa_y^{5/2}$ . The  $\phi_+$  behavior is not dissimilar to that shown in Figs. 1 and 3. However the predicted decrease in  $\phi_z$  is not seen. The reason for the breakdown is that in the simplified calculation it was assumed that  $\phi_+(k)$  could be approximated by its value at the center of the zone. Such an approximation is not satisfactory as is shown in Fig. 5, where  $\phi_+(k)$  is plotted against  $k$ . The deviation from the value at  $k=0$  is very noticeable.

With  $(T_y - T_z)/T_y = 0.1$ ,  $\phi_+(0)$  decreases monotonically to zero for  $(T-T_y)/T_y < 0.1$ . The values of  $\phi_+(0)$  obtained from the asymptotic equation [Eq. (25)] are seen to compare favorably with the computer solutions only for  $(T-T_y)/T_y \lesssim 0.01$ . This value is about 0.1  $(T_y - T_z)/T_y$ . It is also worth noting that the value of  $\phi_+(0)$  at  $(T-T_y)/T_y = 0.1$  is nearly six times as large as the value at  $T = \infty$  which is obtained from the moment analysis. This result appears to be a consequence of the anisotropic exchange as opposed to the single ion anisotropy (see below where  $\phi_+$  at the same temperature is roughly half the value predicted from the moments at  $T = \infty$ ). In the absence of any information on the accuracy of the moment calculation when applied to systems with anisotropic exchange we cannot say whether or not this increase is real.

The behavior of  $\phi_z$  for  $(T_y - T_z)/T_y = 0.1$  is reproduced reasonably well by the asymptotic form apart from some structure which may be spurious, in the

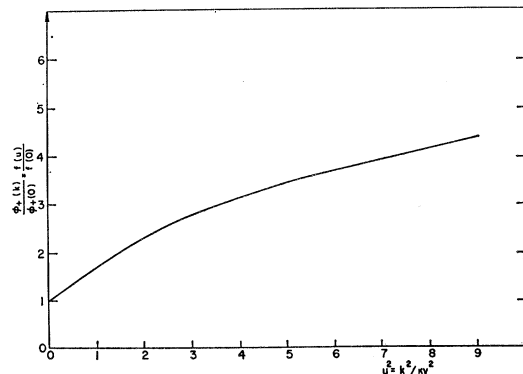


FIG. 5. Decay parameter versus  $k^2$  for  $(T-T_y)/T_y \ll 1$ .

region between  $(T-T_y)/T_y=0.01$  and  $(T-T_y)/T_y=0.03$ . It is apparent that for  $0.01 \lesssim (T-T_y)/T_y \lesssim 0.1$ ,  $\phi_z$  does not depart significantly from its infinite temperature value as determined by the moments. Below this range  $\phi_z$  begins to diverge as  $(T-T_y)^{-1/4}$ , as predicted by Eq. (26). Both  $\phi_+(0)$  and  $\phi_z$  in the region  $(T-T_y)/T_y \lesssim 0.1$  differ greatly from the values predicted by the conventional theory. In the conventional theory it is assumed that  $\phi_+(0)$  and  $\phi_z$  near the Curie point are related to the infinite-temperature values by the equations<sup>18</sup>

$$\begin{aligned}\phi_+(0, T) &= \phi_+(0, \infty) \chi_0(T) / \chi_y(T), \\ \phi_z(T) &= \phi_z(\infty) \chi_0(T) / \chi_z(T),\end{aligned}$$

where  $\chi_0(T)$  is the free-ion susceptibility.

For the smaller anisotropy,  $\phi_+(0)$  below  $(T-T_y)/T_y=0.1$  is seen to go through a maximum at  $(T-T_y)/T_y=0.01$  before finally approaching zero. Furthermore the value at  $(T-T_y)/T_y=0.1$  is approximately 19 times as large as the infinite temperature limit predicted from the moments. The function  $\phi_z$  at the same temperature has approximately half its infinite temperature value, while the divergent behavior does not occur until  $(T-T_y)/T_y \lesssim 5 \times 10^{-3}$ . It is to be noted that for each of the anisotropies the scaling laws are followed when  $(T-T_y) \lesssim 0.1(T_y-T_z)$ . From Eqs. (19)–(22) this is seen to correspond to the region  $\kappa_y^2 \lesssim 0.1\kappa_z^2$ .

The discussion up to now has been confined to calculations based on a model Hamiltonian with an anisotropic exchange interaction. While the self-consistent theory is appealing, there is no reason to prefer it to the conventional theory until a comparison can be made with experiment. Although no inelastic neutron scattering experiments have been reported for systems which are rigorously described by the Hamiltonian (1), there are available measurements of a parameter analogous to  $\phi_+(0)$  in terbium.<sup>14</sup> At 226°K, Tb orders in a spiral array with the magnetization in the basal plane, while at 216°K it undergoes a second transition to a fully ferromagnetic structure. As we noted in Sec. I, Als-Nielsen *et al.* have argued that the pitch angle of the spiral is sufficiently small that Tb may be treated in the first approximation as a uniaxial ferromagnet having a single-ion anisotropy of the form  $D(S_\alpha^z)^2$  ( $D > 0$ ).<sup>14</sup> We have undertaken a calculation of  $\phi_+(0)$  using where possible, parameters identical to those of Tb. That is, we took  $\sum_\beta J_{\alpha\beta}^2$  and  $D$  to have the values given in Ref. 14

$$\begin{aligned}\sum_\beta J_{\alpha\beta}^2 &= 0.1 \text{ meV}^2, \\ D &= 0.2 \text{ meV}.\end{aligned}$$

We inferred a value for the ratio  $(T_y-T_z)/T_z$  from the paramagnetic Curie temperatures, 239 and 195°K, given by Hegland *et al.*<sup>21</sup> The results of this “first-prin-

<sup>21</sup> D. E. Hegland, S. Legvold, and F. H. Spedding, Phys. Rev. **131**, 158 (1963).

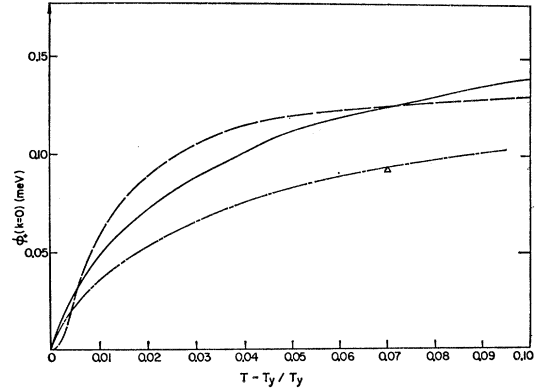


Fig. 6. Decay parameter for the component of spin in the easy plane of magnetization in the long-wavelength limit versus temperature. —, experimental values of Als-Nielsen *et al.* - - -, values calculated using  $\kappa_y \sim (T-T_y)^{1/2}$  and  $\kappa_z \sim (T-T_z)^{1/2}$ . — · —, values calculated using  $\kappa_y \sim (T-T_y)^{1/2}$  and  $\kappa_z \sim (T-T_z)^{1/2}$  normalized to agree with experiment at  $T-T_y=0.07T_y$ .

cles” calculations were quite pleasing. At  $(T-T_y)/T_y=0.07$  we obtain a value of  $\phi_+(0)$  that was within 25% of the measured value. This is to be compared with the results of the conventional theory which predicts a value of  $\phi_+$  at this temperature which is approximately one tenth the measured value.

Because the model calculation was made for a bcc lattice with nearest neighbor interactions whereas Tb has a hexagonal structure with long-range interactions we also display the temperature dependence of  $\phi_+(0)$  after normalizing  $\phi_+$  to the experimental value at  $(T-T_y)/T_y=0.07$ . Our results, scaled in this way, are compared with experiment in Fig. 6. It is seen that the two curves differ by no more than 14 percent over the entire temperature range. We have also calculated  $\phi_+(0)$  assuming  $\kappa_y$  and  $\kappa_z$  varied as  $(T-T_y)^{0.66}$  and  $(T-T_z)^{0.66}$ , respectively, a functional dependence close to what was observed for  $\kappa_y$  in the experiment.<sup>14</sup> The agreement between experiment and theory was somewhat degraded. This may indicate that close to  $T_y$  the detailed shape of  $\phi_+$  in Tb may be determined in part by the anisotropy terms which give rise to the spiral ordering. These, of course, we have neglected.

The agreement between experiment and theory indicated in Fig. 6 is quite good considering the many approximations that have gone into the calculation. Because of this we believe that the self-consistent theory is a significant improvement over the conventional theory. We emphasize, however, that there are still questions as to the applicability of the model to Tb. For this reason additional experiments on systems which are closer to the model Hamiltonian are called for. It is apparent from Eqs. (12)–(14) that the self-consistent theory involves only the parameters of the Hamiltonian, the parallel and perpendicular susceptibilities, and  $\kappa_y$  and  $\kappa_z$ . If this information can be obtained from experiment then a really rigorous test of the self-consistent theory can be made.

Finally we want to comment briefly on the question of dynamic scaling laws. As noted above, Kawasaki<sup>6</sup> has concluded that the characteristic frequencies of a uniaxial ferromagnet with an easy plane of magnetization are related. To a certain extent his analysis is supported by our calculations. If, as predicted by the Green's function calculations in the Appendix, the spin-wave frequency behaves as  $\langle S_z \rangle k$ , then the scaling law arguments predict a characteristic frequency proportional to  $k^{3/2}$  in the transition region and characteristic frequencies  $k^2 \kappa_y^{-1/2}$  and  $\kappa_y^{3/2}$ , in the hydrodynamic region above  $T_y$  (in this discussion we assume  $\eta=0$ ). The hydrodynamic behavior is of course in agreement with Eqs. (25) and (26). It is our feeling that Kawasaki's analysis is more general than the self-consistent calculation. We therefore take the view that agreement with the dynamic scaling laws is a desirable feature of the theory, but do not argue that the self-consistent theory together with the Green's-function analysis justifies the scaling laws.

#### APPENDIX

In this appendix we summarize the results obtained from an analysis of an anisotropic ferromagnet which was carried out using thermodynamic Green's functions.<sup>22</sup> We will derive the functional forms for  $\chi^v(\mathbf{k})$  which were used in Sec. III. In addition we will discuss some of the low temperature properties and the thermodynamic behavior in the critical region. In the interest of simplicity we consider only the case  $S=\frac{1}{2}$ . Our Hamiltonian is similar to (1) except that we include the interaction with an external field perpendicular to the symmetry axis. We choose a coordinate system such that the  $z$  axis is along the field and the  $x$  axis is parallel to the axis of symmetry. [Note that  $(S_x, S_y, S_z)_{\text{Appendix}} \leftrightarrow (-S_z, S_y, S_x)_{\text{Text}}$ .] We have

$$H = -g\mu_B \mathcal{H} \sum_{\alpha} S_{\alpha}^z - \sum_{\alpha, \beta} \{ J_{\alpha\beta} [S_{\alpha}^x S_{\beta}^y + S_{\alpha}^y S_{\beta}^x] + K_{\alpha\beta} S_{\alpha}^z S_{\beta}^z \}. \quad (\text{A1})$$

We introduce the retarded Green's functions

$$G_{+-}^{\alpha\beta}(t) = -i\theta(t) \{ \langle S_+^{\alpha}(t) S_-^{\beta} \rangle - \langle S_-^{\beta} S_+^{\alpha}(t) \rangle \} \quad (\text{A2})$$

and

$$G_{--}^{\alpha\beta}(t) = -i\theta(t) \{ \langle S_-^{\alpha}(t) S_-^{\beta} \rangle - \langle S_-^{\beta} S_-^{\alpha}(t) \rangle \}, \quad (\text{A3})$$

where  $\theta(t)$  is the unit step function and the brackets denote an ensemble average. The equations of motion for  $G_{+-}^{\alpha\beta}$  and  $G_{--}^{\alpha\beta}$  follow in a straightforward fashion from the Hamiltonian (A1). We decouple the equations by making a random-phase approximation (RPA) in which the operator  $S_z^{\alpha}$  is replaced by its expectation value  $\langle S_z^{\alpha} \rangle$  in the higher-order Green's functions arising

from the commutators of  $S_+^{\alpha}$  and  $S_-^{\alpha}$  with  $H$ .<sup>23</sup> In this way we obtain a set of equations involving only  $G_{+-}^{\alpha\beta}$  and  $G_{--}^{\alpha\beta}$ . After Fourier-transforming these equations with respect to the space and time variables we obtain a set of algebraic equations that are readily solved for the transformed Green's functions  $\mathcal{G}_{+-}(k, \omega)$  and  $\mathcal{G}_{--}(k, \omega)$  which are defined by

$$\mathcal{G}_{\pm-}(\mathbf{k}, \omega) = \frac{1}{2\pi} \int dt e^{i\omega t} \sum_{\mathbf{k}} e^{-i\mathbf{k} \cdot (\alpha - \beta)} G_{\pm-}^{\alpha\beta}(t). \quad (\text{A4})$$

The solutions that are found can be written

$$\mathcal{G}_{+-}(\mathbf{k}, \omega) = \frac{\hbar}{2\pi} \times \frac{2\langle S_z \rangle [\hbar\omega + U_{\mathbf{k}}]}{(\hbar\omega)^2 - U_{\mathbf{k}}^2 + \{ \langle S_z \rangle [J(\mathbf{k}) - K(\mathbf{k})] \}^2}, \quad (\text{A5})$$

$$\mathcal{G}_{--}(\mathbf{k}, \omega) = \frac{\hbar}{2\pi} \times \frac{2\langle S_z \rangle^2 [K(\mathbf{k}) - J(\mathbf{k})]}{(\hbar\omega)^2 - U_{\mathbf{k}}^2 + \{ \langle S_z \rangle [J(\mathbf{k}) - K(\mathbf{k})] \}^2}, \quad (\text{A6})$$

where

$$U_{\mathbf{k}} = g\mu_B \mathcal{H} + \langle S_z \rangle \{ 2J(0) - [J(\mathbf{k}) + K(\mathbf{k})] \}, \quad (\text{A7})$$

with

$$J(\mathbf{k}) = \sum_{\alpha} J_{\alpha\beta} e^{i\mathbf{k} \cdot (\alpha - \beta)}, \quad (\text{A8})$$

and similarly for  $K(\mathbf{k})$ .

For a spin- $\frac{1}{2}$  system the temperature and magnetic-field dependence of  $\langle S_z \rangle$  is obtained from a solution of the equation

$$\{ 2\langle S_z \rangle \}^{-1} = N^{-1} \sum_{\mathbf{k}} (U_{\mathbf{k}} / \hbar\omega_{\mathbf{k}}) \coth(\frac{1}{2}\beta\hbar\omega_{\mathbf{k}}), \quad (\text{A9})$$

where

$$\hbar\omega_{\mathbf{k}} = (U_{\mathbf{k}}^2 - \{ \langle S_z \rangle [J(\mathbf{k}) - K(\mathbf{k})] \}^2)^{1/2} \quad (\text{A10})$$

and  $\beta = 1/k_B T$ .

Equation (A9) is a special case of the more general relationship<sup>22</sup>

$$\langle S_-^{\beta} S_+^{\alpha} \rangle = N^{-1} \sum_{\mathbf{k}} e^{i\mathbf{k} \cdot (\alpha - \beta)} \int_{-\infty}^{\infty} d\omega i \{ e^{\beta\hbar\omega} - 1 \}^{-1} \times [\mathcal{G}_{+-}(\mathbf{k}, \omega + i\epsilon) - \mathcal{G}_{+-}(\mathbf{k}, \omega - i\epsilon)], \quad (\text{A11})$$

where the limit  $\epsilon \rightarrow 0+$  is understood. More specifically, (A9) is obtained from (A11) by setting  $\alpha = \beta$ , using (A5) for  $\mathcal{G}_{+-}$  and making use of the result

$$\langle S_-^{\alpha} S_+^{\alpha} \rangle = \frac{1}{2} - \langle S_z \rangle, \quad (\text{A12})$$

which holds for  $S = \frac{1}{2}$ .

We first discuss the implications of (A9) for the low-temperature (spin-wave) region in the limit  $\mathcal{H} = 0$ .

<sup>22</sup> D. N. Zubarev, Usp. Fiz. Nauk **71**, 71 (1960) [English transl.: Soviet Phys.—Usp. **3**, 320 (1960)].

<sup>23</sup> N. N. Bogolyubov and S. V. Tyablikov, Dokl. Akad. Nauk SSSR **126**, 53 (1959) [English transl.: Soviet Phys.—Doklady **4**, 604 (1959)].

Writing  $\langle S_z \rangle = \frac{1}{2} - \langle \Delta S_z \rangle$  we find for  $\langle \Delta S_z \rangle \ll \frac{1}{2}$

$$\langle \Delta S_z \rangle = \frac{1}{2} N^{-1} \sum_{\mathbf{k}} (U_{\mathbf{k}} - \hbar\omega_{\mathbf{k}}) / \hbar\omega_{\mathbf{k}} + N^{-1} \sum_{\mathbf{k}} [e^{\beta\hbar\omega_{\mathbf{k}}} - 1]^{-1} U_{\mathbf{k}} / \hbar\omega_{\mathbf{k}}. \quad (\text{A13})$$

Equation (13), which is identical to the result obtained in the linear spin wave analysis (provided  $\langle S_z \rangle$  is replaced by  $\frac{1}{2}$  in  $U_{\mathbf{k}}$  and  $\omega_{\mathbf{k}}$ ) shows that the anisotropic ferromagnet has a finite zero-point spin deviation. Furthermore, if we write  $J(\mathbf{k}) = r - sk^2$  and  $K(\mathbf{k}) = u - vk^2$  for small  $k$  then the magnon energy

$$\hbar\omega_{\mathbf{k}} = 2\langle S_z \rangle \{ J(0)^2 - [J(\mathbf{k}) + K(\mathbf{k})]J(0) + J(\mathbf{k})K(\mathbf{k}) \}^{1/2} \quad (\text{A14})$$

is seen to vanish linearly with  $k$ , i.e.,<sup>24</sup>

$$\hbar\omega_{\mathbf{k}} \approx s^{1/2} (r - u)^{1/2} k. \quad (\text{A15})$$

As a consequence, the magnon contribution to the specific heat varies as  $T^3$  for small  $T$ , while the temperature-dependent term in  $\langle \Delta S_z \rangle$  varies as  $T^2$ .

The Curie temperature,  $T_c$ , is obtained from the limiting behavior of (A9) as  $\langle S_z \rangle$  approaches zero in the absence of a magnetic field. We have

$$\{2\langle S_z \rangle\}^{-1} = 2k_B T_c N^{-1} \sum_{\mathbf{k}} U_{\mathbf{k}} (\hbar\omega_{\mathbf{k}})^{-2} \quad (\text{A16})$$

or

$$\frac{1}{k_B T_c} = \frac{2}{N} \sum_{\mathbf{k}} \frac{1}{J(0) - J(\mathbf{k})} \left\{ 1 + \frac{J(\mathbf{k}) - K(\mathbf{k})}{2[J(0) - J(\mathbf{k})]} \right\} \times \left\{ 1 + \frac{J(\mathbf{k}) - K(\mathbf{k})}{J(0) - J(\mathbf{k})} \right\}^{-1}, \quad (\text{A17})$$

which is a straightforward generalization of the result obtained for the isotropic ferromagnet in the RPA.<sup>22</sup> A detailed study shows that  $\langle S_z \rangle$  varies  $(T_c - T)^{1/2}$  for

<sup>24</sup> T. Matsubara and H. Matsuda, Progr. Theoret. Phys. (Kyoto) 16, 569 (1956).

$T \lesssim T_c$  in agreement with the behavior found for the isotropic ferromagnet in the RPA.<sup>22</sup>

In the paramagnetic region we can obtain functional forms for the transverse  $\langle S_k^y S_{-k}^y \rangle$  and longitudinal correlation functions directly from the Green's functions by making use of (A11) and similar equations involving the other Green's functions. Defining the susceptibility per spin,  $\chi_{\perp}$ , by

$$\chi_{\perp} = \langle S_z \rangle / g\mu_B \mathfrak{H}C, \quad (\text{A18})$$

we find, after some calculation,

$$\langle S_{\mathbf{k}}^x S_{-\mathbf{k}}^x \rangle = N \sum_{\beta} e^{-i\mathbf{k} \cdot (\alpha - \beta)} \langle S_{\alpha}^x S_{\beta}^x \rangle = N k_B T \{ \chi_{\perp}^{-1} + 2[J(0) - K(\mathbf{k})] \}^{-1} \quad (\text{A19})$$

and

$$\langle S_{\mathbf{k}}^z S_{-\mathbf{k}}^z \rangle = N k_B T \{ \chi_{\perp}^{-1} + 2[J(0) - J(\mathbf{k})] \}^{-1}, \quad (\text{A20})$$

which is equal to  $\langle S_{\mathbf{k}}^y S_{-\mathbf{k}}^y \rangle$  above  $T_c$  ( $\mathfrak{H}C = 0$ ). It should be noted that these equations are valid for general spin.

Because of the approximate equality

$$\chi^{\nu}(\mathbf{k}) \approx \beta \langle S_{\mathbf{k}}^{\nu} S_{-\mathbf{k}}^{\nu} \rangle,$$

which holds for small  $\mathbf{k}$ , we have used the functional forms (A19) and (A20) in the expressions for  $\chi^{\nu}(\mathbf{k})$  in the main text. Rather than values for  $\chi$  which are obtained self-consistently from the Green's functions<sup>25</sup> we have used the molecular field result Eq. (22). Also, we have written  $\langle S_{\mathbf{k}}^x S_{-\mathbf{k}}^x \rangle$  in the form

$$\langle S_{\mathbf{k}}^x S_{-\mathbf{k}}^x \rangle = N k_B T \{ \chi_{\perp}^{-1} + 2[K(0) - K(\mathbf{k})] \}^{-1}, \quad (\text{A21})$$

where

$$\chi_{\perp}^{-1} = \chi_{\perp}^{-1} + 2[J(0) - K(0)] = 3k_B (T - T_{\parallel}) / S(S+1), \quad (\text{A22})$$

with

$$T_{\parallel} = T_c - \frac{2}{3} S(S+1) [J(0) - K(0)] / k_B. \quad (\text{A23})$$

<sup>25</sup> A detailed calculation shows that the susceptibility obtained from the Green's-function formalism [Eq. (A9)] diverges as  $(T - T_c)^{-2}$  near the Curie point. This is the same functional form as is obtained for the isotropic ferromagnet [H. Mori and K. Kawasaki, Progr. Theoret. Phys. (Kyoto) 28, 690 (1962)].