Temperature dependence of the EPR linewidth of $CrBr_3$ near T_C †

M. S. Seehra* and R. P. Gupta‡

Department of Physics, West Virginia University, Morgantown, West Virginia 26506

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Measurements of the temperature dependence of the EPR linewidth ΔH of $CrBr_3$ at 9 GHz and between 20 and 550 °K are reported. The data were taken for the applied field $H_0 \parallel c$ axis and $H_0 \perp c$ axis. The most significant features of the data are (i) a general narrowing of the line as T is lowered from the high-temperature side, (ii) the increasing anisotropy between the two directions as T approaches the Curie point T_C so that near T_C, $(\Delta H)_\parallel/(\Delta H)_\perp \simeq 2$, and (iii) the observation of a crossover from narrowing to broadening for $(\Delta H)_\parallel$ near 50 °K. For $34 < T < 50$ °K, $(\Delta H)_\parallel$ increases as T decreases, confirming the predictions of recent theories of Kawasaki, Huber, and Tomita and Kawasaki. The effect of the magnetic field on the spin dynamics in a ferromagnet as reflected via EPR is discussed. An extension of Huber's calculations to a uniaxial ferromagnet satisfactorily explains the observed anisotropy of the linewidth. Some other ferromagnetic systems, where the crossover effect might be observed, are suggested.

I. INTRODUCTION

The behavior of the electron-paramagnetic-resonance (EPR) linewidth and the spin-spin relaxation rate near the Curie point T_C of a ferromagnet has been studied theoretically by several groups[1-3] in recent years. Kawasaki[1] predicted that as T approaches T_C^+, the EPR linewidth ΔH in a ferromagnet should increase as $\kappa^{-3/2}$ in the small-field limit, i.e.,

$$\Delta H \propto \kappa^{-3/2}, \quad H_0 \ll H_{ex}(\kappa a)^{5/2}. \qquad (1)$$

In the above, κ is the inverse correlation length, a is the lattice constant, and H_0 and H_{ex} are, respectively, the resonance and the exchange fields. In a more recent calculation Huber[2] has arrived at the same result (i.e., $\Delta H \propto \kappa^{-3/2}$) in the zero-field case. These two calculations, done for a Heisenberg system with dipole-dipole interaction as a perturbation, are valid only in the critical region above T_C. Tomita and Kawasaki,[3] on the other hand, have investigated the EPR linewidth in zero field for a uniaxial ferromagnet in the whole paramagnetic region using Green's-function methods. This calculation predicts that as T is lowered toward T_C, the EPR line should first narrow and then broaden as T approaches T_C^+, the crossover occurring near the reduced temperature $\epsilon = (T - T_C)/T_C \simeq 0.5$. Furthermore, the crossover temperature, which physically corresponds to the correlation length $\kappa^{-1} \simeq a$, is independent of the nonzero magnitude of the uniaxial anisotropy. Thus these three calculations predict that in zero field or in the small-field limit as defined by the Kawasaki condition [Eq. (1)], the EPR linewidth in the critical region in a ferromagnet should increase as T approaches T_C^+.

The temperature dependence of the EPR linewidth near T_C has been reported in several ferromagnets, viz., EuO,[4] $CrBr_3$,[5] $K_2CuCl_2 \cdot 2H_2O$,[6] and Ni.[7] In all cases only narrowing of the EPR line has been observed as T approaches T_C^+. In other words, the predicted crossover from narrowing to broadening above T_C has not been observed. However, these experiments were done in resonance fields of 3–10 kOe, whereas the calculations are valid either in zero field or in the small-field limit. Therefore, a direct comparison between the theories and the experiments is not appropriate without taking into account, in some way, the effect of the applied magnetic field. For example, it is well known that the singularity in the isothermal susceptibility of a ferromagnet at T_C is suppressed and the ferromagnetic transition is broadened as the magnetic field is increased from zero.[8]

To check the validity of the theories,[1-3] one has to choose a system in which $H_0 \ll H_{ex}$ in order to satisfy Eq. (1). (Note that below the crossover temperature $\kappa a < 1$ so that the Kawasaki condition will eventually break down for $H_0 \neq 0$ at some temperature near T_C. Huber's calculation also predicts the linewidth to remain finite at T_C.) In $K_2CuCl_2 \cdot 2H_2O$ ($T_C = 1.2$ °K) H_0 (at 9 GHz) $\simeq H_{ex}$ so that this is not an appropriate system for this purpose. In the case of Ni, a metal, it is somewhat doubtful whether the use of localized Heisenberg Hamiltonian is appropriate. Both EuO ($T_C \simeq 69$ °K) and $CrBr_3$ ($T_C \simeq 32.5$ °K) have high enough transition temperatures so that these systems may be expected to satisfy the Kawasaki condition at X-band frequencies. Earlier EPR linewidth measurements in both EuO[4] and $CrBr_3$[5] were made at 24 GHz ($H_0 \simeq 8.5$ kOe). Therefore, we decided to reexamine the EPR linewidth of $CrBr_3$, a uniaxial ferromagnet,[9] at a lower frequency of 9 GHz ($H_0 \simeq 3.2$ kOe). The purpose of these measurements was to sort out the effect of applied magnetic field on spin dynamics as reflected via EPR by comparison with the data at 24 GHz, and thereby check the validity of the theories.[1-3] Our measurements have clearly confirmed the existence of the crossover temperature predicted by the theories. The results of these measurements are presented and discussed in the paper.[10]

II. EXPERIMENTAL PROCEDURES

The EPR linewidth was measured at X-band (~ 9-GHz) frequencies using a standard microwave spectrometer employing bolometer detection and field modulation. The samples of $CrBr_3$ used in these measurements were kindly provided by Remeika and Geschwind of Bell Laboratories. These samples, in the form of thin platelets, were preserved in a dessicator when not in use. Since the magnetic susceptibility of a ferromagnet becomes very large near T_C, overloading of the sample cavity and consequent spurious broadening[11] of the EPR line was observed in some initial experiments below liquid-nitrogen temperatures. To avoid this, a waveguide piece shorted at one end was used instead of a cavity. Also the size of the sample used was quite small, viz., $2\times 2 \times 0.01$ mm with mass $\simeq 10^{-3}$ g and experiments with different size samples convinced us that we were measuring the intrinsic linewidth. The uncertainty in the linewidth is estimated to be about $\pm 2\%$.

For temperatures above about 55 °K, experimental procedure was essentially the same as described elsewhere.[12] Temperatures below 55 °K were obtained with the use of an Andonian variable-temperature liquid-helium Dewar. The temperatures were stabilized to within 0.1 °K near T_C by using Artronix model No. 5301E temperature controller. The temperatures were measured using a copper-constantan thermocouple imbedded in the waveguide in the vicinity of the sample and thermocouple voltages were measured with a Leeds and Northrup K-5 potentiometer.

III. EXPERIMENTAL RESULTS

The temperature dependence of the experimentally measured EPR linewidth (peak-to-peak magnetic field separation in the absorption derivative) is shown in Fig. 1. Measurements were made from temperatures below T_C to about 550 °K. The motivation for our measurements above room temperature was to determine the constant high-temperature linewidth in order to compare it with the calculated infinite-temperature exchange-narrowed linewidth. As is evident from Fig. 1 some temperature dependence of the linewidth is still evident even at the highest temperatures of our measurements. Unfortunately, we were not equipped to take data at higher temperatures.

From Fig. 1 we note that as T approaches T_C^+, the anisotropy of the linewidth for $\vec{H}_0 \parallel c$ axis (the easy axis) and $\vec{H}_0 \perp c$ axis increases. This anisotropy near T_C is shown on a more expanded scale in Fig. 2. An important feature of the data is the line broadening observed for $\vec{H}_0 \parallel c$ axis for $T < 50$ °K. As the temperature is lowered from the high-temperature side, the parallel linewidth $(\Delta H)_\parallel$ decreases, reaching a minimum near 50 °K and then increases for lower temperatures. A significant change in the slope of the $(\Delta H)_\perp$-vs-T plot is also noticeable in the same temperature range. Below about 34 °K, $(\Delta H)_\parallel$ sharply decreases with decreasing temperatures. This is the ferromagnetic region since large changes in the resonance fields were observed because of the demagnetizing fields. NMR[13] and specific-heat[14] measurements in $CrBr_3$ give $T_C \simeq 32.5$ °K. We find that the linewidth is maximum at 34 °K. (Let us call this temperature T_m.) Since our measurements are made in a magnetic field of about 3.2 kOe, a broadening of the critical region resulting in $T_m > T_C$ is not surprising.[8] At T_m, we find $(\Delta H)_\parallel/(\Delta H)_\perp \simeq 2$. We will show later that this results from the uniaxial symmetry of $CrBr_3$.

In Fig. 3 we have compared our measurements at 9 GHz with the data of Dillon and Remeika at 24 GHz[15] for the parallel direction. The two measurements at 9 and 24 GHz agree quite well for $T > 70$ °K although data up to only about 100 °K are shown in order to show the details in the critical region. Unfortunately, the data at 24 GHz are not as detailed as the present measurements. However, it is clear that there are large discrepancies between the two measurements in the critical region. The most dramatic differences are the presence of the crossover effect (from narrowing to broadening) near 50 °K in the measurements at 9 GHz and a further upward shift of T_m from 34 °K at 9 GHz to about 43 °K at 24 GHz. The latter is a further evidence of the broadening of the critical region when magnetic field is increased. The crossover temperature corresponds to $\epsilon \simeq 0.5$, in excellent agreement with the predicted value.[3]

IV. DISCUSSION

A. Effect of magnetic field

From the data shown in Fig. 3, it is clear that the magnetic field, necessary to observe the resonance, has considerable effect on the EPR linewidth near T_C. Although the magnetic field has been shown to suppress the singularity in the EPR linewidth in antiferromagnets,[16,17] the effect in ferromagnets is perhaps qualitatively different. Unlike the antiferromagnetic case, the magnetic field in a ferromagnet couples directly to the long-wavelength fluctuations in the order parameter. Also note that in Fig. 2, the line broadening observed below the crossover temperature is considerably less than the theoretical prediction of $\Delta H \sim \epsilon^{-1}$ if we assume $\kappa \sim \epsilon^{2/3}$. Now we discuss the effect of the applied field below the crossover tem-

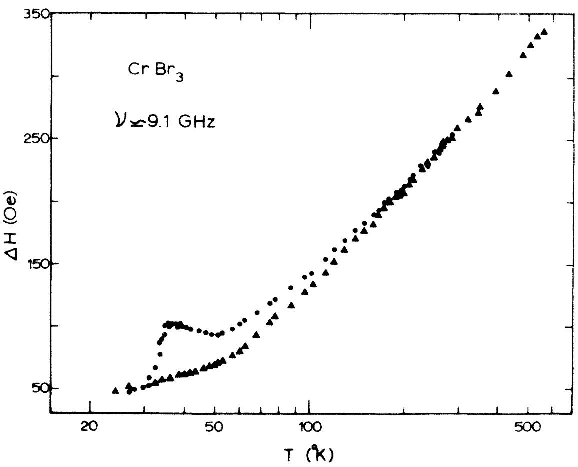

FIG. 1. Measured peak-to-peak linewidth ΔH is plotted against temperature on a semilog plot. Solid circles are for $\vec{H}_0 \parallel c$ axis and solid triangles are for $\vec{H}_0 \perp c$ axis. For the sake of clarity only a small number of data points are plotted at lower temperatures.

perature.

First we note that the Kawasaki condition [Eq. (1)] is valid for $CrBr_3$ at 9 GHz at least for a certain temperature range below the crossover temperature. Following the discussion given by Kawasaki,[1] we estimate that in $CrBr_3$, $H_{ex} \sim 43$ kOe so that H_0 (at 9 GHz) is about an order of magnitude smaller than H_{ex}. Below the crossover temperature, $\kappa a (< 1)$ becomes smaller as T is lowered. Therefore, the Kawasaki condition would start to break down as T approaches T_C.

Another effect that appears to be important in suppressing the predicted enhancement of the linewidth is the magnetic field dependence of the correlation length κ^{-1}. Leichner and Richards[18] (LR) have discussed the field dependence of the correlation length κ^{-1} at T_C. They have shown, using constant-coupling approximation, that for spin $S = \frac{1}{2}$ one obtains

$$\kappa^{-1} \propto (k_B T_C / \mu_B H_0)^{1/2} . \tag{2}$$

Static scaling laws[19] for the Ising model yield (for $T = T_C$)

$$\kappa^{-1} \propto (H_0)^{-2/5} \tag{3}$$

if one uses the values of the critical exponent for three-dimensional systems. What is important in Eqs. (2) and (3) is that the correlation length is suppressed as the magnetic field is increased. Since the predicted enhancement of the linewidth [Eq. (1)] is related to the correlation length becoming very large as T_C is approached, suppression of the range of the correlation by the magnetic field suppresses the enhancement. We believe that this is the reason why we have not observed a more pronounced enhancement of the linewidth below the crossover temperature and also why earlier measurements at 24 GHz failed to show the crossover behavior and consequent enhancement of the linewidth.

Leichner and Richards[18] have presented a detailed analysis of the temperature dependence of the EPR linewidth in a ferromagnet in a finite magnetic field using the moment method. They have applied their analysis to the linewidth behavior in $K_2CuCl_2 \cdot 2H_2O$ [where H_0 (at 9 GHz) $\simeq H_{ex}$)] and Ni and found satisfactory agreement in the critical region. In both cases, as noted earlier, no crossover behavior as reported here was observed. Clearly, observations in $CrBr_3$ cannot be adequately explained by the moment calculations of LR. This is probably due to the fact that the moment calculations of LR,[18] using second and fourth moments only, take into account only the short-range correlations as also noted by Kawasaki.[1] For $K_2CuCl_2 \cdot 2H_2O$, $H_0 \simeq H_{ex}$ and the cor-

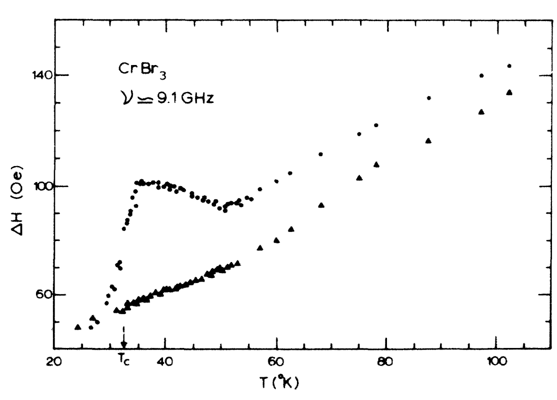

Fig. 2 Measured linewidth is plotted against temperature in the critical region for $\vec{H}_0 \parallel c$ axis (solid circles) and for $\vec{H}_0 \perp c$ axis (solid triangles).

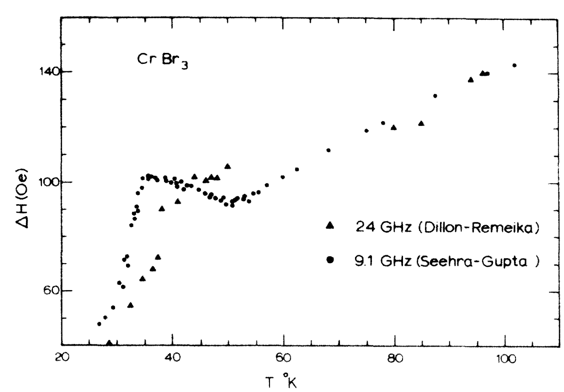

FIG. 3. Comparison of the linewidths at 9 and 24 GHz in the critical region for $\vec{H}_0 \parallel c$ axis. The data at 24 GHz were taken from a graph in Ref. 5.

relation length $\kappa^{-1} \simeq 0.8a$ at T_C as calculated by LR. Clearly, the long-range correlations are suppressed and the moment calculations appear to give a good account of the linewidth behavior in the critical region. On the other hand, our data in $CrBr_3$ at 9 GHz, in particular the crossover effect, tend to show that the long-range correlations are important even in a field of 3.2 kOe. The moment calculations therefore do not explain our observations in $CrBr_3$.

B. Linewidth anisotropy in a uniaxial ferromagnet near T_C

One of the significant features of the data in Fig. 2 is the increasing anisotropy of the linewidth between $(\Delta H)_{\parallel}$ and $(\Delta H)_{\perp}$ as T approaches T_C. At $T = T_m$, $(\Delta H)_{\parallel}/(\Delta H)_{\perp} \simeq 2$ as already noted. Ferromagnetic resonance and magnetization studies have shown that the anisotropy in $CrBr_3$ is nearly uniaxial.[9] We now show that the observed anisotropy of the linewidth in $CrBr_3$ near T_C can be explained on the basis of uniaxial anisotropy. The calculations given below follow the procedure used by Huber[2] for the cubic system.

Huber has discussed that the zero-field spin-spin relaxation time can be written

$$\frac{1}{T_2^\alpha} = (\chi_T^\alpha)^{-1} \int_0^\infty dt\, (\dot{M}^\alpha(t),\ \dot{M}^\alpha(0)), \tag{4}$$

where

$$\dot{M}^\alpha = (i\hbar)^{-1}[M^\alpha, \mathcal{H}] \tag{5}$$

and

$$M^\alpha = g\mu_B \sum_i S_i^\alpha . \tag{6}$$

In the above, $\alpha = x, y, z$ and the g value is assumed to be independent of α. Also $\mathcal{H}$ is the Hamiltonian, the relaxation function for an operator A is given by

$$(A(t), A) = \int_0^\beta d\lambda \left\langle \exp\left[\left(\lambda + \frac{it}{\hbar}\right)\mathcal{H}\right] A \times \exp\left[-\left(\lambda + \frac{it}{\hbar}\right)\mathcal{H}\right] A^+ \right\rangle - \beta\langle A\rangle\langle A^+\rangle, \tag{7}$$

where $\beta = 1/k_B T$ and $\langle\ \rangle$ denotes a statistical average. The isothermal susceptibility χ_T^α is given by

$$\chi_T^\alpha = (M^\alpha, M^\alpha). \tag{8}$$

Huber has calculated T_2 for a magnetic system governed by the Heisenberg exchange interaction with dipole-dipole interaction as the perturbation. However, the anisotropy in $CrBr_3$ is nearly uniaxial as ferromagnetic resonance and magnetization studies[9] have shown. To discuss this case we start with the Hamiltonian

$$\mathcal{H} = -\frac{1}{N} \sum_q J(q)\vec{S}(q) \cdot \vec{S}(-q) - \frac{1}{N} \sum_q \vec{S}(q) \cdot D(q) \cdot \vec{S}(-q), \tag{9}$$

where $S(q)$, the Fourier transform of S_j, is given by

$$S(\vec{q}) = \sum_j e^{i\vec{q}\cdot\vec{r}_j} S_j . \tag{10}$$

In Eq. (9), the first term is the familiar Heisenberg exchange interaction, the second term is the anisotropy with D as a traceless tensor, and the sum is over the Brillouin zone. It is assumed that the total anisotropy, both crystalline and dipolar, is contained in $D(q)$ and that the applied field $H_0 = 0$. Near T_C in a ferromagnet, the fluctuations with $q \leq \kappa$ are the ones which are important for the linewidth anomaly, with the dominant contribution coming from the neighborhood of $q = 0$. Therefore, we separate out the critical part of the anisotropy ($\mathcal{H}_a^c$) and write it as

$$\begin{aligned}\mathcal{H}_a^c &= -\frac{1}{N} \sum_{q \leq \kappa} \vec{S}(q) \cdot D(0) \cdot \vec{S}(-q) \\ &= -\frac{1}{N}\, D_{\parallel}(0) \sum_{q \leq \kappa} S^z(q) S^z(-q) + D_{\perp}(0) \\ &\quad \times \sum_{q \leq \kappa} [S^x(q) S^x(-q) + S^y(q) S^y(-q)]\end{aligned} \tag{11}$$

for a uniaxial system. It follows from Eqs. (5) and (11) that M^z commutes with $\mathcal{H}_a^c$, where M^x and M^y do not. Using this in Eq. (4) yields

$$1/T_{2_\parallel} = C/T\chi_{\parallel} \tag{12}$$

and

$$1/T_{2_\perp} = [C + f(\epsilon)]/T\chi_{\perp} . \tag{13}$$

In writing Eqs. (12) and (13) we have separated the critical and the noncritical contribution to the spin-spin relaxation rate. Here C represents the noncritical contributions [contributions for $D(q)$ for $q \neq 0$] and $f(\epsilon)$ represents the critical contribution to the relaxation rates. In the Appendix we have given a calculation for $f(\epsilon)$ under certain assumptions where it is shown that near T_C, $f(\epsilon)$ diverges as $\chi_{\parallel}^{7/4}$. Calculation of C is considerably more difficult since we do not know enough about the temperature dependence of the noncritical modes. However, it is expected that the temperature dependence near T_C is dominated by the critical part $f(\epsilon)$ and that C is a slowly varying function of T.

To relate the EPR linewidth with the spin-spin relaxation time we note that the EPR linewidth measures the relaxation rate for spin fluctuations normal to the static field. In the short-correlation-time limit (viz., $g\mu_B H_0/\hbar\Gamma_0 \ll 1$, where Γ_0 is the decay rate for $q = 0$ fluctuations) one can write[20]

$$(\Delta H)_{\parallel} = (\hbar/g\mu_B)(1/T_{2\perp}) \tag{14}$$

and

$$(\Delta H)_\perp = (\hbar/2g\mu_B)(1/T_{2\parallel} + 1/T_{2\perp}). \quad (15)$$

With the use of Eqs. (12)–(15), we get

$$\frac{(\Delta H)_\parallel}{(\Delta H)_\perp} = \frac{2}{1 + (\chi_\perp/\chi_\parallel)[1 + f(\epsilon)/C]^{-1}}. \quad (16)$$

Near T_C the critical contribution dominates so that $f(\epsilon) \gg C$. Also only $\chi_\parallel$ is singular as $T \to T_C^+$, whereas $\chi_\perp$ remains finite at T_C. Therefore, in the critical region we may also take $\chi_\parallel \gg \chi_\perp$.[21] Using this information in Eq. (16) we get $(\Delta H)_\parallel \simeq 2(\Delta H)_\perp$, as observed near T_C in $CrBr_3$. In the other limit, i.e., for $T \gg T_C$, the critical contribution is expected to be negligible as compared to the noncritical contribution [i.e., $f(\epsilon) \ll C$]. Also susceptibility measurements in $CrBr_3$[21] have shown that for $T \gg T_C$, $\chi_\parallel \simeq \chi_\perp$. (This is expected since for $T \gg T_C$, the static susceptibility is determined mainly by the predominant Heisenberg exchange interaction.) Using $\chi_\parallel = \chi_\perp$ and $f(\epsilon) \ll C$ in Eq. (16), we get $(\Delta H)_\parallel \simeq (\Delta H)_\perp$, a result observed above about 7 T_C in $CrBr_3$.

We note that the above conclusions, in agreement with the observation reported in this paper, are based on the uniaxial symmetry.[22] It is also noted that if the anisotropy were mainly of the single-ion type, $D(q)$ would be independent of q leading to $(\Delta H)_\parallel = 2(\Delta H)_\perp$ at all temperatures. Since this is contrary to observations, it may be concluded that the dipole-dipole interaction makes important contributions to $D(q)$ in $CrBr_3$. From Eqs. (12)–(15) it can also be seen that the general narrowing of the line with decreasing temperatures results from the enhancement of susceptibilities. The increasing anisotropy between $(\Delta H)_\parallel$ and $(\Delta H)_\perp$ with decreasing temperatures results from $\chi_\parallel$ becoming larger than $\chi_\perp$ and $f(\epsilon)$ becoming larger than C. Thus, the observed anisotropy in the linewidth of $CrBr_3$ and its temperature dependence can be accounted for by using an extension of Huber's formalism to a uniaxial case. It would have been useful to plot $(\Delta H)_\parallel T\chi_\perp$ in order to directly look at the critical part $f(\epsilon)$. Unfortunately, data for $\chi_\perp$ are not available in the whole temperature region of interest.

C. Other suitable systems

A question arises whether the crossover effect observed in $CrBr_3$ might be observed in other ferromagnets. We have already noted that a suitable system might be EuO. In $CdCr_2Se_4$ ($T_C \simeq 140$ °K)[23] there is a slight hint of the crossover effect although the data are not detailed enough to make a strong case. Furthermore, these measurements were made at 13 GHz. Therefore, it might be worthwhile to carefully reexamine the temperature dependence of the linewidth in this material at a lower frequency. In $RbNiF_3$, a ferrimagnet, there is also an indication of the crossover effect.[24] Here again the data in the critical region are not detailed enough. Furthermore, it is doubtful whether the theories[1–3] are directly applicable to a ferrimagnet.

Although it is doubtful that the use of the Heisenberg Hamiltonian is valid in Ni, it might be worthwhile to reexamine its linewidth at a lower frequency since the reported measurements[7] were made at frequencies of 23 and 32 GHz.

V. CONCLUDING REMARKS

As a matter of comparison among the various calculations we note that with $H_0 = 0$, the moment calculations[18] predict that $\Delta H = 0$ at T_C, whereas according to the other theories[1–3] discussed in this paper ΔH at T_C is considerably enhanced as compared to its value in the high-temperature limit. Presumably, this difference arises because of the contributions from the long-range correlations which are not included in the moment calculations. As the magnetic field is increased from zero, the long-range correlations are increasingly suppressed and in the limit that $H_0 \simeq H_{ex}$, the moment calculations may give a good description of the linewidth behavior near T_C.

The data and the discussion presented in this paper have shown that the study of the EPR linewidth can provide useful information on the spin dynamics if the measurements are made at low enough magnetic fields. Although we were not able to make a quantitative check on the theories because they do not include the magnetic field explicitly, this work has clearly shown that the anisotropy induced spin-spin relaxation is quite important in the decay of long-wavelength spin fluctuations near T_C. Consequently, this should be taken into account in interpreting the neutron scattering data in the long-wavelength limit.

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APPENDIX

Here we outline the calculation for $f(\epsilon)$ which represents the critical contribution to $(1/T_{2\perp})$ for a uniaxial ferromagnet. We follow the procedure used by Huber[2] for a similar calculation in the case of cubic system with dipolar anisotropy. The four-spin correlation functions appearing in $(\dot{M}^\alpha(t), \dot{M}^\alpha)$ are decoupled into the products of two two-spin relaxation functions via the random-phase approximation (RPA) and we further assume that the fluctuations are isotropic in spin space. With these assumptions one obtains for the critical part of the relaxation time the expression

$$\left(\frac{1}{T_{2\perp}}\right)_C = \frac{4k_B T[D_\parallel(0) - D_\perp(0)]^2 g^2\mu_B^2}{N^2\hbar^2\chi_\perp} \times \sum_q \int_0^\infty dt(S(q,t),\, S(q))^2. \qquad \text{(A1)}$$

Near T_C, we take $g^2\mu_B^2(S(q,t),\, S(q)) = \chi_\parallel e^{-\Gamma_q t}/(1+q^2\kappa^{-2})$, with $\Gamma_q = \Lambda q^2(1+q^2\kappa^{-2})$, Λ being the diffusion constant. (Huber[2] has discussed the justification for the above relations.) Substituting these values in Eq. (A1), replacing the summation over q by an integration from $q=0$ to $q=\kappa$, and evaluating the integral, yields

$$\left(\frac{1}{T_{2\perp}}\right)_C = \left(\frac{4}{23\pi}\right)\frac{k_B T[D_\parallel(0) - D_\perp(0)]^2 v\chi_\parallel^2}{g^2\mu_B^2\hbar^2\Lambda\kappa^{-1}N\chi_\perp}, \qquad \text{(A2)}$$

where v is the volume per spin. Comparison of Eq. (A2) with Eq. (13) gives

$$f(\epsilon) = \left(\frac{4}{23\pi}\right)\frac{k_B T^2[D_\parallel(0) - D_\perp(0)]^2 v\chi_\parallel^2}{g^2\mu_B^2\hbar^2\Lambda\kappa^{-1}N}. \qquad \text{(A3)}$$

According to our current understanding of second-order phase transitions,[1,19] $\chi_\parallel \sim \kappa^{-2}$ and $\Lambda \sim \kappa^{1/2}$. This leads to $f(\epsilon)$ diverging as $\chi_\parallel^{7/4}$, since all other quantities in Eq. (A3) are either temperature independent or slowly varying in the critical region.

One can also include the anisotropy of the two-spin relaxation function in the above calculations. We note that in the integrand of Eq. (A1), the relaxation function actually appears as $(S_\parallel(q,t),\, S_\parallel(q))\,(S_\perp(q,t),\, S_\perp(q))$. To make further progress one needs to distinguish between parallel and perpendicular values for Γ and κ^{-1}. The result of this analysis is that the singularity in $(1/T_{2\perp})_C$ would not be as enhanced since $\chi_\perp$ and $\kappa_\perp^{-1}$ are expected to remain finite at T_C.

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*Alfred P. Sloan Foundation Research Fellow.

‡Present address: Federal Electric Corp., Vandenburg Air Force Base, Calif.

[1] K. Kawasaki, Prog. Theor. Phys. 39, 285 (1968).

[2] D. L. Huber, J. Phys. Chem. Solids 32, 2145 (1971). These calculations predict that the zero-field linewidth eventually saturates at a temperature where $\chi_T/\chi^0(T_C) \simeq Ja^3/g^2\mu_B^2$. Here χ_T is the isothermal susceptibility, χ^0 is the Curie susceptibility for the noninteracting system, J is the dominant exchange integral in the system, and a is the lattice constant.

[3] K. Tomita and T. Kawasaki, Prog. Theor. Phys. 44, 1173 (1970).

[4] D. E. Eastman, Phys. Lett. A 28, 136 (1968).

[5] J. F. Dillon, Jr. and J. P. Rameika, *Proceedings of the Eleventh Colloque Ampere, Eindhoven*, edited by J. Smit (North-Holland, Amsterdam, 1962), p. 480.

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

[7] S. M. Bhagat and S. M. Chicklis, Phys. Rev. 178, 828 (1969); M. B. Salamon, Phys. Rev. 155, 224 (1967).

[8] L. Flax, Phys. Rev. B 5, 977 (1972).

[9] J. F. Dillon, Jr., J. Appl. Phys. 335, 1191 (1962); see also, J. F. Dillon, Jr., J. Phys. Soc. Jap. 19, 1662, (1964).

[10] A preliminary account of this work has been reported. See M. S. Seehra and R. P. Gupta, Bull. Am. Phys. Soc. 18, 341 (1973).

[11] M. S. Seehra, Rev. Sci. Instrum. 39, 1044 (1968).

[12] R. P. Gupta, M. S. Seehra, and W. E. Vehse, Phys. Rev. B 5, 92 (1972).

[13] S. D. Senturia and G. B. Benedek, Phys. Rev. Lett. 17, 475 (1966).

[14] L. D. Jennings and W. N. Hansen, Phys. Rev. 139, A1694 (1965).

[15] Data at 24 GHz plotted in Fig. 3 were taken from a graph of Ref. 5. Also we used $(\Delta H)_{\text{p-p}} = (\Delta H)_{1/2}/\sqrt{3}$ for a Lorentzian line shape to convert $(\Delta H)_{1/2}$ (full width at half-maximum) to $(\Delta H)_{\text{p-p}}$ (peak to peak).

[16] M. S. Seehra, J. Appl. Phys. 42, 1290 (1971).

[17] R. P. Gupta and M. S. Seehra, Phys. Lett. A 33, 347 (1970).

[18] P. K. Leichner and P. M. Richards, Phys. Rev. B 7, 453 (1973).

[19] L. P. Kadanoff, Physics (N. Y.) 2, 263 (1966).

[20] A discussion on this is given by D. L. Huber, Phys. Rev. B 6, 3180 (1972). In a more recent work, P. W. Verbeek [Ph.D. thesis, (University of Leiden, The Netherlands, 1973) (unpublished). See also W. M. DeJong and J. C. Verstelle, Phys. Lett. A 42, 297 (1972)] has shown that in the short-correlation-time limit so that both the secular and nonsecular terms of the dipolar interaction contribute to line broadening, the linewidth $(\Delta H_{1/2})_i = (\hbar/g_i\mu_B)\,(T_{2j}^{-1} + T_{2k}^{-1})$, where i, j, k refer to the principal axes and $\Delta H_{1/2}$ is the full width at half-maximum. Equations (14) and (15) are equivalent to the above equation except for a numerical factor of order unity which takes into account different definitions of the linewidth. Clearly, the ratio $(\Delta H)_\parallel/(\Delta H)_\perp$ in Eq. (16) is unaffected. Also note that in $CrBr_3$ we find $g_\parallel \simeq g_\perp$.

[21] Experimental measurement of $(\chi_\parallel - \chi_\perp)$ in $CrBr_3$ [I. Tsubokawa, J. Phys. Soc. Jap. 15, 1664 (1960)] between 43 and 293 °K show that for $T \to T_c^+$, $\chi_\parallel \gg \chi_\perp$, but for $T \gg T_c$ (e.g., at $T = 293$ °K) $\chi_\parallel \simeq \chi_\perp$.

[22] Similar anisotropy of the EPR linewidth was observed in the uniaxial antiferromagnet MnF_2. [See M. S. Seehra, Phys. Rev. B 6, 3186 (1972).] Theoretical discussion of the EPR linewidth anomaly in antiferromagnets near T_N has been given by D. L. Huber [Phys. Rev. B 6, 3180 (1972)].

[23] R. C. LeCraw, H. Von Philipsborn, and M. D. Sturge, J. Appl. Phys. 38, 965 (1967).

[24] E. Toyota and K. Hirakawa, J. Phys. Soc. Jap. 30, 692 (1971).