Field and temperature dependence of critical magnetic relaxation in the anisotropic ferromagnet $CrBr_3$

J. Kötzler and W. Scheithe

Institut für Festkörperphysik, Fachgebiet Technische Physik, Technische Hochschule Darmstadt, D-6100 Darmstadt, Federal Republic of Germany

(Received 7 March 1978)

We present measurements of the dynamic longitudinal susceptibility near the Curie temperature of $CrBr_3$ in magnetic fields up to 30 mT applied parallel to the easy magnetic axis. The frequency dependence of χ is monodispersive except very close to T_c, where a second unknown relaxation process comprising a small part of the total χ appears at low frequencies. Above T_c the critical slowing down of the main (fast) process is explained with the help of recent mode-coupling results as arising from spin-spin relaxation in an uniaxial ferromagnet. With rising magnetic field the relaxation rate increases steeply below the dipolar crossover ($\epsilon < 0.03$), while above it a gradual decrease is found. Both observations seem to be inconsistent with existing ideas about the field effect on critical relaxation. Below T_c the fast (para-) process is observed to speed up again.

I. INTRODUCTION

In ferromagnets the magnetic field couples directly to the order parameter, the homogeneous magnetization, and thus it can strongly influence the critical behavior around the magnetic ordering temperature. As far as static properties are concerned, e.g., susceptibility and magnetization, the field suppresses the critical anomalies being observed on a large number of uniaxial and cubic ferromagnets, among them the archetypal examples $CrBr_3$,[1] Gd,[2] and Ni,[3] EuO.[4] Much less information is available so far about the magnetic field effect on the critical dynamics of ferromagnets, for instance, on the magnetic relaxation rate Γ, characterizing the long-time decay of the order parameter to an equilibrium state, $M(t\to\infty)-M_{eq}\propto\exp(-\Gamma t)$. Though several theoretical treatments of the critical behavior of Γ in zero magnetic field exist,[5-11] the field dependence of Γ has received only a little attention.

Above T_c Maleev[9] predicts for uniaxial ferromagnets that Γ will increase anomalously even in extremely small fields, which do not affect the static susceptibility. Kawasaki[5] assumes the magnetic field to be of no importance as long as the Larmor precession cannot disturb the dynamics of the critical fluctuations, i.e., $\gamma B \ll \Gamma_{q\approx\xi^{-1}}$ is satisfied. Here $\Gamma_{q\approx\xi^{-1}}$ denotes the damping constant of the critical magnetization modes having wavelengths comparable to the correlation length ξ. Assuming to stay within this zero-field limit for the critical relaxation, several experimental workers attempted to interpret the critical behavior of magnetic resonance linewidths of Ni,[12] EuO,[13] $CrBr_3$,[14] and Gd,[15] in terms of the cited theoretical results. In no case, however, could convincing agreement be achieved and this has mainly been ascribed to the non-negligible effect of the magnetic fields of at least 300 mT applied in these experiments at X-band frequencies. Only very recently, a systematic study of the critical behavior of EPR linewidths in the cubic ferromagnet $CdCr_2Se_4$[16] quantitatively confirmed the theoretical zero-field results. In this system the linewidths proved to be sufficiently small, so that (by reducing the microwave frequency to 0.7 GHz) the field of resonance could be lowered to 25 mT. Thereby, at least in a certain (outer) part of the critical region Kawasaki's condition, $\gamma B \ll \Gamma_{\xi^{-1}}$, could be shown to hold, in which case the EPR linewidth of a cubic ferromagnet delivers the zero-field limit of the longitudinal relaxation rate Γ.[5,13]

A much more suitable but from the experimental point of view, also more elaborate method for examining the field dependence of Γ is to measure it directly via the frequency dependence of the longitudinal susceptibility of the order parameter which, in case of the aforementioned exponential decay, takes the Debye form

$$\chi(\omega;T,B)=\frac{\chi_{eq}(T,B)}{1+i[\omega/\Gamma(T,B)]} \quad . \tag{1}$$

By this method one eliminates the resonance condition and is therefore able to measure Γ in arbitrarily small fields starting from $B=0$. So far in low fields, $B \lesssim 20$ mT experimental values of $\Gamma(T,B)$ are available only for the cubic ferromagnet, EuS.[17] Above T_c and for $0 \lesssim B \lesssim 20$ mT the critical behavior of the corresponding kinetic Onsager coefficient, being independent of the shape of the sample, could simply be described in terms of the internal equilibrium susceptibility,

$$\chi_{eq,i} = (\chi_{eq}^{-1} - N)^{-1},$$

$$\Gamma\chi_{eq} \equiv L(T,B) = f(\chi_{eq,i}(T,B)) , \qquad (2)$$

which is consistent with the dynamic scaling hypothesis.[18] While in EuS the critical fluctuations can be assumed to be governed by the specific anisotropy of the dipolar interaction,[19] experimental investigations of Γ in cubic ferromagnets with much stronger Heisenberg exchange interactions $CdCr_2Se_4$, $CdCr_2S_4$ are in progress with the aim to extend the examination of the field effect on Γ into the direction of the exchange critical regime.[20]

In view of these current activities with *cubic* materials, we found it natural and interesting to proceed a step further by examining the influence of the field on the critical spin relaxation in an uniaxial ferromagnet like the hexagonal insulator $CrBr_3$. In this system, in addition to the Heisenberg exchange and the dipolar interaction, two more magnetic anisotropies are present. As will be explained below in greater detail, uniaxial dipolar critical behavior should prevail for $\epsilon \ll 0.03$, while at larger ϵ two-dimensional Heisenberg behavior should dominate. It is believed on general grounds that dynamic critical properties are much more sensitive to details of the magnetic Hamiltonian than static critical properties are[21] and, therefore, we expect new effects to occur. For the zero-field case and by using the known magnetic interaction parameters for $CrBr_3$,[22] we intend to perform a quantitative check of the theoretical estimates existing for the paramagnetic side. Below T_c the present understanding of the critical relaxation seems to be much less advanced. We are aware of only one result obtained experimentally by Rutten and Verstelle[23] on the uniaxial ferromagnet $CuRb_2Br_4 \cdot 2H_2O$ ($T_c = 1.874$ K), who could interpret their $\chi(\omega)$ data in terms of a sum of two Debye functions above $1 - T/T_c \approx 0.03$. Assuming a predomain state in this critical part of the ferromagnetic region, the fast and slow relaxation process, respectively, were ascribed to paramagnetic spins and strongly coupled spin clusters, rather than to the relaxation of the mobile wall spins and of those in domains, as it is usually observed in the more saturated state.

The remainder of the paper is organized as follows: in Sec. II we give a brief description of the experimental setup used for measuring the longitudinal dynamic susceptibility. The results are presented in Sec. III, followed by the discussion in Sec. IV. The summary and the principal conclusions of this paper are contained in Sec. V.

II. EXPERIMENTAL PROCEDURES

The sample under study was an ellipsoid with principal axes of $5.0 \times 3.5 \times 2.5$ mm^3 cut from a good quality single crystal. The short axis which coincided with the easy magnetic direction was always oriented parallel to the static and alternating magnetic fields. So all relaxation experiments probed the dynamics of the order parameter of $CrBr_3$. Figure 1 shows the experimental arrangement. By means of a sapphire rod providing good thermal contact to the Ge thermometer, the sample was placed into an inductance which together with a variable capacitor forms the resonant circuit held at (constant) low temperature. It is weakly coupled to a broad-band magic tee (Anzac type H9 and HH108), separating the reflected from the incident signal. The phases of both voltages were compared in a vector voltmeter (hp 8405 A) detecting changes of the resonance condition caused by a change of $\chi'(\omega)$. By means of the error signal the voltage controlled oscillator (Schomandl MS 200 M) was pulled to the new frequency (counted by hp 5264 L) at constant magnetic field as a function of temperature, or *vice versa*. In the present case of small differences between the frequencies of the empty (ω) and the filled (ω_f) resonator, $\omega - \omega_f \ll \omega$, the dispersion of the external dynamic susceptibility, $\chi'(\omega) = \mathrm{Re}\chi(\omega)$, could be easily determined from

$$\chi'(\omega) = \frac{2}{\eta} \frac{\omega - \omega_f}{\omega} . \qquad (3)$$

In the interesting range of frequencies 10–120 MHz the filling factor η does not depend on ω, because we used the same inductivity. The absolute value of η was determined from an adjustment

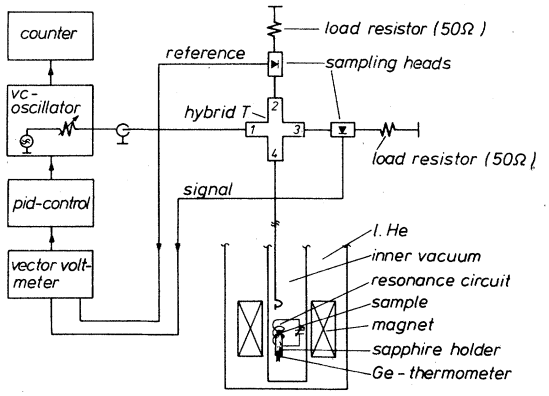

FIG. 1. Block scheme of the arrangement measuring the dispersion of the dynamic susceptibility by frequency counting. pid: proportional integral differential control.

of $\chi'(\omega/2\pi = 10\text{MHz})$ to the isothermal susceptibility at $T \gtrsim 33.5$ K and $B = 0$, where no relaxation could be detected (also supported by the absence of any absorption). The isothermal susceptibility was measured at 78 Hz by a calibrated mutual inductance bridge (Oxford Instruments MIB 70). As an example, Fig. 2 shows $\chi'(\omega)$ curves as measured around T_c in zero magnetic field and calibrated according to Eq. (3). Identifying the susceptibility plateau below T_c with $1/N$, we find, for the demagnetization factor, $N = 0.48(2)$, as compared to 0.47(1) estimated from the sample geometry. An independent check of the calibration has been made by fitting the internal 78-Hz susceptibility, $\chi_{T,i} = (\chi_T^{-1} - N)^{-1}$, to the power law

$$\chi_{T,i}(T; B = 0) = C\epsilon^{-\gamma}, \tag{4}$$

($\epsilon \equiv T/T_c - 1$) using the critical exponent $\gamma = 1.215$ from the magnetization measurements of Ho and Litster.[1] As is illustrated by Fig. 3 reasonable agreement exists between Eq. (4) and the experiment at $\epsilon \lesssim 0.07$. The critical amplitude $C = 0.023(4)$ also is consistent with 0.025 from Ref. 1, thus confirming the calibration. On the other hand, our Curie temperature $T_c = 32.60(2)$ K is much closer to the result of Senturia and Benedek,[24] 32.56(2) than to that of Ref. 1, $T_c = 32.844$ K.

Also by the mutual-inductance technique we measured the isothermal zero-field susceptibility

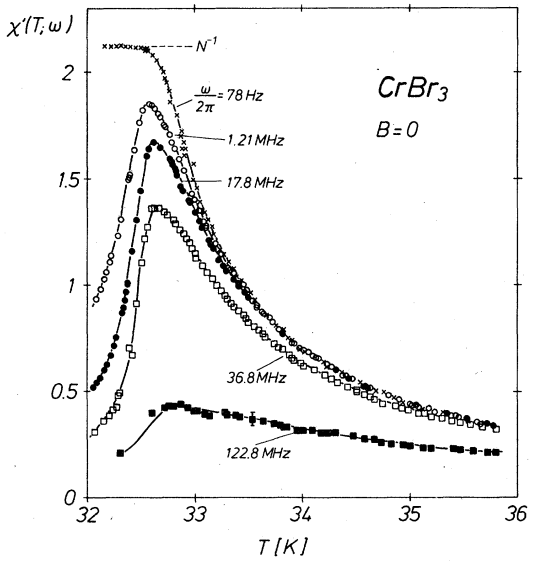

FIG. 2. Temperature dependence of the zero-field dispersion of $CrBr_3$ around the Curie temperature, evaluated from recorded resonance frequencies [cf. Eq. (3)] and, at 78 Hz, from a mutual inductance measurement. The rf field is aligned parallel to the easy magnetic axis. Full curves are drawn as guides to the eye.

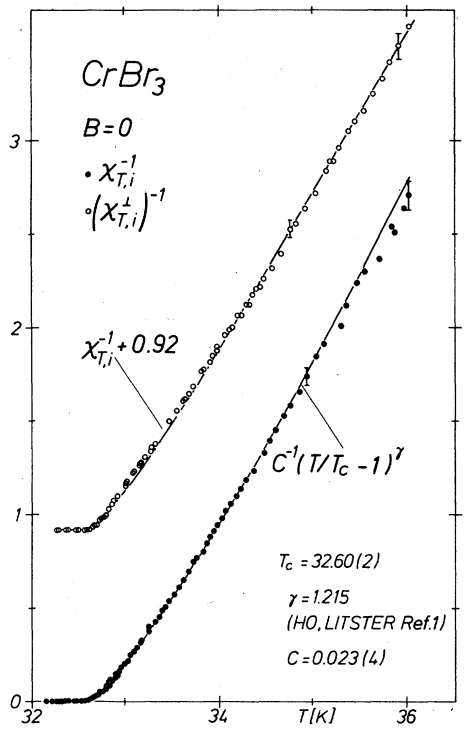

FIG. 3. Inverse of the *internal* zero-field susceptibility of $CrBr_3$ measured at 78 Hz parallel and perpendicular to the easy axis. Full curve through $\chi_{T,i}^{-1}$ represents a power-law fit, assuming $\gamma = 1.215$ from Ref. 1. The inverse of the perpendicular susceptibility can be described by displacing $\chi_{T,i}^{-1}$ by $\Delta\chi^{-1} = 0.92$.

$\chi_T^{\perp}$ in the heavy direction. As is evident from Fig. 3, the internal values $(\chi_{T,i}^{\perp})^{-1} = (\chi_T^{\perp})^{-1} - N^{\perp}$ can adequately be described by displacing the inverse of the measured order parameter susceptibility according to

$$[\chi_{T,i}^{\perp}(T, B = 0)]^{-1} = [\chi_{T,i}(T, B = 0)]^{-1} + \Delta\chi^{-1}, \tag{5}$$

where the displacement term $\Delta\chi^{-1} = 0.92$ is experimentally defined. We shall refer to this result in Sec. IV.

III. ORDER-PARAMETER RELAXATION

The frequency dependencies of the dispersion in zero and finite magnetic fields are reproduced in Fig. 4 for $\omega/2\pi \geq 1$ MHz. It is obvious that all curves are very closely described by the real part of the Debye function [Eq. (1)]

$$\chi'(\omega; \epsilon, B) = \frac{\chi_{p1}(\epsilon, B)}{1 + [\omega/\Gamma(\epsilon, B)]^2}, \quad \frac{\omega}{2\pi} \geq 1 \text{ MHz}. \tag{6}$$

As noted above, this behavior corresponds to a time-dependent magnetization, tending exponentially towards equilibrium with characteristic time

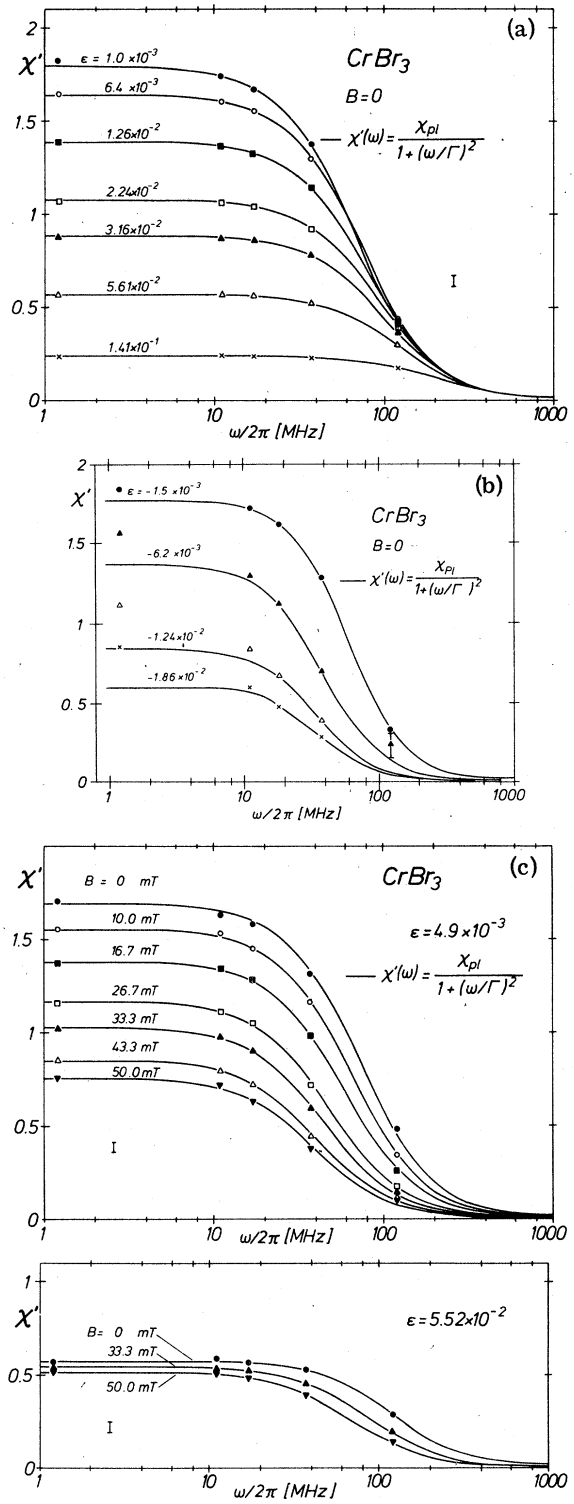

FIG. 4. Frequency dependence of the longitudinal magnetic dispersion at various temperatures in zero field above (a) and below (b) T_c and in finite magnetic fields parallel to the easy axis of $CrBr_3$ (c). Full curves indicate fits to the real part of the Debye function, Eq. (6).

$1/\Gamma$. In the following we will consider, separately for zero and finite magnetic field, the critical behavior of the quantities of primary interest, i.e., the relaxation rate $\Gamma(\epsilon, B)$ and of the plateau susceptibility $\chi_{pl}(\epsilon, B)$, both extracted from fitting the measured dispersion curves to Eq. (6). Because $\Gamma(\epsilon, B)$ still depends on the shape of the sample, it is meaningful to correct it for demagnetization in order to deal with the internal relaxation rate Γ_i, the quantity usually discussed by the theory. For this purpose we use the general relation between external and internal dynamic susceptibilities

$$[\chi(\omega)]^{-1} = [\chi_i(\omega)]^{-1} + N$$

(see e.g., Ref. 25) which, in a frequency region, where $\chi(\omega)$ is described by a single Debye function, like Eq. (6), leads to

$$\Gamma_i(\epsilon, B) = \Gamma(\epsilon, B)[1 - N\chi_{pl}(\epsilon, B)] \,. \tag{7}$$

A. Results in zero magnetic field

We first consider the measured amplitudes of the Debye function depending on the state of equilibrium to which the magnetization relaxes. In the right-hand part of Fig. 5 $\chi_{pl}(\epsilon > 0, B = 0)$ is compared to the isothermal susceptibility measured at 78 Hz. Apparently both susceptibilities agree up to $\chi_T \approx 1.2$, i.e., the whole magnetization of the sample reaches thermal equilibrium during $1/\Gamma$ for $\epsilon > 0.02$. Near T_c, however, χ_{pl} remains below χ_T with a maximum difference of 15% at T_c. In order to obtain more information about this effect we carried out an additional dispersion

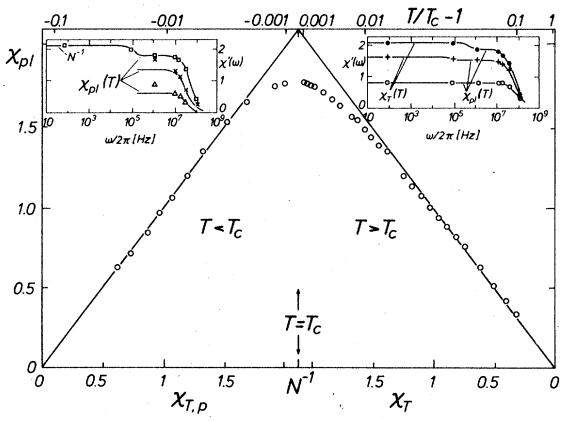

FIG. 5. Comparison between the isothermal susceptibilities χ_T and $\chi_{T,p}$, calculated according to Eq. (4) ($T > T_c$) and Eq. (8) ($T < T_c$), and χ_{pl}, determined from fits of the high-frequency dispersion to the Debye law (Eq. 5). Near T_c a second relaxation region appears, indicating a dynamical rounding [cf. insets, showing $\chi'(\omega)$ at $\epsilon = 0.0316$ (○), 0.0064 (+), 0.0010 (●), −0.0015 (□), −0.0062 (×), −0.0186 (△)].

measurement at 90 kHz by means of a double T bridge.[26] As illustrated by the inset to Fig. 5 (right-hand side) a new relaxation appears, establishing thermal equilibrium between the whole magnetic system and the lattice at lower frequencies only. The results at 90 kHz indicate the relaxation rate of this slow process to be of some orders of magnitude smaller than Γ.

Passing to the ferromagnetic part of the critical region, we find from the left-hand side of Fig. 5 (inset) the amplitude of this slow relaxation process, $1/N - \chi_{p1}$, to increase with $|\epsilon|$ because the plateau susceptibility of the fast relaxation in the MHz region χ_{p1} is reduced. Moreover this relaxation is tending to merge with the fast process so that below $\epsilon \approx -0.02$ it is no longer possible to describe the dispersion $\chi'(\omega)$ by a well separated Debye function nor by a sum of two. Thus, restricting ourselves to $\epsilon > -0.02$, we analyzed first $\chi_{p1}(\epsilon < 0, B = 0)$ by adjusting its internal values to the conventional power law

$$\chi_{p1,i}(\epsilon < 0, B = 0) = C'(-\epsilon)^{-\gamma'} , \qquad (8)$$

obtaining a good quality fit for $-0.02 \lesssim \epsilon \lesssim -0.003$ with $\gamma' = 1.29(5)$ and $C' = 0.0053(5)$. Within the error margins our results are consistent with those obtained from magnetization data under the scaling constraint $\gamma = \gamma' = 1.215(15)$,[1] arriving at $C' = 0.0061(5)$. Thus, it is tempting to identify the plateau susceptibility measured below T_c with the (external) isothermal paramagnetic susceptibility, $\chi_{p1} = \chi_{T,p}(\epsilon < 0, B = 0)$. This would mean, that the relaxation process occuring in the MHz region brings the paramagnetic spins to equilibrium, while the other spins, being strongly coupled within long-range-ordered clusters[23] are still blocked. On the left-hand side of Fig. 5, χ_{p1} is plotted vs $\chi_{T,p}$ as calculated from the power law [Eq. (8)]. Similarly, as above T_c, the agreement is rather good up to $\chi_{T,p} = 1.5$ ($\epsilon \lesssim -0.004$), from whereon χ_{p1} levels off joining the corresponding curve from $T \geq T_c$ at the Curie temperature. At present we cannot offer a satisfying explanation for this "dynamic rounding" in the transition region (note that the rounding of the static susceptibility occurs much nearer to T_c for $\epsilon < 0.001$). Perhaps this effect can be attributed to sample inhomogeneities having a different relaxation behavior near T_c, which may have been created near the surface during the shaping procedure of the rather soft $CrBr_3$ material (tending to cleave perpendicular to the hexagonal axis).

The temperature dependence of the internal relaxation rate Γ_i [Eq. (7)] in zero magnetic field is displayed in Fig. 6. Its characteristic feature is the critical slowing down leading to an absolute minimum of Γ_i at T_c which takes, however, a

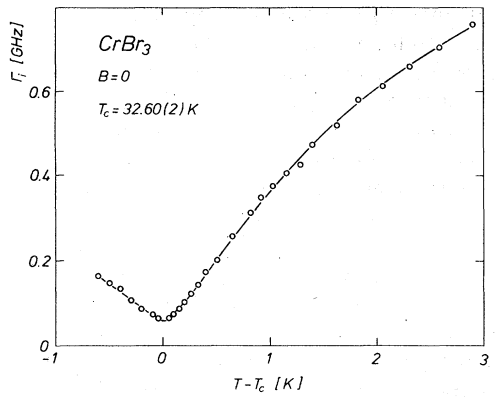

FIG. 6. Temperature dependence of the internal relaxation rate Γ_i around T_c in zero magnetic field. Full curve serves as guide to the eye.

finite value due to the rounding of χ_{p1} [cf. Eq. (7)]. As mentioned above, in the ferromagnetic region Γ_i becomes less well defined with decreasing temperature ($\epsilon < -0.02$), because $\chi'(\omega)$ at $\omega/2\pi \gtrsim 1$ MHz can no longer be explained by a single Debye function.

B. Relaxation in finite magnetic field ($T > T_c$)

In Fig. 7 the field dependence of the internal relaxation rate is presented for several temperatures near T_c. For the sake of consistence, the magnetic field shown there is also corrected for the demagnetizing field, $B_i = B - NM(\epsilon, B)$, with the help of the magnetization data of Ho and Litster.[1] Figure 7 shows us qualitatively quite different behaviors of Γ_i. At $\epsilon > 0.01$ the decay rate decreases slowly with rising field, whereas Γ_i becomes practically independent of B_i at $\epsilon \approx 0.01$. In the immediate vicinity of the Curie

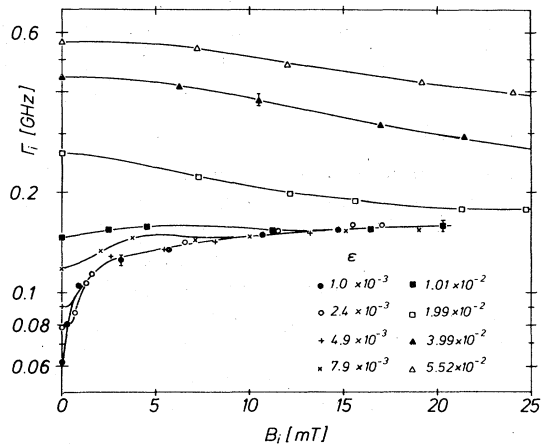

FIG. 7. Internal relaxation rate of the magnetization vs. internal magnetic field at different temperatures above T_c.

temperature Γ_i exhibits a steep increase with magnetic field, which soon reaches a limiting value of 0.16 GHz being independent of B_i and ϵ.

The behavior of χ_{pl} in finite magnetic fields is shown in Fig. 8. For reasons of comparison the isothermal susceptibility $\chi_T(\epsilon, B)$ calculated from the existing magnetization data[1] is indicated, too. At first, i.e., at low fields, the difference between χ_T and χ_{pl} becomes larger with increasing B. This would be consistent with the conclusion that near T_c the magnetic relaxation of $CrBr_3$ is dominated by spin-spin interactions[27] establishing thermal equilibrium within the spin-system only, and, therefore, the plateau susceptibility should be equal to the adiabatic susceptibility of the spin-system. According to thermodynamics, χ_{ad} is given by $\chi_T - T(\partial M/\partial T)_B^2/c_B\mu_0$ with $(\partial M/\partial T)_B^2$ increasing from zero (at $T \geq T_c$) when a magnetic field is applied. Thus χ_{ad} remains always smaller than χ_T in finite magnetic fields above T_c. Unfortunately, the existing data about the specific heat at constant field c_B (Ref. 28) are not precise enough near T_c to allow a reliable estimate of the difference $\chi_T - \chi_{ad}$. In Sec. IV, however, which discusses the decay rate in more detail, we will obtain additional arguments favoring a dominance of the spin-spin interactions in the critical relaxation. At large magnetic fields χ_T and χ_{pl} tend to coincide which cannot be explained by the disappearance of $(\partial M/\partial T)_B^2/c_B$ alone. This result, of course, also requires that the low-frequency relaxation process observed in low fields has merged in the relaxation region of the bulk of the susceptibility located between 10 and 100 MHz, similarly as it is seen in some distance from the critical point at zero field (cf. Fig. 5).

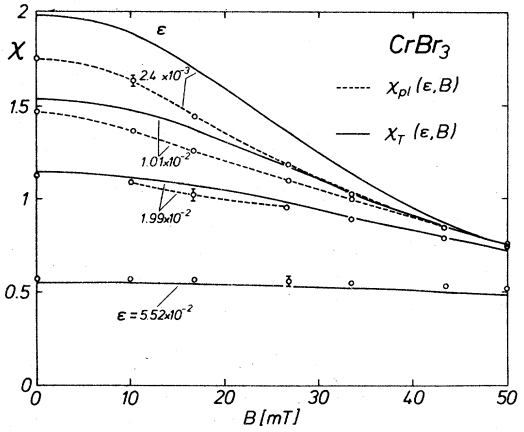

FIG. 8. Magnetic-field dependence of the plateau susceptibility χ_{pl} determined from Debye fits to the high-frequency relaxation region [Eq. (5)]. For comparison are shown the isothermal susceptibilities calculated from the magnetization data of Ref. 1. Error bars contain the combined uncertainties of measurement and calculation of χ_T.

IV. DISCUSSION

As noted in the introduction, it is the kinetic (transport) coefficient of the homogeneous magnetization

$$L \equiv \Gamma\chi_{pl} ,$$

which is of primary concern in critical dynamics. By considering L instead of Γ, we eliminate the thermodynamic slowing down caused by the static susceptibility and, moreover, we deal with a quantity which, as we shall see, is intimately related to the dynamics of the magnetic fluctuations. We also recall the fact that L constitutes a material constant, independent of its (ellipsoidal) form, in which field effects enter only via the internal magnetic field. As a principal result of this work Fig. 9 represents the temperature dependence of the kinetic coefficient of $CrBr_3$ at constant B_i in the critical region above T_c as well as at $B = 0$ below T_c. On the paramagnetic side of T_c and in zero field, L decreases gradually for $\epsilon \gtrsim 0.01$ while in the immediate vicinity of T_c it behaves noncritically. This is in contrast to the region below T_c [Fig. 9(b)], where L diminishes approximately like $(-\epsilon)^{-0.78} \propto \chi_{pl,i}^{0.60}$ for $\epsilon < -0.004$, i.e.,

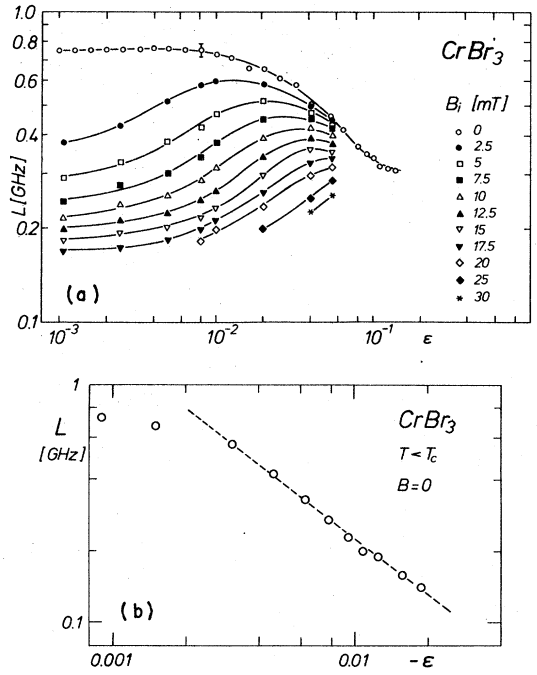

FIG. 9. Kinetic coefficient of the homogeneous easy-axis magnetization, $L = \Gamma\chi_{pl}$ vs reduced temperature: (a) at different internal magnetic fields above T_c; and (b) at zero field below T_c.

outside the range of the dynamic rounding.

In finite fields at $T > T_c$, L exhibits a simpler behavior than Γ_i: the overall effect of B_i is to reduce the kinetic coefficient, especially in the immediate neighborhood of T_c. Thus the steep rise of Γ_i at small ϵ and B_i in Fig. 7 must entirely be ascribed to the strong decrease of the thermodynamic factor $\chi_{\mathrm{pl},i}^{-1}$, which overrides the suppression of L by the magnetic field. With increasing ϵ the field effect on L becomes less significant which appears to be consistent with Kawasaki's idea[5] outlined in the introduction: due to the growing decay rate of the critical fluctuations, $\Gamma_{q\approx\xi^{-1}}$, the field-induced Larmor precession loses its importance for the critical dynamics. In Secs. IV A and IV B these behaviors of $L(\epsilon, B)$ are explained in more detail, separately for the cases of zero- and finite-magnetic field at $T > T_c$.

A. Kinetic coefficient in zero field

Recently Maleev[9] and Finger,[11] employing different theoretical methods, calculated the kinetic coefficient of spin-spin relaxation in ferromagnets considering simultaneously anisotropic exchange and dipolar interaction on a cubic lattice. For uniaxial systems both treatments arrive qualitatively at the same results (in contrast to the planar and Heisenberg cases). We will base our discussion on Finger's mode-coupling calculation because this formulation also allows quantitative estimates for L, which in the case of the cubic ferromagnets EuO (Ref. 29) and $CdCr_2Se_4$,[16] have been shown to be in extremely good agreement with experiment.

The essential quantities entering in the mode-coupling equations are the static magnetic susceptibilities at long wavelengths describing the correlations between the modes of the magnetization components $\vec{M}_{\vec{q}\to 0}$ parallel and perpendicular to the easy axis

$$\chi_T(\vec{q}\to 0) = \frac{\chi_{T,i}/\xi^2}{\xi^{-2} + q^2 + q_d^2(q_z/q)^2} \ , \tag{9}$$

$$\chi_T^{\perp}(\vec{q}\to 0) = \frac{\chi_{T,i}/\xi^2}{\xi^{-2} + q^2 + q_a^2} \ , \tag{10}$$

respectively. q_d and q_a represent wave vectors characterizing the dipolar and exchange anisotropies, and the critical behavior of L depends on whether the inverse correlation length ξ^{-1} is small or large compared to q_d and q_a. In the latter case, both anisotropies do not affect the critical correlations ($q \approx \xi^{-1}$) and their dynamics are dominated by exchange-induced isotropic spin diffusion. In this region, the kinetic coefficient of the homogeneous magnetization is expected to exhibit a "critical speeding up," first predicted by Kawasaki[5] and Huber.[7] From Finger's work[11] we obtain

$$L_{\mathrm{iso}} = Q q_d^{1/2} (q_d \xi)^{7/2}, \quad \xi^{-2} \gg q_a^2, q_d^2 \ , \tag{11}$$

where

$$Q = \hbar^{1} \left(\frac{V_{\mathrm{spin}}}{8\pi^3} \frac{\chi_{T,i}}{\xi^2} G_d k_B T \right)^{1/2} , \tag{12}$$

with $G_d = (g\mu_B)^2 / V_{\mathrm{spin}} \mu_0$. Roughly speaking this speeding up can be ascribed to the suppression of the well-known exchange narrowing of the dipolar forces responsible for the magnetic relaxation by the increasing short-range order when approaching T_c.[30] In the reverse case, i.e., near T_c (where $\xi^{-1} \ll q_a, q_d$), both anisotropies reduce the degrees of freedom for the critical fluctuations. This leads to the so-called conventional behavior[31] of the relaxation rate $\Gamma \propto \chi_T^{-1}$ defined by a non-critical kinetic coefficient. This behavior is definitely observed on $CrBr_3$ below $\epsilon = 0.01$. Riedel and Wegner[6] initially derived this result for isotropic exchange. For the case of uniaxial exchange Finger's generalized treatment[11] yielded the following explicit equations for L depending on the relative strengths of the exchange and dipolar anisotropy:

$$L_d = Q q_d^{1/2}, \quad \xi^{-2} \ll q_a^2 \ll q_d^2 \ , \tag{13}$$

$$L_a = L_d (q_d/q_a)^{7/2}, \quad \xi^{-2} \ll q_d^2 \ll q_a^2 \ . \tag{14}$$

In order to compare these findings quantitatively with $CrBr_3$, we have to take account of the hexagonal lattice structure and other sources of anisotropy (e.g., single ion a [32]) in this ferromagnet, all differing from the underlying theoretical model. Near T_c ($\xi^{-2} \ll q_a^2, q_d^2$), however, it is possible (see Appendix) to approximate the correlation functions of $CrBr_3$ by Eqs. (9) and (10), and to estimate the parameters in Eqs. (9) and (10) by the following expressions:

$$\chi_{T,i}/\xi^2 = G_d/J_2 \ , \tag{15}$$

$$q_d^2 = (G_d/J_2)[1/(1+\chi_{T,i}^{\perp})] , \tag{16}$$

$$q_a^2 = (G_d/J_2)(1/\Delta\chi) \ . \tag{17}$$

Here J_2 denotes the second spatial moment of the exchange interactions defined by Eq. (A7), describing the dispersion of the transverse (equal to "easy dipolar"[19,33]) modes of the order parameter, i.e., of $S_{\vec{q}}^x$ propagating perpendicularly to the z axis. The exchange integrals in $CrBr_3$ have been carefully measured by Samuelsen *et al.*[22] up to the fifth nearest neighbors leading to $J_2 = 171$ (10) μeV nm^2. The numerical values involved in the

TABLE I. Comparison between measured and calculated [Eqs. (12)–(17)] kinetic coefficients of the uniaxial ferromagnets $CrBr_3$ and $GdCl_3$(Ref. 17). The following columns contain the parameters involved in the calculation being estimated by using the magnetic coupling constants from Refs. 22 and 34. All data refer to $T > T_c$, $B = 0$.

	T_c [K]	L [GHz]	L_a [GHz]	L_d [GHz]	G_d [μeV]	J_2 [μeVnm^2]	q_a^2 [nm^{-2}]	q_d^2 [nm^{-2}]
$CrBr_3$	32.60(2)	0.76(4)	0.54(9)	1.73(5)	26.6	171(10)	0.143(9)	0.074(4)
$GdCl_3$	2.211(1)	19(2)	6(1)	11(1)	28.0	2.20(8)	4.4(8)	3.3(4)

calculation of the kinetic coefficient L_a and L_d are compiled in Table I. For $CrBr_3$ it is evident that $q_a^2 > q_d^2$, but the limiting condition of very strong uniaxial anisotropy underlying Eq. (14) is not fulfilled. Nonetheless, calculating the kinetic coefficient from Eq. (14) we obtain $L_a = 0.54$ (9) GHz, being only slightly smaller than the measured value $L = 0.76(4)$ GHz. L is, moreover, bounded from above by $L_d = 1.73(5)$ GHz, calculcated from Eq. (13) for the other limiting case of dominant dipolar anisotropy. Obviously our measured L lies within the crossover region between uniaxial and dipolar anisotropy and we argue that it can perhaps be estimated to better accuracy by a numerical treatment of the appropriate mode-coupling equations instead of using the asymptotic formula (14). This method was utilized recently by Raghavan and Huber[10] to calculate the critical relaxation in the exchange to dipolar crossover region of cubic ferromagnets, the results of which were shown recently to be consistent with measurements on EuO,[29] and $CdCr_2Se_4$.[16]

Table I also includes corresponding data of the low-temperature ferromagnet $GdCl_3$,[17] where the exchange coupling is known up to the second nearest neighbors.[34] Owing to the smaller exchange interaction and uniaxial anisotropy in this system the kinetic coefficient is predicted to be more than one order of magnitude larger than in $CrBr_3$ and also should be closer to L_d than to L_a. From Table I it is evident that again the experimental value confirms these expectations.

It might be interesting to estimate the temperature range where the conventional behavior should hold for $CrBr_3$. Using Eq. (15), the inequalities $\xi^{-2} \ll q_d^2, q_a^2$ can be reduced to $\chi_{T,i} \gg 1 + \chi_{T,i}^{\perp}$, which ceases to be valid for $\chi_{T,i}(\epsilon^*) \approx 1 + \chi_{T,i}^{\perp}(\epsilon^*)$. Inserting our experimental susceptibilities for $CrBr_3$ we obtain $\epsilon^* = 0.03$, where the measured L has in fact already left the plateau (Fig. 9). Obviously, around ϵ^* a dynamic crossover occurs which is complicated by the fact that not only do the dipolar and uniaxial anisotropies become less important here for the critical fluctuations ($q \approx \xi^{-1}$), but that just in this same region $CrBr_3$ becomes a quasi-two-dimensional Heisenberg ferromagnet[35] because of the weakness of J_{ij} parallel to the c axis.[36] The two-dimensional (2-D) short-range order can be observed, e.g., by the significant reduction of the Curie temperature relative to the mean-field value $T_0 = 73.5$ K following from Eq. (A5) with exchange parameter of Ref. 22. One may also conjecture, that the critical exponent of the static susceptibility, $\gamma = 1.215$, represents an effective value[19] in the crossover between 2-D Heisenberg [large γ (Ref. 35)] and uniaxial dipolar ($\gamma = 1.07$),[37] rather than a short-range 3-D Ising exponent ($\gamma = 1.25$). The critical behavior of L in such a 2-D ferromagnet has not yet been calculated. Richards[38] argued that Γ should strongly diverge approaching T_c, which, however, is not born out by our data at large ϵ.

B. Kinetic coefficient in finite field

In order to compare the magnetic-field effect on the kinetic coefficient of $CrBr_3$ with the results of the corresponding investigation on EuS,[17] where L was found simply to depend on ϵ and B_i via the static susceptibility, i.e., $L(\epsilon, B_i) = f(\chi_{pl,i}(\epsilon, B_i))$ we have plotted L vs $\chi_{pl,i}$ in Fig. 10. It is evident that, in contrast to EuS, where all data in this representation collapsed onto a single curve, the

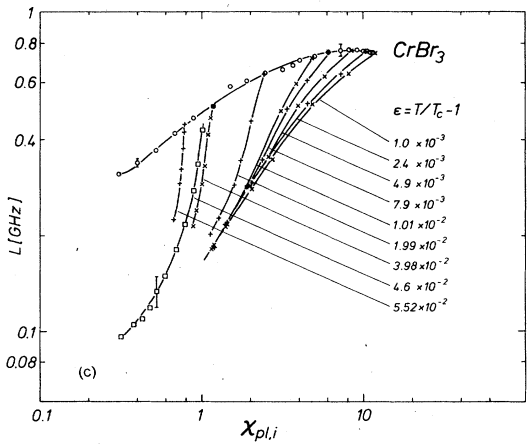

FIG. 10. Kinetic coefficient of the order parameter vs internal plateau susceptibility. Upper curve corresponds to $B = 0$, all others are for constant temperature and internal fields $B_i \leq 25$ mT, except for $\epsilon = 0.046$, where B_i extends up to 165 mT.

transport coefficient of $CrBr_3$ does not show this simple behavior. Especially in low fields there is a considerable divergence of the L isothermes from the curve at $B=0$. Looking for a confluence, one finds an indication at small ϵ and large B_i, but unfortunately there are not enough data to support this trend. Thus, it is convenient to assume that the different field effect on the critical dynamics of $CrBr_3$ is associated with its uniaxial anisotropy and/or with the quasi-two-dimensionality of the magnetic system. The latter should manifest itself for $\chi_{T,i} < 2$, where the magnetic field effect on the $L - \chi_{pl,i}$ plot appears to be strong. As noted above no theory is available to date for critical relaxation in 2-D ferromagnets, neither for $B=0$ nor finite fields.

On the other hand, for $\epsilon \ll \epsilon^*$, $CrBr_3$ can be characterized as a 3-D uniaxial dipolar ferromagnet and, thus it might be appropriate to compare the results from this region to Maleev's calculation.[9] Recalling the steep increase of Γ_i in small fields, $B_i < 1$ mT (Fig. 7), his prediction that Γ_i should increase anomalously in small fields, appears to be satisfied. However, considering the kinetic coefficient we find from Maleev's work [Eq. (48)] that L also increases with growing magnetic field. Such a behavior is inconsistent with our observation (Fig. 9) of a strong reduction of L even in very small fields.

A final remark should be devoted to previous EPR work by Seehra and Gupta[14] on $CrBr_3$ near its Curie temperature. The authors based their discussion of the critical behavior of the linewidth on the zero field estimates for the relaxation rates of the order parameter and of the perpendicular magnetization. Our results show, however, that this assumption leads to incorrect conclusions near T_c ($\epsilon < \epsilon^*$), because the critical behavior of Γ is strongly modified even in moderate fields up to 20 mT (Fig. 7) while the EPR measurements were carried out at 320 mT. In the quasi-two-dimensional region of $CrBr_3$ ($\epsilon > \epsilon^*$) L also significantly depends on the field, indicating that Kawasaki's condition (deduced for isotropic 3-D ferromagnets) as applied to $CrBr_3$ in Ref. 14 does not hold here.

V. SUMMARY AND CONCLUSIONS

In the preceding sections we have described and discussed the temperature and field dependence of the relaxation rate Γ of the order parameter around the Curie temperature of the uniaxial ferromagnet $CrBr_3$, where Γ was extracted from the Debye shape of $\chi'(\omega)$ at $\omega/2\pi \geq 1$ MHz. The observed conventional critical behavior of Γ in zero magnetic field for $0 < \epsilon < 0.01$ is in full qualitative and good quantitative agreement with recent results of the mode-coupling approach assuming the spin-spin relaxation to be dominant. Though no quantitative estimate exists so far for the magnitude of the spin-lattice interaction in $CrBr_3$, the corresponding kinetic coefficient L_{sl} is expected to be essentially independent of temperature in the critical region[27] and, therefore, we can infer from our results, Fig. 9, that $L_{sl} < 0.3$ GHz. Thus, L_{sl} is significantly smaller than the measured $L = 0.76$ GHz, so that our quantitative comparison for L is not seriously affected. Somewhat above, at $\epsilon > 0.01$, a dynamic cross over to nonconventional behavior is found which might originate from the increasing two dimensionality of the critical fluctuations.

When passing T_c we found Γ to behave continuously and most likely to arise at $T < T_c$ from the relaxation of paramagnetic spins in the ferromagnetic phase which would be consistent with an earlier observation on $RbCu_2Br_4 \cdot 2H_2O$.[23] In contrast to above T_c, Γ exhibits unconventional behavior ($\Gamma_i \propto \chi_{pl,i}^{-0.4}$) which remains unexplained as well as the appearance of a second relaxation process in the vicinity of T_c, $-0.004 \leq \epsilon \leq 0.02$, denoted as dynamical rounding, affecting a small part of the total susceptibility.

Applying a magnetic field, the relaxation rate rises steeply at low fields for $\epsilon < 0.03$, while at $\epsilon > 0.03$ a gradual decrease is observed. Both results differ distinctly from the field effect as observed on the cubic dipolar ferromagnet EuS.[17] They cannot be understood within the frame of existing ideas and have to be clarified. In view of these results it might also be advantageous to revise the simple model of $CrBr_3$ as a 3-D–Ising ferromagnet so far used to explain the critical behavior of the static magnetization[1] in the $B_i - \epsilon$ region examined here.

ACKNOWLEDGMENTS

We are much indebted to Dr. P. Radhakrishna, Laboratoire L. Brillouin, Gif-sur-Yvette, for providing us with the single crystal of $CrBr_3$. It is a pleasure to thank Professor G. Weber for a critical reading of the manuscript, G. Raffius for help with the 78-Hz measurement and W. Finger for valuable discussions. This work was a project of the Sonderforschungsbereich 65 "Festkörperspektroskopie" financed by special funds of the Deutsche Forschungsgemeinschaft.

APPENDIX: STATIC MAGNETIC CORRELATIONS IN $CrBr_3$ ($T > T_c$)

From Samuelsen's *et al.* neutron work[22] and Bené's EPR measurements on Cr^{3+} pairs on BiI_3,[32]

it follows that the magnetic Hamiltonian of $CrBr_3$ consists of a dominant Heisenberg exchange plus an anisotropic part

$$H = -\sum_{i \neq j} J_{ij} \vec{S}_i \cdot \vec{S}_j + H_A \, .$$

H_A contains contributions from dipolar, exchange, and single-ion interactions

$$H_A = \sum_{i \neq j} \vec{S}_i \cdot \vec{D}_{ij} \cdot \vec{S}_j - \sum_{i \neq j} J^z_{ij} S^z_i S^z_j - \sum_i D (S^z_i)^2 , \quad \text{(A1)}$$

$$D^{\alpha\beta}_{ij} = \frac{1}{2} \frac{(g\mu_B)^2}{4\pi\mu_0} \left(\frac{\delta^{\alpha\beta}}{r^3_{ij}} - 3 \frac{r^\alpha_{ij} r^\beta_{ij}}{r^5_{ij}} \right) . \quad \text{(A2)}$$

While accurate numbers for J^z_{ij} and D are not yet available, it seems to be certain that the exchange is responsible for the alignment parallel to c ($J^z > 0$), while the smaller crystalline $D < 0$ and dipolar fields both direct the magnetization into the perpendicular direction. To discuss the critical correlations of such an uniaxial ferromagnet with dipolar interaction we follow the treatment given in detail by Ref. 39. As starting point we use the mean-field approximation (MFA, e.g., Ref. 40), where the susceptibilities of the order parameter and of the heavy modes of the magnetization can easily be shown to take the form

$$\chi_T(\vec{q} \to 0) = \chi^{zz}_T(\vec{q} \to 0) = \frac{\lambda}{T - T_0 + \rho_\perp q^2_\perp + \rho_z q^2_z + \lambda / [1 + \chi^\perp_{T,i} (q_\perp/q)^2](q_z/q)^2} \quad \text{(A3)}$$

$$\chi^\perp_T(\vec{q} \to 0) = \tfrac{1}{2} [\chi^{xx}_T(\vec{q} \to 0) + \chi^{yy}_T(\vec{q} \to 0)] = \lambda / (T - T^\perp_0 + \rho_\perp q^2_\perp + \rho_z q^2_z) \, . \quad \text{(A4)}$$

Here λ denotes Curie's constant, $S(S+1)G_d/3k_B$, and the Curie temperature is given by

$$T_0 = \frac{2S(S+1)}{3k_B} \left(\sum_{j(\neq i)}^{\text{L.S.}} (J_{ij} + J^z_{ij} + D^{zz}_{ij}) + D \right) \quad \text{(A5)}$$

and

$$T^\perp_0 = \frac{2S(S+1)}{3k_B} \sum_{j(\neq i)}^{\text{L.S.}} [J_{ij} + \tfrac{1}{2}(D^{xx}_{ij} + D^{yy}_{ij})] , \quad \text{(A6)}$$

where the summations are to be carried out within the Lorentz sphere. Similarly the expansion coefficients of the $\vec{q}$-dependent terms are determined by the magnetic interactions: they reduce to the simple form in Eqs. (A3) and (A4) when the small part H_A is neglected, with

$$\rho_\perp = \frac{S(S+1)}{3k_B} \sum_{j(\neq i)} J_{ij} (\vec{q}_\perp \cdot \vec{r}_{ij})^2 q^{-2}_\perp = \frac{S(S+1)}{3k_B} J_2 \, , \quad \text{(A7)}$$

$$\rho_z = \frac{S(S+1)}{3k_B} \sum_{j(\neq i)} J_{ij} (r^z_{ij})^2 = \frac{S(S+1)}{3k_B} J^z_2 \, . \quad \text{(A8)}$$

With these preparations we can now write

$$\chi_T(\vec{q} \to 0) = \frac{G_d}{J_2} \Bigg/ \left[\frac{T - T_0}{\rho_\perp} + q^2 + \frac{G_d/J_2}{1 + \chi^\perp_{T,i} (q_\perp/q)^2} \times \left(\frac{q_z}{q} \right)^2 + \left(\frac{J^z_2}{J_2} - 1 \right) q^2_z \right] , \quad \text{(A9)}$$

$$\chi^\perp_T(\vec{q} \to 0) = \frac{G_d}{J_2} \Bigg/ \left[\frac{T - T_0}{\rho_\perp} + q^2 + \frac{T_0 - T^\perp_0}{\rho_\perp} + \left(\frac{J^z_2}{J_2} - 1 \right) q^2_z \right] . \quad \text{(A10)}$$

Approaching T_c, we know from the theory[19] that the first term of the denominator in Eq. (A9) is renormalized by the critical fluctuations, whereas the other entities are assumed to retain their mean-field values. Consequently, by comparing Eq. (A9) with Eq. (9), the homogeneous susceptibility is found to be related to the correlation length ξ by Eq. (13). Due to the depolarization term in Eq. (A9) $\propto (q_z/q)^2$, arising from the long-range part of the dipolar interaction, it is only the correlation function between the transverse order-parameter modes, $S^z_{\vec{q}_\perp \vec{e}_z}$ that diverges near T_c. Thus near T_c one is allowed to neglect $q^2_z \to 0$ in Eq. (A9) and to replace $(q_\perp/q)^2$ by one, so that we in fact arrive at the form of Eq. (9) assumed by the mode-coupling theory with q^2_d given by Eq. (16). It might be noteworthy that both results, Eqs. (15) and (16), have been found to be consistent with experimental findings on the uniaxial ferromagnets $LiTbF_4$,[41,42] and $GdCl_3$,[39] respectively. Applying the same arguments to $\chi^\perp_T(\vec{q} \to 0)$ one obtains, from (A10), Eq. (20) with $q^2_d = (T_0 - T^\perp_0)/\rho_\perp$. Since the parameters of H_A involved in $T_0 - T^\perp_0$ [Eqs. (A5) and (A6)] are not yet known precisely, we measured the homogeneous perpendicular susceptibility

near T_c. The experimental result, Eq. (5), is in full agreement with what follows from Eq. (A10):

$$[\chi_T^{\perp}(q=0)]^{-1} \equiv (\chi_{T,i}^{\perp})^{-1} = (\chi_{T,i})^{-1} + (\xi^2/\chi_{T,i})q_a^2, \quad \text{(A11)}$$

from which one obtains directly Eq. (17) using Eq. (15). Comparing our measured value of $\Delta\chi = \chi_{T,i}^{\perp}(T_c, M=0) = 1.09$ with $\chi_{T,i}^{\perp}(T_c, 185\ \text{mT}) = M/B_A = 0.83$, following from Dillons[43] measurement of the anisotropy field by ferromagnetic resonance at 23.4 GHz, $B_A(T_c) = 200$ mT, and the corresponding magnetization $M(T_c) = 185$ mT,[43] a slight discrepancy is observed which indicates that $\chi_{T,i}^{\perp}$ diminishes with increasing magnetization. This is also supported by the neutron[22] and FMR results both yielding $B_A = 650$ mT at 6 K, where $M \approx 340$ mT, and thus $\chi_{T,i}^{\perp}(6\ \text{K},\ 340\ \text{mT}) \approx 0.52$.

[1]J. T. Ho and J. D. Litster, Phys. Rev. Lett. 22, 603 (1969); Phys. Rev. B 2, 4523 (1970).
[2]M. N. Deschizeaux and G. Develey, J. Phys. (Paris) 32, 2–3 Suppl. C1-648 (1971).
[3]J. S. Kouvel and J. B. Comley, Phys. Rev. Lett. 20, 1237 (1968).
[4]M. Menyuk, K. Dwight, and T. B. Reed, Phys. Rev. B 3, 1689 (1971).
[5]K. Kawasaki, Prog. Theor. Phys. 39, 285 (1967); in *Phase Transitions and Critical Phenomena*, edited by C. Domb and M. S. Green (Academic, New York, 1976), Vol. VA, p. 166.
[6]E. Riedel and F. Wegner, Phys. Rev. Lett. 24, 730 (1970).
[7]D. L. Huber, J. Phys. Chem. Solids 32, 2145 (1971).
[8]S. V. Maleev, Phys. Lett. 47A, 111 (1974); Phys. Lett. 55A, 491 (1976).
[9]S. V. Maleev, Zh. Eksp. Teor. Fiz. 69, 1398 (1975) [Sov. Phys. JETP 42, 713 (1976)].
[10]R. Raghavan and D. L. Huber, Phys. Rev. B 14, 1185 (1976).
[11]W. Finger, Phys. Lett. 60A, 165 (1977).
[12]M. B. Salamon, Phys. Rev. 155, 224 (1967); F. Spörel and F. Biller, Solid State Commun. 17, 833 (1975).
[13]M. S. Seehra and D. L. Huber, in AIP Conf. Proc. 24, 261 (1975); M. S. Seehra and W. S. Sheers, Physica (Utr.) 85B, 142 (1977).
[14]M. S. Seehra and R. P. Gupta, Phys. Rev. B 9, 197 (1974).
[15]P. Burgardt and M. S. Seehra, Phys. Rev. B 16, 1802 (1977).
[16]J. Kötzler and H. v. Philipsborn, Phys. Rev. Lett. 40, 790 (1978).
[17]J. Kötzler, G. Kamleiter and G. Weber, J. Phys. C 9, L 361 (1976).
[18]B. I. Halperin and P. C. Hohenberg, Phys. Rev. 177, 952 (1969); one of us (J.K.) is grateful to Professor M. Suzuki, University of Tokyo, for a valuable discussion about this point.
[19]M. E. Fisher and A. Aharony, Phys. Rev. Lett. 30, 559 (1973); A. D. Bruce, J. Phys. C 10, 419 (1977).
[20]S. Ikeda (private communication).
[21]B. I. Halperin, P. C. Hohenberg and S. K. Ma, Phys. Rev. B 10, 139 (1974).
[22]E. J. Samuelsen, R. Silberglitt, G. Shirane, and J. P. Remeika, Phys. Rev. B 3, 157 (1971).
[23]W. L. C. Rutten and J. C. Verstelle, Physica (Utr.) 86–88B, 564 (1977).
[24]S. D. Senturia and G. B. Benedek, Phys. Rev. Lett. 17, 475 (1966).
[25]W. Finger, Physica (Utr.) 90B, 251 (1977).
[26]E. Blank, diploma work TH Darmstadt (1968) (unpublished); J. Kötzler, Physica (Utr.) 60, 375 (1972).
[27]D. L.Huber and M. S. Seehra, J. Phys. Chem. Solids 36, 723 (1975).
[28]L. D. Jennings and W. N. Hansen, Phys. Rev. 139, A1694 (1965).
[29]J. Kötzler, W. Scheithe, R. Blickhan, and E. Kaldis, Solid State Commun. (to be published, 1978).
[30]K. Tomita and T. Kawasaki, Prog. Theor. Phys. 44, 1173 (1970).
[31]L. van Hove, Phys. Rev. 93, 1374 (1954).
[32]R. W. Bené, Phys. Rev. 178, 497 (1969).
[33]A. I. Larkin and D. E. Khmel'nitzkii, Zh. Eksp. Teor. Fiz. 56, 2087 (1969) [Sov. Phys. JETP 29, 1123 (1969)].
[34]R. J. Birgeneau, M. T. Hutchings, and W. P. Wolf, Phys. Rev. 179, 275 (1969).
[35]L. J. de Jongh and A. R. Miedema, Adv. Phys. 23, 1 (1974).
[36]H. L. Davis and A. Narath, Phys. Rev. 134, A433 (1964).
[37]J. Kötzler and W. Scheithe, Solid State Commun. 12, 643 (1973); R. Frowein and J. Kötzler, Z. Phys. B 25, 279 (1976).
[38]P. M. Richards, in *Proceedings of the International School of Physics E. Fermi*, LIX Course, edited by K. A. Müller and A. Rigamonti (North-Holland, Amsterdam, 1976), p. 574.
[39]J. Kötzler, Phys. Lett. A 59, 483 (1977); (unpublished).
[40]H. Thomas, in *Proceedings of the International School of Physics E. Fermi*, LIX Course, edited by K. A. Müller and A. Rigamonti (North-Holland, Amsterdam, 1976), p. 659.
[41]J. Als-Nielsen, Phys. Rev. Lett. 37, 1161 (1976).
[42]R. Frowein and J. Kötzler (unpublished).
[43]J. F. Dillon, Jr., J. Appl. Phys. 33, 1191 S (1962).
[44]J. F. Dillon, Jr., H. Kamimura, and J. P. Remeika, J. Appl. Phys. 34, 1240 (1963).