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PHENOMENOLOGICAL ANALYSIS OF THE TEMPERATURE DEPENDENT MAGNETIC SUSCEPTIBILITY WITHIN THE FIELD-INDUCED GRIFFFITHS PHASE OF $FeCl_2$ [1]

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In the presence of an applied axial magnetic field H_a the uniaxial antiferromagnet $FeCl_2$ shows fluctuating domain-like antiferromagnetic correlations above the phase boundary $T_c(H_a)$. They give rise to a field-induced Griffiths phase at temperatures T within $T_c(H_a) < T < T_N$. The fluctuating correlations are detected by SQUID measurements of the lowfrequency out-of-phase susceptibility χ''. A distinct change of curvature of χ'' vs T at the Néel temperature T_N is explained in terms of fluctuating distributions of demagnetizing fields and, hence, transition temperatures. Within Landau theory a phenomenological T_c distribution allows unequivocal modeling of χ'' vs T at $T > T_c(H_a)$. For sub-tricritical fields the analysis may be extended to below $T_c(H_a)$ by taking into account both critical fluctuations at $T_c(H_a)$ and non-critical Griffiths-type fluctuations within $T_c(H_a) < T < T_N$. Best fits reveal discontinuities of $d^2\chi''/dT^2$ in the vicinity of T_N thus confirming the expected high-T boundary of the field-induced Griffiths phase.

1. Introduction

The Griffiths phase conjecture [1] is based on the idea of "local" phase transitions in a diluted system due to the finite probability of arbitrarily large pure and differently diluted clusters. For a ferromagnet with diamagnetic dilution, x, the magnetization is expected to be a non-analytic function of the magnetic field for any temperature, T, within the so-called Griffiths phase $T_c(x) < T < T_c(x = 0)$. On the one hand, if x does not exceed the percolation limit, x_p, static critical behavior characterises the global phase transition at $T_c(x) > 0$. On the other hand, anomalies, if any [2], are expected within the Griffiths phase. However, Griffiths's original rigorous results, which are based on the theorem of Lee and Yang, give no hint how the weak singularities in

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the magnetization could influence the magnetic properties. To the best of our knowledge, there is still no convincing experimental evidence of its very existence up to now. E.g., the wide temperature regime of extreme slowing-down of the dipolar relaxation in the relaxor ferroelectric $PbMg_{1/3}Nb_{2/3}O_3$ (PMN) has tentatively been attributed to a Griffiths phase [3]. However, this interpretation is seriously cast in doubt by invoking random-field mechanisms to be responsible for the diffuse phase transition of PMN [4]. An experimental hint at a dynamical signature of the Griffiths phase in the dilute Heisenberg antiferromagnet $KMN_{0.3}Ni_{0.7}F_3$ was derived from inelastic neutron scattering experiments [5]. They were explained by use of the theoretical prediction [6,7,8] that the time dependent spin-autocorrelation function should decay more slowly than exponentially within the Griffiths phase in contrast to the purely exponential decay expected for the paramagnetic regime. This conjecture is confirmed by recent Monte Carlo studies of diluted Ising ferromagnets [9]. However, clear experimental verification of this subtlety is still lacking.

The uniaxial antiferromagnet $FeCl_2$ shows in the presence of an applied axial magnetic field H_a quasicritical domain-like fluctuations within the temperature regime $T_c(H_a) < T < T_c(H_a = 0) \equiv T_N$. They give rise to anomalous contributions to the magnetic loss function χ'' at low frequencies $0.1 < f < 10Hz$. Analogously as argued for the Griffiths phase in diluted ferromagnets we attribute the temperature regime $T_c(H_a) < T < T_N$ to a field-induced Griffiths phase. [10] Compared to the diamagnetic dilution x, which reduces the ferromagnetic order, the homogeneous external magnetic field weakens the antiferromagnetic order of the metamagnet. The field-induced Griffiths phase has its experimental manifestation in the temperature dependence of χ''. In particular we showed that $d\chi''/dT$ changes kinklike at T_N. We suggest that fluctuating demagnetizing fields locally diminish the applied field such that internal fields, H_i, drive local phase transitions at $T_c(H)$, where $0 \le |H| \le H_a$. Hence, analogously to the Griffiths scenario of the diluted ferromagnet [1] we expect local antiferromagnetic phase transitions throughout the above temperature regime, which is conventionally denoted as paramagnetic. In contrast with the conventional Griffiths phase, which is due to a static distribution of local concentrations x_1 with $0 \le x_1 \le x$, however, a dynamic distribution of local fields $0 \le |H| \le H_a$ is involved. This is accounted for in our interpretation, which considers fluctuating domain-like antiferromagnetic correlations giving rise to the observed contribution to χ'' at low frequencies. The aim of this paper is to show that the assumption of distribution averaged quasicritical fluctuations [10] may explicity be used to model an analytic expression for the temperature dependence of χ'' for external magnetic fields above and below the applied tricritical field $H_t = 0.84$ MA/m.

2. Experimental details and results

A commercial SQUID magnetometer (Quantum Design MPMS 5S) was used to measure the temperature dependence of the in-phase and out-of-phase parallel magnetic susceptibility in the low-frequency range $0.1 < f < 10$ Hz at constant fields $398 < H_a < 1592$ kA/m. A Bridgman grown $FeCl_2$ crystal was prepared in a dry helium atmosphere to obtain a thin (0001) oriented sheet of size 3.4 x 2.3 x 0.28 mm^3. In order

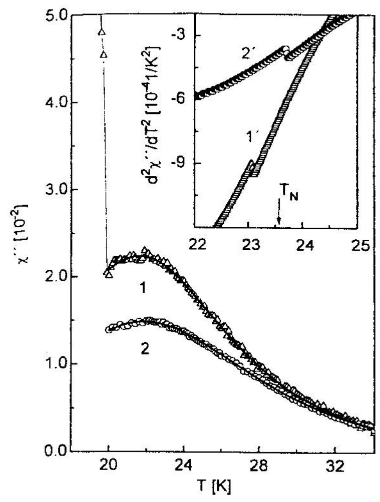

Fig. 1. χ'' vs T of $FeCl_2$ measured at f=5 Hz and H_a=0.96 MA/m (curve 1) and H_a=1.43 MA?m (curve 2). The solid lines are least-squared fits to Eq. 8. Their second derivatives with respect to T are shown in the inset as curve 1′ and 2′, respectively. T_N is marked by an arrow.

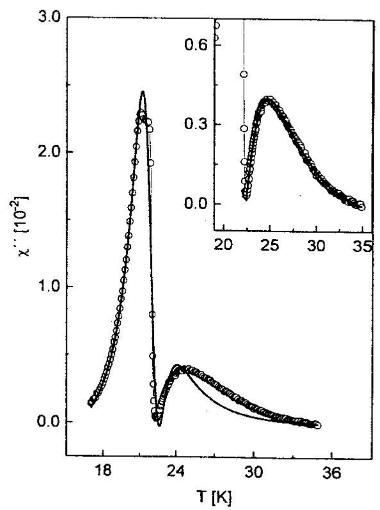

Fig. 2. χ'' vs T of $FeCl_2$ measured at f=5 Hz and H_a=0.6 MA/m (circles) and least-squares fit (solid line) to the data within $17 \le T \le 35$ K. The inset shows χ'' vs T for $22 \le T \le 35$ (circles) with a best-fit (full line) for the data within $22.4K \le T \le 35K$.

to determine the Néel temperature, T_N, we measured the temperature dependence of the magnetization M vs T in a very low field, $H_a = 0.8$ kA/m, and obtained $T_N = 23.7$ K from the peak of dM/dT. Figure 1 shows the out-of-phase component χ'' of the ac susceptibility versus temperature at constant frequency $f = 5$ Hz and field values $H_a = 0.96$ (curve 1) and 1.43 MA/m (curve 2). Figure 2 shows χ'' vs T at $f = 5$ Hz for $H_a = 0.6$ MA/m. As a rule, both positive and negative slopes, $d\chi''/dT$, are observed within the Griffiths range $T_c(H_a) < T < T_N$. This is most pronounced for external fields $H_a < H_t$. As shown in Figure 2 χ'' vs T starts at $T > T_c(H_a)$ with values close to zero and positive slope, passes a maximum and decays gently as observed for $H_a > H_t$ (Figure 1). In the latter case χ'' vs T starts with a finite value at the upper mixed phase boundary, $T > T_c(H_a)$. It will be shown that the entire variety of different curves χ'' vs T can be modeled by the same analytic expression.

3. Theory and comparison with experimental results

The metamagnetic transition of $FeCl_2$ is characterised by a mixed phase region where antiferromagnetic and quasi-ferromagnetic ("saturated paramagnetic") phases coexist (Figure 3). This static domain state is caused by inhomogeneous demagnetizing fields.

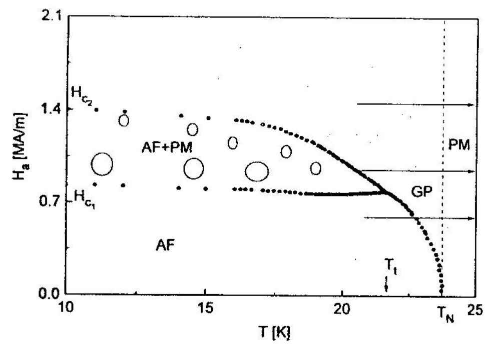

Fig. 3. H_a vs T phase diagram of $FeCl_2$ (solid circles [18]) with elliptical antiferromagnetic domain-symbols (full lines) in the mixed phase (AF+PM, limited by the transition fields H_{c1} and H_{c2}) and symbols for domainlike fluctuations in the field induced Griffiths phase (GP, dotted lines). Tricritical Néel temperatures, T_t and T_N, are indicated by an arrow and a vertical dashed line, respectively. Horizontal arrows denote the isomagnets, along which the ξ'' vs T data in Figures 1 and 2 are taken.

In analogy with the behavior of Ising ferromagnets, where domainlike order parameter fluctuations above the phase transition are well known [11,12,13], we assume that the static inhomogeneity of the mixed phase becomes dynamic above the phase transition. A signature of this behavior is already found in the field dependence of the static magnetization. It shows rounding close to the paramagnetic saturation which has to be interpreted in terms of fluctuating demagnetizing fields. They locally allow for antiferromagnetic order and, hence, reduce the total magnetization [10]. A similar situation applies to a horizontal passage through the H_a-T phase diagram (Figure 3) at $H_a > H_c$ or H_{c2}, respectively. Here we expect a fluctuating distribution of the local internal fields H_i at any fixed value of $T \geq T_c(H_a)$. The Fourier transform of the demagnetizing field can be expressed within the magnetostatic approximation as $\underline{h}(\underline{q}) = -(\underline{q}\underline{m}(\underline{q}))\underline{q}/q^2$ where $\underline{m}(\underline{q})$ is the Fourier transform of the magnetization. Owing to the unambiguous relationship $T_c = T_c(H_a)$ at the static phase boundary (Figure 3) simultaneously a distribution of different phase transition temperatures within $T_c(H_a) \leq T_c \leq T_N$ is encountered. Within the concept of local transition temperatures [14] we introduce the probability $P(T,T_c)\mathrm{d}T_c$ to find a critical temperature within the interval $[T_c, T_c + \mathrm{d}T_c]$ at a given temperature T. The quasicritical fluctuations of the antiferromagnetic order parameter η, which are involved for any T_c with finite probability can be described within the framework of the Landau theory of fluctuations near second-order phase transitions [15,16]. The autocorrelation function $S = \langle|\underline{\eta}(\underline{q})^2|\rangle$ is given by

$$S = (k_B T)/[V(\phi_{\eta\eta} + Dq^2)] \tag{1}$$

where V is sample volume, $\phi_{\eta\eta}$ the second derivative of the Landau expansion of the Gibbs free energy density ϕ at the equilibrium point $\underline{\eta} = \underline{\eta}_e$. It is given by

Table 1 Best-fit parameters to Eq. (8) and quality parameter Q (see text) for the χ'' vs T data presented in Figures 1 (H_a = 0.96 and 1.43 MA/m, fitting range $19.9 \leq T \leq 35$K) and 2 (H_a = 0.6 MA/m, fitting ranges $17 \leq T \leq 35$K and $22.4 \leq T \leq 35$K, respectively).

H_a fitting range	0.96 MA/m $19.9 \leq T \leq 35$K	1.43 MA/m $19.9 \leq T \leq 35$K	0.6 MA/m $17 \leq T \leq 35$K	0.6MA/m $22.4 \leq T \leq 35$K
P	3.4067	3.5150	0.1546	0.4022
b	118.14	172.91	29.166	71.111
T_c	17.303	15.008	22.005	20.323
T_N	23.086	23.717	25.851	27.358
A_0/Dq^2	0.9248	1.0238	1.1698	1.3524
Q	$6.598\ 10^{-8}$	$1.057\ 10^{-8}$	$1.073\ 10^{-6}$	$2.488\ 10^{-9}$

$$\phi_{\eta\eta} = mA_0(T/T_c' - 1) \tag{2}$$

with $m = -2$ and $+1$ for $T < T_c'$ and $T > T_c'$, respectively. $1/2\ A_0(T/T_c' - 1)$ and $D/2$ are the expansion coefficients of the quadratic and the gradient term of ϕ, respectively. From the fluctuation-dissipation theorem it is known that χ'' is related to the power spectrum of the magnetization fluctuations [15]. These induce fluctuations of the antiferromagnetic order parameter by coupling of $\underline{m}$ to η at site $\underline{r}$ via the relation [17] $2\underline{m}(\underline{r}) = (\underline{c}\nabla)\eta(\underline{r})$, where $\underline{c}$ is the basis vector along [0001]. Hence, χ'' is also related to the autocorrelation function of the antiferromagnetic order parameter $S = \langle|\underline{\eta}(\underline{q})^2|\rangle$. One expects the proportionality $S \propto \chi''$, although χ'' refers to the ferromagnetic response. Averaging the order parameter fluctuations by the T_c-distribution function $P(T, T_c)$ yields

$$\chi'' \propto \frac{1}{T}\int_{T_c(H)}^{T_N} P(T,T_c')S(T,T_c')\mathrm{d}T_c'. \tag{3}$$

The factor $1/T$ is the classical residuum of the Bose-Einstein factor which enters the fluctuation-dissipation theorem.

In order to obtain an analytic expression for χ'' vs T we propose the following Lorentzian ansatz for the distribution function:

$$P(T,T_c') = \frac{1}{\pi}\frac{\varepsilon}{\varepsilon^2 + (T_c - T_c')^2} \tag{4}$$

where $T_c = T_c(H_a)$, $T_c \leq T_c' \leq T_N$, $\varepsilon = b/T$ and b=constant. Eq (4) ensures the following physically motivated properties:

(i) $P(T,T_c')$ maximises at $T_c' = T_c$, where the global phase transition takes place with contributions to χ'' by one order of magnitude large than within the Griffiths regime $T_c' > T_c$;

(ii) $P(T,T_c')$ decreases as $T_c' \to T_N$, where T_N denotes the largest possible critical temperature;

(iii) $P(T,T_c')$ becomes narrower with increasing T, since $\varepsilon = b/T$ is proportional to its halfwidth,; at $T \gg T_c$ only contributions referring to $T_c' \sim T_c$ will be relevant.

The normalization factor of the probability density P,

$$N = \frac{1}{\pi}\arctan\left(\frac{T_N - T_c}{\varepsilon}\right), \tag{5}$$

as obtained by integrating $P(T,T_{c'})$ over the Griffiths regime, $T_c \le T_{c'} \le T_N$, is weakly T dependent. However, inspection shows that its temperature dependence is negligible when compared with that of P and S. Hence, for simplicity we treat N as a constant which is absorbed by the constant b and the unknown proportionality constant between χ'' and the averaged order parameter fluctuations. Inserting equations (1) and (4) into (3) yields

$$\chi'' \propto \frac{\varepsilon}{\pi V}\int_{T_c}^{T_N}\frac{T_{c'}}{(\varepsilon^2 + (T_c - T_{c'})^2)(mA_0(T - T_{c'}) + Dq^2T_{c'})}\mathrm{d}T_{c'} \tag{6}$$

The main contributions to the integral arise in the quasicritical temperature regime, $T_{c'} \sim T$. Hence, after substituting $T_{c'} = T - z$ into (6) and integrating with respect to z it is meaningful to make a series expansion of the integrand up to first order in z. This yields

$$\chi'' \propto \frac{\varepsilon}{\pi V}\times$$

$$\times \int_{T-T_N}^{T-T_c}\left[\frac{1}{(\varepsilon^2 + (T_c - T)^2)Dq^2} + z\frac{2Dq^2T^2(T - T_c) - mA_0T(\varepsilon^2 + (T_c - T)^2)}{((\varepsilon^2 + (T_c - T)^2)Dq^2T)^2}\right]\mathrm{d}z \tag{7}$$

Note that m is also a function of temperature which can be taken into account by splitting the integral into two parts with $T > T_{c'}$ and $T < T_{c'}$, respectively. After integration we obtain

$$\chi'' \propto \frac{\varepsilon}{\pi V Dq^2(\varepsilon^2 + (T_c - T)^2)}\times$$

$$\times\begin{cases} T_N - T_c + \frac{T-T_c}{(\varepsilon^2+(T_c-T)^2)}((T-T_c)^2 - (T-T_N)^2) \\ -\frac{A_0}{Dq^2}\left(\frac{1}{2}(T-T_c)^2 + (T-T_N)^2\right) & \text{if } T \le T_N \\ T_N - T_c + \left(\frac{T-T_c}{(\varepsilon^2+(T_c-T)^2)} - \frac{A_0}{2Dq^2T}\right)((T-T_c)^2 - (T-T_N)^2) & \text{if } T > T_N \end{cases} \tag{8}$$

It can easily be shown that $\mathrm{d}^2\chi''/\mathrm{d}T^2$ changes steplike at $T = T_N$. This was already shown by the general consideration in [10] and is, hence, not due to the special choice of the distribution function.

The full lines in Figure 1 show the least-squares fits of Eq. (8) to the χ'' vs T data within and above the field-induced Griffiths phase for external fields above the tricritical field H_t. The four best-fit parameters b, T_c, T_N and A_0/Dq^2, which enter Eq. (8), are summarised in Table 1. The proportionality constant P contains the factor $(\pi V Dq^2)^{-1}$. The quality parameter Q is defined as

$$Q = \frac{\sum_{i=1}^{N}(f(x_i) - Y_i)^2}{N - n} \tag{9}$$

where N is number of data points, n the number of fitting parameters, (x_i, Y_i) are the data points and $f(x_i)$ are the corresponding values of the fitting function. The data which are involved in the fitting procedures are those between the beginning and the end of the full lines drawn in the figures.

The two parameters T_c and T_N correspond to the onset and the high temperature boundary of the field-induced Griffiths phase, respectively. In accordance with the general arguments outlined in [10] the inset in Figure 1 shows steplike changes of the curvature of the least-squares fits to curves 1 and 2 (curves 1' and 2'). These steplike changes coincide with the experimental value of T_N within errors of 2.6 and 0.1%, respectively. Curve 1 shows that χ'' steeply increases when lowering T to below $T_c(H_a)$. This is due to non-critical loss mechanisms owing to the static disorder of the mixed phase which are not included in our model. The fitting parameter T_c indicates the onset of the regime of quasicritical fluctuations. It coincides within an error of 12.2% with the experimental value T_c = 19.7K. Curve 2 does not include additional non-critical losses, because the applied field H_a = 1.43 MA/m slightly exceeds the phase boundary for all temperatures (cf Figure 3). Hence, there is no critical temperature responsible for a phase transition. This is qualitatively taken into account by the low value of $T_c(H_a)$ = 15.008K. Moreover, the width of the T_c-distribution function, which is proportional to the parameter b, becomes larger with increasing field. This is compatible with an increasing number of local phase transition temperatures which are involved, when the critical temperature of the global phase transition is reduced. Note that this argument must break down for applied fields much higher than H_{c2}. Owing to increasing homogeneity of the magnetization additional contributions to the demagnetizing field arising from $\mathrm{div}M \neq 0$ are no longer allowed. Hence, even locally, the total compensation of the applied field is no longer possible.

We now examine the behavior of χ'' vs T for the sub-tricritical field H_a = 0.6 MA/m. In a first attempt it seems to be reasonable to fit only the data above the deep minimum of χ'' vs T at $T >$ 22.4K to Eq. (8). The inset in Figure 2 shows the result. Table 1 summarises the resulting parameters. However, the good quality of the fit as evidenced by an extremely small Q-value is cast in doubt by the large deviation between the real and the fitted T_N value: ΔT_N = 3.6K. Furthermore, in comparison to both other fits, the ratio of the parameter A_0/Dq^2 is about 30% larger. This can be interpreted as an increase of the correlation length involved. In fact, the global second-order phase transition is accompanied by critical fluctuations with diverging correlation length. The maximum of χ'' vs T at $T_c = 21.2$K marks this transition. In contrast to the maximum in the the χ'' vs T curves which arise from the disorder of the mixed phase, the critical fluctuations are included in the model. Hence, we can extend the fitting procedure to the entire temperature range. The result is shown in Figure 2. The best fitting curve seems qualitatively to describe all relevant features of the data. On the one hand, the global phase transition is correctly modeled. On the other hand, the Griffiths regime, which begins already at the large maximum of χ'' vs T, is qualitatively described. In contrast with the high field behavior of χ'', the fluctuations have a prominent minimum within the Griffiths regime. It is due to the vanishing fluctuations of the global phase transition above $T_c(H_a)$. This minimum is followed by a superposition of quasicritical fluctuations which cause χ'' to increase again. In addition, it is satisfying to observe that T_N approaches its expected value.

4. Conclusion and outlook

It is shown that the uniaxial antiferromagnet $FeCl_2$ in an axial field gives rise to a Griffiths phase-like phenomenon. The temperature dependence of its out-of phase susceptibility can be described within the framework of order parameter fluctuations, which are averaged by a physically motivated T_c - distribution function. The different characteristics of χ'' vs T for fields above and below the tricritical field can be modeled.

The poor quality of the least-squares fit of the data obtained for a sub-tricritical field shows that the calculation requires further improvements. Within our model two straightforward refinements are obvious. On the one hand, higher members of the series expansion, which was introduced to solve the averaging integral (6), have to be taken into account. On the other hand, integration in q-space has to be considered. The model deals with a single wave vector q. This is reasonable in the case where only quasicritical fluctuations are involved. The main contributions to the fluctuations and, hence, to χ'' arise from the smallest q value, which corresponds to the restricted antiferromagnetic correlation length owing to the local character of the fluctuating demagnetizing fields. However, the global phase transition is affected by the average internal field. Hence, the smallest q value of the critical fluctuations is limited by the finite sample dimensions. This explains the more rapid thermal decay of the critical fluctuations compared with that of the quasicritical fluctuations which are responsible for the Griffiths regime. Very probably the model would be improved by properly taking into account two different scales of correlation lengths.

The analysis of the frequency dependence of χ'' is another remaining challenge. It would allow to include non-equilibrium effects i.e. dynamical rounding. There is presently no hope to find a correct analytic expresion of the T_c distribution function without further assumptions. Monte Carlo simulations might be appropriate to describe the complicated dynamic demagnetizing processes.

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