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# Magnetic neutron-scattering investigation of the field-induced Griffiths phase in $\text{FeCl}_2$

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Quasielastic neutron scattering has been utilized to investigate the Griffiths phase induced by an axial magnetic field in  $\text{FeCl}_2$  at temperatures  $T_c(H) \leq T \leq T_N$ , where  $T_c(H)$  is the transition temperature in an external magnetic field  $H$ . We present the temperature dependence of the integrated neutron-scattering intensity at fixed scattering vectors for various magnetic fields. On cooling below  $T_N$  the antiferromagnetic short-range fluctuations decrease due to an increase of the volume fraction which is already antiferromagnetically ordered. A weighted average of the local contributions to the antiferromagnetic susceptibility quantitatively describes the scattering cross section. [S0163-1829(96)08534-7]

## I. INTRODUCTION

Recently the concept of a field-induced Griffiths phase has been used to explain the temperature dependence of the complex low frequency magnetic susceptibility above the phase boundary  $T_c(H)$  of various Ising-type antiferromagnets,  $\text{FeCl}_2$ ,<sup>1,2</sup>  $\text{FeBr}_2$ ,<sup>3</sup> and  $\text{Fe}_{1-x}\text{Zn}_x\text{F}_2$ ,<sup>4</sup> in axial magnetic fields  $H$ . The feature that these systems have in common is that fluctuating demagnetizing fields locally diminish the applied field in a way such that the internal fields,  $H_i$ , drive local phase transitions within the temperature range  $T_c(H) \leq T \leq T_N$ . The quasicritical behavior within this temperature interval characterizes the field-induced Griffiths phase. The weighted average of the local quasicritical contributions to the susceptibility allows a quantitative description of the uniform magnetic susceptibility,  $\tilde{\chi}_u$  vs  $T$ , for all three systems.<sup>2-4</sup> However, since  $\tilde{\chi}_u$  measures the ferromagnetic response, the susceptibility data contain no direct information about the local antiferromagnetic order and order parameter fluctuations. The indirect influence of the quasicritical antiferromagnetic fluctuations on the ferromagnetic response function is merely caused by the coupling of the primary to the secondary order parameter. In contrast with susceptibility measurements, the magnetic neutron scattering cross section is directly related to antiferromagnetic order parameter fluctuations. Hence, magnetic neutron scattering experiments can give direct proof of the magnetometrically motivated model.

## II. EXPERIMENTAL DETAILS

The experiments were carried out on the DN3-neutron-diffractometer at the reactor Siloe of C.E.N.G./Grenoble,

France. A graphite monochromator was used to select a beam of thermal neutrons with wavelength  $\lambda = 0.153$  nm.  $\lambda/2$  contamination of the incident beam was eliminated by a pyrolytic graphite filter. A parallelepiped-shaped Bridgman grown sample of  $\text{FeCl}_2$  with edge lengths  $a = 11$ ,  $b = 15$ , and  $c = 6$  mm was mounted in a cryomagnet. Axial fields up to 4 MA/m are applied along the pseudo-hexagonal  $c$  axis allowing full sample rotations around the field direction. The temperature of the sample was varied between 10 and 60 K and stabilized within an accuracy of about 0.1 K. The  $\Delta Q_k$  resolution of the diffractometer was measured around the  $Q_k = 0$  position of the nuclear (1,0,2)-Bragg peak. Using the reciprocal lattice units (r.l.u.) of the double hexagonal cell<sup>5</sup> it is given by  $\Delta Q_k = 0.009$  (r.l.u.).

## III. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 1 shows the field dependence of the integrated neutron-scattering intensity for the magnetic (1,0,-1) Bragg peak of  $\text{FeCl}_2$  at  $T = 10$  K. Within the antiferromagnetic phase, the increase of the applied field does not affect the Bragg intensity. However, on entering the mixed phase region at  $H_{c1} = 0.88$  MA/m the Bragg intensity decreases linearly with increasing magnetic field. The decrease of the scattering intensity measures the decrease of the antiferromagnetic volume fraction with respect to the coexisting paramagnetic phase. This coexistence occurs below the tricritical point and reflects the well-known spread of the first order spin-flip transition line due to demagnetizing field effects.<sup>6</sup> Above the upper mixed phase boundary at  $H_{c2} = 1.11$  MA/m the antiferromagnetic Bragg intensity is nearly independent of the field. However, the small scattering intensity above  $H_{c2}$  gives a first hint of remaining antiferromagnetic contributions of the Griffiths type.

The magnetic scattering intensity at the (1,0,-1)-Bragg

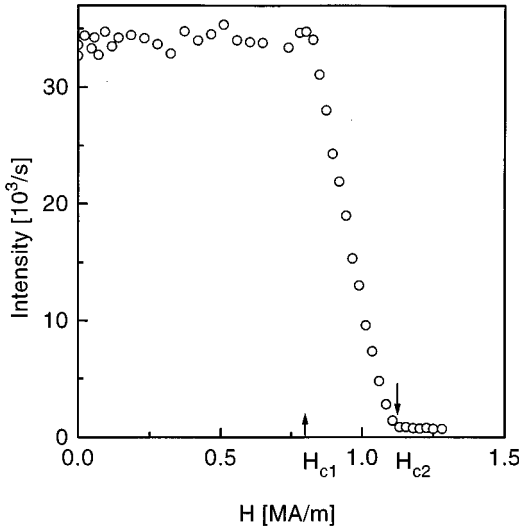


FIG. 1. Field dependence of the neutron-scattering intensity of the  $(1,0,-1)$  Bragg peak of  $\text{FeCl}_2$  at  $T=10$  K. The arrows indicate the boundaries of the mixed phase at  $H_{c1}=0.88$  MA/m and  $H_{c2}=1.11$  MA/m, respectively.

peak is proportional to the square of the antiferromagnetic order parameter. It would now be interesting to inspect the  $q$  dependence of the scattering intensity in the vicinity of  $(1,0,-1)$  in order to study, e.g., the antiferromagnetic correlation length and its dependence on temperature and magnetic field. However, such a program requires complete absence of Bragg peak intensity which was not achieved in our present experiments owing to incomplete filtering of higher harmonics during the second half of our beam time. Hence we restricted ourselves to the consideration of the neutron-scattering intensity at the fixed scattering vector  $\kappa=(1,-0.06,-1)$  r.l.u. at  $H=0$  (a), 0.95 (b), 1.27 (c), 1.35 (d), 1.43 (e), and 1.91 MA/m (f), respectively (Fig. 2). Note that the curves (c)–(f) are shifted with respect to each other for sake of clarity (see caption). The scattering intensity for  $\kappa=(1,-0.06,-1)$  r.l.u. measures the temperature dependence of the superposition of the parallel and the perpendicular order parameter susceptibility,  $\chi_{\parallel}$  and  $\chi_{\perp}$ , where the notation  $\parallel$  and  $\perp$  refer to the  $c$  axis, respectively.<sup>7</sup> In zero field the second-order phase transition at the Néel temperature,  $T_N$ , is associated with the divergence of the staggered parallel susceptibility

$$\chi_{\parallel}(q=0)=\chi^{\pm}|T-T_N|^{-\gamma}. \quad (1)$$

In the case of three-dimensional (3D) Ising model systems  $\gamma=1.25$  and  $\chi^+/\chi^-=4.8$  are the critical exponent and amplitude ratio, respectively.<sup>8</sup> Due to the strong uniaxial anisotropy as, e.g., realized in  $\text{FeCl}_2$ ,  $\chi_{\perp}$  does not contribute to the scattering intensity within a good approximation.<sup>7</sup> The scattering cross section is then proportional to the wave vector dependent order parameter fluctuations, which are related to  $\chi_{\parallel}(q)$  and approximately given by<sup>9</sup>

$$S(T_N, T, q) = \frac{k_B T}{V} \frac{1}{1/\chi_{\parallel}(q=0) + Dq^2}, \quad (2)$$

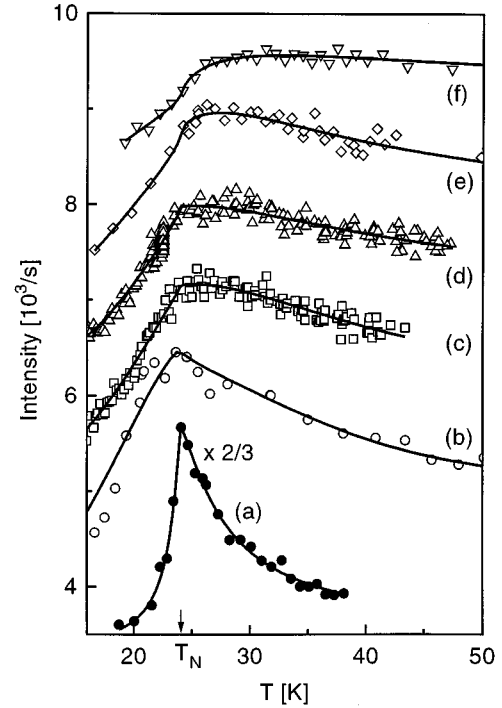


FIG. 2. Temperature dependence of the neutron-scattering intensity at fixed scattering vector  $\kappa=(1,-0.06,-1)$  r.l.u. (symbols) for  $H=0$  (a), 0.95 (b), 1.27 (c), 1.35 (d), 1.43 (e), and 1.91 MA/m (f) with corresponding best fits (solid lines) to Eq. (2) (a) and Eq. (6) (b)–(f), respectively. Curve (a) is scaled by a factor of  $2/3$ . The curves (c)–(f) are shifted by  $n \times 10^3$  counts/s ( $n=1, 2, 3,$  and  $4$ , respectively) with increasing magnetic field. The Néel temperature  $T_N$  is indicated by an arrow.

where  $q \neq 0$  is the value of the difference between the scattering vector and the position of the corresponding antiferromagnetic Bragg peak in the reciprocal space.  $D$  is the coefficient of the second order  $q$  expansion of the magnetic exchange and  $V$  is the scattering volume. The solid line (a) in Fig. 2 shows the best fit of the neutron scattering data for zero magnetic field (solid circles) to Eq. (2). It includes a temperature independent background and a proportionality factor, which mediates between  $S$  and the scattering intensity. By use of the above values of  $\gamma$  and  $\chi^+/\chi^-$  we obtain  $T_N=24.04$  K from the best fit, which finally contains four parameters.

Curves (b)–(f) in Fig. 2 indicate a remarkable change in the temperature dependence of the scattering intensity for  $H \neq 0$ . On cooling below  $T_N$  all curves show a steep decrease of the scattering intensity. This is due to the gradual vanishing of antiferromagnetic short-range correlations. It reflects the sequence of local phase transitions, which take place in the Griffiths phase on cooling below  $T_N$ . The reduction of the scattering intensity due to local quasicriticality is expected to shift the maximum scattering intensity towards  $q=0$ . Hence the steep decrease of the scattering intensity indicates the onset of the field-induced Griffiths phase at its high temperature boundary,  $T=T_N$ .

A quantitative description of the scattering data is performed by the calculation of a weighted average of the local antiferromagnetic contributions to the susceptibility as determined by the corresponding fluctuations of the order param-

eter. Analogously to the description of the magnetic susceptibility we use the well-tried<sup>2-4</sup> phenomenological Lorentzian distribution of local critical temperatures  $T'_c$

$$P(T'_c, T) = \varepsilon / \{ [\varepsilon^2 + (T_c - T'_c)^2] \arctan[(T_N - T_c) / \varepsilon] \}, \quad (3)$$

with  $T_c \leq T'_c \leq T_N$ ,  $\varepsilon = b/T$ , and  $b(H) > 0$ . The measured scattering intensity is then given by a weighted correlation function,

$$I(T, q) \propto \int_{T_c}^{T_N} dT'_c P(T'_c, T) S(T'_c, T, q), \quad (4)$$

where the local correlation function  $S(T'_c, T, q)$  is given by Eq. (2) inserting a modified staggered susceptibility,

$$\chi_{\parallel}(q=0) = \chi^{\pm} |T - T'_c|^{-\gamma}. \quad (5)$$

Since the local quasicritical scattering maximizes at  $T \approx T'_c$  we approximate the slowly varying distribution function by  $P(T'_c = T, T)$ . Further we linearize  $S(T'_c, T, q)$  with respect to the quantity  $1/\chi_{\parallel}(q=0)$ , which is assumed to be small compared with  $Dq^2$  for  $T'_c \approx T$ . These approximations lead to the expression

$$I(T, q) = \frac{K}{\arctan\left(\frac{T_N - T_c}{\varepsilon}\right) [\varepsilon^2 + (T - T_c)^2]} \left( \begin{array}{ll} \chi^{-} (\gamma + 1) Dq^2 (T_N - T_c) - (T - T_c)^{\gamma+1} & \text{if } T_c < T \leq T_N \\ -\frac{\chi^{-}}{\chi^{+}} (T_N - T)^{\gamma+1} & \text{if } T > T_N \end{array} \right). \quad (6)$$

The solid lines (c)–(f) in Fig. 2 show the results of the best fits of Eq. (6) to the scattering data for  $1.27 \leq H \leq 1.91$  MA/m.  $K$ ,  $\gamma$ ,  $\chi^{-}/\chi^{+}$ ,  $b$ ,  $\chi^{-}(\gamma+1)Dq^2$ , and a constant background  $I_0$  are employed as fitting parameters, while  $T_N = 24.04$  K is known from the zero field scattering data [curve (a)].  $T_c(H > H_{c2}) = 0$  K is fixed in accordance with the phase diagram of  $\text{FeCl}_2$ .<sup>1</sup>  $I_0$  accounts for the incoherent background scattering and the neglected regular contributions. Our best-fit values are weakly varying between  $I_0 = 4930$  and  $5240$  counts/s. For  $H = 0.95$  MA/m  $< H_{c2}$  [curve (b)] it is necessary to introduce  $T_c$  as an additional fitting parameter. We find  $T_c = 11.3$  K, which is a reasonable result when considering the steep decrease of  $T_c(H)$  with increasing magnetic field.

Despite the simplicity of our ansatz, Eq. (4), and the approximation leading to Eq. (6), the results of our fitting procedure look quite satisfactorily. However, a critical comment on the values obtained for the exponent  $\gamma$  and the amplitude ratio  $\chi^{-}/\chi^{+}$  is necessary. From the best fits we obtain unexpectedly small and even negative values,  $\gamma = 0.16, -0.05, -0.14, -0.17$ , and  $-0.22$  for  $H = 0.95, 1.27, 1.35, 1.43$ , and  $1.91$  MA/m, respectively. We believe this to be due to the fact that our ansatz accounts for neither the crossover from conventional to tricritical behavior within the interval  $T_t = 21.7$  K  $\leq T \leq T_N$  nor the fact that first-order transitions are involved for  $T < T_t$ . The strong decrease of the effective  $\gamma$  values with increasing magnetic field is in accordance with the increasing contribution of local first-order transitions to the field-induced Griffiths phase. Unexpectedly large amplitude ratios emerge. They vary between 0.8 and 6 without any systematic dependence on the magnetic field. However, large values of  $\chi^{-}/\chi^{+}$  are accompanied by large error bars. Hence

the fitting procedure seems to favor the smallest value,  $\chi^{-}/\chi^{+} = 0.8$ , which comes close to the amplitude ratio of the 3D Ising model.

#### IV. CONCLUSION

The coupling of the magnetic moments of neutrons to the antiferromagnetic order parameter allows experiments which confirm the conjectured<sup>1,2</sup> field-induced Griffiths phase in  $\text{FeCl}_2$ . The high-temperature boundary  $T = T_N$  of the field-induced Griffiths phase is clearly visible by a steplike change of slope and a subsequent steep decrease of the integrated scattering intensity with decreasing temperature. From the experimental point of view this is much more convincing than the uniform magnetic susceptibility data, which merely exhibit a kink of the derivative,  $d\chi''/dT$ , at  $T = T_N$ .<sup>1-3</sup> Our now-classical phenomenological theory,<sup>1-4</sup> using a weighted average of local  $q$ -dependent susceptibility contributions, is particularly appropriate to describe the neutron-scattering data. In particular, in a neutron-scattering experiment the  $q$  value is a well-defined quantity which is fixed by the scattering geometry. This is less clear when discussing uniform susceptibility data<sup>1-4</sup> by introducing *ad hoc*  $q$  values, which formally suppress the divergence of the local susceptibility. Hence our concept of local quasicriticality becomes more convincing in the case of neutron-scattering experiments.

#### ACKNOWLEDGMENTS

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