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Crossover from transient spin structures to the field-induced Griffiths phase of $FeBr_2$

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Abstract

In the presence of an applied axial magnetic field H_a the uniaxial antiferromagnets $FeCl_2$ and $FeBr_2$ show fluctuating domain-like antiferromagnetic correlations above the phase boundary $T_c(H_a)$. They are detected by SQUID measurements of the low frequency out-of-phase susceptibility χ'' and indicate a field-induced Griffiths phase at temperatures $T_c(H_a) < T < T_N$. In contrast to $FeCl_2$, important additional frustration-induced intraplanar non-critical contributions to χ'' vs. T are found in $FeBr_2$. For external fields above the $T_c(H_a)$ line, $H_a > 2.6$ MA/m, they are shown to superimpose linearly on the Griffiths contributions. These dominate at $H_a = 2.67$ MA/m and are unequivocally modeled within the Landau theory of fluctuations near phase transitions by introducing a Lorentzian T_c distribution.

The Griffiths phase conjecture was theoretically introduced for diluted Ising-ferromagnets [1]. It 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. However, despite a possible dynamical signature of the Griffiths phase in inelastic neutron scattering data of $KMn_{0.3}Ni_{0.7}F_3$ [2] and recent Monte Carlo simulations [3], its clear experimental verification is still lacking. A more favorable situation is encountered in an analogous experimental realisation of a Griffiths phase-like phenomenon. It was recently detected on the uniaxial antiferromagnet $FeCl_2$ in an applied axial magnetic field [4]. Domain-like antiferromagnetic correlations are created by fluctuating demagnetizing fields and, hence, transition temperatures due to the unambiguous relationship $T_c = T_c(H_a)$. Within the temperature regime $T_c(H_a) < T < T_c(H_a = 0) \equiv T_N$ the quasicritical order parameter fluctuations give rise to anomalous contributions to the magnetic loss function χ'' at low frequencies $0.1 < f < 10$ Hz. Regions with local antiferromagnetic correlations are analogous to the non-diluted clusters, which are responsible for the Griffiths phase in diluted ferromagnets. As shown in Ref. [4], these antiferromagnetic fluctuations are suitably described within the framework of the Landau theory of fluctuations near second-order phase transitions [5]. In addition, the concept of local transition temperatures, which is accounted for by a Lorentzian T_c distribution function, allows one to model the temperature dependence of the out-of phase susceptibility χ'' within and above the Griffiths regime $T_c(H_a) < T < T_N$. An approximate analytic expression is given by [6]

$$\chi'' \propto \frac{\epsilon}{\pi V D q^2 (\epsilon^2 + t_c^2)} \times \begin{cases} T_N - T_c + \dfrac{t_c (t_c^2 - t_N^2)}{\epsilon^2 + t_c^2} \\ \quad - \dfrac{A_0}{D q^2 T} \left(\tfrac{1}{2} t_c^2 + t_N^2 \right) & \text{if } T \le T_N, \\ T_N - T_c + \left(\dfrac{t_c}{\epsilon^2 + t_c^2} - \dfrac{A_0}{2 D q^2 T} \right) \\ \quad \times \left(t_c^2 - t_N^2 \right) & \text{if } T > T_N, \end{cases}$$

with $t_c = T - T_c$, $t_N = T - T_N$, A_0 and D the Landau expansion coefficients of the quadratic and gradient term of the Gibbs free energy density, $\epsilon = b/T$ the temperature-dependent width of the T_c distribution, V the sample volume and q the wave-vector of the order parameter fluctuations. While the field-induced Griffiths phase is driven by inhomogeneous demagnetizing fields, $FeBr_2$ shows an important additional intrinsic loss mechanism. As outlined in Ref. [7], non-critical fluctuations are attributed to intraplanar frustration. It gives rise to transient non-uniform spin structures, which carry excess magnetisation. Their location in the H_a–T phase diagram is shown in Fig. 1. Below the $T_c(H_a)$ line they appear only on the

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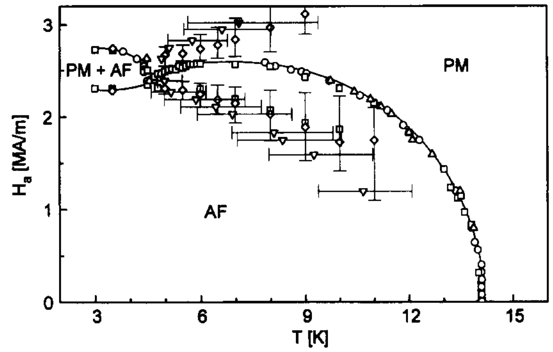

Fig. 1. H_a–T phase diagram (data points with eye-guiding lines) and regimes of strong non-critical fluctuations (data points with bars indicating full widths at 0.6 maximum) obtained from M vs. T (○) and H_a (□), χ' vs. T (Δ), χ'' vs. T(▽) and H_a (◇) [7].

sublattice with magnetisation antiparallel to H_a, whereas above $T_c(H_a)$ they are assumed to spread over all Fe^{2+} layers by symmetry.

Fig. 2a (circles) shows χ'' vs. T measured by SQUID magnetometry at $H_a = 3.02$ MA/m and constant frequency $f = 5$ Hz obtained for a [0001] oriented Bridgman-grown single crystal with ~ 0.2 mm thickness. The non-critical fluctuations above the phase transition are responsible for the maximum of χ'' vs. T at $T = 7.1$ K (solid symbol in Fig. 1). With increasing temperature, χ'' decreases due to the thermal decay of spin clusters. However, contributions from the field-induced Griffiths phase cause a delay of the thermal decay in the vicinity of the 'Griffiths-temperature' $T_N = 14.1$ K. With decreasing external field, the frustration-induced fluctuations shift to lower temperatures and thus gradually separate from the Griffiths-like contributions appearing at higher T. At $H_a = 2.67$ MA/m this is indicated by a clear minimum close to zero at $T = 8.5$ K (Fig. 2a, squares).

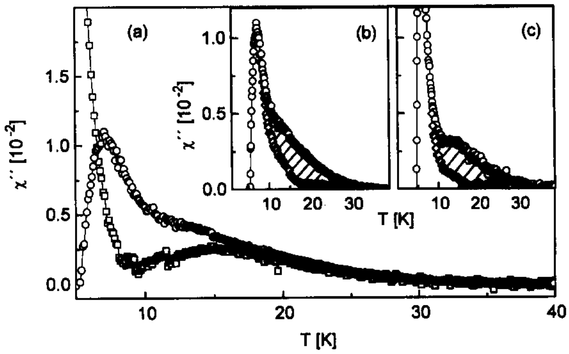

Fig. 2. (a) χ'' vs. T measured at $f = 5$ Hz and $H_a = 2.67$ (□) and $H_a = 3.02$ MA/m (○). The solid line is a least-squares fit to the theory (see text). The insets show χ'' vs. T for $H_a = 3.02$ (b) and $H_a = 2.86$ MA/m (c) before (○) and after (◇) subtracting the Griffiths-type contribution, see text. The hatched areas indicate the excess in χ'' due to the Griffiths contributions.

The solid line in Fig. 2a shows the least-squares fit of the above function to these data. T_N, T_c, A_0/Dq^2, b and an additional proportionality constant are involved as fit parameters. The result from the fitting procedure, $T_N = 13.97$ K, comes close to the experimental value, $T_N = 14.1$ K, which was obtained by the temperature dependence of the low-field magnetization. This demonstrates that the concept of the field-induced Griffiths phase can be extended from the prototype $FeCl_2$ [4] to the frustrated Ising system $FeBr_2$. Under the assumption that the field-dependent change of χ'' vs. T is mainly caused by the frustration-induced fluctuations, we are able to separate these and the virtually constant Griffiths contributions from each other. As a result, Figs. 2b and c show χ'' vs. T for $H_a = 3.02$ and 2.86 MA/m, respectively, before and after subtracting the Griffiths-type contribution. This is taken from the fit of the data at $H_a = 2.67$ MA/m (Fig. 2a) and represented by the hatched areas in Figs. 2b and c. As expected, the frustration-induced fluctuations increase with increasing field from $H_a = 2.86$ to 3.02 MA/m. This is consistent with the phase diagram as discussed in Ref. [7]. It shows that in contrast to the Griffiths contributions, the frustration-induced fluctuations develop their main intensity far from the phase transition line. This is observed above and also below [7] the $T_c(H_a)$ line. This remarkable property and the interplay of the frustration-induced and three-dimensional critical fluctuations are still under investigation.

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