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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 $\chi''$ 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 $\chi''$ 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 $T_c(H_a) = T_c(H_a)$. Within the temperature regime $T_c(H_a) < T < T_c(H_a = 0) = T_N$ the quasicritical order parameter fluctuations give rise to anomalous contributions to the magnetic loss function $\chi''$ 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 $\chi''$ within and above the Griffiths regime $T_c(H_a) < T < T_N$. An approximate analytic expression is given by [6]

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

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...
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 $\chi''$ 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 $\sim 0.2$ mm thickness. The non-critical fluctuations above the phase transition are responsible for the maximum of $\chi''$ vs. $T$ at $T = 7.1$ K (solid symbol in Fig. 1). With increasing temperature, $\chi''$ 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).

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 $\chi''$ 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 $\chi''$ 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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References