August 2000

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Binek, Christian, "Transient spin structures at the antifero-to-paramagnetic phase boundary of FeBr2" (2000). *Christian Binek Publications*. 47.
https://digitalcommons.unl.edu/physicsbinek/47

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Journal of Magnetism and Magnetic Materials 140–144 (1995) 1557–1558

journal of magnetism and magnetic materials

# Transient spin structures at the antiferro-to-paramagnetic phase boundary of $FeBr_2$

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## Abstract

Excess magnetization and anomalous susceptibility loss is observed on antiferromagnetic $FeBr_2$ in an axial field $H$ below and above its $H$–$T$-phase boundary between the multicritical temperature ($T_m = 4.6$ K) and the Néel temperature ($T_N = 14.1$ K). These and other unusual properties of $FeBr_2$ are attributed to frustration-induced intraplanar non-critical spin fluctuations, which can be simulated within the framework of a triaxial version of the 2D-ANNNI model.

Both $FeBr_2$ and $FeCl_2$ are considered as textbook examples of Ising-type metamagnets [1] with tricritical points on their magnetic field ($H$) vs. temperature ($T$) phase boundary. Their layered structures consist of triangular $Fe^{2+}$ planes with ferromagnetic (FM) intra-layer coupling, separated from each other by two diamagnetic halide layers and coupled by weak antiferromagnetic (AF) exchange, $J'$. In spite of their qualitative similarities both phase diagrams show significant differences [2], which are not understood by simply scaling with the relevant interaction parameters [3]. Recently [4] it was argued that the in-plane frustration in $FeBr_2$ might be at the origin of the unusual features of its $H$–$T$ phase diagram.

This idea is pursued in the present paper. We report on measurements of the magnetization $M$ and of the complex susceptibility $\chi' - i\chi''$ as functions of $H$ and $T$. Precursor anomalies are observed below the second-order phase transition (PT) line, $H_c$ vs. $T$, between the multicritical and Néel temperature, $T_m = 4.6$ K and $T_N = 14.1$ K. They are attributed to transient non-uniform spin structures within that sublattice, whose FM order is reversed as $H > H_c$. These anomalies are described within a model closely related to the well-known [5] two-dimensional (2D) ANNNI model. The 2D spin fluctuations are non-critical similarly to those observed on the edge layers in the $\langle 3 \rangle$-phase of the conventional 3D-ANNNI model [6].

The magnetization was measured by SQUID magnetometry on [0001] oriented single crystal samples with thickness $t \approx 0.2$ mm. Fig. 1 shows $M$, $dM/dH_a$, $\chi'$ and $\chi''$ vs. applied field $H_a$ at $T = 6.0$ K. The second-order PT at $H_c = 2.54$ MA/m yields, as expected [2], sharp peaks of both $dM/dH_a$ and $\chi'$. Below $H_c$, however, they are asymmetrically broadened and reveal a plateau, $dM/dH_a \approx$ const. This seems to hint at an intermediate spin-flop phase with continuously rising magnetization. However, field-induced in-plane spin components were excluded by recent Mössbauer investigations [7] and the prominent peak of $\chi''$ vs. $H_a$ (Fig. 1) is incompatible with spin-flop long-range order (LRO). It rather hints at strong fluctuations centered at about $0.88H_c$. Interestingly, another peak of $\chi''$ appears in the paramagnetic (PM) phase at $H \approx 1.07H_c$, whereas the critical fluctuations at $H \approx H_c$ are nearly suppressed.

These results are corroborated by the $T$ dependences of $M$, $\chi'$ and $\chi''$ shown in Fig. 2 in the range $2 \leq T \leq 16$ K for constant fields $0.4 \leq H_a < 2.9$ MA/m. The mixed AF + PM phase yields virtually constant isomagnets $M(T)$ at $T_c < T_m$ up to kink points (arrows) for $2.4 \leq H_a \leq 2.8$

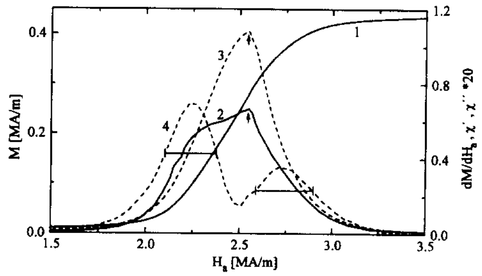

Fig. 1. $M$ (1), $dM/dH_a$ (2), $\chi'$ (3) and $\chi''$ (4) (both at frequency $f = 20$ Hz) vs. $H_a$ measured on $FeBr_2$ at $T = 6.0$ K. $H_c = 2.54$ MA/m and field ranges of strong non-critical fluctuations (see text) are indicated by arrows and bars, respectively.

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*SSDI* 0304-8853(94)00909-0

MA/m. Inflection points of $M(T)$ and sharp cusps of $\chi'(T)$ and $\chi''(T)$ (arrows) determine $T_c(H_a)$ on the second-order PT line. The $H_a$–$T$ phase diagram thus obtained is shown in Fig. 2c. It largely agrees with previous results [2]. Below the phase boundary, in the range $1.6 \leq H_a \leq 2.4$ MA/m, convex-shaped $M(T)$ and $\chi'(T)$ curves and prominent, albeit rounded peaks of $\chi''(T)$ are observed. They coincide with those of $\chi''(H_a)$ (Fig. 1). By plotting the full widths of 60% of the maximum of $\chi''(H_a)$ (Fig. 1) and $\chi''(T)$ (Fig. 2b) a strip-shaped regime of 'strong non-critical fluctuations' is defined in the $H_a$–$T$ plane of Fig. 2c.

We propose that these fluctuations are due to intraplanar frustration as illustrated in Fig. 2 (bottom), where ANNNI-type ⟨3⟩ structures [5] with finite correlation length appear on the down-spin sublattice. In the case of $FeBr_2$ we propose a triaxial version of the 2D-ANNNI model ('TNNNI') with 6 nearest ($J_1 > 0$) and 6 third-nearest neighbors ($J_3 < 0$). In fact, Monte Carlo (MC) simulations of this model with $J_3 = -0.9J_1$, $T = 0.1J_1$ and $H = 0$ (Fig. 3a) show long-lived triple stripe patterns along [1000], [0100] and [0010], respectively. Very probably they define the ground state of the 2D-TNNNI model at sufficiently large $|J_3/J_1|$. They resemble double stripes and 4 × 4 checkerboard patterns of the biaxial 2D-ANNNI model ground state [5]. In order to simulate the situation of $FeBr_2$ ($J_3 = -0.21J_1$, $J_2 = -0.02J_1 \approx 0$ [8]) at $T_m < T < T_N$ and $H_a < H_c \approx J'$, we performed MC calculations letting $J_3 = -0.3J_1$, $T = 0.7J_1$ and $H = -0.1J_1$. Although this system maintains its down-spin LRO, it reveals surprisingly long-lived mesoscopic up-spin clusters (Fig. 3b), which resemble the above triple stripes.

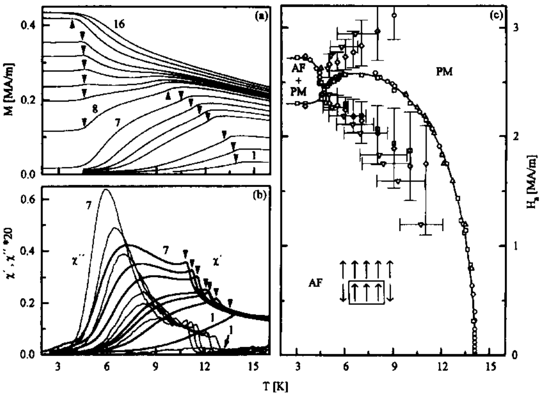

Fig. 2. (a) $M$ vs. $T$, and (b) $\chi'$ and $\chi''$ (both at 20 Hz) vs. $T$ measured on $FeBr_2$ at applied fields, (a) 0.4 (1), 0.8 (2), 1.2 (3), 1.8 (4), 1.9 (5), 2.0 (6), 2.22 (7), 2.4 (8), 2.46 (9), 2.5 (10), 2.55 (11), 2.6 (12), 2.63 (13), 2.7 (14), 2.8 (15), 2.87 MA/m (16), and (b) 0.8 (1), 1.6 (2), 1.75 (3), 1.8 (4), 2.0 (5), 2.1 (6), 2.2 MA/m (7). PTs are indicated by arrows. (c) $H_a$–$T$ phase diagram (data points with eye-guiding lines) and regimes of strong non-critical fluctuations (data points with bars and pictogram with typical 2D spin fluctuations; see text) obtained from $M$ vs. $T$ (○) and $H_a$ (□), $\chi'$ vs. $T$ (△), $\chi''$ vs. $T$ (▽) and $H_a$ (◇) (Figs. 1, 2a and b).

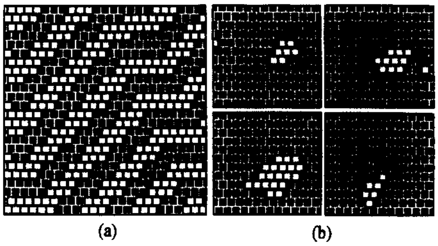

Fig. 3. MC simulated spin configurations (white: up-spins, black: down spins) of the 2D triangular Ising ferromagnet with (a) $J_3 = -0.9J_1$, $T = 0.1J_1$, $H = 0$ at equilibrium, and (b) $J_3 = -0.3J_1$, $T = 0.7J_1$, $H = -0.1J_1$ at four MC steps distance.

Very probably these spin fluctuations cause the excess contributions to $M$ and $\chi'$ as confirmed also by preliminary MC simulations. Their dynamics gives rise to magnetic noise and, hence, to peaks of $\chi''$. Furthermore, we propose that they also give rise to anomalously small intraplanar AF correlation lengths observed at $H = 0$ above $T_N$ [8]. In addition they probably enhance the stability of the AF phase by entropy against the spin-flipping field. This explains the unusual balloon-like shape of the $H$–$T$ phase line at $T > T_m$. The peak of $\chi''$ at $H > H_c$ (Fig. 1) is probably also due to 2D-TNNNI fluctuations appearing, however, on all $Fe^{2+}$ layers. The suppression of $\chi''$ at the crossover from 2D non-critical to 3D critical fluctuations involving $J'$ in addition to $J_1 + J_3$ at $H \approx H_c$ (Figs. 1 and 2b) is hitherto poorly understood.

*Acknowledgements:* We thank W. Selke for useful discussions and DFG (SFB166) for financial support.

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