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Polydispersivity of non-critical field-induced fluctuations in FeBr2

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Polydispersivity of non-critical field-induced fluctuations in $FeBr_2$

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Abstract

The field and temperature dependence of the complex low-frequency susceptibility of the metamagnet $FeBr_2$ exhibits large anomalies in the vicinity of the second-order phase boundary $H_c(T)$. The low-frequency losses reflect polydispersive dynamics with a broad distribution of relaxation times peaking at $\tau \approx 0.2$ s. The dispersion, χ versus f, is well described within Chamberlin's model of dynamically coupled domains. The heterogeneous glass-like response gives rise to the intuitive picture of a magnetic fluid of high viscosity.

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Recent SQUID investigations of the magnetization, M, and of the complex AC susceptibility, $\chi = \chi' - i\chi''$, of the metamagnetic antiferromagnet $FeBr_2$ in axial magnetic fields revealed large anomalies in the vicinity of the antiferromagnetic (AF) to paramagnetic (PM) phase boundary [1]. This renewed the experimental [2–5] and theoretical [6, 7] interest in this layered hexagonal antiferromagnet. According to Selke [6], the extraordinarily large coordination number, $N = 20$, of nearest interplanar AF bonds associated with a relatively large ratio of the AF interplanar and the ferromagnetic intraplanar exchange is essential for the occurrence of anomalies in the magnetization M and the specific heat c_m. They are reminiscent of a new first-order phase boundary, which is conjectured to connect a critical endpoint, CEP, at $T_{CE} = 4.6$ K (traditionally identified as tricritical point, TCP) with a bicritical endpoint, BCE, at a temperature $T_{BE} > T_{CE}$ [3, 4, 7].

Most remarkably, the anomalies in the magnetic loss function χ'' have been found at unusually low frequencies, $f \approx 20$ Hz [1], i.e. orders of magnitudes below typical spin-wave frequencies, $f \approx 10^{10}$ Hz. They are obviously due to slowly fluctuating transient spin structures, which are close to be domain forming, but still far from becoming static. It is the purpose of the present paper to present a first systematic investigation of the dynamics involved in these fluctuations.

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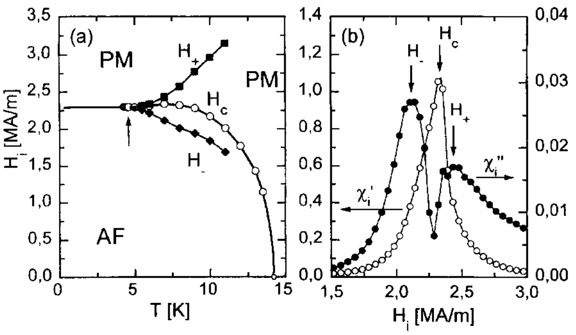

Fig. 1. (a) Magnetic phase diagram, H_i versus T from peak positions of $\chi_i'(H_i)$ (open circles) and $\chi_i''(H_i)$ (solid squares and diamonds) as shown for $T = 7$ K in (b). Arrow in (a) indicates position of CEP at $T_{CE} = 4.6$ K.

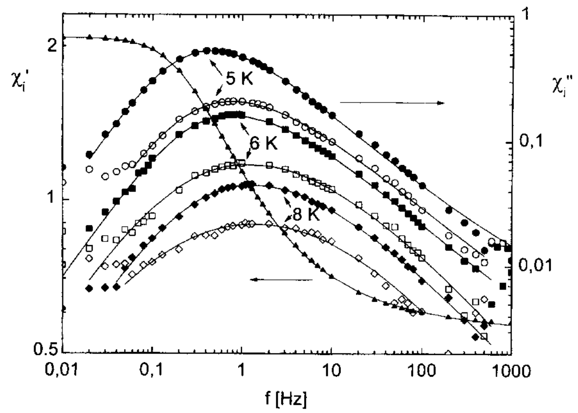

Fig. 2. $\chi_i''(f)$ for $T = 5$, 6, 8 K at $H_i = H_-(T)$ (solid symbols) and $H_i = H_+(T)$ (open symbols). $\chi_i'(f)$ is shown for $T = 5$ K and $H_i = H_-(T)$.

Fig. 1a shows the magnetic phase diagram (PD) of $FeBr_2$, plotted as internal field H_i versus T. It is extracted from χ' and χ'' data measured at constant frequency $f = 7$ Hz after correction for demagnetizing fields. The internal field is given by $H_i = H - NM$, where H is the applied field and the demagnetizing factor $N = 0.76$, calculated from $M(H)$ data. For the internal susceptibility then follows $\chi_i \equiv \partial M/\partial H_i = \chi/(1 - N\chi)$. For $T > T_{CE} = 4.6$ K (Fig. 1a, arrow) the real part χ_i' peaks at the second-order phase-transition line $H_c(T)$ (open circles), as shown in Fig. 1b for $\chi_i'(H_i)$-data (open circles) at $T = 7$ K. A similar peak is found along the metamagnetic transition line of first-order below T_{CE}. The non-critical lines $H_-(T)$ and $H_+(T)$ (Fig. 1a solid diamonds and squares, respectively) are extracted from the successive peak positions of the broad anomalies in the $\chi_i''(H_i)$-curves as shown in Fig. 1b for $T = 7$ K. The absence of an anomaly of χ_i'' at H_c is probably due to the low measuring frequency, which falls way below the critical relaxation frequencies expected at $f \approx 10^{10}$ Hz. Hence, only χ_i' is sensitive to the phase transition.

Fig. 2 shows the dispersion of $\chi_i''(f)$ at frequencies $10^{-2} \leqslant f \leqslant 10^3$ Hz for $T = 5$ K at $H_+ = 2.30$ and $H_- = 2.28$ MA/m (open and solid circles, respectively), $T = 6$ K at $H_+ = 2.34$ and $H_- = 2.22$ MA/m (open and solid squares, respectively) and $T = 8$ K at $H_+ = 2.57$ and $H_- = 2.01$ MA/m (open and solid diamonds, respectively). In addition, one representative dispersion step of $\chi_i'(f)$ is shown for $T = 5$ K at $H_- = 2.28$ MA/m (solid triangles).

The solid lines are results from best fits of Chamberlin's model of dynamically coupled domains [8, 9] to the data of $\chi_i'(f)$ and $\chi_i''(f)$. His model connects the dynamics of the transient domain structures with heterogeneous magnetic response. According to the representation of time-dependent relaxation processes in terms of a superposition of Debye-type exponential relaxations, one obtains

$$\chi''(f) = \phi_0 \int \mathrm{d}x\, n(x)\, x[f/w(x)]/\{1 + [f/w(x)]^2\}, \quad (1)$$

where ϕ_0 is an amplitude, $n(x)$ is the distribution function of domains of size x and $w(x)$ is the inverse relaxation time. In contrast with an Arrhenius-ansatz, however, the relaxation rate is given by $w(x) = w_\infty \exp(-C/x)$ with possible coefficients $C < 0$ and $C > 0$. Since the non-critical fluctuations are transient structures in thermal equilibrium, the size distribution $n(x)$ is of Gaussian-type, $n(x) = \exp[-(x - \langle x \rangle)^2]$, where x is normalized to the width of $n(x)$.

Fig. 3 shows the results of the fitting procedures at $H_i = H_-$ (solid symbols) and H_+ (open symbols), for fixed temperatures $T = 5$, 6 and 8 K, respectively. Surprisingly, both sets of parameters show the same qualitative temperature dependence.

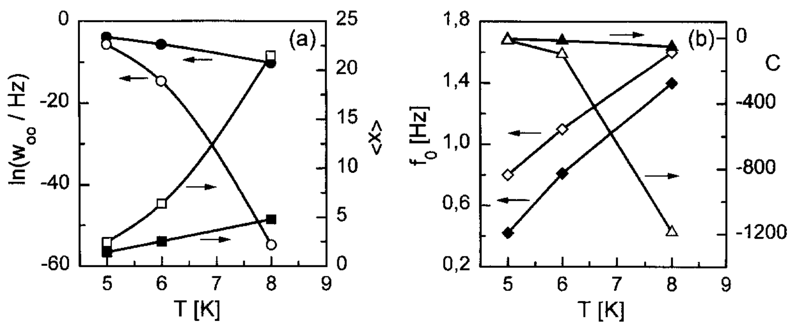

Fig. 3. Fitting parameters $\ln(w_\infty)$ (circles), $\langle x \rangle$ (squares) and C (triangles) versus T of the data in Fig. 2 to Eq. (1) and peak positions f_0 of the $\chi_i''(f)$-curves (diamonds). Open symbols represent data for $H_i = H_+(T)$ and solid symbols for $H_i = H_-(T)$, respectively.

This result and the similarity of the corresponding $\chi_i''(f)$-curves suggest that the H_-- and H_+-fluctuations are of the same nature, probably due to the conjectured AF II phase [6, 7]. While the normalized cluster size $\langle x \rangle$ increases, the correlation coefficient C and the largest relaxation rate w_∞ decrease with increasing temperature. In addition to these fitting parameters, Fig. 3b shows the peak frequencies, f_0, of the $\chi''(f)$-curves. They decrease from $f \approx 1.5$ to ≈ 0.5 Hz with decreasing temperature. Presumably this indicates a freezing tendency of the transient spin structures into a static domain state, which accompanies the conjectured [3, 4, 7] first-order phase-transition line between the CEP and the BCE. Its very existence, however, cannot be evidenced by this finding and still remains an open question.

In conclusion, the polydispersive and heterogeneous low-frequency response of the non-critical fluctuations resembles the dynamics of glass-forming liquids or spin glasses. Being quite uncommon for ergodic magnetic systems it stresses the anomalous behaviour of $FeBr_2$.

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