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Critical behavior and metastability of the magnetization in the random-field Ising system $Fe_{0.7}Mg_{0.3}Cl_2$

U. A. Leitão and W. Kleemann
Angewandte Physik, Universität Duisburg, 4100 Duisburg 1, Federal Republic of Germany

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Optical Faraday rotation was used to measure the uniform magnetization M vs T of the random-field Ising-model (RFIM) system $Fe_{0.7}Mg_{0.3}Cl_2$ in fields $65 \leq H \leq 500$ mT. In disagreement with previous specific-heat results, but in accordance with the "standard" RFIM system $Fe_{1-x}Zn_xF_2$, critical exponents $\phi = 1.41 \pm 0.05$ and $\tilde{\alpha} \sim 0$ are obtained from $(\delta M/\delta T)_H$ and $(\delta M/\delta H)_T$. Dynamical rounding due to the finite measurement time scale, $\tau \sim 100$ s, occurs below $|t| \sim 0.003$. Excess magnetization ΔM is found in the field-cooled domain state in agreement with previous computer simulations. Scaling as $\Delta M \propto H^2$ and the essential time independence are compatible with "opaque" potential-barrier models.

Many aspects of the random-field Ising model (RFIM)[1] have found their realization in Fishman-Aharony systems,[2] i.e., in diluted uniaxial antiferromagnets (AF's) in a uniform magnetic field H applied parallel to the easy c axis.[3] Despite the considerable progress made from both the theoretical and the experimental points of view, open questions are still quite numerous. Current interest is focused mainly on systems with dimension $d = 3$ with the goal a better understanding of (i) static critical behavior and scaling, both of which seem to hint at an effective dimensionality $\tilde{d} = 2$,[3] (ii) critical dynamics,[4] which seems to involve unusually large critical exponents $\tilde{z}$,[5] or even "activated" dynamic scaling,[6] and (iii) metastability as evidenced by field-cooling- (FC) induced domain states,[7] whose interdependence with the critical dynamics[5,6] is still far from being clear.

Recently, studies on $Fe_{1-x}Zn_xF_2$ of the ac susceptibility, $\chi'(\omega)$,[4] and of the uniform magnetization, M,[8] have proved to be particularly suitable for testing both static and dynamic scaling predictions. However, effects due to FC-induced metastability remained undetected in these investigations. This was surprising, since according to computer simulations[9] distinct differences between FC and zero-field-cooled (ZFC) magnetization should be observable at large enough RF. Presumably this condition was not fulfilled in the previous experiments[4,8] at moderate fields, $H \sim 1$ T. A more favorable situation is met in our present investigation of $Fe_{0.7}Mg_{0.3}Cl_2$, which has been known to exhibit random field (RF) effects in both magnetic neutron scattering[10] and specific-heat, c_m, measurements.[11] For the first time we shall report on the expected[9] enhancement of M upon FC in comparison with that obtained after ZFC.

Under ZFC conditions both $(\partial M/\partial T)_H$ and $(\partial M/\partial H)_T$ diverge logarithmically as in the classical RFIM system $Fe_{1-x}Zn_xF_2$.[8] This signifies $\tilde{\alpha} \sim 0$, in sharp contrast with the specific-heat result, $\tilde{\alpha} \sim -1$.[11] Moreover, also our exponent $\phi = 1.41 \pm 0.05$ describing the crossover from random-exchange to RF behavior, disagrees with that obtained from c_m-vs-T data,[11] but confirms the value expected theoretically[2,12] and observed in $Fe_{1-x}Zn_xF_2$.[3,8] In our opinion the reliability of our data is warranted by the local resolution of the Faraday-rotation (FR) technique[8] for the measurement of M, which avoids falsifying concentration gradients of the mixed-crystal samples to a large extent. Our new results are also of interest in the recent discussion[11,13] on Fisher renormalization of the critical exponents in the case of Fishman-Aharony systems. $\tilde{\alpha} \sim 0$ would imply a trivial renormalization factor $(1-\tilde{\alpha})^{-1} \sim 1$, thus leaving unaffected the values of *all exponents* hitherto obtained on three-dimensional (3D) RFIM systems.[3]

The RFIM regime of $Fe_{0.7}Mg_{0.3}Cl_2$ is restricted to fields $H \lesssim 0.4$ T owing to the occurrence of spin-flip transitions at about 0.6 T.[14] A full account of the H vs T phase diagram obtained from FR measurements along with a detailed study of the tricritical point will be given elsewhere.

All measurements were carried out on Bridgman-grown samples, which exhibit typical concentration gradients of $\Delta x/\Delta l \sim 0.005$ cm^{-1} along the growth direction lying in the basal plane. Since the narrow HeNe laser beam (diameter 0.6 mm) probes the sample along its c axis (thickness ~ 0.2 mm), rounding of the transition due to Δx is believed to be no larger than $\delta t = 1.5 \times 10^{-3}$ $(t = T/T_c - 1)$. The FR angle θ, induced by the magnetic field of a superconducting coil directed along the c axis, was measured with a resolution of $\delta\theta = 0.006°$ by use of a computer-controlled compensation method.[8] The temperatures were stabilized to within 1 mK prior to each measurement and measured with a carbon-glass resistor.

Typical θ-vs-T curves for $H = 130$ and 260 mT are shown in Fig. 1. As in $FeCl_2$,[15] the FR is proportional to the uniform magnetization, as verified by comparison with recent[16] M vs T data obtained on $Fe_{0.716}Mg_{0.284}Cl_2$. Our curves were taken upon field heating (FH) after ZFC. The inset of Fig. 1 shows a FH-FC cycle with $H = 260$ mT around the maximum-slope temperature, $T_c = 12.54$ K, after ZFC. It is seen that the FC magnetization exceeds the ZFC signal, $\Delta\theta = \theta(\text{FC}) - \theta(\text{ZFC}) > 0$, at temperatures $T < T_{\text{eq}}$, where $T_{\text{eq}} \sim 12.65$ K. It was observed that the FC branch of the cycle is fully reversible on subsequent FH. This strongly hints at frozen-in metastability in the FC state, which will be recalled in more detail

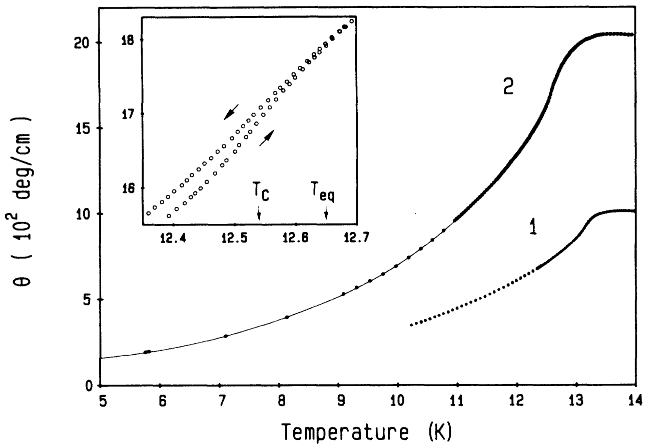

FIG. 1. Temperature dependence of the Faraday rotation measured with $\lambda = 633$ nm on $Fe_{0.7}Mg_{0.3}Cl_2$ at (1) $H = 130$ mT and (2) 260 mT (with eye-guiding line) applied after ZFC. The inset shows a temperature cycle at $H = 260$ mT around $T_c = 12.54$ K and $T_{eq} = 12.65$ K (arrows) after ZFC.

below. Let us first discuss the equilibrium properties attributed[3] to the ZFC configuration.

Figure 2 shows the derivative $(\delta\theta/\delta T)_H$ vs T in the vicinity of $T_c(H)$ for different field strengths. Similarly as observed[8] in $Fe_{1-x}Zn_xF_2$ sharp peaks appear thus defining $T_c(H)$. On increasing H, they become more symmetric and shift to lower temperatures according to the crossover scaling relation[2]

$$T_N - T_c(H) = bH^2 + cH^{2/\phi} , \qquad (1)$$

where ϕ is the crossover exponent and bH^2 denotes the mean-field correction. Assuming the validity of the relation[11]

$$b(x) = b(0)T_N(0)/T_N(x) ,$$

we obtain $b(0.3) = 2.62$ K/T^2 inserting $b(0) = 1.49$ K/T^2,[11] $T_N(0) = 23.60$ K, and $T_N(0.3) = 13.45$ K. Then a least-squares fit of the internal field H_i vs $T_c(H_i)$ to Eq. (1) (Fig. 2, inset) for $H_i \leq 320$ mT yields $\phi = 1.41 \pm 0.05$. Very satisfactorily this value is near to that of the standard 3D RFIM system $Fe_{1-x}Zn_xF_2$, $\phi = 1.42 \pm 0.03$,[3,8] in agreement with theoretical predictions.[12]

It is noted that our value of ϕ lies well outside the error margins of that obtained[11] from c_m vs T data, $\phi = 1.25 \pm 0.11$. One reason for the obvious discrepancy is suspected to lie in the lack of demagnetization correction in the former study.[11] Our data were corrected for this effect using the demagnetization factor achieved from FR data of the mixed phase in the spin-flip regime following the procedure of Dillon *et al.*[15] However, the correction turned out to be virtually constant, $H_i \sim 0.75H$ for $H_i \leq 320$ mT. Hence, the value of ϕ does not change on replacing H_i by the applied field H in Eq. (1). More likely the reliability of the previous[11] ϕ value may have suffered from severe drawbacks of the experimental method involved: Taken at a speed of 0.12 K/s the c_m vs T curves were *not* in equilibrium (e.g., peak shifts up to 0.1 K occur between FH and FC), whereas our data rely on point-by-point equilibration and subsequent waiting times of 10^2 s. It is, however, unclear how these uncertainties do change the measured H-T phase diagram *systematically*. We rather believe that systematic errors originate from the unavoidable T_c spread due to the concentration gradients in relatively large sample volumes. Because of their asymmetry, the c_m vs T curves[11] peak at temperatures T_c, which are lower than the sample averages $\overline{T}_c$.[17] It is easily seen that the misfit, $\overline{T}_c - T_c$, in-

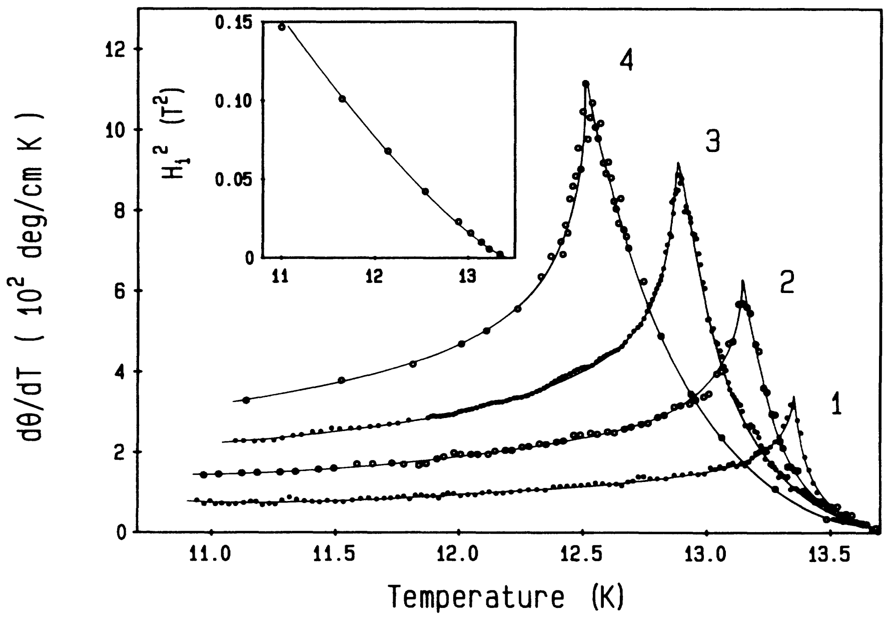

FIG. 2. $(\delta\theta/\delta T)_H$ vs T for ZFC $Fe_{0.7}Mg_{0.3}Cl_2$ measured at (1) $H = 65$, (2) 130, (3) 195, and (4) 260 mT, respectively. The solid lines are guides for the eye. The inset shows H_i^2 vs $T_c(H_i)$ best fitted to Eq. (1) (solid line).

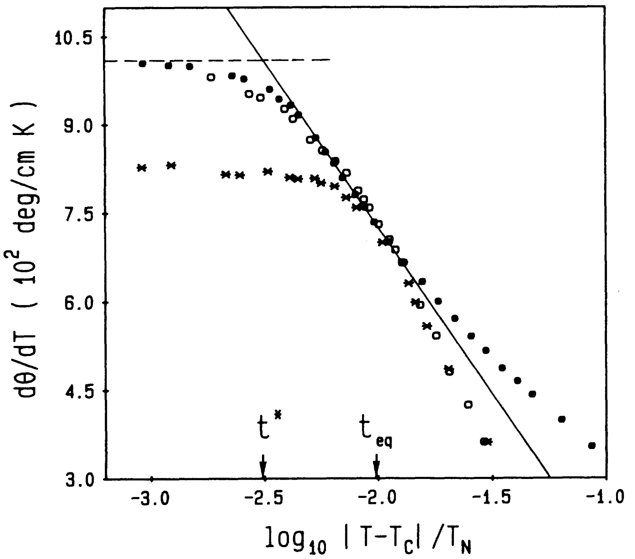

FIG. 3. $(\delta\theta/\delta T)_H$ vs $\log_{10}|t|$ for $H=260$ mT (cf. Fig. 2), where $t=(T-T_c)/T_N$ with $T_c=12.54$ K and $T_N=13.45$ K, after ZFC (filled circles: $t<0$, open circles: $t>0$) and upon FC (asterisks: $t>0$), respectively. t_{eq} (arrow) corresponds to T_{eq} in Fig. 1. The horizontal line through the peak height and the $\log_{10}|t|$ line intersect at t^* (arrow).

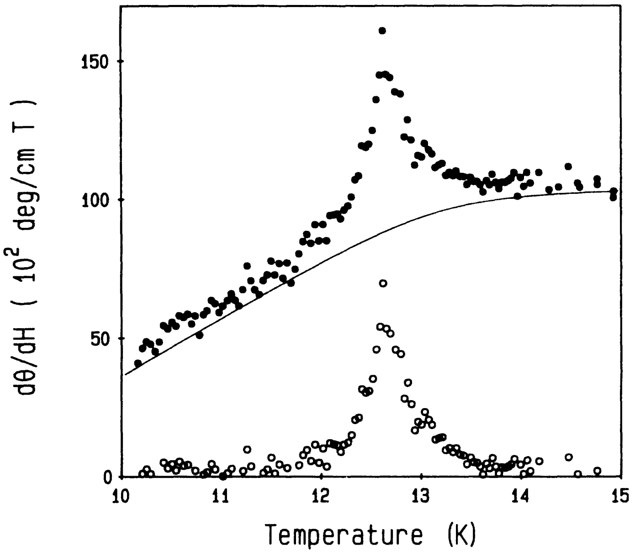

FIG. 4. $(\delta\theta/\delta H)_T$ vs T for ZFC $Fe_{0.7}Mg_{0.3}Cl_2$ measured at $H=260$ mT with $\delta H=8$ mT (filled circles) and its singularity (open circles) after subtracting a standard background function (solid line, see text).

creases with increasing H. Hence, the emerging H vs $\bar{T}_c$ phase line bends more strongly than would the H vs $\bar{T}_c$ curve, thus yielding too small a value of ϕ. Significantly smaller errors of this kind are expected in our FR measurements, since they probe about 30-times-smaller sample volumes and yield symmetric singularities of $(\delta\theta/\delta T)_H$, hence $T_c \sim \bar{T}_c$.

A plot of $(\delta\theta/\delta T)_H$ vs $\log_{10}|t|$ for $H=260$ mT in Fig. 3 reveals a straight line for the ZFC data within $t^*=0.003 \leq |t| \leq 0.02$ thus indicating a specific-heat exponent $\tilde{\alpha}\sim 0$ in close agreement with previous FR experiments.[8] Rounding occurs below t^*, which is significantly larger than the Δx-induced rounding interval $\delta t \sim 0.0015$. Albeit less clearcut than in the $Fe_{1-x}Zn_xF_2$ study,[8] we believe the excess rounding to be due to the extreme slowing down of the critical dynamics of the RFIM system[4-6] on the time scale of our experiment ($\tau \sim 10^2$ s).

Much stronger rounding, starting below $|t| \sim 0.01$ for $H=260$ mT, appears upon FC (Fig. 3). This temperature is close to the equilibrium boundary, $t_{eq} \sim 0.009$ (Fig. 1, inset), below which the system is believed to condense into a domain state.[3] RFIM critical behavior is, hence, restricted to the range $t_{eq} < t < 0.02$ under FC conditions, where the upper boundary presumably indicates crossover into the random exchange regime.[3]

The "dc" susceptibility corresponding to $(\delta\theta/\delta H)_T$ is determined as the difference of two θ vs T curves taken at $H \pm \delta H/2$ with $H=260$ mT and $\delta H=8$ mT (Fig. 4). It shows similar critical behavior as does $(\delta\theta/\delta T)_H$ (Fig. 2) in its singular part. This is obtained by subtracting a smooth regular background,[8] which would correspond to $\chi'(\omega \rightarrow \infty)$. A logarithmic plot reveals a symmetric straight line in the range $0.003 < |t| < 0.06$, thus confirming the critical exponent $\tilde{\alpha}\sim 0$. The onset of dynamic rounding below $t^*=0.003$ is, again, believed to be due to the finite "frequency" of the experiment, $\omega = \tau^{-1}$ with $\tau \sim 10^2$ s.

As shown previously,[8] both $(\delta\theta/\delta T)_H$ and $(\delta\theta/\delta H)_T$ are related to second derivatives of the free energy F. They are expected to scale as $H^y|t|^{-\tilde{\alpha}}$ with different amplitude exponents y_1 and y_2, respectively. Whereas for $Fe_{1-x}Zn_xF_2$ the experimental values $y_1 \sim 0.6$ and $y_2 \sim 1.0$ are close to the theoretical predictions,[8] the values found for $Fe_{0.7}Mg_{0.3}Cl_2$, $y_1 = 1.1 \pm 0.2$ and $y_2 = 1.3 \pm 0.5$, seem to differ significantly. As outlined above, demagnetization effects are not expected to account for this discrepancy. We rather believe that the proximity of the tricritical point ($T_t \sim 8.5$ K, $H_t \sim 0.6$ T) plays a crucial role. Presumably the exponents y do change, if the ordinary field-induced crossover from antiferromagnetic to spin-flip behavior would properly be accounted for in the scaling form of F. Unfortunately, to the best of our knowledge theoretical predictions for the scaling properties of weak-anisotropy randomly diluted AF's in a uniform field H are lacking up to now.

Recently[11,13] it was argued that Fisher renormalization might affect the RFIM critical behavior of Fishman-Aharony systems. Since the RF varies like $M(T)$ at constant applied field H, the phase transition is not observed along a path parallel to the T axis in the RF vs T phase diagram. Renormalized exponents, e.g., $\tilde{\alpha}' = -\tilde{\alpha}/(1-\tilde{\alpha})$, are expected to appear, if $\tilde{\alpha} \geq 0$.[13] However, in view of $\tilde{\alpha}'=0$ emerging from this and previous[3,8] investigations, one readily obtains the trivial renormalization $\tilde{\alpha}=\tilde{\alpha}'=0$, $\tilde{\gamma}=\tilde{\gamma}'$, etc. Nontrivial renormalization is thus presently a mere speculation, unless future very low frequency measurements will reveal asymptotic exponents $\tilde{\alpha}>0$ as recently predicted.[18]

Let us finally discuss the $\Delta\theta$ effect appearing upon a ZFC-FH-FC cycle around $T_c(H)$ and $T_{\rm eq}(H)$ (Fig. 1, inset). Figure 5 shows $\Delta\theta$ vs t for different values of H. The reversibility temperatures $t_{\rm eq}$ are of the order 10^{-2} and increase with increasing H. Scaling with $t_{\rm eq} \propto H^{2/\phi}$ (after mean-field correction) is expected,[3] but cannot safely be confirmed because of the large scatter of our data points. Maximum values $\Delta\theta_m$ are obtained at $t_m \sim -t_{\rm eq}$. On further lowering T we find a monotonous decrease of $\Delta\theta$, closely resembling that of θ (ZFC) vs T and yielding residual values $\sim\Delta\theta_m/10$ at liquid-helium temperature. Most remarkable is the proportionality of $\Delta\theta_m$ with H^2 (Fig. 5, inset). Furthermore, virtually *no* time dependence of $\Delta\theta$ was found within $5\,{\rm s} < \tau < 7200$ s after quenching from above $t_{\rm eq}$, $t \sim 0.05$, into metastable states at constant temperatures $|t| \lesssim 0.02$. All of these properties were reproduced on different samples for $H \lesssim 0.4$ T, reasonable signal-to-noise ratios requiring $H > 50$ mT.

The $\Delta\theta$ effect is qualitatively compatible with recent mean-field computer experiments on randomly diluted Ising AF's:[9] (i) The M vs T curves smoothly split just below $M_{\max}$, where $M({\rm FC}) > M({\rm ZFC})$ in the low-field limit; (ii) the excess magnetization of FC samples is reversible at all T; (iii) ΔM is preferentially concentrated at the AF domain walls, which mainly nucleate at vacancies of the spin system. Sufficiently far from T_c (in our experiments tentatively at $t \lesssim t_m$) the walls become smooth and well defined. Hence, assuming a system of domains with average radius R and constant excess surface magnetization density, one readily finds $\Delta M \propto R^{-1}$. Since R generally depends on both the RF and the time τ, $R(H,\tau) \propto H^{-\nu_H} r(\tau)$,[7] we thus expect

$$\Delta\theta \propto H^{\nu_H}/r(\tau) \ . \qquad (2)$$

Experimentally, we find $\nu_H = 2.0 \pm 0.1$ (Fig. 5, inset) in perfect agreement with $\nu_H = 2$ predicted for broad domain walls in $d = 3$.[7] Time independence, $r(\tau) \sim$ const, as observed for $\tau > 5$ s, seems to agree with Bruinsma and Aeppli's[7] low-temperature prediction of frozen domains with size R and "opaque" potential barriers. Within experimental accuracy, however, very weak time dependence $r(\tau) \propto \ln(\tau/\tau_0)$ with $\tau_0 \sim 10^{-13}$ s, as predicted by Villain[7] for thermal depinning of domain walls, may not completely be ruled out. This would better agree with our experimental condition, $T \lesssim T_c$, and with recent neutron data[10] on $\rm Fe_{0.7}Mg_{0.3}Cl_2$ showing weak time dependence of the FC state. Systematic studies to check very weak time dependence of $\Delta\theta$ are presently under way. The final discussion should also account for possible failure of our basic assumption $\Delta M \propto R^{-1}$ under isothermal growth conditions.

The decrease of $\Delta\theta$ for $t < t_m(H)$ is primarily due to the decrease of the RF being proportional to $M(T)$, hence the close correspondence with θ(ZFC) vs T (Fig. 1). It remains to be shown if a strict proportionality $\Delta\theta \propto \theta^{\nu_H}$ holds, as suggested by relation (2). This test would shed more light on the very origin of the excess *uniform* magnetization in the FC state, which seems to be related to the simultaneous decrease of the *staggered* magnetization.[10] Future discussions should also be extended to other dilute AF, showing either very large ($\rm Mn_{1-x}Zn_xF_2$, Ref. 19) or nearly negligible ($\rm Fe_{1-x}Zn_xF_2$, Ref. 20) ΔM effects.

In conclusion, our FR data have provided evidence that solid solutions of $\rm Fe_{1-x}Mg_xCl_2$ fit well with many predictions for the RFIM in both the ZFC equilibrium and the metastable FC domain state. Strong similarity with the only hitherto undisputed model system, $\rm Fe_{1-x}Zn_xF_2$, is now apparent. It will be interesting to test and compare other important properties, e.g., dynamical scaling observed via $\chi'(\omega)$ or $(\delta\theta/\delta H)_T$ vs ω. It is hoped that ex-

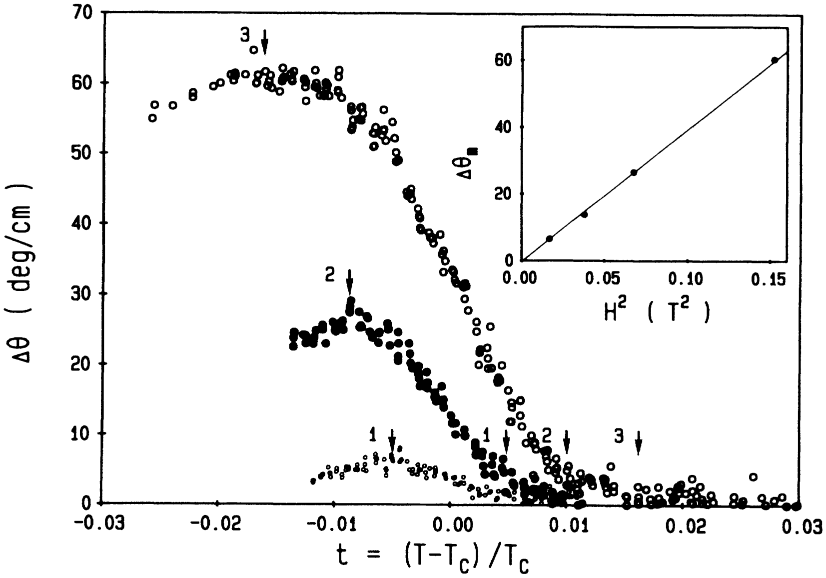

FIG. 5. Excess Faraday rotation $\Delta\theta$ vs $t = T/T_c - 1$ of $\rm Fe_{0.7}Mg_{0.3}Cl_2$ cycled around $T_c(H)$ (see Fig. 1) measured at (1) $H = 130$ mT ($T_c = 13.15$ K), (2) 260 mT ($T_c = 12.54$ K), and (3) 390 mT ($T_c = 11.62$ K). The respective temperatures $t_{\rm eq} > 0$ and $t_m < 0$ (see text) are indicated by arrows. The inset shows $\Delta\theta_m$ vs H^2 together with a best linear fit (solid line).

periments of this kind will eventually help to find the true asymptotic exponents predicted theoretically.[18,21] Furthermore, in $Fe_{1-x}Mg_xCl_2$ a detailed study of the competition between RF and spin-flip processes near to the tricritical point will be of special interest.

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