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1995 J. Phys.: Condens. Matter 7 L1

(http://iopscience.iop.org/0953-8984/7/1/001)

J. Phys.: Condens. Matter 7 (1995) L1–L6. Printed in the UK

LETTER TO THE EDITOR

Critical behaviour of the linear and non-linear magnetic susceptibilities of $FeCl_2$

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Received 24 October 1994

Abstract. The linear and non-linear uniform magnetic susceptibilities of the three-dimensional Ising antiferromagnet $FeCl_2$ were studied by using quasi-static magneto-optical Faraday rotation. Scaling analysis of their critical exponents at $H = 0$, $2\beta' = 0.83 \pm 0.05$ and $\gamma' = -0.19 \pm 0.05$, allows us to determine the exponent of the specific heat, $\alpha = 0.15 \pm 0.04$, which is in good agreement with other experiments and theoretical predictions.

Scaling analysis shows [1] that the non-linear magnetic susceptibility of dilute antiferromagnets in uniform fields $\boldsymbol{H}$ should diverge at the Néel temperature, T_N. Experimentally this prediction was quantitatively confirmed on the layered system $Fe_{0.7}Mg_{0.3}Cl_2$ by measurements of the magneto-optical Faraday rotation [2]. In particular the random-exchange–random-field crossover exponent $\Phi = 1.44$ was deduced from an analysis of both the linear and the non-linear susceptibilities. It was argued [2] that similar information should be obtained from non-linear susceptibility studies on pure Ising antiferromagnets in a longitudinal field.

The analysis starts from the scaling properties of the free-energy density in the vicinity of T_N,

$$g(t, h, H^2) = L^{-d} g(L^y t, L^x h, L^z H^2) \tag{1}$$

where $t = T/T_N - 1$ and h and H are the ordering (staggered) and non-ordering (uniform) fields, respectively. By setting $h = 0$ and the scaling parameter $L = |t|^{-1/y}$ one readily obtains with $d/y = 2 - \alpha$ (d = dimension):

$$g(t, H^2) = |t|^{2-\alpha} f(H^2 |t|^{-\Phi}). \tag{2}$$

In the pure case the specific heat exponent α governs the asymptotic critical behaviour along the λ-line, $T_t < T \leqslant T_N$, where T_t is the tricritical temperature [3]. Lacking new asymptotic critical behaviour, the 'crossover' exponent $\Phi = z/y$ merely describes the well-known [4] mean-field shift of the critical temperature,

$$T_c(H) = T_N - aH^{2/\Phi} \tag{3}$$

with $\Phi = 1$ and $a > 0$. Inspection of the non-asymptotic tricritical behaviour with $\alpha_t \neq \alpha$, however, reveals interesting field dependences of the 'susceptibilities' $\partial^2 g/\partial T^2$, $\partial^2 g/\partial H^2$

0953-8984/95/010001+06\$19.50

and $\partial^2 g/\partial T\,\partial H$ [5]. They formally correspond to those found for the asymptotic random-field critical behaviour of diluted antiferromagnets in fields with $H \neq 0$ [6].

Here we consider the critical behaviour in the limit $H \to 0$. Equation (2) readily yields the leading divergences of the linear and the non-linear susceptibilities [1],

$$\chi_{\mathrm{L}} = (\partial^2 g/\partial H^2)_{H=0} \propto |t|^{2\beta'} \qquad \text{with } 2\beta' = 2 - \alpha - \Phi \tag{4}$$

and

$$\chi_{\mathrm{NL}} = (\partial^4 g/\partial H^4)_{H=0} \propto |t|^{\gamma'} \qquad \text{with } \gamma' = 2 - \alpha - 2\Phi. \tag{5}$$

Fisher's relation [7] $\chi_{\mathrm{L}} \propto |t|^{1-\alpha}$ and a novel proportionality, $\chi_{\mathrm{NL}} \propto |t|^{-\alpha}$, emerge on inserting the mean-field exponent $\Phi = 1$. Clearly, the exponents $2\beta' = 0.89$ and $\gamma' = -0.11$, as expected for the pure $d = 3$ Ising system with $\alpha = 0.11$ [8], are sharply distinct from those of the diluted one, $2\beta' = 0.62$ and $\gamma' = -0.82$, where $\Phi = 1.44$ and $\alpha = -0.06$ [2]. It is the aim of this letter to check the predictions of (4) and (5) for the first time on the pure Ising antiferromagnet $FeCl_2$. By simultaneously measuring χ_{L} and χ_{NL} both exponents, Φ and α, can be obtained unambiguously. In particular (5) seems promising for determining the specific heat exponent α with high precision.

$FeCl_2$ *crystallizes in the rhombohedral* D^5_{3d} *structure. It consists of ferromagnetically* ordered layers, stacked antiferromagnetically along the pseudo-hexagonal c-axis [9]. The in-plane interaction is ferromagnetic between the next-nearest neighbours, $J_1/k_{\mathrm{B}} = 10$ K. The inter-plane interaction is antiferromagnetic, $J_2/k_{\mathrm{B}} = -0.5$ K. $FeCl_2$ undergoes a first-order antiferromagnetic-to-paramagnetic phase transition when exposed to a magnetic field, H_{a}, applied along the c-axis below the tricritical temperature, $T_{\mathrm{t}} = 21.7$ K. In spite of the remarkable two-dimensional behaviour of the magnetic and crystallographic properties and *owing to the large single-ion anisotropy,* $D/k_{\mathrm{B}} = 10$ K, $FeCl_2$ *shows asymptotic critical* behaviour of the $d = 3$ Ising model for the whole of the λ-line, i.e. for $T_{\mathrm{t}} < T \leqslant T_{\mathrm{N}}$ [10].

The experiments were carried out on a Bridgman-grown sample, which was prepared in a glove box filled with dry helium in order to avoid hydration. A disk-shaped sample with diameter 6 mm and thickness 100 μm was obtained by successively peeling-off layers parallel to the cleavage plane perpendicularly to the c-axis. The sample was mounted in an optical gas-flow cryostat allowing us to control temperatures within 5 and 100 K, equipped with a superconducting solenoid for magnetic fields, $H_{\mathrm{a}} \leqslant 4$ MA m^{-1}. The long-time thermal stability is $\delta T = 2$ mK. The field resolution is better than 800 A m^{-1}. The linearly polarized light beam of a laser diode at wavelength $\lambda = 670$ nm travels, precisely aligned parallel to the easy axis, through the sample and is detected after passing a rotating combination of an elasto-optic modulator with an analyser, as described previously [11]. The Faraday rotation angle, θ, is expected to be proportional to the homogeneous magnetization M [10, 12], and was measured with an accuracy of $\delta\alpha = 5 \times 10^{-4}$ deg.

T_{N} was obtained from isomagnetic measurements at low applied magnetic fields on field heating after zero-field cooling from $T \sim 50$ K $\sim 2T_{\mathrm{N}}$ to 10 K. The inset of figure 1 shows field-heating curves at various fields, $50.2 \leqslant H_{\mathrm{a}} \leqslant 597$ kA m^{-1}. The inflexion points marking $T_{\mathrm{c}}(H_{\mathrm{a}})$ are indicated by open circles. A plot of $T_{\mathrm{c}}(H_{\mathrm{a}})$ versus H_{a} is shown in figure 1. Anticipating the relationship (3) with $\Phi = 1$ we are able to determine T_{N} from a fit to the data within the range $0 \leqslant H_{\mathrm{a}} \leqslant 300$ kA m^{-1} (arrow in figure 1). The best-fitted solid line refers to $T_{\mathrm{N}} = 24.037 \pm 0.005$ K and $a = (3 \pm 1) \times 10^{-6}$ K kA^{-2} m^2. The Neél temperature is in excellent agreement with previous measurements [5, 10, 12] thus providing evidence for the high purity of the sample.

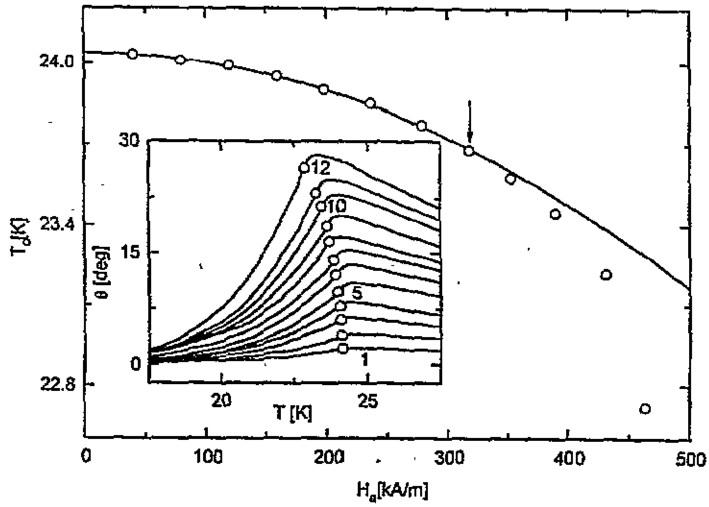

Figure 1. Critical temperature, T_c, versus applied magnetic field H_a. The solid line is the best fit to $T_c(H_a) = T_N - aH_a^2$, with $T_N = 24.037 \pm 0.005$ K and $a = (3 \pm 1) \times 10^{-6}$ K kA^{-2} m^2. The arrow indicates the upper fitting limit. The inset shows isomagnetic Faraday rotation curves, θ versus temperature, T, for H_a = 50.2 (1), 99.5 (2), 149.6 (3), 199.0 (4), 249.1 (5), 299.3 (6), 392.4 (7), 398.0 (8), 448.1 (9), 496.3 (10), 548.4 (11), 597 kA m^{-1} (12). The inflexion points, marking $T_c(H_a)$, are indicated by open circles.

The isothermal magnetic field dependence of the Faraday rotation angle, θ, was recorded at 44 different temperatures, $15.14 \leqslant T \leqslant 25.86$ K, after zero-field cooling from $T \sim 50\ \mathrm{K} \sim 2T_N$ to the measuring temperature, figure 2. After correction for demagnetizing fields θ was analysed using an expansion in terms of odd powers of the internal magnetic field, $H_i = H_a - N\theta$ [2]:

$$\theta = \chi_L H_i + \tfrac{1}{3}\chi_{NL} H_i^3 + O(H_i^5). \tag{5}$$

The demagnetizing factor, N, was estimated from isothermal magnetization curves at low temperatures, $T = 17.91$ K, as shown in figure 2, inset, and is assumed to be independent of the temperature [2, 12]. χ_L and χ_{NL} define the intersection with the ordinate axis and the slope in the limit $H \to 0$, respectively, in a plot of $\partial\theta/\partial H_i$ versus H_i^2.

Figure 3 shows the linear and non-linear susceptibilities, χ_L and χ_{NL}, respectively, versus temperature, T. Obviously, the zero-field ordering temperature, T_N, is well defined by the singularity of χ_{NL}. On the other hand, T_N coincides with the inflexion point of χ_L, which lies well below the maximum at $T_m = 24.42$ K. This is in excellent agreement with the data on the diluted system, $Fe_{0.7}Mg_{0.3}Cl_2$ [2] and theory [7] where T_m is expected to be significantly larger than T_N. In contrast, the AC susceptibility measurements on $Fe_{0.46}Zn_{0.54}F_2$ reveal a coincidence of T_N and T_m at $H_i = 0$ [13]. The origin of this difference was argued [2] to lie in different behaviours of the short-range correlations in the fluoride and the chloride system. Recently, however, random-stress-induced, symmetry-allowed piezomagnetic moments were measured on both FeF_2 and $Fe_{0.47}Zn_{0.53}F_2$ in small residual magnetic fields, $H > 8$ A m^{-1} [14, 15]. They give rise to an extra peak of χ_L at T_N which might dominate over and thus suppress the nearby intrinsic peak at T_m. Very probably this is the main reason for the coincidence of T_N and T_m in $Fe_{0.47}Zn_{0.53}F_2$ [13].

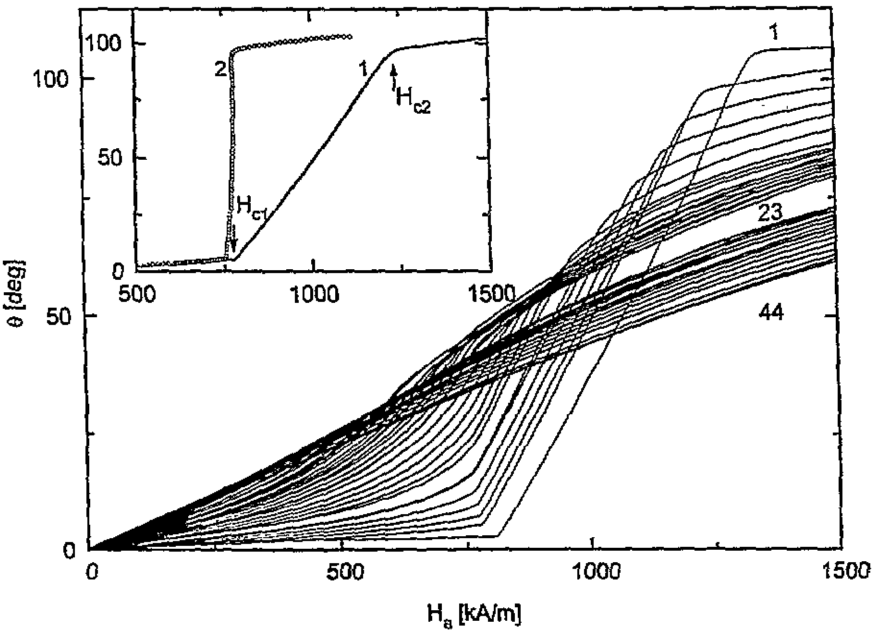

Figure 2. Faraday rotation angle, θ, versus applied magnetic field, H_a, for T = 15.14 (1), 17.91 (2), 18.40 (3), 18.85 (4), 19.38 (5), 19.99 (6), 20.21 (7), 20.53 (8), 20.79 (9), 21.03 (10), 21.27 (11), 21.49 (12), 21.74 (13), 21.96 (14), 22.20 (15), 22.29 (16), 22.39 (17), 22.49 (18), 22.58 (19), 22.68 (20), 22.79 (21), 22.84 (22), 22.89 (23), 23.01 (24), 23.10 (25), 23.19 (26), 23.27 (27), 23.37 (28), 23.48 (29), 23.58 (30), 23.68 (31), 23.78 (32), 23.89 (33), 24.01 (34), 24.10 (35), 24.19 (36), 24.28 (37), 24.38 (38), 24.48 (39), 24.72 (40), 24.95 (41), 25.22 (42), 25.40 (43) and 25.86 K (44). Additionally, θ versus H_a (1) and H_i (2), $H_i = H_a - N\theta$, is shown in the inset. N is obtained from the slope of curve 1, $N = (H_{c2} - H_{c1})/(\theta(H_{c2}) - \theta(H_{c1}))$, where H_{c1} and H_{c2} are indicated by arrows.

Figure 4 shows a double-logarithmic plot of χ_{NL} and $\chi_c - \chi_L$ versus reduced temperature, $|t|$, for $T < T_N$, where $\chi_c = \chi_L(T_N)$; see figure 3. $\chi_c = 15.72$ deg kA^{-1} m and T_N = 24.037 K are found to optimize the linearity of the double-logarithmic plots of figure 4 within $10^{-3} \leqslant |t| \leqslant 2 \times 10^{-2}$. Obviously, the range of linearity is smaller than that found for $Fe_{0.7}Mg_{0.3}Cl_2$ [2]. Assuming power laws, equation (4) with $\chi_c - \chi_L$ replacing χ_L and equation (5), least-squares fits yield $2\beta' = 0.83 \pm 0.05$ and $\gamma' = -0.19 \pm 0.05$ and, after conversion, $\Phi = 1.02 \pm 0.1$ and $\alpha = 0.15 \pm 0.04$. The comparatively large errors refer to the poor statistics of the data points. Nevertheless, the agreement with expected values, $\Phi = 1$ [4] and $\alpha = 0.12$ [10], is very good. In particular the expected difference to the 'random' exponents of the diluted system $Fe_{0.7}Mg_{0.3}Cl_2$, $\Phi = 1.44$ and $\alpha = -0.06$ [2], is convincingly demonstrated.

In conclusion, the unconventional scaling function, equation (2), derived from an expansion around T_N rather than around T_t [3] and involving the irrelevant non-ordering field $\boldsymbol{H}$, has proven to be useful for determining the critical exponent α without ambiguity. Both linear and non-linear magnetic susceptibilities were measured on the prototypical $d = 3$ Ising antiferromagnet $FeCl_2$ using Faraday rotation techniques. It will be interesting to investigate χ_L and χ_{NL} in other uniaxial antiferromagnets, e.g. FeF_2 and $Fe_xZn_{1-x}F_2$, in order to corroborate the findings for $FeCl_2$ and $Fe_{0.7}Mg_{0.3}Cl_2$ [2] and to estimate the influence of piezomagnetic perturbations.

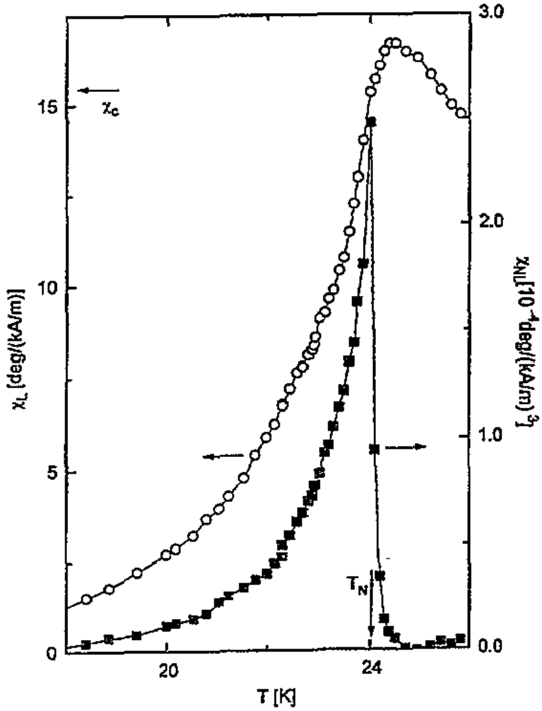

Figure 3. Temperature dependence of the linear and non-linear uniform magnetic susceptibilities, χ_L and χ_{NL}, respectively, as obtained from θ versus H_i (see text). The values of $T_N = 24.037$ K and $\chi_c = 15.72$ deg kA^{-1} m are indicated by arrows.

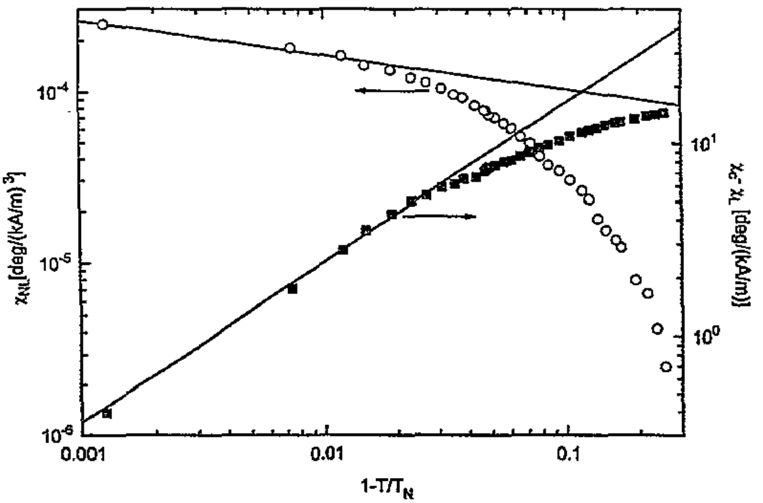

Figure 4. Double-logarithmic plot of $\chi_c - \chi_L$ (solid squares, right-hand scale) and χ_{NL} (open circles, left-hand scale) versus reduced temperature, $1 - T/T_N$, for $T < T_N = 24.037$ K, where $\chi_c = \chi_L(T_N) = 15.72$ deg kA^{-1} m. The solid lines are best fits within $0.001 \leqslant |t| \leqslant 0.02$.

Financial support from the Deutsche Forschungsgemeinschaft via SFB 166 is gratefully acknowledged.

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