STATIC AND DYNAMIC CRITICAL BEHAVIOR IN RANDOM MAGNETS

V. Jaccarino, A. King

To cite this version:
V. Jaccarino, A. King. STATIC AND DYNAMIC CRITICAL BEHAVIOR IN RANDOM MAGNETS. Journal de Physique Colloques, 1988, 49 (C8), pp.C8-1209-C8-1214. <10.1051/jphyscol:19888549>. <jpa-00228760>

HAL Id: jpa-00228760
https://hal.archives-ouvertes.fr/jpa-00228760
Submitted on 1 Jan 1988

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L’archive ouverte pluridisciplinaire HAL, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d’enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.
STATIC AND DYNAMIC CRITICAL BEHAVIOR IN RANDOM MAGNETS

V. Jaccarino and A. R. King

Department of Physics, University of California, Santa Barbara, CA 93106, U.S.A.

Abstract. — Experiments in recent years of critical behavior in random magnets are reviewed. Attention is focused on the random exchange (REIM) and random field (RFIM) Ising model problems, as realized in dilute, insulator antiferromagnets. The static exponents of the $d = 3$ REIM universality class are now measured and agree with theory. REIM to RFIM crossover scaling has been shown to govern the static and dynamic critical behavior in all $d = 3$ systems ($\text{Fe}_2\text{Zn}_{1-x}\text{F}_2$, $\text{Mn}_2\text{Zn}_{1-x}\text{F}_2$, and $\text{Fe}_2\text{Mg}_{1-x}\text{Cl}_2$) studied, with a universal exponent $\phi = 1.42 \pm 0.03$, which satisfies the Aharony inequality $\phi > \gamma$, the REIM susceptibility exponent. So extreme is the slowing down of the critical fluctuations in $d = 3$ REIM systems that it is at the roots of earlier misunderstandings of what was the lower critical dimension of the RFIM and may well limit the accuracy to which the static exponents of the $d = 3$ RFIM universality class may be obtained. Only qualitative models and limited Monte Carlo calculations exist with which to make comparison with experiment.

Introduction

In the last three years, considerable progress has been made in the understanding of random magnetic systems, both from theoretical and experimental points of view. We confine ourselves to the static and dynamic critical behavior of random systems which exhibit long range order below the magnetic phase transition — in particular, the Random Exchange (REIM) and Random Field (RFIM) Ising Model systems [1].

In the main, the experimentally realizable REIM and RFIM systems all turn out to be randomly diluted, uniaxial antiferromagnets, in which a uniform external field $H$ applied colinearly with the spontaneous ordering direction, generates a random field $H_{\text{RF}}$, as was first predicted by Fishman and Aharony [2]. With dilution governing the degree of randomness in the exchange, and the strength of $H_{\text{RF}}$ a function of the dilution and $H$, one may externally control the region of reduced temperature in which crossover from pure Ising to REIM and the crossover from REIM ($H = 0$) to RFIM ($H \neq 0$) take place. Since the pure Ising, REIM and RFIM systems each represent different universality classes, the scaling properties associated with the crossover between any of the three is of special interest, as are the critical exponents and amplitudes.

Effects of concentration gradients

The experimental situation, with respect to the critical behavior of random magnets, has been clouded by uncertainties arising from the existence of concentration gradients in any two component system. For example, we will have recourse to discuss the prototype $d = 3$, REIM and RFIM system $\text{Fe}_2\text{Zn}_{1-x}\text{F}_2$. If the concentration $x$ varied in a macroscopic (nonrandom) manner in a given crystal, then the ordering temperature $T_N$ would be position dependent. Any thermodynamic measurement (e.g. magnetic specific heat $C_M$) will be difficult to interpret unless one knows in advance the concentration profile and how $T_N(x)$ varies with $x$.

A method has been developed to characterize the concentration gradient in an optically anisotropic, mixed crystal [3]. It utilizes the room temperature optical birefringence technique combined with laser scanning profiles of the crystal. The effects such inhomogeneities have on the critical behavior were explicitly determined by computer simulations of the anomaly in $C_M$ and the quasielastic neutron scattering line profile in the REIM system [4]. Examples of these are given in figures 1a and 1b. The derivative of the magnetic birefringence $d (\Delta n) / d T$ (which has been previously shown to be proportional to $C_M$ [5]) vs. $T$ is shown for the laser beam parallel and perpendicular to the gradient. An important conclusion from this study is that the peak in $C_M$ does not occur at the mean transition temperature $T_N$, if the amplitudes $A^\pm$ of the divergence of $C_M^+ = A^+ |t|^{-\alpha}$ are not equal above and below $T_N$ (i.e. $A^+ \neq A^-\)$. Not understanding this point has been the origin of much of the confusion in the interpretation of REIM and RFIM experiments.

REIM critical behaviour

$d = 3$. — Measurements now exist of the following REIM critical exponents: $\alpha (C_m)$, $\nu$ (the correlation length $\xi = \xi_\alpha |t|^{-\nu}$), $\gamma$ (staggered susceptibility $\chi = \chi_\beta |t|^{-\gamma}$), and $\beta$ (order parameter $M = M_0 |t|^{\beta}$) and corresponding amplitude ratios. Those that we judge to be the most accurate [6-8] are collected in table I. Examples used to determine $\gamma$ and $\beta$ in $\text{Fe}_2\text{Zn}_{1-x}\text{F}_2$ are shown in figures 2 and 3, respectively, in which particular attention was paid to using an extremely homogeneously random crystal. Theoretical predictions [9, 10] of the exponents for $d = 3$ REIM universality class are also given in table I, where it is seen the agreement with experiment in each case is quite good. Unfortunately, critical amplitude ratios have only been calculated in the “one-loop” approxi-
Fig. 1. — (a) (Above) Simulations of the critical behavior of \(d(\Delta n)/dT = A^2 |g|^{-\alpha}\) with REIM parameters, in the presence of linear concentration gradients. The amplitude and gradients used correspond to those observed in a sample of Fe\(_{0.8}\)Zn\(_{0.2}\)F\(_2\). The solid curve is for no gradient with \(T_N = 68.6\) K. The two curves with short and long dashes correspond to \(\Delta T_N = 0.2\) and 0.9 K, respectively. The arrows indicate the limits of these variations. The average transition temperatures \(T_N\) are equal to the \(T_N\) of the solid curve (b) (Below) The observed behavior of \(d(\Delta n)/dT vs. T\) in the critical region, with the laser beam parallel (open circles) and perpendicular (solid circles) to growth direction, for the Fe\(_{0.8}\)Zn\(_{0.2}\)F\(_2\) crystal. The solid and dashed lines are the respective simulations of figure 1a [4].

Fig. 2. — Log-log plot of reduced hyperfine field vs. reduced temperature, from Fe\(_{0.9}\)Zn\(_{0.1}\)F\(_2\) Mössbauer Effect studies. The line is a fit to the data of the form \(Bt^\beta (1 + at^\delta)\) so as to include corrections to scaling [8].

Table I. — Experimental values for the REIM critical exponents and amplitude ratios. Comparison with the best theoretical results are given, where available.

<table>
<thead>
<tr>
<th></th>
<th>Experiment</th>
<th>Theory [10]</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\alpha)</td>
<td>(-0.09 \pm 0.03) [6]</td>
<td>(-0.04)</td>
</tr>
<tr>
<td>(\nu)</td>
<td>(0.69 \pm 0.01) [7]</td>
<td>0.68</td>
</tr>
<tr>
<td>(\gamma)</td>
<td>(1.31 \pm 0.03) [7]</td>
<td>1.34</td>
</tr>
<tr>
<td>(\beta)</td>
<td>(0.350 \pm 0.009) [8]</td>
<td>0.349</td>
</tr>
<tr>
<td>(A^+ / A^-)</td>
<td>(1.7 \pm 0.1) [6]</td>
<td></td>
</tr>
<tr>
<td>(\xi_0^I / \xi_0^S)</td>
<td>(1.45 \pm 0.04) [7]</td>
<td></td>
</tr>
<tr>
<td>(X_0^S / X_0^S)</td>
<td>(2.8 \pm 0.2) [7]</td>
<td></td>
</tr>
</tbody>
</table>

mation [11] and, in the case of \(C_m\), the latter fails to give even the correct sign. Clearly, this is an area requiring more extensive theoretical consideration.

\(d = 2\). — Experimental studies have been made on the Rb\(_2\)Co\(_{1-x}\)Mg\(_1\)F\(_4\) REIM system. Some questions [12, 13] exist whether there is a new fixed point in the \(d = 2\) REIM problem or rather that logarithmic corrections should be applied to the pure \(d = 2\) Ising model. Within the rather limited experimental accuracy on these generally poorer quality crystals, all of the observed critical exponents [14, 15] are equal to the pure \(d = 2\) Ising values, namely: \(\alpha = 0, \gamma = 1.75, \nu = 1.0\) and \(\beta = 1/8\), as is the ratio \(R_a = \xi_0^S / (M_0 \xi_0^S)^2 = 0.05\), which, from the hypothesis of two scale-factor universality, is a universal quantity [16]. Whether the observed amplitude ratios \(\xi_0^I / \xi_0^S\) and \(X_0^S / X_0^S\) do or do not agree with their pure Ising counterparts depends upon the assumed form of the neutron quasi-elastic scattering function.

Dynamics; \(d = 3\). — Recent Mössbauer Effect [17] and neutron spin-echo (NSE) [18] studies of Fe\(_{x}\)Zn\(_{1-x}\)F\(_2\) for \(T > T_N\) show strikingly longer relaxation times for the critical fluctuations than are found in the pure Ising system. From the dynamic scaling form \(\Gamma = At^\nu, A(Fe_{0.46}Zn_{0.54}F_2) \approx 7\) \(\mu eV\) [18] is obtained whereas for pure FeF\(_2\) \(\Gamma \approx 4.5\) meV.
Crossover behavior

Two crossover behaviors have been studied experimentally in connection with the $d = 3$ REIM and RFIM systems: 1) pure Ising to REIM and 2) REIM to RFIM. In principle, there is a third – pure Ising to RFIM – but there is, as yet, no system in which it has been evidenced, although some have claimed otherwise; see discussion in [4].

Pure Ising to REIM. – It had been thought [19] the reduced crossover temperature $t_\text{cr}$ which separated the asymptotic Ising and REIM critical behavior regions would scale with the randomness $\Delta J$ in the exchange $J$ as $t_\text{cr} \sim (\Delta J/J)^{1/\alpha}$, with $\alpha = 0.11$, the pure Ising specific heat exponent. Since $\Delta J/J \sim x (1 - x)$ and $\alpha$ is so small, one would not expect the crossover to be manifest (i.e. $t_\text{cr} \geq 10^{-3}$) until $x$ was quite small (i.e. $x < 0.6$). Instead, there is overwhelming evidence that the crossover is virtually complete for $x < 0.9$. This conclusion follows from recent Mössbauer Effect studies [8] of the magnetization exponent $\beta$ but was already implicit in the early birefringence and neutron scattering measurements of the REIM critical exponents. It is now believed [20] the proportionality factor in the scaling relation $t_\text{cr} \sim [x (1 - x)]^{1/\alpha}$ may differ from unity by a much as $|x_c (1 - x_c)|^{1/\alpha}$ where $x_c \approx 0.9$.

REIM-RFIM. – One of the earliest [2] and most interesting predictions associated with the RFIM problem was that related to crossover behavior. Apart from a mean-field correction, the shift in the ordering temperature $T_c (H)$ should scale with the field as

$$\Delta T_c (H) = c H^{\phi/\gamma}$$

where $\phi$ is the crossover exponent and the strength of the random field $H_{\text{RF}}$ is proportional to $H$. Recently Aharony [21] has demonstrated that, at $d = 3$, $\phi \neq \gamma$, the susceptibility exponent of the REIM fixed point (as would be the case in nonrandom systems), but rather the inequality $1.05 \leq \phi/\gamma \leq 1.1$ obtains. Accurate birefringence [22], and capacitance [23] measurements on Fe$_{0.46}$Zn$_{0.54}$F$_2$ give $\phi = 1.42 \pm 0.03$. When combined with the value $\gamma = 1.31 \pm 0.03$ (see Tab. 1) this yields $\phi/\gamma \approx 1.08 \pm 0.05$. Likewise, in two separate Faraday rotation studies of Fe$_{0.7}$Mg$_{0.3}$Cl$_2$ it has been observed that $\phi = 1.41 \pm 0.05$ [24] and $\phi = 1.44 \pm 0.04$ [25]. In addition, in the Mn$_{x}$Zn$_{1-x}$F$_2$ system it has been found [26] that $\phi = 1.43 \pm 0.03$, independent of $x$ in the range $0.4 \leq x \leq 0.8$. Thus $\phi$ is universal in that it is system and dilution independent. This puts to rest a long standing controversy on the nature of the crossover, the origin of which is now traceable to misinterpretations of just where $T_c (H)$ occurs as a function of $H$ in crystals with unknown or uncharacerized concentration gradients (see Ref. [4]).

An interesting achievement of one crossover study was the first measurement of nonlinear susceptibility of a REIM system [25]. One expects [21] that $x_{nl} \sim |t|^\gamma'$, where $\gamma' = 2 - \alpha - 2\phi$. Since $\gamma' = -0.82 \pm 0.03$, $x_{nl}$ has a divergent behavior at $T_N$. This is seen in figure 4 where the linear $x_1$ and $x_{nl}$ of Fe$_{0.7}$Mn$_{0.3}$Cl$_2$ are displayed.

It should be remarked that the REIM to RFIM crossover manifests itself in both equilibrium and nonequilibrium behavior as well as the critical dynamics of the RFIM problem.

RFIM critical behavior

DYNAMICS. – By far the most exciting recent development in the RFIM problem has been the recognition of the extreme slowing down associated with the dynamic critical behavior. Villain [27] and Fisher [28] have proposed an activated dynamics model. In it the characteristic relaxation time $\tau$ of a critical fluctuation increases exponentially with the correlation length $\xi$ as the transition is approached, $\tau \sim \exp (\xi^\theta)$ , where the parameter $\theta$ is bounded by $1 \leq \theta \leq 2$, rather than the conventional power law divergence $\tau \sim \xi^\phi$. Because of this exponential slowing down, measurements on very different time scales shown only a weak variation of a rounding temperature $t^*$ where non-equilibrium effects first set in. So extreme is this that experiments normally thought of a “static” (e.g. several minutes per point) exhibit non-equilibrium dynamical effects near $T_c (H)$. Thus the dynamics is easy to see but difficult to measure quantitatively, since small changes must be measured over very wide frequency (or time) ranges. Conversely, the range of “static” RFIM behavior is severely limited by the dynamics, and may preclude accurate determination of RFIM exponents. For this reason we begin by discussing dynamics before statics.

Fisher has argued that the dominant slow fluctuations of scale $\xi$ will occur on exponentially long time scales: i.e. the dynamic scaling is on logarithmic time scales rather than with a single characteristic relaxation time, leading to scaling functions such as the one for the specific heat $C_m \sim |t|^{-\alpha} f (\ln \omega/\xi^\phi)$. At
the critical point, $C_m$ must be independent of $\xi$, and its peak value should scale as $[C_m]_p \sim |\ln \omega|^{\alpha/\nu_T}$. The apparent width of the transition should scale as $t^* \sim |\ln \omega|^{-1/\nu_T}$.

If $H_{RF}$ is small (as is the case in the experiments), the critical behavior near $T_c$ will also be affected by the crossover from the REIM fixed point at $H = 0$, $T = T_N$. Lengths are then measured in units of the crossover length $L_0 \sim \xi_{cr} \sim \xi_{cr}^{-\nu} \sim H_{RF}^{-2\nu/\phi}$ and times in units of $\tau_0 \sim L_0^{\phi}$, where $\nu$ and $\phi$ (the dynamic scaling exponent) correspond to zero random field (REIM) exponents.

EXPERIMENTS AC SUSCEPTIBILITY. – The real part of the uniform RFIM ac susceptibility $\chi'(\omega)$ is expected to have the same singular behavior as does the specific heat $C_m \sim |\omega|^{-\alpha}$. Since both $d(\Delta n)/dT$ [29] and $C_m$ [30] measurements yield a symmetric, logarithmic divergence ($\alpha = 0$) so should $\chi'(\omega)$. Indeed this is found, but measurements from 2 Hz to 1 kHz result in a much more rounded peak than $C_m$ or $d(\Delta n)/dT$ data on the same Fe$_{0.46}$Zn$_{0.54}$F$_2$ crystal. Moreover, the peak value $[\chi'(\omega)]_p$ decreases and $t^*$ increases gradually with increasing frequency [31]: See figure 5. $[\chi'(\omega)]_p$ and $t^*$ were first analyzed in terms of the classical (power-law) dynamic scaling, where $t^*(\omega) \sim \omega^{1/2\nu}$ and $[\chi'(\omega)]_p \sim \omega^{-\alpha/\nu_T}$; from which the unusual value of $\nu_T \sim $ 14 was found: (See Fig. 6). But the data are equally well fit by the “activated” scaling form [28], which leads to $t^*(\omega) \sim |\ln \omega|^{-1/\nu_T}$,

$$[\chi'(\omega)]_p \sim |\ln \omega|^{-1/\nu_T}.$$ 

At present one cannot determine which form better fits the data.

FARADAY ROTATION. – Faraday rotation (FR) was used to measure the uniform magnetization $M$ vs. $T$ in the RFIM system [32]. Studies were made of $(\delta M/\delta T)_H$ and $(\delta M/\delta H)_{T}$, between 0.2 and 1.9 Tesla, on a Fe$_{0.47}$Zn$_{0.53}$F$_2$ crystal. In the RFIM limit, it was shown that $(\delta M/\delta T)_H$ and $(\delta M/\delta H)_{T}$ diverge as $t^* \sim 1 \times 10^{-3}$ and the peak value of the FR, measured at the same field. A similar extrapolation using the activated scaling form agrees satisfactorily with the FR data, provided $\theta \sim 1$. Thus, even with a combined range in $\omega$ of 6 decades from the two experiments, one can not determine which form yields the better fit to the data.

BIREFRINGENCE. – An analysis of birefringence data, on the same sample used in the $\chi'(\omega)$ experiments, yields a value for $t^* \sim 1 \times 10^{-3}$, in agreement with the FR results [22]. In addition, the field scaling of $d(\Delta n)/dT \sim C_m$ has been studied. As a consequence of the REIM to RFIM crossover, the field dependence of the static scaling is introduced into the dynamic scaling [26]. The strong static $H^y$ dependence of the peak height and $H^{2\nu/\phi}$ dependence of $t^*$ swamp the weak log $H$ dynamic dependence of these quantities, and the entire shape of the peaks appears to be described by static scaling alone. The amplitude $H^y$ has been shown [32] to contain two terms, with $y = 0.13$ and 1.56. Experiments on Mn$_{0.55}$Zn$_{0.45}$F$_2$ [26] and Fe$_{0.6}$Zn$_{0.4}$F$_2$ [22] agree with the $y = 0.13$ expected in low $H$ and show, in addition, rounding of the transition by concentration gradients must also be considered [26]: (See Fig. 7). Higher-field data on Fe$_{0.6}$Zn$_{0.4}$F$_2$ [22] can be fit only when the expected $y = 1.56$ term is included.
Fig. 7. Peak amplitude of $d(\Delta n)/dT$ vs. the mean-field corrected shift in $T_c(H)$, at several values of $H \neq 0$ in $\text{Mn}_{0.55}\text{Zn}_{0.45}\text{F}_2$. The upper curve is the expected weak random field $H^{0.12}$ scaling in the absence of gradient rounding. The lower curve is the result of a calculation that includes the measured gradient induced rounding $\delta_f \text{N}$ observed at $H = 0$. Inset shows the complete $d(\Delta n)/dT$ data at $H = 4.9, 9.8, 14.7$, and $19$ kOe (indicated by the arrows in main figure) in the RFIM region. The solid lines are the calculated behavior of $d(\Delta n)/dT$ vs. $T$ at each value of $H$ and include static and dynamic scaling effects as well as those arising from the gradient induced rounding [26].

**NEUTRON SCATTERING.** Extreme critical slowing in the order-parameter fluctuations has been observed using quasielastic neutron scattering in the same crystal of $\text{Fe}_{0.46}\text{Zn}_{0.54}\text{F}_2$ [33]. Unusual behavior is seen in the temperature dependence of the intensity $I(q)$ at the antiferromagnetic Bragg peak position, $I(1, 0, 0)$. A peak in $I(q)$ occurs near $T_c(H)$, for $H = 1.5, 1.9$, and $3.0$ T, whose width $W(H)$ scales as $H^{2/\phi}$, as would be expected from RFIM dynamical crossover scaling, (as do $T_c(H)$ and $T_{eq}(H)$), as shown in figure 8.

More direct evidence for the extreme critical slowing down of the order-parameter fluctuations is seen in two ZFC experiments illustrated in figure 9. In the inset, the height of the peak in $I(1, 0, 0)$ near $T_c(H)$ is found to be greater when measurements are taken more slowly. In the main figure, the sample was cooled in $H = 0$ to the temperature indicated, then $H$ raised to the designated value as quickly as possible, and then counting begun. In all cases, a nearly logarithmic increase in counting rate was observed over a $30 - 10^6$ sec time domain [33].

**STATICS.** From the birefringence studies it is known that $\alpha = 0.00 \pm 0.03$, $A/A' \simeq 1$ [29]. Except for the preliminary results of an earlier study of the $\text{Fe}_{0.46}\text{Zn}_{0.54}\text{F}_2$ crystal [34] there has been no attempt to accurately determine the static $d = 3$ RFIM critical exponents $\nu$, $\gamma$, and $\beta$ associated with this new universality class. The exponents $\nu$ and $\gamma$ and the correlation function exponent $\eta$ must be obtained from an interpretation of the quasi-elastic neutron scattering $I(q)$. However, no rigorous theory exists for $I(q)$ in the RFIM problem. Instead it has generally been assumed that $I(q)$ can be described by a Lorentzian (L) plus Lorentzian-squared (LSQ) form, on the basis of mean field arguments. The measurements of reference [34] assumed this to be the case and it was determined that $\nu = 1.0 \pm 0.15$, $\gamma = 1.75 \pm 0.20$ and $\eta \approx 1/4$. Recently it has been found that, close to $T_c(H)$, $I(q)$ cannot be fit by the L + LSQ form [33]. Since the values of $\nu$, $\gamma$, and $\eta$ are sensitive to the assumed form of $I(q)$, considerable uncertainty in these quantities will remain until new theoretical insight is obtained.

Other than Monte Carlo (MC) simulations, no theoretical predictions of any of the RFIM exponents exist. From the MC results values of $\nu = 1.0 \pm 0.1$, $\gamma = 2.0 \pm 0.5$, $\eta = 0.5 \pm 0.01$ and $\tilde{\eta} = 0.9 \pm 0.1$ have been obtained [35]. The quantity $\tilde{\eta}$ is the disconnected staggered susceptibility exponent; $\chi^{\text{dis}} \sim \xi^{2-\tilde{\eta}}$. Only one
generally agreed upon relation exists and that is the Schwartz inequality; $2(\nu - 1) \geq \hat{\eta} \geq -1$ [35]. Various new RFIM scaling forms have been proposed [28], including the modified hyperscaling relation $(d - \theta)\nu = 2 - \alpha$ and $\theta = \nu - \hat{\eta}$ and $\beta = (1 + \hat{\eta}) / 2$. Exponents derived from the use of these new relations should not be regarded with the same certainty as those directly obtained from the MC simulation.

The (staggered) magnetization exponent $\beta$ is especially interesting, since it is predicted to change from $0.35$ (REIM) to possibly as low as $0.05$ (RFIM) [35]. A measurement by capacitance and dilation techniques [35] yields a bound $\beta \leq 1/8$, confirming a large change does occur.

Acknowledgments

We acknowledge important collaborations with D. P. Belanger, J. L. Cardy, I. B. Ferreira and C. A. Ramos. The work at UCSB has been supported by NSF Grants DMR80-17582 and DMR85-16786.

[18] Belanger, D. P., Farago, B., Jaccarino, V., King, A. R. and Mezei, F. (paper 3P C-3 this Conf.).