Recent Experiments in Ising Antiferromagnets with Quenched Randomness

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Sevcral areas of experimentation using $d = 2$ aiid $d = 3$ Isiiig antiferromagnets with quenched randomness are reviewed. Tlie randomly dilute $Fe_xZn_{1-x}F_2$ aiid $Rb_2Co_xMg_{1-x}F_4$ systems in zero field sliow excellciit random- exchange Ising bcliavior for two and three dimensions, respectively. Tlie same systems in a field are the bcst experimental examples of random-field Ising systems. Low-temperature domain wall dynamics have becn studied in tlie $Fe_xZn_{1-x}F_2$ system. Spin glass-likc bcliavior is observed in $Fe_xZn_{1-x}F_2$ near tlie percolation tliresliold, tliougli tlie system is not a canonical spin glass. Tlie canonical Ising spin glass model is realized in tlie $d = 3$ Ising antiferromagnet $Fe_xMn_{1-x}TiO_3$. Tliese examplcs are chosen to demonstrate tlie opportunity of studying various Ising modcls with quenched randomness using insulating antiferromagnets.

I. Introduction

Iisulating antiferromagnetic crystals provide a rich testing ground for theories of phase transitions. Tliis is made possible by tlie universality of critical parameters cliaractcrizing phase transitions. Tlie only properties needed to describe tlie asymptotic bcliavior upon approach to a phase transition are the basic symmetrics of the system, for example tlie spatial dimension or the lattice geometry. Hence, theorists may study tlie simplest Hamiltonians having only tlic necessary symmetries aiid experimentalists need only to study simple, well understood magnetic systems witli tlie corresponding symmetries. Insulating antiferromagnets are often ideal in tliis regard. Tlie interactions between tlie magnetic spins decrease extremely rapidly with distance. Therefore, only a very few (often onc or two) interactions characterize tlie beliavior of many insulating magnetic crystals extraordinarily well. This simplicity lias helped both theorists and experimentalists make tremendous progress on pure systems and those with quenched disorder. Tlie subject of tliis review is tlie bcliavior of Is ng systerns witli quenched disorder wliicli can be studied in insulating antiferromagnet by growing crystals from a mixture of two isomorphic substances. For example, a mixed antiferromagnetic and diamagnetic crystal results in a system in which each possible magnetic site is randomly occupied or not. Aside from tlie quenched randomness, sucli a system may still prove quite simple in its microscopic description, with only a very few interactions needed. Tlie rich macroscopic behavior of such simple systems is surprising and some aspects of the transitions are still not well understood. Experiments in non-equilibrium states at low temperatures also are niost readily studied in dilute magnetic systems since tlic interactions are very simple and easy to model. Tliose aspects of tlie problems that are now understood can be applied to much more complicated substances by virtue of universality.

Tlie simplicity of tlie Hamiltonians needed to describe Ising transitions in randomly dilute antiferromagnets lends itself well to Monte Carlo siniulations. Such siiiiulations describe tlie observed beliavior extremely well. Tliis lias lead to better understanding of tlie theorics aiid experiments in many cases.

We will describe some experiments, tlieories and simulations relating to diluted and mixed antiferromagnetic crystals. We will hopefully give a flavor of some of tlie iiitcresting properties associated witli quenched randomness. Some of tlie topics of current interest in this field will be exemplified by the examples chosen for tliis review. For this particular discussion, we will largely lirnit ourselves to tlie simplest, best characterized and most studied crystuls. We will add comments about related systems when they add to the understanding of tlic models. Other systems have been discussed in previous reviews[1-8].

We begin by briefly reviewing tlie critical behavior of FeF_2, wliicli is an excellent example of a pure three-dimensional ($d = 3$) Ising model system, and K_2CoF_4 aiid Rb_2CoF_4, wliicli are excellent examples of pure $d = 2$ Ising model systems. Tliis will lielp to place the experiinents in tlie random systems into context.

We tlien discuss tlie random-exchange Ising model (REIM), wliicli lias becn most accurately studied for $d = 3$ in tlie magnetically dilute $Fe_xZn_{1-x}F_2$ system well above the percolation thresliold $x_p = 0.24$ witli no applied magnetic field. Tlie experimental critical beliavior measured in this system, both static and dynamic, will be compared witli theoretical results for critical exponents and amplitude ratios. Tlie evolution of metastable domains at low temperaturcs in tlie $Fe_xZn_{1-x}F_2$ system in zero field will be discussed as an example of the dynamics of ordering well below T_N. For d = 2, the best studied Ising case is $Rb_2Co_xMg_{1-x}F_4$. Again, excellent comparisons can be made betwcen tlieory and experiment.

We will briefly discuss tlie random-field Isiiig model (RFIM), which can be gcncratcd by applying a magnetic ficld to tlie dilute antiferromagnet $Fe_xZn_{1-x}F_2$ system for d = 3 and $Rb_2Co_xMg_{1-x}F_4$ for $d = 2$. Evcn though many problems have bccn resolved, tlicre are fundamental open questions about the $d = 3$ RFIM tramition after years of intense experimental aiid theoretical study.

We will discuss tlie beliavior of thc threedimensional Ising model close to tlie percolation threshold, primarily using the rcsults from the Ising system $Fe_xZn_{1-x}F_2$ system. Upon approach to tlie percolation tliresliold in tlie Ising system, spin glass-like beliavior becomcs dominant. This is surprising since, although tlie systcm possesses randomness, it apparcntly lacks tlie othcr essential ingrcdicnt of true spin glasscs, namely frustration.

A particularly clear and simple Ising spin glass system is tlie mixed antifcrromagnet $Fe_xMn_{1-x}TiO_3$. In tliis insulator botli frustration and randomness are present in tlie sliort-range interactions and much of tlie prcdicted spin glass behavior[8,9] has been clearly observed.

II. Tlie Pure Ising Model

Tlie Ising niodel is one of the simplest systems exhibiting a phase transition, witli eacli spin having only two possible states. Tlie simplicity of tlie system on a microscopic scale belies tlie intricate and rich behavior on the macroscopic scale, especially witli qucnclicd site randomness. A simple pure Ising antiferromagnetic model Hamiltonian is

$$\mathcal{H} = \sum_{<i,j>} J S_i S_j \ , \tag{1}$$

where there is only one essential iiiteraction betwecn neigliboring spins of strength $J > 0$ and $S_i = \pm 1$. In a real system universality dictates tliat tlie asymptotic critical bchavior will be described by tlie simple Hamiltonian above as long as the interactions are sliort-range. For weaker anisotropy, longer-range interactions, or multiple interactions, one must simply mcasure the behavior closer to tlie transition, i.e. the system will eventually cross over to the correct asymptotic behavior.

Tlie pure FeF_2 system lias becn shown to be an excellent d = 3 Ising system. Tlie critical exponent α and amplitude ratio A^+/A^- for tlie specific hcat

$$C_p = A^{\pm}|t|^{-\alpha} + B \tag{2}$$

from birefringcncc aiid pulsed specific licat tcchniques[10] agrce preciscly witli tlic results of many theoretical tecliniques. Tlic correlation length for fluctuations

$$\xi = \xi_0^{\pm}|t|^{-\nu} \tag{3}$$

and tlie staggered susceptibility

$$\chi_s = \chi_0^{\pm}|t|^{-\gamma} \ , \tag{4}$$

have been obtained from neutron scattering experiments[11], tlic staggcred magnetization

$$M_s = M_0|t|^{\beta} \ , \tag{5}$$

wliicli is only nonzero for $t < 0$, from Mössbauer experiments[12], and tlic dynamic critical beliavior for tlie relaxation timcs

$$\tau \sim \xi^z \ . \tag{6}$$

from spin-eclio neutron scattering tcchniques[13]. All of tlie critical behavior measured ovcr tlie critical range $|t| < 0.02$ in pure FeF_2 is in supcrb agrccment with theory[14–18]. Tlie obscrvation of asymptotic behavior over sucli a largc range of $|t|$ is a result of tlic strong single-ion anisotropy aiid tlie simplicity of tlie magnetic interactions in pure FeF_2. Tlie magnetic ions form a tetragonal body-centered lattice and tlie excliangc interaction betwccn tlie body center and corncr ions is tlie only significant one[19].

Tlie critical paramctcrs measurcd for tlie purc Ising model using tlie FeF_2 system are summarized in Table I along witli relevant theoretical rcsults. It is important to realize tliat the universal ratios of tlie amplitudes are just as important in cliaracterizing tlie critical behavior as tlic cxponcnts. Tlic rcsults demonstrate tliat FeF_2 is an exemplary Ising system and, upon dilution, an ideal one to study the effects of random-excliangc aiid random-fields for d = 3.

Tlie pure d = 2 Ising transition is very well reprcscnted by tlie isomorphic antiferromagncts K_2CoF_4 and Rb_2CoF_4. Tlie interactions betwecn the planes of magnetic C_0^{2+} ions are extremely small coinpared to tlic interactions within tlie planes. Tlie specific heat behavior lias been measured[20] using birefringence tecliniqucs in Rb_2CoF_4. From tlie Onsager solution[21] of tlie $d = 2$ Ising model, we expect tlie specific licat to be consistciit with tlie asymptotic logarithmic form

$$C_p = A\ln(|t|) + B \ . \tag{7}$$

The data indeed agree well witli this form. Thc critical behavior of ξ, χ_s and M_s have been obtained using ncutron scattering tcchniques[22]. Tlic static critical be- pavior cxponents and amplitudc ratios arc consistcnt with thcory[21,23]. Iii addition, tlic two-scale univcrsal ratio[24]

$$R_s = \chi_0^+ \kappa_0^{+\ d} / M_0 \ , \tag{8}$$

lias bccn dcmonstratcd[22,25] in thcsc compounds. Thc rcsults of cxpcrimcnts oii thcsc systcms and compar- isons to thcory are summarizcd iii Tablc I. Thcsc sys- tcms, whcn di utcd, arc obvioiis clioiccs for investigat- ing random-cxchangc aiid random-ficld cffccts in two dimcnsions.

Tablc I: Expcrimcntal aiid Thcorctical Purc Isiiig Crit- ical Paramctcrs. Thc dcfinitions for tlic cxponcnts aiid amplitudcs can bc found iii Eq. (2 -8). 1111 $d = 3$ ex- pcrimcntal pa-amctcrs are obtaincd from FeF_2. Tlie paramctcrs foi $d = 2$ wcre obtaincd using Rb_2CoF_4 or K_2CoF_4. Supcrscripts + or - rcfcr to tliosc obtaincd for ν and γ using only data for 1 > 0 and 1 < 0, rcspec- tivcly.

$d = 2$ PURE ISING

	Experiment	Theory
α	0.00 ± 0.01^a	$0(\log\|t\|)^b$
A^+/A^-	1.01 ± 0.00^a	$1(\log\|t\|)^b$
ν	$1.02 \pm 0.05^{+c}$	1^b
	1.12 ± 1.13^{-c}	
κ_0^+/κ_0^-	0.54 ± 0.06^c	$1/2^d$
γ	$1.82 \pm 0.07^{+c}$	$7/4^e$
	1.92 ± 0.20^{-c}	
χ_0^+/χ_0^-	32.6 ± 3.7^c	37.33^d
β	0.155 ± 0.02^c	$1/8^b$
R_s	$0.0565^c, 0.043^e$	0.051^f

$d = 3$ PURE ISING

	Experiment	Theory
α	0.11 ± 0.005^g	0.11 ± 0.003^h
A^+/A^-	0.54 ± 0.025^i	0.55^h
ν	0.64 ± 0.01^j	0.630 ± 0.001
κ_0^+/κ_0^-	0.53 ± 0.01^j	0.52^i
γ	1.25 ± 0.02^j	1.240 ± 0.001^h
χ_0^+/χ_0^-	4.6 ± 0.2^j	4.8^i
β	0.325 ± 0.005^k	0.325 ± 0.001^h
Z	2.1 ± 0.1^l	1.05 ± 0.07^m

a)ref.[20]; b)ref.[21]; c)ref.[22]; d)ref.[23,17]; e)ref.[25]; f)ref.[24]; g)ref.[10]; h)ref.[14]; i)ref.[15]; j)ref.[11]; k)ref.[12]; l)ref.[13]; m)ref.[18].

An important point should bc made concerning tlie mcasurcment of tlie staggcrcd magnetization with ncu- tron scatteriiig tcchniqucs. Iii tlie $d = 3$ case, tlie sam- plcs arc typically of such high crystalline quality that those ncutrons aligned well enough to Bragg scatter do so iii tlie first few microns of tlie crystal. This results in a saturation cffcct in tlic obscrvcd scattcring intcnsity, an effect known as extinction. Rccently, neutron Bragg scattcring in cpitaxial thin films lias bccii succcssful in showing thc propcr bchavior[26] of tlic staggcred magne- tization iii pure and dilutc Isiiig antifcrromagnets. For tlie case of $d = 2$ magnetic systems, cxtinction is not a problem bccausc tlic order paramctcr scattcring takes place along onc-dimcnsional rods iii reciprocal space aiitl iiot at points as is tlic case for $d = 3$. 11s a rcsult, tlic scatteriiig is iiot saturatcd iii bulk samples aiid tlie propcr critical bchavior is observcd.

111. Tlic Random-Exchange Isiiig Model

A siniplc random-cxchange Ising model Hamilto- nian for a single iiitcractioii site-diluted antiferromag- nct is

$$\mathcal{H} = \sum_{<i,j>} J\epsilon_i\epsilon_j S_i S_j \ , \tag{9}$$

where ϵ_i is iiiiity if tlic site is occupied aiid zero oth- crwise. No cxternal field is applied in this model aiid there are no frustrated iiitcractioiis. This model can be realized to high accuracy for $d = 3$ in tlic $Fe_xZn_{1-x}F_2$ antiferromagnet[27]. Similarly, tlic corresponding two- dimensional REIM is extremely well represented by the magnetically dilute antiferromagnet $Rb_2Co_xMg_{1-x}F_4$.

Tlic most basic effect of dilution on an unfrustrated Isiiig system is the lowering of tlic traiisitioii tempera- ture $T_N(x)$. There is a limit, tlie percolation threshold at concentration x_p, below which no traiisitioii can take place since tlie magnetic spins cannot form a long-range network. For concentrations well above x_p, it is found from both CPA theory[28] and experiments[29] in tlie $d = 3$ Ising system $Fe_xZn_{1-x}F_2$ tliat $T_n(x)/T_N(1) \approx x$. For tlie $d = 2$ system $Rb_2Co_xMg_{1-x}F_4$ an approxi- mately linear decrease is observed above tlic percola- tioii threshold[30]. Tlie traiisitioii temperature inevitably drops precipitously near tlic percolation tlircsliold for both $d = 2$ aiid $d = 3$.

Tlic specific heat critical beliavior in a $d = 3$ REIM system is fundamentally different from tlie pure case. As first pointed out by Harris[31], tlie specific heat can- not be positive for a system with quenched dilution if tlie hyperscaling relation $2 - \nu d = a$ holds, as it does in tlie REIM. (It does not hold for tlie RFIM.) Iii tlie $d = 3$ Isiiig case this implies a crossover from tlic pure tran- sition for wliicli a > 0. Tlie greater tlic dilution, tlie larger tlie region of reduced temperature over which tlie REIM behavior is observed. Contrary to early theoretical expectations[32], tlie crossover occurs quite rapidly[33,34]. The new, REIM specific heat critical be- havior lias been measured[35] for $Fe_{0.6}Zn_{0.4}F_2$ and more recently[36] for $Fe_{0.85}Zn_{0.15}F_2$, using tlie optical bire- fringence technique[37]. Tlie latter case is shown in Fig. 1. Tlie most recent preliminary result $\alpha = -0.9 \pm 0.02$, using Eq. 2 agrees very well with theory[38,39] aiid tlie previous experiment. The most recent amplitude ratio, $A^+/A^- = 1.55 \pm 0.15$ agrees well with the previous result but does not agree with theories[40,41] which yield $A^+/A^- < 0$. The predicted behavior is unusual. The specific heat would appear as an inflection rather than a cusp. The experimental results are completely incompatible with such behavior for reduced temperatures as small as $|t| = 10^{-4}$. This most likely reflects an inadequacy of the theory and remains an outstanding tlieoretical problem.

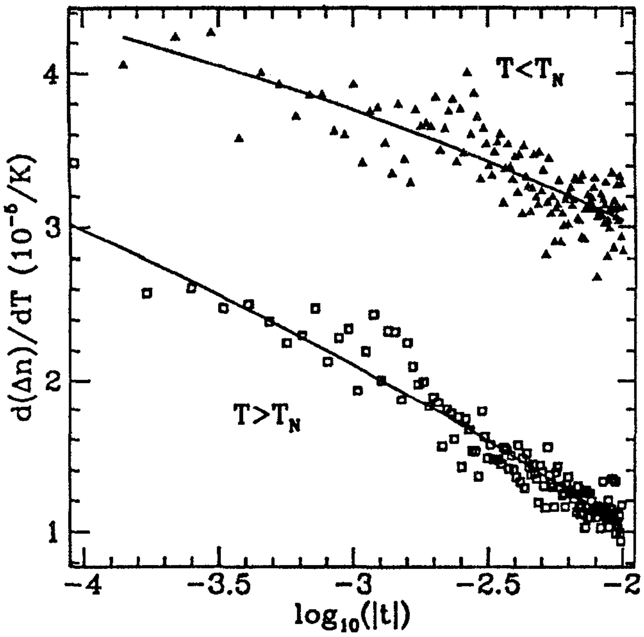

Figure 1: The temperature derivative of the optical birefringence $d(\Delta n)/dT$, which is proportional to tlie magnetic specific heat[37], vs. T for the $d = 3$ Ising antiferromagnet $Fe_{0.85}Zn_{0.15}F_2$. The solid curves indicate a fit to several sets of data using the power law behavior in Eq. 2 yielding the parameters given in Table II. The data indicate a cusp and not the inflection suggested by theory[41].(Wang and Belanger[36]).

Quasielastic neutron scattering studies[42] of $Fe_{0.46}Zn_{0.54}F_2$ have yielded the critical parameters for ξ, and χ using Lorentzian Iine shapes, which work well despite predictions for non-Lorentzian contributions[43]. Mossbauer experiments[33,44] have been used to determine the exponent for M,. Spin-echo neutron scattering techniques[13] have shown that the critical dynamics of $Fe_{0.46}Zn_{0.54}F_2$ are well approximated by conventional theory[18], but with relaxation times two orders of magnitude longer than for pure FeF_2. All of the measured critical parameters and their calculated values[45-47] for the d = 3 REIM are listed in Table II.

The d = 2 specific heat, best exemplified by measurements using the optical birefringence technique[48] in tlie $Rb_2Co_{0.85}Mg_{0.15}F_2$ system, is a marginal case with respect to the Harris criterion, since in the pure system α is zero (logarithmic divergence). Theories[45,46] for the random-exchange behavior predict tlie asymptotic behavior

$$C_p \sim \log(\log(|t|)) \qquad (10)$$

The experimental data are well dcscribed by a logarithmic divergence, as sliown in Fig. 2. It would be incredibly difficult in experiments to distinguisli bctween fits of tlie data to tlie doublc logaritlım in Eq. 10 and fits to tlie siiigle logarithin of Eq. 7. Hence, tlie experiments are consistent with tlie theories, tliough perhaps not a definitive test of them. In the two-dimensional system $Rb_2Co_xMg_{1-x}F_4$, neutron scattcring measurements[49] provide the critical beliavior of ξ, χ and M,. In addition, a two-scale universality analysis yields a value close to that obtained in tlie pure material. Tlie experimental and tlieoretical results are summarized in Table II.

Table II: Experimental and Theoretical Random-Exchange Ising Critical Parameters. Tlie definitions for tlie exponents and amplitudes can be found in Eq. (2-8). All d = 3 cxpcrimental paramcters are obtaincd from $Fe_xZn_{1-x}F_2$. The parameters for d = 2 were obtained using $Rb_2Co_xMg_{1-x}F_4$. Superscripts + or - refer to those obtained for ν and γ using only data for $t > 0$ and $t < 0$, respectively.

$d = 2$ RANDOM-EXCHANGE ISING

	Experiment	Theory
α	$\approx O(\log\|t\|)^a$	$O(\log(\log\|t\|))^b$
A^+/A^-	0.05 ± 0.10''	$1(\log(\log\|t\|))^b$
ν	$1.08 \pm 0.06^{+c}$	$1^d, 1002^e$
	1.58 ± 0.52^{-c}	
κ_0^+/κ_0^-	0.98 ± 0.02'	
γ	$1.75 \pm 0.07^{+c}$	1.753^e
	2.6 ± 0.6^{-c}	
χ_0^+/χ_0^-	19.1 ± 5.0^c	-
β	0.13 ± 0.02'	-
R_s	0.062 ± 0.01'	-

d = 3 RANDOM-EXCHANGE ISING

	Experiment	Theory
α	-0.09 ± 0.02^f	$-0.099, -0.03^h - 0.01^e$
A^+/A^-	1.55 ± 0.15^f	-0.5'
ν	0.69 ± 0.01^j	$0.70^g, 0.68^h, 0.67^e$
κ_0^+/κ_0^-	0.69 ± 0.02^j	0.83'
γ	1.31 ± 0.03^j	$1.39^g, 1.34^h, 1.32^e$
χ_0^+/χ_0^-	2.8 ± 0.2^j	1.7^i
β	0.35 ± 0.01^k	$0.349 \pm 0.002^h, 0.348^e$
Z	1.7 ± 0.2'	2.3^m

a)ref.[48]; b)ref.[45,46]; c)ref.[49]; d) pure value; e)ref.[47]; f)ref.[36]; g)ref.[38]; h)ref.[39]; i)ref.[40]; j)ref.[42]; k)ref.[33,44]; l)ref.[13]; m)ref.[18].

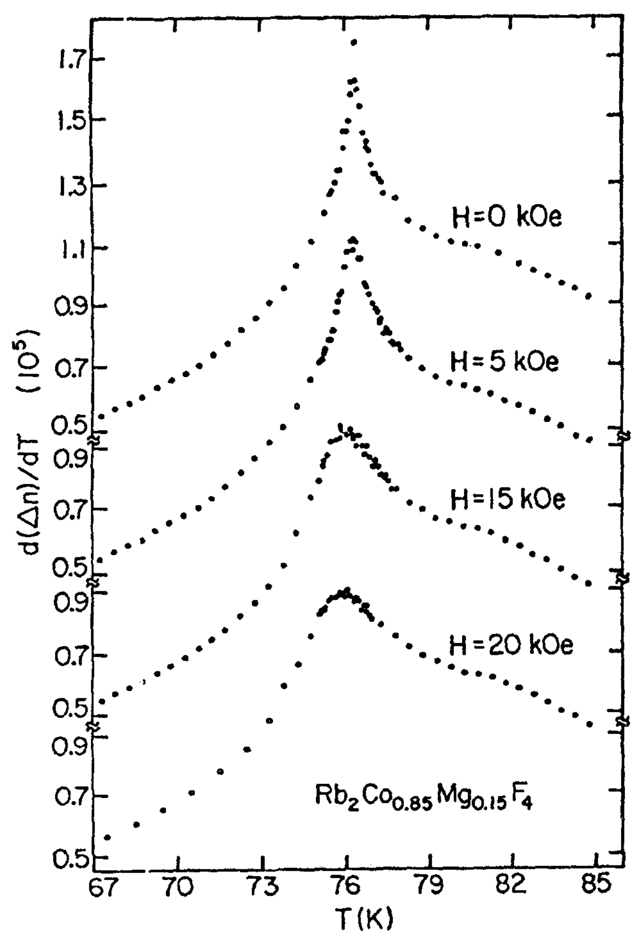

Figure 2: $d(\Delta n)/dT$ vs. T for tlie d = 2 Ising antiferromagnet $Rb_2Co_{0.85}Mg_{0.15}F_4$ in applied fields I' = 0, 0.5, 1.5, and 2.0T. The data sets are offset for clarity. Tlie H = 0 peak is approximated by a symmetric logaritlimic divergence. Tlie rounding of tlie peak increases with H and ɔccurs in equilibrium, indicating that the transition is destroyed by tlie random-field. (Ferreira, et al.[48]).

IV. Tlie Random-Field Ising Model

A randorr-field Ising ferromagnet may be represented most simply by a Hamiltonian such as

$$\mathcal{H} = -\sum_{<i,j>} JS_iS_j - \sum_i h_iS_i, \qquad (11)$$

where the field satisfies the conditions $[h_i]_{av} = 0$ and $[h_i^2]_{av} = h_r^2$, with $[\ ]_{av}$ signifying an average over tlie quenched disorder. It is quite difficult to realize such a Hamiltonian directly in a real magnet with a controllable random-field strength. Instead, the RFIRI is studied in dilute Ising antiferromagnets with a uniform field applied in the spin ordering direction. A simple Hamiltonian corresponding to an antiferromagnet with random field is

$$\mathcal{H} = \sum_{<i,j>} J_{ij}\epsilon_i\epsilon_j S_iS_j - \sum_i H\epsilon_i S_i\ , \qquad (12)$$

where H is a uniform field. The analogy of tlie two systems was first pointed out by Fishman and Aharony[32]. In the original random-field ferromagnetic model[50], tlie local fluctuations in the net random field compete with tlie long-range ferromagnetic order. In the dilute antiferromagnet, the local fiuctuations in tlie sublattice populations tends locaily to favor one sublattice over the other to point along tlie uniform field, in direct competition with tlie antiferromagnetic long-range order. This analogy was taken further by Cardy[51], wlio sliowed that tlie static critical behavior of a ferromagnet in a random field is ideiitical to tliat of a dilute antiferromagnct in a uniform field. Tlic random and uniform fields are related by

$$h_r^2 = \frac{x(1-x)[T^{MF}/T]^2(g\mu_B SH/k_BT)^2}{[1+\Theta^{MF}(x)/T]^2}\ , \qquad (13)$$

where x is tlie concentration, T^{MF} is tlie pure system mean-field transition temperature and Θ^{MF} tlie Curie-Weiss susceptibility parameter.

Tlie early investigations of tlie RFIM were marked by a controversy over whether a phase transition actually took place for $d = 3$. Tliis was a particularly difficult theoretical question to answer, though some of tlie first experiments[52], using the birefringence technique to measure tlie specific heat critical behavior, yielded strong evidence for a transition. In contrast to tliese conclusions, poor sample quality and a poor appreciation for tlie strong liysteresis in tlie dilute antiferromagnets did result in some groups claiming that tlie transition was destroyed. Tlie rigorous theories of Imbrie[53] and Bricmont and Kupiainen[54] proved tlie validity of the conclusions of the early birefringence measurements.

Efforts now are being made to fully calculate[55] and measure the d = 3 RFIAI critical behavior. From neutron scattering measurements[56], the correlation length appears to diverge with an exponent v ≈ 1, and the exponent for ξ is approximately $\gamma = 1.75$. The specific heat appears to be close to a symmetric, logarithmic divergence[52]. Curiously, these values are all close to those of the pure d = 2 Ising model. Early theoretical works in fact predicted an effective dimensional reduction in tlie critical behavior[57–59]. However, it was supposed to be from three to one dimension. Since there is no transition in tlie d = 1 Ising model, tlie implication was that there would be no transition for the $d = 3$ RFIM, a result later shown to be incorrect. Dimensional reduction is no longer generally supported by theorists.

The one exponent which is still only roughly determined[60] is the staggered magnetization exponent,

β. This is primarily a result of tlie scvcre extinction of tlic neutron scattcring intensity encountered in tlie $d = 3$ crystals. Tlie measurement of this exponent is of crucial importance for testing tlie RFIAI critical bchavior tlicories.

Tlic crossovcr from tlic random-exchange to the random-field Ising behavior depends upon tlic strength of tlie random-field which in turn varics linearly with tlie strength of tlie applied field[34]. Tlic random-field scaling behavior dictates tliat tlie traiisition boundary in tlic $II - T$ phase diagram varies as

$$T_c(II) - T_N \approx II^{2/\phi} \tag{14}$$

where the crossover exponent $4 \approx 1.1\gamma$ witli γ being tlie random-exchange staggered susceptibility exponent[61]. This behavior has been verified[62] by experiments on $Fe_xZn_{1-x}F_2$, with tlic results $\gamma = 1.31 \pm 0.03$ aiid tlie value from many measurements[63] $\phi = 1.42 \pm 0.03$. Some of the carlier experiments leading to tlie conclusion tliat tlic pure value of γ were adversely affected by concentration gradients and tlic practice of taking tlic transition to be at tlic peak in tlie specific heat, as shown by simulations[64].

The dynamics of tlie d = 3 RFIAI have now been convincingly demonstrated, using ac susceptibility measurements[65,66], to be activated in $Fe_xZn_{1-x}F_2$ in agreement with theory[67,68]. Spin-echo neutron scattering measurements show[13] tliat, on tlie very sliort time scale of nanosccoiids, tlie system crosses over to random-field dynamics at much larger reduced temperatures and much lower applied fields than measurements which are conducted on time scales of seconds, such as specific heat or quasielastic neutron scattering. Extremely slow dynamics are manifest in two distinct ways near tlie transition. First, when cooling in a field (FC) or heating in a field after cooling in zero field (ZFC), equilibrium is lost iicar tlie traiisition. Upon FC, the system cannot achieve long-range order aiitl tlic data differ from tlic ZFC data below an equilibrium boundary $T_{eq}(II)$ which lies just above $T_c(II)$ aiitl scales iii precisely the same manner. Of coiirse, with a small enough random field and limited instrumental resolution, the FC procedure will appear to yield long-range order[69].

Closer to $T_c(II)$, tlic critical bcliavior is rounded by tlic critical slowing, as exemplified by tlic ZFC Faraday rotation data[7'] in Fig. 3. Shapiro[71,5] has proposed a tlicory based on tlie slow dynamics which predicts tlie observed critical exponents.

Tlic neutron scattering line shapes observed in tlie $d = 3$ RFIM systems are far from tlic Lorentzian forin which adequately describes tlie scattering iii pure FeF_2 aiid in $Fe_xZn_{1-x}F_2$ in zero field well above tlic percolation threshold. Mean-field tlicory predicts a squared Lorentzian line shape and it has been commonly assumed tliat tlie mean-field argument adequately explains tlic unusual observed line shapes. However, al-

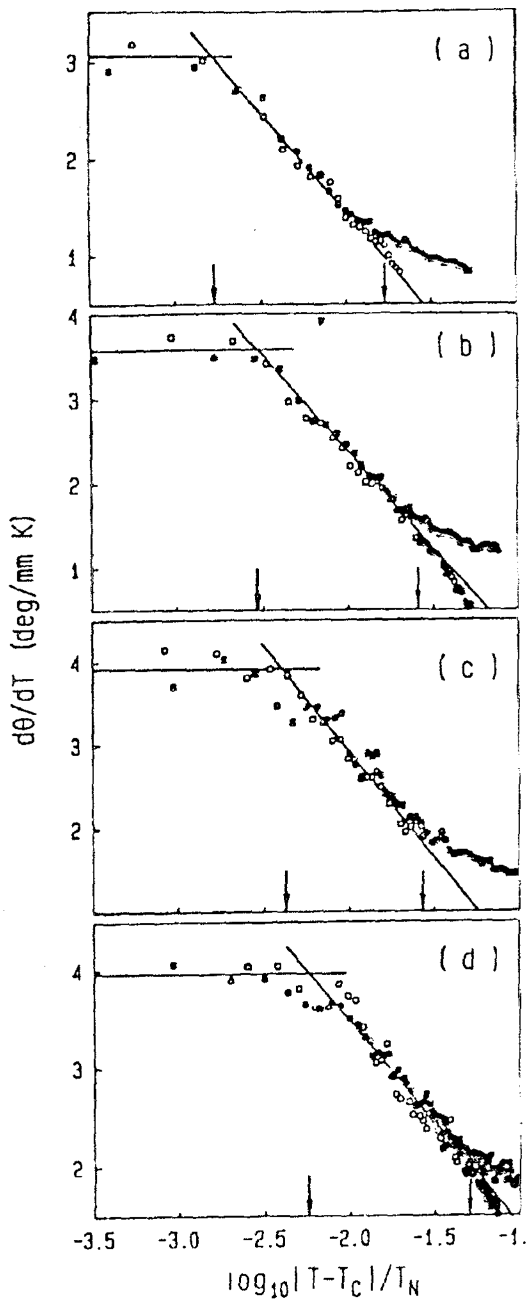

Figure 3: $d\theta/dT$, which is proportional to the magnetic specific heat[78], vs. $log_{10}(|t|)$ upon FC in tlic $d = 3$ Ising antiferromagnet $Fe_{0.47}Zn_{0.53}F_2$ for $\mathbf{II} = 2, 3, 4$ and$5T$ in (a), (b), (c), and (d), respectively. In each figure the filled symbols are for $T < T_c(II)$ and tlie open ones for $T > T_c(II)$. Tlic right arrow iiidicntes where tlie random-exchange to random-field crossover occurs. Tlic left arrow in cacli case indicates tlic onset of rounding from RF dynamics. Both tlic region of RF dynamics and tlic region of RFIM critical behavior increase in size witli increasing field. Data between tlie arrows in each case are seen to approximate a symmetric, logarithmic divergence. (Pollak, et. al.[70]).

though tlic squared Lorentzian works well for low temperature FC scans and scans above T_{eq} it does not work well[62] for ZFC scans below $T_c(II)$. Furthermore, non-Lorentzian liiie sliapcs are observed[72] at zero field in tlic nonequilibriuin region near tlie percolation threshold but not observed[73] in tlie $d = 2$ case for $II > 0$ near T_N where tlie destruction of tlie transition occurs in equilibrium. All of these observations suggest a nonequilibrium origin to tlie non-Lorentzian line shapes iicar $T_c(II)$ for $d = 3$.

For tlie $d = 2$ RFIM system $Rb_2Co_{0.85}Mg_{0.15}F_4$, tlic transition is observed to be destroyed[48], as shown in Fig. 2, iii agreement with theory[50]. No hysteresis or extremely slow dynamics are observed near tlie destroyed transition, indicating equilibrium beliavior. At low temperatures, however, metastable long-range order induced by ZFC decays only when tlie rounded traiisitioii is approached sufficiently closely. Tliis occurs closer to the transition as tlie measurement time scale decreases, as iiidicated by tlic comparison of high field pulsed magnetization experiments[74] aiid neutron scattering measurements[75] wliicli differ in tlie measurement time scale by eight orders of magnitude.

V. Metastable Domain Wall Dynamics

Random-exchange interactions not oiily affect tlie phase transitioii to long-range order in a profound way, but also tlie eqiiilibratioii dynamics for $T << T_N$. Tlie fact tliat tlic interactions in $Fe_xZn_{1-x}F_2$ are so well understood and are very simple makes tliis system ideal for investigating tlic dynamics of metastable domain walls at low temperature. Domain walls may be conveniently introduced into the system by FC. The length scale associated with tlic domains, as evident from tlie widths of neutron scattering liiie profiles[76], decreases as tlie strength of tlie applied field upon FC increases. Two distinct kinds of dynamics can be investigated. With a field applied tlic evolution of tlic doniains will be influenced by pinning from tlic random field as well as from vacancies. Oiice tlie field is removed, on tlie otlier hand, the oiily pinning remaining is from tlic vacancies. We shall discuss tlie latter case first.

At low enough temperatures, it lias been shown from neutron scattering[77] and Faraday rotation experiments[78,70] tliat tlie domain walls do not move macroscopically even after tlic field is removed. A common misconception is tliat tlic width of tlic neutron scattering profiles yields directly tlie size of metastable domains. Computer simulations[79] demonstrate, however, tliat tlie magnetic systein forms essentially only two domains tliat are incredibly intertwined, as shown in Fig. 4. At best, tlie non-Lorentzian width represeiits tlie typical distance needed to pass from one domain to tlie otlier. Nowak and Usadel[80] suggest tliat tlic domains are fractal iii structure. The line sliape lias been studied with Monte Carlo simulations[81] and has some unusual properties associated with tlic fractal-like structure.

Tlie fact tliat tlie domain structure does not evolve oii a macroscopic scale at low temperatures indicates tliat tlie thermal fluctuations are iiot strong enough to overcome tlie vacancy pinning. The walls evolve on a microscopic scale, however. Early experiments[70] showed a time dependence to tlie remanent magnetization after tlie field is turned off at low temperatures. Tlie remanent magnetization originates from tlie domain walls formed upon FC. Tlie domain walls have magnetization since, iipon cooling in a field, tlie spins on either side of tlic domain wall are parallel to tlic field to reduce tlic local random-field free energy. Once tlie field is removed, there is no energetic advantage to alignment with tlic field. Tlie domain walls then evolve in sucli a way as to locally minimize tlie exchange energy, i.e. tlic siirface area of tlie walls is reduced. Spins along tlic wall which were predominantly aligned with tlic field will flip as tlie wall translates by oiie lattice spacing. Tliis provides tlie mechanism by wliicli microscopic wall movements, driven by tlie exchange energy, reduce tlic net magnetization[82].

Nattermann and Villain[6,83] initiated tlie idea tliat iiiicroscopic wall movements aiid not macroscopic motion constituted the mechanism for tlic decay of tlie remanent magnetization and attempt to explain tlic original experimental results. Tliey proposed tlic decay

$$\Delta M = M_0 \ln(|t|)^{-\psi} + B \;, \qquad (15)$$

where $\psi \approx 0.4$ aiid B is a constant volume term wliicli is small, to describe tlie experimental results[70] for $Fe_{0.47}Zn_{0.53}F_2$. Later Monte Carlo simulation data obtained by Nowak and Usadel[84] usiiig a simple-cubic lattice showed better agreement with tlie power law behavior

$$M_s \sim |t|^{-x} \qquad (16)$$

aiid attributed tlic beliavior to a lack of a cliaracteristic length scale associated witli tlie fractal doniaiiis.

Further experimental studies[85] and Monte Carlo simulations[82] indicate aiiotlicr expression wliicli does an cxcclleiit job of describing tlie data for tlie body-centered tetragoiial lattice of dilute $Fe_{0.47}Zn_{0.53}F_2$ and a simulated body-cciitered cubic lattice. Tlie expression,

$$M_s = M_0 \exp[(-A \ln|t|)^{\psi}] \;, \qquad (17)$$

yields exceptionally good fits of tlie data witli y independent of tlie field aiid temperature. A suitable tlieory for tliis form is lacking.

Tlie dynamics associated witli domain wall pinning by random fields have been studied[**] by employing tlie FC procedure at temperatures not far below $T_C(II)$ and measuring tlie time dependence of tlie uniform magnetization in tlie presence of tlie field usiiig SQUID techniques. Apparently, at sucli temperatures tlie random-field pinning dominates over tlie vacancy pinning aiid many of the observations are consistent witli the theoretical models for low-temperature random-field activated dynamics[S7]. This implies that there must be many small activated domains as opposed to the fractal-like, two-dnmain, frozen structure existing at low temperature.

VI. Tlie Percolation Threshold in the Ising Model

The d = 3 Ising model system $Fe_xZn_{1-x}F_2$ near tlie percolation threshold provided some experimental surprises. Earlier experiments[88] on the weakly anisotropic, isomorphic antiferromagnet $Mn_xZn_{1-x}F_2$ were interpreted in terms of a geometric correlation lengtli, κ_G, aiid a thermal correlation lengtli. For all concentrations above the percolation threshold in the weakly anisotropic system, long-range order was observed and equilibrium behavior seemed to prevail for all temperatures. Such is not the case for tlie Ising system. In zcro field, even a few percent above the percolation tlireshold, long-range order does not develop. Instead, near tlie percolation threshold a spin glass-like pliase is encountered[7,89,90]. Yet tliis system cannot be considercd a canonical spin glass wliicli requires two ingredients, randomness and frustration. In zero field, there is little or no frustration[19] in $Fe_xZn_{1-x}F_2$. Nevertheless, in tlie H − T phase diagram there is a boundary wliicli is completely analogous to tlie spin glass de Almeida-Thouless (AT) boundary[91] including tlie scaling exponent $\phi \approx 3.4$. This was first observed in magnetization studies by Montenegro, et al. [89,90]. Even in zcro field no long-range order develops[72] for $x \leq 0.27$ and unusual line sliapes are evident for $T < 12K$. A bit further above tlie percolatioii threshold, at x = 0.31, both tlie usual random-field beliavior at low *11* and tlie spin glass-like beliavior at higher H exist[90,92], as shown in tlie H − T phase diagram in Fig. 5. Tlie random-field beliavior at low *11* is tlie same as that seen for all H at higher concentrations $x \geq 0.46$. At higher fields, tlie equilibrium boundary scaling with $\phi \approx 3.4$ appears. No long-range order persists in the region at high field. It has been shown[79] in Monte Carlo simulations, in which it is assured that there are no frustrating interactions, that a phase diagram very much like tliat seen in $Fe_xZn_{1-x}F_2$ emerges. It is interesting to note that the observation of such behavior may be indicative of quenclied randomness rather tlian simply a manifestation of a spin glass systeni, as is commonly assumed. The underlying physics causing the spin glass-like behavior in $Fe_xZn_{1-x}F_2$ must be related to tlie extremely slow dynamics associated with tlie Ising percolation threshold[93]. A complete understanding of the behavior in this system is important in itself and may shed light on the spin glass problem as well.

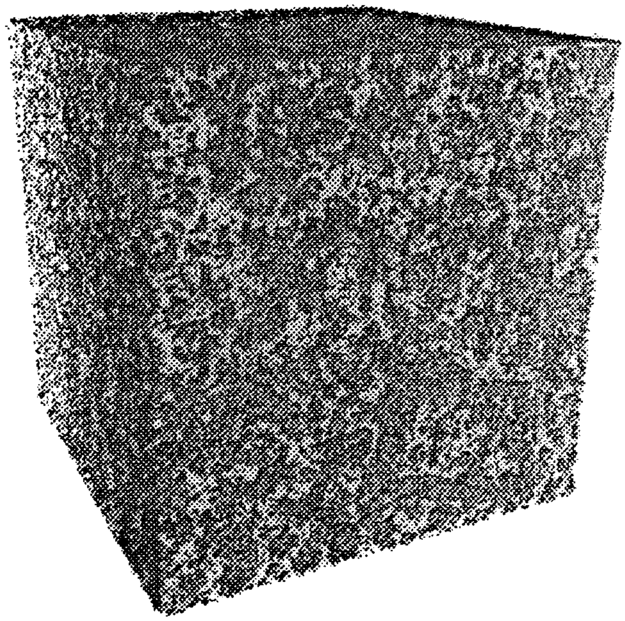

Figure 4: A computer simulation of a dilute (x = 0.5) antifcrromagnet cooled in a magnetic field to low temperature. Tlie lattice size is 99 x 99 x 98 and it was FC witli $H = 3J$. Tlie light and dark sliades represent spins bclonging to the two connected domains wliicli interweave in a fractal-like structure. (Nowak and Usadel [79]).

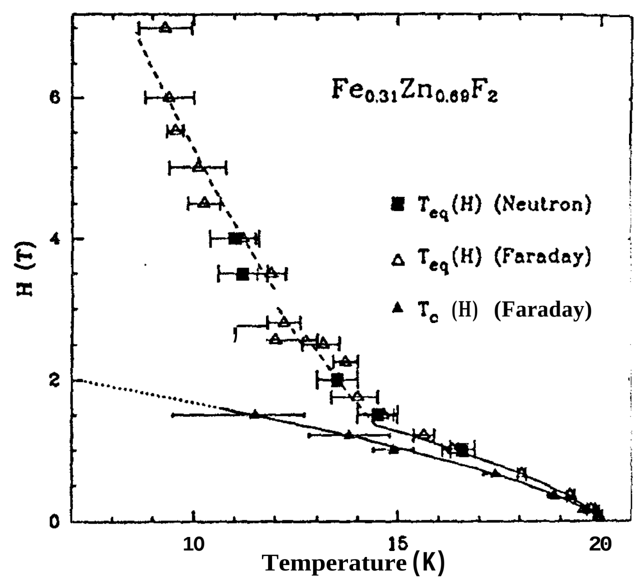

Figure 5: *11* vs. T phase diagram for $Fe_{0.31}Zn_{0.69}F_2$. For H < 1.5T, $T_{eq}(H)$ and $T_c(H)$ scale witli $\phi = 1.4$, tlie RFIM crossover esponent. For $H > 2T$, $T_{eq}(H)$ changes curvature and scales witli $\phi = 3.4$. Tlie region below $T_c(H)$ exhibits aiitiferromagnetic long-range order. The region below $T_{eq}(H)$ and above $T_c(H)$ is not antiferromagnetic. (Montenegro, et al.[92]).

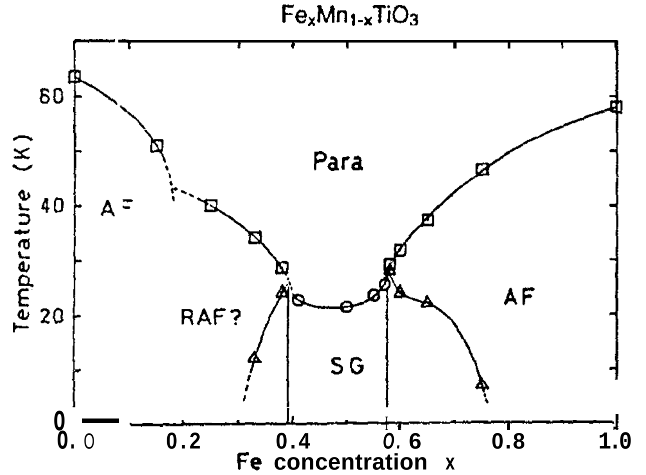

Figure 6: The $x - T$ phase diagram for the spin glass system $Fe_xMn_{1-x}TiO_3$. For concentrations $0.4 < x < 0.6$, a spin glass phase is encountered as T is lowered. Just to either side of this region, upon lowering T, a transition from paramagnetism to antiferromagnetism occurs and, at lower T, a mixed antiferromagnetic/spin glass region is entered. (Yoshizawa, et al.[96]).

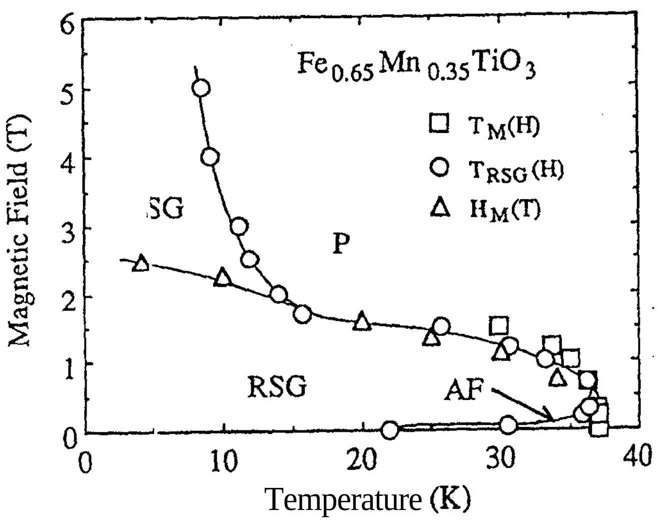

Figure 7: The $H - T$ phase diagram for the spin glass system $Fe_{0.65}Mn_{0.35}TiO_3$. At $H = 0$, the system undergoes a transition to antiferromagnetic long-range order and then, at lower T enters the mixed region. At high H, a spin glass region separates the mixed region from the paramagnetic region. There are significant similarities and differences between this spin glass phase diagram and the one for near the Ising percolation threshold shown in Fig. 5. (Yoshizawa, et al.[96]).

VII. Ising Spin Glass in Mixed Antiferromagnets

Finally, we discuss an example of an Ising spin glass system, $Fe_xMn_{1-x}TiO_3$, formed by the random mixing of two Ising antiferromagnets[94-96]. The pure systems $FeTiO_3$ and $MnTiO_3$ are both Ising antiferromagnets with spin alignments along the same crystalline axis but with very different spin arrangements in the ordered state. In the mixed system, each spin encounters a random environment of ferromagnetic and antiferromagnetic interactions with its neighbors. Hence, interactions in the mixed system possess both randomness and frustration, the two essential ingredients for a canonical spin glass. This is perhaps the finest example of an insulating antiferromagnetic Ising spin glass.

Mean-field theories for spin glasses yield a phase diagram in which the spin glass transition is encountered as the temperature decreases in the presence of a large amount of randomness. For the case of less randomness, the sample undergoes a transition to antiferromagnetic long-range order and then, at lower temperature, a mixed region is entered as the AT boundary is crossed. The mixed region corresponds to both long-range order and a spin glass order. Between the mixed region and the spin glass region is a vertical boundary. In the $Fe_xMn_{1-x}TiO_3$ system[94], the spin glass region occurs for concentrations between $x \approx 0.4$ and 0.6 as shown in the $x - T$ phase diagram in Fig. 6. This is the region of greatest randomness. Antiferromagnetic regions, with mixed regions at lower temperatures, exist to either side of the spin glass region, consistent with the mean-field diagram.

Elastic scattering measurements at the antiferromagnetic scattering point in sample at a concentration $x = 0.60$ in the antiferromagnetic/mixed region show an abrupt increase in intensity at the Bragg point upon lowering the temperature through the antiferromagnetic transition, as would be expected for a transition to antiferromagnetic long-range order. Then, upon crossing the AT line, the intensity sharply decreases. In the mixed region, the width of the Bragg scattering peak remains resolution limited and no sign of hysteresis is observed. This indicates that the observed behavior is the equilibrium behavior and, therefore, the long-range antiferromagnetic order decreases upon entering the mixed region. Concomitantly, the short-range antiferromagnetic order increases as the mixed phase is entered. The correlation length for antiferromagnetic fluctuations appears to diverge at the antiferromagnetic transition and shows another maximum at the crossing of the AT line. At low temperatures, the diffuse scattering profiles have a width reflecting a geometric disorder which appears to vanish at the boundary between the mixed and spin glass phases. Sharp excitations are supported[95] by the system only for $q < \kappa_G$.

The $H - T$ phase diagrams of this material for

x = 0.6, 0.65 and 0.75 have bccn measured[96]. An example is shown in Fig. 7. At low fields an antiferromagnetic region exists but a spiii glass region dominates nt higher field. Some aspccts of this phase diagram resemble that of $Fe_{0.31}Zn_{0.69}F_2$ shown in Fig. 5. There is a transition from paramagnetism to antiferromagnetism at low fields aiid at high fields there is an AT-like borindary between tlie paramagnetic and spiii glass regions. However there are significant differences. No clear reentrance region is defined in tlie $Fe_{0.31}Zn_{0.69}F_2$ system *as* there is in $Fe_{0.65}Mn_{0.35}TiO_3$. Tlic Bragg scattering shows no hysteresis in $Fe_{0.65}Mn_{0.35}Tio_3$, whereas it is evident in $Fe_{0.31}Zn_{0.69}F_2$. Lorentzian line shapes are adequate for $Fe_{0.65}Mn_{0.35}TiO_3$ for $H = 0$, but not for $Fe_{0.31}Zn_{0.69}F_2$ at low temperatures. The differences and similarities are interesting and need to be understood for a complete undcrstaiiding of both the spin glass aiid Isiiig percolation threshold models.

VIII. Concluding Remarks

We have seen several examples of models of randomness realized by simple insulating antiferromagnets. The experiments done on these aiid other antiferromagnets with quenched randomness have helped tremendously iii tlic understanding of tlie general role randomness plays in phase transitions and the behavior of ordered systems. Tlie simple nature of tlic interactions lias allowed theoretical treatments aiid computer simulations to elucidate tlie observed behavior. Experiments on $d = 3$ REIM systems confirmed tlie prediction of a crossover to a new universality class. Only minor problems still need to be solved to bring REIM experiments and tlicory iiito complete agreement. Strong evidence for tlie $d = 3$ RFIM transition was observed in some experiments long before theorists finally proved its existence. Theories, simulations and experiments are now directed at understanding tlic phase transitioii iii detail. Near tlie percolation threshold tlic observation of spiii glass-like behavior in Ising antiferromagnets has been corroborated by computer simulations and is indicated by some tlicory, but a complete understanding is still lackiiig. A better understanding of tlie common appearance of a AT line in canonical Isiiig spin glasses and iii dilute Ising antiferromagnets near the percolation threshold is needed. Tlie decay of tlie remanent magiictization in dilute Ising antiferromagnets for $T < T_N$ may lead to a better understanding of nonequilibrium processes if proper tlicories can be developed to explain tlic observed behavior. Tlie Isiiig spiii glass lias been well realized in a mised Ising antiferromagnet. Most of the predicted spin glass beliavior has been verified by the experiments. Antiferromagnets will surely continue to play a crucial role in future investigations of systems with quenched disorder.

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