Recent Experiments in Ising Antiferromagnets with Quenched Randomness

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Several areas of experimentation using $d = 2$ and $d = 3$ Ising antiferromagnets with quenched randomness are reviewed. The randomly dilute $Fe_2Zn_{1-x}F_2$ and $Rb_2Co_2Mg_{1-x}F_4$ systems in zero field show excellent random-exchange Ising behavior for two and three dimensions, respectively. The same systems in a field are the best experimental examples of random-field Ising systems. Low-temperature domain wall dynamics have been studied in the $Fe_2Zn_{1-x}F_2$ system. Spin glass-like behavior is observed in $Fe_2Zn_{1-x}F_2$ near the percolation threshold, though the system is not a canonical spin glass. The canonical Ising spin glass model is realized in the $d = 3$ Ising antiferromagnet $Fe_2Mn_{1-x}TiO_3$. These examples are chosen to demonstrate the opportunity of studying various Ising models with quenched randomness using insulating antiferromagnets.

I. Introduction

Ising antiferromagnetic crystals provide a rich testing ground for theories of phase transitions. This is made possible by the universality of critical parameters characterizing phase transitions. The only properties needed to describe the asymptotic behavior upon approach to a phase transition are the basic symmetries of the system, for example the spatial dimension or the lattice geometry. Hence, theorists may study the simplest Hamiltonians having only the necessary symmetries and experimentalists need only to study simple, well understood magnetic systems with the corresponding symmetries. Insulating antiferromagnets are often ideal in this regard. The interactions between the magnetic spins decrease extremely rapidly with distance. Therefore, only a very few (often one or two) interactions characterize the behavior of many insulating magnetic crystals extraordinarily well. This simplicity has helped both theorists and experimentalists make tremendous progress on pure systems and those with quenched disorder. The subject of this review is the behavior of Ising systems with quenched disorder which 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 the quenched randomness, such a system may still prove quite simple in its microscopic description, with only a very few interactions needed. The 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 most readily studied in dilute magnetic systems since the interactions are very simple and easy to model. Those aspects of the problems that are now understood can be applied to much more complicated substances by virtue of universality.

The simplicity of the Hamiltonians needed to describe Ising transitions in randomly dilute antiferromagnets lends itself well to Monte Carlo simulations. Such simulations describe the observed behavior extremely well. This leads to better understanding of the theories and experiments in many cases.

We will describe some experiments, theories and simulations relating to diluted and mixed antiferromagnetic crystals. We will hope to give a flavor of some of the interesting properties associated with quenched randomness. Some of the topics of current interest in this field will be exemplified by the examples chosen for this review. For this particular discussion, we will largely limit ourselves to the simplest, best characterized and most studied crystals. We will add comments about related systems when they add to the understanding of the models. Other systems have been discussed in previous reviews$^{1-8}$.

We begin by briefly reviewing the critical behavior of $FeF_2$, which is an excellent example of a pure threedimensional ($d = 3$) Ising model system, and $K_2CoF_4$ and $Rb_2CoF_4$, which are excellent examples of pure $d = 2$ Ising model systems. This will help to place the experiments in the random systems into context.

We then discuss the random-exchange Ising model (REIM), which has been most accurately studied for $d = 3$ in the magnetically dilute $Fe_2Zn_{1-x}F_2$ system
well above the percolation threshold \( x_p = 0.24 \) with no applied magnetic field. The experimental critical behavior measured in this system, both static and dynamic, will be compared with theoretical results for critical exponents and amplitude ratios. The evolution of metastable domains at low temperatures in the \( Fe_xZn_{1-x}F_2 \) 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 \( Rb_2Co_2Mg_{1-x}F_4 \). Again, excellent comparisons can be made between theory and experiment.

We will briefly discuss the random-field Ising model (RFIM), which can be generated by applying a magnetic field to the dilute antiferromagnet \( Fe_xZn_{1-x}F_2 \) system for \( d = 3 \) and \( Rb_2Co_2Mg_{1-x}F_4 \) for \( d = 2 \). Even though many problems have been resolved, there are fundamental open questions about the \( d = 3 \) RFIM transition after years of intense experimental and theoretical study.

We will discuss the behavior of the three-dimensional Ising model close to the percolation threshold, primarily using the results from the Ising system \( Fe_xZn_{1-x}F_2 \) system. Upon approach to the percolation threshold in the Ising system, spin glass-like behavior becomes dominant. This is surprising since, although the system possesses randomness, it apparently lacks the other essential ingredient of true spin glasses, namely frustration.

A particularly clear and simple Ising spin glass system is the mixed antiferromagnet \( Fe_xMn_{1-x}TiO_3 \). In this insulator both frustration and randomness are present in the short-range interactions and much of the predicted spin glass behavior has been clearly observed.

### II. The Pure Ising Model

The Ising model is one of the simplest systems exhibiting a phase transition, with each spin having only two possible states. The simplicity of the system on a microscopic scale belies the intricate and rich behavior on the macroscopic scale, especially with quenched site randomness. A simple pure Ising antiferromagnetic model Hamiltonian is

\[
\mathcal{H} = \sum_{\langle i,j \rangle} J S_i S_j ,
\]

where there is only one essential interaction between neighboring spins of strength \( J > 0 \) and \( S_i = \pm 1 \).

In a real system universality dictates that the asymptotic critical behavior will be described by the simple Hamiltonian above as long as the interactions are short-range. For weaker anisotropy, longer-range interactions, or multiple interactions, one must simply measure the behavior closer to the transition, i.e. the system will eventually cross over to the correct asymptotic behavior.

The pure \( FeF_2 \) system lies within an excellent \( d = 3 \) Ising system. The critical exponent \( \alpha \) and amplitude ratio \( A^+ / A^- \) for the specific heat

\[
C_p = A^\pm |t|^{-\alpha} + B
\]

from birefringence and pulsed specific heat techniques agree precisely with the results of many theoretical techniques. The correlation length for fluctuations

\[
\xi = \xi_0 |t|^{-\nu}
\]

and the staggered susceptibility

\[
\chi_s = \chi_0^+ |t|^{-\gamma}
\]

have been obtained from neutron scattering experiments, the staggered magnetization

\[
M_s = M_0 |t|^\beta
\]

which is only nonzero for \( t < 0 \), from Mössbauer experiments, and the dynamic critical behavior for the relaxation times

\[
\tau \sim \xi^\nu .
\]

from spin-echo neutron scattering techniques. All of the critical behavior measured over the critical range \( |t| < 0.02 \) in pure \( FeF_2 \) is in superb agreement with theory. The observation of asymptotic behavior over such a large range of \( |t| \) is a result of the strong single-ion anisotropy and the simplicity of the magnetic interactions in pure \( FeF_2 \). The magnetic ions form a tetragonal body-centered lattice and the exchange interaction between the body center and corner ions is the only significant one.

The critical parameters measured for the pure Ising model using the \( FeF_2 \) system are summarized in Table 1 along with relevant theoretical results. It is important to realize that the universal ratios of the amplitudes are just as important in characterizing the critical behavior as the exponents. The results demonstrate that \( FeF_2 \) is an exemplary Ising system and, upon dilution, an ideal one to study the effects of random-exchange and random-fields for \( d = 3 \).

The pure \( d = 2 \) Ising transition is very well represented by the isomorphic antiferromagnets \( K_2CoF_4 \) and \( Rb_2CoF_4 \). The interactions between the planes of magnetic \( C_0^+ \) ions are extremely small compared to the interactions within the planes. The specific heat behavior lies has been measured using birefringence techniques in \( Rb_2CoF_4 \). From the Onsager solution of the \( d = 2 \) Ising model, we expect the specific heat to be constant with the asymptotic logarithmic form

\[
C_p = A \ln(|t|) + B.
\]

The data indeed agree well with this form. The critical behavior of \( \xi, \chi_s \) and \( M_s \) have been obtained using
neutron scattering techniques. The static critical behavior exponents and amplitude ratios are consistent with theory.\textsuperscript{21,23} In addition, the two-scale universal ratio\textsuperscript{24}

\[ R_s = \lambda_+^{\nu} \kappa_0^{\sigma_d} / M_0, \]

has been demonstrated in these compounds. The results of experiments on these systems and comparisons to theory are summarized in Table I. These systems, when diluted, are obvious choices for investigating random-exchange and random-field effects in two dimensions.

Table I: Experimental and Theoretical Pure Ising Critical Parameters. The definitions for the exponents and amplitudes can be found in Eqs. (28). III d = 3 experimental parameters are obtained from FeF\textsubscript{2}. The parameters for d = 2 were obtained using Rb\textsubscript{2}CoF\textsubscript{2} or K\textsubscript{2}CoF\textsubscript{3}. Superscripts + or - refer to those obtained for \( \nu \) and \( \gamma \) using only data for 1 > 0 and 1 < 0, respectively.

\[ d = 2 \text{ PURE ISING} \]

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Experiment</th>
<th>Theory</th>
</tr>
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<tbody>
<tr>
<td>( \alpha )</td>
<td>0.00 ± 0.01 \textsuperscript{a}</td>
<td>0 (log</td>
</tr>
<tr>
<td>( \lambda^+ / \lambda^- )</td>
<td>1.01 ± 0.00 \textsuperscript{a}</td>
<td>1 (log</td>
</tr>
<tr>
<td>( \nu )</td>
<td>1.02 ± 0.03 \textsuperscript{c}</td>
<td>1</td>
</tr>
<tr>
<td>( \kappa_0^{\sigma_d} )</td>
<td>1.2 ± 1.13 \textsuperscript{c}</td>
<td>1/2 \textsuperscript{d}</td>
</tr>
<tr>
<td>( \gamma )</td>
<td>1.02 ± 0.07 \textsuperscript{c}</td>
<td>1/4 \textsuperscript{e}</td>
</tr>
<tr>
<td>( \lambda^+ / \lambda^- )</td>
<td>32.6 ± 3.7 \textsuperscript{c}</td>
<td>37.33 \textsuperscript{d}</td>
</tr>
<tr>
<td>( \beta )</td>
<td>0.155 ± 0.02 \textsuperscript{e}</td>
<td>1/8 \textsuperscript{f}</td>
</tr>
<tr>
<td>( R_s )</td>
<td>0.0565\textsuperscript{e}, 0.043 \textsuperscript{e}</td>
<td>0.051 \textsuperscript{f}</td>
</tr>
</tbody>
</table>

\[ d = 3 \text{ PURE ISING} \]

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Experiment</th>
<th>Theory</th>
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<tbody>
<tr>
<td>( \alpha )</td>
<td>0.1 ± 0.005 \textsuperscript{g}</td>
<td>0.11 ± 0.003 \textsuperscript{h}</td>
</tr>
<tr>
<td>( \lambda^+ / \lambda^- )</td>
<td>0.51 ± 0.02 \textsuperscript{g}</td>
<td>0.55 \textsuperscript{h}</td>
</tr>
<tr>
<td>( \nu )</td>
<td>0.61 ± 0.01 \textsuperscript{j}</td>
<td>0.630 ± 0.001</td>
</tr>
<tr>
<td>( \kappa_0^{\sigma_d} / \kappa_0^{\sigma_d} )</td>
<td>0.53 ± 0.01 \textsuperscript{j}</td>
<td>0.52 \textsuperscript{j}</td>
</tr>
<tr>
<td>( \gamma )</td>
<td>1.25 ± 0.02 \textsuperscript{j}</td>
<td>1.240 ± 0.001 \textsuperscript{b}</td>
</tr>
<tr>
<td>( \lambda^+ / \lambda^- )</td>
<td>4.6 ± 4.8 \textsuperscript{j}</td>
<td>4.8 \textsuperscript{j}</td>
</tr>
<tr>
<td>( \beta )</td>
<td>0.325 ± 0.005 \textsuperscript{c}</td>
<td>0.325 ± 0.001 \textsuperscript{h}</td>
</tr>
<tr>
<td>( Z )</td>
<td>2.1 ± 0.1 \textsuperscript{o}</td>
<td>1.05 ± 0.07 \textsuperscript{m}</td>
</tr>
</tbody>
</table>

a) ref.\textsuperscript{20}; b) ref.\textsuperscript{21}; c) ref.\textsuperscript{22}; d) ref.\textsuperscript{23,24}; e) ref.\textsuperscript{25}; f) ref.\textsuperscript{26}; g) ref.\textsuperscript{10}; h) ref.\textsuperscript{11}; i) ref.\textsuperscript{12}; j) ref.\textsuperscript{13}; k) ref.\textsuperscript{14}; l) ref.\textsuperscript{15}; m) ref.\textsuperscript{16}; n) ref.\textsuperscript{17}.

An important point should be made concerning the measurement of the staggered magnetization with neutron scattering techniques. In the d = 3 case, the samples are typically of such high crystalline quality that those neutrons aligned well enough to Bragg scatter do so in the first few microns of the crystal. This results in a saturation effect in the observed scattering intensity, an effect known as extinction. Recently, neutron Bragg scattering in epitaxial thin films has been successful in showing the proper behavior\textsuperscript{26} of the staggered magnetization in pure and dilute Ising antiferromagnets. For the case of d = 2 magnetic systems, extinction is not a problem because the order parameter scattering takes place along one-dimensional rods in reciprocal space at i = 0.5 at points as is the case for d = 3. In a result, the scattering is not saturated in bulk samples and the proper critical behavior is observed.

111. The Random-Exchange Ising Model

A simple random-exchange Ising model Hamiltonian for a single interaction site-diluted antiferromagnet is

\[ H = \sum_{<i,j>} J \epsilon_i \epsilon_j S_i S_j, \]

where \( \epsilon \) is +1 if the site is occupied and 0 otherwise. No external field is applied in this model and there are no frustrated interactions. This model can be realized to high accuracy for d = 3 in the Fe\textsubscript{2}Zn\textsubscript{1-x}F\textsubscript{2} antiferromagnet.\textsuperscript{27} Similarly, the corresponding two-dimensional REIM is extremely well represented by the magnetically dilute antiferromagnet Rb\textsubscript{2}Co\textsubscript{2}Mg\textsubscript{1-x}F\textsubscript{4}.

The most basic effect of dilution on an unfrustrated Ising system is the lowering of the tricritical temperature \( T_N(x) \). There is a limit, the percolation threshold at concentration \( x_p \), below which no tricritical points can exist since the magnetic spins cannot form a long-range network. For concentrations well above \( x_p \), it is found from both CPA theory\textsuperscript{28} and experiments\textsuperscript{29} in the d = 3 Ising system Fe\textsubscript{2}Zn\textsubscript{1-x}F\textsubscript{2} that \( T_N(x) / T_N(1) \approx x \). For the d = 2 system Rb\textsubscript{2}Co\textsubscript{2}Mg\textsubscript{1-x}F\textsubscript{4} an accurately linear decrease is observed above the percolation threshold.\textsuperscript{30} The tricritical temperature inevitably drops precipitously near the percolation threshold for both d = 2 and d = 3.

The specific heat critical behavior in a d = 3 REIM system is fundamentally different from the pure case. As first pointed out by Harris, the specific heat cannot be positive for a system with quenched dilution if the hyperscaling relation \( 2 - \nu d = 0 \) holds, as does in the REIM. (It does not hold for the RFM.) In the d = 3 Ising case this implies a crossover from the pure transition for which a > 0. The greater the dilution, the larger the region of reduced temperature over which the REIM behavior is observed. Contrary to early theoretical expectations,\textsuperscript{29} this crossover occurs quite rapidly.\textsuperscript{29,31} The new, REIM specific heat critical behavior has been measured\textsuperscript{32} for Fe\textsubscript{0.9}Zn\textsubscript{0.1}F\textsubscript{2} and more recently\textsuperscript{36} for Fe\textsubscript{0.85}Zn\textsubscript{0.15}F\textsubscript{2}, using the optical birefringence technique.\textsuperscript{37} The latter case is shown in Fig. 1. The most recent preliminary result \( \alpha = -0.9 \pm 0.02 \), using Eq. 2 agrees very well with theory\textsuperscript{35,39} and the
Quasielastic neutron scattering studies\textsuperscript{42} of Fe\textsubscript{0.46},Zn\textsubscript{0.54}F\textsubscript{2} have yielded the critical parameters for $\xi$, and $\chi$ using Lorentzian line shapes, which work well despite predictions for non-Lorentzian contributions\textsuperscript{43}. Mossbauer experiments\textsuperscript{33,34} have been used to determine the exponent for $M_\perp$.  Spin-echo neutron scattering techniques\textsuperscript{15} have shown that the critical dynamics of Fe\textsubscript{0.46}Zn\textsubscript{0.54}F\textsubscript{2} are well approximated by conventional theory\textsuperscript{15}, but with relaxation times two orders of magnitude longer than for pure FeF\textsubscript{2}.  All of the measured critical parameters and their calculated values\textsuperscript{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\textsuperscript{48} in the Rb\textsubscript{2}Co\textsubscript{0.85}Mg\textsubscript{0.15}F\textsubscript{4} system, is a marginal case with respect to the Harris criterion, since in the pure system $\alpha$ is zero (logarithmic divergence). Theories\textsuperscript{45,46} for the random-exchange behavior predict the asymptotic behavior

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

The experimental data are well described by a logarithmic divergence, as shown in Fig. 2.  It would be incredibly difficult in experiments to distinguish between fits of the data to the double logarithm in Eq. 10 and fits to the single logarithm of Eq. 7.  Hence, the experiments are consistent with the theories, though perhaps not a definitive test of them.  In the two-dimensional system Rb\textsubscript{2}Co\textsubscript{0.85}Mg\textsubscript{0.15}F\textsubscript{4}, neutron scattering measurements\textsuperscript{49} provide the critical behavior of $\xi$, $\chi$ and $M_\perp$.  In addition, a two-scale universality analysis yields a value close to that obtained in the pure material.  The experimental and theoretical results are summarized in Table II.

### Table II: Experimental and Theoretical Random-Exchange Ising Critical Parameters

<table>
<thead>
<tr>
<th>$d = 2$ RANDOM-EXCHANGE ISING</th>
<th>Experiment</th>
<th>Theory</th>
</tr>
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<tbody>
<tr>
<td>$\alpha$</td>
<td>$\approx O(\log(</td>
<td>t</td>
</tr>
<tr>
<td>$A^+/A^-$</td>
<td>$0.05 \pm 0.10^{c}$</td>
<td>$1^{d}, 100^{e}$</td>
</tr>
<tr>
<td>$\nu$</td>
<td>$1.08 \pm 0.06^{c}$</td>
<td>$1^{d}, 100^{e}$</td>
</tr>
<tr>
<td>$\kappa_\perp^{+}/\kappa_\perp^{-}$</td>
<td>$0.98 \pm 0.02^{c}$</td>
<td>$1^{d}, 100^{e}$</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>$1.75 \pm 0.07^{c}$</td>
<td>$1.753^{e}$</td>
</tr>
<tr>
<td>$\beta$</td>
<td>$0.13 \pm 0.02^{c}$</td>
<td>$-^{e}$</td>
</tr>
<tr>
<td>$R_\perp$</td>
<td>$0.062 \pm 0.01^{c}$</td>
<td>$-^{e}$</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>$d = 3$ RANDOM-EXCHANGE ISING</th>
<th>Experiment</th>
<th>Theory</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\alpha$</td>
<td>$-0.09 \pm 0.02^{f}$</td>
<td>$-0.099, -0.03^{h} - 0.01^{e}$</td>
</tr>
<tr>
<td>$A^+/A^-$</td>
<td>$1.55 \pm 0.15^{f}$</td>
<td>$-0.5^{e}$</td>
</tr>
<tr>
<td>$\nu$</td>
<td>$0.69 \pm 0.01^{j}$</td>
<td>$0.70^{g}, 0.68^{h}, 0.67^{e}$</td>
</tr>
<tr>
<td>$\kappa_\perp^{+}/\kappa_\perp^{-}$</td>
<td>$0.69 \pm 0.02^{j}$</td>
<td>$0.63^{g}$</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>$1.31 \pm 0.03^{j}$</td>
<td>$1.39^{g}, 1.34^{h}, 1.32^{e}$</td>
</tr>
<tr>
<td>$\chi_\perp^{+}/\chi_\perp^{-}$</td>
<td>$2.8 \pm 0.2^{i}$</td>
<td>$1.7^{i}$</td>
</tr>
<tr>
<td>$\beta$</td>
<td>$0.35 \pm 0.01^{k}$</td>
<td>$0.349 \pm 0.002^{h}, 0.348^{e}$</td>
</tr>
<tr>
<td>$Z$</td>
<td>$1.7 \pm 0.2^{m}$</td>
<td>$2.3^{m}$</td>
</tr>
</tbody>
</table>

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].
antiferromagnetic behavior of a ferromagnet in a random field is identical to that of a dilute antiferromagnet in a uniform field. The random and uniform fields are related by

\[ h_r^2 = \frac{x(1-x)[T^{MF}/T]^2(\mu_B S H/k_B T)^2}{[1 + \Theta^{MF}(x)/T]^2}, \]

where \( x \) is the concentration, \( T^{MF} \) is the pure system mean-field transition temperature and \( \Theta^{MF} \) the Curie-Weiss susceptibility parameter.

The early investigations of the RFIM were marked by a controversy over whether a phase transition actually took place for \( d = 3 \). This was a particularly difficult theoretical question to answer, though some of the first experiments, using the birefringence technique to measure the specific heat critical behavior, yielded strong evidence for a transition. In contrast to these conclusions, poor sample quality and a poor appreciation for the strong hysteresis in the dilute antiferromagnets did result in some groups claiming that the transition was destroyed. The rigorous theories of Imbrie and Bricmont and Kupiainen proved the validity of the conclusions of the early birefringence measurements.

Efforts now are being made to fully calculate and measure the \( d = 3 \) RFIAI critical behavior. From neutron scattering measurements, the correlation length appears to diverge with an exponent \( \nu \approx 1 \), and the exponent for \( \xi \) is approximately \( \gamma = 1.75 \). The specific heat appears to be close to a symmetric, logarithmic divergence. 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 the critical behavior. However, it was supposed to be from three to one dimension. Since there is no transition in the \( d = 1 \) Ising model, the 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 is the staggered magnetization exponent.
β. This is primarily a result of the severe extinction of the neutron scattering intensity encountered in the d = 3 crystals. The measurement of this exponent is of crucial importance for testing the RFIM critical behavior theories.

The crossover from the random-exchange to the random-field Ising behavior depends upon the strength of the random-field which in turn varies linearly with the strength of the applied field\[^{39}\]. The random-field scaling behavior dictates that the transition boundary in the H = T phase diagram varies as

\[ T_{c}(H) - T_{N} \approx H^{2/\phi} \]  \((14)\)

where the crossover exponent \(4 \approx 1.17\) with \(\gamma\) being the random-exchange staggered susceptibility exponent\[^{61}\]. This behavior has been verified\[^{32}\] by experiments on Fe\(_{2}\)Zn\(_{1-x}\)F\(_{2}\), with the results \(\gamma = 1.31 \pm 0.03\) and the value from many measurements \(\phi = 1.42 \pm 0.03\). Some of the earlier experiments leading to the conclusion that the pure value of \(\gamma\) were adversely affected by concentration gradients and the practice of taking the transition to be at the peak in the specific heat, as shown by simulations\[^{64}\].

The dynamics of the d = 3 RFIM have now been convincingly demonstrated, using ac susceptibility measurements\[^{65,66}\] to be activated in Fe\(_{2}\)Zn\(_{1-x}\)F\(_{2}\) in agreement with theory\[^{67,68}\]. Spin-echo neutron scattering measurements show\[^{19}\] that, on the very short time scale of nanoseconds, the 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 the transition. First, when cooling in a field (FC) or heating in a field after cooling in zero field (ZFC), equilibrium is lost faster the transition. Upon FC, the system cannot achieve long-range order until the data differ from the ZFC data below an equilibrium boundary \(T_{c}(H)\) which lies just above \(T_{c}(H)\) and scales in precisely the same manner. Of course, with a small enough random field and limited instrumental resolution, the FC procedure will appear to yield long-range order.

Closer to \(T_{c}(H)\), the critical behavior is rounded by the critical slowing, as exemplified by the ZFC Faraday rotation data\[^{7}\] in Fig. 3. Shapiro\[^{7,15}\] has proposed a tlcibory based on the slow dynamics which predicts the observed critical exponents.

Tlc neutron scattering line shapes observed in the d = 3 RFIM systems are far from the Lorentzian form which adequately describes the scattering in pure FeF\(_{2}\) and in Fe\(_{2}\)Zn\(_{1-x}\)F\(_{2}\) in zero field well above the percolation threshold. Mean-field tlicory predicts a squared Lorentzian line shape and it has been commonly assumed that the mean-field argument adequately explains the unusual observed line shapes. However, al-
though the squared Lorentzian works well for low temperature FC scans and scans above $T_c$, it does not work well\textsuperscript{62} for $ZFC$ scans below $T_c$. Furthermore, non-Lorentzian line shapes are observed\textsuperscript{72} at zero field in the nonequilibrium region near the percolation threshold but not observed\textsuperscript{73} in the $d = 2$ case for $H > 0$ near $T_N$ where the destruction of the transition occurs in equilibrium. All of these observations suggest a non-equilibrium origin to the non-Lorentzian line shapes near $T_c(II)$ for $d = 3$.

For the $d = 2$ RFM system $Rb_2Co_8.85Mn_{0.15}F_6$, the transition is observed to be destroyed\textsuperscript{48} as shown in Fig. 2, iii agreement with theory\textsuperscript{50}. No hysteresis or extremely slow dynamics are observed near the destroyed transition, indicating equilibrium behavior. At low temperatures, however, metastable long-range order induced by ZFC decays only when the rounded transition is approached sufficiently closely. This occurs closer to the transition as the measurement time scale decreases, as indicated by the comparison of high field pulsed magnetization experiments\textsuperscript{74} and neutron scattering measurements\textsuperscript{75} which differ in the measurement time scale by eight orders of magnitude.

V. Metastable Domain Wall Dynamics

Random-exchange interactions not only affect the phase transition to long-range order in a profound way, but also the equilibrium dynamics for $T \ll T_N$. The fact that the interactions in $Fe_2Zn_{1-x}F_2$ are so well understood and are very simple makes this system ideal for investigating the dynamics of metastable domain walls at low temperature. Domain walls may be conveniently introduced into the system by FC. The length scale associated with the domains, as evident from the widths of neutron scattering line profiles\textsuperscript{76}, decreases as the strength of the applied field upon FC increases. Two distinct kinds of dynamics can be investigated. With a field applied the evolution of the domain will be influenced by pinning from the random field as well as from vacancies. Once the field is removed, on the other hand, the only pinning remaining is from the vacancies. We shall discuss the latter case first.

At low enough temperatures, it has been shown from neutron scattering\textsuperscript{77} and Faraday rotation experiments\textsuperscript{78,70} that the domain walls do not move macroscopically even after the field is removed. A common misconception is that the width of the neutron scattering profiles yields directly the size of metastable domains. Computer simulations\textsuperscript{79} demonstrate, however, that the magnetic system forms essentially only two domains that are incredibly intertwined, as shown in Fig. 4. A: best, the non-Lorentzian width represents the typical distance needed to pass from one domain to the other. Nowak and Usadel\textsuperscript{80} suggest that the domains are fractal in structure. The line shape has been studied with Monte Carlo simulations\textsuperscript{81} and has some unusual properties associated with the fractal-like structure.

The fact that the domain structure does not evolve over a macroscopic scale at low temperatures indicates that the thermal fluctuations are not strong enough to overcome the vacancy pinning. The walls evolve on a microscopic scale, however. Early experiments\textsuperscript{70} showed a time dependence to the remanent magnetization after the field is turned off at low temperatures. The remanent magnetization originates from the domain walls formed upon FC. The domain walls have magnetization since, upon cooling in a field, the spins on either side of the domain wall are parallel to the field to reduce the local random-field free energy. Once the field is removed, there is no energetic advantage to alignment with the field. The domain walls then evolve in such a way as to locally minimize the exchange energy, i.e., the surface area of the walls is reduced. Spins along the wall which were predominantly aligned with the field will flip as the wall translates by one lattice spacing. This provides the mechanism by which macroscopic wall movements, driven by the exchange energy, reduce the net magnetization\textsuperscript{82}.

Nattermann and Villain\textsuperscript{63} initiated the idea that microscopic wall movements aid macroscopic motion constituted the mechanism for the decay of the remanent magnetization and attempt to explain the original experimental results. They proposed the decay

\[ M_t = M_0 \exp \left[ -\frac{\phi}{\psi} \right] + B, \]

where $\psi \approx 0.4$ and $B$ is a constant volume term which is small, to describe the experimental results\textsuperscript{70} for $Fe_0.47Zn_{0.53}F_2$. Later Monte Carlo simulation data obtained by Nowak and Usadel\textsuperscript{85} using a simple-cubic lattice showed better agreement with the power law behavior

\[ M_t \sim \left| t \right|^{-x}, \]

aid attributed the behavior to a lack of a characteristic length scale associated with the fractal domains.

Further experimental studies\textsuperscript{86} and Monte Carlo simulations\textsuperscript{82} indicate a new expression which does an excellent job of describing the data for the body-centered cubic lattice of dilute $Fe_0.47Zn_{0.53}F_2$ and a simulated body-centered cubic lattice. The expression

\[ M_t = M_0 \exp \left[ -\frac{\phi}{\psi} \right], \]

yields exceptionally good fits of the data with $\psi$ independent of the field and $\psi$ dependent on temperature. A suitable theory for this form is lacking.

The dynamics associated with domain wall pinning by random fields have been studied\textsuperscript{87} by employing the FC procedure at temperatures not far below $T_c(II)$ and measuring the time dependence of the uniform magnetization in the presence of the field using SQUID techniques. Apparently, at such temperatures the random-field pinning dominates over the vacancy pinning aid
many of the observations are consistent with the theoretical models for low-temperature random-field activated dynamics. This implies that there must be many small activated domains as opposed to the fractal-like, two-domain, frozen structure existing at low temperature.

VI. The Percolation Threshold in the Ising Model

The d = 3 Ising model system Fe_xZn_{1-x}F_2 near the percolation threshold provided some experimental surprises. Earlier experiments on the weakly anisotropic, isomorphic antiferromagnet Mn_xZn_{1-x}F_2 were interpreted in terms of a geometric correlation length, \( \kappa_G \), and a thermal correlation length. 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 the Ising system. In zero field, even a few percent above the percolation threshold, long-range order does not develop. Instead, near the percolation threshold a spin glass-like phase is encountered. Yet this system cannot be considered a canonical spin glass which requires two ingredients, randomness and frustration. In zero field, there is little or no frustration in Fe_xZn_{1-x}F_2. Nevertheless, in the H - T phase diagram there is a boundary which is completely analogous to the spin glass de Almeida-Thouless (AT) boundary including the scaling exponent \( \phi \approx 3.4 \). This was first observed in magnetization studies by Montenegro, et al. Even in zero field no long-range order develops for \( x \leq 0.27 \) and unusual line shapes are evident for \( T < 12 \)K. A bit further above the percolation threshold, at \( x = 0.31 \), both the usual random-field behavior at low \( H \) and the spin glass-like behavior at higher \( H \) exist, as shown in the H - T phase diagram in Fig. 5. The random-field behavior at low \( H \) is the same as that seen for all \( H \) at higher concentrations \( x \geq 0.46 \). At higher fields, the equilibrium boundary scaling with \( \phi \approx 3.4 \) appears. No long-range order persists in the region at high field. It has been shown in Monte Carlo simulations, in which it is assumed that there are no frustrating interactions, that a phase diagram very much like that seen in Fe_xZn_{1-x}F_2 is emergent. It is interesting to note that the observation of such behavior may be indicative of quenched randomness rather than simply a manifestation of a spin glass system, as is commonly assumed. The underlying physics causing the spin glass-like behavior in Fe_xZn_{1-x}F_2 must be related to the extremely slow dynamics associated with the Ising percolation threshold. A complete understanding of the behavior in this system is important in itself and may shed light on the spin glass problem as well.
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. 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, 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 by the system only for $q < k_G$.

The $H - T$ phase diagrams of this material for
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27. Many of the highest quality crystals used in the $d = 3$ experiments have been grown at the UCSB Materials Preparation Laboratory by N. Nighman.
42. J. P. Hill, T. R. Thurston, R. W. Erwin, M. J. Ramstacl and R. J. Birgeneau, Phys. Rev. Lett. 66, 3281 (1991). In this work on a weakly anisotropic system, the authors claim to be the first to observe the RFIM transition, disregarding all previous work on this subject in which the critical behavior has been studied. At low enough field and large enough magnetic concentration the line shapes will of course be resolution limited as observed in this work. The problem is in interpretations relying solely on the width of the diffuse scattering peaks for which a precise theoretical form is unknown. The same method has been used to incorrectly deduce that there is no RFIM transition, and that the transition is first order, both in the same system used above.
H. Montenegro, dependence

Guggenheim, magnetization dependent.

A. R. King, V. Jaccarino, unpublished.


Iliaas recently been pointed out by T. Nattermann in a private communication that power law behavior is also consistent with the original N-V picture of microscopic domain wall rearrangements if the power law dependence of AE on the effective exchange scale L is replaced by a logarithmic one. This transforms the logarithmic time dependence of the remanent magnetization given in the original N-V theory into a power law dependence.


