

Final Report
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1 Progress

1.1 The random-field Ising model at high magnetic concentration

During the project period we have largely completed our characterization of the critical behavior of the random-field Ising model (RFIM) transition in three dimensions ($d = 3$). This has been a long sought-after goal, the transition having been first observed in the early 1980's[1, 2]. Our thinking about the phase transition in the RFIM was greatly advanced when we discovered a few years ago that one has to work at concentrations $x > x_v = 0.76$, the magnetic vacancy percolation threshold concentration[3, 4]. As described below, at lower concentrations, the transition is obscured by metastable domains. Once we realized this, we were able devote considerable effort to understanding the RFIM critical behavior to test theories and predictions from computer simulations. We applied neutron scattering, x-ray scattering, optical, specific heat, and other techniques to achieve a nearly complete characterization of this important phase transition, devoting most of our effort to one particular high magnetic concentration crystal $\text{Fe}_{0.85}\text{Zn}_{0.15}\text{F}_2$. The $\text{Fe}_x\text{Zn}_{1-x}\text{F}_2$ system has been well documented and studied as an ideal realization of the RFIM when subjected to an applied field along its easy axis. The significance of this particular phase transition is that it represents one of the major classes of randomness in ordering processes, distinct from random-exchange and spin-glass ordering. This baffling problem of ordering in the presence of random fields has now been brought by our experiments to a much higher level of understanding.

The early studies of the RFIM for three dimensions were very controversial. To achieve sufficiently large random fields, a large amount of dilution was required for the available magnetic fields. It was not appreciated that the large number of vacancies would result in domain formation that would obscure the phase transition in scattering experiments, though not in specific heat experiments. Our group and collaborators have always maintained that there was an underlying phase transition and our recent experiments bear this out.

Our group[5] and others[6], using Monte Carlo simulations, showed that large numbers of vacancies allow the magnetic system to break into two domains with intricate domain walls that take great advantage of magnetic vacancies to avoid domain wall formation energies[7]. Hence, the conventional wisdom based on the domain wall energy arguments are inapplicable. Our Monte Carlo results[3] indicated that domains would not form for magnetic concentrations x above the vacancy percolation threshold concentration, $x_v = 0.76$. The stability of the long-range antiferromagnetic order at high magnetic concentrations was first demonstrated[4] in neutron scattering experiments on $\text{Fe}_{0.93}\text{Zn}_{0.07}\text{F}_2$ in a field $H = 7$ T. Unfortunately, the field was not large enough at this high concentration to ensure that the results were not unduly influenced by crossover effects and we certainly could not study the critical behavior as a function of the field strength. Over the past two years or so, we have focused attention on two samples, $\text{Fe}_{0.76}\text{Zn}_{0.24}\text{F}_2$ and particularly $\text{Fe}_{0.85}\text{Zn}_{0.15}\text{F}_2$. The first crystal is just at the vacancy percolation threshold x_v . At low temperatures, where normally one does not expect scattering intensity away from the Bragg point, this crystal exhibits line shapes demonstrating the fractal nature of the vacancy structures[8]. The antiferromagnetic ordering at $x = 0.76$ is unstable above about $H = 6.5$ T, consistent with the trend of lower concentration samples to enter a spin-glass-like region. On the other hand, it shows no evidence of domain formation at low fields in contrast to lower magnetic concentrations. These results are quite in accord with our simulations[3] that indicate $x = 0.76$ is a special boundary in concentration separating crystals dominated by metastable domain behavior from those exhibiting stable long-range order.

At slightly higher magnetic concentration, the crystal of $\text{Fe}_{0.85}\text{Zn}_{0.15}\text{F}_2$ exhibits antiferromagnetic order that is stable to approximately $H = 18$ T[9]. In addition to showing no metastable domain formation at moderate fields, this crystal is far enough away from x_v to avoid any spurious line shape effects from fractal vacancy structures. It is this sample which has allowed us to characterize in great detail the RFIM phase transition. Along with other techniques, we now have a complete set of critical exponents that are self consistent and the transition seems to be a second-order one. Examples of the critical behavior are shown in Fig. 1 and 2 for the specific heat and order parameter critical behaviors, respectively. However, it shows strong hysteresis, something not normally associated with second-order transitions.

The critical behavior of the RFIM is shown in Table I. Both the simulations and experimental results are shown. The experimental exponents are obtained upon cooling in zero field, raising the field and warming across the phase boundary (ZFC). The simulation results are obtained under equilibrium conditions. Note that there is very poor agreement between experiments and simulations. Nevertheless, the results of both are of very high quality and the experimental critical behavior measurements in this sample are measured nearly as accu-

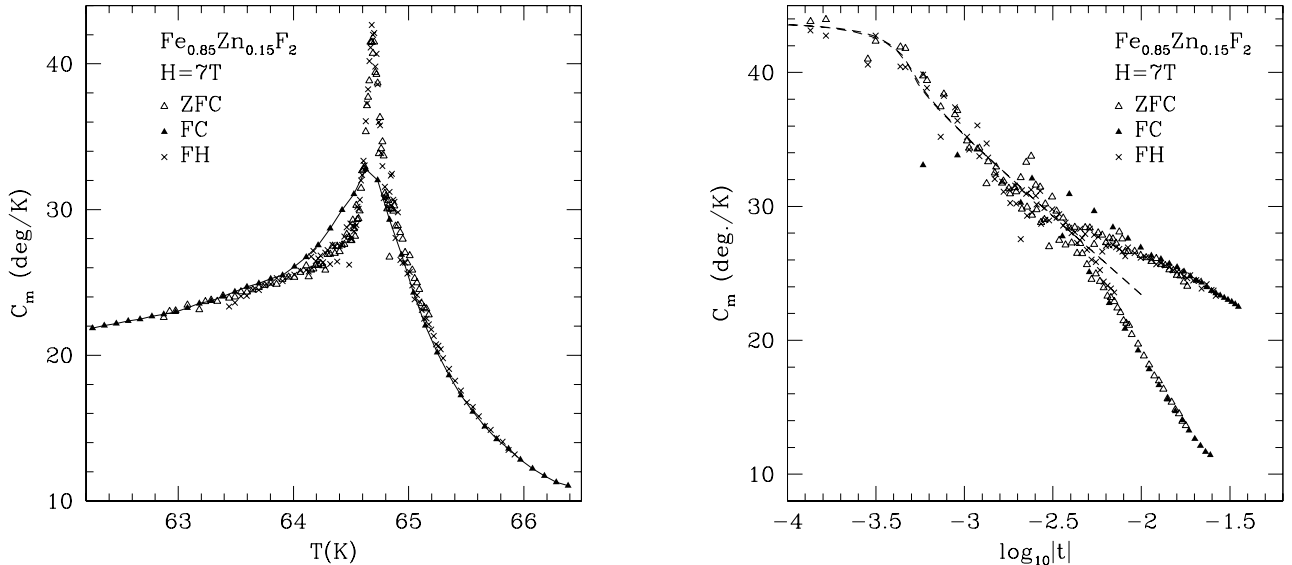


Figure 1: The critical behavior of the temperature derivative of the linear birefringence, which is proportional to the magnetic specific heat. ZFC yields extremely sharp behavior which is symmetric and logarithmic. In the right figure, the dashed line represents the rounding expected by the measured concentration gradient. FC yields a smaller peak. FH (warming after FC) yields a peak essentially identical to ZFC.

rately as the most detailed studies of pure systems. In addition, the experimental exponents are self consistent in that they satisfy the Rushbrooke equation

$$2\beta + \gamma + \alpha = 2 \quad . \quad (1)$$

Adding the values from the table, we obtain a value 2.00 ± 0.09 . So, why is there such stark disagreement between the simulation and experimental results? A hint comes from the fact that if one cools in the field (FC), different behavior is obtained, as illustrated in Fig. 2. Clearly, the second-order transition is not that of a system in equilibrium. There is no requirement that scaling behaviors occur only in equilibrium systems, though most people associate equilibrium with second-order scaling through experience and the fact that first-order transitions are notoriously non-equilibrium. As is the convention, great effort was taken in Monte Carlo simulations to ensure equilibrium. Only small lattices, where equilibrium could be ensured by sufficiently long runs, were studied. Results were also obtained from exact ground state calculations on small lattices, which by definition yield equilibrium results.

An example of the disagreement with experiments can be seen in the simulated specific heat behavior[10]. There is clearly no divergence in the specific heat peak in the simulations

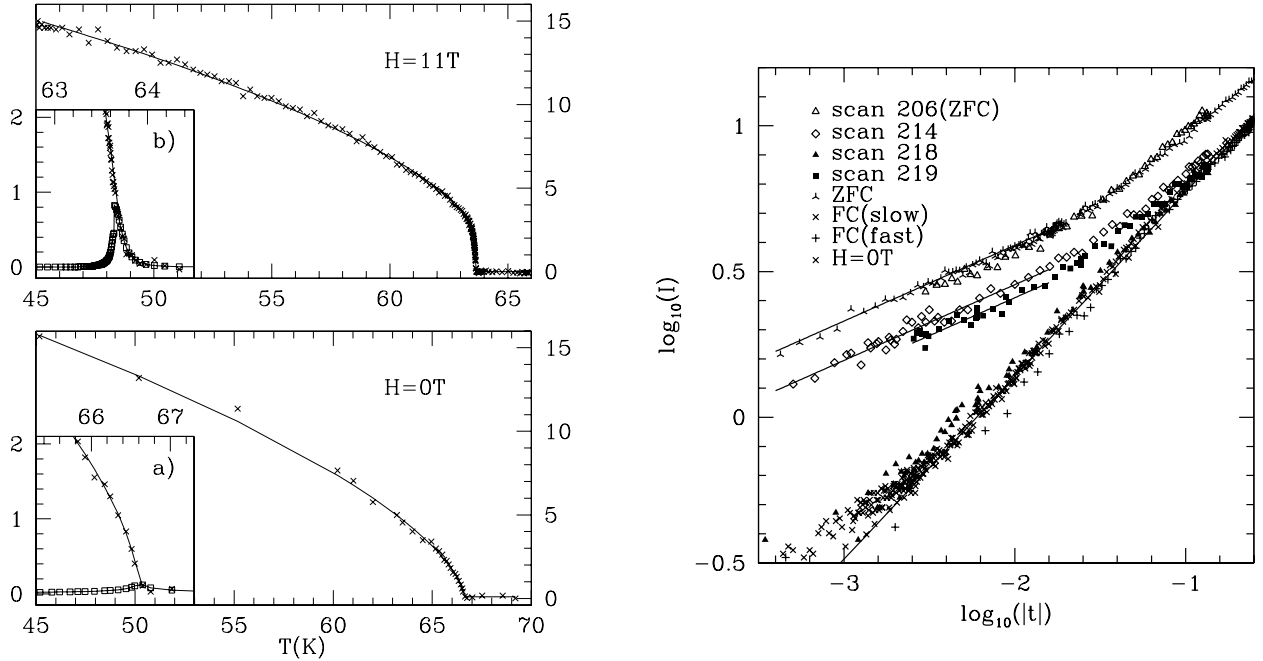


Figure 2: The order parameter, measured using magnetic x-ray scattering in $\text{Fe}_{0.85}\text{Zn}_{0.15}\text{F}_2$ in zero field and for ZFC in a field of $H = 11$ T[15]. When data are shown in a log-log plot, for all data taken upon ZFC, or other procedures where the transition is crossed from below in the field, the exponent $\beta = 0.16$ is quite apparent for small $|t|$. However, FC data fall precisely on $H = 0$ data. Not only is there a large hysteresis, despite the power law behaviors, when cooling in the field, the power law for FC is essentially identical to that with no field.

but, as can be seen in Fig. 1, a clear symmetric, logarithmic behavior is indicated in the experiments. Our group has conducted a Monte Carlo study using large lattices that cannot be equilibrated. Indeed, the results, exemplified in Fig. 3 for the order parameter, are very different from the equilibrated Monte Carlo results, but look similar to the experimental results shown in Fig. 2, further corroborating that the experiments do not reflect equilibrium behavior.

We now know that the RFIM transition is a second-order-like transition that is not in equilibrium. We still need to know what is the diverging length scale that governs the scaling behavior and understand why the hysteresis is so strong. It is quite possible that second-order transitions in many random systems share such attributes, even if the critical points are hidden as in $\text{Fe}_x\text{Zn}_{1-x}\text{F}_2$ for $x < x_v$. The foundation established in our experiments and simulations will help to unravel complex behavior in other ordering systems.

One of the very difficult problems facing us in the characterization of the RFIM ordering processes was that the scattering line shapes were far from mean-field predictions, but theorists provided little insight into what line shapes were suitable for analysis of scattering data. This has implications for any phase transition in systems with quenched randomness, where

exponent	experimental	simulation
α	0.00 ± 0.01^a	-0.5 ± 0.2^d
		-0.55 ± 0.2^e
		-0.63 ± 0.07^f
		-0.01 ± 0.09^g
β	0.16 ± 0.02^b	0.02 ± 0.01^h
		0.017 ± 0.005^g
ν	1.18 ± 0.05^c	1.1 ± 0.2^d
		1.14 ± 0.10^h
		1.32 ± 0.07^f
		1.37 ± 0.09^g
γ	1.68 ± 0.05^c	1.7 ± 0.2^d
		1.5 ± 0.2^e

Table 1: Measured values for the critical exponents obtained from the experiments done on the diluted antiferromagnet in a field and simulations on random field magnets.

^a Reference [2, 41] ^b Reference [15]

^c Reference [13] ^d Reference [42]

^e Reference [43] ^f Reference [10]

^g Reference [11] ^h Reference [12]

the line shapes are unknown and likely to be far from mean-field predictions. One of the accomplishments of our group was to develop a technique that invokes scaling, but allows the line shapes to be analyzed to obtain the critical behavior with little a priori input. We used this[13, 4] technique in analyses of the behavior in $\text{Fe}_{0.85}\text{Zn}_{0.15}\text{F}_2$ and $\text{Fe}_{0.93}\text{Zn}_{0.07}\text{F}_2$ and the technique allowed us to extract the critical exponents. Nevertheless, there was still little comprehension for why the line shapes had such unusual forms far from mean field ones.

The ordering process in random-field systems was examined[14] in exact ground state simulations and line shapes were generated that behaved much like our experimental ones[13]. The ordering process was shown to be rather interesting in that, above the transition, percolating spanning clusters with a fractal structure form, with a length scale that diverges at the transition. We reexamined the line shapes in our experiments as well as in the simulations and found that indeed the line shapes are governed by this fractal structure[13] as demonstrated in Fig. 4. These RFIM results are likely relevant to other phase transitions in random and frustrated systems and similar techniques are surely applicable.

1.2 Magnetic x-ray scattering

In the RFIM section above, the results obtained for the order parameter were obtained using magnetic x-ray scattering techniques at SSRL. This is a new technique to the group and our scattering experiments were the first done using the high field magnet. Fields up

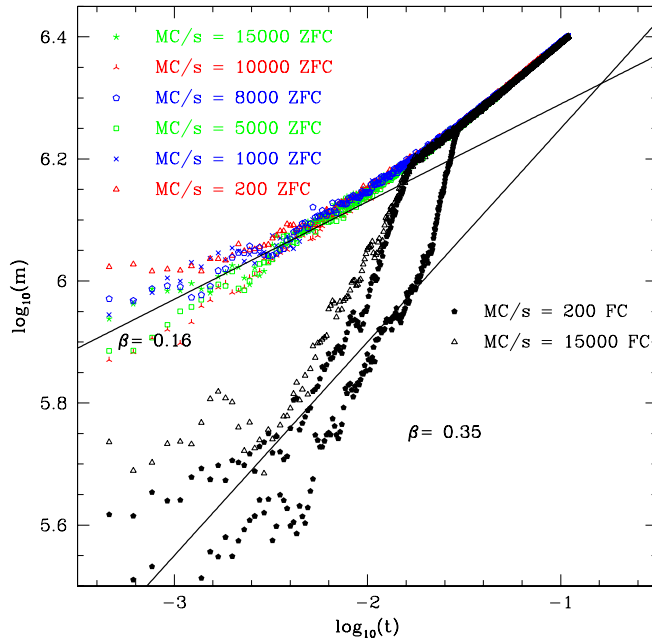


Figure 3: Monte Carlo simulations for a dilute antiferromagnet in a field[44]. The solid lines represent the asymptotic behaviors observed in the x-ray experiments. Several ZFC runs of various rates yield the same critical behavior as the experiments. FC runs are less stable, but tend to the same behavior as the experiments for small $|t|$, but then jump up to the ZFC curves. This is probably an effect of the finite lattice size of 128^3 in the simulations.

to 11 T were used to study[15] the RFIM by measuring the square of the order parameter, given by the Bragg intensity in $\text{Fe}_{0.85}\text{Zn}_{0.15}\text{F}_2$. This measurement could not be accomplished using neutron scattering because of severe extinction problems. Our experiments were featured in the SSRL experiment highlights which now can be found archived at the www-ssrl.slac.stanford.edu/research/highlights_archive/Ising.html web site. This facility will be useful in other systems when extinction is a factor limiting measurements using neutrons.

1.3 The RFIM at low magnetic concentration

While metastable domains obscure the RFIM critical behavior in $d = 3$, and it was a breakthrough to understand that they only occur for $x < x_v$, the domains themselves are of interest. The relaxation of the domain walls after the field is removed was studied through experiments[16] and simulations[5]. We studied the dynamics in both $d = 2$, using $\text{Rb}_2\text{Co}_{0.85}\text{Mg}_{0.15}\text{F}_4$, and $d = 3$, using $\text{Fe}_x\text{Zn}_{1-x}\text{F}_2$. No transition takes place in $d = 2$, but one does take place for $d = 3$. Not only did we characterize the experimental behavior for $d = 3$, we did the simulations to find scaling equations describing the behavior as well. These behaviors were confirmed in later independent studies. These domains have been argued to

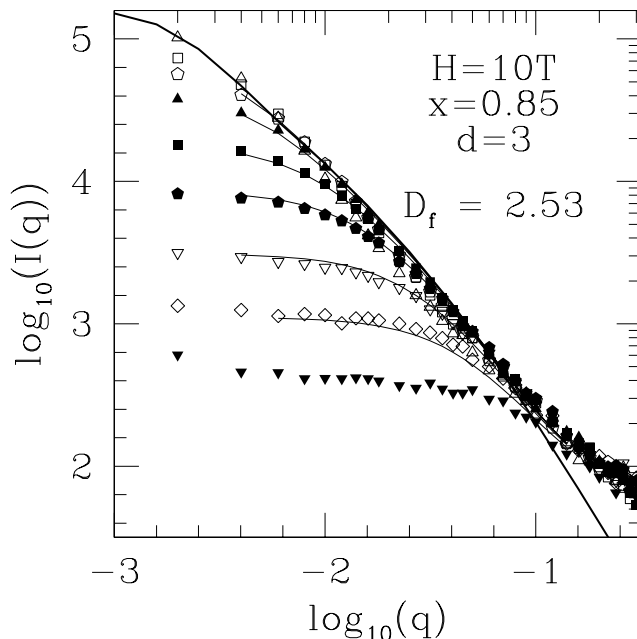


Figure 4: The fractal nature of the $|q| > 0$ line shapes in the RFIM as realized in the dilute antiferromagnet $\text{Fe}_x\text{Zn}_{0.85}\text{F}_2$ above the transition temperature. The solid curve is the percolation fractal behavior $I(q) \sim |q|^{-D_f}$, where $D_f = 2.53$ is the percolation fractal dimensionality. The curvature is from having the instrumental resolution folded in. The data curves are at different temperatures. As the transition temperature is approached, the data follow the fractal behavior for small $|q|$. Hence, the line shape must be governed by the fractal RFIM spanning clusters.

have relevance in magnetic recording devices[17].

1.4 Neutron scattering on films

We measured the critical behavior in FeF_2 [18] and layered $\text{FeF}_2/\text{CoF}_2$ [19] films using reactor sources. We also did extensive studies[20] on the RFIM system $\text{Fe}_{0.52}\text{Zn}_{0.48}\text{F}_2$ which showed that domains form upon either FC or ZFC near the transition temperature and explained why neutron scattering at such a low magnetic concentration could not be used for measuring the order parameter. Most recently, we have demonstrated that spallation sources can also be used in such studies. We measured[21] the ordering in the RFIM system $\text{Fe}_{0.85}\text{Zn}_{0.15}\text{F}_2$, which has a magnetic concentration sufficiently high so that domains do not form. This was the first use of the high field cryostat at the ASTERIX spectrometer at Los Alamos. We have just collected data on the instrumental resolution and will more accurately subtract the critical scattering background using data obtained at this concentration in reactor-based neutron scattering experiments[13]. Our results at Los Alamos were highlighted in the <http://lansce.lanl.gov/research/ASTERIX11T.html> web site archive.

1.5 Chiral phase transitions

Another area that my group has studied under this grant is relevant to proposed experiments to be done over the next three years and therefore warrants review. Our group was the first[22] to characterize the specific heat critical behavior in CsMnBr_3 , a highly frustrated XY antiferromagnet on a triangular lattice. Our results were subsequently verified. The spins can form one of two 120 degree configurations. One cannot be obtained from the other by a global spin rotation and, hence, a symmetry breaking occurs which H. Kawamura predicted to be that of the chiral universality class. We measured the exponent $\alpha = 0.39 \pm 0.09$, a very large value consistent with predictions by Kawamura[24]. These results are also consistent with neutron scattering results[25]. However, some lingering controversy persists since the transition splits when a field is applied. Some have suggested that the $H = 0$ transition is therefore at a multicritical point which influences the critical behavior. We also measured the transition in holmium metal[22]. This case is, in one sense, a much simpler chiral system since the spins choose between two chiralities in their spiral ordering. However, since holmium spins interact through the rather long-ranged RKKY interaction, it was not possible to access the asymptotic critical region to ascertain the specific heat critical exponent. We will propose to do an experiment in a system that has short-range interactions and exhibits a spiral structure. This might possibly be the cleanest test of the chiral universality proposed by Kawamura.

1.6 Ising antiferromagnet on a triangular lattice

Another example of a frustrated system is CsCoBr_3 , an Ising model on a triangular lattice. It was studied by our group[23]. In this case, chirality does not play a role in the transition to long range order. Nevertheless, the frustration results in a new universality class of critical behavior. Instead of a positive specific heat exponent α , as in the unfrustrated $d = 3$ Ising model, the behavior is very close to the $d = 3$ XY system with a slightly negative α .

1.7 The magnetic percolation threshold

The behavior of $\text{Fe}_{0.25}\text{Zn}_{0.75}\text{F}_2$, which is very close to the magnetic percolation threshold was explored[26] using specific heat and birefringence (Δn) techniques. As well as characterizing the magnetic ordering in this system, we showed that this is an example of when the proportionality between $d(\Delta n)/dT$ and the magnetic specific heat C_m breaks down. There is no phase transition to ensure the proportionality[27] and a small frustrating interaction plays an important role in the birefringence. There was much discussion of whether the frustrating interaction was sufficient at the percolation threshold to cause spin-glass behavior[28, 29, 30, 31]. A careful study of the susceptibility and magnetization showed that, although such characteristic spin-glass effects as memory are observed, there seems to be no finite transition temperature, a necessary condition for a canonical $d = 3$ spin glass[32].

1.8 Magnetic ordering in the weakly anisotropic antiferromagnet $\text{Mn}_x\text{Zn}_{1-x}\text{F}_2$

We were able in this system to explore[33], with available fields, the $H - T$ phase diagram, including the spin-glass-like behavior at high fields and the low-field conventional RFIM behavior. The paramagnetic phase boundary exhibits curvature characteristic of the RFIM at low H and a spin-glass at high H .

1.9 Excitations in the dilute Ising antiferromagnet $\text{Fe}_x\text{Zn}_{1-x}\text{F}_2$

We recently completed a far-infrared study[34] of low temperature excitations in $\text{Fe}_x\text{Zn}_{1-x}\text{F}_2$ that complemented a neutron scattering study sometime before that[35]. We also did a series of pulsed field studies of the excitations in this system[36]. The main point of these studies is that, although the mean field predictions for the excitations roughly gives the energy range of the excitations, the details are wrong. The number and separation of the excitations observed is quite different from the mean field calculations. It is still not understood what these fundamental excitations represent, even though the system could not be simpler in structure.

1.10 Structural RFIM system, $\text{DyAs}_x\text{V}_{1-x}\text{O}_4$

An example of the application of RFIM physics to a non-magnetic system is in $\text{DyAs}_x\text{V}_{1-x}\text{O}_4$. We studied[37] the specific heat of this system for $x = 0.15$. The effective short range of interactions nominally puts this system into the same class as $\text{Fe}_x\text{Zn}_{1-x}\text{F}_2$ in an applied field. It does not seem to be completely analogous, however, since we saw the entropy greatly diminished in the structural transition and it was not clear that the critical behavior was the same.

1.11 Magnetic phase transition in UNi_4B

We made specific heat critical behavior measurements in UNi_4B in zero field to complement other measurements characterizing the transition in this compound. The critical exponent and amplitude ratio were indicative of the $d = 3$ Heisenberg universality class.

1.12 Magnetic behavior of $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$

A neutron scattering, susceptibility and specific heat study[38] of the random dipolar magnet were made to study the glassy behavior of the spin correlations at low temperatures.

1.13 Two-dimensional RFIM system $\text{RbCo}_x\text{Mg}_{1-x}\text{F}_4$

Studies were made of the ordering processes in the $d = 2$ RFIM system $\text{RbCo}_x\text{Mg}_{1-x}\text{F}_4$, which has no phase transition, but does show interesting dynamics[39, 40].

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2 Chronological list of recent publications

A complete compilation of papers can be found at the dave.ucsc.edu website.

1. *Stationary states and criticality of the Order-Parameter of the $d = 3$ Random-Field Ising Antiferromagnet $\text{Fe}_{0.85}\text{Zn}_{0.15}\text{F}_2$* , F. Ye, L. Zhou, S. A. Meyer, L. J. Shelton, D. P. Belanger, L. Lu, S. Larochelle, and M. Greven, unpublished.
2. *Optical studies of the specific heat of $d=3$ Random-Field Ising Antiferromagnet $\text{Fe}_{0.85}\text{Zn}_{0.15}\text{F}_2$* , F. Ye, L. J. Shelton, S. A. Meyer, and D. P. Belanger, unpublished.
3. *Magnetic Vacancy Percolation in Dilute Antiferromagnets*, W. C. Barber, F. Ye, D. P. Belanger and J. A. Fernandez-Baca, submitted to Physical Review B.
4. *Simulations of irreversibilities of the random-field Ising model order parameter*, L. J. Shelton, F. Ye, W. C. Barber, L. Zhou, D. P. Belanger, To be published in the proceedings of ICM 2003, Rome.
5. *Percolation fractal dimension in scattering line shapes of the random-field Ising model*, F. Ye, M. Matsuda, S. Katano, H. Yoshizawa, D. P. Belanger, E. T. Seppala, J. A. Fernandez-Baca, M. J. Alava, To be published in the proceedings of ICM 2003, Rome.
6. *The spallation neutron source is taking shape*, T. E. Mason, R. K. Crawford, G. J. Bunick, A. E. Ekkebus, D.P. Belanger, *Appl. Phys. A* **74**, S11-S14 (2002).
7. *Order Parameter Criticality of the $d=3$ Random-Field Ising Antiferromagnet $\text{Fe}_{0.85}\text{Zn}_{0.15}\text{F}_2$* , F. Ye, L. Zhou, S. Larochelle, L. Lu, D. P. Belanger, M. Greven, and D. Lederman, *Phys. Rev. Lett.* **89**, 157202-157205 (2002).

8. *Large Random-Field Behavior Above the Vacancy Percolation Concentration Threshold: $Fe_{0.84}Zn_{0.16}F_2$* , T. Sakon, S. Awaji, M. Motokawa, D. P. Belanger, *J. Phys. Soc. Japan* **71**, 411 (2002).
9. *Monte Carlo Simulations of the Random-Field Ising Model*, W. C. Barber and D. P. Belanger, *J. Magn. Magn. Mater.* **226-230**, 545 (2001).
10. *Scaling properties of the critical behavior of the dilute antiferromagnet $Fe_{0.93}Zn_{0.07}F_2$* , Z. Slanić, D. P. Belanger, and J. A. Fernandez-Baca, *J. Phys.: Condens. Matter* **13** 1711 (2001).
11. *Far infrared spectroscopy on the three-dimensional dilute antiferromagnet $Fe_xZn_{1-x}F_2$* , J. Satooka, K. Katsumata, and D. P. Belanger, *J. Appl. Phys.* **89**, 7047 (2001).
12. *The random field critical concentration in dilute antiferromagnets*, W.C. Barber and D. P. Belanger, *J. Appl. Phys.* **87**, 7049 (2000).
13. *$d = 3$ random field behavior near percolation*, F. C. Montenegro, D. P. Belanger, Z. Slanic and J. A. Fernandez-Baca, *J. Appl. Phys.* **87**, 6537 (2000).
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3 Recent students

Three students working in my group have just left to continue their careers elsewhere after receiving their respective degrees. Feng Ye defended his thesis and will officially receive his PhD in December, 2003. He took a post doctoral position in the neutron scattering group at the Oak Ridge National Laboratory and will do research at the High Flux Isotope Reactor under supervision of Dr. Jaime Fernandez-Baca. His research was primarily on the $Fe_{0.85}Zn_{0.15}F_2$ crystal experiments described above, including the neutron scattering, x-ray scattering, birefringence, and Faraday rotation experiments as well as the Monte Carlo simulations.

As an undergraduate student, Leslie Shelton did her senior thesis using Faraday rotation techniques on $Fe_{0.85}Zn_{0.15}F_2$ and was involved in the Monte Carlo simulations, x-ray scattering experiments at SSRL and neutron scattering at LANL. She is now a first year graduate student in the Physics Department at Brown University.

A second undergraduate student, Stephanie Meyer, was involved in the Faraday rotation and birefringence experiments as well as x-ray scattering experiments at SSRL. She has graduated with a double major in physics and math and is now a first year graduate student in the Physics Department at the University of Colorado, Boulder.

The most recent previous student is William Barber, who graduated with his PhD in 2000. He took a post doctoral position in medical physics and instrumentation at the University of California, San Francisco.