Critical Scaling of the S=5/2 Kagome Antiferromagnet Iron Jarosite

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We report on the scaling of the heat capacity and susceptibility of the kagome lattice antiferromagnet Iron Jarosite ($KFe_3(OH)_6(SO_2)_2$). This material is a candidate quantum spin liquid due to the presence of geometrical frustration and Dzyaloshinsky-Moriya (DM) interactions which shift the ground state away from classical Neel order. Monte Carlo results presented here correctly reproduce the critical exponents of the heat capacity and the general lineshape of the susceptibility, and can be used to infer bounds on the DM interaction parameters.

In iron jarosite, magnetic spins associated with the iron atoms form a kagome lattice with antiferromagnetic coupling in plane and weak ferromagnetic coupling between planes. The local environment of the sites is that of an octahedral OH cage, canted away from the interlayer direction. This asymmetry permits a DM interaction and anisotropic self-energy which has axial anisotropy along the axis of the octahedral cage, canted off the z axis. The direction of the DM vector is given by the Moriya's symmetry rules.[i] The total Hamiltonian is thus given by

$$\mathcal{H} = \sum_{\alpha,\langle ij\rangle} J\, S_i^\alpha \cdot S_j^\alpha + \sum_{\alpha,\langle ij\rangle} D_{ij} \cdot \left(S_i^\alpha \times S_j^\alpha\right) + \sum_{\alpha=1}^{N-1} \sum_{\langle i^\alpha j^{\alpha+1}\rangle} S_i^\alpha \cdot S_j^{\alpha+1} + E \sum_{\alpha,i} \left(S_{\alpha i}^{z'}\right)^2 + D \sum_{\alpha,i} \left(S_{\alpha i}^{x'}\right)^2 + \left(S_{\alpha i}^{y'}\right)^2$$

where the sum over α is over layers and <ij> denote nearest neighbors; in the final interlayer sum, the nearest neighbors are calculated between layers, which are shifted such that a triangle Δ on layer *i* is followed by a triangle centered in the same place oriented as ∇ on layer *i+1*.[ii] Note that since the DM bond is antisymmetric with respect to exchange of spins, there is a definite chirality to the sum over bonds. The self-energy term is calculated with respect to the canted axis of the octahedral OH cage; this local coordinate system is denoted by a primed variable in the above sum.

Previous work on iron jarosite has found critical behavior consistent with both U(1) and Z_2 order parameters; however, the only symmetries respected by the Hamiltonian is lattice translation and time reversal.[iii] Neutron scattering measurements of the magnetic form factor support a magnetization pattern where below T_N the spins are pinned to point along the line from the site toward the center of each triangle on the kagome lattice; this would be a *n=1,d*=2 model.[iv] At temperatures greater than T_N, the form factor shows that the magnetization is approximately fixed in the plane of the kagome lattice, leading to a *n=2, d*=2 model. Finally, at higher temperatures when the interactions forcing the spin into the plane are small compared with the temperature, we expect *n=3,d*=2 scattering from a kagome plane. One further complication is nontrivial coupling between planes; at low temperatures, neutron scattering indicates that the magnetization is coherent across planes, while at T_N=65K this coherence is lost.[v] So below T_N, the more appropriate model would be *d*=3, while above it would be *d*=2. We expect that some of these cases will exhibit topological order; previous work has matched the observed correlation length with the Kostless-Thouless BKT prediction with good success.[vi] This complication motivates our use of Monte Carlo calculations, where explicit specification of the order parameter and dimensionality is not necessary. The hierarchy of interactions is expected to be $J>>D_{ij}\approx J_{inter}>E\approx D$.

In this calculation we neglect the anisotropic self energy terms E and D, since they are obtained from higher orders of perturbation theory than DM. Presence of DM with nontrivial components D_P perpendicular to the bond and D_Z perpendicular to the layer in conjunction with J_{inter} is still sufficient to break U(1) symmetry, so this is expected to be of the same universality class as the system under inclusion of anisotropic self-energy.

Heat capacity data collected on a single crystal sample of iron jarosite collected by Matan et al is shown in Figure 2.[vi] The heat capacity is shown with a phonon background subtracted; this background is collected by measurement of nonmagnetic gallium jarosite and subtracted out as detailed previously. Data at reduced temperature below $t = 0.01$ is strongly influenced by the choice of critical temperature; since the actual sample likely has local disorder and a distribution of critical temperatures, the deviation of the critical exponent here to values below 0.5 is likely not a reflection of new physics at a lower energy scale. The transition from α=0.5 to α=2 may be indicative of crossover phenomena.

Susceptibility data, shown in Figure 3, is calculated by applying a 3T field either perpendicular to the Kagome plane or parallel to it. Critical exponents are not fit since M/H is expected to differ from χ; agreement with the Mote Carlo results

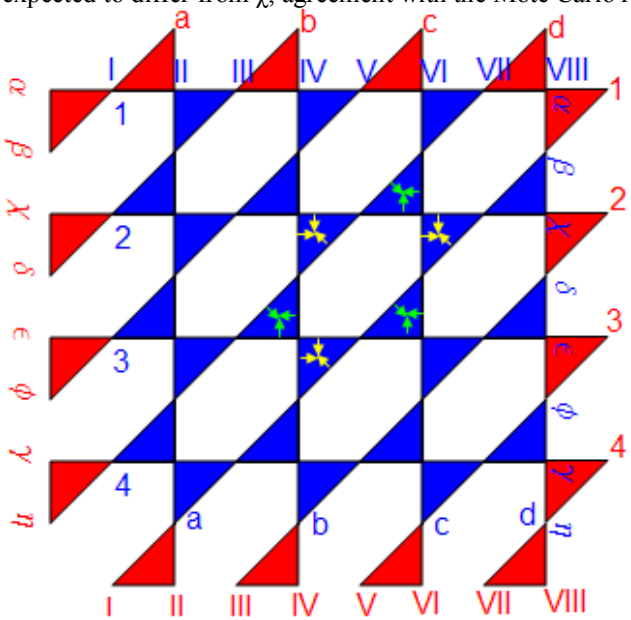

Figure 1: n = 3 layer with 48 Fe sites (corners of blue kagome lattice); virtual sites used in boundary conditions are shown in red. Note upper left, upper right, and bottom right corners are all Fe sites. Induced changes in lattice topology are all at the scale of the length of the lattice. DM vectors shown in green $D_Z>0$, yellow for $D_Z<0$.

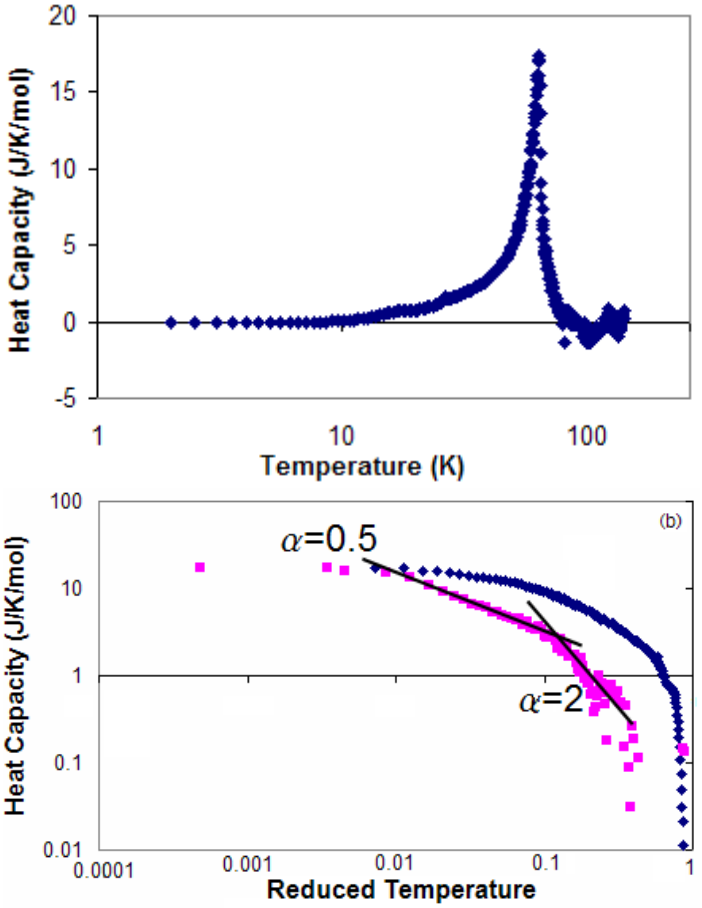

Figure 2: Heat Capacity as a function of temperature (a) and reduced temperature (b). Purple squares indicate data for $T>T_N$, blue diamonds for $T<T_N$. Black lines are critical scaling law behaviors, $C_V \propto t^{-\alpha}$ (courtesy Dr. Matan)

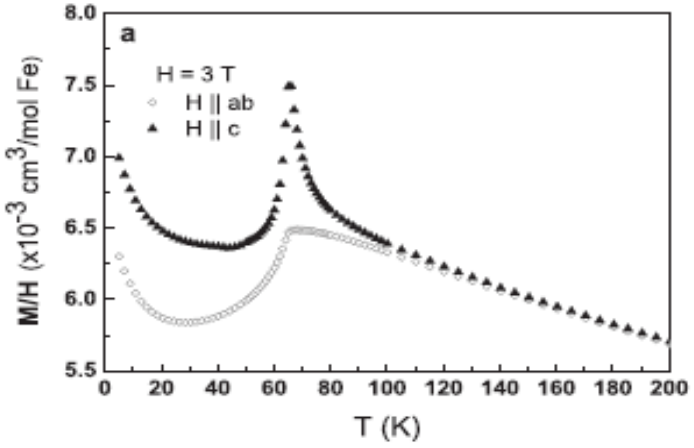

Figure 3: M/H measured with field applied perpendicular and parallel to Kagome plane (courtesy Dr. Matan).

is surprisingly good considering this difference. Note the low T rise is a Curie Tail; we expect local impurities to contribute to the susceptibility as M~1/T. Since our lattices had no impurities, we do not expect to reproduce this feature. Experimentally, we can measure sublattice magnetization through a measure of the intensity at the Bragg peak, which via the Fluctuation-Dissipation theorem can be related to the spin-spin correlation function, which is itself proportional to magnetization squared. This measures the ordered moment, which is not the appropriate quantity to scale as moment.

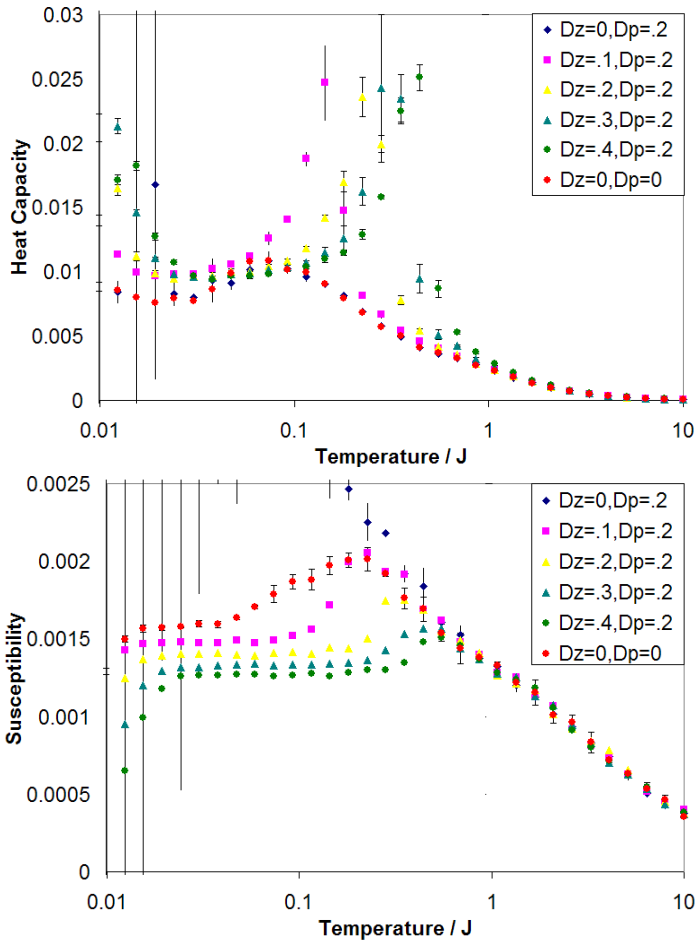

Figure 4: (a) Heat capacity and (b) susceptibility for varying DM values. Relevant parameters are angle step size 10^o and 48 sites in the lattice; the calculation is done over $5*10^6$ Monte Carlo steps for both relaxation and measurement. The rise in C_V and drop in χ below T/J= 0.05 is due to gapping out of interactions due to quantization of the angle of the spin. Reducing the minimum angle step to 2.5^o and increasing the lattice to 108 atoms (runtime increase to $2.4*10^8$ steps) removes low temperature feature but converges to the curve shown for T/J > .05. Equality suggests the parameters are sufficiently large to be independent of the value and greater than the coherence length.

We can view this several ways. First, consider a two parameter Hamiltonian producing critical scaling with exponent α. Imagine adjusting one of the parameters a small amount. It is feasible that this change produces a kink in the scaling law, so that in a local region determined by the size of the perturbation, the critical exponent is a different constant *a*, while outside this region the behavior returns to scaling as α. The position of this kink would indicate a phase transition. Consider a trajectory along the boundary of this transition; there may be a divergent tricritical point along this walk. Previous work on frustrated triangular lattices with tricritical behavior has reproduced some of the better predictions for critical exponents.[vii] The tricritical point can be phrased in mean field theory as resulting from a fixed point with greater than one unstable direction. Thus we are lead to consider the topology of the multiparameter space (J, J_{inter}, D_P, D_Z) under RG. While such nonlinear sigma model calculations are outside of the scope of this work, the RG picture provides a useful framework for thinking about crossover phenomena and potential corrections to the critical scaling of thermodynamic quantities. Previous Monte Carlo work on fixed

D_Z/J, D_P/J	T_N/J	α
0, 0	0	0.50
0.1, 0	0.16	0.50
0.2, 0	0.27	0.59
0.3, 0	0.36	0.58
0.4, 0	0.43	$0.55^{<}$,$0.73^{>}$
0, 0.2	0	0.50
0.1, 0.2	0.15	0.49
0.2, 0.2	0.26	0.55
0.3, 0.2	0.35	0.59
0.4, 0.2	0.41	$0.48^{<}$,$0.80^{>}$

Table 1: Values of the critical temperature and heat capacity exponent for various values of DM parameters in 2D with angle step size 10^o and 108 sites in the lattice; the calculation is done over $1.2*10^8$ Monte Carlo steps for both relaxation and measurement. Symbols < and > denote fits to $T<T_N$ and $T>T_N$ separately, which are not statistically advantageous in runs other than for $D_Z = 0.4$.

point structure and critical exponents in the presence of DM for a kagome lattice has focused on single layer simulations.[viii]

We present several Monte Carlo calculations: single layer and 3D, with varying DM interactions. For a single layer with varying D_{ij}, we produce figure 4. From this, we can identify the peak temperature as approximately T/J=|D|. Higher-statistic simulations around T/J between 0.05 and 0.65 allow us to identify the critical exponent associated with the transition. These values are given in table 1. Fits were performed over the region excluding a temperature interval around the peak of width 0.06, to avoid restricting the critical scaling behavior to the induced finite sample size error. For D_Z=0.4, D_P=0, we were unable to fit to the same scaling exponent on both sides of the critical transition. This is consistent with the observed differences between the behavior above and below the transition. It is possible that the underlying physics is of unbinding of topological defects as discussed in terms of correlation length in previous works.[vi]

It is important to note that the observed value of α=0.5 is entirely consistent with our simulations in a range of DM parameters, unlike previously reported Monte Carlo work on triangular antiferromagnets or applications of known universality classes for various values of *n*, *d*.[ix] α=2 behavior is obtained in the asymptotic limit of the heat capacity in our simulations, between T=3J to 10J with respect to a T=0 critical point. However, since D_{ij}<<J, we expect that this temperature scale would be at T_{sample}>>T_N, which we associate with D_{ij}. We expect that the dynamics below the interplane decoherence temperature will involve switching of correlatelated regions. Thus it can be argued that the effective interaction is scaled by a monontonic function of the coherence length as clusters of coherent sites interact across layers. If we argued that the temperature of the transition was actually proportional to Dij times a cluster size, we could argue that at higher temperatures, the clusters had decohered, the interaction had weakened, and the temperature would shift down. Our Monte Carlo simulations are desensitized to coherent effects due to the small lattice size, so it is possible this feature is missed in the Monte Carlo simulations. It is also possible this is purely an experimental error due to subtraction; note that in the subtracted version of the heat capacity, we have a large negative region, clearly a nonphysical artifact of attempting to equate phonon modes in Ga jarosite with those in Fe jarosite. Such error will undoubtedly have effect at lower temperatures, and could easily cause the observed dependence. It is more interesting to hope that such behavior is a crossover phenomena, where as we reduce temperature we move into a regime where the RG flow is toward a T=0 critical point. From crossover transition theory, we know that it is possible to obtain the infinite temperature limit at arbitrarily small temperature, since the relative quantity is '$x=h/t^{\Delta}$', where *h* is a generalized parameter and Δ is the appropriate scaling. As temperature and field both go to zero, it is possible to obtain either the $x=0$ or $x=\infty$ limit, depending on how *h* vanishes. We suggest that the α=2 behavior may be due to a similar process in the multiparameter space where $h \in \{h, D_P, D_Z, J_{inter}\}$. While RG is a purely mathematical construct for understanding the fixed point structure, it is useful to phrase the result in terms of RG to understand the characteristics of the model Hamiltonian which might produce this behavior.

The susceptibility in 2D shows qualitative agreement with that of M/H measured in plane, but we see no difference in susceptibility in and out of the plane with only one Kagome layer. This motivated our examination of a 3D lattice. Results for heat capacity in 3D are summarized in Figure 5. Susceptibility in absence of a field appeared qualitatively similar

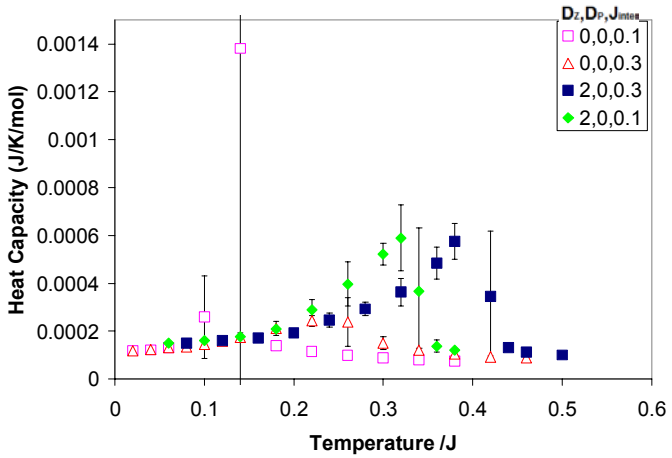

Figure 5: Heat capacity for angle step size 5^o, 363 sites in plane, 20 layers; simulation with between $2*10^8$ to $4*10^8$ steps depending on convergence. Inset table shows the associated D_Z, D_P, and J_{inter} values for each data set.

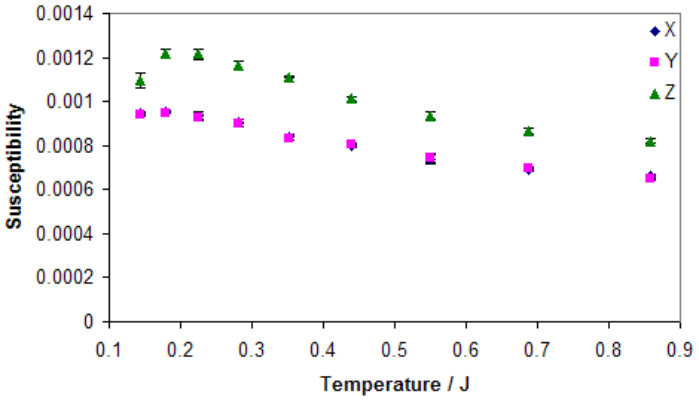

Figure 6: Susceptibility measured along X, Y, and Z for D_{ij} = 0, J_{inter} = -0.1. Monte Carlo simulation done with angle step size 10^o with 48 sites per layer and 4 layers over $3*10^7$ steps for both relaxation and measurement. Similar results are obtained with nontrivial DM vectors. Unlike 2D lattices, the susceptibilities are anisotropic around the peak.

to what was obtained in 2D, but showed directional anisotropy favoring χ_Z over χ_X or χ_Y near the transition temperature T_N. Data illustrating this difference is presented in Figure 6. It is important to note that although we have changed from *d*=2 to d=3, the heat capacity is unaffected except for when D_{ij}=0, illustrating that the dynamics are predominantly in plane, or in the RG language, that we are flowing into a J_{inter}=0 fixed point. Experimentally, it has been observed that below the temperature. The susceptibility shown in Figure 6 becomes anisotropic at a temperature scale that is greater than J_{inter} in the absence of DM interactions. This factor shifting the transition from J_{inter} to a temperature scale several times J_{inter} likely arises from dynamics governed by correlations between coherent regions of layers. If this is the case, our small lattice may be significantly limiting the critical scaling, which may explain why we do not observe the very sharp measured peak in susceptibility along Z.

Our Monte Carlo studies have reproduced the measured heat capacity scaling exponent α=0.5, unlike previous theoretical work modeling this system as either $XY \times Z_2$, Heisenberg spins, or a stacked triangular lattice.[ix] A possible mechanism for α=2 arising via crossover phenomena from a T=0 fixed point has been discussed, although it is unclear if the experimental data is truly representative of this critical exponent. Susceptibility lineshapes measured in the plane are in good agreement with simulation results, while measurements normal to the plane show anisotropies, but are missing a well defined peak. An explanation for the difference in critical behavior is that the susceptibility is a direct measurement of a correlation, and the limited lattice size may show up here to a stronger degree than it affects the heat capacity.

A primary direction for further application of this simulation is in measuring correlations. Testing correlations in plane would provide insight into if the observed correlation length scaling law is due to Kostless-Thoules vortices, as has been previously suggested.[vi] Interplane correlations should reproduce the decohering transition between layers observed in neutron scattering. Preliminary results here do observe decays in correlation as a function of temperature, but the variance is very high and was not seen to diminish as the run length increased. Current statistics do not permit distinguishing between a power and exponential decays as a function of tempreature, which we would expect below and above the unbinding transition. In this system the lattice is tripartite; we examined measuring both correlations to all atoms and only the same of element of the basis, and neither showed clear decay as a function of position with the current lattice sizes of 48 and 108 atoms. It is possible that these behaviors are only true well outside the core, and we need to scale the lattice size up substantially before we can see these mean field results.

Visualization of the spin configuration as a function of Monte Carlo step could illustrate spin dynamics, which are expected to show both spin wave and vortex like behavior. Finally, we could approach more accurate values of D_{ij}, J_{inter}; we expect these may be on the order of 0.01 J; to probe such scales, we need to greatly reduce the gap in excitations by increasing the number of allowed values of angle each spin can have. Doing so will likely amount to a large increase in runtime; one way to combat further runtime increases is to take better advantage of the symmetry of the problem and change the axis of rotation locally to that of the octahedral cage. This could allow our model to more naturally represent spin wave excitations. Testing for the presence of topological defects or singularities is another avenue for further research; since much of this was done with a small lattice, it is likely that we need to scale up the lattice so that the core energy would not dominate. The largest lattice ever tested here was for 363 points in each kagome plane with 20 such planes, but an insufficient number of data points could be obtained to draw conclusions about vortex structures.

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