A metamagnetic critical point in a three dimensional frustrated antiferromagnet

Nic Shannon,1,2 Karlo Penc,3 and Yukitoshi Motome4

1 Max-Planck-Institut für Physik komplexer Systeme, Nüthnitzer Str. 38, 01187 Dresden, Germany
2 H. H. Wills Physics Laboratory, University of Bristol, Tyndall Ave, BS8-1TL, UK.
3 Research Institute for Solid State Physics and Optics, H–1525 Budapest, P.O.B. 49, Hungary
4 RIKEN (The Institute of Physical and Chemical Research), Wako, Saitama 351–0198, Japan

(Dated: March 23, 2022)

The competition between different forms of order is central to the problem of strong correlation. This is particularly true of frustrated systems, which frequently exist at or near to a zero–temperature critical point. Here we show that a state with a half–magnetization plateau but no long range order can arise when a three dimensional frustrated antiferromagnet is tuned to a critical point bordering a metamagnetic state. We use classical Monte Carlo simulation and low–temperature expansion techniques to accurately characterize this “spin pseudogap” state, and show how its properties relate to those of the critical point. Our results provide an example of three dimensional spin model which can be used to study the relationship between gap and “pseudogap”, — i.e. long range and preformed local order — near a metamagnetic critical point.

PACS numbers: 75.10.-b, 75.10.Hk 75.80.+q

In recent years the concept of “quantum criticality” has become central to efforts to understand correlated electron systems. This rapidly growing body of work rests on the simple idea that where there is a zero–temperature phase transition between two different ordered phases, the finite temperature properties of the paramagnetic phase are controlled by the critical point which separates them. In order to test this hypothesis experimentally, it is necessary to identify systems with a suitable control parameter which can be used to tune through the critical point. Mechanical pressure, chemical pressure, doping and magnetic field have all been used successfully to this end. Two widely discussed examples are the underdoped cuprate superconductors, whose “pseudogap phase” has been suggested to originate in a quantum critical point as a function of doping, and Sr3Ru2O7 where non–Fermi liquid behavior appears to be associated with a metamagnetic transition with strongly suppressed Tc.

Theoretical attempts to understand these phenomena have largely concentrated on renormalization group analysis of phenomenological field theories. However it is also interesting to ask what happens in microscopic models, especially where these are accessible to a variety of different approaches. Can we construct concrete examples of systems with a zero–temperature critical point? What are the nature of the correlations at this point, and in the paramagnetic phase connected to it? What does a “pseudogap” look like at a microscopic level?

In this Letter we show how a state with a gap to spin excitations — a “spin pseudogap” — but no long–range magnetic order, can arise near a metamagnetic critical point in a microscopic model. Our main results are summarized in the phase diagram Fig. 1. The model which we use to explore these ideas is the antiferromagnetic (AF) Heisenberg model on the highly frustrated pyrochlore lattice, in applied magnetic field h:

\[
\mathcal{H} = J \sum_{\langle i,j \rangle} \left[ \mathbf{S}_i \cdot \mathbf{S}_j - b (\mathbf{S}_i \cdot \mathbf{S}_j)^2 \right] + J_3 \sum_{\langle i,j \rangle''} \mathbf{S}_i \cdot \mathbf{S}_j - h \sum_i S_i^z. \tag{1}
\]

In order to make the problem accessible to large scale simulation, we consider the classical limit of the problem...
$S = |S| \rightarrow \infty$. The dominant effect of quantum fluctuations in frustrated systems is known to be a tendency towards collinearity, and we characterize this through an effective biquadratic interaction $b$.

An additional third–neighbour interaction $J_3$ is used to tune the system away from a critical point at $J_3 = 0$. The model was recently shown to offer an explanation of the broad half–magnetization plateaux observed in the spinel oxides CdCr$_2$O$_4$ and HgCr$_2$O$_4$. In these materials, strong effective biquadratic interactions $b \sim 0.1–0.2$ arise from the coupling of spins to the lattice.

For simplicity, we focus below on the limiting case of ferromagnetic (FM) $J_3 \rightarrow 0^-$. For FM $J_3$, the model exhibits $q = 0$ four–sublattice long–range magnetic order at low temperatures. The most dramatic feature of its zero–temperature phase diagram is a first order transition in applied field into a metastable “plateau” state with $m = 1/2$ — exactly half the saturation magnetization — protected by a gap to the lowest lying spin excitation. This state is of the type illustrated in Fig. 2(a).

We have performed extensive Monte Carlo (MC) simulations of Eq. 1 at finite temperatures, complemented by classical low–temperature expansions about both ordered and disordered states. These confirm that the phase diagram given in Fig. 2 remains valid up to a transition temperature $T_c$ whose energy scale is set by $|J_3|$. However for $|J_3| \rightarrow 0$, the transition temperature for each of the different ordered phases vanishes and the system exists at a critical point.

So much for long range order — what about metamagnetism? In Fig. 3 we illustrate the magnetization process of Eq. 1 for $J_3 = 0$. Data are taken from MC simulations of Eq. 1 using a Metropolis algorithm with local spin update. For convenience we assume a (large) value of $b = 0.6$, and work in units such that $J = S = 1$. It is clear from Fig. 3(a) that the magnetization plateau is alive and well — in fact from the magnetization process alone it is essentially impossible to distinguish these results from those for the four–sublattice ordered phase for FM $J_3$. The strong suppression of the magnetization susceptibility at low temperatures shown in Fig. 3(b) suggests the existence of a well defined gap, of similar magnitude to that in the nearby ordered phase. How should we reconcile these results with the vanishing transition temperature for the ordered plateau state at $J_3 = 0$?

In Fig. 4 we present MC results for the reduced spin–spin correlation function

$$Q(r_{ij}) = \langle S_i \cdot S_j \rangle - \langle m \rangle^2,$$

and the measure of collinearity

$$P(r_{ij}) = \frac{3}{2} \left[ \langle (S_i \cdot S_j)^2 \rangle - \frac{1}{3} \right].$$

As shown in Fig. 4(a), spin correlations exhibit a liquid–like structure. After an initial AF oscillation on the scale of a near–neighbour they decay very rapidly to zero. Within the range accessible to MC simulation, the envelope for this decay appears to cross over smoothly between an exponential decay at high temperatures and a $1/r^3$ power–law behavior at low temperatures, as shown in Figs. 4(b) and (c). The collinearity $P(r_{ij})$, on the other hand, does exhibit long range order — see Figs. 4(d) and (e).

We can understand these results as follows — the biquadratic interaction $b$, which we have introduced to characterize quantum effects, favors collinear states. The pyrochlore lattice is corner–sharing network of tetrahedra. For $h \simeq 4$, the $b$ term acts to select $uuud$ states with exactly three up and one down spins in each tetrahedron and, consequently, $m = 1/2$. The $q = 0$ ordered phase [Fig. 2(a)] is an example of such states. But there are many more — in fact there is a one–to–one mapping between $uuud$ states on a pyrochlore lattice and the $\sim 1.3N^2$ dimer coverings of the dual diamond lattice.

Exactly at the critical point, for $T = 0$ and $J_3 = 0$, the system exists in an equally weighted superposition of

FIG. 2: (Color online) Half–magnetization plateau states $(uuud$ states) on a pyrochlore lattice with exactly three up (black) and one down (white) spins per tetrahedron. (a) $uuud$ state with long–range four–sublattice order, as considered in \[4\]. (b) A schematic picture of $uuud$ state with no long range order of the type found at the critical point.

FIG. 3: (Color online) Dependence of (a) magnetization $m$ and (b) uniform magnetic susceptibility $\chi$ on magnetic field $h$ for $b = 0.6$ and $J_3 = 0$, showing the existence of the magnetization plateau in the absence of long–range magnetic order. Symbols (lines) denote the data for $L = 16$ ($L = 8$). The dashed lines show the $T = 0$ results for long–range four–sublattice order.
FIG. 4: (Color online) Absence of long–range magnetic order in the spin–pseudogap state for \( b=0.6, J_3 = 0 \) and \( h=4 \).

(a)—(c) The reduced spin correlation function \( Q(r) \), defined by Eq. (2), decays exponentially at high temperatures, crossing over to a power law behavior \( Q(r) \sim 1/r^3 \) at low temperatures. (d), (e) However the measure of collinearity \( P(r) \), defined by Eq. (3) does exhibit long range order at low temperatures. All the results are calculated along [110] chains for \( L = 16 \). Distances are measured in units of the nearest–neighbor bond length.

We find that the spectrum is always bounded above and below by the same gaps \( \Delta_2 \) and \( \Delta_1 \), which are set by the same nodeless excitations. In the four–sublattice order.

duced \( uuud \) state these reduce to the highest and lowest energy excitations with \( q = 0 \), respectively. One of the zero modes survives at finite \( b \) as a non–dispersing excitation at finite energy. Variation in entropy between different \( uuud \) states is negligible compared with the entropy of the \( uuud \) manifold, and so fluctuations alone cannot drive the system to order. The insensitivity of the excitation spectrum to the type of \( uuud \) state considered explains why the thermodynamic properties of the disordered phase for \( J_3 = 0 \) — in particular its half–magnetization plateau — are so similar to those of the ordered phases for finite \( |J_3| \).

We can go further and calculate the asymptotic decay of spin correlations in the region of the critical point. We do this by constructing an effective electrodynamics for the \( uuud \) “dimer” manifold in close analogy with a recent treatment of the \( uudd \) “ice” manifold [8]. In this approach, the entropy term \( T \ln |\mathbf{M}|/2 \) is written in terms of a course–grained polarization field \( \mathbf{P} \) which measures deviations from \( uuud \) order. We find that the spin correlations in the \( uuud \) manifold decay as \( 1/r^3 \), just like those of the \( uudd \) manifold. This result is in perfect agreement with the results of our MC simulation at low temperatures — see Fig. 4(c).

Thus the situation in the immediate vicinity of the zero–temperature critical point for \( J_3 = 0 \) is fairly clear. A power law behavior of spin correlations coexists with a finite gap to spin excitations and a well defined plateau in the magnetization process. Since this gap is not associated with long range order — at least in any conventional sense — we adopt the terminology “spin pseudogap” to describe this state.

But how does this “spin pseudogap” state emerge from the paramagnet at high temperatures? And how does it relate to the state for FM \( J_3 \) with long–range magnetic order? To answer these questions, we introduce mea-
group irreducible representation of the tetrahedral symmetry

...sures of both local and global order, based on the relevant
irreducible representation of the tetrahedral symmetry group $T_d$:

$$
\lambda^\text{local}_{T_2} = \frac{2}{N} \sum_{\text{tetra}} \Lambda^2_{T_2}, \quad \lambda^\text{global}_{T_2} = \frac{2}{N} \left[ \sum_{\text{tetra}} \Lambda_{T_2} \right]^2,
$$

where, following Eq. (5), $\Lambda_{T_2} = (\Lambda_{T_2,1}, \Lambda_{T_2,2}, \Lambda_{T_2,3})$, and $\Lambda_{T_2,1} = (S_2 \cdot S_3 - S_1 \cdot S_4)/\sqrt{2}$, etc., are defined in terms of the spins on an individual tetrahedron. We also consider the related susceptibilities, $\chi_{\lambda}(T) = [(\lambda^2) - (\lambda^2)]/T$.

In Fig. 6 we show MC results for the specific heat, local and global “order” parameters $\lambda_{T_2}$, and their related susceptibilities. A broad peak in the specific heat is observed at $T \approx b$, with most of the entropy coming from the selection of collinear states. At the same time, the measure of local order $\lambda^\text{local}_{T_2}$ increases rapidly, and the related susceptibility $\chi^\text{local}_{T_2}$ shows a broad peak. However none of this signals the onset of long–range four–sublattice $uudd$ order of the type found for FM $J_3 < 0$ — the relevant order parameter $\chi^\text{global}_{T_2}$ tends to zero in the thermodynamic limit, and the related susceptibility $\chi^\text{global}_{T_5}$ appears to diverge only as $T \to 0$.

The peak in $\chi^\text{local}_{T_2}$ or, equivalently, the peak in the specific heat $c$ defines a crossover temperature $T^*$ for entering the spin pseudogap state, with its associated magnetization plateau. The crossover also corresponds to an emergence of the power law behavior in spin correlations and the rapid development of collinearity in Fig. 6. $T^*$ is set by $b$ — in fact, we have confirmed that for $b < J$, it scales nearly linearly with $b$. Meanwhile, the global order parameter monitors a transition for long–range ordered phase for finite $J_3$. By tracking both $T^*$ and $T_c$ as a function of $J_3$, we can map out the phase boundary of the true, long–range ordered plateau phase, and the extent of the finite–temperature spin pseudogap “phase” associated with the critical point at $J_3 = 0$. Results for $b = 0.6$ and $h = 4$ are shown in Fig. 4. In the vicinity of the critical point, we can also estimate $T_c$ for this (1$^\text{st}$ order) transition within the low–temperature expansion by comparing the change in internal energy with the entropy change going from the gapped to the pseudogapped states: The prediction is $T_c \sim 30|J_3|$, the dashed line in Fig. 1. This phase diagram summarizes the relationship between gap and “pseudogap”, or order and disorder, near a metamagnetic critical point.

For small $|J_3|$ where $T_c < T^*$, the system exhibits a magnetization plateau in the absence of long range order. As the system is cooled from the spin–pseudogap state into the ordered phase, the preformed local order in $\Lambda_{T_2}$ becomes global order in $\sum_{\text{tetra}} \Lambda_{T_2}$. This provides an intriguing magnetic analogy for the way in which preformed superconducting order is believed to emerge from preformed local cooper pairs in underdoped cuprates.

So far as Cr spinels are concerned, our results suggest that a material which exhibits a magnetization plateau while maintaining the full (cubic) symmetry of the pyrochlore lattice, need not be magnetically ordered [10].

In the light of our results it also seems highly probable that the spin–1/2 Heisenberg model on a pyrochlore lattice exhibits a half–magnetization plateau because of strong quantum fluctuations. Within a spin–wave approximation, our calculations show that the spin gap protecting the plateau state is again finite and set by a nodeless wave function, and so is insensitive to whether the system is ordered or disordered.

We conclude by noting that it is possible to construct a phase diagram similar to Fig. 1 for Eq. (1) near critical point with $J_3 = 0$ and $h = 0$. In this case the competing ground states belong to the $uudd$ “ice” manifold, and the correlations at the critical point are of the spin–liquid form described in [11]. However there are two important distinctions from the case of $h = 4$. Firstly, neither ordered nor disordered $uudd$ phases possesses a gap to spin excitations. And secondly, for $h = 0$, the collinearity Eq. (6) is a well defined nematic order parameter.

While we have been unable to construct any local object which can serve as order parameter for the spin–pseudogap state for $h = 4$, we cannot rule out the possibility that $T^*$ signals the onset of some exotic non–local order [12]. This remains as a subject for future investigation.

Acknowledgements It is our pleasure to acknowledge stimulating discussions with T. Momoi, H. Takagi, O. Tchernyshyov, H. Tsunetsugu and M. E. Zhitomirsky. This work was supported under a Grant–in–Aid for Scientific Research (No. 16GS050219) and NAREGI from the Ministry of Education, Science, Sports, and Culture of Japan, Hungarian OTKA T049607 and SFB 463 of the DFG.
[1] S. A. Griegera et al., Science 294, 329 (2001).
[2] See e.g. “Quantum Phase Transitions”, S. Sachdev, Cambridge University Press (2001).
[3] C. L. Henley, Phys. Rev. Lett. 62, 2056 (1989).
[4] K. Penc, N. Shannon, and H. Shiba, Phys. Rev. Lett. 93, 197203 (2004).
[5] H. Ueda et. al., Phys. Rev. Lett. 94, 047202 (2004); unpublished.
[6] Y. Motome, K. Penc, and N. Shannon, cond-mat/0506500; cond-mat/0506598.
[7] We typically perform $10^5$–$10^6$ MC samplings for measurements after $10^4$–$10^5$ steps for thermalization. System sizes are measured in units of the the 16 spin cubic unit cell, so that the total number of spins $N = 16 \times L^3$. Because of the frustration inherent in Eq. (1), the autocorrelation time becomes extremely large at low temperatures. Data are shown only where simulations have converged and are known to be ergodic.
[8] J. F. Nagler, Phys. Rev. 152, 190 (1966); J. Mat. Phys. 7, 1484 (1966).
[9] See e.g. C. L. Henley, cond-mat/0407005 also S. V. Izakov et al., Phys. Rev. Lett. 93, 167204, (2004); M. Hermel et al., Phys. Rev. B 69, 064404 (2004). In the case of the uuud manifold we must associate three vertex arrows with each down spin (and one with each up spin), in order to obtain a divergence–free field $P$.
[10] c.f. the classical Heisenberg model on a Kagomé lattice, M. E. Zhitomirsky, Phys. Rev. Lett. 88, 057204 (2002).
[11] J. N. Reimers, Phys. Rev. B 45, 7287 (1992). R. Moessner and J. Chalker, Phys. Rev. Lett 80, 2929 (1998); Phys. Rev. B 58, 12049 (1998).
[12] X–G Wen, cond-mat/0508020.