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# Quadrupolar quantum criticality on a fractal

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We study the ground state ordering of quadrupolar ordered  $S = 1$  magnets as a function of spin dilution probability  $p$  on the triangular lattice. In sharp contrast to the ordering of  $S = 1/2$  dipolar Néel magnets on percolating clusters, we find that the quadrupolar magnets are quantum disordered at the percolation threshold,  $p = p^*$ . Further we find that long-range quadrupolar order is present for all  $p < p^*$  and vanishes first exactly at  $p^*$ . Strong evidence for scaling behavior close to  $p^*$  points to an unusual quantum criticality without fine tuning that arises from an interplay of quantum fluctuations and randomness.

## I. INTRODUCTION

Quantum spin models with random vacancies provide a rich playground for the study of the combined effects of strong interactions and quenched disorder that are relevant to experimental measurements on a number of doped magnetic alloys.<sup>1</sup> Magnetic order in higher dimensions is generally stable to a small concentration of vacancies. On the other hand, if its moments are diluted beyond the percolation threshold, the system breaks up into zero-dimensional clusters and hence magnetic order must be lost. What is the nature of the magnetic quantum phase transition as the dilution is varied?

The answer to this question is most thoroughly understood for the randomly diluted transverse field Ising model.<sup>2</sup> The expected ground state phase diagram<sup>3</sup> as a function of the transverse field  $g$  and site dilution probability  $p$ , has three kinds of quantum phase transitions, Fig. 1. For  $g > g_c$  the transition takes place at a value of  $p$  smaller than the percolation threshold  $p_c$ , the critical point is then described by the random transverse field Ising model which possesses an infinite randomness fixed point<sup>4,5</sup>; the physics of percolation however plays no role (“R” in Fig. 1). For  $g < g_c$  on the other hand percolating clusters are magnetically ordered and the singularities at the transition are determined by those of classical percolation (“P” in Fig. 1)<sup>6,7</sup>. Finally exactly at  $g = g_c$  a critical point is obtained: Crucial to our discussion, quantum criticality at the percolation threshold (“C” in Fig. 1) requires *fine tuning* of the quantum fluctuations and hence “C” is not the generic transition for Ising magnets on dilution.

Another family of quantum spin models where this question has been addressed in detail are bipartite Néel ordered Heisenberg models. For random depletion of the square lattice it was found that percolating clusters have long range order,<sup>7–9</sup> the destruction is thus akin to Fig. 1 “P”. The surprising stability of the Néel order has been traced back to the effect of uncompensated Berry phases which result in “orphan” moments.<sup>10,11</sup> In the bilayer geometry, with bond dilution that circumvents random Berry phases, a phase diagram similar to Fig. 1 was found<sup>12,13</sup>, with quantum phase transitions corresponding to “P”<sup>14</sup> in some regions and corresponding to “R” in others, in contrast to the Ising case the critical phe-

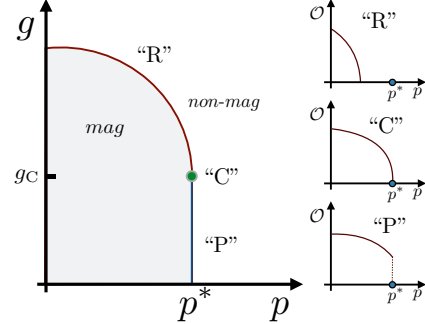


FIG. 1. Cartoon  $T = 0$  phase diagram for transverse field Ising model in the dilution probability  $p$  and transverse field  $g$  plane. For the largest cluster on a two-dimensional lattice occupied with probability  $p$ , the transition from magnetic to non-magnetic phases can be of three types as illustrated for the Ising order parameter  $\mathcal{O}$  vs.  $p$ : “R” the magnetic transition occurs before the percolation threshold and is hence expected to be identical to the random transverse field Ising model. “P” the percolation quantum transition where the magnet remains ordered at the percolation threshold. “C” at which the order parameter vanishes continuously at  $p^*$  achieved by fine tuning  $g$  to a special value  $g_c$ . For the  $S = 1$  quadrupolar magnets discussed here, we present detailed evidence that the order vanishes like “C”, but with no fine tuning, leading to an unusual quantum criticality on a fractal cluster.

nomena here is controlled by a finite disorder fixed point with conventional scaling.<sup>15</sup> The critical point “C” which requires fine tuning has also been studied.<sup>16</sup> A variety of phase diagrams and critical phenomena can be accessed by dilution of Néel magnets in geometries different from the single and bilayer.<sup>16,17</sup>

The two examples of dipolar ordered magnets (Ising and Heisenberg) make it clear that the details such as symmetry of the order parameter and Berry phases play a crucial role in determining how quantum magnetism is destroyed by random dilution. In this work we address the dilution transition in a different kind of system: quadrupolar ordered (also referred to as spin nematic) magnets in their most common  $S = 1$  realization.<sup>18</sup> The study of the quadrupolar phase of  $S = 1$  magnets has become increasingly popular motivated by their possible sighting in some triangular lattice Ni based magnets (see

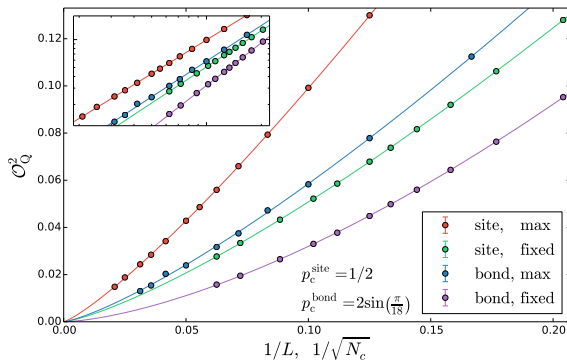


FIG. 2. Thermodynamic extrapolation of the  $T = 0$  quadrupolar order parameter  $\mathcal{O}_Q^2$  at the percolation threshold. We present results of simulations of the model Eq. (1) using both site diluted and bond diluted triangular lattices at their respective  $p^*$ . For each case we have carried out the averages for finite-size scaling in two ways: using an ensemble with a *fixed* cluster of size  $N_c$  or an ensemble of the largest *max* cluster on an  $L \times L$  lattice. In all four cases, we find  $\mathcal{O}_Q^2$  vanishes in the thermodynamic limit at  $p^*$ . The inset shows the same data on a log-log scale.

e.g.<sup>19–24</sup>). Experimental studies of Zn replacement of Ni in  $\text{NiGa}_2\text{S}_4$  provide a direct experimental motivation for the site diluted  $S = 1$  magnets we study here<sup>25</sup>. As we shall describe below, in contrast to what has been observed for Néel order in  $S = 1/2$  magnets and transverse field Ising models, we find here that quadrupolar order vanishes exactly at the percolation threshold *without any fine tuning*, resulting in new quantum critical behavior.

## II. MODEL

Just as the bilinear Heisenberg model is the archetype for realizing Néel order, the biquadratic Heisenberg interaction is the archetypical model system that realizes quadrupolar order in  $S = 1$  magnets. We shall consider the following Hamiltonian,

$$H = - \sum_{\langle ij \rangle} J_{ij} (\vec{S}_i \cdot \vec{S}_j)^2, \quad (1)$$

where  $\langle ij \rangle$  denote nearest neighbors in the triangular lattice, and  $\vec{S}_i$  are the  $S = 1$  Pauli matrices on the  $i^{\text{th}}$  site. The model possesses an explicit physical  $\text{SO}(3)$  internal spin symmetry. We use standard uncorrelated random site and bond dilution: For site dilution  $J_{ij} = |J|r_i r_j$  where  $r_i = 0$  with probability  $p$  and  $r_i = 1$  with probability  $1 - p$ . For bond dilution  $J_{ij} = |J|\chi_{ij}$  where  $\chi_{ij} = 0$  with probability  $p$  and  $\chi_{ij} = 1$  with probability  $1 - p$ . While site dilution is closer to experimental realizations such as Zn doped  $\text{NiGa}_2\text{S}_4$ , bond dilution provides an alternate access to the percolation threshold.

Our numerical results were obtained using stochastic

series expansion<sup>26</sup> with an efficient sign-problem free algorithm that has been described and tested previously<sup>27</sup>. We calculate averages on finite size clusters at finite-temperature,  $T$ . To study the quadrupolar order in the magnet we define the quadrupolar order parameter through the equal time two point correlation function of the quadrupolar order parameter.

$$\mathcal{O}_Q^2 = \langle \frac{1}{N_c} \sum_{a,i,j} \langle \hat{Q}_i^{aa} \hat{Q}_j^{aa} \rangle \rangle_p, \quad (2)$$

where the quadrupolar order parameter is defined in terms of the local  $S = 1$  Pauli matrices as  $\hat{Q}^{ab} = \frac{\hat{S}^a \hat{S}^b + \hat{S}^b \hat{S}^a}{2} - \frac{2}{3} \delta^{ab}$ ,  $N_c$  is the size of the cluster,  $\langle \dots \rangle$  is the quantum mechanical average over the thermal density matrix and  $\langle \dots \rangle_p$  is a disorder average over realization of the ensemble of clusters.

## III. NUMERICAL RESULTS

It is now well established that the model Eq. (1) without any depletion ( $p = 0$ ) has quadrupolar order with ferromagnetically aligned directors<sup>22,27,28</sup> (i.e.  $\mathcal{O}_Q^2$  in Eq. 2 is finite in the ground state in the thermodynamic limit). We begin our numerical study by asking the following question: As  $p$  is increased does the quadrupolar order parameter eventually vanish like “R”, “C” or “P” (referring to Fig. 1). As we have discussed in the introduction, generic expectations would be either “R” or “P”.

In Fig. 2 we show extrapolations of the quadrupolar order parameter to the thermodynamic limit at the percolation threshold. We define two different ensembles for finite-size scaling which allow us to approach the thermodynamic limit in two different ways, with fixed cluster size  $N_c$  (on an infinite underlying lattice) and the largest cluster on a finite  $L \times L$  lattices, following<sup>8</sup>. We have also studied both bond and site dilution at the percolation threshold. Consistently across all finite-size scaling schemes we find that the order parameter vanishes at the percolation threshold. For a similar scaling analysis for the Néel order, see Fig. 10 in Ref. 8 which clearly shows the ordering of  $S = 1/2$  Heisenberg model on the percolating cluster in two dimensions. In contrast here we find the quadrupolar order vanishes in the thermodynamic limit. This data clearly eliminates the “P” possibility since this requires the percolating clusters to be magnetically ordered. We have taken great care to make sure our data is equilibrated and in the limit of  $T = 0$ . The ground state limit requires extraordinarily low temperatures because of the weak links that connect percolating clusters. Details are provided in the Appendix.

Now that we have shown that the percolating cluster are magnetically disordered, the generic expectation is that the quadrupolar order vanishes before the percolation threshold is reached at some  $p < p^*$  as illustrated for “R” in Fig. 1. To address this question quantitatively

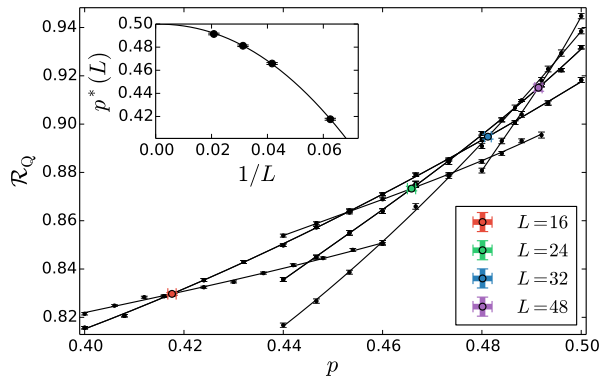


FIG. 3. Crossing of the Binder ratio  $\mathcal{R}_Q$  for the site diluted triangular lattice, demonstrating that the order parameter vanishes first at  $p = p^*$  with high precision. The main panel shows a zoom in of  $\mathcal{R}_Q$  as a function of the site occupation probability  $p$  close to the percolation threshold. The crossing points between  $(L/2, L)$  pairs are marked with errors. Inset shows the  $p(L)$  value at the crossings of  $(L/2, L)$  pairs. The crossing data extrapolates accurately to the percolation threshold of the site diluted triangular lattice,  $p^* = 0.5$  with a power law fit. This numerical evidence establishes that the quadrupolar order parameter is finite for all  $p < p^*$  and vanishes first exactly at the percolation threshold as illustrated in case “M” in Fig. 1.

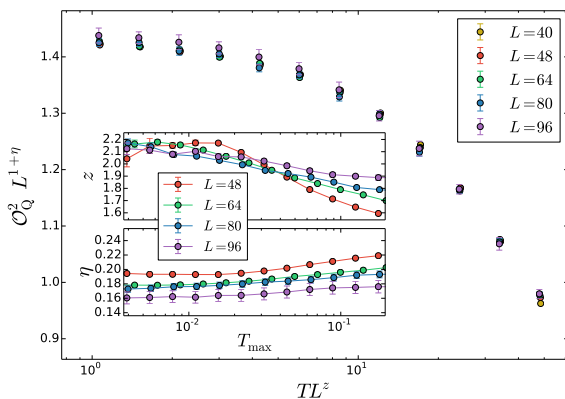


FIG. 4. Finite temperature scaling collapse of the quadrupolar order parameter at  $p = p^*$ . Here we are testing the quantum critical scaling form  $\mathcal{O}_Q^2 = \frac{1}{L^{1+\eta}} \mathcal{F}(L^z T)$ . The main panel shows the collapse of the order parameter at  $p = p^*$ , using the values  $z = 1.987$  and  $\eta = 0.178$  for the exponents. The inset illustrates the finite size corrections and convergence of the critical exponents: the values of  $z$  and  $\eta$  obtained from pair-wise collapse for  $(L, L/2)$  are graphed as a function of the maximum temperature data used in the collapse. The drifts on the largest system sizes at the smallest temperatures are relatively small allowing us to make reliable estimates for the exponents and their error windows:  $z = 2.1(2)$  and  $\eta = 0.17(2)$ .

we study a quadrupolar “Binder” ratio  $\mathcal{R}_Q \equiv \frac{\langle \mathcal{O}_Q^4 \rangle}{\langle \mathcal{O}_Q^2 \rangle^2} \cdot \mathcal{R}_Q$

is expected to be monotonically *decreasing* with  $L$  in a quadrupolar ordered phase. When graphed as a function of the tuning parameter  $p$ ,  $\mathcal{R}_Q$  data for different  $L$  are expected to cross at the phase transition to a non-magnetic phase. Our zero temperature data for the case of site dilution in Fig. 3 clearly shows that the  $\mathcal{R}_Q$  crosses and as the thermodynamic limit is reached the crossing point approaches the percolation threshold  $p^* = 0.5$  with high accuracy. This establishes that the quadrupolar order vanishes continuously as the percolation threshold is approached. This allows us to eliminate the possibility “R”, in which the order parameter vanishes before the percolation threshold is reached. We hence conclude that the order parameter vanishes first precisely at the percolation threshold  $p^*$ , as illustrated in the cartoon “C” shown in Fig. 1.

The vanishing of quadrupolar order right at the percolation threshold raises the interesting possibility that at  $p = p^*$  the system is quantum critical and the quadrupolar correlation possess scale invariance.

Random systems are well known to display a range of novel scaling behavior. We present evidence however that our model possesses conventional power law scaling. In order to test the scaling hypothesis, we first study the dynamic scaling in imaginary time of the order parameter at  $p = p^*$ . In Fig. 4 we show scaling collapse of the order parameter data at the percolation threshold  $p = p^*$  for the maximum sized cluster on an  $L \times L$  lattice as a function of the putative scaling variable  $L^z T$ . For a quantum critical system, we expect the scaling form  $\mathcal{O}_Q^2 = \frac{1}{L^{1+\eta}} \mathcal{F}(L^z T)$ . Note the absence of a tuning parameter (we only require the system to be at the percolation threshold) that would normally be expected for instance from previously studied phase diagrams of quantum rotor models in Fig. 1 (the tuning parameter there is the choice of  $g = g_C$ ). We find an excellent collapse with small finite size corrections to the exponents.

An alternate test of scaling can be made by varying  $p$  away from the percolation threshold. In Fig. 5 we study the scaling of the order parameter with the deviation from the percolation threshold  $p - p^*$ . We find excellent scaling behavior assuming the simple scaling form  $\mathcal{O}_Q^2 = \frac{1}{L^{1+\eta}} \mathcal{G}((p - p^*) L^{1/\nu})$ . The value of  $\eta$  so obtained is in excellent agreement with the estimate obtained from the previous scaling analysis at the percolation threshold Fig. 4.

#### IV. DISCUSSION

We have presented extensive evidence that in two dimensions quadrupolar order vanishes continuously as a function of dilution right at the percolation threshold. At the percolation threshold the systems shows conventional scaling behavior symptomatic of a finite disorder fixed point. Our study is an unusual example of quantum criticality at the percolation threshold without any fine tuning. We contrast this with the generic phase di-

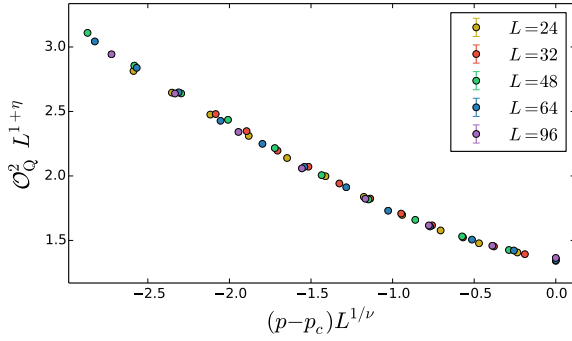


FIG. 5. Scaling collapse of the order parameter as a function of the deviation from the percolation threshold  $p-p^*$ . A good collapse is found without any corrections to scaling with  $\eta = 0.196$  and  $\nu = 0.977$ . We have collected this data assuming  $z = 2$  and hence fixed  $T = 1/L^2$ . The critical exponents were used for this collapse. From pair-wise collapse similar to Fig. 4 we find  $\nu = 1.01(5)$  and  $\eta = 0.18(1)$ . The value obtained for  $\eta$  is consistent with the collapse in Fig. 4.

agram Fig. 1 where at the percolation threshold, tuning of quantum fluctuations to a special value if required to observe quantum criticality.

An interesting open question is whether the critical exponents we have found are universal or can vary continuously as non-universal properties of the magnet are varied at the percolation threshold. Such varying exponents were reported in a study of specific diluted dimerized  $S = 1/2$  magnet<sup>16</sup>. Interestingly, the dynamic critical exponent  $z$  we find from our scaling analysis is numerically somewhat close to  $d_f = 91/48 \approx 1.8958\dots$  (the fractal dimension of a percolating cluster). A number of previous works have found or predicted such scaling at the percolation threshold and our finding could be consistent with such behavior<sup>13,14,17</sup>, though numerically  $z = 2$  would also be consistent with our value within errors. A complete theory of the unusual behavior and scaling we have found here is an interesting direction for future work.

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## Appendix A: Equilibration of disordered clusters

In order to ensure the proper equilibration of our disordered clusters, we have adopted the same equilibration and measurement protocol as in Ref. 8. For each disorder realization we begin by performing  $N_e$  equilibration sweeps at some large initial temperature ( $\beta \approx 1$ , setting  $|J| = 1$ ), followed by  $N_m$  more measurements sweeps, then again  $N_e$  equilibration sweeps, and finally  $N_m$  measurement sweeps. Once this process is complete we perform  $\beta$  doubling on our configurations (see appendix B) and start the process again. Thus for each disorder realization we have two separate measurement cycles at many different values of  $\beta$ .

Separating out two distinct measurement segments allows us to check the equilibration of our disordered clusters. In order to achieve the best QMC averages with minimal computational time, we have set  $N_m = 2N_e$ . We can then study the percent difference of our two measurement segments (each averaged over disorder realizations) as a function of  $N_m$  to determine its optimal value, which is given in Fig. 6. We observe that at the percolation threshold (where the clusters are most fragmented), the difference between our first and second measurement cycles becomes statistically insignificant when  $N_m \approx 64$ . We therefore cautiously set  $N_m = 200$  throughout the course of our numerical studies.

## Appendix B: Zero temperature convergence

The most computationally expensive component of our numerical studies is to converge our configurations to the ground state on disordered clusters. In order to achieve this, extremely large values of the inverse temperature are needed (relative to that of a clean lattice). Fur-

thermore, the free energy landscape of configurations is rugged, and thus quenching a randomly initialized starting configuration to low temperature abruptly may leave

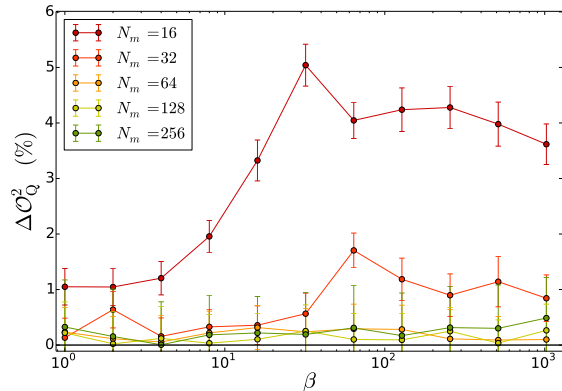


FIG. 6. Here we show the percent difference of the quadrupolar order parameter between the first and second measurement segments ( $\Delta Q_Q^2$ ) as a function of  $\beta$  for several different values of  $N_m = 2N_e$ . The difference between the two segments becomes statistically insignificant over the whole temperature range near  $N_m = 64$ , indicating that we have sufficiently equilibrated our configurations beyond this number of sweeps. We therefore cautiously set  $N_m = 200$  throughout the course of our numerical studies.

it stuck in a local minimum. In order to circumvent this issue, and to efficiently reach the low temperatures required with minimal equilibration, we implement the  $\beta$  doubling procedure<sup>8</sup>. The procedure works as follows: given a disorder realization, we begin the equilibration and measurement cycles at some initial high temperature ( $\beta_0$  on the order of unity). Configurations at high temperatures are relatively easy to equilibrate, since there are few operators acting in the operator string. After the equilibration and measurement cycles, the value of  $\beta$  and the QMC configuration are both doubled. In terms of the configuration, this corresponds to repeating the operator sequence twice. This procedure gives another valid partition function configuration that is close to being equilibrated at an inverse temperature  $2\beta_0$ . From here the equilibration and measurement sequence is again carried out. The process is continued until the final target value  $\beta_{\max}$  is reached.

In order to illustrate the convergence of our zero temperature data, in Fig. 7 we show the binder ratio for max clusters on an  $L = 32$  lattice near the site percolation threshold for all of our  $\beta$  doubled values. We see that very large values of  $\beta$  are required to converge to the ground state. This is a reflection of the fact that  $z$  is close to 2 at this disordered quantum critical point, meaning that doubling the size of the lattice would require quadrupling  $\beta$  to remain near the ground state.

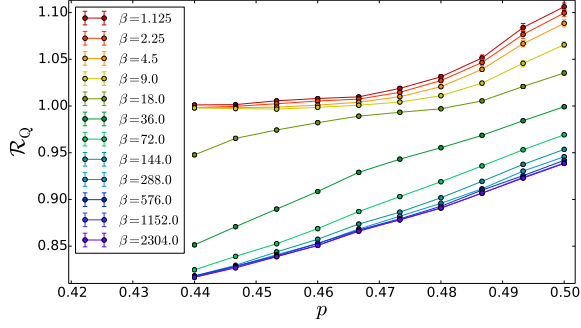


FIG. 7. Here we show the zero temperature convergence of the quadrupolar binder ratio near the site diluted percolation threshold for max clusters on an  $L = 32$  lattice. Each one of the  $\beta$  values was obtained by the  $\beta$  doubling procedure explained in appendix B. We note that very large values of  $\beta$  are required to converge to the ground state, especially near the percolation threshold ( $p_c = 0.5$ ) where we observe quantum critical behavior.