Quantum Criticality and Percolation in Dimer-Diluted Two-Dimensional Antiferromagnets

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The $S = 1/2$ Heisenberg model is considered on bilayer and single-layer square lattices with couplings J_1, J_2, with each spin belonging to one J_2-coupled dimer. A transition from a Néel to disordered ground state occurs at a critical value of $g = J_2/J_1$. The systems are here studied at their dimer-dilution percolation points p^*. The multicritical point (g^*, p^*) previously found for the bilayer is not reproduced for the single layer. Instead, there is a line of critical points $(g < g^*, p^*)$ with continuously varying exponents. The uniform magnetic susceptibility diverges as $T^{-\alpha}$ with $\alpha \in [1/2, 1]$. This unusual behavior is attributed to an effective free-moment density $\sim T^{1-\alpha}$. The susceptibility of the bilayer is not divergent but exhibits remarkably robust quantum-critical scaling.

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A challenging and important aspect of quantum-phase transitions [1] is the influence of disorder (randomness) on the critical behavior, in the ground state as well as in the finite-temperature quantum-critical scaling regime [2,3]. For one-dimensional quantum spin systems a real-space renormalization group (RG) scheme [4] has rigorously established [5] a strong-disorder *random singlet* fixed point which controls the low-energy behavior for any strength of the disorder. This is an unusual type of quantum criticality with dynamic exponent $z = \infty$, which leads to, e.g., a susceptibility diverging as $T^{-1}\ln^{-2}(T)$. The RG procedure has also been carried out numerically for various two-dimensional (2D) models, although in this case there is no rigorous proof of its validity. In the random transverse Ising model a random singlet phase was found [6], but only conventional critical points (finite z) were found for SU(2) symmetric (Heisenberg antiferromagnet) systems [7]. However, the disorder leads to unusual properties in the ordered and disordered phases [7,8], e.g., quantum Griffiths effects.

In the 2D $S = 1/2$ Heisenberg model an order-disorder transition occurs at a critical strength of a coupling pattern favoring singlet formation on dimers, plaquettes, etc. [2,3]. There is ample evidence from quantum Monte Carlo (QMC) studies [9–11] that this transition is in the 3D $O(3)$ universality class, as predicted theoretically [2,12]. According to the Harris criterion [13], disorder should be relevant at this transition. The applicability of efficient quantum Monte Carlo methods [14], in combination with a well developed RG scheme, makes dimerized Heisenberg models very well suited for exploring disorder effects on quantum-phase transitions. Recent studies have demonstrated a variety of scaling behaviors [7,15–19]. The case of dilution disorder is particularly interesting, as it includes the special case of quantum criticality at the classical percolation point, where geometrical and spin fluctuations are simultaneously divergent [15,16]. Here two cases of such multicriticality are studied using QMC simulations—a bilayer and a dimerized single-layer model with random dilution of dimers. The systems are illustrated in Fig. 1 along with schematic phase diagrams. The dramatically different behaviors at the percolation threshold is the main focus of this Letter. The bilayer has a single point of quantum criticality where the susceptibility scales to zero as $T \to 0$ [15,16]. In contrast, the single layer exhibits a line of quantum-critical points where the susceptibility is divergent.

The 2D Heisenberg model with random site dilution has been studied extensively because of its relevance to Zn doped cuprate antiferromagnets [20,21]. It was for a long time believed that site or bond dilution leads to a quantum-phase transition before the geometric percolation point is reached [22–24]. However, recent QMC studies have shown that the transition coincides with the percolation point [25,26] and that the percolating cluster is ordered

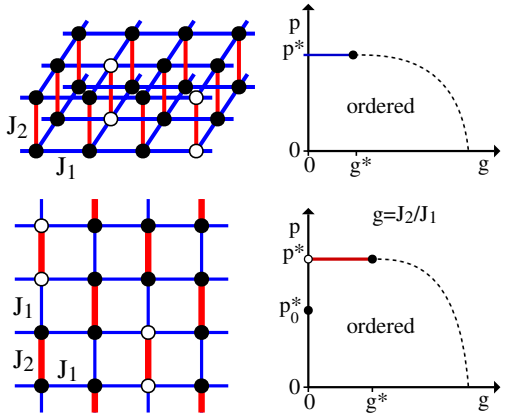

FIG. 1 (color online). The lattices studied (left) and their $T = 0$ phase diagrams (right). The couplings are $J_1\mathbf{S}_i \cdot \mathbf{S}_j$ and $J_2\mathbf{S}_i \cdot \mathbf{S}_j$, with $J_1, J_2 > 0$ (in the bilayer all interlayer couplings are J_2 and in the single layer the vertical bonds alternate J_1, J_2). Dimer dilution corresponds to removing J_2-coupled pairs—such removed dimers are indicated by open circles. For the bilayer, the percolating cluster is ordered on the line $(0 \le g < g^*, p^*)$, whereas the single-layer is quantum critical for $(0 < g < g^*, p^*)$.

[26]. The critical exponents pertaining to equal-time correlations (ν, β, and η) are therefore those of classical percolation. Other exponents (α, γ, δ) are given by combinations of percolation exponents and the dynamic exponent of the spin clusters [27].

The long-range order of the Heisenberg antiferromagnet on percolating clusters, which have fractal dimensionality $d = 91/48$ [28], implies that other couplings have to be introduced in order to realize a dilution-driven quantum-phase transition [26]. This has been explored recently with the dimer-diluted bilayer model [15,16]. In the pure Heisenberg bilayer, with intralayer and interlayer couplings J_1 and J_2, there is an order-disorder transition at $g = J_2/J_1 \approx 2.52$ [11] (due to the tendency to singlet formation across the planes). If this system is diluted by randomly removing single spins, order is induced in the disordered phase because moments localize in the neighborhood of the vacancies. These moments interact and order at $T = 0$ [29]. By instead diluting the two layers symmetrically, i.e., removing dimers consisting of nearest-neighbor spins on opposite planes, no localized moments form and a phase transition takes place at a coupling g which depends on the dilution fraction p, as shown in Fig. 1. At the geometrical percolation point, which clearly is the same as for a single site-diluted layer, $p^* \approx 0.41$ (hole concentration) [28], there is a multicritical point (g^*, p^*) at which the long-range order on the percolating cluster vanishes and the spins are quantum critical. A critical coupling $g^* \approx 0.15$ and dynamic exponent $z^* \approx 1.3$ were found in two independent QMC calculations [15,16]. The generic transition for $p < p^*$ has also been studied in detail by Monte Carlo simulations of an analogous 3D classical Heisenberg model with columnar defects [17]. Here an exponent $z \approx 1.3$ was found (but $z^* \neq z$ is expected because of the different cluster dimensionality). Long-range order in the presence of quantum fluctuations on the line ($g < g^*$, p^*), which was believed not to be possible for a continuous order parameter [30], was recently related to the fracton dimensionality of the percolating cluster [31].

The question now arises as to the generality of the behavior found in the bilayer model. On its percolating cluster each spin has a neighbor in the opposite layer with which it correlates at low temperature. Magnetization fluctuations are thus quenched as $T \to 0$. Here the dimerized single-layer model is used to investigate the role of the bilayer symmetry upon dilution. Without disorder, the system has a quantum-phase transition, in the same universality class as the bilayer, at $J_2/J_1 \approx 2.5$. With dimer dilution again corresponding to random removal of J_2 dimers, the percolation point $p^* = 1/2$ because the dimers are connected as a triangular lattice [28]. The $T = 0$ phase diagram, outlined in Fig. 1, is similar to that of the bilayer in that there is a finite segment of the phase boundary at $p = p^*$, terminating at a point (g^*, p^*) beyond which the transition occurs for $p < p^*$. However, as will be discussed in detail below, there is a striking difference: Whereas the percolating cluster of the bilayer has Néel order for $0 \leq g < g^*$ [15,16], the percolating cluster of the single dimerized layer is quantum critical on the whole line ($0 < g \leq g^*$, p^*), with $g^* \approx 1.25$. On this line, the magnetization fluctuations are not completely quenched as $T \to 0$, leading to a divergent susceptibility, $\chi \sim T^{-\alpha}$, with $\alpha \to 1/2$ for $g \to g^{*-}$ and $\alpha \to 1$ for $g \to 0$. For $g = 0$, a Curie susceptibility is expected on account of the percolating cluster breaking up into smaller pieces when all the J_2 couplings vanish (the percolation point here is $p_0^* \approx 0.29$). Some of these clusters contain an odd number of spins. For $g > 0$, the divergent susceptibility can then be attributed to effectively isolated subclusters with net moments, which are gradually "frozen out" as the temperature is reduced. The form $\chi \sim T^{-\alpha}$ corresponds to a free-moment density scaling as $T^{1-\alpha}$. This remarkable behavior will here be demonstrated on the basis of large scale QMC (stochastic series expansion [14]) calculations. Only results exactly at the percolation threshold, $p = p^*$, will be discussed. The numerical techniques and special methods developed for studies of random systems at ultra-low temperature are discussed in detail in Ref. [26].

The temperature dependence of the uniform susceptibility $\chi(T)$ of the bilayer close to the multicritical point was discussed before in Ref. [15] (averages over all clusters were presented in Ref. [16]). Figure 2 shows a more extensive set of high-precision results for the largest cluster on $L \times L$ lattices with $L = 256$ ($L \to \infty$ converged for the temperatures shown) at $p^* = 0.407\,253\,8$ [32]. Averages over several thousand dilution realizations were taken. The temperature is scaled according to the expected quantum-critical form, $\chi = a + bT^{d/z-1}$ [3], where a and b are constants and $a = 0$ at a quantum-critical point. Using the fractal dimension $d = 91/48$ and adjusting z to obtain a linear χ versus $T^{d/z-1}$, the same dynamic exponent, $z = 1.36 \pm 0.01$, is found for all $0 < g \lesssim g^*$. An improved estimate $g^* = 0.118 \pm 0.006$ is also obtained. Note that

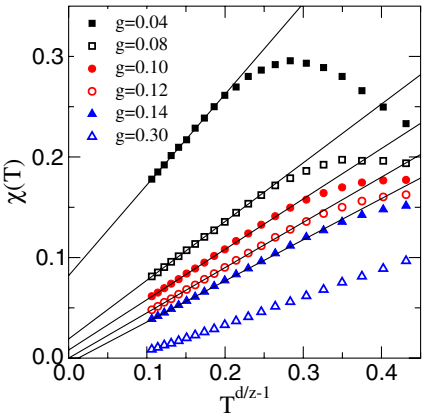

FIG. 2 (color online). Temperature scaling of the susceptibility of the infinite percolating bilayer cluster for different coupling ratios g, using a dynamic exponent $z = 1.36$.

bilayer criticality can be expected only for $T < g$, which is indeed the case in Fig. 2.

In a clean quantum-critical system, there is a low-temperature crossover of $\chi(T)$ to a "renormalized classical" behavior when $g < g_c$, at a temperature of the order of the spin stiffness ρ_s [3]. No such crossover is seen in Fig. 2, however. Although it cannot be completely excluded that there is a crossover at still lower temperature, one can also argue that there should be no crossover, because $\rho_s = 0$ on the percolating cluster [15,26] (although there is long-range order—this unusual behavior has also been discussed in Ref. [33]) and there is no apparent energy scale, except T, to govern the long-distance physics of the spins on this fractal network. Thus the multicritical point (p^*, g^*) may control the $T < g$ temperature dependence for all $0 \leq g \leq g^*$ at p^*. Considering that the scaling in Fig. 2 extends down as low as to $T = J/256$ and to couplings $g \approx g^*/3$, the results appear to support such an unusual manifestation of quantum criticality on a fractal percolating cluster.

Turning now to the single-layer model at its dimer percolation point, $p^* = 1/2$ for $g > 0$, the disorder-averaged uniform susceptibility of the largest cluster was calculated for L up to 256, down to $T = J/512$ ($L \to \infty$ converged). Here a special point (g^*, p^*) is found which separates qualitatively different behaviors of the uniform susceptibility. As seen in Fig. 3(a), at $g^* = 1.247 \pm 0.001$, χ is linear in T and approaches a finite value as $T \to 0$. For $g > g^*$ the susceptibility drops to zero and for $g < g^*$ it diverges. As shown in Figs. 1(b) and 1(c), the divergence is of the form $\chi \sim T^{-\alpha}$, with α very close to $1/2$ for $1 \lesssim g < g^*$ and $\alpha \to 1$ for $g \to 0$. Such a divergence can be interpreted as a temperature dependent fraction $\sim T^{1-\alpha}$ of effectively free magnetic moments.

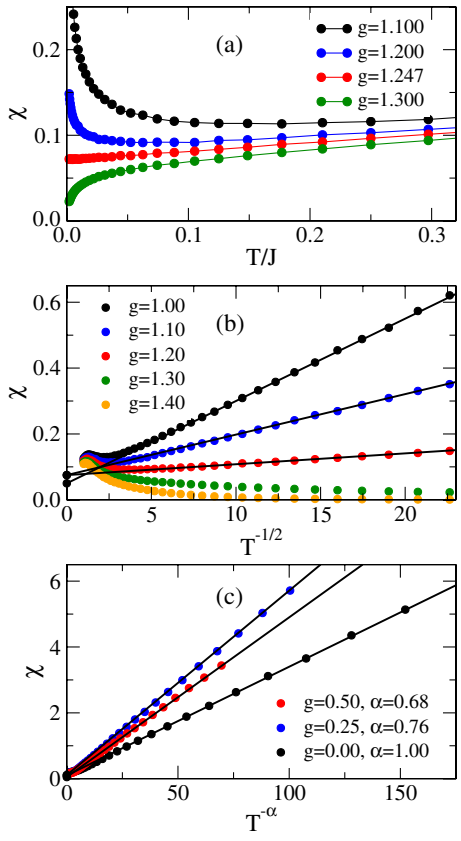

FIG. 3 (color online). Temperature dependence of the susceptibility of the percolating single-layer cluster. (a) At and close to the coupling $g^* \approx 1.247$. (b) $T^{-1/2}$ scaling for $1 < g \lesssim g^*$. (c) Scaling with varying exponent for $g < 1$.

As already noted, exactly at $g = 0$ the percolating cluster is broken up into smaller subclusters and then a Curie behavior, $\chi \sim T^{-1}$, is expected on account of clusters with a net spin. For small but nonzero g, one might then have expected a crossover from Curie behavior when $T > g$ to a finite susceptibility as $T \to 0$. Instead, it appears that coupling the subclusters leads to a g dependent power-law temperature scaling of the number of effectively free moments. The effective couplings of these moments to each other must thus have a g dependent power-law distribution, leading to a self-similar structure of free moments different from that of the underlying fractal cluster. It is remarkable that this behavior persists also when $g \approx 1$, where the picture of weakly connected subclusters is not obviously relevant.

The bilayer percolating cluster is ordered at $T = 0$ for $g < g^*$ [15,16]. This is not the case for the single dimerized layer at p^*. Instead, quantum-critical fluctuations are observed for $0 < g \leq g^*$. Consider the staggered structure factor $S(\pi, \pi)$ and susceptibility $\chi(\pi, \pi)$ of a cluster of N_c spins,

$$S(\pi, \pi) = \frac{1}{N_c} \sum_{i,j} P_{ij} \langle S_i^z S_j^z \rangle,$$

$$\chi(\pi, \pi) = \frac{1}{N_c} \sum_{i,j} P_{ij} \int_0^\beta d\tau \langle S_i^z(\tau) S_j^z(0) \rangle,$$

where $P_{ij} = (-1)^{x_i + y_i - x_j - y_j}$. At a quantum-critical point, these quantities, averaged over disorder, should scale with the system size as $\langle S(\pi, \pi)/N_c \rangle \sim L^{\gamma_s}$, $\langle \chi(\pi, \pi)/N_c \rangle \sim L^{\gamma_\chi}$, with $\gamma_s = -(z + \eta)$ and $\gamma_\chi = -\eta$ (normalizing by $N_c \sim L^d$ before disorder-averaging leads to some reduction of statistical fluctuations). Figure 4 shows results for $g = 1$. The observed scaling gives $z \approx 3.0$ and $\eta \approx -1.7$. The dynamic exponent can be compared with the expected quantum-critical susceptibility; $\chi \sim T^{-\alpha}$ with $\alpha = 1 - d/z$ [3]. With the exponent $\alpha = 1/2$ obtained from $\chi(T)$ in Fig. 3(b), it is apparent that this relationship does not hold here [z extracted from $S(\pi, \pi)$ and $\chi(\pi, \pi)$ should be the actual dynamic exponent]. At the special point g^* the extracted $\alpha = 0$ and $z \approx d$ (not shown here) are in fact consistent with this relationship. This is also the case at the bilayer multicritical point [15]. The single-layer quantum criticality on the line $(0 < g < g^*, p^*)$ thus appears to be fundamentally different. It should be noted that the behavior is not consistent with a Griffiths phase [7], as the spin correlations in that case should be exponentially decaying, in contrast to the power law seen in Fig. 4.

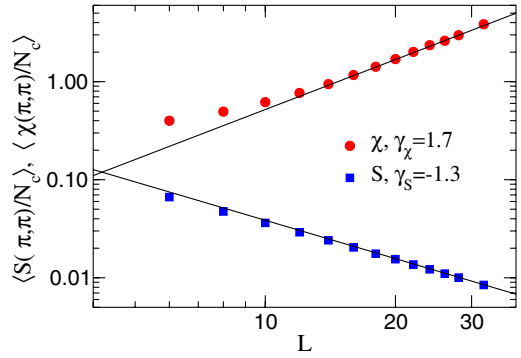

FIG. 4 (color online). Finite-size scaling of the staggered structure factor and susceptibility, normalized by the cluster size, at intradimer coupling $g = 1$. The lines show scaling with exponents indicated in the legends. The results where obtained at T sufficiently low to give the ground state.

An extended line of quantum-critical points was not anticipated on the basis of a real-space RG approach developed recently for quantum rotors on a percolating cluster [31]. The critical line discovered here for the single-layer model is more similar to the 1D random singlet phase [5], in that there is a temperature dependent fraction of effectively free moments. In the random singlet phase $z = \infty$ whereas in the model studied here z is finite and diverges in the limit $g \to 0$. A temperature dependent fraction of effective moments has also been observed in a 2D model of interacting localized moments [34]. However, there the asymptotic $T \to 0$ susceptibility is always Curie-like, and there is long-range order at $T = 0$. An RG calculation for random frustrated moments in the continuum shows a $\chi(T)$ divergence with varying exponent [35]. The ground state properties were not accessible in that study.

The bilayer multicritical point (p^*, g^*) has been argued [15,16] to influence finite-temperature properties of single-layer Zn doped cuprate antiferromagnets, for which a dynamic exponent $z \approx 1.4$ was found in neutron scattering experiments [21]. However, although g^* is small (≈ 0.12) it is difficult to explain how bilayer quantum criticality could be realized when $T \gg g = 0$ (due to an expected crossover at $T \approx g$; see Fig. 2). Physical realizations of single-layer dimer dilution are not immediately obvious. Nevertheless, the results presented here serve to illustrate rich and surprising behaviors arising from the interplay of classical percolation and quantum fluctuations, going beyond previous examples of scaling in percolating fractal structures [36].

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