Quantum Versus Geometric Disorder in a Two-Dimensional Heisenberg Antiferromagnet

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We present a numerical study of the spin-1/2 bilayer Heisenberg antiferromagnet with random interlayer dimer dilution. From the temperature dependence of the uniform susceptibility and a scaling analysis of the spin correlation length we deduce the ground state phase diagram as a function of nonmagnetic impurity concentration p and bilayer coupling g. At the site percolation threshold, there exists a multicritical point at small but nonzero bilayer coupling $g_m = 0.15(3)$. The magnetic properties of the single-layer material $La_2Cu_{1-p}(Zn, Mg)_pO_4$ near the percolation threshold appear to be controlled by the proximity to this new quantum critical point.

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Quantum phase transitions (QPTs) in the presence of disorder are the subject of considerable current interest as they exhibit rich new physics but are rather poorly understood. Experimental examples include the cuprate superconductors [1], heavy fermion compounds [2], metal-insulator [3] and superconductor-insulator transitions [4], quantum Hall effect [5], and quantum magnets [6]. A significant amount of theoretical and numerical work has been devoted to the study of the random Ising chain in a transverse field [7], the simplest quantum many-body system with quenched disorder, and to its analog in higher dimensions [8,9]. Partly because of its relevance to cuprate superconductivity, the spin-1/2 square-lattice Heisenberg antiferromagnet (SLHAF) has attracted enormous interest [10]. While the effects of a single nonmagnetic impurity are well understood [11], there exist few theoretical results for finite impurity concentrations [12]. Given the considerable challenges, theory for quantum systems with disorder is often guided by insight provided by numerical work.

In the absence of disorder, the nearest-neighbor (NN) SLHAF can be driven through a quantum phase transition by introducing a parameter analogous to the transverse field for the Ising model. For example, this is achieved by introducing frustrating next-NN couplings [13] or by coupling two square lattices to form an antiferromagnetic bilayer [14]. It had been argued that random site or bond dilution of the SLHAF in the extreme quantum limit of spin-1/2 may lead to a nontrivial quantum phase transition [9,15,16]. However, recent experimental [17] and numerical [17,18] work suggests that the ground state remains ordered for nonmagnetic impurity concentrations p up to the site percolation threshold p_*, and that the critical cluster at $p = p_*$ appears to have a nonzero staggered moment [18], which would imply that the percolation transition is classical.

In this Letter, we present numerical results for the site-diluted spin-1/2 NN bilayer antiferromagnet, with disorder that is fully correlated between the layers ("dimer" dilution). At zero bilayer coupling, this problem reduces to the previously studied diluted spin-1/2 SLHAF. By increasing the strength of the bilayer coupling from zero, we are able to increase quantum fluctuations beyond those for the spin-1/2 SLHAF. Our finite-temperature results allow us to extract the ground state phase diagram as a function of the strength of quantum fluctuations and the degree of disorder, and they reveal a new multicritical point at $p = p_*$ for small, but nonzero bilayer coupling. This phase diagram resembles that of the diluted two-dimensional Ising ferromagnet in a transverse field [9]. The present results are relevant to recent experimental findings for the model spin-1/2 SLHAF $La_2Cu_{1-p}(Zn, Mg)_pO_4$ [17].

We study the Hamiltonian

$$H = J \sum_{\langle i,j\rangle, n=1,2} \epsilon_i \epsilon_j \mathbf{S}_{i,n} \cdot \mathbf{S}_{j,n} + J_\perp \sum_i \epsilon_i \mathbf{S}_{i,1} \cdot \mathbf{S}_{i,2}, \quad (1)$$

where J ($J_\perp$) is the antiferromagnetic planar (bilayer) coupling, the first sum is over all planar NN pairs, and $\epsilon_i = 0$ ($\epsilon_i = 1$) with probability p ($1 - p$). We define the reduced bilayer coupling $g = J_\perp/J$ and work with units in which $J = a = k_B = g\mu_B = \hbar = 1$ (a is the lattice constant). If a site is removed on one layer, the corresponding site in the adjacent layer is also removed. This constraint preserves the percolation properties of the square lattice. It also ensures that there are no unpaired "dangling" spins. Unlike for the diluted SLHAF ($g = 0$), the uniform susceptibility χ_u for the bilayer then contains no Curie-like term. The temperature dependence of χ_u can then be used to determine the critical bilayer coupling $g_c(p)$ for the diluted system. We use the loop-cluster Monte Carlo method [19], which has provided good results for the diluted spin-1/2 ladder [20] and SLHAF [17]. Simulations are performed on large lattices of up to $N = 512 \times 512 \times 2$ sites, to temperatures as low

as $T = 1/300$, with Trotter numbers of $20/T$, and by averaging 10 to 100 random configurations. By keeping the planar lattice size significantly larger than the correlation length $\xi(g, p, T)$, we are able to avoid finite-size effects. Several concentrations are chosen, spanning the range from the pure system up to $p = 0.5$, beyond the site percolation threshold $p_* = 0.407\,253\,79(13)$ [21]. In addition to $\chi_u(g, p, T)$ and $\xi(g, p, T)$, we also compute the staggered susceptibility $\chi_{st}(g, p, T)$. The susceptibilities are defined as $NT\chi_u = [\langle(\sum_i S_i^z)^2\rangle]$ and $NT\chi_{st} = [\langle[\sum_i (-1)^i S_i^z]^2\rangle]$, where the sum is over all sites N and $[\cdots]$ indicates the disorder average. The instantaneous staggered correlation function is given by $NC(\mathbf{r}) = \mathrm{sgn}(\mathbf{r})[\langle\sum_i S_i^z S_{i+\mathbf{r}}^z\rangle]$, where $\mathrm{sgn}(\mathbf{r}) = 1\,(-1)$ for $\mathbf{r}$ separating sites on the same (different) sublattice. The correlation length is obtained from the behavior $C(r) \sim e^{-|\mathbf{r}|/\xi}$ at large instances.

At the critical coupling of the pure bilayer, $\chi_u(g_c, 0, T) \sim T$ [14]. In the quantum disordered phase ($g > g_c$), the strong bilayer coupling leads to a nonzero singlet-triplet gap, which prevents long-range order in the ground state, and χ_u is exponentially small at low temperature. In the ordered phase ($g < g_c$), χ_u approaches a nonzero value proportional to the spin stiffness [22]. These different characteristic behaviors have allowed the determination of the critical bilayer coupling for the pure system from finite-T numerics [14]. This should also be possible at nonzero p, as long as there exists an ordered phase. Figure 1 shows some of our results for χ_u for the pure system, at the intermediate concentration of $p = 25\%$, and at a concentration just below the percolation threshold. Note that T is scaled by g, so that results for different concentrations fall into approximately the same horizontal range. Fitting the low-T data to $\chi_u = c_1 + c_2 T^\lambda$ gives excellent results, as shown by the dashed lines in Fig. 1. We interpret positive (negative) values of c_1 as indicative of an ordered (quantum disordered) ground state. For $p = 0$, we find $g_c \approx 2.525$, in very good agreement with $g_c = 2.525(2)$ obtained previously [14]. The critical coupling decreases with increasing p, and λ decreases from its value of 1 for the pure system to around 0.7 near the percolation threshold. We conclude that order persists at $g = 0.10$ even at $p = 417/1024$ (40.722...%), very close to $p = p_*$ (40.725...%).

In the ordered phase, the low-T correlation length is expected to be exponential in inverse temperature as for the pure SLHAF [12,17], while $\xi \sim T^{-1/z}$ at the quantum critical point [23], where z is the dynamic critical exponent. The crossover temperature T_{dev} at which ξ deviates from a power law scales with the deviation from the critical point as $T_{\mathrm{dev}} \sim |g - g_c|^\phi$ (p can be substituted for g for cuts at fixed g). Similarly, in the disordered phase, the low-T correlation length approaches a constant value, but follows power-law behavior at higher temperatures. Figure 2 shows scaling plots for four cuts across the phase boundary. For the pure bilayer, Fig. 2(a), for which the low-T physics can be mapped to the quantum nonlinear σ model [13], the Euclidean time direction is equivalent to the spatial dimensions (Lorentz invariance) and $z = 1$. At zero temperature, Euclidean time extends to infinity, so that the critical exponents of the pure system are those of the *three-dimensional classical* Heisenberg model, for which $\nu \approx 0.705$ and $\gamma \approx 1.39$ [24]. Since $\nu = \phi/z$, we therefore have fixed $\phi = 0.705$ in Fig. 2(a). We obtain excellent scaling with $g_c = 2.5215(10)$, slightly lower than the value obtained from Fig. 1(a) and in previous work [14]. A similar result is obtained for χ_{st} (not shown) using $\gamma = 1.39$.

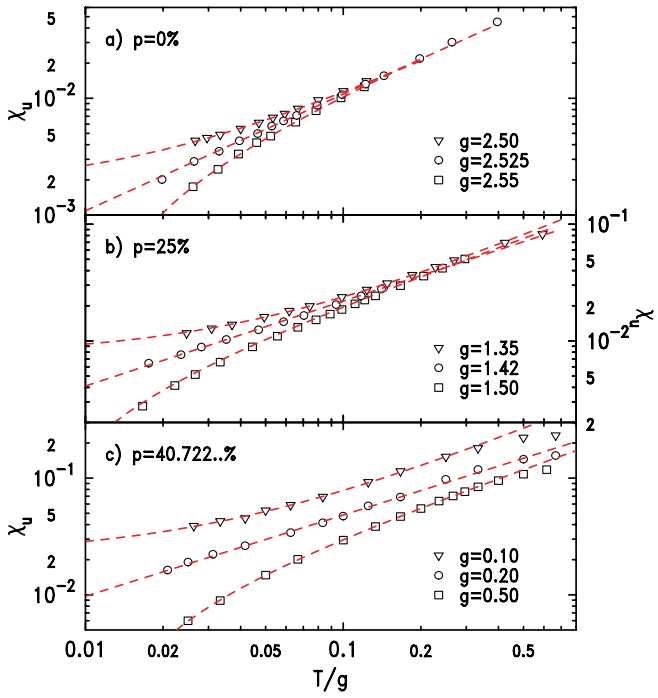

FIG. 1 (color online). Temperature dependence of the uniform susceptibility. (a) Pure bilayer: $g_c \approx 2.525$ and $\chi_u(g_c, 0, T) \sim T$, as found previously [14]. (b) $p = 0.25$: $g_c \approx 1.42$ and $\chi_u \sim T^{0.73(5)}$. (c) Just below the percolation threshold: g_c is significantly reduced, and $\chi_u \sim T^{0.70(7)}$.

At $p = 25\%$ [Fig. 2(b)], we find $z = 1.07(2)$ and $\phi = 0.95(5)$, as well as $g_c(p = 0.25) = 1.412(6)$, consistent with Fig. 1(b). Scaling as a function of p at fixed $g = 1.42$ (not shown) gives good results using the same exponents and $p_c(g = 1.42) = 24.85(15)\%$. Near $p = p_*$, scaling as a function of p is easier to obtain, and our results for $g = 0.20$ and $g = 0$ (single-layer) are shown in Figs. 2(c) and 2(d), respectively. For $g = 0.20$, the exponents are $z = 1.3(1)$, $\phi = 1.45(10)$, and $p_c = 39.9(3)\%$. Recent neutron scattering results for $La_2Cu_{1-p}(Zn, Mg)_pO_4$ [17] as well as numerical work [16–18] indicate that $p_c = p_*$ for the spin-1/2 SLHAF. Therefore, at $g = 0$, we fixed $p_c = p_*$. As is evident from Fig. 2(d), this gives excellent scaling, with $z = 1.65(5)$ and $\phi = 1.8(1)$.

Sandvik [18] concludes that the percolating cluster of the NN SLHAF at $p = p_*$ has a nonzero ordered moment per spin, so that even in the extreme quantum limit of

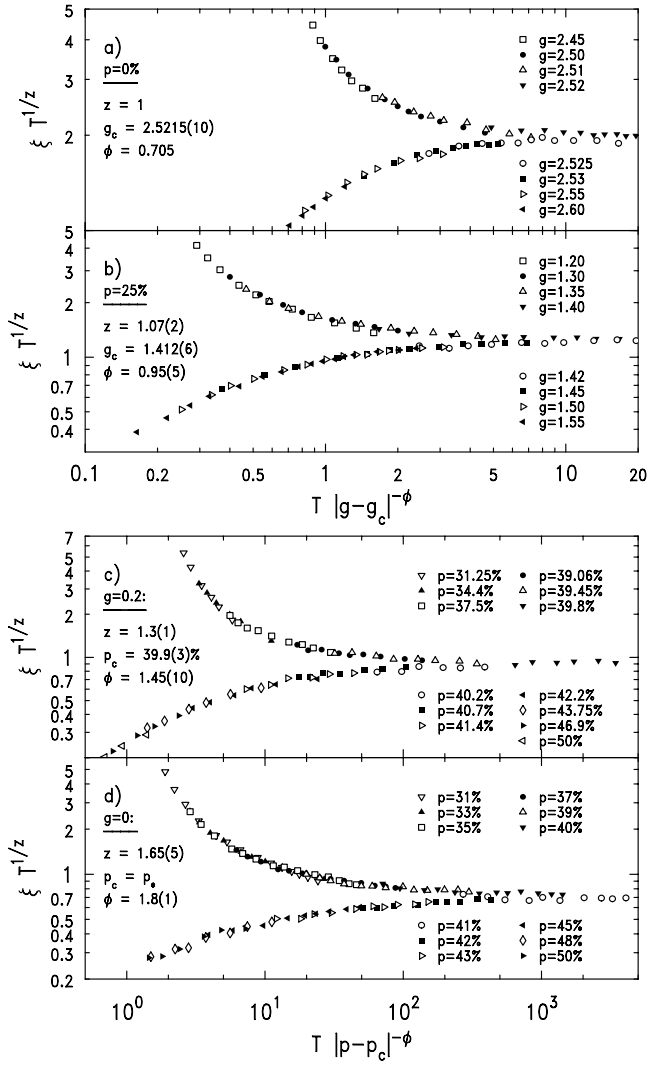

FIG. 2. Scaling plots of the correlation length (a) for the pure spin-1/2 bilayer Heisenberg antiferromagnet, (b) at fixed nonmagnetic concentration $p = 0.25$, and at fixed bilayer couplings (c) $g = 0.20$ and (d) $g = 0$ (SLHAF).

spin-1/2 the percolation transition at $T = 0$ is still classical. If this picture is correct, then one would expect the moment per spin on the critical cluster to vanish at some nonzero value $g_m = g_c(p_*)$. This is indeed consistent with our findings. Figure 3 shows the extrapolated ground state phase diagram, and we obtain $g_m = 0.15(3)$. The exponents for $g = 0.10$ are very similar to those at $g = 0.20$, and we estimate $z_m = 1.33(7)$, $\nu_m = 1.11(14)$, and $\gamma_m = 2.69(31)$. Calculations of the temperature dependence of the correlation length at this multicritical point agree with this estimate for z_m.

The Harris criterion is often used to determine whether the exponents at classical phase transitions should change in the presence of weak disorder: for disorder to be relevant, ν of the *pure system* has to satisfy $\nu < 2/d$ [25]. More generally, it is expected that the *disordered* system satisfies $\nu \geq 2/d$ [26]. For a quantum phase transition, the dimension to use is the number of dimensions

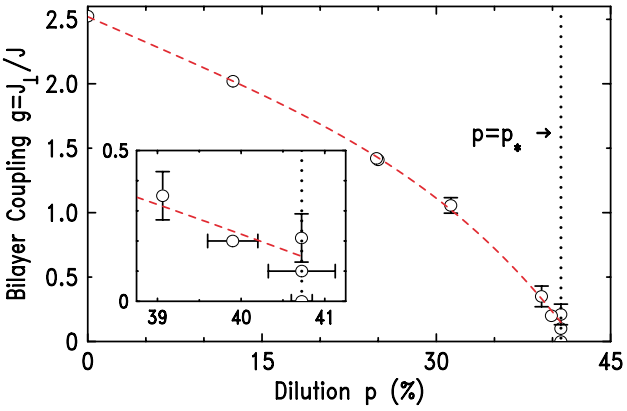

FIG. 3 (color online). Phase diagram of the diluted bilayer. The dashed line is a guide to the eye. The percolation threshold is marked by the dotted vertical line. The inset is a close-up view of the region near the percolation threshold.

in which there is disorder, in our case $d = 2$. Since for the pure bilayer $\nu \approx 0.705 < 1$, we expect disorder to be relevant [23].

The meaning of the exponents obtained along the phase boundary ($0 < p < p_*$) is not entirely clear. As argued above, disorder is expected to be a relevant perturbation of the pure system. This should lead to a unique set of critical exponents for the randomly diluted bilayer Heisenberg antiferromagnet, different from those at $g_c(0)$ and g_m, and from the classical percolation exponents. There are no special values of p between $p = 0$ and $p = p_*$, and the exponents appear to vary continuously along the phase boundary. This is qualitatively similar to the two-dimensional Ising magnet, for which quenched disorder is a relevant perturbation, and which appears to exhibit (finite-temperature) transitions with concentration-dependent exponents [27]. However, these Ising exponents also show a slight temperature dependence, and it has been argued that this indicates that these are not the true exponents, which should be observable only at temperatures asymptotically close to the transition temperature, but rather effective exponents [27]. A similar masking might occur in the present system, so that true critical exponents would be revealed only at asymptotically low temperatures. The critical points at $g_c(0)$ and g_m influence the intermediate-T physics of systems nearby in parameter space. One might expect that at $p \approx 25\%$ the extracted exponents are close to the true critical exponents: $p = 25\%$ appears to be far away from both the pure system and from the percolation threshold, so that disorder effects might be large, while ξ at the temperatures of our study is still much larger than the percolation length, which diverges at $p = p_*$. However, we obtain $\nu = \phi/z = 0.89$, in violation of the criterion $\nu \geq 1$ [26]. Consequently, we believe that the exponents extracted for $0 < p < p_*$ are effective, rather than true critical exponents.

The scaling dimension for a QPT is $d_q = d + z$, where z can differ from 1 in the disordered case. Using

$z_m = 1.33$, $\nu_m = 1.11$, and $\gamma_m = 2.69$, the hyperscaling relationship $\beta = (d_q\nu - \gamma)/2$ gives $\beta_m \approx 0.5$ for the multicritical point. Our corresponding effective values for $g = 0$ are $z = 1.65(5)$, $\nu = 1.09(9)$, $\gamma = 2.65(27)$, and $\beta \approx 0.67(13)$. Previous finite-size scaling results for the NN SLHAF resulted in spin-dependent critical exponents, with $z = 2.54(8)$, $\nu = 1.23(16)$, and $\beta = 0.50(7)$ for spin-1/2 [16]. Based on our results, these exponents have to be viewed as effective exponents due to the influence of the nearby multicritical point. This is consistent with the claim that the spin-1/2 NN SLHAF at $p = p_*$ should exhibit asymptotic percolation critical behavior [18], for which $\nu_p = 4/3 \approx 1.33$, $\gamma_p = 43/18 \approx 2.39$, and $\beta_p = 5/36 \approx 0.14$ [28]. Experiments for $La_2Cu_{1-p}(Zn, Mg)_pO_4$ near $p = p_*$ revealed $\xi \sim T^{-\nu_T}$, with $\nu_T \approx 0.7$, which is very close to the effective value $1/z \approx 0.61$ ($g = 0$) and to $1/z_m \approx 0.75$ ($g_m = 0.15$).

In summary, we have mapped out the phase diagram of the spin-1/2 bilayer Heisenberg antiferromagnet with quenched disorder in the form of interlayer dimer dilution. Varying the bilayer coupling has allowed us to further increase quantum fluctuations beyond those of the spin-1/2 NN SLHAF and to investigate the joint effects of quantum fluctuations and quenched disorder. Our results for the pure bilayer agree with previous work. We find that the critical coupling $g_c(p)$ decreases with increasing p but remains nonzero even at the percolation threshold $p = p_*$. The point $g_m = g_c(p_*)$ is a new multicritical critical point, and we obtain estimates of several critical exponents. Quenched disorder is expected to be a relevant perturbation to the pure quantum system and to lead to new critical behavior at nonzero p below the percolation threshold. The critical exponents along the phase boundary ($0 < p < p_*$) appear to change continuously, but it is likely that the true critical behavior is masked by finite-temperature effects and by the effective proximity to either the pure fixed point or the new multicritical point. The small value of $g_m \approx 0.15$ indicates that the intermediate-temperature properties of the spin-1/2 SLHAF $La_2Cu_{1-p}(Zn, Mg)_pO_4$ near $p = p_*$ [17] may be controlled by this nearby quantum critical point. We note that the experimental system might be best described by considering a nonzero, frustrating next-NN exchange of about $0.05 - 0.10J$ [17,29], which, in effect, would place it even closer to the multicritical point. The phase diagram of Fig. 3 is qualitatively similar to that for the randomly diluted 2D Ising model in a transverse field [9]. We note that recent numerical work [30], which uses a different approach, gives $g_m(p_*) = 0.16(1)$ with $z_m = 1.28(2)$, in very good agreement with our results.

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