Impurities near an antiferromagnetic-singlet quantum critical point

T. Mendes-Santos,[1,2] N. C. Costa,[1] G. Batrouni,[3,4,5] N. Curro,[2] R. R. dos Santos,[1] T. Paiva,[1] and R. T. Scalettar[2]

[1]*Instituto de Física, Universidade Federal do Rio de Janeiro, Caixa Postal 68.528, 21941-972 Rio de Janeiro Rio de Janeiro, Brazil*

[2]*Department of Physics, University of California, Davis, California 95616, USA*

[3]*Université Côte d'Azur, INLN, CNRS, France*

[4]*MajuLab, CNRS-UNS-NUS-NTU International Joint Research Unit UMI 3654, Singapore*

[5]*Centre for Quantum Technologies, National University of Singapore, 2 Science Drive 3, Singapore 117542*

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Heavy-fermion systems and other strongly correlated electron materials often exhibit a competition between antiferromagnetic (AF) and singlet ground states. Using exact quantum Monte Carlo simulations, we examine the effect of impurities in the vicinity of such an AF-singlet quantum critical point (QCP), through an appropriately defined "impurity susceptibility" $\chi_{\rm imp}$. Our key finding is a connection within a single calculational framework between AF domains induced on the singlet side of the transition and the behavior of the nuclear magnetic resonance (NMR) relaxation rate $1/T_1$. We show that local NMR measurements provide a diagnostic for the location of the QCP, which agrees remarkably well with the vanishing of the AF order parameter and large values of $\chi_{\rm imp}$.

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I. INTRODUCTION

In materials, such as the cuprate superconductors, mobile impurities introduced, e.g., via the replacement of La by Sr, are known to destroy antiferromagnetic (AF) order very rapidly [1,2]. Long-range spin correlations are somewhat more robust to static scatterers, e.g., via Zn substitution for Cu in the same materials [3–5]. This competition of AF and chemical doping is a central feature of many other strongly correlated systems, including Li doping in nickel oxides [6,7], spin chains [8], and ladders [9] and has been explored by quantum Monte Carlo (QMC) approaches in single band fermion models [10] and their strong-coupling spin limits [11].

Materials with multiple fermionic bands or localized spins in multichain or multilayer geometries offer an additional richness to the effect of impurities on AF since even in the clean limit they can exhibit a quantum critical point (QCP) separating AF and singlet phases. Although impurities reduce AF deep in the ordered phase, nearer to the QCP, they can increase AF and even, beginning in the quantum disordered phase, *induce* AF by breaking singlets [8,9]. This has recently been explored in heavy-fermion materials where Cd doping of superconducting $CeCoIn_5$ induces long-range magnetic order [12]. The underlying mechanism is believed to be a local reduction of conduction electron-local moment (c-f) hybridization on the Cd sites, suppressing the singlet energy gain. The experimental observation that the NMR spectra linewidths broaden with Cd substitution indicates that Cd impurities induce AF puddles around them. The size of these AF regions shrinks with pressure, which increases this hybridization towards its value in the absence of disorder, ultimately yielding a revival of superconductivity (SC). However, as indicated by NMR relaxation rate $1/T_1$ measurements, the resulting phase is quite heterogeneous [13] not unlike the stripe and nematic orders which coexist with superconductivity in the cuprates. Prior theoretical work examined domains within a mean-field theory of competing AF and SC orders [13].

A useful initial route to a better understanding of the mechanisms of the evolution of the NMR relaxation rate $1/T_1$ is to single out the contributions from the spin degrees of freedom. This is the approach we follow here in which two antiferromagnetically coupled layers give rise to a competition between an interlayer-singlet-rich and an AF phase. By allowing dilution in the second layer, thus breaking interlayer singlets, one mimics the reduction of c-f hybridization. In addition, from a pragmatic perspective, accessible system sizes for QMC simulations of itinerant fermion systems are not sufficiently large to encompass multiple impurities and carefully study finite-size effects. However, it is known that many of the qualitative features of itinerant AF models, such as the Hubbard Hamiltonian, are reflected in their spin counterparts [14,15], notably the successful modeling of the Knight-shift anomaly, certain aspects of which can be captured either with descriptions in terms of localized spins [16] or itinerant electrons [17]. This paper reports QMC simulations of a disordered bilayer Heisenberg Hamiltonian, characterizing its physics within an exact treatment of quantum many-body fluctuations. Key findings are as follows: (i) An appropriately defined impurity susceptibility captures both the inhibition of AF order deep in the ordered phase and its sharp enhancement near the QCP; (ii) quantitative determination of the AF regions induced by impurities and the criterion for their coalescence into a state with long-range order at experimentally relevant temperature scales; (iii) verification of the suggestion that $1/T_1$ is very weakly temperature dependent at the QCP in the clean system. We also establish that the local value of $1/T_1$ at an impurity site increases abruptly at the QCP and resembles the behavior of the clean system "far away" from the impurity.

QMC, in combination with analytic scaling arguments, has previously been used to study the loss of magnetic order and multicritical points in bilayers where the dilution at a given site discards simultaneously the spins in *both* layers [18]. Interesting topological considerations arise from the removal of a *single* spin from a bilayer system in the singlet phase since an unpaired spin-1/2 object is left behind [19]. QMC has been used to study the spin texture produced by a single impurity [20] as well as the onset of AF order in lattices of dimerized chains [21].

II. MODEL AND METHODS

We consider the spin-1/2 AF Heisenberg bilayer Hamiltonian,

$$H = \sum_{\langle ij\rangle,\alpha} J^{\alpha}\vec{S}_i^{\alpha}\cdot\vec{S}_j^{\alpha} + g\sum_i \vec{S}_i^{1}\cdot\vec{S}_i^{2}, \tag{1}$$

where subscripts i,j denote spatial sites on a square lattice and superscripts $\alpha = 1,2$ label the two layers. We study the case when the intraplane exchange constants $J^{\alpha} = J$ are the same, and we choose $J = 1$ to set the energy scale. g is the interlayer exchange.

The Heisenberg bilayer model considered here describes the competition between AF order and singlet formation, such as in the Kondo effect. The spin-1/2 Heisenberg Hamiltonian has been studied widely as a model of quantum magnetism, in particular, as the AF parent compounds of the cuprate superconductors. In that context, the establishment, via QMC simulations, that long-range order (LRO) occurs in the ground state of the Heisenberg model on a square lattice, i.e., $g = 0$ in Eq. (1) [22] was followed by the demonstration that LRO is also present at $T = 0$ in the half-filled fermion Hubbard Hamiltonian [23,24], emphasizing similarities between the two models, in the insulating phase of the latter.

In the absence of disorder, the AF-singlet transition has been located to high accuracy through finite-size extrapolation of the AF order parameter. The square of the order parameter sums the spin-spin correlations throughout the lattice, normalized to the lattice volume N. If those correlations are short ranged (e.g., decaying exponentially), the local contributions to the sum, when divided by N, vanish. If the correlations extend over the entire lattice, then the order parameter is nonvanishing. In practice, in QMC simulations, careful finite-size scaling is essential to demonstrate LRO. See also Eq. (1) and Fig. (S1) of the Supplemental Material for further discussions [25]. Our focus here will be on the nature of these correlations in the neighborhood of an impurity,

$$\langle m^2\rangle = \left\langle \left(\frac{1}{N}\sum_i (-1)^{x_i+y_i+\alpha} S_i^{\alpha}\right)^2\right\rangle. \tag{2}$$

For the symmetric case [26,27] $J^1 = J^2$, the critical interlayer exchange is $g_c = 2.5220$. For the "Kondo-like" lattice where one of the intralayer J's is zero, $g_c = 1.3888$.

As in Ref. [26], we use the stochastic series expansion (SSE) method to obtain $\langle m^2\rangle$. SSE samples terms in a power expansion of $e^{-\beta\hat{H}}$ in the partition function using operator loop (cluster) updates to perform the sampling efficiently [28]. Here we consider bilayer systems with $N = 2\times L\times L$ and L up to 100 sites.

We also evaluate the NMR relaxation rate, given by the low-frequency limit of the dynamic susceptibility,

$$\frac{1}{T_1} = T\lim_{\omega\to 0}\sum_q A^2\frac{\chi''(q,\omega)}{\omega}, \tag{3}$$

where A is the hyperfine coupling and T is the temperature. We obtain $1/T_1$ using the long imaginary-time behavior of the spin-spin correlation function,

$$\frac{1}{T_1} = \frac{A^2}{\pi^2 T}\langle S_i(\tau = \beta/2)S_i(\tau = 0)\rangle; \tag{4}$$

the regime of validity of Eq. (4) is discussed in Ref. [29].

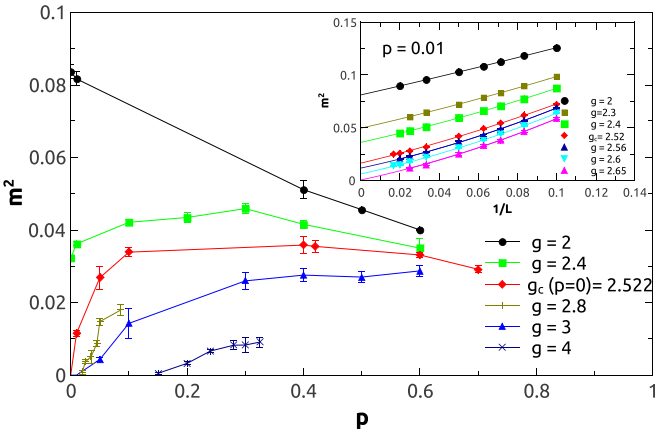

FIG. 1. The square of the staggered magnetization $\langle m^2\rangle$ as a function of the impurity concentration p for different g's. In the AF phase with $g = 2 < g_c(p = 0) = 2.522$, impurities reduce the order. The effect of impurities near the QCP and in the singlet phase is discussed in the text. The inset: Finite-size scaling of $\langle m^2\rangle$ for $p = 0.01$. The position of the QCP is increased to $g_c(p = 0.01) = 2.65$. Data were averaged over 120 disorder realizations. The inverse temperature is $\beta = 80$.

III. AF DOMAINS AND IMPURITY SUSCEPTIBILITY

For a given lattice size L and one disorder realization (i.e., random removal of a fraction p of the spins on layer $\alpha = 2$), we perform the simulations to obtain the quantities of interest; these are then averaged over about 120 disorder realizations. The inset of Fig. 1 shows an example of the size dependence of the AF order parameter thus calculated for a given impurity concentration and different values of g: The intercepts with the vertical axes provide the extrapolated ($L\to\infty$) values for the given p appearing in the main body of the figure. As expected, impurities decrease $\langle m^2\rangle$ deep within the AF phase ($g = 2$) where they act to reduce the average coordination number of the lattice and hence the tendency to order. Closer to the QCP, a different behavior emerges. Impurities begin to inhibit singlet formation by leaving unpaired moments on their partner sites. The AF order parameter, which had been disrupted by singlet formation, therefore increases with p for $g \lesssim g_c$ and does so especially sharply at $g = g_c$. For $g > g_c$, sufficiently large p can induce AF order, even though these larger interplanar couplings would result in singlet formation in the pure case. The appearance of a finite p_c for $g > g_c$ is discussed further below.

The effect of impurities on the AF order parameter can be characterized by an impurity susceptibility,

$$\chi_{\rm imp} = \left.\frac{d\langle m^2\rangle}{dp}\right|_{p=0}, \tag{5}$$

which, as shown in Fig. 2, has a sharp peak at g_c. The effect of impurities is especially large close to the QCP

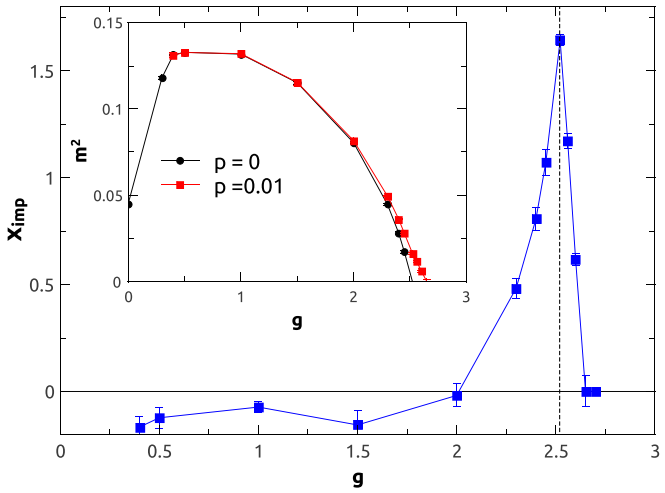

FIG. 2. The impurity susceptibility $\chi_{\rm imp}$ is sharply peaked at g_c (vertical dashed line): Impurities induce AF order. Away from g_c, $\chi_{\rm imp} < 0$: Impurities reduce the AF order parameter. The inset shows the g dependence of $\langle m^2\rangle$ for $p = 0.01$ (squares) and the clean system (circles); these data were used to obtain $\chi_{\rm imp}$. Both the shift in g_c and the large effect of impurities at the QCP are evident. Data for $\langle m^2\rangle$ result from extrapolations to $L = \infty$. The inverse temperature is $\beta = 80$.

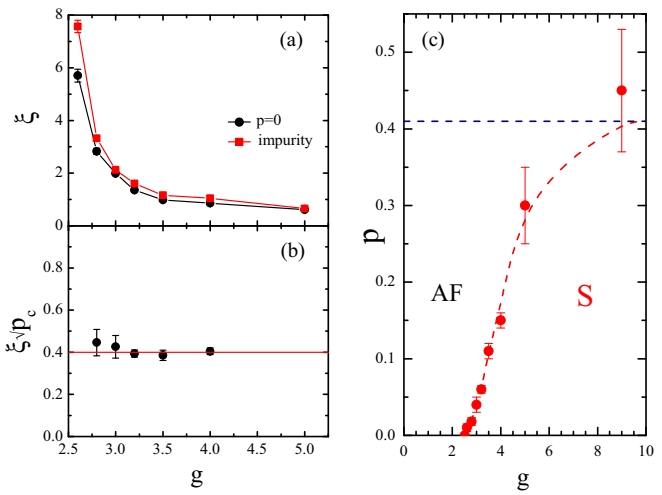

FIG. 3. (a) Correlation length ξ as a function of g. Data are shown around a single impurity (square) and for the clean system (circles). (b) $\xi\sqrt{p_c}$ is roughly constant, consistent with a picture where AF order occurs when the mean impurity separation $\langle l\rangle$ is proportional to ξ. Data for inverse temperatures $\beta = 40,80$ were compared to ensure convergence to the ground state. L up to 100 was used to calculate ξ. (c) The AF order parameter at fixed $\beta = 80$ exhibits a sharp crossover indicating the position of the enhanced range of AF order created by the spin-1/2 impurities.

where the system in delicately poised between two phases. Farther away from the QCP in the AF phase $g \lesssim 2$, the impurity susceptibility is negative as in the two-dimensional (2D) Heisenberg model with site dilution [30].

For $g > g_c$, impurities induce AF order in an otherwise singlet phase [31]. We estimate the critical impurity concentration as follows: Prior to the establishment of order, the coupling between two regions centered at sites i and j will oscillate in phase with an amplitude which decays exponentially [32–34] $J_{\rm eff} \approx J(-1)^{-|i-j+1|}\exp(-\langle l\rangle/\xi)$. Here $\langle l\rangle$ is the mean impurity separation, and ξ is the correlation length in the clean system. For 2D, $\langle l\rangle = 1/\sqrt{p}$. Assuming that the AF order will set in when the average distance between the impurities is on the same scale as ξ yields $\xi\sqrt{p_c} \approx 1$. For a dilute system, we compute ξ by embedding a single impurity in the lattice and evaluating the decay of the spin-spin correlation in its vicinity; see the Supplemental Material [25]. Figure 3(a) shows the resulting ξ, and panel (b) validates the picture that the critical concentration of impurities to induce AF order occurs when $\langle l\rangle = 1/\sqrt{p} \propto \xi$.

There are several subtleties to this argument. At $T = 0$, an exponentially small interaction between impurities can induce order [35]. (This occurs despite the fact that some impurity pairs, which are sufficiently close spatially, lock into singlets [36].) This suggests $p_c = 0$ throughout the singlet phase—an arbitrarily small number of impurities will order despite the rapid decay of their coupling. The effect of these very small couplings is, however, seen only at extremely low T, a fact that is reflected in SSE simulations [35] by the need to study inverse temperatures $\beta \sim 10^4$–10^5 (except very close to the QCP where ξ diverges). In contrast, β which is two to three orders of magnitude smaller is sufficient to reach the ground state on lattices of $L \sim 60$ studied here.

The ordering temperatures in Cd-doped $CeCoIn_5$ are about 2–5 K, and the c-f coupling is reported to be around 49 meV so that $T_c \sim 10^{-2}J$. Thus a more refined interpretation of Fig. 3(b) is that, although AF likely exists at infinitesimal p_c strictly at $T = 0$, panel (b) gives the effective critical impurity concentration to induce AF in the singlet phase at experimental temperature scales [37]. Figure 3(c) shows the position of this sharp crossover in the AF order parameter.

IV. UNIVERSAL BEHAVIOR OF THE NMR RELAXATION RATE

The NMR spin-relaxation rate $1/T_1$ provides an experimental window into doped heavy-fermion materials. Secondary spectral peaks and broadening of the main line implicate the presence of inhomogeneous environments [13]. Here we provide a quantitative description of the effect of impurities on $1/T_1$ and demonstrate that these provide a crisp signature at the QCP.

The main panel of Fig. 4(a) shows the evolution of $1/T_1$ with interlayer coupling at different fixed temperatures for the clean case: It follows the same trend as the AF order parameter $\langle m^2\rangle$, i.e., it initially rises as the two planes are coupled, has a maximum for $g \approx 0.5$, and then decreases to small values at the QCP. The inset of Fig. 4(a) emphasizes the common crossing point at $g_c \sim 2.52$, which is indicative of a very weak T dependence of $1/T_1$ as the QCP is approached. Indeed, this behavior is consistent with early predictions [38] of universality in clean two-dimensional quantum antiferromagnets, according to which $1/T_1 \sim T^\eta$ with $\eta \approx 0.0375$ [39] and in the Kondo lattice model [40]; more on this in the Supplemental Material [25].

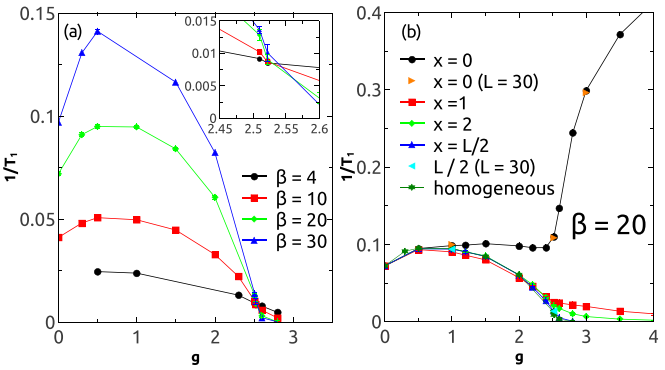

FIG. 4. (a) $1/T_1$ as a function of g for different values of β. The linear size is $L = 50,60$. The inset: blowup of the crossing point. $g < g_c$, $g = g_c$, and $g > g_c$. (b) g dependence of $1/T_1$ for a system with a single removed spin. $x = 0,1,2,L/2$ are different horizontal distances from the impurity. See the text. For the impurity system the linear lattice size is $L = 20,30$.

The behavior of $1/T_1$ in the presence of disorder is shown in Fig. 4(b). We consider the simplest case of a single spin removed from one layer and compute the relaxation rate of spins in the pure layer as a function of distance x on a horizontal line from the removed site. $x = 0$ corresponds to the removed spin's immediate partner, whereas $x = 1,2$ are near and next-nearest neighbors and finally at $x = L/2$, far away from the impurity. For $x = 0$ the partner shows a sharp QCP signature. Above $g = g_c$ when all the other spins are locked in singlets, the free spin-1/2 left behind by spin removal has a greatly enhanced $1/T_1$. Meanwhile, the relaxation rate is small for all other sites. For $g = 0$ the spins on the undiluted plane are decoupled from the second layer and hence share a common value of $1/T_1$ regardless of impurities. Figure 4(b) indicates this independent plane behavior extends out to $g \lesssim g_c$ for $x \geqslant 1$. The curve for $x = 0$ breaks away for $g \geqslant 1$ and has a sharp increase at the QCP. Comparison with the results for the clean system shows that $1/T_1$ on the farthest spin is unaffected by the impurity as observed experimentally [37].

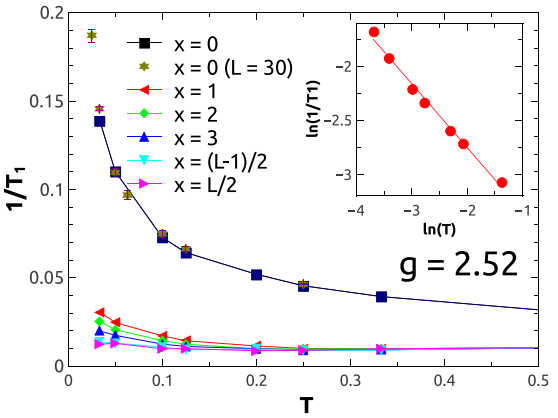

FIG. 5. T dependence of $1/T_1$ for separations x from the impurity. See the text. The inset: determination of the η' exponent. Linear lattices sizes were $L = 20,30$, somewhat smaller than in previous figures because of the necessity to compute the imaginary time-dependent correlation functions.

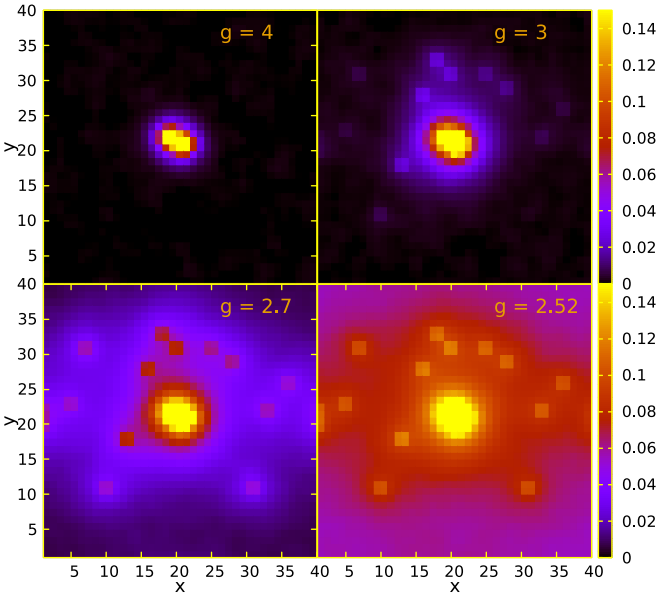

FIG. 6. AF correlation $C(\mathbf{r})$ of the spin at an impurity site with the other spins in the layer $\alpha = 1$. $g = 4$, 3, 2.7, and 2.52. See the text.

We conclude by computing the T dependence of $1/T_1$ at the QCP $g = 2.52$ for this same collection of sites. As emphasized by Fig. 5, $1/T_1$ is weakly temperature dependent away from the impurity site. For the spin left behind at $x = 0$, $1/T_1$ increases substantially as T is lowered and can be described by a power law (the inset). Sachdev *et al.* have argued [19] that the imaginary-time autocorrelation function of an impurity at the QCP scales as $S_i(\tau)S_i(0) \sim \tau^{\eta'}$, implying, through Eq. (4), that $1/T_1 \sim T^{(\eta'-1)}$. Here we obtain $\eta' = 0.41 \pm 0.03$, in agreement with Ref. [20].

Finally, we study correlations between AF puddles believed to form around Cd sites. Figure 6 shows the AF correlation function $C(\mathbf{r}) = (-1)^{\vec{r}}\langle S_{\mathbf{i}}^{1,z} S_{\mathbf{i}+\mathbf{r}}^{1,z}\rangle$, around an impurity site $\mathbf{i}$, at the center of the lattice. For $g = 4$ only spins in the close vicinity of an impurity at the lattice center are correlated with it. When $g = 3$ other impurity locations become evident. Correlations start to become substantial over the whole lattice at $g \sim 2.7$. For an impurity density $p = 0.01$ (16 impurities on a 40×40 lattice), enhanced spin-correlation values $g_c = 2.52$ (bottom right panel) are consistent with the establishment of a nonzero order parameter value of $m^2 \sim 0.02$ in Fig. 1.

V. CONCLUSIONS AND OUTLOOK

Exploration of randomness and dilution effects on magnetic and superconducting order is crucial to understanding disordered strongly interacting quantum systems, such as heavy fermions and cuprates. Impurities reduce order but also nucleate ordered domains which, when sufficiently proximate, coalesce to create long-range order [41–43]. We brought together exact QMC calculations of the effect of impurities on spin correlations/domains and the NMR relaxation rate as a system is tuned through a QCP.

Our key conclusions are as follows: (i) The impurity susceptibility, defined as the response of the AF order parameter

to the removal of a small number of spins, exhibits a sharp peak at the QCP so that low disorder concentrations readily lead to long-range order; (ii) the critical concentration p_c for randomness to induce long-range AF order in the singlet phase, at moderate β, is well described by $\xi\sqrt{p_c} \sim 0.4$, where ξ is the spin-correlation length at $g > g_c$; and (iii) verification that the NMR relaxation rate is nearly temperature independent at the QCP and that an abrupt increase in the local value of $1/T_1$ on an impurity site provides a clear signature of the QCP.

Our paper focused on localized Heisenberg spins. Analogous studies dealing with itinerant electrons, such as the periodic Anderson model, are underway [44]. In that case, an impurity is modeled by a site with reduced c-f hybridization $V^* < V$, and similar to our case, it becomes increasingly effective at inducing AF correlations as the AFM-singlet transition is approached.

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