Susceptibility of the 2D $S = \frac{1}{2}$ Heisenberg antiferromagnet with an impurity

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We use a quantum Monte Carlo method (stochastic series expansion) to study the effects of a magnetic or nonmagnetic impurity on the magnetic susceptibility of the two-dimensional Heisenberg antiferromagnet. At low temperatures, we find a log-divergent contribution to the transverse susceptibility. We also introduce an effective few-spin model that can quantitatively capture the differences between magnetic and nonmagnetic impurities at high and intermediate temperatures.

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Static impurities can be introduced into the CuO$_2$ planes of the high-$T_c$ cuprates as a means of probing their electronic correlations and excitations; some unwanted impurities and defects are also always present in "pure" systems. Impurities coupled to a two-dimensional (2D) host system hence constitute an important class of quantum many-body problems. When Cu ions in hole-doped CuO$_2$ planes are substituted with Zn, experiments indicate that local magnetic moments are induced at the Cu sites neighboring the nonmagnetic Zn impurities. This behavior can be reproduced in spin-gapped Heisenberg antiferromagnets, e.g., in systems of weakly coupled spin-$\frac{1}{2}$ ladders. Although spin models cannot address questions specifically related to a metallic or superconducting host, static impurities in various gapped (paramagnetic) and gapless (antiferromagnetic) Heisenberg systems are important limiting models for understanding the physics of quantum spin defects. They also have direct experimental realizations in the cuprates.

Several studies have addressed static vacancies and added spins in 2D $S = 1/2$ Heisenberg models. The localized moments forming in spin-gapped systems are now well understood. Recently, a universal behavior was predicted for the $T > 0$ impurity effects in systems that have long-range order at $T = 0$ (i.e., in the "renormalized classical" regime), as well as in systems close to a quantum-critical point. Some predictions for quantum-critical systems have been confirmed numerically, whereas other aspects of the theory remain unsettled. The predictions in the renormalized-classical regime have not yet been tested.

In this Letter, we consider the 2D square-lattice spin-$\frac{1}{2}$ Heisenberg antiferromagnet with (i) a vacancy (nonmagnetic impurity) or (ii) an off-plane added spin (magnetic impurity) coupled to a single host spin. We perform quantum Monte Carlo calculations, using the stochastic series expansion (SSE) technique, and determine the effects of the two different impurities on the uniform magnetic susceptibility. Our results confirm quantitatively a classical-like longitudinal Curie contribution resulting from alignment of the impurity moment with the local Néel order. However, we also find evidence of a logarithmic divergence as $T \to 0$ in the transverse component of the impurity susceptibility, instead of the predicted $T$ independence at low $T$. We point out qualitative differences between the vacancy and the added spin at high and intermediate $T$ and explain these in terms of an effective few-spin model.

We begin by defining three spin-$\frac{1}{2}$ Hamiltonians describing the models investigated:

\begin{align}
H_0 &= J \sum_{\langle i,j \rangle} S_i \cdot S_j, \\
H_- &= J \sum_{\langle i,j \rangle \neq 0} S_i \cdot S_j, \\
H_+ &= J \sum_{\langle i,j \rangle} S_i \cdot S_j + J_\perp S_x \cdot S_0,
\end{align}

where $\langle i,j \rangle$ denotes a pair of nearest-neighbor sites on a periodic $L \times L$ lattice. $H_0$ is the standard Heisenberg model. In $H_-$ the spin $S_0$ has been removed, creating a vacancy. We also study systems with two vacancies at maximum separation on different sublattices. In $H_+$ an added spin-$\frac{1}{2}$, $S_x$, is coupled to one of the spins, $S_0$, in the plane. We here only consider $J_\perp = J$.

We have calculated the total susceptibilities

\begin{equation}
\chi^z_k = \frac{1}{T} \left\langle \left( \sum_{i=1}^{N} S^z_i \right)^2 \right\rangle,
\end{equation}

with $k = 0, -, +$ corresponding to $H_0$, $H_-$, and $H_+$. We have used square lattices with $L = 4, 8, 16, 32$, and 64, at temperatures down to $T = J/32$. The number of spins $N = L^2$ for $k = 0$ and $L^2 \pm 1$ for $k = \pm$. In order to determine the effects of the impurities, we follow Ref. and define the impurity susceptibilities

\begin{equation}
\chi^{z,\pm}_{\text{imp}} = \chi^z_{\pm} - \chi^z_0.
\end{equation}

In the case of two vacancies, the impurity susceptibility is further normalized by a factor $\frac{1}{2}$, so that for $L \to \infty$ it should be the same as for a single vacancy.

The temperature dependence of $\chi^{z,\pm}_{\text{imp}}$ was discussed by Sachdev et al. on the basis of quantum field theory.
They concluded that at $T = 0$, the component $\chi_{\text{imp}}^\parallel$ parallel to the direction of the Néel order vanishes, while the perpendicular component $\chi_{\text{imp}}^\perp = C_3 / \rho_s$, where $C_3$ is a constant and $\rho_s$ is the spin stiffness of the host. For $T > 0$ the correlation length $\xi$ grows exponentially as $T \to 0$. For a general impurity spin, a moment of magnitude $T > 0$, the quantum mechanical magnitude might have been naively expected. The full low-$T$ impurity susceptibility is hence $\chi_{\text{imp}} = \frac{1}{3} T^2 + \frac{2}{3} \chi_{\text{imp}}^\perp$.

In order to have a simple system which reproduces this expected low-$T$ behavior, and also accounts for non-universal behavior at higher $T$ for different types of impurities, we introduce a simple effective few-spin model. The idea is to model a large Néel-ordered region by a classical vector $\mathbf{N}$. Three Hamiltonians corresponding to the original models (1) are defined:

$$H_0^{\text{eff}} = \alpha \mathbf{S}_0 \cdot \mathbf{S}_e + r \mathbf{N} \cdot \mathbf{S}_e - h_z M_0^z,$$
$$H_\perp^{\text{eff}} = r \mathbf{N} \cdot \mathbf{S}_e - h_z M_0^z,$$
$$H_\parallel^{\text{eff}} = \alpha \mathbf{S}_0 \cdot \mathbf{S}_e + r \mathbf{N} \cdot \mathbf{S}_e + J_{\perp} \mathbf{S}_a \cdot \mathbf{S}_0 - h_z M_a^z.$$ (4a, 4b, 4c)

The effective Hamiltonian for the vacancy model, $H_0^{\text{eff}}$, contains a single spin $\mathbf{S}_e$ representing a remnant spin-$\frac{1}{2}$ due to the sublattice asymmetry caused by the vacancy. It is coupled to the unit vector $\mathbf{N}$, representing the orientation of the local Néel order of the host antiferromagnet. The magnitude of this order is absorbed into the effective coupling $r$. In $H_0^{\text{eff}}$ we “reinsert” the spin $\mathbf{S}_0$ that was removed in the vacancy model, and couple it to $\mathbf{S}_e$. Further, including the Heisenberg interaction between the host spin $\mathbf{S}_0$ and the added spin $\mathbf{S}_a$, we arrive at the effective Hamiltonian for the added spin model $H_\parallel^{\text{eff}}$.

We determine the impurity susceptibilities (3) of the effective models in the same way as for the original models. In Eqs. (1) we have explicitly indicated how an applied external field, which defines the $z$ direction, couples to the systems. The magnetization operators are $M_k^z = S_k^z$, $M_0^z = S_0^z + S_e^z$, and $M_a^z = S_a^z + S_0^z + S_e^z$. The susceptibilities for $k = 0, -, +$, corresponding to $H_0^{\text{eff}}$, $H_\perp^{\text{eff}}$, and $H_\parallel^{\text{eff}}$, can be written in the form

$$\chi_k = \frac{\partial}{\partial h_z} \langle M_k^z \rangle = \frac{1}{3} \chi_k^\parallel + \frac{2}{3} \chi_k^\perp,$$ (5)

where $\parallel$ and $\perp$ refer to the directions parallel and perpendicular to the vector $\mathbf{N}$. After straightforward diagonalization of the effective Hamiltonians in the $\parallel$ basis, the two components can be evaluated using

$$\chi_k^\parallel = \frac{1}{T} \langle (M_k^\parallel)^2 \rangle,$$ (6a)
$$\chi_k^\perp = \int_0^{1/T} d\tau \langle M_k^\parallel (\tau) M_k^\perp (0) \rangle.$$ (6b)

In an average over all orientations of $\mathbf{N}$ relative to the fixed $z$ axis has been taken. The coupling of the impurity spin to $\mathbf{N}$, therefore, and in accordance with Ref. 12, leads to a classical-like low-$T$ divergence: $\chi_{\text{imp}} \sim \frac{1}{T} \frac{1}{\rho_s^2} + \frac{2}{3} \chi_{\text{imp}}^\perp$, where the second term is constant at low $T$ (instead of having the $r = 0$ form $\frac{1}{T}$).

We do not attempt to derive values for the couplings $\alpha$ and $r$ [however, $J_{\perp} = J = \frac{1}{2}$, and $r/J = 1.75$, which give a reasonable over-all agreement with the SSE calculations. We will show that although the effective model is highly simplified, it captures some of the differences between the vacancy and the added spin.

Next, we present the results of the SSE calculations for the models (1) and compare with the corresponding effective models. The impurity susceptibilities of interest, Eq. (2), are defined as differences between two extensive susceptibilities for $L = 64$ simulation data and the effective model.

In Fig. 1 an average over all orientations of $\mathbf{N}$ relative to the fixed $z$ axis has been taken. The coupling of the impurity spin to $\mathbf{N}$, therefore, and in accordance with Ref. 12, leads to a classical-like low-$T$ divergence: $\chi_{\text{imp}} \sim \frac{1}{T} \frac{1}{\rho_s^2} + \frac{2}{3} \chi_{\text{imp}}^\perp$, where the second term is constant at low $T$ (instead of having the $r = 0$ form $\frac{1}{T}$).

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The single-vacancy impurity susceptibility multiplied by $4T$ is shown in Fig. 1. At high $T$ the data for different $L$ are indistinguishable, while at low $T$ finite-size effects are clearly seen for $L \leq 16$. The range of $T$ considered, all finite-size effects are eliminated within statistical errors for $L = 64$. The observed behavior at high $T$ is easily understood as the total susceptibility is then just the sum of Curie contributions of each independent spin; $\chi_{\text{imp}}^\perp (T \to \infty) = (L^2 - 1)/4T^2 - L^2/4T^2 = \frac{1}{4T}$. At low $T$ the $S = \frac{1}{2}$ ground state of $H_\perp$ and $S = 0$ of $H_0$ lead to the observed Curie behavior $\chi_{\text{imp}}^\perp = \frac{1}{4T}$ for small $L$. **FIG. 1: Impurity susceptibilities for different system sizes $L$ with one vacancy. Error bars are smaller than the symbols. The inset shows a comparison between $L = 64$ simulation data and the effective model.**
As $L$ grows this finite-size effect vanishes, and we observe the predicted $\frac{1}{12T}$ contribution arising from the longitudinal part, in analogy with the effective model. Hence the nearly constant $4T\chi_{\text{imp}}^{z,1} \approx \frac{1}{3}$ for $L = 32$ and 64. The effective model reproduces the behavior reasonably well, as shown in the inset of Fig. 1.

Looking more carefully at the SSE data, the predicted low-$T$ constant behavior in the quantity $\chi_{\text{imp}}^{z,1} - \frac{1}{12T}$, which should reduce to $\frac{2}{3}\chi_{\text{imp}}^{1,1}$ as $T \to 0$, is not observed. In Fig. 2 (solid circles) we present results for $\chi_{\text{imp}}^{z,1} - \frac{1}{12T}$ for $L = 64$ (data for smaller $L$ indicate that there are no significant finite-size effects for $L = 64$). It shows an apparent logarithmically divergent behavior, roughly from the onset $T$ of renormalized-classical behavior in the pure 2D Heisenberg susceptibility, $T/J \approx 0.3$. This is also approximately where the corresponding effective-model result converges to a constant.

Results for two vacancies are shown in Fig. 3. The high-$T$ behavior has the same explanation as for the single vacancy. At low $T$, the moments due to the two vacancies, which reside on different sublattices, are pinned by the Néel order antiparallel to each other, resulting in a vanishing $\chi_{\text{imp}}^{||}$. Hence, in this case $\chi_{\text{imp}}^{z,1}$ does not diverge for small $L$ and we do not, therefore, multiply our results by $T$. The inset in Fig. 3 shows a comparison between one and two vacancies for $L = 16$. The $T$ at which the two curves deviate corresponds to a correlation length of the same order as the separation between the two impurities, $\xi \approx L/2$, i.e., above this $T$ the two impurities couple to different Néel domains and behave as independent vacancies. Since $\xi$ diverges exponentially, the point of deviation moves very slowly towards $T = 0$ as $L$ grows. At low $T$ the resulting $\chi_{\text{imp}}^{z,1} / T$ for two vacancies shows little $T$ dependence for large $L$. However, no sign of convergence of the plateau value is seen as the system size grows. If $\chi_{\text{imp}}^{z,1}$ is finite as $T \to 0$, we would expect such a convergence at low $T$ (with a peak at intermediate $T$ for very large $L$, where the cut-off of the $\frac{1}{12T}$ divergence occurs at low $T$). It should be noted that in a finite system there will always be some interactions also between the $\perp$ components of the two impurity spins at low $T$. We can therefore not expect the behavior for two vacancies to be given in a straightforward way by the single-vacancy results in Fig. 2. The roughly $\ln(L)$ divergence of the plateau height in Fig. 3 is, however, fully in line with a log-divergent $\chi_{\text{imp}}^{z,1}$ for a single vacancy.

Results for $\chi_{\text{imp}}^{z,1}$ for systems with an off-plane added spin are shown in Fig. 4. The high-$T$ behavior, as well as the low-$T$ behavior for small systems ($L = 4, 8$), is understood by the same arguments as for the vacancy. Note that in this case the high-$T$ impurity susceptibility is $\frac{1}{12T}$, instead of $\frac{1}{12T}$ for the vacancy. Again, we believe that all finite-size effects are eliminated for $L = 64$ down to $T = J/32$. Contrary to the behavior for the vacancy in Fig. 4 the $T$ at which $4T\chi_{\text{imp}}^{z,1}$ assumes an almost constant value $\frac{1}{3}$ has not yet been reached at $T = J/32$. Moreover, we note the substantial differences between $\chi_{\text{imp}}^{z,1}$ and $\chi_{\text{imp}}^{z,1}$ at intermediate $T$. In particular, the shoulder seen in Fig. 4 for $T \sim 0.2 - 1.2$ has no counterpart in Fig. 4. This feature is clearly due to the internal structure of the added spin impurity and is reproduced very well by the effective model. In Fig. 2 (open circles) we present the results for $\chi_{\text{imp}}^{z,1} - \frac{1}{12T}$ for $L = 64$. As in the vacancy case, it appears to be log-divergent at low $T$. Note also that the effective model describes the added spin impurity very well down to quite low tem-

FIG. 2: $\chi_{\text{imp}}^{z,1} - \frac{1}{12T}$ for systems of size $L = 64$ with a vacancy and an added spin. The straight lines are fits to the low-$T$ simulation data.

FIG. 3: Impurity susceptibility for systems with two vacancies. The inset shows results for $L = 16$ systems with one and two vacancies.
Fig. 4: Impurity susceptibilities for different system sizes $L$ with an off-plane added spin. The inset shows a comparison with the effective model.

temperatures. It captures the behavior better than for the vacancy, but a better agreement for the vacancy can also be achieved by using slightly different $\alpha$ and $r$.

Summarizing our results, we have confirmed the predicted $\ln(1/T)$ contribution to the impurity susceptibility. However, we also find a logarithmic divergence of $\chi_{\text{imp}}^{\perp}$ instead of the constant $\frac{2}{3}\chi_{\text{imp}}^{\perp}$ predicted for this quantity at low $T$ (and reproduced with our effective model). We cannot, of course, completely exclude an approach to a constant at still lower temperatures, but such a slow convergence had also not been anticipated. We note that the separation of $\chi_{\text{imp}}^{\perp}$ into transverse and longitudinal components, Eq. (6), is strictly correct for the 2D Heisenberg model only at $T = 0$. However, the very sudden cutoff of the divergence seen in the two-vacancy data in Fig. 3 supports the notion of a component aligning very strongly to the local Néel order (which becomes the global order at the $L$-dependent crossover $T$) and justifies the relation $\frac{2}{3}\chi_{\text{imp}}^{\perp} = \chi_{\text{imp}}^{\perp} - \frac{1}{2}\chi_{\text{imp}}^{\parallel}$ also at relatively high temperatures. Hence, our results are most naturally interpreted as a log divergent $\chi_{\text{imp}}^{\perp}$. This is in fact in line with a Green’s function calculation by Nagaosa et al. [8]. They found that for a system with a vacancy, the frequency dependent impurity susceptibility at $T = 0$, $\chi_{\text{imp}}^{\perp}(T = 0, \omega)$, was log divergent when $\omega \to 0$. In view of the renormalized-classical picture, this is consistent with exact results [19] for the classical 2D Heisenberg model. An anomalous perpendicular susceptibility was also recently noted for the $S = 1/2$ model at finite impurity concentration [20].

The results presented here call for a reexamination of the field theory [3] of quantum impurities in the renormalized-classical regime. Very recent efforts to explain the log divergence, motivated by our numerical findings, have indicated that the impurity moment acquires a previously unnoticed correction of order $T \ln(1/T)$ to its leading order value $S$ in the renormalized-classical regime [21, 22], and that this can be interpreted as a log-divergent contribution to $\chi_{\text{imp}}^{\perp}$ as proposed here (results in the quantum-critical regime [3] remain unchanged).

Apart from the log divergence, the effective few-spin model that we have introduced here gives a good description of the impurity susceptibility at high and intermediate $T$. In particular, it captures very well the nonmonotonic $T$ dependence that we have found in the case of an added spin.

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