# Knight Shifts around Vacancies in the 2D Heisenberg Model

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The local response to a uniform field around vacancies in the two-dimensional spin-1/2 Heisenberg antiferromagnet is determined by numerical quantum Monte Carlo simulations as a function of temperature. It is possible to separate the Knight shifts into uniform and staggered contributions on the lattice which are analyzed and understood in detail. The contributions show interesting long- and short-range behavior that may be of relevance in NMR and susceptibility measurements. For more than one impurity, remarkable nonlinear enhancement and cancellation effects take place. We predict that the Curie impurity susceptibility will be observable for a random impurity concentration even in the thermodynamic limit.

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The deliberate use of substitutional impurities in strongly correlated electron systems has become a valuable tool for controlled studies of the underlying correlations [1–5]. Doping of nonmagnetic Zn ions in antiferromagnetic $CuO_2$ planes is known to lead to a surprisingly large reduction of $T_c$ [1] and induces local staggered magnetic moments around the impurity sites [2]. It has now become quite standard to analyze the local magnetic moments around static magnetic and nonmagnetic impurities in an antiferromagnetic background for a deeper understanding of the correlated states [3–5].

Theoretically the Knight shifts around impurities and vacancies have been studied for many low-dimensional antiferromagnets [6–12], which typically show a strong enhancement of the antiferromagnetic order. The detailed behavior of the staggered magnetic moments can even give rather exotic results, such as an increasing amplitude as a function of distance [7], and show renormalization effects [11,12]. The magnetization pattern can often be interpreted as the interference of incoming and scattered quasiparticle excitations [11] or in terms of a pruned valence bond basis [8].

The sum of all Knight shifts adds up to the total susceptibility $\chi_1$. The resulting impurity susceptibility $\chi_{\rm imp} = \chi_1 - \chi_0$ has been studied in detail in more recent theoretical studies [13–16]. In systems with long-range magnetic order the leading temperature dependence of the impurity susceptibility from one single vacancy is given by a *classical* Curie spin which has been confirmed by numerical simulations for the 2D Heisenberg antiferromagnet [14], where a subleading logarithmic term has also been established [13,14]:

$$\chi_{\rm imp} = \chi_1 - \chi_0 = \frac{S^2}{3T} + \frac{1}{3\pi\rho_s}\ln\left(\frac{C}{T}\right). \quad (1)$$

Here $\chi_0$ is the total susceptibility without any impurities, $\rho_s$ is the spin stiffness, and the limit of large correlation length $\xi(T)$ and system size $\xi(T) > L \to \infty$ is assumed.

We now want to examine how this impurity susceptibility is distributed on the lattice by considering the linear *local* response to a small uniform magnetic field (Knight shift) at each site

$$\chi(\mathbf{r}) = \beta \sum_i \langle S_i^z S_{\mathbf{r}}^z \rangle. \quad (2)$$

We find that long and short-range patterns of the Knight shifts contribute to the impurity susceptibility differently. We are able to study the interference of several impurities with nontrivial cancellation and enhancement effects over long distances, which allows us to make predictions for a finite impurity density.

In general we can write $\chi(\mathbf{r})$ as a sum of uniform and staggered parts on the lattice

$$\chi(\mathbf{r}) = \chi_{\rm uni}(\mathbf{r}) + (-1)^{r_x + r_y}\chi_{\rm stag}(\mathbf{r}), \quad (3)$$

the amplitudes of which are both slowly varying on the scale of one lattice spacing. In order to extract those two components we extrapolate the data on the even sublattice to the odd sublattice and vice versa and define $\chi_{\rm uni/stag}(\mathbf{r}) = [\chi_{\rm even}(\mathbf{r}) \pm \chi_{\rm odd}(\mathbf{r})]/2$. The model is the 2D Heisenberg spin-1/2 antiferromagnet $H = J\sum_{\langle i,j\rangle} \mathbf{S}_i \cdot \mathbf{S}_j$, where $\langle i, j\rangle$ denotes nearest-neighbor sites on a periodic square lattice. The quantum Monte Carlo program we developed uses the loop algorithm in a single cluster variety implemented in continuous time [17], which gives efficient updates even at very low temperatures on a lattice of $100 \times 100$ sites. For convenience, temperatures and energies are given in units of $J = 1$.

The Knight shift data around one single vacancy are remarkably isotropic as a function of geometrical distance at all temperatures as is shown in Fig. 1 and in the inset of Fig. 2 for the staggered part. Slight anisotropic deviations near the edges ($|\mathbf{r}| \sim 40$–$50$) can be seen due to the finite system size and periodic boundary conditions. The remarkable isotropy on an anisotropic lattice is an indication that the observed behavior is likely due to spin wave excitations, which have an isotropic dispersion at low energies.

 

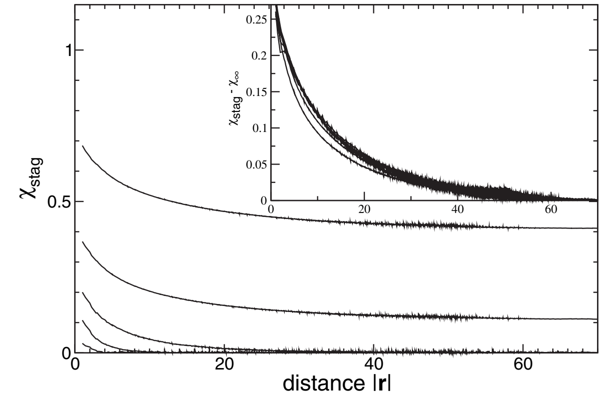


FIG. 1. All data points of $\chi_{\rm stag}(\mathbf{r})$ as a function of distance $|\mathbf{r}|$ from one vacancy for $T = 1$, 0.5, 0.33, 0.2, and 0.1 from below. Inset: All curves collapse to one almost universal, temperature independent shape after subtracting the limiting constant of the induced order $\chi_\infty$ for $0.02 \le T \le 0.2$.

At high temperatures $T$ the impurity effect is confined within the correlation length $\xi(T)$, which has an exponential dependence with $1/T$ [18]. Already for $\beta = 5$ the impurity affects the whole system in our case. This is commonly referred to as the zero temperature limit where Néel order effects can be observed in 2D. In finite systems the effects of this order can be observed already for finite temperatures when the correlation length exceeds the system size $\xi(T) > L$ (in our case when $\beta \gtrsim 5$). However, even at the lowest temperatures $T$ we always remain in the thermodynamic limit; i.e., a large number of excited states contribute to a possible symmetry breaking and the energy level spacing remains very small compared to $T$.

The origin of a classical Curie impurity susceptibility in Eq. (1) can intuitively be understood by considering the classical limit of an Ising system with $N$ spins of size $S$ (here $S = \frac{1}{2}$). Removing one spin on the even sublattice at (0, 0) does not affect the long-range order of such a system. In that case all the spins on the odd sublattice form one large effective spin of size $NS/2$, while the spins on the even sublattice have total spin of $NS/2 - S$. The difference of these two large spins will leave a net classical impurity spin of size $S$, which is free to rotate toward an applied magnetic field. In the 2D Heisenberg antiferromagnet, a similar mechanism can be invoked, except that the long-range order amounts only to about 61% of the classical limit [9,19]. As can be seen in Fig. 1 we can indeed find a field-induced longitudinal long-range staggered order which approaches a limiting constant $\chi_\infty = \chi_{\rm stag}(\infty)$. The value of $\chi_\infty$ approaches a Curie behavior of about $0.6\frac{S^2}{3k_BT}$ as can be seen in the inset of Fig. 3, consistent with the fact that only about 60% of the classically possible long-range order couples to the field. The corresponding contribution to the impurity susceptibility is given by

$$\chi_{\rm imp-lr} = \sum_{\mathbf{r}\neq\mathbf{0}} (-1)^{x+y+1}\chi_\infty = \chi_\infty \sim 0.6\frac{S^2}{3k_BT}. \qquad (4)$$

Since this contribution from long-range order only amounts to 60% of the leading impurity susceptibility in Eq. (1), the remaining 40% and subleading contributions of $\chi_{\rm imp}$ must arise from a more local disturbance of Knight shifts in the vicinity around the vacancy.

The uniform part of the Knight shifts is found to drop off very fast with the distance from the impurity to a value $\chi_{\rm pure}$, which corresponds to the bulk susceptibility in a system without impurities $\chi_0/L^2$. This is shown in Fig. 2 for various temperatures as a function of distance. Closer inspection shows that the behavior on distance is not completely isotropic, since the enhancement is stronger along the lattice axes (at integer distances $|\mathbf{r}| = 1, 2, 3$). The sum of this uniform part gives a net contribution to the impurity susceptibility

$$\chi_{\rm imp-uni} = \sum_{r\neq 0} \chi_{\rm uni}(\mathbf{r}) - \chi_0, \qquad (5)$$

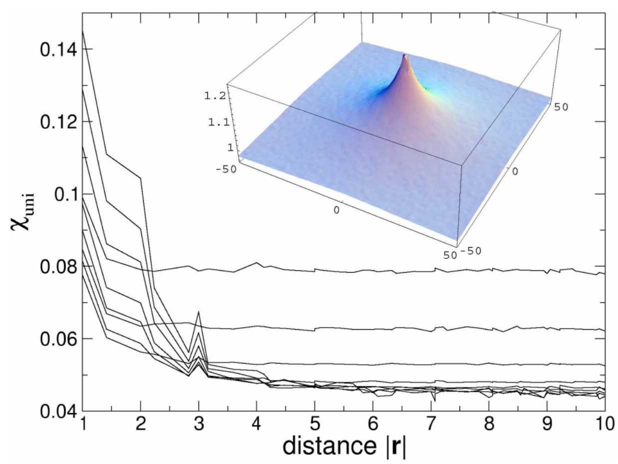


FIG. 2 (color online). The uniform part $\chi_{\rm uni}(\mathbf{r})$ as a function of distance $|\mathbf{r}|$ for $T = 0.5$, 0.33, 0.2, 0.1, 0.067, 0.05, 0.033, 0.025, and 0.02 (from above at $\mathbf{r} = 10$). Inset: The staggered part at $T = 0.05J$.

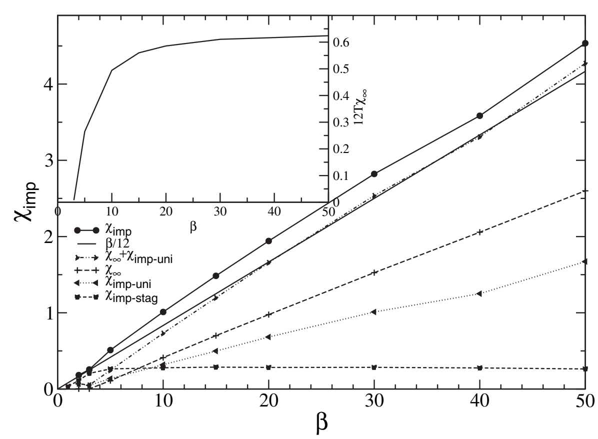


FIG. 3. The different contributions to the impurity susceptibility as a function of $\beta$. The error in the uniform impurity susceptibility is about 0.2 because it is a difference of two large values. Inset: The ratio $12T\chi_\infty$ as a function of $\beta$.

which turns out to contain the remaining 40% of the classical Curie impurity susceptibility of $1/12T$ in Eq. (1). This is shown in Fig. 3 in comparison to other contributions to the impurity susceptibility, where $\chi_{\text{imp}-\text{uni}}$ is seen to be proportional to $\beta$ and the sum $\chi_\infty + \chi_{\text{imp}-\text{uni}}$ follows the predicted Curie behavior $1/12T$ closely.

Finally, there is a staggered pattern of Knight shifts localized around the impurity $\chi_{\text{stag}}(\mathbf{r}) - \chi_\infty$ as shown in the inset of Fig. 1. Interestingly, the dependence on temperature is very weak both in shape and magnitude. The net contribution to the impurity susceptibility

$$\chi_{\text{imp}-\text{stag}} = \sum_{r\neq 0}(-1)^{x+y+1}[\chi_{\text{stag}}(\mathbf{r}) - \chi_\infty] \qquad (6)$$

is also small and contributes only to higher orders of $\chi_{\text{imp}}$ as shown in Fig. 3 compared to the other contributions. Obviously $\chi_{\text{imp}} = \chi_{\text{imp}-\text{stag}} + \chi_{\text{imp}-\text{lr}} + \chi_{\text{imp}-\text{uni}}$.

We now consider the case of two impurities that are separated sufficiently far away from each other, so that any quantum interference can be neglected. As long as the correlation length $\xi$ is smaller than the interimpurity distance, no interference can be found and the impurity contributions to the Knight shifts and the susceptibility simply add. In the low temperature limit the correlation length grows beyond the system size and the staggered order extends through the entire system. In this case, it makes a significant difference if the two vacancies are on the same or on different sublattices.

For two vacancies on *different* sublattices the field-induced long-range order cancels exactly, giving no contribution to the impurity susceptibility. Remarkably, the uniform contribution is also strongly affected as shown in Fig. 4 for impurities located at $(0, 0)$ and $(48, 49)$ in comparison to the uniform Knight shift data of just one impurity. Even though the impurities are located a distance of $|\mathbf{r}| \sim 69$ sites apart, we find a strong reduction of the uniform Knight shifts within just a few lattice sites around each impurity in Fig. 4. This reduction goes far beyond simple interference and must be taken as a collective effect of the entire system. The resulting impurity contribution $\chi_{\text{imp}-\text{uni}}$ from the uniform Knight shifts in Eq. (5) is small and has no Curie-like temperature behavior; i.e., only the subleading corrections remain. On the other hand, the *local staggered* enhancement of the Knight shifts is remarkably unaffected by interference effects, but has the opposite phase around each impurity as shown in the inset of Fig. 4. Therefore, the sum in Eq. (6) gives exactly twice the magnitude of $\chi_{\text{imp}-\text{stag}}$ compared to one single impurity [the opposite phase cancels with the minus sign from the different sublattice in Eq. (6)]. This part $\chi_{\text{imp}-\text{stag}}$ now becomes the main contribution to the impurity susceptibility, but has no divergent Curie-like behavior as discussed above. This is in agreement with findings in Ref. [14] that the impurity susceptibility of two impurities on different sublattices corresponds to subleading terms only.

For two vacancies on the *same* sublattice we find a doubling of both the induced long-range order $2\chi_\infty$ and the uniform enhancement as shown in Fig. 4. Since two vacancy sites contribute in Eqs. (4) and (5), this implies that the Curie-like impurity susceptibilities $\chi_{\text{imp}-\text{lr}}$ and $\chi_{\text{imp}-\text{uni}}$ are *quadrupled* as if two virtual spins form a triplet. However, the local staggered enhancement of Knight shifts close to each impurity again remains almost completely unaffected, compared to the single impurity case in the inset of Fig. 1. The Curie-like contributions $\chi_{\text{imp}-\text{lr}}$ and $\chi_{\text{imp}-\text{uni}}$ therefore increase *quadratically* with the number of same-sublattice impurities $N$, while the subleading contribution $\chi_{\text{imp}-\text{stag}}$ increases linearly with $N$ independent of sublattice $\chi_{\text{imp}} \sim N^2/12T + N\mathcal{O}[\ln(C/T)]$.

In the case of two impurities that are very close to each other, quantum effects are bound to also play a role and a comparison with the interference of two single impurities is not useful. For two neighboring impurities we find that also subleading contributions to the impurity susceptibility are smaller [14].

In an infinite system with a small random impurity concentration $\rho$ the impurities act independently at high and intermediate temperatures. As the temperature is lowered, vacancies within the distance of the exponentially growing correlation length $\xi$ start to feel each other; i.e., the effective number of correlated impurities within an antiferromagnetic domain is $N \propto \xi^2\rho$. The subleading contribution from the local staggered part $\chi_{\text{imp}-\text{stag}}$ always adds independent of sublattice and will therefore be of order $N$, but is not divergent with temperature. For the Curie-like contributions $\chi_{\text{imp}-\text{lr}}$ and $\chi_{\text{imp}-\text{uni}}$ the difference of impurities on the two sublattices plays an important role, which is of order $|N_A - N_B| \sim \sqrt{N}$ [9]. Our simulations

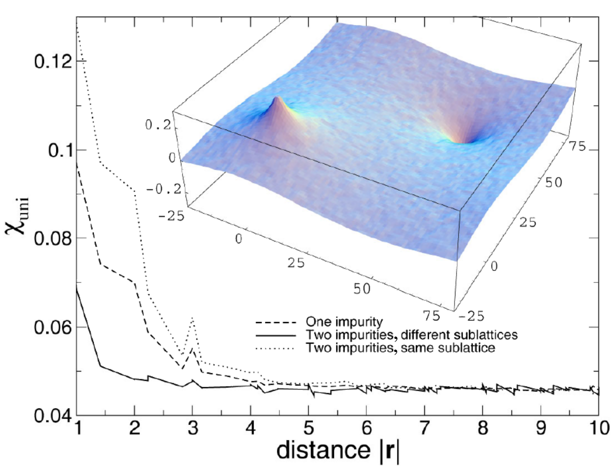


FIG. 4 (color online). The uniform part of the Knight shifts around an impurity at $(0, 0)$ as a function of distance at $\beta = 20$. For a second impurity on the same sublattice $(48, 48)$ (dotted line) we find twice the uniform enhancement compared to the single impurity case (dashed line). For a second impurity on the other sublattice $(48, 49)$ (solid line) there is a strong reduction. Inset: Staggered part for two impurities on different sublattices.

show that the induced long-range order is proportional to the excess of impurities on one sublattice $|N_A - N_B|\chi_\infty \propto \xi\sqrt{\rho}\chi_\infty$, while the uniform enhancement is proportional to $\pm(N_A - N_B)$ on the sublattice $A/B$ up to subleading terms, i.e., *negative* on the minority sublattice. The contributions $\chi_{\mathrm{imp-lr}}$ and $\chi_{\mathrm{imp-uni}}$ in the sums (4) and (5) therefore both increase quadratically with the difference $(N_A - N_B)^2 \sim N$. Hence we predict that a Curie susceptibility of $\rho S^2/3k_BT$ survives even in the thermodynamic limit, contrary to conjectures made in Ref. [9]. In NMR experiments induced staggered Knight shifts of order $\xi\sqrt{\rho}\chi_\infty B$ throughout the lattice should be directly observable.

In conclusion, we have analyzed the local response around vacancies in the 2D Heisenberg antiferromagnet in the low temperature limit. These Knight shifts can be analyzed in terms of staggered and uniform contributions, which are mostly isotropic on the 2D lattice. In the presence of a small magnetic field a single vacancy induces a staggered pattern of Knight shifts over the entire lattice in the low temperature limit, giving a longitudinal Néel order $\chi_\infty \sim 0.6/12T$. This field-induced order corresponds to a free classical moment accounting for 60% of the Curie impurity susceptibility. The remaining 40% of the magnetic moment is found to be located in a uniform enhancement of Knight shifts in the immediate neighborhood of the vacancy. Additionally, a staggered pattern in a finite range around the vacancy is also found, which turns out to have a largely temperature independent, universal shape, and gives rise to subleading corrections of $\chi_{\mathrm{imp}}$.

When more than one vacancy is introduced in the low temperature limit, the Curie terms in the impurity susceptibility cancel if the vacancies are on different sublattices with a remarkable long-distance reduction of the uniform Knight shifts around each vacancy. For vacancies on the same sublattice, both the induced long-range order and the uniform part increase, leading to a Curie-like impurity susceptibility that increases quadratically with the number of impurities. Both uniform and long-range staggered parts appear to be contributing to virtual classical impurity spins that are ferromagnetically or antiferromagnetically linked for impurities on the same/opposite sublattice. The local staggered enhancement around each impurity remains unaffected by interference and temperature in any case, contributing to subleading corrections of $\chi_{\mathrm{imp}}$. In the thermodynamic limit, cancellation and nonlinear enhancement effects even out from statistical arguments, leaving a Curie susceptibility of $\rho/12T$.

Some of the results can be generalized for other antiferromagnets in higher dimensions and/or with higher spin. In particular, a vacancy will always induce a longitudinal antiferromagnetic response $\chi_\infty$ over the entire lattice, the magnitude of which is linked to the order parameter as $T \to 0$ [80% in Eq. (4) for a 3D spin-1/2 model]. The remaining impurity susceptibility in Eq. (1) must be found in a uniform enhancement around the vacancies. The virtual classical impurity spins are antiferromagnetically or ferromagnetically linked depending on the sublattice as seen above. In the spin-1/2 chain, no long-range order and no Curie impurity susceptibility is found. However, the induced Knight shifts increase with a distance from the vacancy [7] and boundary terms give a Curie contribution that is logarithmically reduced for low temperatures from nonuniversal boundary effects [20]. For vacancies in quantum antiferromagnets with a gap, a more local antiferromagnetic pattern is induced [8,9], leading to a Curie susceptibility corresponding to virtual quantum spins, which are not linked to each other. The effect of substitutional impurities with higher spin is much less clear, although a coupling to the antiferromagnetic order can again be expected.

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