Universal Alternating Order around Impurities in Antiferromagnets

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The study of impurities in antiferromagnets is of considerable interest in condensed matter physics. In this Letter we address the elementary question of the effect of vacancies on the orientation of the surrounding magnetic moments in an antiferromagnet. In the presence of a magnetic field, alternating magnetic moments are induced, which can be described by a universal expression that is valid in any ordered antiferromagnet and turns out to be independent of temperature over a large range. The universality is not destroyed by quantum fluctuations, which is demonstrated by quantum Monte Carlo simulations of the two-dimensional Heisenberg antiferromagnet. Physical predictions for finite doping are made, which are relevant for experiments probing Knight shifts and the order parameter.

The intentional doping of antiferromagnetic materials has become a useful tool in order to study the complicated physics in the context of high temperature superconductivity and quantum magnetism [1–6]. Large alternating magnetic moments around static nonmagnetic impurities are observed in Knight shift experiments when a uniform field is applied [3–6]. Theoretical studies have shown that vacancies in low-dimensional antiferromagnetic backgrounds give rise to locally enhanced antiferromagnetic correlations [7–14], which strongly depend on the microscopic model and temperature in the low-dimensional models.

In this work, we show that in generic *ordered* antiferromagnets the alternating local moments in the vicinity of vacancies can be quantitatively described by a universal expression which only depends on the field B, but is surprisingly independent of temperature, quantum fluctuations, and microscopic details. The mechanism which gives rise to the alternating moments is a local tilting of the order parameter due to the broken sublattice symmetry by impurities. In contrast to the pure sample, where the order parameter is always confined in the plane normal to the field, a large alternating order *parallel* to the field is induced as schematically depicted in Fig. 1. The calculations agree remarkably well with quantum Monte Carlo (QMC) simulations without any adjustable parameters even in two dimensions $D = 2$, where quantum fluctuations are strongest.

The typical antiferromagnetic Hamiltonian

$$H = J\sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j - \sum_j B S_j^z \qquad (1)$$

describes the magnetic behavior realistically even for rather complex materials despite its simplicity. We consider systems with bipartite lattices of dimension $D \geq 2$, where the sum in Eq. (1) runs over nearest neighbor sites. Generically, the dominant interaction $J > 0$ comes from the Coulomb forces via the exchange mechanism and is therefore isotropic. The rotational symmetry is broken by applied and crystal fields B in units of $g\mu_B$, which are typically small compared to the interaction $B < J$. The direction of the field defines the z axis of our coordinate system, which does not need to coincide with any of the lattice directions. For bipartite lattices of dimension $D \geq 2$ the model system (1) is known to have finite Néel order at sufficiently low temperatures for both quantum and classical spins $\mathbf{S}_i$ of any size s [15]. The ordered state remains stable over a large range of perturbations by impurities and frustrating interactions.

In order to obtain an intuitive picture of the physical behavior, let us first consider a highly simplified model of the Hamiltonian (1). The long-range order spreads over the entire sample, so it might be justified to describe all ordered spins on one sublattice A by a common direction $\hat{n}_A = (\sin\theta_A \sin\phi_A, \sin\theta_A \cos\phi_A, \cos\theta_A)$ and analogously for sublattice B. In this case, the interaction energy is always minimized by a relative azimuthal angle $\phi_A - \phi_B = \pi$, so that the effective energy is given just in terms of the polar angles
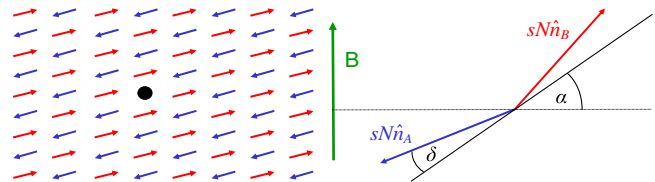

FIG. 1 (color online). Schematic illustration of the effect of a vacancy in an ordered antiferromagnet. The spins "cant" with an angle δ towards the field corresponding to a small *uniform* magnetization. Because of the broken sublattice symmetry the order may be "tilted" by an angle α relative to the plane normal to the field corresponding to an induced *alternating* magnetization around the impurity.

$$
\begin{aligned}
E_{\text{eff}} &= JzNs^2 \hat{n}_A \cdot \hat{n}_B - sNB(n_A^z + n_B^z) \\
&= JzNs^2 \cos(\theta_A + \theta_B) - sNB(\cos\theta_A + \cos\theta_B) \\
&= JzNs^2 (2\sin^2\delta - 1) - 2sNB\cos\alpha \sin\delta,
\end{aligned}
\tag{2}
$$

where z is the number of nearest neighbors and N is the total number of sublattice sites in the sample. The angle $\delta = (\pi - \theta_A - \theta_B)/2$ corresponds to a uniform "canting" of all spins on both sublattices towards an applied magnetic field as shown in Fig. 1. The angle $\alpha = (\theta_B - \theta_A)/2$ measures the "tilt" of the antiferromagnetic order relative to the plane that is normal to the field.

Below saturation $B < 2szJ$ the energy is minimized by setting $\sin\delta = \frac{B}{2szJ}\cos\alpha$ which gives an effective low energy description for α

$$
E_{\text{eff}}(\alpha) = \frac{NB^2}{2zJ}\sin^2\alpha + E_0,
\tag{3}
$$

where $E_0 = -JNs^2 z - NB^2/2zJ$. The physical interpretation of this simple model is textbook knowledge [17]: all spins align slightly towards the magnetic field $m = s\langle\sin\delta\rangle = B\chi_\perp \cos\alpha$ with a susceptibility $\chi_\perp \sim 1/2zJ$ that is largest when the magnetic field is perpendicular to the order and therefore there is a small energy gain for the Néel order to be in the plane normal to B (i.e., $\alpha = 0$). Since the energy gain is small, the order may point in another direction in realistic materials where the sublattice symmetry is broken. A common source of sublattice symmetry breaking is disorder and impurities which is the topic of this Letter.

Let us first consider a single vacancy in the framework of the simple model above by reducing the size of the corresponding sublattice vector by one spin $\mathbf{N}_A = s(N-1)\hat{n}_A$. Starting from Eq. (2) the susceptibility for δ remains the same for large N. However, the size of the two sublattice spins is not equally large and therefore a net coupling to the field remains in the effective energy as a function of the z component of the alternating order $n_z = \sin\alpha$

$$
E_{\text{eff}}(n_z) = NB^2\chi_\perp n_z^2 - sBn_z + E_0,
\tag{4}
$$

where we have also used that the dependence on δ is small and irrelevant in the direct coupling term. Even though N is large, the second term will ensure that the expectation value of the impurity induced alternating order along the field n_z is always nonzero

$$
\begin{aligned}
\langle n_z \rangle &= \frac{1}{Z}\int_{-1}^{1} dn_z e^{-\beta E_{\text{eff}}(n_z)} n_z \\
&= \frac{s}{BN\chi_\perp}\left(\frac{1}{2} - \frac{e^{-B^2 N\chi_\perp \beta}}{\int_{-1}^{1} e^{-x^2 B^2 N\chi_\perp \beta}dx}\right),
\end{aligned}
\tag{5}
$$

where we have assumed the thermodynamic limit $N \gg \beta J$ ($\beta = 1/k_B T$). In the limit of small and large fields, respectively, we find

$$
\langle n_z \rangle = \begin{cases} sB/3T & \text{for } N\chi_\perp B^2 \ll T \\ s/2NB\chi_\perp & \text{for } N\chi_\perp B^2 \gg T \end{cases}.
\tag{6}
$$

In the first case of very small fields, the alternating response to a uniform field is described by a classical susceptibility, which also directly follows from Eq. (4) if only the second term is kept (i.e., $\chi_\perp B \to 0$). Therefore, a tilting of the order parameter of order $\alpha \sim B/T$ is expected which is larger than the uniform canting $\delta \sim B/J$ in the ordered phase $T \ll J$. By QMC simulations it was shown that a corresponding alternating order is induced throughout the lattice by a single vacancy in the limit of linear response [11], which is consistent with the assumption that α describes the tilting of all spins. The corresponding impurity susceptibility is given by a classical Curie behavior $s^2/3T$, as first predicted in Ref. [2] and confirmed by QMC simulations in Ref. [18] in the limit of linear response $N\chi_\perp B^2 \ll T$. This limit is only relevant in the case where the domain size N is restricted by disorder or boundaries.

However, if N is macroscopic, the limit $N\chi_\perp B^2 \gg T$ is already reached for any naturally occurring background field, so that the second case in Eq. (6) is the more interesting limit for the description of realistic impurity effects. The induced alternating magnetization decreases with increasing field and the behavior is independent of temperature since corrections from the second term in Eq. (5) are exponentially small in the macroscopically large scaling variable $N\chi_\perp B^2\beta$. This remarkable behavior will survive even in more refined models and give rise to a universal temperature independent description as we will see.

In order to make the model more realistic, the angle α can be interpreted as a *local* tilting that is dependent on position in order to reflect the fact that the first term in Eq. (4) is an effective potential that acts on all spins in the lattice, while the second term arises from the vacancy locally at the origin. There is an energy cost to change the direction of the order parameter from one lattice site to a neighboring lattice site corresponding to the so-called spin stiffness ρ_s, so that Eq. (4) has to be generalized to an energy functional for n_z

$$
E[n_z(\mathbf{r})] = \int d^D\mathbf{r}\left\{\frac{\rho_s}{2}[\nabla n_z(\mathbf{r})]^2 + \frac{\chi_\perp}{2}B^2 n_z^2(\mathbf{r})\right\} - sBn_z(0),
\tag{7}
$$

where we have replaced the sum over both sublattices by an integral for convenience. The energy density in the first term is reminiscent of the nonlinear sigma model [19,20], but only for one component and without the imaginary time direction describing the quantum fluctuations.

In order to calculate the expectation value of $\langle n_z(\mathbf{r_0})\rangle$ at any position $\mathbf{r_0}$, it is useful to define a generating partition function

$$
Z_\gamma = \int \mathcal{D}[n_z(\mathbf{r})]\exp\{-\beta E[n_z(\mathbf{r})] + \gamma n_z(\mathbf{r_0})\}.
\tag{8}
$$

The expectation value is then given by the logarithmic derivative

$$
\langle n_z(\mathbf{r_0})\rangle = \partial_\gamma \ln Z_\gamma|_{\gamma=0}.
\tag{9}
$$

In momentum space the partition function becomes

$$Z_\gamma = \int \mathcal{D}[n_z(\mathbf{q})]e^{\int d^D\mathbf{q}[-\beta E_\mathbf{q}|n_z(\mathbf{q})|^2 + I_\mathbf{q}(\gamma)n_z(\mathbf{q})]}, \quad (10)$$

where

$$E_\mathbf{q} = (\rho_s q^2 + \chi_\perp B^2)/2 \quad (11)$$

$$I_\mathbf{q}(\gamma) = (\beta Bs + \gamma \cos\mathbf{q} \cdot \mathbf{r_0})/(2\pi)^{D/2}. \quad (12)$$

The expectation value is therefore

$$\langle n_z(\mathbf{r_0})\rangle = \partial_\gamma \ln Z_\gamma|_{\gamma=0}$$

$$= \partial_\gamma \int d^D\mathbf{q} \ln \int dn_z e^{-\beta E_\mathbf{q} n_z^2 + I_\mathbf{q}(\gamma)n_z}|_{\gamma=0}$$

$$= \partial_\gamma \int \frac{d^D\mathbf{q}}{(2\pi)^{D/2}} \frac{I_\mathbf{q}^2(\gamma)}{2\beta(\rho_s q^2 + \chi_\perp B^2)}\bigg|_{\gamma=0}$$

$$= \int \frac{d^D\mathbf{q}}{(2\pi)^D} \frac{Bs\cos(\mathbf{q}\cdot\mathbf{r_0})}{(\rho_s q^2 + \chi_\perp B^2)} \quad (13)$$

$$= \begin{cases} \frac{sB}{2\pi\rho_s}K_0(\frac{B}{c}r) & D = 2 \\ \frac{sB}{4\pi\rho_s r}e^{-Br/c} & D = 3 \end{cases} \quad (14)$$

where $c = \sqrt{\rho_s/\chi_\perp}$ is known as the spin-wave velocity and K_0 is the modified Bessel function. This result is remarkable in two ways: first of all, it turns out to be completely independent of temperature and secondly, it is independent of the underlying detailed microscopic model. Therefore, the formula in Eq. (13) can be taken as a universal description for all antiferromagnets in the ordered phase. Variations in the lattice structure, frustration, quantum effects, and the detailed microscopic parameters only renormalize the spin stiffness ρ_s and the uniform susceptibility $\chi_\perp$, but not the functional behavior in Eq. (14). For spins close to the vacancy $r \gtrsim 1$ the tilting $n_z \sim sB/4\pi\rho_s$ remains typically less than saturation, but larger than the uniform canting $\alpha > \delta$, so that spins on the same sublattice as the vacancy tend to align *against* the field.

It can be checked that the functions in Eq. (14) are solutions of the diffusion equation

$$B^2 \chi_\perp n_z = \rho_s \nabla^2 n_z \quad (15)$$

that also follows from minimizing the energy functional Eq. (7). In lattices where the spin stiffness is not isotropic, the result can be generalized by taking ρ_s as an anisotropic diffusion coefficient.

As a concrete example, we will now consider the spin-1/2 Heisenberg model on a 2D square lattice, which is possibly the most studied antiferromagnetic model, since it has received much attention in connection with high temperature superconductivity, but also because it is an interesting case where quantum fluctuations strongly compete with Néel order.

In Monte Carlo simulations we have used the stochastic series expansion with directed loop updates [21] in order to extract the magnetic moments $\langle m_z \rangle$ around a single va-

cancy in small magnetic fields in the ordered phase $\xi(T) \gg L$ (here $L = 128$) as shown in the inner inset of Fig. 2. In the plane perpendicular to the field the order is fluctuating, so that $\langle m_x \rangle = \langle m_y \rangle = 0$. For the moments parallel to the field, we expect to find a large staggered magnetization according to Eq. (14)

$$\langle m_z(\mathbf{r})\rangle = (-1)^r m_{\max} \frac{sB}{2\pi\rho_s} K_0\left(\frac{B}{c}r\right) \quad (16)$$

in addition to the less interesting small uniform canting δ. Here $m_{\max} \approx 0.308$ is the maximum order in the 2D Heisenberg model which is reduced from $s = 1/2$ due to quantum fluctuations. In fact, there are no adjustable parameters in Eq. (16) since all relevant parameters have long been established to high precision by independent methods [16].

$$m_{\max} \approx 0.308, \qquad \rho_s \approx 0.18J,$$
$$\chi_\perp \approx 0.065/J, \qquad c = \sqrt{\rho_s/\chi_\perp} \approx 1.67J. \quad (17)$$

By extrapolating the numerical data for $m_z(\mathbf{r})$ on the even and the odd sublattice separately and taking half the difference, we extracted the staggered magnetization $m_{\text{alt}}(\mathbf{r})$ around a vacancy. The resulting alternating amplitude m_{alt} is completely isotropic and can be plotted as a function of the geometrical distance $r = |\mathbf{r}|$ only as shown in Fig. 2 for different fields and temperatures. While the size of m_{alt} is proportional to the field, the dropoff is shortened for higher fields so that the integrated amplitude
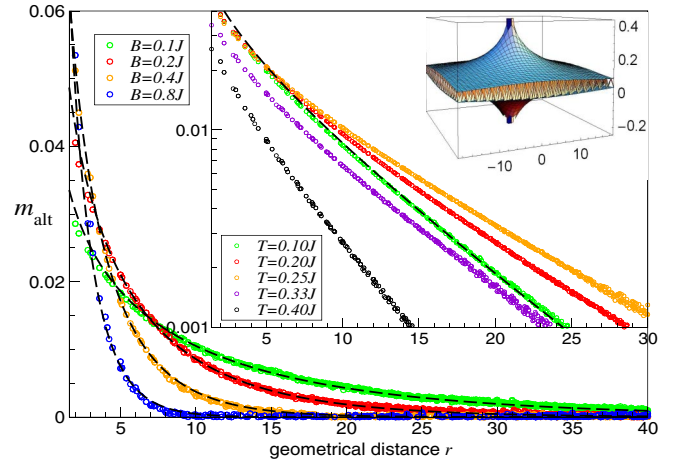

FIG. 2 (color online).   The alternating response m_{alt} as a function of geometrical distance r from a vacancy in a 2DHAF at different fields and $T = 0.025$ J from QMC simulations compared to the universal theoretical prediction $m_{\max}n_z$ in Eq. (16) *without any adjustable parameters* (dashed black lines). Inset: m_{alt} at $B = 0.2$ J. Even at higher $T = 0.1$ J no deviations from Eq. (16) can be seen on a logarithmic scale. At still higher T the induced order first *increases* for 0.3 J $\gtrsim T \gtrsim 0.2$ J and then decreases again for $T \gtrsim 0.3$ J. Inner inset: magnetic moments at $B = 0.1$ J and $T = 0.1$ J alternating between the extrapolated amplitudes on even and odd sublattices.

decreases with increasing field as also reflected in the simple model of Eq. (5). The agreement with Eq. (16) is remarkably good even on a logarithmic scale and for widely different fields and temperatures, which we take as confirmation for the general validity of the result in Eq. (13). Since there were no adjustable parameters, we conclude that the quantitative predictive power for static expectation values of the hydrodynamic model in Eq. (7) is not changed by quantum fluctuations. From a theoretical point of view this means that the renormalized classical model [19,20] can be taken for quantitative calculations anywhere in the antiferromagnetic phase, while microscopic details only affect the values of the constants in Eq. (17). In particular, close to a critical point ρ_s and m_{max} become vanishingly small, but the model remains valid.

A breakdown of the universal formula in Eq. (13) must occur at the transition temperature to the disordered phase. In the 2D simulations we find indeed that any temperature dependence is exponentially suppressed until the Kosterlitz-Thouless temperature is approached [22] $T_{KT} \sim 0.2$ J. However, the induced alternating magnetization is surprisingly *enhanced* by increasing temperature near T_{KT} as shown in the inset of Fig. 2. Only at still higher temperatures $T \gtrsim 0.3$ J the induced order is finally reduced as expected, leading to a nonmonotonic behavior with temperature. While we have no explanation of this exotic effect in terms of our model, we hope that future works on this topic may uncover this mystery.

Finally, we would like to generalize our results to finite impurity concentrations ρ. For higher fields/small concentrations $\rho < (B/c)^D$ the impurities are sufficiently far apart to be treated independently (dilute limit). In this case, the above conclusions are unchanged and the magnetic order is tilted locally in the vicinity of each vacancy. At smaller fields (larger concentrations) $\rho > (B/c)^D$ the impurities become correlated and enhance (annihilate) the tilting effect depending if they are on the same (opposite) sublattices [11]. In this disorder limit all impurities become correlated and the tilting is again nearly uniform throughout the lattice, with an effective total impurity strength that is given by the difference of the vacant sites on each sublattice $|N_A - N_B| \sim \sqrt{\rho N}$ in a domain of size N. In this case the simple model in Eq. (4) remains valid with the effective size of the spin s in the last term replaced by $s\sqrt{\rho N}$. The average universal tilt in Eq. (6) throughout the domain is then given by $m_{max}s\sqrt{\rho}/2\sqrt{N}B\chi_\perp$.

In conclusion, we have analyzed the induced alternating magnetization around vacancies in ordered antiferromagnets in quantitative detail. Large alternating moments are induced parallel to the field, which corresponds to a tilting of the order parameter. The induced order decays with distance at a rate that is independent of temperature and inversely proportional to the field c/B. From a theoretical point of view we have demonstrated that the renormalized classical description gives an intuitive insight into the mechanism on how the alternating magnetization arises.

At the same time, the theory gives good quantitative agreement with QMC simulations even in 2D where quantum fluctuations are large. At large impurity densities $\rho > (B/c)^D$ the impurities become correlated and give rise to a tilt of the order throughout the sample towards the field direction. The predicted effects can be observed by Knight shift experiments like NMR and μSR, or by investigating the order directly via magnetic neutron scattering. Numerically we find an *enhancement* of the induced order near the Kosterlitz-Thouless transition $T_{KT} \gtrsim 0.2$ J which is counterintuitive and calls for further investigation.

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