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"Non-magnetic impurities in two- and three-dimensional Heisenberg antiferromagnets"

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Non-magnetic impurities in two- and three-dimensional Heisenberg antiferromagnets

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Abstract

In this letter we study in a large-S expansion effects of substituting spins by non-magnetic impurities in two- and three- dimensional Heisenberg antiferromagnets in a weak magnetic field. In particular, we demonstrate a novel mechanism where magnetic moments are induced around non-magnetic impurities when magnetic field is present. As a result, Curie-type behaviour in magnetic susceptibility can be observed well below the Neel temperature, in agreement with what is being observed in $La_2Cu_{1-x}Zn_xO_4$ compounds.

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Recently, there has been increasing interests in the effects of substituting Cu by non-magnetic ions (Zn) in high-T_c cuprates, where interesting effects were discovered for compounds in the underdoped regime [1]. In particular, magnetic susceptibility measurement [2] and other experiments [3] seem to indicate that local magnetic moments are generated as Cu ions were replaced by Zn, both in the underdoped High-T_c Yttrium and Bismuth compounds [2,3], and for *undoped* Lanthanium compound [4]. Based on RVB theories of the $t-J$ model, Nagaosa and Ng [5] have constructed explanations on how local magnetic moments can be generated in these compounds in the disordered phase of two-dimensional antiferromagnets. Their theory is believed to be applicable to the underdoped regime of High-T_c compounds. However the theory is not applicable to the La compound where Curie-type behaviour is observed in magnetic susceptibility at temperatures well below the Neel temperature where long-ranged antiferromagnetic order exists.

In the presence of long-ranged antiferromagnetic order, the low temperature properties of clean antiferromagnets can be understood at least qualitatively using a semi-classical theory ($1/S$ expansion). In this letter we shall generalize this approach to include the effects of non-magnetic impurities in the presence of a uniform magnetic field. Notice that in the absence of magnetic field, a similar study has been performed by Bulut *et. al.* [6] where no 'free' magnetic moments were found to be induced around non-magnetic impurities. We shall show that local magnetic moments can be induced by non-magnetic impurities once magnetic field is applied on the system through a novel mechanism. Notice that straightly speaking, thermal fluctuations destroy long-ranged magnetic order at two dimension at any finite temperature. Thus our analysis at two dimension can only be applied to layered systems like Lanthanium cuprates where the magnetic coupling between different layers is much weaker than intra-layer coupling.

To begin with, we first consider the zero temperature problem of classical spins interacting antiferromagnetically under a uniform magnetic field B, with a single non-magnetic impurity replacing spin at site i_0. We shall consider the Neel state in the absence of magnetic field to be ordered in the z-direction, with the uniform magnetic field B applied on the $+x$-direction. The classical ground state in the absence of magnetic field has spins all pointing in $+z$ direction for spins in A-sublattice and spins all pointing in $-z$ direction for spins in B-sublattice. In the presence of magnetic field, the spins tilt to the $+x$-direction to minimize the magnetic energy. Let θ_i^A be the tilted angle away from the z-axis for spin on site i on A-sublattice and θ_j^B be the corresponding angle for spin on site j on B-sublattice. It is easy to show that the classical energy E_{cl} is given in the limit when $\theta^{A(B)}$ are small (weak B-field limit) by

$$E_{cl}/S^2 = -\sum_{<i\neq i_0,j>}(1-\frac{(\theta_i^A+\theta_j^B)^2}{2}) - B'\sum_{i\neq i_0}\theta_i^A - B'\sum_j \theta_j^B, \tag{1}$$

where $< i \neq i_0, j >$ are nearest neighbor sites in the square (cubic) lattice excluding contributions from the non-magnetic impurity at site i_0 and $B' = g\mu_B B/S$. Notice that the only effect of non-magnetic impurity is to remove the spin at site i_0 in our theory. We have also set the spin-coupling $J = 1$ in Eq. (1). Notice that the energy expression for E_{cl} is valid only to order $O(B^2)$.

Next we consider the continuum limit of the energy expression (1). Introducing symmetric and antisymmetric angle variables

$$\begin{aligned}\theta_s(\vec{x}) &= S(\theta^A(\vec{x})+\theta^B(\vec{x})),\\ \theta_a(\vec{x}) &= \frac{S}{2}(\theta^A(\vec{x})-\theta^B(\vec{x})),\end{aligned} \tag{2}$$

we obtain after some straightforward algebra

$$E_{cl} = \int\frac{d^dx}{a_o^d}\left[(d)\theta_s(\vec{x})^2 - B'S\theta_s(\vec{x}) + (\nabla\theta_a(\vec{x}))^2 + \frac{\theta(x_<)}{v_a}(\theta_s(\vec{x})\hat{a}_o.\nabla\theta_a(\vec{x}) - \theta_a(\vec{x})\hat{a}_o.\nabla\theta_s(\vec{x}))\right] + E_0 \tag{3}$$

where $E_0 = -NS^2d/2$, N = number of sites in the system, a_o is lattice spacing and $\hat{a}_o$ is a radial vector pointing away from i_0. v_a is the volume enclosed by the sphere (circle in 2D) with radius a_o. $x_< = a_o - |\vec{x}-\vec{x}_0|$. For an impurity located at $B-$ sublattice, $\hat{a}_o \to -\hat{a}_o$.

The various terms appearing in Eq. (2) can be understood rather easily. First of all, in the limit $a_o \to 0$, $x_i \to x_j$ in Eq. (1) and the only contribution to E_{cl} would be the first two θ_s terms in Eq. (2). It is also clear that in the limit $a_o \to 0$, terms proportional to θ_a or θ_a^2 do not appear in E_{cl}, and the only contribution from θ_a can only appears as $(\nabla\theta_a)^2$. Appearance of non-magnetic impurity at site i_0 breaks the symmetry between $A-$ and $B-$ sublattices and introduces local coupling between the θ_s and θ_a fields. Notice that additional coupling between θ_a and θ_s fields will appear if we take into account higher order terms in B in our energy expression E_{cl}. However, in the absence of impurities, these terms do not break the symmetry between $A-$ and $B-$ sublattices or the symmetry of interchanging θ^A and θ^B fields.

Minimizing E_{cl} with respect to the θ_s field we obtain

$$\theta_s(\vec{x}) = \frac{1}{2d}\left(B'S - \frac{2\theta(x_<)}{v_a}\hat{a_o}.\nabla\theta_a(\vec{x})\right). \tag{4a}$$

Putting θ_s back into E_{cl} and minimizing with respect to θ_a field, we obtain for $|\vec{x}-\vec{x_0}| \geq a_o$.

$$\nabla^2\theta_a(\vec{x}) = \frac{2B'Sa_o}{dv_a}\delta^d(|\vec{x}-\vec{x_0}| - a_o). \tag{4b}$$

Notice that in the presence of magnetic field, the non-magnetic impurity acts as a source term for the θ_a field of strength $2B'S$ in the limit $a_o \to 0$. As a result, $\theta_a(\vec{x}) \sim (2B'S)ln(|\vec{x}-\vec{x_0}|)$ in two dimension, and the corresponding 'electric field' energy cost $\sim \int d^2x(\nabla\theta_a)^2$ is diverging logarithmically as size of system goes to infinity.

The divergence of magnetic energy to order $O(B^2)$ indicates that the (classical) response of a non-magnetic impurity to external magnetic field is intrinsically *non-linear* [7] and suggests that quantum effect may play an important role in determining the correct *linear* response of non-magnetic impurities to external magnetic field. In the following we shall derive in the continuum limit the $1/S$ (spinwave) Lagrangian in the presence of background θ_s and θ_a fields, and shall show that the infra-red divergence in classical energy can be cured by formation of local magnetic moment around non-magnetic impurity once quantum effects are considered.

To derive the spinwave Hamiltonian in the presence of magnetic field, we rotate our co-ordinate system locally on each site such that the local $z-$axis is always along the 'classical' spin direction. In this co-ordinate system, the Hamiltonian becomes

$$H = \sum_{<i\neq i_0,j>} \left[cos(\theta_i+\theta_j)\left(S_i^{(z)}S_j^{(z)} + S_i^{(x)}S_j^{(x)}\right) + S_i^{(y)}S_j^{(y)} + sin(\theta_i+\theta_j)\left(S_i^{(z)}S_j^{(x)} - S_i^{(x)}S_j^{(z)}\right)\right]$$
$$-g\mu_B B\sum_{i\neq i_0}(S_i^{(z)}sin\theta_i + S_i^{(x)}cos\theta_i) - g\mu_B B\sum_j(cos\theta_j S_j^{(x)} - sin\theta_j S_j^{(z)}). \quad (5)$$

The Hamiltonian can be rewritten in Schwinger-boson representation of spins in the usual way. In the large-S limit, we may write

$$Z_{i\uparrow}^A = \bar{Z}_{i\uparrow}^A = Z_{j\downarrow}^B = \bar{Z}_{j\downarrow}^B = \sqrt{(2S)},$$

where $\bar{Z}(Z)_{i\sigma}^{\alpha}$'s are spin σ Schwinger-boson creation (annihilation) operators for site i on sublattice α. Expanding the Hamiltonian to order $O(S)$, we obtain to order $O(B^2)$,

$$H = E_{cl} + H_{1/S},$$

where

$$H_{1/S} = S\sum_{<i\neq i_0,j>}\left[(1-\frac{(\theta_i+\theta_j)^2}{2})\left(\bar{Z}_{i\downarrow}^A Z_{i\downarrow}^A + \bar{Z}_{j\uparrow}^B Z_{j\uparrow}^B\right) + (1-\frac{(\theta_i+\theta_j)^2}{4})\left(Z_{i\downarrow}^A Z_{j\uparrow}^B + \bar{Z}_{i\downarrow}^A \bar{Z}_{j\uparrow}^B\right)\right]$$
$$+g\mu_B B\sum_{i\neq i_0}(\theta_i)\bar{Z}_{i\downarrow}^A Z_{i\downarrow}^A + g\mu_B B\sum_j(\theta_j)\bar{Z}_{j\uparrow}^B Z_{j\uparrow}^B. \quad (6)$$

To further analyse our system we again go to the continuum limit. Introducing symmetric and anti-symmetric boson fields

$$\phi(\vec{x}) = \frac{1}{\sqrt{2}}(Z_\downarrow^A(\vec{x}) - \bar{Z}_\uparrow^B(\vec{x})),$$
$$\pi(\vec{x}) = \frac{1}{\sqrt{2}}(Z_\downarrow^A(\vec{x}) + \bar{Z}_\uparrow^B(\vec{x})), \quad (7)$$

and integrating out the $\pi(\vec{x})$ field, we obtain in the continuum limit an effective Lagrangain for $\phi(\vec{x})$ field [8,9]. Details of the technique can be found in ref. [8] and we shall not repeat them here. For $\vec{x}$ away from the impurity site ($a_o < |\vec{x}-\vec{x}_0|$), we obtain in imaginary time

$$L_{1/S} \sim \int d\tau\int d^dx\frac{1}{4dS}\left[(\frac{\partial}{\partial\tau}-e\theta_a)\phi^+(\frac{\partial}{\partial\tau}+e\theta_a)\phi + m^2\phi^+\phi + c^2|\nabla\phi|^2\right], \quad (8)$$

where $e = B'$ and $m^2 = S^2B'^2/2$. $c^2 \sim 4d(JS)^2$ is the spinwave velocity. We have set $\theta_s(\vec{x}) = B'S/2d$ and have neglected $(\nabla\theta_a)^2$ terms in deriving $L_{1/S}$. The later is of higher order ($O(1/S)$) compared with the corresponding term in E_{cl}. The most striking feature of the effective lagrangian is that the θ_a field now appears as the τ-component of a U(1) gauge field coupling to a charge boson field ϕ, with the dynamics of the θ_a field governed by E_{cl}. In particular, in the presence of B field, the non-magnetic impurity appears as electric charge generating static electric field coupling to the charge bosons. Notice that the effective charge of the non-magnetic impurity changes sign when it is moved from one sublattice to another, indicating that the 'sign' of the charge is in fact a sublattice index [8,9].

The logarithmic divergence in 'electric field' energy in presence of non-magnetic impurity in E_{cl} in two dimension will be removed if bosons of opposite 'electric' charge are nucleated from vacuum to screen the effective electric field, forming effective local magnetic moments around the impurity. The number of bosons nucleated from vacuum is $\sim 2S$, as can be seen easily by counting the number of 'charges' carried by the non-magnetic impurity. The magnitude of magnetic moment formed around the impurity is thus $\sim S$. The energy 'cost' for nucleating the bosons can be computed by solving the corresponding Schrödinger equation for charge bosons moving in scalar potential of external charge of magnitude $2Se$ [10]. To capture qualitatively the physics, we estimate the energy 'cost' by using a variational wavefunction $\psi(\vec{r}) \sim Ae^{-|\vec{r}|/\xi_1}$, where ξ_1 is determined variationally. Minimizing the energy, we find that ξ_1 is of order $\xi_1 \sim (JSc^2/(g\mu_B B)^3)^{1/2})$. Notice that $\xi_1 \to \infty$ as $B \to 0$ or $S \to \infty$, indicating that the formation of magnetic moment around non-magnetic impurity is a non-perturbative quantum effect which cannot be captured by usual spinwave theory. The energy 'cost' for nucleating the bosons is of order $E_1 \sim (2S)(m + 2SB'^2 ln(\xi_1/a_o))$.

Next we discuss the situation when there is finite concentration of impurities n randomly distributed in the system, which is the case of experimental interests. In our continuum theory, the presence of finite concentration of random impurities is equivalent to putting finite concentration of 'charges' with magnitude $2SB'$ and random sign in our effective charge boson system. The behaviour of the system is very different from the one impurity case, since now electric fields originating from opposite charges will cancel, with remaining 'electric field' energy of order $\sim nE_e$, where $E_e \sim (2SB')^2 ln(l/a_o)$ on average, $l \sim n^{-1/d}$ is the average distance between non-magnetic impurities. For $E_e > E_1 \sim 2Sm$ or equivalently $l >> \xi_o \sim a_o e^{JS/(2g\mu_B B)}$, it is energetically favourable to nucleate charge bosons from vacuum to screen the electric field, and magnetic moments will be formed around non-magnetic impurities. The magnetic moments couple to each other weakly with $J_{eff} \sim Je^{-l/\xi_1}$ implying that Curie behaviour will be present in magnetic susceptibility down to very low temperature $\sim J_{eff}$. As concentration of non-magnetic impurities increases, l decreases until $l \sim \xi_o$. At this point a transition occurs where it becomes energetically unfavourable to nucleate bosons from vacuum to screen the electric field, i.e. there will be no magnetic moments forming around non-magnetic impurities at zero temperature when $l \leq \xi_o$. The boson-non-magnetic impurity bound states become excited states of the spin system!

The boson-non-magnetic impurity bound states can still be observed as effective local magnetic moments at finite temperature $T \geq m \sim g\mu_B B$ if $E_e + 2Sm > E_1$ (corresponding roughly to $\xi_1 < l$). At this energy range, the boson-impurity bound state appears as stable excited state of the spin system with excitation energy $\sim (E_1 - E_e)/2S < m$. At finite temperature $T \geq m$, these states will be occupied by thermally excited bosons, forming effective free magnetic moments around non-magnetic impurities. It is important to emphasize that the formation of excited boson bound state around non-magnetic impurity is only possible when there is a gap m in the boson excitation spectrum. In the absence of non-magnetic impurity, a direct computation of the spinwave spectrum indicates that the spinwave spectrum splits into two branches at small momentum q, with one branch carrying a gap larger than m in our effective Lagrangian, and the other branch gapless as $q \to 0$. The contribution from the splitting of the two branches to $H_{1/S}$ cancels to order $O(B^2)$, leading to our effective Lagrangian with only massive bosons. The splitting of the boson spectrum reappears if we include terms to order $O(B^4)$ in $H_{1/S}$, and the boson bound state around non-magnetic impurity will ceased to be good eigenstate of the effective Lagrangian (the bound state turn into resonant state). However, the decay rate of the resonant state $\sim B^4$ is small compared with the energy of the resonant state $\sim B^2$ in the limit of weak magnetic field, indicating that the physical picture of boson bound state around non-magnetic impurity is still a good approximation to the system in weak field limit.

As concentration of impurities increases and distances between impurities decreases further the overlap between boson bound state wavefunctions at different impurity sites increases and a boson impurity band is formed (recall that the size of the boson bound state wavefunction is of order ξ_1). For $l \leq \xi_1$, the bandwidth W of this impurity band is of order $\sim (\sqrt{m^2+(c/l)^2}-m)$ and the effective impurity potential strength is of order $v \sim 2SB'^2/(1/l)^2$. We find that the screening of effective impurity potential by charged bosons is weak and can be neglected. Estimating the scattering life time τ we find that bosons with energy $E \leq E_l \sim cl/\xi_1^2$ are strongly localized in this impurity band using the criteria $E\tau \leq 1$. Notice that the number of localized boson states $\sim n^{-1}$ and decreases as n increases in this regime. The magnetic susceptibility will show Curie behaviour at temperature above $T_l \sim E_l/k_B$. For $T \leq T_l$, the number of effective magnetic moments contributing to magnetic susceptibility decreases as temperature lowers. Roughly speaking, the contribution from non-magnetic impurities to the magnetic susceptibility can be described by a magnetic field and temperature dependent density of local moments $n(B,T)$ which has the following properties: (i) $n(B,T) \sim 0$ at the region $g\mu_B B >> k_B T$, (ii) $n(B,T) \sim n$ for $l \geq \xi_1$, and $\sim (n\xi_1^4)^{-1}$ for $l \leq \xi_1$ at the region $k_B T >> g\mu_B B + E_l$, and is smoothly interpolating between the two regions. As temperature $T \rightarrow 0, n(B,T) \rightarrow 0$ and the contribution to magnetic susceptibility from impurities is of order $\delta\chi \sim 4S^2 n ln(l/a_o)$.

Our theory can be extended to three dimension in a straightforward way. We find that in the weak-field limit, no magnetic moment is formed even in the limit of one single impurity at zero temperature because of absence of infra-red divergence associated with Coulomb potential in $3D$. However, bound states of bosons can be formed at $T \geq g\mu_B B$ as in two dimensional case, and can still result in Curie-type behaviour in magnetic susceptibility for $l >> \xi_1 \sim (c/g\mu_B B)^3$. As concentration of impurities increases further, the boson bound states turn into impurity band and the effective number of 'free' magnetic moments $n(B,T)$ decreases when temperature is lower than the boson impurity band bandwidth as in two dimension.

Summarizing, in this paper we have carried out an analysis of the effects of replacing spins by non-magnetic impurities in Heisenberg antiferromagnets in magnetic field in two- and three- dimensions in a semi-classical $1/S$ expansion. We find within a continuum approximation that magnetic moments will form around non-magnetic impurities once magnetic field is put on the system, resulting in Curie-type behaviour in magnetic susceptibility when concentration of impurities is not too high, and temperature is not too low. Notice that similar continuum theories have been sucessfully applied to explain behaviours of clean two-dimensional quantum Heisenberg antiferromagnets at low temperature [11], and to the effect of non-magnetic impurities in one dimensional spin chains (edge states) [12,13] Our theory is able to explain observation of Curie-type behaviour in magnetic susceptibility in $La_2Cu_{1-x}Zn_xO_4$ compounds at temperature well below Neel temperature. In particular, the vanishing of local moments at $T \leq g\mu_B B$ is a theoretical prediction which can be tested experimentally.

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