Non-magnetic Impurities in Spin Gap Systems

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The effect of non-magnetic impurities discussed for both the two-leg Heisenberg ladder system and other spin liquids with excitation gap. It is shown that the random depletion of spins introduces a random Berry phase term to the non-linear $\sigma$ model. The classical nature of the antiferromagnetic correlation is enhanced by the topological decoherence, and the staggered susceptibility shows more singular behavior at low temperatures than the uniform antiferromagnetic Heisenberg chain.

KEYWORDS: quantum spin chain, Heisenberg ladder, random spin system

Recently a class of quantum antiferromagnets which has a ground state with a gap in the spin excitation spectrum has attracted much attention. These systems are characterized either by a local singlet correlation of valence-bond-solid state as found in spin-Peierls systems and even-leg spin ladders or by resonating-valence-bond (RVB) state as in underdoped cuprates. In such systems it was found that the presence of non-magnetic impurities replacing magnetic ions often leads to a Curie behavior of the uniform susceptibility with a Curie constant proportional to the impurity concentration. This can be understood basically from the idea that, due to the depletion of spins, some spins lose their partners to form a singlet. They appear, therefore, as nearly independent spin doublet degrees of freedom. However, the more striking experimental feature is the appearance of the antiferromagnetic long-range order (AFLRO) induced by the non-magnetic impurities. This property has been observed essentially in all the systems mentioned above, and thus seems to be a universal phenomenon in gapped quantum spin liquids. This effect has recently been analyzed in terms of the phase Hamiltonian for the spin-Peierls system by Fukuyama et al. The presence of impurities leads to a local weakening of the dimerization, which induces enhanced local antiferromagnetic correlations. The AFLRO can then emerge at low temperatures, because the local antiferromagnetic moments around the impurities correlate in phase with each other. The phase Hamiltonian approach has also been applied to the two-leg spin ladder system with a single non-magnetic impurity. In this case the phase field forms a soliton at the impurity position, which corresponds to an impurity-induced spin degree of freedom ($S = 1/2$). Local antiferromagnetic correlation appears in the vicinity of the impurity, and would turn to an in-phase (quasi) long-range ordered state in the ground state. Sigrist and Furusaki pointed out that low-energy physics of the lightly depleted spin ladder system is described by a spin-$\frac{1}{2}$ Heisenberg model with random ferromagnetic and antiferromagnetic couplings. They found that at low temperatures the uniform susceptibility follows the Curie law while the specific heat shows power-law behavior, $C \sim T^{2\alpha}$, where the exponent $\alpha$ is expected to be equal to or smaller than 0.25. On the other hand, in a recent numerical study Furukawa et al. found a significant enhancement of the staggered susceptibility $\chi(Q)$ in the presence of non-magnetic impurities. The asymptotic behavior, however, could not be obtained because the size of the system they studied is not large enough.

In this paper we study the effect of non-magnetic impurities on gapped spin liquids via a mapping to the non-linear $\sigma$ model with a random topological term. In this formulation the appearance of the AFLRO originates from the recovery of the topological Berry phase term, which suppresses quantum fluctuations such that the classical nature of the AFLRO is enhanced. We find that the generalized susceptibility $\chi(Q, T)$ diverges with a non-trivial exponent, $\chi(Q, T) \sim T^{-1-2\alpha}$, in the low-temperature limit. This divergence is much stronger than the spin-$\frac{1}{2}$ antiferromagnetic Heisenberg chain. This picture also applies to higher-dimensional systems where quantum fluctuations suppress the AFLRO and generate the spin gap phase, even though the behavior of $\chi(Q, T)$ depends on the dimensionality.

For concreteness we consider the two-leg spin-$S$ Heisenberg ladder as an example of gapped spin liquids. We use the functional integral method and write the partition function $Z$ as

$$Z = \int DS_i(\tau) e^{-A}. \tag{1}$$

The action $A$ of the system is given by

$$A = iS \sum_i \omega(\{S_i(\tau)\}) + \int d\tau \sum_{ij} JS_i(\tau) \cdot S_j(\tau). \tag{2}$$

The first term is the Berry phase contribution, where
The effective action $A_{\text{eff}}$ is given by

$$A_{\text{eff}} = -i S \sum_n (-1)^{l_n} \omega(\{I_n(\tau)\})$$

$$+ \frac{g}{2 \omega_1} \sum_{k, \omega_1} \left[ \frac{(\omega_1)}{c} \right]^2 + k^2 + \frac{1}{l_0^2} \Omega(k, \omega) \cdot \Omega(-k, -\omega)$$

$$+ \frac{g}{2 \omega_1} \sum_{\omega_n} \lambda_n(\omega) \cdot I_n(\omega) - \sum_k \frac{e^{ikx_{in}}}{\sqrt{L}} \Omega(k, \omega)$$,

where $\omega_1$ is the Matsubara frequency for bosonic fields, and $(-1)^{l_n} = +1 (-1)$ if $x_{in}$ is on the A-sublattice (B-sublattice). In eq. (9) we have introduced a mass $c/l_0$ in the second term. This corresponds to taking saddle-point approximation for a Lagrange multiplier field representing the constraint $|\Omega| = 1$. This can be justified, in the limit of small impurity concentration, by noting that $l_0$ is the finite correlation length of the pure two-leg spin ladder. We then integrate over $\Omega$ to obtain

$$A_{\text{eff}} = -i S \sum_n (-1)^{l_n} \omega(\{I_n(\tau)\})$$

$$+ \frac{g}{2 \omega_1} \sum_{k, \omega_1} \left[ \frac{(\omega_1)}{c} \right]^2 + k^2 + \frac{1}{l_0^2} \Omega(k, \omega) \cdot \Omega(-k, -\omega)$$

$$+ \frac{g}{2 \omega_1} \sum_{\omega_n} \lambda_n(\omega) \cdot I_n(\omega) - \sum_k \frac{e^{ikx_{in}}}{\sqrt{L}} \Omega(k, \omega)$$,

where we have ignored an additive constant coming from integration over $\Omega$, and

$$K_{m,n}(\omega_l) = \int_{-\infty}^{\infty} \frac{dk}{2\pi} \frac{\sin(x_{in} - x_{in})}{k^2 + k_0^2} = \frac{1}{2k_0} e^{-k_0|x_{in} - x_{in}|}$$

with $k_0 = \left( (\omega_l/c)^2 + l_0^2 \right)^{1/2}$. For the case where the concentration $\delta$ of the non-magnetic impurities is very small ($\alpha/\delta \gg l_0$), we can approximate $K^{-1}$ as

$$(K^{-1})_{m,n} \approx 2k_0 \left( \delta_{m,n} - \delta_{m,n+1} e^{-k_0|x_{in} - x_{in}|} \right).$$
After integrating over $\lambda$, we obtain, for $|\omega_n| \ll c/l_0$,

$$ A_{\text{eff}} = -iS \sum_n (-1)^{\nu_n} \omega(\{I_n(\tau)\}) $$

$$ -\int_0^\beta d\tau \sum_n \tilde{J} e^{-|x_{n-1}-x_{n+1}|/l_0} I_n(\tau) \cdot I_{n+1}(\tau), \quad (13) $$

where $\tilde{J} = c k_0/g$. Equation (13) can be identified with the action for the random-exchange Heisenberg model (REHM),

$$ H = -\sum_n J_n S_n \cdot S_{n+1}, \quad (14) $$

where $S_n = (-1)^{\nu_n+1} I_n$. The exchange interaction $J_n$ is given by $J_n = \tilde{J} (-1)^{\nu_n+1} \exp(-|x_{n-1}-x_{n+1}|/l_0)$, which is random in sign as well as in magnitude. [8] Note that the two spins $S_n$ and $S_{n+1}$ are coupled ferromagnetically (antiferromagnetically) if they are on the same (different) sublattice(s). [8] It is important to notice that the classical nature of $I$ is more enhanced in the randomly depleted Heisenberg ladder than in the pure spin-ladder, and thus there is no topological term for the staggered spin $I_n$, which is typically of the order of $Je^{-\lambda_0/\delta}$. Therefore Table I is not intended to exhaust all the universality classes of the quantum spin chains. [18] Here some remarks are in order on the random singlet phase, in which spins far apart form weakly bound singlet pairs in a random manner. [18, 20] This phase is realized in the random antiferromagnetic Heisenberg model where the exchange couplings are all antiferromagnetic but are random in their magnitude. In this case the Berry phase term is not affected by the randomness, and the singlet formation of spins plays an essential role. We did not include this phase in Table I, because the randomness is not of topological nature. Therefore Table I is not intended to exhaust all the universality classes of the quantum spin chains.

Finally, we briefly discuss the two-dimensional case for which the mapping to the non-linear $\sigma$ model can be used in exactly the same way as in the one-dimensional case. For example, in the Heisenberg antiferromagnet on the square lattice, Berry phases of each row cancel as in the spin ladder, and thus there is no topological term for the slowly varying field $\Omega$. [18] For $S = 1/2$ the dimensionless coupling constant $g \sim 1/S$ is not large enough for the quantum fluctuations to destroy the AFLRO. However, as the mobile holes are doped into the antiferromagnet (e.g., doped high-Tc cuprates), the AFLRO is rapidly suppressed and eventually destroyed. These holes are localized when Zn impurities are doped sufficiently. In this case both the Zn impurities and the localized holes contribute to the random Berry phase terms in the non-linear $\sigma$ model. Then we expect that the antiferromagnetic correlation is enhanced and the AFLRO is recovered with non-magnetic impurities, which is indeed observed in a recent experiment on the underdoped cuprates with the (pseudo) spin gap. [3] The problem of the coexistence of the mobile holes and Zn impurities is left for future study.

Before summarizing the results, we point out that a
non-linear $\sigma$ model with a random topological term has also been discussed in the context of the localization of a particle in a random magnetic field. It is an interesting future problem to explore further this connection between these two apparently different problems, the random-exchange Heisenberg model and the random flux problem.

In summary, we have studied the effects of non-magnetic impurities in gapful spin liquids. We have shown that the depletion of spins induces the random Berry phases, leading to the destructive interference of quantum fluctuations. The effective low-energy model for the randomly depleted spin ladder is the random-exchange Heisenberg chain, in which the generalized susceptibility for the staggered moment diverges as $T^{-1-2\alpha}$ ($\alpha \approx 0.22$). The correlation of the staggered spin moment becomes more classical due to the decoherence by the random Berry phase term.

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[1] M. Hase, I. Terasaki and K. Uchinokura: Phys. Rev. Lett. 70 (1993) 3651.
[2] For a recent review see E. Dagotto and T. M. Rice: Science 271 (1996) 618.
[3] L. P. Regnault, J. P. Renard, G. Dhalenne and A. Revcolevschi: Europhys. Lett. 32 (1995) 579.
[4] M. Azuma, Y. Fujishiro, M. Takano, T. Ishida, K. Okuda, M. Nohara and H. Takagi: preprint.
[5] P. Mendels et al. Phys. Rev. B 49 (1994) 10035; S. Uchida: private communications.
[6] H. Fukuyama, T. Tanimoto and M. Saito: J. Phys. Soc. Jpn. 65 (1996) 1183.
[7] H. Fukuyama, N. Nagaosa, M. Saito and T. Tanimoto: J. Phys. Soc. Jpn. 65 (1996) No. 8.
[8] M. Sigrist and A. Furusaki: J. Phys. Soc. Jpn. 65 (1996) No. 8.
[9] N. Furukawa et al.: preprint.
[10] See, for example, E. Fradkin: *Field Theories of Condensed Matter Systems*, (Addison-Wesley, Redwood City, 1991).
[11] F. D. M. Haldane: Phys. Rev. Lett. 50 (1983) 1153.
[12] D. Sénéchal: Phys. Rev. B 52 (1995) 15319.
[13] G. Sierra: J. Phys. A 29 (1996) 3299.
[14] E. Westerberg, A. Furusaki, M. Sigrist and P. A. Lee: Phys. Rev. Lett. 75 (1995) 4302.