Spin Waves in Random Spin Chains

Xin Wan and Kun Yang
National High Magnetic Field Laboratory and Department of Physics,
Florida State University, Tallahassee, Florida 32306

Chenggang Zhou and R. N. Bhatt
Department of Electrical Engineering and Princeton Materials Institutes,
Princeton University, Princeton, New Jersey 08544
(Dated: December 26, 2021)

We study quantum spin-1/2 Heisenberg ferromagnetic chains with dilute, random antiferromagnetic impurity bonds with modified spin-wave theory. By describing thermal excitations in the language of spin waves, we successfully observe a low-temperature Curie susceptibility due to formation of large spin clusters first predicted by the real-space renormalization-group approach, as well as a crossover to a pure ferromagnetic spin chain behavior at intermediate and high temperatures. We compare our results of the modified spin-wave theory to quantum Monte Carlo simulations.

One-dimensional (1D) quantum Heisenberg systems with random ferromagnetic-antiferromagnetic (FM-AFM) couplings are of interest due to the interplay between quantum fluctuations and disorder. An archetypical system is Sr₃CuPt₁₋ₓIrₓO₆ alloy of the pure compounds Sr₃CuPtO₆ (antiferromagnet) and Sr₃CuIrO₆ (ferromagnet). Another example is organic radical alloys. These systems are often referred to as quasi-1D random spin chains since the interchain coupling cannot be neglected at low temperatures where three-dimensional (3D) ordering dominates. Curie-like magnetic susceptibility with impurity-concentration dependent Curie constant has been reported for these systems.

Theoretically, random spin chains have been studied by high-temperature series expansions, quantum transfer matrix method, quantum Monte Carlo (QMC) simulations, density matrix renormalization group method, and most noticeably, the real-space renormalization-group (RSRG) method. In a simple RSRG picture, the Curie-like susceptibility occurs because ferromagnetic correlations lead to spin correlations that form spin clusters whose average size ⟨N⟩ and effective spin ⟨S_eff⟩ grow stochastically (⟨N⟩ ∝ A S_eff²) at low temperatures. The coefficient A depends on the distribution of random couplings, and, in general, increases with concentration of ferromagnetic couplings. Therefore, the magnetic susceptibility per spin (χ) varies with temperature (T) as

\[ χ = \frac{1}{3T} \frac{S^2_{eff}}{N} \propto \frac{1}{T} \]  

i.e. Curie-like, in the low-T limit. Here, we assume μ = k_B = 1 for convenience.

In the RSRG treatment, one decimates the spins that are coupled by the strongest coupling in the system, and renormalizes the couplings among the remaining spins perturbatively. This procedure generates effective spins with effective (in general weaker) couplings, so that excitations at lower and lower energy scale can be probed. It becomes obvious that an initial broad distribution of spin couplings ensures the quick approach to the asymptotic renormalization-group (RG) behavior. However, it is not the case for a Heisenberg spin chain with nearest-neighboring exchanges +J or −J with certain probabilities (a minimal model for real systems). In fact, an RSRG study of random spin chains has suggested the possibility of having a crossover region spanning more than five orders of magnitude in T before the true RSRG scaling regime can be reached. This suggests that the crossover physics may be more relevant to experimental findings.

In this paper, we model a random spin chain \( (S = 1/2) \) by the following Heisenberg Hamiltonian

\[ H = \sum_i J_i S_i \cdot S_{i+1}, \]  

where the coupling strength \( J_i \) is randomly chosen between \( J_{AF} \) (AFM) with probability \( p \) and \( −J_F \) (FM) with probability \( 1 − p \) (see Fig. 1). We are primarily interested in the dilute doping regime, \( i.e. \ p \ll 1 \), where RG-predicted Curie behavior is expected to occur at lower \( T \) with decreasing \( p \). In the absence of AFM couplings \( (J_{AF} = 0) \), the system is a collection of independent FM spin segments with exponentially distributed numbers of spins. For small \( p \), the static magnetic susceptibility \( χ \) of the isolated spin segments shows a crossover from an ordinary Curie behavior at high temperatures \( (T > J_F) \) to a characteristic \( χ \propto 1/T^2 \). This FM-chain behavior has been revealed by the modified spin-wave theory. Finite amount of \( J_{AF} \) introduces correlations between FM segments, which form spin clusters that grow, in a random walk fashion, both in size and total spin at decreasing \( T \). Statistical analysis shows that the RG-predicted Curie susceptibility

\[ χ_{RG} = \frac{S^2}{3T} \frac{1 − p}{p} \]  

is expected at low \( T \). Therefore, for dilute and not-too-weak AFM couplings, two crossover between distinctive \( T \)-dependence can be observed in this system.
Unfortunately, by generating effective couplings perturbatively, the RSRG method keeps discarding high-energy excitations and, thus, is not expected to describe correct physics well before the onset of the true RG behavior. To address these higher energy scales (at intermediate and high temperatures), we have generalized the modified spin-wave theory (MSW) to random spin systems, with dominating ferromagnetic couplings, to study the crossover behavior from high-temperature, short-distance physics to low-temperature, long-distance physics. Our data suggests an unambiguous crossover from FM spin-chain to random spin-chain behavior, which becomes wider for smaller $p$. Comparing with our QMC simulations using continuous time loop cluster algorithm, we discuss the limitations of the MSW method.

The technical details of the application of the MSW theory to random spin chains have been presented elsewhere. We summarize the MSW approach below. The main idea of the MSW theory is to introduce a chemical potential (equivalent to a uniform magnetic field) for the spin-wave excitations to ensure zero-magnetization of a FM segment, so that zero-magnetization of a segment implies zero-magnetization for every spin. With the physical significance of restoring the rotational symmetry broken during the introduction of spin waves, these zero-magnetization constraints ensure not only the finite number of spin waves at finite $T$, but the appropriate correlations between spin waves in different FM segments as well. A bosonic version of the generalized Bogoliubov transformation has been developed for this approach.

Figure 1 (left panel) shows the static magnetic susceptibility per spin $\chi$ for a random Heisenberg spin chain of 600 spins, distributed in 30 FM segments of average length 10 spins, $i.e.$ $p = 0.1$, coupled by 59 AFM couplings with $J_{AF} = 0.5J_F$. Three distinctive temperature regimes can be identified in the curve. For $T/J_F > 3$, $\chi$ can be fit to an ordinary Curie law of the form $\chi = C/T$. For $0.2 < T/J_F < 3$, $\chi$ rises with decreasing $T$, following the trend of a infinitely-long, pure FM spin chain. This implies that spins start to correlate, forming independent FM segments. The AFM couplings are still too weak to affect thermodynamics of the random spin chain at $T > 0.2J_F$. Below $T = 0.2J_F$, $\chi$ is in good agreement with the low-$T$, RG-predicted Curie behavior (Eq. 3 for $p = 0.1$). It is worth pointing out that we can identify the crossover temperature from the FM spin-chain physics to random spin-chain physics as the cross point of Eq. 3 and 4.

$$\chi_{FM} = \frac{1}{4} \left[ 0.1667J_F \frac{T}{T^2} + 0.581J_F^{1/2} \frac{T^{3/2}}{T^{3/2}} + 0.68 \frac{T}{T} + \ldots \right].$$

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In the dilute doping limit (small $p$), we have

$$\frac{T_X}{J_F} = \frac{p}{1 - p} \left[ 0.50 + 3.04 \sqrt{\frac{p}{1 - p}} + O \left( \frac{p}{1 - p} \right) \right].$$

For $p = 0.1$, $T_X = 0.17J_F$. The right panel in Fig. 2 shows averaged $\chi$ for 7 random realizations of 300 spins in 30 segments with $J_{AF} = 0.5J_F$. In this study, we require that the total magnetization of the corresponding classical ground state be zero, so $\chi$ vanishes at low-$T$ limit. This is responsible for the drop below $T < 0.02$, where there are no more segments to carry out RSRG.
FIG. 3: Averaged susceptibility $\chi$ of random Heisenberg spin chains of 360 spins in 30 FM segments, with AFM impurity couplings $J_{AF} = 0.5J_F$, for the MSW method (14 samples) and the QMC simulations (28 samples). The dashed line ($\chi = 11/12T$) is the low-$T$ Curie susceptibility expected by the RSRG approach (Eq. 3). The inset shows the same data on a log-log scale. The dotted line is the low-$T$ expansion of $\chi$ of a pure spin-1/2 FM chain (of infinite length) i.e. Eq. 4.

to lower $T$. Nevertheless, the crossover in the vicinity of $T_X = 0.17J_F$ is clearly visible.

We compare, in Fig. 3, results of $\chi$ averaged over different random realizations, obtained from both the MSW theory (14 samples) and QMC simulations (28 samples). For a system of 360 spins in 30 FM segments ($p = 1/12$) with AFM couplings ($J_{AF} = 0.5J_F$) between neighboring segments, we plot $\chi$ on both linear scale and log-log scale (inset). On the log-log scale, we can easily identify the crossover from the FM spin-chain physics to random spin-chain physics around $T_X = 0.13J_F$. The two methods are in good agreement, suggesting that the MSW theory can be generalized to a certain class of random spin chains. Near the crossover, the MSW method overestimates $\chi$, according to the QMC simulations. The MSW theory is not exact even in pure FM segments; up to 5% error can be observed for a 12-spin FM segment near this crossover temperature. In addition, by enforcing a periodic boundary condition for each FM segment, we add an extra FM coupling per segment, which can enhance the FM behavior of the system. We require that $\chi$ drop to zero at $T \to 0$; the MSW method seems to feature a faster drop for such a short chain, compared to the QMC simulations.

The modified spin-wave theory to random spin chains has been applied to one-dimensional Heisenberg ferrimagnets. In this case, two constraints corresponding to zero-magnetization on each of the two sublattices should be applied simultaneously, similar to the multiple zero-magnetization constraints on all FM segments in random spin chains. The linear combinations of the two chemical potentials (local fields), in our terminology, have the physical interpretations of uniform magnetic field and staggered magnetic field, which ensure both magnetization and staggered magnetization be zero. The MSW study on ferrimagnetic chains revealed that the zero-staggered-magnetization constraint keeps number of spin waves finite at both high-$T$ and low-$T$ limit, while the zero-magnetization constraint does only at the low-$T$ limit. We point out that the two chemical potentials are equivalent to the two kinds of Lagrangian multipliers in the Schwinger-boson mean-field study of the ferrimagnetic chains.

This work was supported by NSF grants No. DMR-9971541 (XW and KY), No. DMR-9809483 (CZ and RNB), the State of Florida (XW), and the Alfred P. Sloan Foundation (KY). Three of us (XW, KY and RNB) thank the Aspen Center for Physics for hospitality when this work has been written up.

1. T. N. Nguyen, P. A. Lee, and H.-C. zur Loye, Science 271, 489 (1996).
2. K. Mukai, K. Suzuki, K. Ohara, J. B. Jamali, and N. Achiwa, J. Phys. Soc. Jpn. 68, 3078 (1999).
3. S. H. Irons, T. D. Sangrey, K. M. Beauchamp, M. D. Smith, and H.-C. zur Loye, Phys. Rev. B 61, 11594 (2000).
4. A. Furusaki, M. Sigrist, P. A. Lee, K. Tanaka, and N. Nagaosa, Phys. Rev. Lett. 73, 2622 (1994).
5. A. Furusaki, M. Sigrist, E. Westerberg, P. A. Lee, K. Tanaka, and N. Nagaosa, Phys. Rev. B 52, 15930 (1995).
6. B. Frischmuth and M. Sigrist, Phys. Rev. Lett. 79, 147 (1997).
7. B. Frischmuth, M. Sigrist, B. Ammon, and M. Troyer, Phys. Rev. B 60, 3388 (1999).
8. B. Ammon and M. Sigrist, J. Phys. Soc. Jpn. 68, 1018 (1999).
9. K. Hida, J. Phys. Soc. Jpn. 66, 330 (1997).
10. T. Hikihara, A. Furusaki, and M. Sigrist, Phys. Rev. B 60, 12116 (1999).
11. E. Westerberg, A. Furusaki, M. Sigrist, and P. A. Lee, Phys. Rev. Lett. 75, 4302 (1995).
12. E. Westerberg, A. Furusaki, M. Sigrist, and P. A. Lee, Phys. Rev. B 55, 12578 (1997).
13. M. Takahashi and M. Yamada, J. Phys. Soc. Jpn. 54, 2808 (1985); M. Yamada and M. Takahashi, ibid. 55, 2024 (1986).
14. B. B. Beard and U.-J. Wiese, Phys. Rev. Lett. 77, 5130 (1996).
15. X. Wan, K. Yang, and R.N. Bhatt, Phys. Rev. B 66, 014429 (2002).
16. M. Takahashi, Prog. Theor. Phys. Suppl. 87, 233 (1986); Phys. Rev. Lett. 58, 168 (1987).
17. S. Yamamoto and T. Furui, Phys. Rev. B 57, 14008 (1998); S. Yamamoto, T. Fukui, K. Maisingher, and U. Schollwock, J. Phys.: Condens. Matter 10, 11033 (1998).
18. C. Wu, B. Chen, X. Dai, Y. Yu, and Z.-B. Su, Phys. Rev. B 60, 1657 (1999).