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Dynamics of weakly coupled random antiferromagnetic quantum spin chains

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We study the low-energy collective excitations and dynamical response functions of weakly coupled random antiferromagnetic spin-1/2 chains. The interchain coupling leads to Néel order at low temperatures. We use the real-space renormalization-group technique to tackle the intrachain couplings and treat the interchain couplings within the random phase approximation (RPA). We show that the system supports collective spin wave excitations, and calculate the spin wave velocity and spectra weight within RPA. Comparisons will be made with inelastic neutron scattering experiments on quasi-one-dimensional disordered spin systems such as doped CuGeO₃.

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Antiferromagnetic (AF) quantum spin chains have been of interest to physicists since the early days of quantum mechanics.¹ The one-dimensional nature of such systems allowed for tremendous theoretical progress both in clean systems by using exact solution and field theory mapping^{1,2} and disordered systems within renormalization-group framework.3-8 While such one-dimensional models have remarkably rich physics, in general, they do not give a complete description of real systems. Real spin chain compounds, such as CuGeO₃ (Ref. 9) and KCuF₃,¹⁰ always have some weak interchain couplings present, which can change the physics at lowest energy and/or temperature. For example, strictly one-dimensional (1D) models do not exhibit phase transitions into states with broken symmetry, while real spin chain systems often develop Néel order at very low temperatures due to the weak (3D) interchain couplings. It is thus important to study the effects of these interchain couplings to fully understand the low-energy and/or temperature physics of real spin chain compounds.

In this paper we study the low-energy collective excitations and dynamical response functions of weakly coupled, disordered AF spin-1/2 chains. Our work is motivated in part by the experimental studies on doped CuGeO₃. In the absence of doping, it is a spin-Peierls system in which the spins dimerize and form a gapped, nonmagnetic ground state. Upon doping, the system becomes disordered, and both dimerization and spin gap get suppressed. Amazingly, when doping reaches certain levels the spins become Néel ordered at low temperature, which has been observed experimentally in Zn- and Si-doped CuGeO₃.¹¹⁻²⁰ Since these experimental discoveries a number of theoretical papers have addressed the static Néel ordering in these systems²⁴⁻³⁶ using meanfield theory. On the other hand the collective excitations and dynamical response functions which have been studied experimentally using inelastic neutron scattering,²⁰ have not been studied theoretically thus far. The collective excitations and dynamical response functions are the subjects of the present work. We go beyond mean-field theory by allowing the Néel order parameter to fluctuate, and treat the interchain coupling using the random-phase approximation (RPA), while tackling the intrachain coupling using the real-space renormalization-group (RSRG) method.^{3,4} The RSRG technique has been proven to be powerful in obtaining magnetic and thermodynamics properties of random spin chains. Various numerical techniques²¹⁻²³ have also been deployed to

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study random chains and their results agree with those obtained by RSRG.

Our strategy here is similar to that of Schulz,³⁷ who studied weakly coupled pure chains. We find that despite the presence of disorder, the Néel state supports linearly dispersing spin waves, in agreement with experiments. We also obtain the spin wave velocity and the spectra weight of spin waves in the dynamical response function in terms of microscopic parameters of the system; this allows for detailed quantitative comparison between theory and inelastic neutron scattering experiments in the future.

Consider weakly coupled spin-1/2 antiferromagnetic chains with *z* nearest neighbor for each chain. The suitable Hamiltonian to describe this system is given by

$$H = \sum_{i,\vec{n}} J_{i,\vec{n}} \mathbf{S}_{i,\vec{n}} \cdot \mathbf{S}_{i+1,\vec{n}} + J_{\perp} \sum_{i,\vec{n},\vec{\delta}} \mathbf{S}_{i,\vec{n}} \cdot \mathbf{S}_{i,\vec{n}+\vec{\delta}},$$
(1)

where *i* is the site index along the chain, \vec{n} is the chain index, and δ is the index summed over the nearest neighbors. The intrachain couplings $J_{i,\vec{n}}$ are drawn from a random distribution function $P(J_{i,\vec{n}})$ (but with independent realizations for each chain), while the interchain coupling J_{\perp} is taken to be constant. Both of the intrachain and interchain couplings are taken to be positive. Treating the interchain couplings in the mean-field approximation,^{36,37} the presence of these couplings is effectively replaced by a staggered field which is responsible for long-range ordering at low temperature. The mean-field approximation for the interchain couplings can be described as follows: for a given site *i*, the staggered field acting on this site is determined by the magnetization of neighboring sites siting on neighboring chains. In general, the staggered field resulting from averaging the magnetization of the neighboring sites will be random. However, in the limit of infinite coordination number z (say in the limit of large dimensionality), the number of neighboring sites contributing to the staggered field at site *i* becomes infinitely many and the fluctuations are suppressed; the staggered field becomes uniform in this limit because it becomes the average of the magnetization over infinitely many neighboring sites (see below). With this simplifying approximations at hand, the original problem is reduced to a random AF spin chain in the presence of uniform staggered field

$$H_{1\mathrm{D}} = \sum_{i} J_i \mathbf{S}_i \cdot \mathbf{S}_{i+1} - h \sum_{i} (-1)^i S_i^z.$$
 (2)

The staggered field h is obtained through mean-field selfconsistency condition

$$h = zJ_{\perp}m, \tag{3}$$

where *m* is the disorder-averaged staggered magnetization. The staggered magnetization at site *i* is $m_i = (-1)^i \langle S_i \rangle$. The resulting effective 1D problem can be solved using the RSRG method,^{3,4} from which the phase diagrams of the systems have been obtained for various cases.³⁶

In the present work we go beyond the static mean-field approximation and calculate the dynamical response functions by treating interchain couplings within the RPA, from which we also obtain the collective mode spectrum of the system. Within the RPA the dynamic susceptibility is given by³⁷

$$\widetilde{\chi}_{\rm RPA}^{\alpha\beta} = \frac{\widetilde{\chi}_{\rm 1D}^{\alpha\beta}}{1 - zJ_{\perp}\widetilde{\chi}_{\rm 1D}^{\alpha\beta}},\tag{4}$$

where $\tilde{\chi}_{1D}^{\alpha\beta}$ is the disorder-averaged single chain susceptibility matrix in the presence of the staggered field that satisfies Eq. (3). The expression given above is valid for transverse and longitudinal dynamic susceptibility. In this work we focus on the transverse response function since the transverse part couples more directly to the collective excitation of the system than the longitudinal part does. Further motivation to study the transverse susceptibility is provided by recent experiments which focus on the transverse part; hence, $\alpha\beta$ = +- in Eq. (4). It is worth noting that although we concentrate on the transverse dynamic response in our calculation, the formalism developed here can be readily applied to obtain the longitudinal dynamic response and to study other random spin chains.

Let us continue our discussion on the transverse dynamic response. For a specific disorder configuration, the chain susceptibility $\tilde{\chi}_{1D}^{\alpha\beta}$ represents the dynamical response of the chain at wave vector q' to an external perturbation at wave vector q; in general q' can be of any value due to the presence of disorder, which breaks the translational symmetry. The symmetry is restored, however, once disorder averaging is performed (in fact, the system is self-averaging), except for the doubling of unit cell by the staggered field. Thus a perturbation with wave vector q also induces response at another wave vector $q'=q+\pi$, in addition to the usual response at q'=q. Hence the chain transverse susceptibility matrix is represented by a 2×2 matrix:

$$\widetilde{\chi}_{1\mathrm{D}}^{+-}(q,\omega) = \begin{pmatrix} \chi_{1\mathrm{D}}^{+-}(q,q,\omega) & \chi_{1\mathrm{D}}^{+-}(q,q+\pi,\omega) \\ \chi_{1\mathrm{D}}^{+-}(q+\pi,q,\omega) & \chi_{1\mathrm{D}}^{+-}(q+\pi,q+\pi,\omega) \end{pmatrix}.$$

As a consequence of this, we can rewrite the RPA susceptibility, Eq. (4) as follows:

$$\widetilde{\chi}_{\text{RPA}}^{+-}(q,\omega) = \frac{1}{D(q,\omega)} \begin{pmatrix} \chi_{11}(q,\omega) & \chi_{12}(q,\omega) \\ \chi_{21}(q,\omega) & \chi_{22}(q,\omega) \end{pmatrix}, \quad (5)$$

where $D(q, \omega)$ is the determinant of $1-zJ_{\perp}\tilde{\chi}_{1D}^{+-}(q, \omega)$. Thus the calculation of $\tilde{\chi}_{RPA}^{+-}(q, \omega)$ reduces to the calculation of $\chi_{1D}^{+-}(q, q', \omega)$.

PHYSICAL REVIEW B 72, 020403(R) (2005)

To calculate $\chi_{1D}^{+-}(q,q',\omega)$, we use the RSRG method. In the RSRG scheme, one assumes that the two spins that are coupled by the highest-energy bond form a ground state on this bond (singlet when there is no field); this bond is eliminated and new effective bonds between the remaining spins are generated perturbatively; this process is repeated until the ultimate low-energy limit is reached. Within this scheme the single chain response functions are approximated by the sum of those strongly coupled bonds that form during the RG process, and the coupling among different pairs of spins are neglected, as outlined in Ref. 38. This approximation is asymptotically exact in the low-energy limit. The difference between the present case and that of Ref. 38 is that in addition to the AF bonds we also have the staggered field, which complicates the RG process. However, it has been shown earlier³⁶ that in the limit of a weak field (corresponding to weak interchain coupling), its effect on the RG flow is negligible and we thus do not consider it here. We thus start by considering a spin pair connected by a strong bond in the presence of a staggered field whose Hamiltonian is given by

$$H_0 = \Omega \mathbf{S}_1 \cdot \mathbf{S}_2 + h(S_1^z - S_2^z), \tag{6}$$

where Ω is the bond connecting the spin pair which we will identify as the cutoff of the system at a given stage of RG and *h* is the uniform staggered field as defined in Eq. (3); the eigenstates of this Hamiltonian are

$$\begin{aligned} |0\rangle &= 1/\sqrt{1+C_0^2}(C_0|++\rangle+|--\rangle), \\ |1\rangle &= 1/\sqrt{1+C_1^2}(C_1|++\rangle+|--\rangle), \\ |2\rangle &= |++\rangle; \quad |3\rangle &= |--\rangle, \end{aligned}$$
(7)

with the corresponding energy eigenvalues:

$$E_0 = -\Omega/4 - \sqrt{(\Omega/2)^2 + h^2},$$

$$E_1 = -\Omega/4 + \sqrt{(\Omega/2)^2 + h^2},$$

$$E_2 = E_3 = \Omega/4,$$
(8)

where the coefficients $C_0 = 2h/\Omega - \sqrt{1 + (2h/\Omega)^2}$ and $C_1 = 2h/\Omega + \sqrt{1 + (2h/\Omega)^2}$. Equipped with the spectrum of the spin pair, we proceed to calculate the dynamic structure factor for the pair. The T=0 spectral representation of the dynamic structure factor for a spin pair in the presence of uniform staggered field is

$$S_{pair}^{+-}(q_1, q_2, \omega) = \sum_{m} \langle 0|S_{-q_1}^+|m\rangle \langle m|S_{q_2}^-|0\rangle \,\delta(\omega - \Delta E)$$

=
$$\frac{(C_0 + e^{-iq_1l})(C_0 + e^{iq_2l})}{1 + C_0^2} \,\delta(\omega - \Omega/2)$$

-
$$\sqrt{(\Omega/2)^2 + h^2}, \qquad (9)$$

where $S_q^{\pm} = S_1^{\pm} + e^{iql}S_2^{\pm}$ is the Fourier transform of the spin operator, l is the distance between two spins, $|0\rangle$ is the ground state, and $|m\rangle$ are the excited states $|++\rangle$ and $|--\rangle$ of such pair as written explicitly in Eq. (7). The system can only be excited to states different m_z value because the operator S_q^{\pm} connects states with different m_z value; the transition can only happen from the ground state to the states $|++\rangle$ and $|--\rangle$.

DYNAMICS OF WEAKLY COUPLED RANDOM...

To calculate the dynamics structure factor of the whole chain, we use the joint distribution function of bond length and strength, characterized in detail in Ref. 4, and sum up contributions of all the strongly coupled bonds that are formed through the RSRG process. We follow the procedure outlined in Ref. 38 closely to obtain the dynamic structure factor of a single chain by summing the contribution from strongly coupled pairs; the dynamic structure factor for a single chain is given by

$$S_{1\mathrm{D}}^{+-}(q_1, q_2, \omega) = n_{\Gamma_{\Omega}} \int dl d\zeta P(\zeta, l; \Gamma_{\Omega}) S_{pair}^{+-}(q_1, q_2, \omega),$$
(10)

where $P(\zeta, l; \Gamma_{\Omega})$ is the joint distribution of the bond-length and strength. We follow the definitions outlined in Ref. 4 to denote $n_{\Gamma_{\Omega}}$ as the fraction of spins left at energy scale Γ_{Ω} , $\zeta = \ln(\Omega/J)$ as the log energy scale, $\Gamma_{\Omega} = \ln(\Omega_0/\Omega)$ as the log cutoff of the energy scale, Ω_0 as the nonuniversal energy cutoff of the original Hamiltonian, and Ω as the energy cutoff of the renormalized problem. The transverse susceptibility for a single chain is then obtained by integrating the dynamic structure factor:

$$\chi_{1\rm D}^{+-}(q_1, q_2, \omega) = \int \frac{d\omega'}{\pi} \frac{S_{1\rm D}^{+-}(q_1, q_2, \omega')}{\omega' - \omega - i\epsilon},$$
 (11)

where $q_1, q_2=q$ or $q+\pi$ and $S_{1D}^{+-}(q_1, q_2, \omega)$ is given by Eq. (10).

We would like to study the collective excitations of the system, whose spectrum is given by the singularity of $\tilde{\chi}_{\text{RPA}}^+$, Eq. (4), or the condition $D(q, \omega)=0$. The rotational invariance of the system combined with the self-consistent condition Eq. (3) requires $D(q=0, \omega=0)=0$, because the staggered field can be rotated without affecting the self-consistency. As a consequence the energy of the collective mode vanishes as the wave vector q goes to zero. Thus to obtain the collective mode dispersion it is natural to expand the quantities $\chi_{1\text{D}}^{+-}(q,q,\omega), \ \chi_{1\text{D}}^{+-}(q,q+\pi,\omega), \ \chi_{1\text{D}}^{+-}(q+\pi,q,\omega), \ \text{and} \ \chi_{1\text{D}}^{+-}(q+\pi,q,\omega)$

$$\chi_{1\mathrm{D}}^{+-}(q,q,\omega) \simeq \chi_{1\mathrm{D}}^{+-}(0,0,0) + a_{q,q}\omega^{2} + b_{q,q}q^{2},$$

$$\chi_{1\mathrm{D}}^{+-}(q,q+\pi,\omega) \simeq a_{q,q+\pi}\omega + ib_{q,q+\pi}q,$$

$$\chi_{1\mathrm{D}}^{+-}(q+\pi,q,\omega) \simeq a_{q,q+\pi}\omega - ib_{q,q+\pi}q,$$

$$\chi_{1\mathrm{D}}^{+-}(q+\pi,q+\pi,\omega) \simeq \chi_{1\mathrm{D}}^{+-}(\pi,\pi,0) + a_{q+\pi,q+\pi}\omega^{2} - b_{q,q}q^{2},$$

(12)

where the expansion coefficients are given by

$$\begin{split} a_{q,q} &= 2 \int \frac{d\omega'}{\pi} \frac{S_{1\mathrm{D}}^{+-}(0,0,\omega')}{\omega'^3}, \\ b_{q,q} &= -\frac{2l_v}{15} \int \frac{d\omega'}{\pi} \frac{C_0}{1+C_0^2} \ln(\Omega_0/\Omega) \frac{1}{\omega'} \frac{\omega'^2 + h^2}{\omega'^2 - h^2}, \\ a_{q,q+\pi} &= 2 \int \frac{d\omega'}{\pi} \frac{S_{1\mathrm{D}}^{+-}(0,\pi,\omega')}{\omega'^2}, \\ b_{q,q+\pi} &= \frac{4}{3} \int \frac{d\omega'}{\pi} \frac{C_0}{1+C_0^2} \frac{1}{\ln(\Omega_0/\Omega)} \frac{1}{\omega'^2} \frac{\omega'^2 + h^2}{\omega'^2 - h^2}, \end{split}$$

PHYSICAL REVIEW B 72, 020403(R) (2005)

$$a_{q+\pi,q+\pi} = 2 \int \frac{d\omega'}{\pi} \frac{S_{1D}^{+-}(\pi,\pi,\omega')}{{\omega'}^3},$$
 (13)

where

$$S_{1D}^{+-}(0,0,\omega) = \frac{(1+C_0)^2}{1+C_0^2} \frac{1}{l_v \ln^3(\Omega_0/\Omega)} \frac{1}{\omega} \frac{\omega^2 + h^2}{\omega^2 - h^2},$$

$$S_{1D}^{+-}(0,\pi,\omega) = \frac{1-C_0^2}{1+C_0^2} \frac{1}{l_v \ln^3(\Omega_0/\Omega)} \frac{1}{\omega} \frac{\omega^2 + h^2}{\omega^2 - h^2},$$

$$S_{1D}^{+-}(\pi,\pi,\omega) = \frac{(1-C_0)^2}{1+C_0^2} \frac{1}{l_v \ln^3(\Omega_0/\Omega)} \frac{1}{\omega} \frac{\omega^2 + h^2}{\omega^2 - h^2},$$
 (14)

where $l_v = a/[\ln(\Omega_0/J)]$ is the microscopic length scale determined by the initial bond distribution; we use \bar{x} to denote the variance of x. Using the condition that $D(q, \omega) = \det[1 - zJ_{\perp}\tilde{\chi}_{1\mathrm{D}}^{+-}(q, \omega)] = 0$ for $\omega = q = 0$, we obtain quartic equation in ω

$$A\omega^4 + B\omega^2 + C = 0, \qquad (15)$$

where the coefficients A, B, and C are

$$A = (zJ_{\perp})^{2}a_{q,q}a_{q+\pi,q+\pi},$$

$$B = -zJ_{\perp}[1 - zJ_{\perp}\chi_{1D}^{+-}(0,0,0)]a_{q+\pi,q+\pi}$$

$$+ (zJ_{\perp})^{2}[(a_{q+\pi,q+\pi} - a_{q,q})b_{q,q}q^{2} - a_{q,q+\pi}^{2}],$$

$$C = [zJ_{\perp}(1 - zJ_{\perp}\chi_{1D}^{+-}(0,0,0) \quad b_{q,q}$$

$$- ((zJ_{\perp})^{2}b_{q,q+\pi}^{2})]q^{2} - (zJ_{\perp})^{2}b_{q,q}^{2}q^{4}.$$
 (16)

The solution to this quartic equation gives us the spin-wave dispersion of the system. To the leading order of the wave vector q we obtain a linear dispersing spin wave $\omega = v_s q$, with

$$v_s \simeq \sqrt{\frac{\pi}{90}} z J_\perp m \ln^{3/2} (\Omega_0 / z J_\perp m) l_v. \tag{17}$$

In obtaining the result above for the spin wave velocity, we have explicitly worked in the limit $J_{\perp} \rightarrow 0$.

We now calculate the dynamic structure factor within RPA, which can be accessed through inelastic neutron scattering (INS) experiment. To obtain the dynamic structure factor near q=0, we take the imaginary part of the upper left component of the RPA susceptibility matrix, Eq. (5), i.e., $S_{\text{RPA}}=\mathcal{I}(\chi_{11}(q,\omega)/D(q,\omega))$,

$$S_{\text{RPA}}(q,\omega) = \mathcal{I}\left[\frac{\chi_{11}(q,\omega)}{A\omega^4 + B\omega^2 + C - i\delta}\right],$$
(18)

where a small imaginary part $i\delta$ has been introduced to shift the pole in the determinant $D(q, \omega)$ to slightly above the real axis. The pole in the determinant is realized when $\omega = v_s q$, where v_s is the spin-wave velocity given in Eq. (17). A straightforward calculation results in a simple form of the RPA dynamic structure factor near q=0:

$$S_{\rm RPA} = \frac{1}{3} \frac{v_s q}{z J_\perp} \delta(\omega - v_s q).$$
(19)

Following the same procedure, we also obtain the dynamic structure factor near $q = \pi + \delta q$, $S_{\text{RPA}} = \mathcal{I}(\chi_{22}(q, \omega)/D(q, \omega))$. The result is as follows:

$$S_{\rm RPA} = \frac{1}{2} \frac{z J_{\perp} m^3 \ln^2(\Omega_0 / z J_{\perp} m)}{v_s(\delta q)} \delta(\omega - v_s(\delta q)), \qquad (20)$$

where m is disorder-averaged staggered magnetization and Ω_0 is the nonuniversal cutoff for the chain. Our calculation predicts that a sharp peak develops at the pole where ω $=v_{s}q$. The intensity of the peak is proportional to the wave vector $q(1/\delta q)$ near $q=0(\pi)$. Within the random phase approximation framework we only get a sharp peak of the intensity, as is shown by the δ function in the dynamic structure factor. The peak shows a more pronounced contribution from the dynamic structure factor near $q = \pi$ because the long-range staggered configuration is realized near this wave vector. We comment here that while our expression for v_s [Eq. (17)] involves parameters of the random distribution $(\Omega_0 \text{ and } l_n)$ that cannot be measured directly, our results on the spectral weight can be compared directly with the intensity of inelastic neutron scattering experiment, once v_s is determined from the measurement; this is because Eq. (19) involves v_s and other measurable quantities only, and the same combination of Ω_0 and and other measurable quantities appears in Eqs. (17) and (20) (l_v) is of order one lattice spacing for generic distributions). Thus our results allow for a detailed quantitative comparison with future experiments.

A few years earlier, Martin *et al.* studied the excitation spectrum of doped CuGeO₃ using inelastic neutron scattering,¹⁷ and found sharp propagating spin-wave excitations when the system is Néel ordered, despite the fact that the Néel phase is stabilized by disorder. They found the spin-

PHYSICAL REVIEW B 72, 020403(R) (2005)

wave spectrum to be linear. Our results agree with these experimental findings, and it is clear that such propagating excitations must be collective modes stabilized by interchain couplings, as single random chains do not support such propagating modes.³⁸ In the present work we assume there is no dimerization, while in doped CuGeO₃ dimerization survives and coexists with Néel order when doping level is sufficiently low. It is straightforward to generalize the present approach to the case with dimerization,^{5,36} as well as finite temperature and chains with other spin sizes. Recently, Masuda et al.³⁹ studied the dynamic spin-spin correlation of a new compound $BaCu_2(Si_{1-x}Ge_x)_2O_7$ using inelastic neutron scattering. This system can be described very well by antiferromagnetic spin-1/2 chains with random exchange due to the random distribution of Si and Ge atoms. The experimental data on the dynamic structure factor on this compound fit the universal scaling form predicted in Ref. 38 very well. Our theoretical work proposed here could be of relevance to this new experimental realization of the random exchange antiferromagnetic spin-1/2 chain; in particular it would be interesting to study the collective excitation on this compound in the ordered phase and compare it with the results obtained here. We hope that the present work will motivate future experiments that will study the spectral weights of the spin waves in detail, and test the predictions made here on them

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