

Low-energy dynamical characteristics of a quantum spin chain with magnetic impurities

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In the framework of the exactly solvable model, using nonperturbative methods, the temperature and magnetic field behavior of the NMR relaxation rate (and low-energy dynamical structure factor) for the antiferromagnetic Heisenberg spin-1/2 chain with magnetic impurities is calculated. For a single magnetic impurity, totally screened at low temperatures by spinons of the chain, the critical exponents, which determine the low-temperature and magnetic field behavior, get renormalized. For the finite concentration of impurities with the random distribution of the impurity-chain coupling the weak disorder in the distribution does not produce essential changes to the dynamical structure factor. Contrary, the strong disorder yields drastically different behavior of the low-energy dynamical characteristics, comparing to the homogeneous chain. Our results are generic, and they qualitatively agree in limiting cases with the results of perturbative calculations for disordered spin chains and with the data of experiments on quasi-one-dimensional spin compounds.

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I. INTRODUCTION

During recent years the interest in low-dimensional quantum systems has grown substantially, due to several reasons. First, last decades celebrated a great progress in synthesizing materials with low-dimensional electron subsystems, such as oxides of transition metals (such as copper and vanadium) or organic compounds. Second, the theory of one-dimensional-correlated electron models is one of the most developed parts of many-body quantum theory, which permits to calculate in many cases exactly their characteristics. Finally, one-dimensional quantum systems are often used to realize the ideas of the quantum information. The construction of quantum computers based on qubits (elementary cells, which states are entangled, hence, permit to manipulate with them using the quantum coherence), and the use of the quantum communication, where the entanglement also plays the major role, are among the main directions in the application of one-dimensional quantum systems.

The one-dimensional Heisenberg model with antiferromagnetic interactions between neighboring spins 1/2 is the seminal model of the modern quantum physics. The eigenstates of this model are strongly entangled, and, thus, the quantum coherence is manifested in numerous characteristics of the model. The model is integrable, i.e., infinite series of integrals of motion (which commute mutually and with the Hamiltonian of the model) are possible to construct. In real low-dimensional spin systems one usually deals not only with the homogeneous system but also with a system with imperfections, e.g., impurities. We can distinguish between magnetic and nonmagnetic impurities (latter effectively cut the spin chain into segments with open edges). Due to peculiarities in the one-dimensional density of states, impurities can drastically change the behavior of one-dimensional spin systems, in particular, the presence of impurities in an electron chain can yield a localization of eigenstates. This is why, one mostly needs to take into account impurities in one-dimensional systems nonperturbatively. It is possible to real-

ize in the framework of integrable spin models with impurities.¹ Static characteristics, such as the magnetic susceptibility, specific heat, etc., were calculated exactly for those models using the Bethe ansatz. However, the calculation of dynamical characteristics is more difficult, and only few exact results were obtained for spin chains with impurities.

Important low-energy dynamical characteristics of the spin system can be measured in the nuclear magnetic resonance (NMR) experiments. The main NMR characteristics are the Knight shift of the position of the NMR, and the relaxation time T_1 . The Knight shift for antiferromagnetic interactions is proportional to the static (local) magnetic susceptibility of the system. The NMR relaxation rate T_1^{-1} can be presented as²

$$T_1^{-1} = (g^2 \mu_B^2 \gamma_N^2 / 2) \times \int dq [F^z(q) S^{zz}(q, \omega_N) + F^x(q) S^{xx}(q, \omega_N)], \quad (1)$$

where γ_N is the gyromagnetic ratio of the nuclear spin, ω_N is the resonance frequency of nuclear spins, $F^{z,x}(q)$ are the hyperfine form factors of nuclear spins, parallel and perpendicular to the static magnetic field B , $S^{\mu\nu}(q, \omega_N)$ ($\mu\nu=x, z$) are the components of the tensor of the dynamical structure factor (DSF) of the spin chain, also parallel and perpendicular to B , g is the g factor of magnetic ions, and μ_B is Bohr's magneton. $\hbar\omega_N \ll J, H$, where $H = g\mu_B B$ and J defines the exchange interaction between electron spins, hence, the limit $\omega_N \rightarrow 0$ can be applied. For the transverse components we have $S^{xx} = S^{yy}$, because of the rotational symmetry perpendicular to the field direction. The NMR relaxation rate of the chain with embedded nonmagnetic impurities, which cut the chain into finite-size fragments, was studied in Ref. 3. There open boundary conditions produce divergent behavior of the relaxation rate $T_1^{-1} \sim T[\ln(T_0/T)]^2$, where $T_0 \sim J$ is a constant of order of the exchange integral.

In this paper, using nonperturbative methods, we find the generic low-energy dynamical properties of an antiferromagnetic spin-1/2 chain with *magnetic* impurities. We show that the behavior of the NMR relaxation rate in the chain is drastically changed due to the presence of single impurities, and the finite concentration of magnetic impurities with randomly distributed parameters.

II. INTEGRABLE MODEL OF A SPIN CHAIN WITH MAGNETIC IMPURITIES

Consider the spin chain with magnetic impurities, in which the impurity is characterized by the value of the impurity-host coupling, equal or smaller than the exchange constant in the chain $J > 0$ (in that case we expect the absence of impurity-caused local levels for the generic situation with periodic boundary conditions¹), and/or by the value of the impurity spin $S'_j \neq 1/2$. The Hamiltonian of the integrable lattice version of the Heisenberg spin-1/2 chain with magnetic impurities has the form^{1,4}

$$\mathcal{H} = J \sum_j [h_{j,j+1} + x \tilde{h}_{j,j+1}], \quad (2)$$

where

$$h_{j,j+1} = (\mathbf{S}_j \cdot \mathbf{S}_{j+1}) \quad (3)$$

describes the interaction in the host chain, and $x = 4/[4\theta_j^2 + (2S'_j + 1)^2]^{-1}$, and

$$\begin{aligned} \tilde{h}_{j,j+1} = & [\theta_j^2 - 2S'(S' + 1)](\mathbf{S}_j \cdot \mathbf{S}_{j+1}) + [(\mathbf{S}_j + \mathbf{S}_{j+1}) \cdot \mathbf{S}'_j] \\ & + [(\mathbf{S}_j \cdot \mathbf{S}'_j), (\mathbf{S}_{j+1} \cdot \mathbf{S}'_j)] + 2i\theta_j[(\mathbf{S}_j \cdot \mathbf{S}'_j), (\mathbf{S}_{j+1} \cdot \mathbf{S}'_j)], \end{aligned} \quad (4)$$

where $[a, b]$ ($\{a, b\}$) denote commutator (anticommutator) of the operators a and b and the impurity spin \mathbf{S}'_j is coupled to two neighboring spins, \mathbf{S}_j and \mathbf{S}_{j+1} , of the host chain. Each impurity has the properties of a forward and backward scatterer.^{1,4} It is characterized by two parameters, its spin S'_j and real θ_j , which defines the value of the impurity-host couplings⁴ $J'_j = J/(\theta_j^2 + [S'_j + (1/2)]^2) \leq J$. Even for $S'_j = 1/2$ the impurity couplings differ from the host one due to $\theta_j \neq 0$. Despite the complicated form of the lattice integrable Hamiltonian, in the long-wave limit [from the renormalization-group (RG) viewpoint two last terms are less relevant than two-spin couplings] the impurity-host interaction is $J'_j(\mathbf{S} \cdot \mathbf{S}')$, i.e., it has the standard form of the local exchange coupling between the spins of the impurity and the host. Without the loss of generality we consider such a concentration of impurities, that they do not directly interact with each other. The model describes generic features of magnetic impurities (each impurity is characterized by the different coupling to the host) in the spin-1/2 Heisenberg chain, because the results of exact Bethe ansatz calculations for this model^{1,4} agree with the results of RG calculations⁵ for the “standard” Heisenberg chain with random values of exchange couplings between spins.

The behavior of each impurity depends on the value of its spin. We can introduce the effective Kondo temperature for each impurity,^{1,4} as

$$T_{K,j} = \frac{\pi J}{2} \exp(-\pi \sqrt{[(J/J'_j) - [S'_j + (1/2)]^2]}). \quad (5)$$

For $S' > 1/2$ the situation is analogous to the underscreened Kondo impurity in a metal.⁶ For $T > aT_{K,j}$, where a is a non-universal constant and in the ground state for $H > bT_{K,j}$, where $b = 2\sqrt{\pi/e}$, the impurity behaves as asymptotically free spin with the value¹ S'_j . On the other hand, at $T = 0$ for $H < bT_{K,j}$ and for $T < bT_{K,j}$ at $H = 0$, spinons (elementary excitations of the spin chain) partly screen the impurity, and it behaves as the asymptotically free spin¹ ($S'_j - 1/2$).

III. DYNAMICAL CHARACTERISTICS OF A SPIN CHAIN WITH A SINGLE MAGNETIC IMPURITY

The approach, similar to Refs. 1 and 4, yields that the dynamical characteristics of the spin chain with a single impurity in the main approximation are the sum of two contributions.

A. Contribution to the NMR linewidth from a homogeneous spin chain

The first contribution comes from the host chain itself. The asymptotic behavior of the correlation functions of the chain can be calculated in the framework of the conformal-field theory.¹ The most important contribution for $S^{zz}(q, \omega \rightarrow 0)$ comes from $q = 0$ and $q = \pi(1 - 2m)$, where $0 \leq m \leq 1/2$ is the magnetic moment per site of the host chain. At $H_s = 2J$ the quantum phase transition to the spin-saturated phase takes place. For $H > H_s$ in the ground state we have $m = 1/2$ and the relaxation rate for the spin chain (with and without impurities) is exponentially small due to that gap, and we will not consider it below. For $S^{xx}(q, \omega \rightarrow 0)$ the main contribution comes from $q = \pi$ and $q = 2\pi m$. For the longitudinal component of the DSF at $q \sim 0$ we have

$$S_h^{zz} = \frac{2KT}{\pi v^2}, \quad (6)$$

where K is the critical exponent and v is the Fermi velocity of a spinon. Dependencies $v(H)$ and $K(H)$ can be obtained from the Bethe ansatz solution.¹ Recently a simple ansatz for the magnetic field behavior of the velocity v and exponent K , valid in the interval $0 \leq H \leq H_s$, was proposed⁷

$$\begin{aligned} v &= (\pi J/2) \left(\left[1 - \frac{H}{H_s} \right] \left[1 - \frac{H}{H_s} + \frac{2H}{\pi J} \right] \right)^{1/2}, \\ K &= f/\sqrt{4f^2 - 3H^2}, \quad f = \pi J \left[1 - \frac{H}{H_s} \right] + H. \end{aligned} \quad (7)$$

The behavior of $v(H)$ and $K(H)$, given by those expressions, agrees with the Bethe ansatz. For $q \sim \pi(1 - 2m)$ we get

$$S_s^{zz} \sim \frac{C_1 \cos(2\pi K)}{v} B^2[(K/2), 1 - K] \left(\frac{2\pi T}{v} \right)^{2K-1}, \quad (8)$$

where $B(x, y) = \Gamma(x)\Gamma(y)/\Gamma(x+y)$ is the beta function and C_1 is the magnetic field-dependent multiplier.⁸ For the trans-

verse component of the DSF at $q \sim \pi$, we have

$$S_s^{xx} \sim \frac{C_2 |\cos(2\pi\gamma')|}{v} |B^2[(\gamma'/2), 1 - \gamma']| \left(\frac{2\pi T}{v} \right)^{2\gamma'-1}, \quad (9)$$

where $\gamma' = 1/(4K)$ and C_2 is magnetic field dependent. At $H=0$ the transverse component of the DSF at $q \sim 2\pi m$, calculated in the field-theory approach, diverges, which is known to be incorrect. That transverse component of the DSF has to behave as the homogeneous longitudinal component for small H and like the staggered transverse component near H_s . Marginal operators (from the RG viewpoint) introduce logarithmic corrections to the asymptotic behavior of correlation functions of the spin chain at low temperatures.¹ Those corrections can be taken into account, see, e.g., Ref. 9, which yields the additional multiplier to the DSF, $\sqrt{\ln(24.27J/k_B T)/(2\pi)^{3/2}}$.

B. Contribution to the NMR linewidth from the underscreened magnetic impurity

The other contribution comes from the asymptotically free spin (either of the value S' or of the value $(S' - 1/2)$, depending on T , H , and $T_{K,j}$). Its behavior is well known, see, e.g., Ref. 10. If the concentration of $S'_j > 1/2$ impurities is finite, contributions to the DSF from each impurity come separately (except of the case, when all spins of impurities and all θ_j are almost equal, when impurities form the impurity band, see the discussion below). This behavior of the dynamical characteristics of a single $S'_j > 1/2$ impurity in the integrable spin chain Eq. (1) is physically clear; it supports the generic nature of the studied model.

C. Dynamical characteristics of a spin chain with the totally screened magnetic impurity

For large values of the temperature and the external magnetic field the single-impurity spin $S'_j = 1/2$ behaves as the asymptotically free spin $1/2$. However, in the ground state for $H < bT_{K,j}$ and for $T < aT_{K,j}$ at $H=0$, spinons totally screen the impurity spin.⁴ The static magnetic susceptibility of the impurity is finite at $T \rightarrow 0$ or at $H \rightarrow 0$. Its value is inverse proportional to $T_{K,j}$ (not to the velocity of a spinon, as for the homogeneous chain).¹ The dynamical characteristics of the impurity are calculated in the similar way, as the ones for the homogeneous chain. Namely, the asymptotic behavior of correlation functions is given by the conformal-field theory, in which $T_{K,j}$ plays the role of the “velocity” for the impurity.⁴ The exponents are calculated as finite-size corrections to the Bethe ansatz equations,¹ using the Euler-MacLaurin formula. Parameters, which describe how the impurity affects the exponents, (n_{imp} and d_{imp} , see below) renormalize the values of the quantum numbers (zero modes in the bosonization), which define the change in magnetization and the transfer from the one Fermi point to the other one due to excitations. The new (renormalized, due to n_{imp} and d_{imp}) quantum numbers determine the conformal weights, and, hence, the exponents for correlation functions in the conformal-field theory. The DSF for the impurity are given by similar expressions as for the contribution from the

homogeneous chain, where we have to replace $\pi J/2 \rightarrow T_{K,j}$ in the expression for the velocity and exponents for the staggered components of the DSF as

$$K \rightarrow K[(1 + d_{imp})^2 + (n_{imp}/2K)^2],$$

$$\gamma' \rightarrow \frac{1}{4K} [(1 + n_{imp})^2 + (2Kd_{imp})^2]. \quad (10)$$

From the calculations of finite-size corrections to the energy of the considered integrable model we get

$$n_{imp} = \int_{-A}^A dx \sigma(x), \quad d_{imp} = \int_A^\infty dx \frac{\sigma(-x) - \sigma(x)}{2}, \quad (11)$$

where $\sigma(x)$ satisfies the equation

$$\sigma(x) + \frac{1}{2\pi} \int_{-A}^A dy \frac{4\sigma(y)}{(x-y)^2 + 4} = \frac{1}{\pi[1 + (x + \theta_j/2)^2]}. \quad (12)$$

n_{imp} is related to the magnetic moment of the impurity via $n_{imp} = (1/2) - m_{imp}$. The limits of integration $A > 0$ play the role of Fermi edges. They are H dependent: $A=0$ for $H=H_s$ and $A=\infty$ for $H=0$. n_{imp} monotonically decreases from $1/2$ to 0 and d_{imp} increases from 0 to $\pi^{-1} \tan^{-1}(\theta_j/2)$ with the growth of H from zero to H_s . It means that the exponents in Eq. (10), which determine the behavior of correlation functions, and, hence, of the DSF and NMR linewidth, change their values from $K=5/8$ and $\gamma'=9/8$ in the absence of the external magnetic field, at $H=0$, to $K=[1 + \pi^{-1} \tan^{-1}(\theta_j/2)]^2$ and $\gamma'=(1/4) + [\pi^{-1} \tan^{-1}(\theta_j/2)]^2$ at the saturation field, $H=H_s$.

The calculation of the renormalization of exponents is totally analogous to that in the recently developed approach, which takes into account the nonlinearity of the dispersion relations in the spinless Luttinger liquid.¹¹ The difference is in the definition of n_{imp} and d_{imp} . In Ref. 11 the artificial impurity has the parameters, related to the characteristics of a high-energy excitation (or a hole). Hence, magnetic impurities, studied in our paper, can also produce additional features in the high-energy dynamics of the spin chain.

IV. FINITE CONCENTRATION OF MAGNETIC IMPURITIES

A. NMR linewidth of a spin chain with randomly distributed impurity-chain couplings

Now consider the situation with the finite concentration of $S' = 1/2$ impurities, more realistic in NMR experiments. Suppose the impurity-host couplings (or θ_j) are distributed randomly. In such a case the model permits to integrate the expression for T_1^{-1} (in which we replace $\pi J/2 \rightarrow T_{K,j}$) over $T_{K,j}$ with the probability distribution of effective Kondo temperatures $P(T_{K,j})$ (or, equivalently, over θ_j with the probability distribution of the impurity-host coupling constants). For the weak disorder, i.e., if $P(T_{K,j})$ is a narrow function (e.g., for a narrow Gaussian distribution), the static properties are similar to the ones of the chain with a single impurity,⁴ which agrees with the results of perturbative calculations for a spin chain with the finite concentration of magnetic impu-

rities with the random distribution of impurity-host interactions¹² and experiments on spin-chain materials with the finite concentration of magnetic impurities.¹⁰ We obtain that dynamical characteristics of the inhomogeneous spin chain behave qualitatively similar to those of a single impurity, studied above. More interesting situation takes place if $P(T_{K,j})$ is wide (a strong disorder). Here for some impurities $T_{K,j} < T$, and, therefore, these impurities are not screened by spinons. It produces the divergency of the low T (or low H at $T=0$) of the magnetic susceptibility for the inhomogeneous chain. For example, such a situation takes place for $P(T_{K,j}) \sim (T_{K,j}/G)^\lambda / T_{K,j}$, which takes place in many experiments on spin-chain systems with randomly distributed magnetic impurities,¹³ valid till the maximal value of $T_{K,j}$, $G \sim \pi J/2$ (the value of the exponent $\lambda < 1$ is concentration dependent¹⁴). In this case for $H=0$ we have $\chi(T) \sim (T/G)^\lambda / T$, see Ref. 15, i.e., divergent low-temperature static susceptibility. It is easy to estimate the influence of the strong disorder in the distribution of the couplings between magnetic impurities and the host chain on the components of the DSF, neglecting marginal corrections. So, for the considered components of the DSF and the NMR linewidth T_1^{-1} in the absence of the external magnetic field, at $H=0$, become

$$T_1^{-1} \sim S_h^{zz} \sim S_s^{zz} \sim S_s^{xx} \sim S_h^{xx} \sim \frac{T^{\lambda-1}}{G^\lambda}, \quad (13)$$

i.e., instead of the growth with the temperature for the homogeneous longitudinal component and temperature-independent behavior for the “staggered” part of the longitudinal DSF, staggered and “homogeneous” parts for the transverse component of the DSF for the homogeneous chain, the strong disorder produces weak divergencies of those components of the DSF at low temperatures. Similar divergencies $T_1^{-1} \sim S^{\mu\nu} \sim H^{\lambda-1}$ are in the ground state for $H \rightarrow 0$. For the marginal case $\lambda=1$ we have the logarithmic divergencies of the static magnetic susceptibility¹⁵ $\chi \sim (1/G) \ln(a'J/T)$ for low temperatures, where a' is a non-universal constant. In this case the homogeneous longitudinal and homogeneous transverse components of the DSF (and, thus, the NMR relaxation rate $1/T_1$) become temperature independent due to this kind of disorder,

$$T_1^{-1} \sim S_h^{xx} \sim S_h^{zz} \sim \text{const.}, \quad (14)$$

while the staggered longitudinal and staggered transverse components of the DSF and the NMR relaxation rate become logarithmically divergent

$$T_1^{-1} \sim S_s^{zz} \sim S_s^{xx} \sim \ln(a'J/T). \quad (15)$$

An external magnetic field at $T \neq 0$ lifts the degeneracy, and, therefore, removes the divergencies for the static and dy-

namical characteristics of the spin chain with the strong disorder in the distribution of the impurity-chain couplings. Marginal perturbation, which produces logarithmic corrections to the behavior of the homogeneous chain, yields sub-leading (mostly logarithmic) corrections to the DSF for the chain with random impurities, cf. Ref. 12.

B. Finite concentration of similar magnetic impurities

For the finite concentration of the magnetic impurities which have the same values of S'_j and/or θ_j , the situation is different from the case with a single impurity or with the random distribution of couplings between impurities and the host chain. This case was analyzed, e.g., in Refs. 1 and 16. Here impurities' levels are organized in impurities' bands. Those bands produce additional Fermi seas, the van Hove singularities of which yield additional quantum phase transitions, governed by the external magnetic field. Continua of low-energy excitations for each additional band contribute to the low-temperature DSF in a similar way, as spinons contribute to the DSF of the homogeneous chain. The description of the behavior of critical exponents in this situation is complicated because the exponents for each band are not independent.

V. CONCLUSIONS

In summary, using nonperturbative methods in the framework of the exactly solvable model, we have found the temperature and magnetic field behavior of the NMR relaxation rate and related to it low-energy dynamical structure factor for the antiferromagnetic Heisenberg spin-1/2 chain with magnetic impurities. Our calculations show that for single magnetic impurities in the most interesting case of totally screened impurity at low temperatures, the critical exponents, which determine the low- T and $-H$ behavior, are renormalized due to the presence of the impurity. In the case of the finite concentration of impurities, for the random distribution of the impurity-chain couplings, we have found that the weak disorder does not produce essential changes to the dynamical structure factor. On the other hand, the strong disorder yields drastically different behavior of the low-energy dynamical characteristics, comparing to the homogeneous case. Our results are generic, they qualitatively agree with the results of perturbative RG calculations for disordered spin chains¹² and with the data of experiments¹³ on quasi-one-dimensional spin organic and nonorganic compounds.

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