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Effect of Intersite Repulsion on the Correlation Functions and Thermodynamics of an Ising Chain with Annealed Magnetic Disorder

V. V. Val'kov[a, *] and M. S. Shustin[a, b, **]

[a]*Kirensky Institute of Physics, Krasnoyarsk Scientific Center, Siberian Branch, Russian Academy of Sciences, Krasnoyarsk, 660036 Russia*

[b]*Siberian Federal University, Krasnoyarsk, 660041 Russia*

**e-mail: vvv@iph.krasn.ru*

***e-mail: mshustin@yandex.ru*

Abstract—An exact solution for a model describing the equilibrium behavior of an ensemble of Ising chains with nonmagnetic intersite repulsion of nearest neighbors and an equilibrium distribution of nonmagnetic impurities is obtained using the transfer matrix technique. The possibility of exciting quantum phase transitions using the intersite repulsion parameter in a system is demonstrated. Proximity to the critical points of these transitions has a substantial effect on the temperature dependence of a system's magnetic susceptibility.

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INTRODUCTION

Low-dimensional magnetic strucutres, including strongly anisotropic organic single-chain magnets (SCMs), have in recent years been widely synthesized [2, 3]. At low temperatures, these compounds are characterized by slow magnetization dynamics and are considered promising in the development of hardware components for memory devices. In experimental studies of these materials, much information about magnetic interaction is obtained by measuring the temperature dependence of static magnetic susceptibility. The effective low-energy model for describing the magnetic properties of such compounds is the 1D Ising model and its generalizations. Rexcently synthesized SCMs [2, 3] can change their magnetic state under external irradiation. The photoinduced change in the states of magnetic centers in such compounds is attributed to a similar change in the states of their electronic subsystems. Since the characteristic times of the dynamics of the magnetic subsytems in SCMs are usually comparable to or much larger than those of their electronic subsystems, it is generally agreed that magnetic ions in the photoinduced states result in an annealed type of magnetic disorder in a system [4]. It is therefore important to study the effect magnetic disorder and intersite correlations have on the temperature dependence of the magnetic susceptibility of SCMs. In this work, we solve this problem using a model that describes an equilibrium ensemble of Ising chains with annealed magnetic vacancies and intersite repulsion of the nearest magnetic sites.

MODEL AND EXACT SOLUTION

Let us consider an equilibrium ensemble of magnetic chains with spin $S = 1/2$ and number N of sites that can contain nonmagnetic impurities. We assume that the characteristic times of the dynamics of the initial magnetic and impurity subsystems are comparable. We may then include the variables of the impurity subsystem in the phase space of the initial subsystem. There are thus three one-site states in a chain: two states $|\sigma\rangle$ corresponding to the magnetic states of a site with spin projection $\sigma \pm 1/2$ onto the quantization axis and one state $|0\rangle$ corresponding to the localization of a nonmagnetic impurity or a vacancy on a site. In addition, we assume that both Ising exchange coupling and nonmagnetic intersite repulsion occur between nearest sites in the magnetic states. Physically, nonmagnetic intersite repulsion can occur via electrostatic or Van der Waals repulsion of the magnetic centers of a chain or enhancement of the system energy due to repulsion of the electron pairs of valence molecular orbitals. The role of vacancies can be played by magnetic ions in the low-spin state ($S = 0$). The Hamiltonian of the system is thus written in the form

$$H = J\sum_{f=1}^{N} S_f^z S_{f+1}^z + V\sum_{f=1}^{N} n_f n_{f+1} - h\sum_{f=1}^{N} S_f^z - \mu\sum_{f=1}^{N} n_f, \quad (1)$$

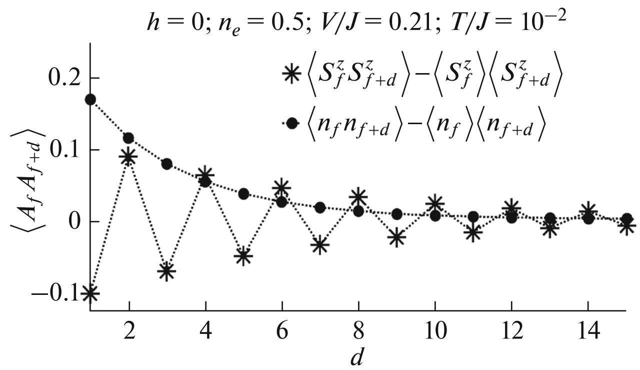

Fig. 1. Dependences of magnetic correlators $\langle S_f^z S_{f+d}^z\rangle$ and $\langle n_f n_{f+d}\rangle$ on the distance between chain sites in the proximity of quantum critical point $V \to V_c = J/4 - h/2$ corresponding to the state with short-range magnetic order and no long-range order.

where $J > 0$ and $V > 0$ are the intensities of Ising exchange and intersite repulsion, respectively; h is the external magnetic field in units of energy; and μ is an indefinite Lagrange multiplier. Operators S_f^z and n_f are the pseudo-spin operator and operator of the number of magnetic particles on the fth site of a chain, respectively. In the basis of one-site states of a chain, these operators have the form $S^z = 0.5\mathrm{diag}(0, 1, -1)$ and $n = \mathrm{diag}(0, 1, 1)$.

We use the transfer matrix technique [5] to study the equilibrium properties of model (1). In contrast to other ways of studying magnetic systems with the annealed disorder [4, 6], our approach based on the transfer matrix technique allows us to calculate both average values and pair correlators easily and accurately, based on observed one-site states $A_f^{(\nu)}$ of a system diagonal in space:

$$\left\langle A_f^{(\nu)}\right\rangle_N = \frac{1}{\Xi}\sum_{\alpha=1}^{N}\langle u_\alpha|A^{(\nu)}|u_\alpha\rangle\lambda_\alpha^N;$$
$$\left\langle A_f^{(\nu_1)}A_{f+d}^{(\nu_2)}\right\rangle_N = \frac{1}{\Xi}\sum_{\alpha_1,\alpha_2=1}^{N} A_{\alpha_1\alpha_2}^{(\nu_1)}A_{\alpha_2\alpha_1}^{(\nu_2)}\lambda_{\alpha_1}^{N-d}\lambda_{\alpha_2}^{d}. \quad (2)$$

Index ν denotes the type of one-site operator; λ_α and $|u_\alpha\rangle$ are the eigenvalues and eigenvectors of the transfer matrix, respectively; and $A_{\alpha\beta}^{(\nu)} = \langle u_\alpha|A^{(\nu)}|u_\beta\rangle$. When we are interested in the properties of the model at specified concentrations of vacancies $\langle n\rangle$ or particles with spin $1 - \langle n\rangle$ (denoted below as n_h and n_e, respectively), means must be calculated using formula (2) and preliminarily solving the equation for indefinite Lagrange multiplier $\langle n\rangle(\mu) = n_e$.

QUANTUM PHASE TRANSITION AND MAGNETIC SUSCEPTIBILITY MODIFICATION

Let us consider the effect intersite repulsion has on the magnetic properties of model (1) at fixed concentration n_e of magnetic centers. Without losing the generality of subsequent results, for the sake of simplicity we consider a semioccupied chain ($n_e = 0.5$). When $h \ll J$, the chain can be in two basic states: one with microscopic separation in phases from antiferromagnetically ordered spin subchains and vacancy subchains, and one with alternating paramagnetic centers and vacancies. The energies of these states are $E_1 \approx Nn_e(V - J/4)$ and $E_2 \approx -Nn_h h/2$, respectively. It can be seen that at zero temperature and $V < V_c = J/4 - h/2$, the phase with the antiferromagnetic subchains is observed; when $V > V_c$, the phase with paramagnetic centers emerges. We shall refer to the transition between these two phases as a quantum phase transition (QPT), assuming that the classical Ising model (1) is the limiting case of the quantum anisotropic Heisenberg model in [2]. We refer to point V_c of this transition as a critical quantum point (CQP). The QPT's existence is confirmed by the calculated dependence of correlation functions $K_S(d) = \left\langle S_f^z S_{f+d}^z\right\rangle$ and $K_n(d) = \langle n_f n_{f+d}\rangle$ on distance d between chain sites. It is known that when $T = 10^{-5}J$ and $V = V_c - \delta$ ($\delta \to 0$), the correlator is $K_S = 0.5S^2$ ($K_n = 0.5$) at even d values and $K_S = -0.5S^2$ ($K_n = 0.5$) at odd d values. When $V = V_c + \delta$, spin correlator K_S is identically zero and K_n acquires values of 0.5 and 0 for even and odd d values, respectively. At finite temperatures, the QPT boundary is spread and the intermediate phase is observed in the proximity of the CQP. Features of this phase are illustrated in Fig. 1 by correlators $K_S(d)$ and $K_n(d)$. It can be seen that this phase is characterized by short-range order but no long-range order.

The QPT determined by the intersite repulsion parameter substantially affects the low-temperature portion of the temperature dependence of the system's magnetic susceptibility $\chi(T)$. This is due to the qualitatively different behaviors of the $\chi(T)$ dependences for the magnetic phases described above: the state at $V < V_c$ is characterized by activation growth in the low-temperature dependence of susceptibility $\chi(T) \sim e^{-\Delta/T}$ typical of an ensemble of antiferromagnetic Ising chains, while the state at $V > V_c$ is a set of paramagnetic centers and the temperature dependence of magnetic susceptibility is described by Curie–Weiss law $\chi(T) \sim C/T$ ($T_N = 0$). A crossover between these two dependences is observed upon variation in V (Fig. 2). It can be seen that minor variation

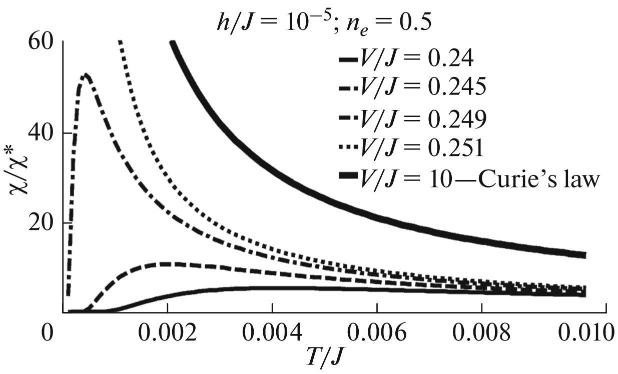

Fig. 2. Evolution of the low-temperature dependence of magnetic susceptibility upon variation in intersite repulsion parameter V/J in the proximity of the quantum critical point of system $V \to V_c = J/4 - h/2$. A crossover from the activation growth law ($V < V_c$) to the Curie–Weiss law ($V > V_c$) is observed.

in parameter V/J near CQP V_c can strongly alter the characteristic peak in dependence $\chi(T)$.

CONCLUSIONS

A model describing the equilibrium behavior of an ensemble of Ising chains with magnetic vacancies and nonmagnetic intersite repulsion between nearest magnetic centers was created. The model qualitatively describes the low-temperature magnetic properties of anisotropic single-chain organic magnets exposed to optical radiation. Using this model's exact solution according to the transfer matrix technique, it was shown that a quantum phase transition can occur in such systems according to the intersite repulsion parameter. The low-temperature portion of the temperature dependence of a system's magnetic susceptibility is substantially altered after passing through the critical quantum point of this transition and crosses over from activation ascendance law $\chi(T) \sim e^{-\Delta/T}$ to Curie–Weiss law $\chi(T) \sim C/T$. The phase with short-range magnetic and no long-range order is observed in the proximity of the critical quantum point. This information can be used to interpret experimental data on the low-temperature behavior of single-chain magnets upon their optical irradiation.

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REFERENCES

1. Prosorova, L.A., Sosin, S.S., Svistov, L.E., et al., *Phys. Rev. B*, 2015, vol. 91, p. 174410.
2. Zhang, W.X., Ishikawa, R., Breedlove, B., and Yamashita, M., *RSC Adv.*, 2013, vol. 3, p. 3772.
3. Liu, T., Zheng, H., Kang, S., et al., *Nat. Commun.*, 2013, vol. 4, p. 2826.
4. Arzhnikov, A.K. and Vedyaev, A.V., *Fiz. Nizk. Temp.*, 1982, vol. 8, p. 1186.
5. Bernot, K., Luzon, J., Caneschi, A., et al., *Phys. Rev. B*, 2009, vol. 79, p. 134419.
6. Lushnikov, A.A., *J. Exp. Theor. Phys.*, 1969, vol. 29, p. 120.

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