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Low Temperature Spectral Properties of 1D Magnets with Twisting and Mutually Orthogonal Arrangement of Easy Planes

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Abstract—The exitation spectrum of a ferrimagnetic Heisenberg chain with four sublattices and twisting mutually ortoghonal easy planes is calculated using a diagram technique of atomic representation. It is shown that at low temperatures, the exitation spectrum of elementary excitations of the system is similar to the spectrum of a magnet with an effective easy-axis type anisotropy directed along the line of easy plane intersection.

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INTRODUCTION

A quasi–one-dimensional magnet, *catena*-$[Fe^{II}(ClO_4)_2\{Fe^{III}(bpca)_2\}](ClO_4)$ (below, *SCM-catena*) with twisted low-spin ($S = 1/2$) and high-spin ($S = 2$) states of magnetic iron ions was recently synthesized. The magnetic states of high-spin ions are formed with the participation of strong single-ion anisotropy of the easy-plane type [1] in an orientation that changes during transitions between high-spin iron ions (Fig. 1). At the same time, experimental investigations [2] show that this compound displays properties typical of magnets with the easy-axis type anisotropy. It therefore seems important to determine the reasons for this discrepancy. In this work, the problem is solved via microscopic calculations of the excitation spectrum of an anisotropic four-sublattice *SCM-catena* and establishing its correspondence with the excitation spectrum of an easy-axis ferrimagnet.

AN *SCM-catena* HAMILTONIAN

The magnetic properties of the single-chain *SCM-catena* magnet, the magnetic structure of which is shown in Fig. 1, is described using the model of a ferrimagnetic Heisenberg chain with single-ion anisotropy of the easy-plane type:

$$\begin{aligned} H_G &= J\sum_f\left[\vec{S}_{f,A}\vec{S}_{f,B} + \vec{S}_{f,B}\vec{S}_{f,C} + \vec{S}_{f,C}\vec{S}_{f,D} + \vec{S}_{f,D}\vec{S}_{f+1,A}\right] \\ &+ 2D\sum_f\left[\left(S^x_{f,A}\right)^2 + \left(\left(S^y_{f,C}\right)^2\right)\right] \\ &- h\left(g_1\left(S^z_{f,A} + S^z_{f,C}\right) + g_2\left(S^z_{f,B} + S^z_{f,D}\right)\right), \end{aligned} \quad (1)$$

where $\vec{S}_{f,A}$ and $\vec{S}_{f,C}$ are vector operators of the spin moment of iron ions in high-spin (*HS*) states with spin $S = 2$ belonging to magnetic cell f (the cell contains four magnetic ions) and residing in positions A and C. These ions are affected by single-ion anisotropy of the easy-plane type. The anisotropy is determined by parameter D. It is important that the orientation of the easy magnetization planes changes in transitioning from one *HS* iron ion to another. For iron ions in positions A, the plane of easy magnetization is the $Y0Z$ plane; for iron ions in positions C, it is the $X0Z$ plane. $\vec{S}_{f,B}$ and $\vec{S}_{f,D}$ are the vector operators of spin moments in low-spin (*LS*) states with spin $S = 1/2$ belonging to magnetic cell f and residing in positions B and D; h is the external magnetic field in energy units; g_1 and g_2 are the g-factors for *HS* and *LS* ions, respectively; and J is the exchange integral between the nearest ions. From the experiment in [2], it is known that $J \approx 20$ K and $D \approx 7$ K.

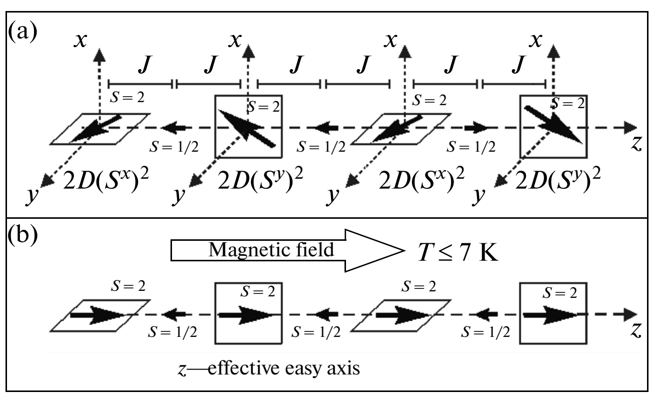

Fig. 1. (a) Mutual orientation of the easy-magnetization planes in *SCM-catena*; (b) Short-range order in a four-sublattice ferrimagnet at $T < 7$ K. The assumed sequence of sublattices in the unit cell is $A–B–C–D$.

In calculating the low temperature excitation spectrum, we consider that at $T \leq 7$ K short range ferrimagnetic ordering has been observed in the *SCM-catena* in experiments (Fig. 1b) [2]. We assume that the spontaneous magnetization of all ions is oriented along z axis. Allowing for the effects of a self-consistent field, the Hamiltonian of the system takes the form

$$H_G = \sum_{f\in A} H_{0,A}(f) + \sum_{g\in B} H_{0,B}(g) + \sum_{k\in C} H_{0,C}(k) + \sum_{l\in D} H_{0,D}(l) + H_{\text{corr}}, \quad (2)$$

where single-ion operators for the introduced four sublattices have the form

$$H_{0,A}(f) = D\left(S_f^x\right)^2 - \bar{h}_1 g_1 S_f^z; \quad H_{0,B}(g) = -\bar{h}_2 g_2 S_g^z;$$
$$H_{0,C}(k) = D\left(S_k^y\right)^2 - \bar{h}_1 g_1 S_k^z; \quad H_{0,D}(l) = -\bar{h}_2 g_2 S_l^z; \quad (3)$$
$$\bar{h}_1 = h + 2J\sigma; \quad \bar{h}_2 = h - 2J\tilde{S}.$$

The following designations were introduced in writing Eq. (3) for the magnetic ordering shown in Fig. 1: $\tilde{S} = \langle S_{f,A}^z \rangle = \langle S_{f,C}^z \rangle$ and $\sigma = -\langle S_{f,B}^z \rangle = -\langle S_{f,D}^z \rangle$.

If we add circular spin operators and the three-component operator $\vec{u} = \{S^z, S^+, S^-\}$, summand H_{corr} in (2) can be written in the form

$$H_{\text{corr}} = J\sum_{\langle fg\rangle}(\Delta\vec{u}_f, V\Delta\vec{u}_g) + J\sum_{\langle gk\rangle}(\Delta\vec{u}_g, V\Delta\vec{u}_k) + J\sum_{\langle kl\rangle}(\Delta\vec{u}_k, V\Delta\vec{u}_l) + J\sum_{\langle lf\rangle}(\Delta\vec{u}_l, V\Delta\vec{u}_f), \quad (4)$$

where $\Delta\vec{u} = \vec{u} - \langle\vec{u}\rangle$, $f \in A$, $g \in B$, $k \in C$, and $l \in D$. Matrix V for isotropic exchange interaction between the nearest magnetic ions has the components

$$V = [1,\ 0,\ 0;\ 0,\ 0,\ 1/2;\ 0,\ 1/2,\ 0].$$

We use the ideology of atomic representation to calculate the excitation spectra; this allows us to consider strong single-ion anisotropy correctly [3]. The atomic representation assumes a diagonal Hamiltonian $H_{0,A}(f)$. This problem can be easily solved via the unitary transformation of group $U(N)$ [4]. We then get a system of self-consistent equations for finding the transformation parameters and effective fields:

$$\tilde{S}(\sigma) = 2\cos^2\beta\cos 2\alpha; \quad \sigma(\tilde{S}) = \frac{1}{2}\tanh\left(\frac{\bar{h}_2}{2T}\right). \quad (5)$$

$$2\bar{h}_1 \sin 2\beta\cos\beta + \sqrt{6}D(\cos\alpha - \sin\alpha)\sin\beta = 0,$$
$$(2D + \bar{h}_1\cos 2\alpha)\sin 2\beta + \sqrt{6}D(\cos\alpha + \sin\alpha)\cos 2\beta = 0. \quad (6)$$

ELEMENTARY EXCITATION SPECTRUM

Let us introduce Green's functions in atomic representation $D_{nm;pq}(fi\tau; f'j\tau') = -\langle T_\tau \tilde{X}_{fi}^{nm}(\tau)\tilde{X}_{f'j}^{pq}(\tau')\rangle$ built upon Hubbard operators $X_{fi}^{nm} = |\Psi_{fi}^n\rangle\langle\Psi_{fi}^m|$. Here i and j are the indices of sublattices and $|\Psi_{fi}^n\rangle$ functions are determined by solving the Schrödinger equation for the single-ion problem for every ion.

The derivation of the Larkin equation, which allows us to obtain the dispersion relation for anisotropic magnets, was described in detail in [3, 4]. The solving of a large number of linear equations is based on the use of matrix elements in atomic representation, split by the indices of the root vectors. Extending this approach to the case of four sublattices, we find that the elementary excitation spectrum of *SCM-catena* is provided by the equation

$$\Delta(q,\omega) = \det\left\|\left[\hat{U}(\omega),\ \hat{0},\ \hat{0};\ \hat{0},\ \hat{\Phi}(\omega),\ \hat{W}(\omega);\ \hat{0}, \hat{W}(\omega), \hat{\Phi}(-\omega)\right]\right\| = 0, \quad (7)$$

where $\hat{0}$ is a three-dimensional zero matrix. Matrices $\hat{U}(\omega)$ and $\hat{W}(\omega)$ have the form

$$\hat{U}(\omega) = J\begin{pmatrix} -1 & u_A(\omega) & 0 & u_A(\omega)e^{-4iq} \\ u_B(\omega) & -1 & u_B(\omega) & 0 \\ 0 & u_A(\omega) & -1 & u_A(\omega) \\ u_B(\omega)e^{4iq} & 0 & u_B(\omega) & -1 \end{pmatrix};$$

$$\hat{W}(\omega) = \frac{J}{2}\begin{pmatrix} 0 & w(\omega) & 0 & w(\omega)e^{-4iq} \\ 0 & 0 & 0 & 0 \\ 0 & -w(\omega) & 0 & -w(\omega) \\ 0 & 0 & 0 & 0 \end{pmatrix},$$

$$u_{A,B}(\omega) = \sum_{\alpha\in A,B} |\gamma_{\parallel}(\alpha)|^2 (\omega + \vec{\alpha}\vec{E})^{-1} b(\vec{\alpha}),$$
$$z_{A,B}(\omega) = \sum_{\alpha\in A,B} |\gamma_{\perp}(\alpha)|^2 (\omega + \vec{\alpha}\vec{E})^{-1} b(\vec{\alpha}); \quad (8)$$
$$w(\omega) = \sum_{\alpha\in A} \gamma_{\perp}(\alpha)\gamma_{\perp}(-\alpha)\left(\omega + \vec{\alpha}\vec{E}^{(HS)}\right)^{-1} b(\vec{\alpha}),$$

where $\gamma_{\parallel}(\alpha)$ and $\gamma_{\perp}(\alpha)$ are, respectively. longitudinal and transverse parameters in the representation of spin operators through Hubbard operators. $\hat{U}(\omega)$ matrix determines the longitudinal branches of the elementary excitations of the magnet under consideration.

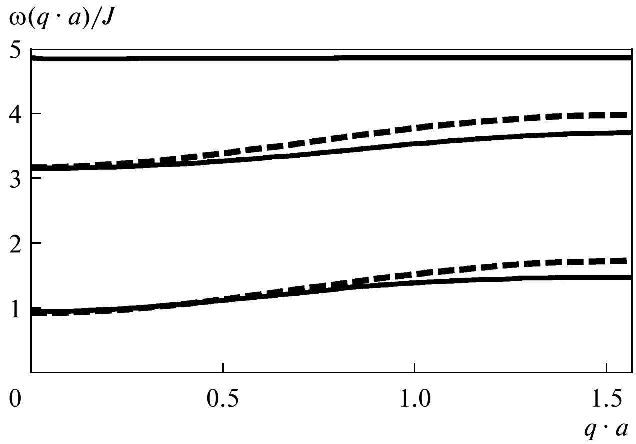

Fig. 2. Low temperature spectrum of elementary excitations in a four-sublattice *SCM-catena* ferrimagnet at $D = J/3$ (solid lines). Dotted lines show the excitation spectrum obtained for a ferrimagnet in which the anisotropy characterized by twisting orientations of easy-magnetization planes is replaced with an effective light axis anisotropy with $D_{ef} = -J/8$.

Matrix $\hat{\Phi}(\omega)$ responsible, along with matrix $\hat{\Phi}(-\omega)$, for the transverse branches of the elementary excitation spectrum, can be obtained from matrix $\hat{U}(\omega)$ via substitution: $J \to J/2$ and $u_{A,B}(\omega) \to z_{A,B}(\omega)$.

Results from the numerical solution to Eq. (7) at $T \ll J$ are shown in Fig. 2. Solid lines are the dependences on the quasi-momentum of the *SCM-catena* excitation spectrum branches. Dotted lines represent the dependences on the quasi-momentum of the spectrum branches for an effective model of a ferrimagnetic Heisenberg chain with single-ion anisotropy of the easy-axis type. It was assumed that $D \to D_{ef} = -J/8$ and the operator expressions $D\left(S_f^x\right)^2$ and $D\left(S_k^y\right)^2$ were replaced with $D_{ef}\left(S_f^z\right)^2$ and $D_{ef}\left(S_k^z\right)^2$. A comparison of these dependences shows that in the low temperature region, *SCM-catena* actually exhibits properties characteristic of easy-axis ferrimagnets.

CONCLUSIONS

Our results show that at low temperatures, the excitation spectrum of *SCM-catena* corresponds to the spectrum of a 1D ferrimagnet with an effective easy magnetization axis directed along the chain axis. In both models, the excitation spectrum had a characteristic gap of width $\Delta \sim J$ and a low (relative to Δ) dispersion of the main excitation branches. This means that on a qualitative level, the energy structure of a one-dimensional four-sublattice ferrimagnet with twisting orientations of the easy-magnetization planes of high-spin iron ions coincides with the single-particle excitation spectrum of a ferrimagnetic Ising chain for which $\Delta = 2JS_1S_2$ and there is no dispersion of branches. This allows us to investigate the thermodynamic properties of *SCM-catena* over a wide range of temperatures using precise calculations of the statistical sum for the one-dimensional Ising model with several sublattices via the transfer matrix method.

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REFERENCES

1. Kajiwara, T., Nakano, M., Kaneko, Y., Takaishi, S., Ito, T., Yamashita, M., Igashira-Kamiyama, A., Nojiri, H., Ono, Y., and Kojim, N., *J. Am. Chem. Soc.*, 2005, vol. 127, p. 10150.
2. Kajiwara, T., Tanaka, H., and Yamashita, M., *Pure Appl. Chem.,* 2008, vol. 80, p. 2297.
3. Val'kov, V.V., Val'kova, T.A., and Ovchinnikov, S.G., *J. Exp. Theor. Phys.*, 1985, vol. 61, p. 323.
4. Val'kov, V.V., *Theor. Math. Phys.*, 1988, vol. 76, p. 766.

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