Spontaneous spin-nematic ordering in a spin-chain system

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The spontaneous onset of the spin-nematic order in spin-chain systems is studied. The ordering persists due to distortions of either magnetic ions, or ligands taking part in the superexchange between magnetic ions. The spin-nematic order yields a nonzero biaxial magnetic anisotropy for the spin chain, violating the U(1) symmetry. The influence of the external magnetic field and nonzero temperature is considered. We predict how the spin-nematic ordering can manifest itself in the temperature and magnetic field behavior of magnetoacoustic characteristics.

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I. INTRODUCTION

Correlations between electrons in condensed matter physics can change the behavior of electrons from the conventional metallic one to liquid states. The examples of the latter are electron liquid crystal states in metals, valence bond solids, or spin liquid states in insulating quantum spin systems [1]. For instance, the standard magnetic order (i.e., the ordering of magnetic dipoles) in quantum spin liquids is suppressed down to the lowest temperatures due to the frustration of spin-spin interactions and/or enhanced quantum fluctuations in low-dimensional systems [2]. While magnetic (dipole) moments remain disordered, higher-rank multipoles can order under special conditions. Unlike the order of magnetic dipoles, e.g., the quadrupolar order does not break the time-reversal symmetry. Quadrupolar ordering in insulating spin systems is often referred to as the spin-nematic one, because that type of ordering is analogous to the known ordering of molecules in nematic phases of liquid crystals [3]. For orbital electron moments the dipole moment of the orbital electron moment is frozen in most of compounds: The orbital order manifests itself first of all in the charge ordering or in the ordering of multipolar moments. The spin-nematic order and the orbital/charge order breaks the rotational symmetry of electron states. Strong nematic fluctuations were observed, e.g., in Fe- and Cu-based superconductors [4] (nematicity is believed to be the essential property for Fe-based superconductors [5]), as well as in strontium ruthenates [6], and high fractional Landau levels [7]. Dipole moments are coupled to the electromagnetic field, while there is no simple field directly coupled to quadrupoles. Hence, the quadrupolar hidden order is frequently difficult to detect in experiments because most of the available techniques are sensitive to dipole moments only. However, some manifestations of the quadrupolar order were observed in several compounds, e.g., in the layered system $NiGa_2S_4$, in CeB_6 and UPd_3, and the high magnetic field-induced spin-nematic order in the spin-chain compound $LiCuVO_4$ [8]. Critical spin-chain systems are almost ideal candidates for the search of the spin-nematic ordering, because magnetic order is suppressed there by quantum and thermal fluctuations [9].

In this paper we propose a different approach for the onset of the spin-nematic order in spin-chain systems, not related to the application of the external magnetic field as in $LiCuVO_4$ [10], i.e., the spontaneous onset of the spin nematicity. We show that due to the coupling of spins to the lattice, the spin-nematic order can appear in quasi-one-dimensional spin systems, even at zero field. Unlike previous approaches, which used approximate, e.g., spin-wave-like, theories to describe the spin subsystem of spin-chain materials [10], our work takes into account spin-spin interactions in spin chains exactly. We also show how magnetoacoustic studies can serve as a good method for the investigation of the spin-nematic ordering in spin-chain systems.

II. SPONTANEOUS SPIN-NEMATIC ORDERING

Let us study the spin-chain system, which is described by the Hamiltonian

$$\mathcal{H}_0 = \sum_n \left[J\left(S_n^x S_{n+1}^x + S_n^y S_{n+1}^y\right) + J_z S_n^z S_{n+1}^z \right], \qquad (1)$$

where $S_n^{x,y,z}$ are the operators of projections of the spin-1/2 situated at the site n, and J, J_z are the values of the exchange coupling between neighboring spins. Notice that for spin-1/2 systems one cannot speak about the single-ion quadrupolar moments. Our goal is to study the onset of the spin-nematic order. Let us limit ourselves with the homogeneous case. Then the spin-nematic order can be characterized by the nonzero value of the operator of the quadrupolar moment at neighboring sites $\langle S_n^x S_{n+1}^x - S_n^y S_{n+1}^y \rangle$. It obviously violates the U(1) symmetry of the spin chain, however, it does not break the time-reversal symmetry. It is easy to show that for the Hamiltonian $\mathcal{H}_0$ such a value is zero for any temperatures $T \geqslant 0$ (in our work we use the energy units in which the Boltzmann constant is unity, $k_B = 1$ for simplicity). Then, let us consider a more general Hamiltonian with the biaxial

magnetic anisotropy

$$\mathcal{H} = \sum_n \left[J_x S_n^x S_{n+1}^x + J_y S_n^y S_{n+1}^y + J_z S_n^z S_{n+1}^z \right], \qquad (2)$$

with $J_x = J(1+x)$, and $J_y = J(1-x)$, where $0 \leqslant x \leqslant 1$. The case $J_z > 0$ corresponds to the antiferromagnetic chain (notice that the sign of J is nonessential in thermodynamics [11]), while $J_z < 0$ describes the ferromagnetic chain. For $-J \leqslant J_z \leqslant J$ the system is critical (with gapless low-energy excitations), and for $J_z < -J$ and $J_z > J$ the system is in the Heisenberg-Ising phase with gapped excitations. First we consider $J \geqslant 0$ for concreteness; a similar approach exists for the ferromagnetic chain. The quadrupolar (spin-nematic) order parameter is nonzero for the system with the Hamiltonian $\mathcal{H}$ for $x \neq 0$ for any $T \geqslant 0$. Hence, the onset of the nonzero parameter x describes the onset of the homogeneous spin-nematic order with nonzero quadrupolar order parameter $\langle S_n^x S_{n+1}^x - S_n^y S_{n+1}^y \rangle$.

Consider the three-dimensional crystal with spin-1/2 magnetic ions, situated at sites of the crystal lattice, with the exchange coupling between spins along one distinguished direction being much stronger than along other directions. Suppose that the one-dimensional spin subsystem of the considered system is described by the Hamiltonian $\mathcal{H}$. The exchange coupling is determined by the effective overlap of the wave functions of neighboring magnetic ions [11]. For most of spin-chain compounds the exchange is indirect (the superexchange) [11], and due to the spin-orbit interaction those overlaps can be anisotropic [12]. The anisotropy of overlaps depends on the relative positions of neighboring magnetic ions, and also on the positions of nonmagnetic ions (ligands) via which the superexchange coupling is realized [13]. Hence, to get the nonzero biaxial magnetic anisotropy $J_x \neq J_y$, i.e., $x \neq 0$, one needs to change the positions of either neighboring magnetic ions, coupled by the superexchange interactions, or the positions of the surrounding ligands. Those shifts are characterized by the distortions u_{xy} (namely such distortions yield $J_x \neq J_y$). Then the loss of the energy of the system due to such distortions can be written in the main approximation in u_{xy} as $Cu_{xy}^2/2$, where C denotes the elastic modulus. For temperatures smaller than the Debye one we can limit ourselves with that term only for the lattice subsystem. On the other hand, the magnetic subsystem of the crystal can gain the energy due to those distortions, because of the onset of the biaxial anisotropy of the exchange couplings, and, hence, the spin-nematic order with the nonzero quadrupolar order parameter $\langle S_n^x S_{n+1}^x - S_n^y S_{n+1}^y \rangle$ appears. For small distortions we can approximate $J_x = J(1 + du_{xy})$ and $J_y = J(1 - du_{xy})$, where d characterizes the exchange-elastic coupling. We see that $x = du_{xy}$, and can rewrite the elastic term as $Cx^2/2d^2$.

III. GROUND STATE

Our aim is to show that for some conditions the situation with $x \neq 0$ can be realized. Our approach is analogous to Peierls' description of the dimerization of electron chains [14]. The energy of the ground state of the system with the Hamiltonian $\mathcal{H}$ (let us consider the most interesting case with $J_x \geqslant J_y > J_z \geqslant 0$, i.e., for $x = 0$ it corresponds to the easy-plane anisotropic critical antiferromagnetic chain with gapless excitations) can be exactly obtained by using the famous Baxter's solution [15] (notice that we interchange the indexes $x \leftrightarrow z$ comparing to Baxter's notations).

Two exchange constants can be expressed via the third one as $J_z = \mathrm{cn}(2\zeta, k) J_x$ and $J_y = \mathrm{dn}(2\zeta, k) J_x$. Here $\mathrm{cn}(z, k)$ and $\mathrm{dn}(z, k)$ [and $\mathrm{sn}(z, k)$, see below] are Jacobi elliptic functions of the argument z with the modulus k. The argument ζ and the modulus k determine the magnetic anisotropy of the system. The modulus k is equal to $k = [(J_x^2 - J_y^2)/(J_x^2 - J_z^2)]^{1/2} \equiv 2J[x/(J_x^2 - J_z^2)]^{1/2}$, i.e., nonzero values of x imply nonzero values of k (and vice versa).

The set of quantum numbers, rapidities, $\{x_\alpha\}_{\alpha=1}^{N/2}$, where N is the number of spins in the chain, parametrize all eigenfunctions and eigenstates of the Hamiltonian $\mathcal{H}$. They satisfy the Bethe ansatz equations [16,17]

$$\left[\frac{H[\zeta(x_\alpha + i)]}{H[\zeta(x_\alpha - i)]} \right]^N = -e^{2\pi\nu/G} \prod_{\beta=1}^{N/2} \frac{H[\zeta(x_\alpha - x_\beta + 2i)]}{H[\zeta(x_\alpha - x_\beta - 2i)]}, \qquad (3)$$

where $G = K_k'/\zeta$, $H(x) = \vartheta_1(x/2K_k'; iK_k/K_k')$ is the Jacobi function, with K_k and K_k' being the complete elliptic integrals of the first kind with the modules k and $k' = \sqrt{1-k^2}$, respectively. Notice that

$$\sum_{\alpha=1}^{N/2} \zeta x_\alpha = K_k' \nu' + iK_k \nu, \qquad (4)$$

where ν and ν' are integers. The eigenvalues of the Hamiltonian can be written as

$$E = -\frac{J_x \mathrm{sn}(2\zeta, k)}{2\zeta} \sum_{\alpha=1}^{N/2} \mathbf{a}_1(x_\alpha) - NJ_x R, \qquad (5)$$

where $\mathbf{a}_1(x) = (\zeta/\pi)\{Z(\zeta) + \mathrm{sn}(\zeta, k)\mathrm{cn}(\zeta, k)\mathrm{dn}(\zeta, k)/[\mathrm{sn}^2(\zeta, k) - \mathrm{sn}^2(i\zeta x, k)]\}$, where $Z(x)$ is Jacobi's zeta function with the modulus k, and $R = (1/8) - [\pi \mathrm{sn}(2\zeta, k)/8\zeta][\mathbf{a}_1(0) - \mathbf{a}_1(G)]$.

The ground state in the thermodynamic limit $N \to \infty$ (i.e., filling of all states with negative energies) corresponds to the set of real rapidities x_α distributed in the interval $[-G, G]$. The ground-state energy can be written as [15]

$$E_{gs} = -\frac{J_x}{8} - \frac{\pi J_x}{2K_k'} \mathrm{sn}(2\zeta, k) \sum_{n=1}^{\infty} X, \qquad (6)$$

where

$$X = \frac{\sinh^2[(\tau - \lambda)n] \tanh(\lambda n)}{\sinh(2\tau n)} \qquad (7)$$

and $\tau = \pi K_k/K_k'$, $\lambda = \pi\zeta/K_k'$ $(0 < \lambda < \tau)$. Now the condition of the equilibrium in the ground state implies that x is the solution of the equation

$$\frac{Cx}{d^2} = \frac{\partial E_{gs}}{\partial x}. \qquad (8)$$

Equation (8) is very complicated, and we can limit ourselves with the studying of the situation with small $x \neq 0$ (i.e., small k), which is the physically relevant case. For small k (i.e., $J_x \to J_y$) the ground-state energy can be rewritten

as [15]

$$E_{gs} = -\frac{\left(J_x^2 - J_z^2\right)^{1/4}\left(J_y^2 - Jz^2\right)^{1/4}}{4\sqrt{k'}K_k}\left(\frac{1}{4\sin\mu}\right.$$
$$+\int_{-\infty}^{\infty} dx \frac{\sinh[(\pi-\mu)x]\tanh(\mu x)}{\sinh(2\pi x)}$$
$$+2\sum_{n=1}^{\infty}\frac{q_k^{2n}}{1-q_k^{2n}}\sin^3(\mu n)\cos(\mu n)$$
$$\left.-\frac{2\pi}{\mu}\sum_{n=1}^{\infty}\frac{q_k^{(2n-1)\pi/\mu}}{1-q_k^{(2n-1)\pi/\mu}}\cot\left[\frac{(2n-1)\pi^2}{2\mu}\right]\right), \quad (9)$$

where $\mu = \pi\zeta/K_k$, and $q_k = \exp(-\pi K_k'/K_k)$. The main contribution to Eq. (8) comes from the singular term in the last line of Eq. (9). For k small we have $q_k \approx k^2/16$, and one gets for $0 < \mu < \pi$ (with $\mu \neq \pi/2m$ for m integer) [15]

$$E_{gs}^s = -J_x \sin\mu \frac{\pi}{\mu}\cot\left[\frac{\pi^2}{2\mu}\right]|Y|^{\pi/\mu}, \quad (10)$$

and for $\mu = \pi/2m$ one has

$$E_{gs}^s = -\frac{4mJ_x\sin\mu}{\pi}Y^{2m}\ln|Y|. \quad (11)$$

Here $\cos\mu = J_z/J_x$, and $Y = J_x(J_x - J_y)/8(J_x^2 - J_z^2)$. The analysis of Eq. (8) shows that for μ small (including the case $\mu = \pi/2m$), i.e., for the small anisotropy of the spin system with $\mathcal{H}_0$, the solution with $x \neq 0$ can exist only for small C/d^2J. On the other hand, for μ close to $\pi/2$, i.e., for large enough easy-plane anisotropy of $\mathcal{H}_0$, the solutions with $x \neq 0$ exist for a large interval of values of C/d^2J. We see that the fact of whether the spin-nematic ordering can exist or not, depends on the value of C/d^2J. The values of the elastic modules and exchange constants are known for many spin-chain materials, whilst the exchange-striction parameter d cannot be directly determined from experiments. However, the combination of parameters C/d^2J for realistic spin-chain materials can be in the necessary range of order of 1, which is demanded by the condition of the onset of the spin-nematic ordering there. For example, a similar combination of constants appears in theories describing the spontaneous onset of the spin-Peierls alternation in spin-chain systems [18]. Spin Peierls alternation exists, e.g., in nonorganic materials $CuGeO_3$ and NaV_2O_5 [19]. Figure 1 shows how the surface $a = (C/d^2J)x - (\partial E_{gs}^s)/(\partial x)$ as a function of C/Jd^2 and x crosses zero. One sees that together with $x = 0$, there exists nonzero solutions of Eq. (8). We can show that $E_{gs} + Cx^2/d^2$ is smaller for those nonzero values of x than for $x = 0$ for the case of large easy-plane anisotropy for C/d^2J small. Figure 2 manifests that behavior of the ground-state energy $E_0 = E_{gs} - Cx^2/2d^2$ as a function of x and C/d^2J. However, for larger values of the parameter C/d^2J, the energy of the state with $x = 0$ is the smallest one. It implies the onset of the spin-nematic order in such an easy-plane spin-chain system in the ground state for small C/d^2J. On the other hand, for an almost isotropic spin-chain system the ground-state energy is minimal for $x = 0$, i.e., here the spin-chain system is robust with respect to the biaxial spin-nematic ordering. A similar approach can be applied for the critical ferromagnetic

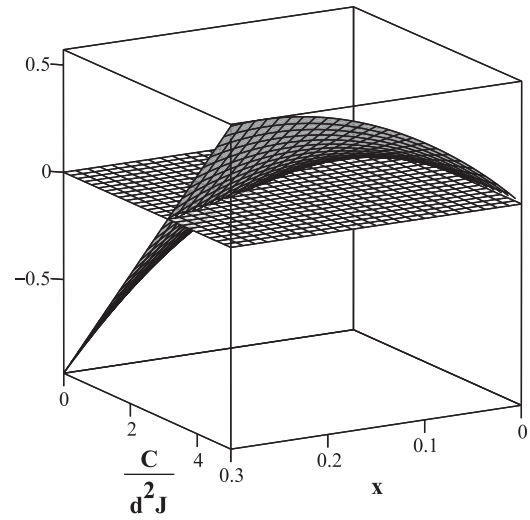

FIG. 1. The surface $a = (C/d^2J)x - (\partial E_{gs}^s/J\partial x)$ as a function of x and C/d^2J. The crossing of the surface with the zero plane defines possible values of x for the antiferromagnetic spin chain with the strong easy-plane anisotropy $\mu = \pi/2 - 0.3$.

spin chain [15]. The case with $J_z > J_y \to J_x > 0$, i.e., the Heisenberg-Ising antiferromagnetic spin chain can also be considered in a similar way [15]. That case is characterized by the ground-state energy being analytic function at $J_x = J_y$ (the same is true for the ferromagnetic easy-axis Heisenberg-Ising case, because of the periodicity of the Jacoby elliptic functions). This is why the onset of the spin-nematic phase is unlikely for those cases. The ground state of the Heisenberg-Ising spin chain is magnetically ordered, and, hence, it is problematic to see the quadrupolar order there.

IV. NONZERO TEMPERATURES

The energy of the system at nonzero temperatures can be described, e.g., by the string hypothesis [16]. The free energy

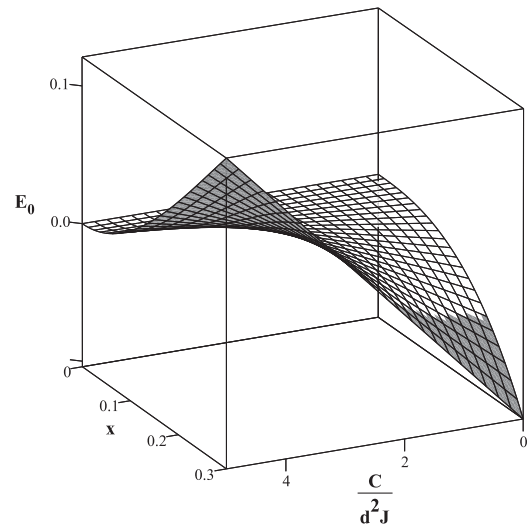

FIG. 2. The ground-state energy $E_0 = E_{gs} + Cx^2/2d^2$ for the antiferromagnetic spin chain with the easy-plane anisotropy $\mu = \pi/2 - 0.3$ as a function of x and C/d^2J.

F (the total free energy is $F_t = F + Cx^2/2d^2$) of the system with the Hamiltonian $\mathcal{H}$ can be written for the critical case as

$$F = E_{gs} - T\int_{-G}^{G} dx \ln[1+\exp(\varepsilon_1(x)/T)]\mathbf{s}_1(x), \quad (12)$$

where $\mathbf{s}_1(x) = \sum_{j=-\infty}^{\infty} s_1(x+2jG)$, $s_1(x) = (1/4\cosh(\pi x/2))$, and $\varepsilon_1(x)$ is the solution of the set of equations

$$\begin{aligned}\varepsilon_1 &= -A\mathbf{s}_1(x) + T\mathbf{D}_{12} \star \ln(1+\eta_2) \\ &\quad + T\mathbf{D}_{11} \star \ln(1+e^{\varepsilon_1/T}), \\ \ln\eta_j &= \mathbf{D}_{j1} \star \ln(1+e^{\varepsilon_1/T}) + \sum_{l=2}^{\infty} \mathbf{D}_{jl} \star \ln(1+\eta_j), \\ j &\geqslant 2, \end{aligned} \quad (13)$$

where $\star$ denotes convolution, and $A = \pi \mathrm{sn}(2\zeta, k)/\zeta$, with

$$\mathbf{D}_{jl}(x) = (1-\delta_{j,m_{i-1}})\delta_{j,l+1}\mathbf{s}_i(x) + \delta_{j,l-1}\mathbf{s}_{i+1}(x) + \delta_{j,l}\mathbf{d}_i(x) \quad (14)$$

for $j = m_i - 1$, with

$$\mathbf{d}_i(x) = \sum_{j=-\infty}^{\infty} d_i(x+2jG), \quad (15)$$

$$d_i(x) = \int_{-\infty}^{\infty} (d\omega/2\pi)\exp(i\omega x) \times \frac{\cosh((p_i - p_{i+1})\omega)}{2\cosh(p_i\omega)\cosh(p_{i+1}\omega)}, \quad (16)$$

$p_i = p_{i-2} - p_{i-1}\nu_{i-1}$, $p_1 = 1$, $p_0 = \pi/\zeta$, $\nu_i = [p_{i-1}/p_i]$, and

$$\mathbf{D}_{jl}(x) = (1-\delta_{j,m_{i-1}})\delta_{j,l+1}\mathbf{s}_i(x) + \delta_{j,l-1}\mathbf{s}_i(x) \quad (17)$$

for $m_{i-1} \leqslant j \leqslant m_i - 1$.

For $T \gg J, J_z$ only one solution $x = 0$ for the equation $(Cx/d^2) = (\partial F/\partial x)$ exists, hence at high temperatures there is no spin-nematic ordering in the considered system. On the other hand, for low temperatures $T \ll J, J_z$ one obtains [16]

$$F = E_{gs} - T^{3/2}\mathbf{s}_1(G)\sqrt{\frac{2\pi}{A\mathbf{s}_1''(G)}}e^{-A\mathbf{s}_1(G)/T}[1+O(T)], \quad (18)$$

where $\mathbf{s}_1(G) = K_k'k'/2\pi$, and $\mathbf{s}_1''(G) = (K_k')^3k^2k'/2\pi$. For small X (i.e., small k) one has $\mathbf{s}_1(G) \approx \ln(4/k)/2\pi$, and $\mathbf{s}_1''(G) \approx (k^2/2\pi)\ln^3(4/k)$, yielding

$$F^s \approx E_{gs}^{sing} - \frac{2T^{3/2}\zeta}{J_x \sin(2\zeta)k\ln^{3/2}(4/k)}\left(\frac{4}{k}\right)^{J_x\sin(2\zeta)/2T\zeta}. \quad (19)$$

i.e., similar singularity as in the ground state persists. It means that at low temperatures $x \neq 0$, i.e., the spin-nematic phase with the nonzero quadrupolar order parameter $\langle S_n^xS_{n+1}^x - S_n^yS_{n+1}^y\rangle$, exists for the antiferromagnetic spin-1/2 chain with the strong easy-plane anisotropy. Similar results can be obtained for thermodynamics of the system with the Hamiltonian $\mathcal{H}$, using the Trotter-Suzuki decomposition of the quantum transfer matrix [11], see Ref. [20]. The onset of the magnetic anisotropy in the isotropic antiferromagnetic spin-chain system with the quadrupolar order parameter $\langle S_n^xS_{n+1}^x + S_n^yS_{n+1}^y\rangle$, which violates the SU(2) symmetry of the spin chain, was considered in Ref. [21].

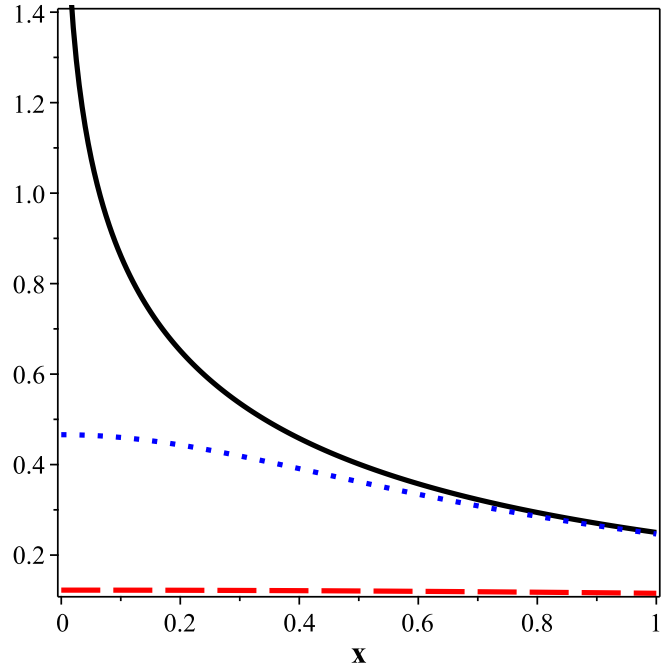

FIG. 3. The right-hand side of Eq. (20) for $H = 0$ and several values of T ($T = 10^{-7}J$, the solid black line; $T = 0.2J$, the dotted blue line; and $T = J$, the dashed red line) as a function of x. When the line $C/2Jd^2$ crosses those lines, the solution for the equation Eq. (20) exists.

V. SPECIAL CASE

More results can be obtained in the special case $J_z = 0$, where eigenstates are free noninteracting fermions [11]. In that case the Hamiltonian of the spin chain can be transformed to the quadratic form of Fermi operators with the help of the Jordan-Wigner transformation [22] $\mathcal{H} = \sum_k \varepsilon_k(b_k^\dagger b_k - 1/2)$, where $b_k^\dagger$ (b_k) creates (annihilates) the fermion for the mode k, and $\varepsilon_k = \{[H - J\cos(k)]^2 + J^2x^2\sin^2(k)\}^{1/2}$. Notice that the case $J_z = 0$ permits us to study the effect of the external magnetic field H applied along the z axis exactly, which is, unfortunately, impossible for the exact solution for $J_x, J_y, J_z \neq 0$.

The ground-state onset of the in-plane anisotropy for $J_z = 0$ case at $H = 0$ was studied in Ref. [23]. For small x one obtains $x = \exp(-\pi C/Jd^2)$, which means the spin-nematic ordering, violating the U(1) symmetry of the spin chain. It is easy to show that for $T \leqslant J$ (at least for temperatures not very high comparing to the value of the exchange coupling J) the equation

$$\frac{C}{2Jd^2} = \frac{J}{2\pi}\int_0^\pi \frac{\sin^2 k}{\varepsilon_k}\tanh\left[\frac{\varepsilon_k}{2T}\right], \quad (20)$$

where $\varepsilon_k = \{[H - J\cos(k)]^2 + J^2x^2\sin^2(k)\}^{1/2}$, has a nonzero solution for x, see Fig. 3. The critical temperature of

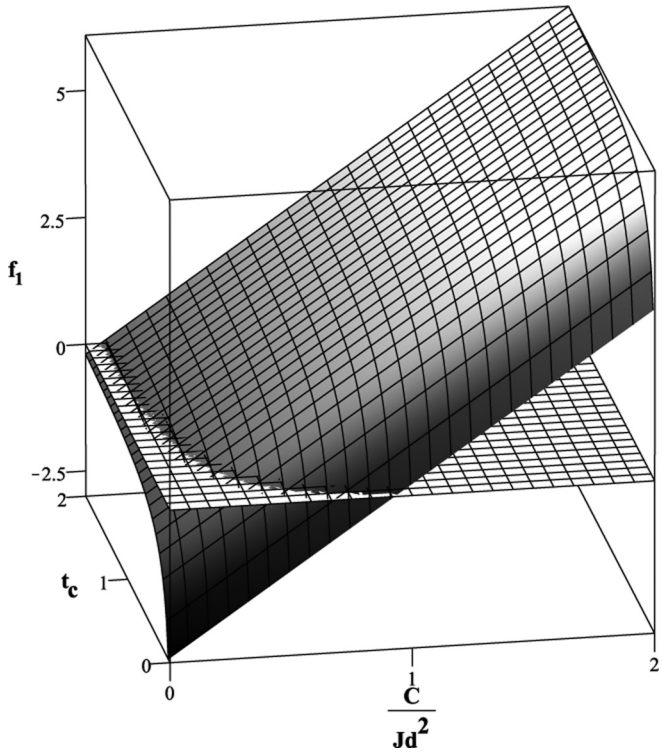

FIG. 4. The cross section of the surface f_1 as the function of C/Jd^2 and $t_c = T_c/J$ with zero plane determines the transition temperature of the spin chain to the spin-nematic ordered phase as a function of C/Jd^2.

the transition to the spin-nematic phase for $J_z = H = 0$ is determined from the condition

$$f_1 = \frac{\pi C}{Jd^2} - \int_0^{J/2T_c} dy \frac{\sqrt{1 - \frac{4T_c^2 y^2}{J^2}}}{y} \tanh(y) = 0. \qquad (21)$$

Figure 4 shows how the surface f_1 as a function of C/Jd^2 and $t_c = T_c/J$ crosses zero plane. The crossing curve determines the critical value of the temperature of the transition to the spin-nematic phase as a function of C/Jd^2. We see that the spin-nematic ordering exists for $C/Jd^2 \leqslant 1$ at nonzero temperatures. The critical temperature grows monotonically with the decrease of the parameter C/Jd^2.

As we pointed out already, the case $J_z = 0$ permits us to obtain the dependence of the onset of the spin-nematic phase in the critical spin chain system as a function of the applied field H. There is only one critical field $H_c = J$, at which the second-order quantum phase transition takes place in the system for $x \neq 0$ (the magnetic susceptibility has the logarithmic feature at that point in the ground state, being nonzero for $H < H_c$ and $H > H_c$). Figure 5 presents the right-hand side of Eq. (20) for $T \to 0$ ($T = 10^{-7}J$) for several values of the magnetic field $H \leqslant H_c$. One can see that there exists the region of values of $C/2Jd^2$, for which the nonzero x is the solution, i.e., the spin-nematic phase exists. For $H = H_c$ the solution can be written in the closed form; it is $x = [\sqrt{1 + (2\pi C/Jd^2)^2} - 1]/2\pi C/Jd^2$. For $H > H_c$ the nonzero solution for x is very small in the ground state and at low temperatures. Hence, the spontaneous spin-nematic ordering is robust with respect to the external magnetic field.

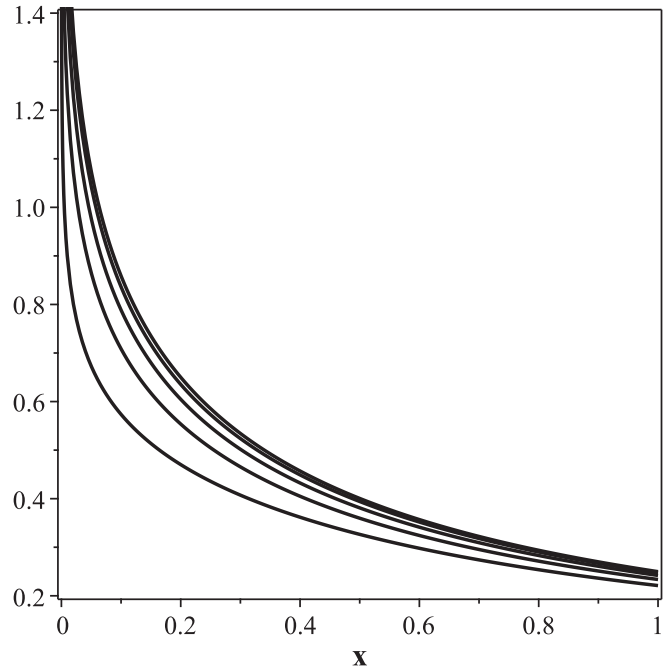

FIG. 5. The low-temperature ($T = 10^{-7}J$) behavior of the right-hand side of Eq. (20) for several values of the field (from bottom to top: $H = 0.1J$, $H = 0.3J$, $H = 0.5J$, $H = 0.7J$, and $H = 0.9J$).

VI. MAGNETOACOUSTIC CHARACTERISTICS

The spin-nematic ordering in spin-chain materials is related to shifts of either magnetic ions themselves, or to shifts of the surrounding ligands. Therefore, the natural way to observe such an ordering is to study the behavior of magnetoacoustic characteristics [24]. The onset of the quadrupolar order parameter $\langle S_n^x S_{n+1}^x - S_n^y S_{n+1}^y \rangle$ is mostly related to the elastic modulus C_{66}. However, the behavior of other elastic modules can also reveal the features, connected with the phase transition to the spin-nematic phase, violating U(1) symmetry. For highly symmetric crystals the behavior of the elastic modules C_{el} is connected with the behavior of the sound velocity v via $C_{el} = \rho v^2$, where ρ is the density of the crystal.

According to Ref. [25] the exchange-striction coupling in magnetic systems yields the renormalization of the velocity of the applied sound, proportional to some spin-spin correlation functions. Those correlation functions can be approximated by the combination of the magnetization and the magnetic susceptibility of the system, for purely spin-dipole case, and by the combination of the quadrupole moment and susceptibility for the system with possible quadrupolar degrees of freedom. References [26] imply the good agreement between the experiments and the theory (for many magnetic systems, including spin-chain compounds) achieved even when taking into account only the homogeneous part of the magnetic susceptibility. The renormalization of the sound velocity v due to the exchange-striction coupling in the general case can be written as

$$\frac{\Delta v}{v} \approx -\frac{v}{\rho V \omega^2}[|g(0)|^2(2M^2\chi + T\chi^2) + h(0)(M^2 + T\chi)], \qquad (22)$$

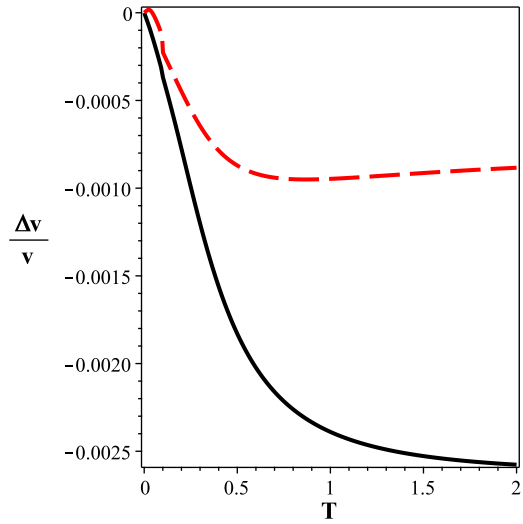

FIG. 6. The temperature behavior of the renormalization of the sound velocity in the spin-chain system with the spin-nematic ordering at $T_c = 0.1J$. The red dashed line describes the transverse sound mode, while the black solid line corresponds to the longitudinal mode.

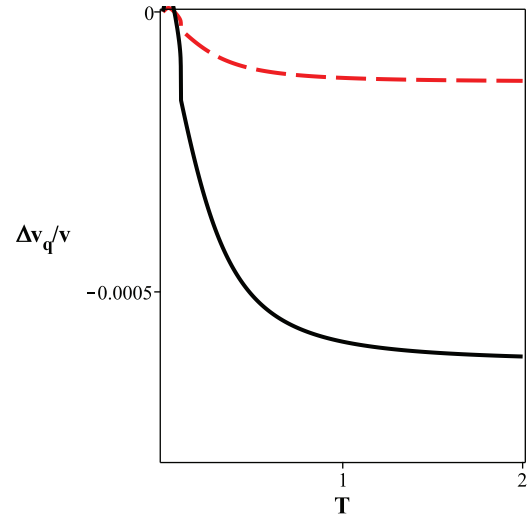

FIG. 7. The temperature behavior of the renormalization of the quadrupolar contribution to the sound velocity $\Delta v_q/v$ in the spin-chain system with the spin-nematic ordering at $T_c = 0.1J$. The red dashed line describes the transverse sound mode, while the black solid line corresponds to the longitudinal mode.

where V is the volume of the crystal, ω is the sound angular frequency, M and χ are the magnetization and the magnetic susceptibility, respectively, and the parameters of the exchange-sound coupling are

$$h = \sum_j e^{i\mathbf{q}\mathbf{R}_{ji}}[1-\cos(\mathbf{k}\mathbf{R}_{ji})](\mathbf{u}_{\mathbf{k}}\cdot\mathbf{u}_{-\mathbf{k}})\frac{\partial^2 J_{ij}^{\beta,\beta'}}{\partial\mathbf{R}_i\partial\mathbf{R}_j},$$
$$g = \sum_j e^{i\mathbf{q}\mathbf{R}_{ji}}(e^{i\mathbf{k}\mathbf{R}_{ji}}-1)\mathbf{u}_{\mathbf{k}}\frac{\partial J_{ij}^{\beta,\beta'}}{\partial\mathbf{R}_i} \tag{23}$$

(taken at $\mathbf{q}=0$), where $\mathbf{k}$ and u are the wave vector and the polarization of the sound wave, $\mathbf{R}_{ji} = \mathbf{R}_j - \mathbf{R}_i$, $\mathbf{R}_j$ is the position vector of the jth site of the magnetic ion, and $J_{ij}^{\beta,\beta'}$ ($\beta, \beta' = x, y, z$) are the exchange couplings between magnetic ions situated at the ith and jth site. On the other hand, the quadrupolar contribution to the renormalization of the sound velocity can be written as

$$\frac{\Delta v_q}{v} \approx -\frac{v}{\rho V\omega^2}\Big[|g_{xy}(0)|^2|Q|^2 + h_{xy}|Q| + T\sum_q |g_{xy}|^2\chi_Q(q))\Big], \tag{24}$$

where $Q = \langle S^x_{\mathbf{R}_i}S^x_{\mathbf{R}_j} - S^y_{\mathbf{R}_i}S^y_{\mathbf{R}_j}\rangle\exp(i\mathbf{q}\mathbf{R}_{ji})$ is the spin-nematic order parameter (the component of the quadrupolar moment), and $\chi_Q(q) = -\partial^2 F/\partial x^2$ is the quadrupolar susceptibility. Obviously, the renormalization of the sound velocity due to spin-nematic ordering is possible only inside the region, where $x \neq 0$ (i.e., the quadrupolar order parameter is nonzero). For the considered model the main contribution from the quadrupolar correction to the sound velocity comes from the homogeneous quadrupolar susceptibility

$$\chi_Q \approx \frac{J^2}{2\pi}\int_0^\pi dk\frac{\sin^2(k)}{\varepsilon_k}\Big[\frac{\varepsilon_k}{2T\cosh^2(\varepsilon_k/2T)} - \frac{J^2x^2\sin^2(k)}{\varepsilon_k^2}\tanh(\varepsilon_k/2T)\Big], \tag{25}$$

where $x(T, H)$ is the solution of Eq. (20). Similar expressions can be derived for the sound attenuation. Figure 6 shows the temperature behavior of the longitudinal (for the wave vector of the sound wave parallel to its polarization) and transverse (for the wave vector and the polarization being perpendicular) sound velocities of a spin-chain material (for

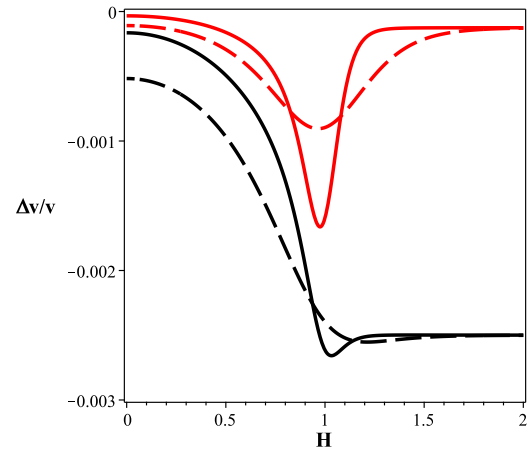

FIG. 8. The magnetic field behavior of the renormalization of the sound velocity in the spin-chain system with the spin-nematic ordering at $T_c = 0.1J$. The red dashed line describes $\Delta v/v$ for $T = 0.15J$, while the red solid solid line describes the case $T = 0.05J$. for the transverse mode; the black dashed and solid lines are related to the cases of $T = 0.15J$ and $T = 0.05J$, respectively, for the longitudinal sound mode.

$J_z = 0$) with the transition to the spin-nematic state at $T_c = 0.1J$. It turns out that namely the transverse mode (related to the u_{xy} distortions, like the C_{66} one) manifests mostly the transition to the spin-nematic ordered state. However, the transition to the spin-nematic phase is more pronounced in the temperature dependence of the total transverse mode of the renormalized sound velocity, where the scale of changes of the sound velocity due to magnetic (dipole) characteristics is smaller.

In Fig. 7 the contribution of the quadrupolar part is shown separately. We see that in fact both longitudinal and transverse parts sound modes are affected by the spin-nematic transition. Notice the smaller scale in Fig. 7 comparing to Fig. 6.

Finally, Fig. 8 manifests the magnetic field behavior of the transverse and longitudinal sound velocities at temperatures below and above phase transition to the spin-nematic phase. The magnetic field behavior of the sound velocity is mostly determined by the spin dipole contribution. Summarizing, the behavior of the magnetoacoustic characteristics can serve as a good tool to detect the spin-nematic ordering in spin-chain systems.

VII. CONCLUSION

In conclusion, we have predicted the spontaneous spin-nematic ordering, violating the U(1) symmetry in spin-chain systems. The predicted order is the result of the competition between the gain of the energy of the spin subsystem, caused by the spin-nematic ordering, and the loss of energy of the lattice due to the distortions of either magnetic ions, or surrounding them nonmagnetic ligands. Our theory is applicable the ground state and for nonzero temperatures, lover than the exchange energies and the Debye temperature. To avoid the magnetic (dipole) ordering the minimal interchain spin-spin coupling in the spin-chain compound has to be smaller than $[Z\chi(k,T)]^{-1}$, where Z is the coordination number, and $\chi(k,T)$ is the inhomogeneous magnetic susceptibility of the spin chain [27]. For the special case the effect of the external magnetic field on the spin-nematic ordering has been also studied. We have shown how such a spin-nematic ordering in a spin-chain material can be observed in the behavior of sound characteristics in magnetoacoustic experiments.

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