

The Geometric Frustration Effects in Nanostructured Magnetic Systems

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We numerically investigate the geometric frustration effects in finite ring-shaped antiferromagnetic $S = 1/2$ Heisenberg models with an odd number spins in a ring. It is shown that, in a single ring, the lattices always display heterogeneous distortions for an arbitrarily large spring constant. However for any nonzero antiferromagnetic interring coupling in a finite two-leg spin ladder structure, a second order magnetoelastic transition (not the thermodynamic transition) takes place from the heterogeneous lattice distortion phase to the uniform phase (without lattice distortion) when the spring constant is increased. For ferromagnetic inter-ring coupling, there exists a critical coupling strength J_{\perp}^c , for $J_{\perp} > J_{\perp}^c$ a first order magnetoelastic transition is present.

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I. INTRODUCTION

Single molecular magnets (SMMs), which are nanostructured spin systems and formed by a finite number of exchange-coupled transition-metal ions, have become extensively investigated topics in condensed matter physics [1–11]. On the one hand, the SMMs can be regarded as the bridge to explore the crossover from microscopic magnetism to collective effects in macroscopic structure. On the other hand, the unique properties in these systems, such as giant magnetostriction [1], resonant tunneling of the magnetization [2, 3] and geometric phase effects [4] promise to open new frontiers in technology and fundamental research. In experiments, plenty of SMMs, especially with high-symmetry ring-shaped structures [5–9], have been synthesized and intensively studied. Recently, in the Cu₈ ring, the ⁶³Cu NQR (nuclear quadrupole resonance) spectrum shows four structurally nonequivalent Cu ions [9], which is perhaps related to the appearance of lattice dimerization. The lattice dimerization is a magnetoelastic (ME) instability, which may originate from the spin-phonon interaction. This phenomenon was first predicted to occur in the infinite $S = 1/2$ Heisenberg antiferromagnetic (AF) chain [12], and afterward, was observed experimentally in the quasi-one-dimensional $S = 1/2$ compound CuGeO₃ [13]. Due to the size effects, in finite spin systems, the spin-phonon interaction becomes stronger, and the lattice distortion behavior is evidently different from that in thermodynamic limit [14–16].

In spin systems, the geometric frustrations have strong effects on the ground state properties. For example, for a square Heisenberg antiferromagnet (HAF), the ground state

has Néel ordering with staggered magnetization [17]. But for the highly frustrated $S = 1/2$ Kagomé lattice HAF, the ground state is disordered [18, 19]. The triangular lattice is another kind of frustration structure. Some results suggest that the above mentioned system has a disordered ground state [20, 21]. For small spin rings of various numbers N of spin sites, many results indicate that [22–24], if N is an even number, then the ground state, which has total spin $S = 0$, is non-degenerate. But if N is an odd number in which frustration is also introduced, then the ground state has total spin $S = 1/2$ and is fourfold degenerate. So the magnetic systems with an odd number spins would indicate novel properties [22, 23, 25].

Theoretically, in finite ring-shaped systems with an even number of spins in a ring, the ME transitions (not the thermodynamic transition) have been investigated by some groups [15, 16]. But to our knowledge, few investigations have been performed on this problem in the systems with an odd number spins in a ring. These are the main motivations in this paper. Our results indicate that, for a single AF $S = 1/2$ ring with an odd number of spins, the system always locates in the lattice distortion phase, and the distortion amplitude at every site is heterogeneous. For the $S = 1/2$ spin-ladder model, in which two AF rings with odd number spins in each ring interact with each other antiferromagnetically and ferromagnetically, the systems may have a second order (continuous) and a first order ME transition from the heterogeneous lattice distortion phase to the uniform phase (without lattice distortion), respectively, as the spring constant is increased.

II. MODELS AND METHODS

In the adiabatic approximation, the Hamiltonian containing the lattice distortion, spin-phonon interaction, and interring coupling can be written as

$$H = J \sum_{j=1,2} \sum_{i=1}^{N \in \text{odd}} (1 + \alpha u_{i,j}) \mathbf{S}_{i,j} \cdot \mathbf{S}_{i+1,j} + J_{\perp} \sum_{i=1}^{N \in \text{odd}} \mathbf{S}_{i,1} \cdot \mathbf{S}_{i,2} + \frac{K}{2} \sum_{j=1,2} \sum_{i=1}^{N \in \text{odd}} u_{i,j}^2, \quad (1)$$

where $\mathbf{S}_{i,j}$ (respectively, $u_{i,j}$) represents the spin-1/2 operator at site i (respectively, the lattice distortion between site i and $i + 1$) in the j th ring ($j = 1$ or 2). K is the lattice spring constant and the last term of the Hamiltonian (1) is the elastic energy. α represents the spin-phonon interaction. Under the scaling $\alpha u_{i,j} \rightarrow \delta_{i,j}$, the spin-phonon interaction strength has been absorbed in the spring constant ($K/J\alpha^2 \rightarrow K$). N ($\in \text{odd}$) is the lattice number in each ring. J (>0) and J_{\perp} ($J_{\perp} >0$, AF coupling; $J_{\perp} <0$, ferromagnetic (FM) coupling) are the respective intraring and interring coupling strengths. J will be taken as the unit of energy. For a ring-shaped structure, $\mathbf{S}_{N+1,j} = \mathbf{S}_{1,j}$.

The lattice distortion of the Hamiltonian (1) with minimal ground state energy is obtained by the following iterative procedure. First, with a random initial distortion pattern $\{\delta_{i,j}\}$, we determine the lowest energy eigenvalue of (1) by the Lanczos diagonalization technique. Then according to the equilibrium condition derived from the Hellman-Feynman theorem [15]

$$K\delta_{i,j} + J \langle \mathbf{S}_{i,j} \cdot \mathbf{S}_{i+1,j} \rangle - \frac{J}{N} \sum_{j=1,2} \sum_{i=1}^{N \in \text{odd}} \langle \mathbf{S}_{i,j} \cdot \mathbf{S}_{i+1,j} \rangle = 0, \quad (2)$$

we update the lattice distortions until the iterations converge and thereby obtain a group of $\{\delta_{i,j}\}$. The results indicate that, for all the cases we investigated, $\delta_{i,1} = \delta_{i,2}$. So we only give the results of $\delta_i = \delta_{i,1}$ (or $\delta_{i,2}$).

III. RESULTS AND DISCUSSION

At first, we set $J_{\perp} = 0$. The system can be regarded as two single rings without interaction and the ground state has total spin $S = 1/2$ for each ring. The lattice distortion of every site δ_i in a ring is depicted in Fig. 1 with $K = 1.0, 2.0,$ and 3.0 for $N = 5$. It can be seen that both δ_1 and δ_3 are negative, but δ_2 and δ_4 are positive. The value of δ_5 is very small. These results indicate that site one and site two (respectively, site three and site four) are away from each other. Site two and site three (respectively, site four and site five) approach each other. The amplitudes of lattice distortion at every site are not equal to one another. The bond between site five and site one is almost unchanged. So, except for site one, the other four lattices form heterogeneous dimerization. In Fig. 2 we give the function relations between the lattice distortion of every site and the spring constant. With increasing spring constant, all of the lattice distortion amplitudes become smaller and smaller. But even for very large spring constant, the lattice distortions do not equal to zero. So we speculate that, for arbitrarily large spring constant, there may always exist heterogeneous lattice distortions in the spin system with odd N . About this problem we will give further evidence later (in Fig. 4). These characters of lattice distortion are greatly different from those in the ring with finite even N . For finite even N , there is a critical spring constant K_c , beyond and below which the systems locate in uniform phase and regular dimerized phase with equal distortion amplitudes at every site, respectively [15].

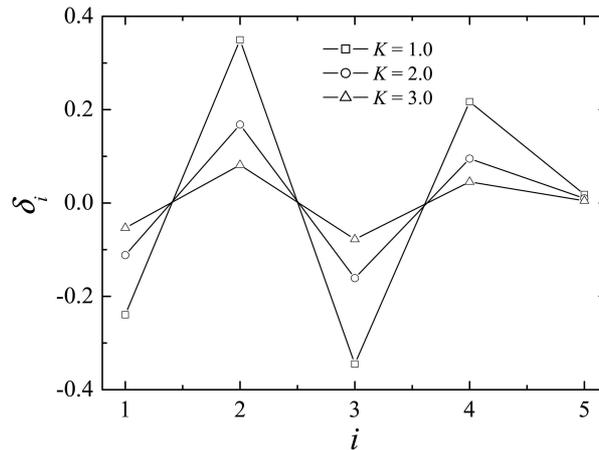


FIG. 1: The lattice distortion of every site δ_i with $K = 1.0, 2.0,$ and 3.0 for $N = 5$.

Next, we study the behavior of the lattice distortion in ring-shaped two-leg spin ladder structures with AF interring coupling. The lattice distortion of every site δ_i versus

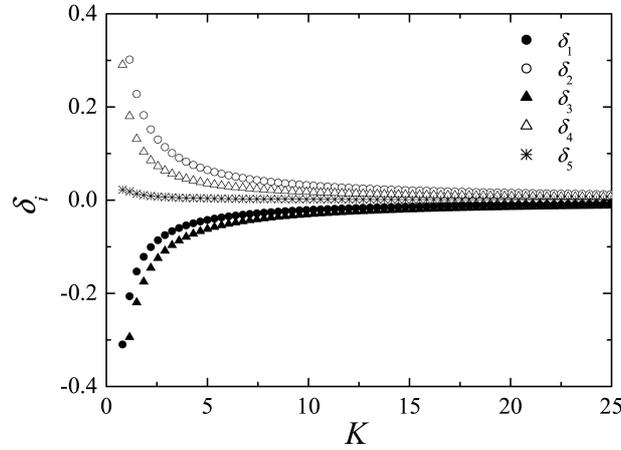


FIG. 2: The K dependence of lattice distortion of every site δ_i with $J_{\perp} = 0.0$ for $N = 5$.

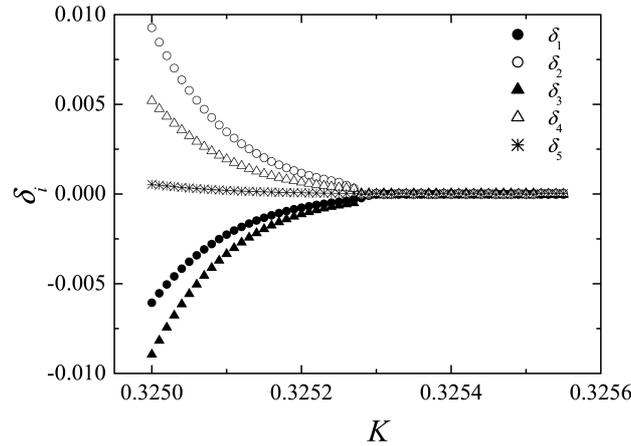


FIG. 3: The K dependence of lattice distortion of every site δ_i with $J_{\perp} = 2.0$ for $N = 5$.

the spring constant is shown in Fig. 3 for $J_{\perp} = 2.0$ and $N = 5$. It can be seen that, for smaller spring constant, the system still locates in the heterogeneous lattice distortion phase. When the spring constant is increased, the distortion amplitude of every site is reduced monotonically and at a critical spring constant K_c , all of the lattice distortions totally disappear simultaneously, and the system becomes a uniform phase. The process of transition is continuous evidently without abrupt jumping of the lattice distortions from a finite value to zero. For larger J_{\perp} , such as $J_{\perp} = 10.0$ which is not displayed in this paper, our results indicate that the transition is still continuous. In Fig. 4, the reciprocals of K_c as functions of J_{\perp} are shown. The solid squares are the numerical results and the solid lines are polynomial fits with the fifth order. It can be found that, with decreasing AF interring coupling, the values of $1/K_c$ are reduced gradually. As $J_{\perp} = 0$, $1/K_c$ also

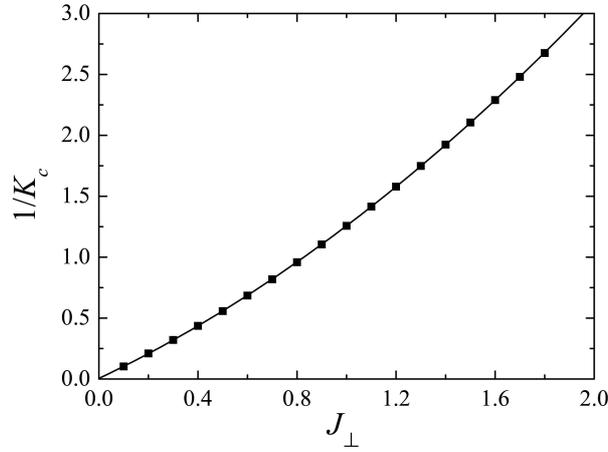


FIG. 4: The J_{\perp} dependence of $1/K_c$ with AF interring coupling for $N = 5$. The solid squares are numerical data and the solid lines are polynomial fit with the fifth order.

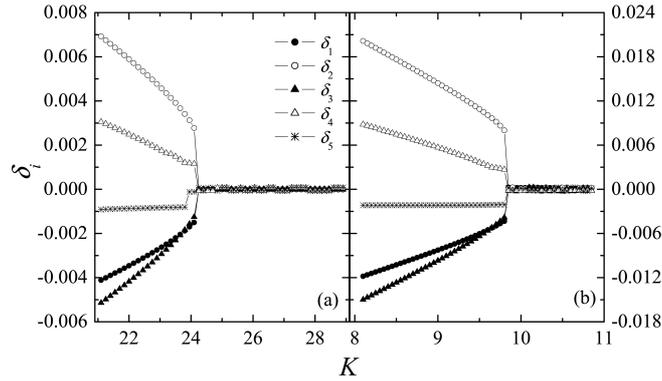


FIG. 5: The K dependence of the lattice distortion of every site δ_i with (a) $J_{\perp} = -2.0$ and (b) $J_{\perp} = -4.0$ for $N = 5$.

equals to zero, which clearly shows that there is no ME transition for a single ring with odd N . That is to say, as $J_{\perp} = 0$, the ground state always locates in heterogeneous lattice distortion phase, which offers evidence for the result in Fig. 2. But for arbitrary nonzero AF interring coupling, there may exist a second order ME transition from the heterogeneous lattice distortion phase to the uniform phase when K is increased.

For FM interring coupling, The K dependence of the lattice distortion of every site δ_i in a ring is given in Fig. 5(a) ($J_{\perp} = -2.0$) and Fig. 5(b) ($J_{\perp} = -4.0$) for $N = 5$. The results indicate that, for both cases, with increasing K , the amplitudes of the lattice distortions decrease monotonically. When K equals a critical value K_c , the lattice distortions jump suddenly from a finite value to zero. These results indicate clearly that a first order ME transition takes place from the heterogeneous lattice distortion phase to the uniform phase.

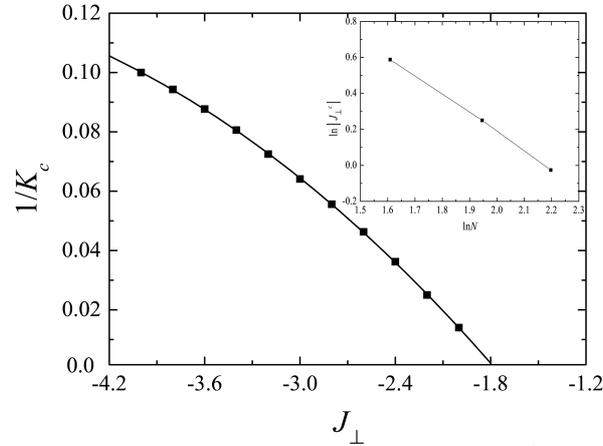


FIG. 6: The J_{\perp} dependence of $1/K_c$ with FM interring coupling for $N = 5$. The solid squares are numerical data and the solid lines are polynomial fits with the fifth order. In the inset, the size effects of J_{\perp}^c are given for $N = 5, 7$, and 9 .

In Fig. 6, the J_{\perp} dependence of $1/K_c$ for $N = 5$ is depicted. The solid squares are the numerical data and the solid lines are polynomial fits with the fifth order. It is shown that, with weakening the interring couplings, the values of $1/K_c$ become smaller and smaller. At $J_{\perp}^c \approx -1.8$, $1/K_c = 0$. That is to say, for a finite two-leg spin-ladder ring-shaped system with odd N , there exist a nonzero FM interring coupling J_{\perp}^c , when $|J_{\perp}| > |J_{\perp}^c|$, the ME transition from the lattice distortion phase to the uniform phase is a first order transition, but when $|J_{\perp}| < |J_{\perp}^c|$, the system always locates in the heterogeneous lattice distortion phase. In the inset of Fig. 6, the values of $\ln|J_{\perp}^c|$ against $\ln N$ are plotted for $N = 5, 7$, and 9 . With increasing N , the values of $\ln|J_{\perp}^c|$ become smaller monotonically. As $N \rightarrow \infty$, $\ln|J_{\perp}^c| \rightarrow -\infty$. Correspondingly, J_{\perp}^c approaches zero. That is to say, in the thermodynamic limit, any nonzero FM interring coupling may induce a first order ME transition in the two-leg spin ladder model with odd N . This is consistent with that for the even N case [16]. In a spin system, the properties of the $S = 1$ Heisenberg chain can be understood by representing the $S = 1$ model as two FM coupled $S = 1/2$ models [26]. The results above mentioned indicate that, for a finite $S = 1$ AF ring with odd number spin, the system may have a first order ME transition.

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