Long-range ferromagnetism in one-dimensional monatomic spin chains

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We show that transition states during spin flips, besides interspin exchange and on-site anisotropy, are necessary for a complete description of monatomic spin chains, and study the dynamical spin system using kinetic Monte Carlo method based on transition-state theory. Using experimental parameters of the monatomic Co spin chains on Pt surfaces, we produce the experimental hysteresis loop at 10 K and show that there is no hysteresis loops at 45 K. There is a phase crossover from the long-range ferromagnetism at low temperature to disordered magnetic states at high temperature. Our systematical simulation shows that the chain-length dependence of the long-range ferromagnetism is very weak and even approaches to zero when the chain length becomes large enough. The crossover temperature is a monotonously increasing function of the sweeping rate of changing magnetic field. The simulations are justified because the quantum effect can be ignored except for very low temperatures. Our theory explains the ferromagnetism of finite one-dimensional spin chains with large magnetic anisotropy such as the monatomic Co chains on Pt surfaces.

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I. INTRODUCTION

Experimental and theoretical studies revealed that magnetic anisotropy energies and magnetic moments in one-dimensional (1D) metal systems and quasi-1D metal stripes can be largely enhanced compared to those in two-dimensional (2D) film and three-dimensional (3D) bulk materials owing to the reduced symmetry and atomic coordinations in these spin systems.[1–6] Because of the extremely large anisotropy, the 1D magnetic materials are very attractive for spintronic applications because of their good thermal stability. On the other hand, the dimensionality of magnetic systems plays a key role in determining their spontaneous magnetization, specific heat, and other thermodynamical quantities.[2,3,7] Recently, Gambardella *et al.* first constructed high-density arrays of Co monatomic chains on the vicinal Pt(997) surface by self-assembly epitaxial techniques,[8] and showed that at 10 K there exists a hysteresis loop, which implies long-range ferromagnetic spin order, and that at 45 K there are no hysteresis loops at all, which means short-range magnetic orders only.[9] This appears to be contradictory to the established result of lattice models that there is no phase transition in 1D systems without long-range interactions,[10–12] such as usual 1D Heisenberg and Ising models. There have been some theoretical studies on this issue,[13–15] but the mechanism of the spin dynamics and the experimental 1D phase change remains unexplained. It is highly desirable to find the mechanism.

In this paper, we study the long-range ferromagnetism and the phase change. Because usual spin exchange lattice models cannot be used to explain this phenomenon, we take into account transition states during single spin flips in addition to the spin exchange and the extremely large anisotropy, and use a kinetic Monte Carlo (KMC) method[16] to simulate the resultant dynamical spin system. This scheme is feasible because of the large magnetic anisotropy. Using experimental parameters of monatomic Co spin chains on Pt surfaces, we produce the experimental magnetization hysteresis loop at 10 K and show that there is no such loops at 45 K. The long-range ferromagnetism exists at low temperature but is replaced by short-range magnetic order at high temperature. There is a phase crossover temperature, approximately 20 K.[9] Furthermore, we show that the phase crossover temperature decreases with the system size decreasing and increases with the sweeping rate of changing the applied magnetic field. The simulations are justified because the quantum effect of the spin dynamics should not be important except at substantially lower temperature. Our theory explains the ferromagnetism of finite 1D spin chains with large magnetic anisotropy such as the monatomic Co chains on Pt surfaces.

The remaining part of this paper is organized as follows. In Sec. II we shall present our model and methodology. In Sec. III we shall present our main simulation result and compare it with experiment. In Sec. IV we shall make some discussions on our result and method in comparison to other Monte Carlo ones. Finally, we shall present our conclusion in Sec. V.

II. MODEL AND METHODOLOGY

According to the experimental results,[9] we take the fact that the easy magnetization axis of the Co chains is perpendicular to the chain direction, the anisotropy energy per Co atom is very large, being the same order of magnitude as the intrachain exchange interaction, and the interchain interactions are shown to be ignorable.[9,13–15] Therefore, these anisotropic monatomic spin chains are really 1D spin ones. We use a Heisenberg model with a large uniaxial magnetic anisotropy to describe the magnetic properties of the 1D monatomic chain. With an external magnetic field **B** applied along the easy axis, the Hamiltonian can be written as

$$H = -\sum_{\langle ij\rangle} J_{ij}\mathbf{S}_i \cdot \mathbf{S}_j - k_u \sum_i (\mathbf{S}_{i,z})^2 - \mu\mathbf{B} \cdot \sum_i \mathbf{S}_i, \qquad (1)$$

where the dot means vector product, the summation $\langle ij\rangle$ is over all nearest neighbor sites i and j, and $\mathbf{S}_i$ is the normal-

ized spin variable at site i. The exchange constant J_{ij}, the uniaxial anisotropy energy k_u, and the magnetic moment μ are chosen according to corresponding experimental values.[9] This Hamiltonian can describe the paramagnetic state at high temperature,[17] but there is no phase transition or change in this model because, as it is well known, there is no phase transition in 1D models without long-range interactions. More factors are needed to describe both the long-range ferromagnetism at low temperature and the disorder magnetic states at high temperature.

The barriers of the transition states during the single spin flip must be taken into account in order to describe the long-range ferromagnetism. The large uniaxial anisotropy makes the spins tend to orient along the easy axis. We can define the two metastable states for a spin to orient along the easy axis. When being in the metastable orientations, the spin variable $\mathbf{S}_i$ reduces to the reduced variable s_i which takes values +1 and −1, corresponding to the spin orientation in the $+z$ and $-z$ direction, respectively. To describe the transition states of a spin, we begin with the given metastable direction and assume the spin can deviate from its current metastable direction, $+z$ or $-z$. We use an angle θ_i to describe the angular deviation of the spin at site i from the initial metastable state. A transition state begins at $\theta_i=0$ ($\cos\theta_i=1$, initial state) and ends at $\theta_i=\pi$ ($\cos\theta_i=-1$, reversed spin). A nonzero θ_i will produce an energy increment

$$e_i = k_u \sin^2\theta_i - h_i(\cos\theta_i - 1), \tag{2}$$

where

$$h_i = \left(\sum_j J_{ij}s_j + \mu B\right)s_i. \tag{3}$$

The energy increment yields a transition-state barrier,

$$\Delta E_i = (2k_u + h_i)^2/4k_u, \tag{4}$$

at $\cos\theta_i=h_i/2k_u$ when $2k_u>|h_i|$, as Figs. 1(a)–1(c) show. The spin reversal is a barrier-limited process. Its rate can be expressed as Arrhenius law

$$R = R_0 \exp(-\Delta E_i/k_BT), \tag{5}$$

where k_B is Boltzmann constant and T is temperature.[16] The prefactor R_0 is taken as 10^{-9} s^{-1} in our simulation. We use the kinetic Monte Carlo method[16] to simulate the dynamics of the spin system. If the condition $2k_u\leq|h_i|$ is satisfied, there is no transition state barriers for some spin reversal processes, as is shown by Figs. 1(d) and 1(e) and we use the Glauber method[18] to calculate the exponential factor of the rates, with the prefactor being kept the same. The expression of the rate implies that our spin processes are thermal activated. This scheme is justified for our simulation because dipolar interactions can be ignored thanks to the extremely large magnetic anisotropy[19,20] and quantum tunnelling comes into action at very low temperature only.[21]

We take the parameters mainly from the experiment.[9] The exchange interaction, nonzero only between the nearest-neighbor spins, is equivalent to $J=7$ meV, and the anisotropy energy is $k_u=0.3J=2.1$ meV. The magnetic field is along the easy axis. We change the applied magnetic field starting from a strong field $-B_0$, for which all spin magnetic moments are aligned along the $-z$ direction. Then the field strength increases by an increment of ΔB gradually to $+B_0$, and then decreases back to $-B_0$ so that a magnetic field sweeping cycle is complete. The sweeping rate of changing applied magnetic field is set to 132 T/s unless specified otherwise. The magnetization is normalized to 1 when all the reduced spin variables take 1. We use free boundary conditions and start with an initial state with all the reduced spin variables taking −1. In the following, each data point is calculated by averaging over 500 independent runs to reduce possible errors.

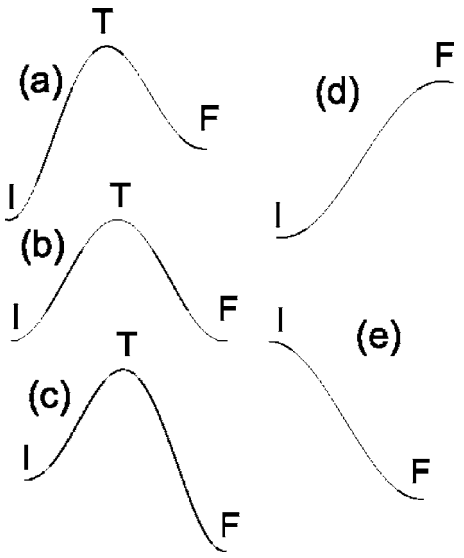

FIG. 1. The schematic transition-state energy for a single spin flip from the initial state (I) to the final state (F). The point T in (a), (b), and (c) is the transition state with maximal energy. There is no transition-state barrier in (d) or (e).

III. MAIN SIMULATION RESULT

Figure 2 shows the magnetization responses to the applied magnetic field for the monatomic chains of $N=80$ at $T=10$ K and 45 K. At the low temperature, $T=10$ K, the magnetization curve exhibits hysteresis, but there is no hysteresis loop at $T=45$ K. In Fig. 2(b) the dashed line represents the magnetization for a chain consisting of isolated

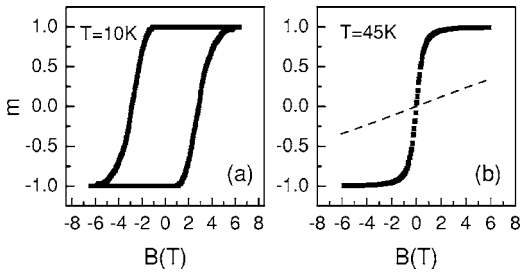

FIG. 2. Magnetization (squares) as a function of the applied field in the one-dimensional monatomic chain of $N=80$ for the exchange interaction $J=7$ meV and the single-site anisotropy energy $k_u=2.1$ meV at $T=10$ K (a) and $T=45$ K (b). The dashed line in (b) is calculated with $J=0$, $k_u=2.1$ meV, and $T=45$ K.

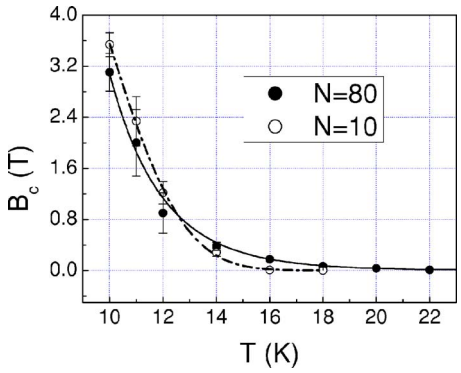

FIG. 3. (Color online) Temperature dependence of the coercive fields above 10 K. The coercive field should stop increasing when the temperature decreases far enough from 10 K and becomes saturated at very low temperature because of the quantum effect. The lines show the fitting of the data with a simple exponential function, $B_c = A \exp[-(T/T_0)^p]$.

spins ($J=0$) with the magnetic moment $\mu=4.0\mu_B$. [This value is taken from the caption of Fig. 3 in the experiment paper[9] and includes the contributions of spin and orbital moments of Co atom and that of induced moments (per Co atom) on Pt neighbors.] Anisotropy energy $k_u=2.1$ meV per atom. Our magnetization curve at $T=10$ K is in agreement with experimental result,[9] and the magnetization curve at $T=45$ K is in good agreement with the experimental one. Moreover, the magnetization for the chain of isolated atoms is very similar to the magnetic curve for an isolated Co atom.[9]

The hysteresis loop is characterized by the critical field at which the magnetization is equivalent to zero. In Fig. 3 we show the temperature dependence of the coercive fields for the two cases of $N=80$ and $N=10$. The values of the coercive field for $N=80$ decrease with temperature increasing from 10 K, and become zero above $T=20$ K. For $N=10$, the coercive field already is zero at 16 K. The crossover temperature for $N=10$ is smaller than that for $N=80$. Below 12.5 K we observe higher coercive field for $N=10$ than for $N=80$. These temperature dependences of the coercive fields above 10 K can be described by a simple function

$$B_c = A \exp[-(T/T_0)^p], \tag{6}$$

where the three parameters A, T_0, and p are the fitting parameters. They take 4778.3 T, 0.5788 K, and 0.70 for $N=80$; and 9.0859 T, 10.16 K, and 4.0 for $N=10$. Below 10 K the curve should deviate from the fitting exponential function because the quantum effect should play some role. Below 5 K the quantum effect cannot be ignored any more,[21] and the coercive field is expected to be saturated when the temperature becomes lower than 5 K.

It is interesting to systematically study the effect of spin chain length on the ferromagnetism and the phase crossover temperature. To make our result general, we express the temperature T and the anisotropy energy k_u in terms of the exchange constant J. Figure 4 shows the magnetization curves as a function of the applied field for the chains of $N=10$, 20, 40, 60, and 80 at temperatures $T=0.197J$ and $0.308J$. Here

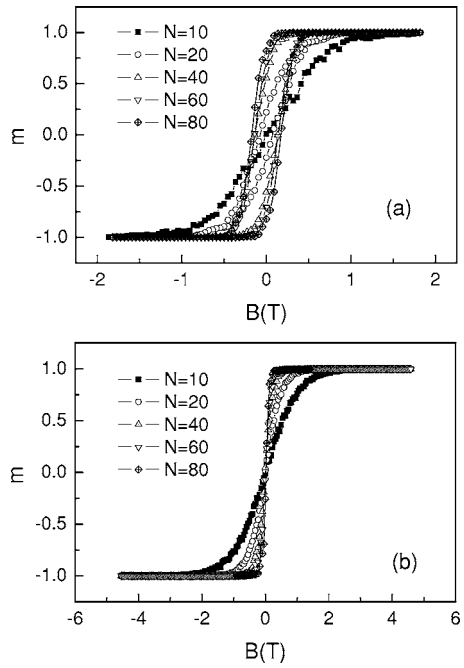

FIG. 4. The magnetic hysteresis curves for different chains with $N=10$, 20, 40, 60, and 80 at two temperatures: (a) $T=0.197J$ and (b) $T=0.308J$. The magnetic anisotropy energy is $k_u=0.3J$. The phase crossover to ferromagnetism takes place at the temperature $T_c=0.246J$ for the system of $N=80$. The crossover temperatures decrease with N decreasing. There is no hysteresis loops above the crossover temperatures.

the magnetic anisotropy energy is $k_u=0.3J$. For $T=0.197J$ the coercive field decreases with N decreasing, as is shown in Fig. 4(a). For the smallest $N=10$ among the five systems, the coercive field already is equivalent to zero. Anyway, the N dependence of the coercive fields becomes very weak for $N\geq 40$. The low-temperature ferromagnetism becomes independent of the length for long enough spin chains. At higher temperatures, for instance, $T=0.308J$, there is no hysteresis loops and the effect of the chain length can be seen in the sense that the magnetization starts to rise at a smaller field for a smaller system (with smaller N), but the coercive field remains zero indicating that there exists short-range magnetic order only, as is shown in Fig. 4(b). These results reveal that the long-range ferromagnetism in long chains is almost independent of the chain length, and the short-range magnetic orders are weakly dependent on the chain length but their key property remains independent of the chain length.

In Fig. 5 we show the effect of the sweeping rate on the coercive field at the temperature 16 K for $N=80$ and $N=40$. The sweeping rate used in doing simulations for Figs. 2–4 is 132 T/s, a reasonable value. For the two N values, the coercive fields increase monotonously with the sweeping rate increasing. When the sweeping rate v decreases down to zero, the coercive fields B_c decrease quickly and are expected to approach zero. This implies that the hysteresis loops will disappear when the sweeping rate is zero.

IV. DISCUSSION

It should be pointed out that the phase crossover cannot be obtained with Hamiltonian (1) alone, as has been shown

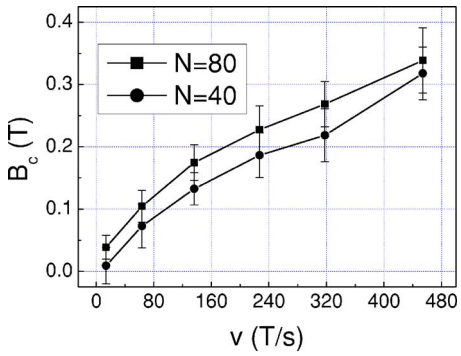

FIG. 5. (Color online) Sweeping rate dependence of coercive fields B_c for $N=80$ and $N=40$ at temperature 16 K. The lines are plotted by simply connecting the nearest points for guiding the eyes. The sweeping rate used in other figures is 132 T/s, a reasonable value.

in the literature.[17] The transition states and their barriers are necessary to the 1D phase crossover in the monatomic spin chains. The barriers are supplied by the large magnetic uniaxial anisotropy energy. The resultant processes are thermally activated. The kinetic Monte Carlo method is the right method for capturing the main properties of the dynamical spin system. The simulation is justified because the quantum tunnelling of the spins can be ignored safely except below 5 K,[21] which is well below our lowest simulation temperature 10 K. Therefore, our main result shows reliably that long-range ferromagnetism and phase crossover can occur in one-dimensional monatomic spin chains of finite lengths without long-range interactions.

This does not imply that the established thermodynamical conclusion that there exists no phase transition in 1D lattice models[10] needs modification or the Mermin-Wagner theorem[12] is wrong. What our theory shows is that the theorem does not make sense for nanoscale systems such as what we have treated. This is because for nanomagnets we do not need infinitely long correlation lengths in time and space. The conventional lattice models, such as that defined in (1), cannot produce any phase transition or crossover in such nanoscale systems. The transition states must be taken into account in order to completely describe these nanoscale systems. Our result should turn out to be useful in designing real nanoscale devices because our result shows that single-domain ferromagnetic orders can be established and last for a long enough time in chainlike nanomagnets with large uniaxial magnetic anisotropy. Such long-range ferromagnetism can be manipulated thanks to the accurate control techniques of adatoms on substrates[9] and the technique of ordered atomic doping.[22]

V. CONCLUSION

In summary, we study one-dimensional monatomic spin chains with giant magnetic anisotropy using a dynamical spin model. In addition to the interspin exchange and on-site anisotropy, transition states during the single spin flip are taken into account in this model. We use the kinetic Monte Carlo method based on transition-state theory to simulate the spin dynamics. Using the experimental parameter values, we produce the experimental ferromagnetic long-range order in the one-dimensional monatomic chains of finite length at low temperatures, and clarify the phase crossover to disordered magnetic states at high temperature. The ferromagnetism is a long-range magnetic order which becomes independent of chain length for long enough chains. The energy barriers of the transition states are essential to the one-dimensional ferromagnetic order and the phase crossover. Our simulation is justified because the quantum effect in these spin systems can be ignored except for very low temperatures which are substantially lower than our lowest simulation temperature. Our theory should be desirable to understand ferromagnetism and phase changes in nanomagnets such as the monatomic Co chains on Pt surfaces.

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