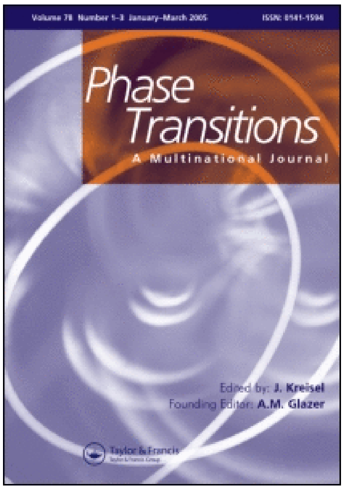

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Investigation of magnetic field effect on surface and finite-site free energy in one-dimensional Ising model of nanosystems

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Investigation of magnetic field effect on surface and finite-site free energy in one-dimensional Ising model of nanosystems

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We investigate a one-dimensional (1-D) Ising model for finite-site systems. The finite-site free energy and the surface free energy are calculated via the transfer matrix method. We show that, at high magnetic fields, the surface free energy has an asymptotic limit. The absolute surface energy increases when the value of f (the ratio of magnetic field to nearest-neighbor interactions) increases, and for $f \geq 10$ approaches a constant value. For the values of $f \geq 0.2$, the finite-site free energy also increases, but slowly. The thermodynamic limit in which physical properties approach the bulk value is also explored.

Keywords: magnetic materials; nanostructures; surface free energy; finite-size free energy

1. Introduction

Since the discovery of single-molecule magnets in 1993 [1,2], the synthesis and physical characterization of molecular nanomagnets have been one of the most active fields in molecular magnetism. Caneschi et al. observed a slow relaxation of the magnetization in a magnetically isolated cobalt (II) nitronyl nitroxide chain [3] and described the main experimental requirements to be fulfilled in the design of such 1-D nanomagnets. One-dimensional (1-D) magnetic models have attracted much interest in recent years, because they are much easier to be treated theoretically than 2-D and 3-D models [4–7], and several quasi 1-D magnetic materials have been discovered. Critical behavior of the 2-D Ising antiferromagnets $K_2 \cdot CoF_4$ and Rb_2CoF_4 have been considered by Samuelsen [8]. Ising transition in a 1-D quarter-field electron system with dimerization has been considered by Tsuchiizu and Orignac [9]. The critical temperature in ferroelectric films described by a transverse spin-1/2 Ising model was studied using the effective field theory along with a probability distribution technique by Htoutou et al. [10].

Most of these systems can be interpreted in terms of Ising models including a nearest-neighbor (NN) exchange interaction whose sign determines the type of short-range order: e.g., ferromagnetic in $CsNiF_3$ and antiferromagnetic in $(CH_3)_4NMnCl_3$ [3].

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As a matter of fact, one may find a strong uniaxial Ising-type anisotropy and significant difference between intra-(J) and inter-chain (J') magnetic interactions in some materials. These magnetic nanowires are called single-chain magnets [11,12]. The dark-brown crystals obtained by assembling $[Mn(5\text{-}MeOsalen)(H_2O)]^+$ and $[(Tp)Fe(CN)_3]^-$ afford the 1-D zigzag chain $[(Tp)Fe(CN)_3Mn(5\text{-}MeOsalen)\cdot 2CH_3OH]_n$ [Tp^-=hydrotris(pyrazolyl) borate, 5-MeOsalen^{2-} = N,N′ ethylenebis (5-methoxyysalicylideneiminate)]. An alternating topology was presented with J_a and J_b Mn(III)-Fe(III) coupling parameters [13]. The anhydrous version of K-titanium alum, on the other hand, consists of layers of Ti^{3+} ions coordinated and interlinked to SO_4^{2-} anions, and provides a good realization of an $s = 1/2$ Ising model on the triangular lattice [14]. $KTi(SO_4)_2 \cdot H_2O$ is a frustrated chain with $s = 1/2$ with the NN interaction ($J = 9.462\,\text{cm}^{-1}$) and next-NN interaction J_1 with $\frac{J_1}{J} = 0.291$ [15]. Other real systems that map onto this model include Cu [2-(2-aminomethyl)] Br_2 ($J_1/J = 0.2$) [16], $(N_2H_5)CuCl_3$ ($J_1/J = 4$) [17,18], $SrCuO_2$ ($J_1/J < -10$) [19,20], $LiCu_2O_2$ ($J_1/J = -1$) [21,22], $LiCuVO_4$ ($J_1/J = -0.78$) [23], and Li_2CuZrO_4 ($J_1/J = -0.3$) [24].

We have previously applied the transfer matrix method to solve the 1-D Ising model in the presence of a magnetic field, taking both nearest and next-NN interactions into account [25]. We employed a numerical method to obtain the eigenvalues of the transfer matrix. Moreover, the heat capacity, magnetization, and magnetic susceptibility versus temperature for different values of the competition factor (the ratio of next-NN to NN interactions) were presented [25].

Surface and site effects are important in nanosystems and have a significant influence on the energy which may not be extensive [26]. Exact solution for the thermodynamic functions of the randomly diluted $s = 1/2$ NN Ising chain in a magnetic field was examined by Wortis [27] in which both site and bond impurities were treated. The system behaves nonanalytically at $T = h = 0$. The divergences of the pure-chain thermodynamics were replaced at nonzero dilution by essential singularities of the Griffiths type at which all functions are finite and infinitely differentiable [27]. Finite-size effect on the static properties of a single-chain magnet was studied by Bogani et al. They investigated the role of defects in the single-chain magnet CoPhOMe by inserting a controlled number of diamagnetic impurities [28]. Finite-size effect in $s = 1/2$ Ising systems showing slow dynamics of the magnetization was investigated by Bogani et al. [29] introducing diamagnetic impurities in a Co^{2+}-radical chain.

In this study, we have explored how the physics of 1-D Ising models is affected by the finite site of the system. We have specially studied effects of magnetic field on the surface free energy and the finite-site free energy.

2. Calculation of surface energy and finite-site free energy

2.1. *NN interactions in the presence of a magnetic field*

The partition function, Z, for the 1-D Ising model with the NN interactions in the presence of a magnetic field in free boundary condition may be written as (see [30] for more details):

$$Z = \lambda_1^N\left(\left(\sin^2 2\phi \sinh(K)\left(1 - \frac{\lambda_2}{\lambda_1}\right)\right) + e^{-K}\right) + \lambda_1^N\left(\left(\sin^2 2\phi \sinh(K)\left(1 - \frac{\lambda_1}{\lambda_2}\right)\right) + e^{-K}\right)\left(\frac{\lambda_2}{\lambda_1}\right)^N, \quad (1)$$

where

$$\lambda_1 = e^K\left(\cosh(h) + \sqrt{\sinh^2(h) + e^{-4K}}\right), \tag{2}$$

and

$$\lambda_2 = e^K\left(\cosh(h) - \sqrt{\sinh^2(h) + e^{-4K}}\right).$$

Therefore, the free energy, A, for the model in the presence of a magnetic field is

$$A = -Nk_BT\ln(\lambda_1) - k_BT\ln\left(\sin^2 2\phi\sinh(K)\left(1-\frac{\lambda_2}{\lambda_1}\right) + e^{-K}\right)$$
$$- k_BT\ln\left(1 + \frac{\sin^2 2\phi\sinh(K)(1-\frac{\lambda_1}{\lambda_2}) + e^{-K}}{\sin^2 2\phi\sinh(K)(1-\frac{\lambda_2}{\lambda_1}) + e^{-K}}\left(\frac{\lambda_2}{\lambda_1}\right)^N\right), \tag{3}$$

where $K = J/kT$, s, and $h = H/kT$ are the reduced spin–spin NN coupling energy, the spin of a site, and the reduced magnetic field, respectively. Also, k_B is the Boltzmann constant and T the absolute temperature.

The Helmholtz free energy can be partitioned into three components: bulk A_{bulk}, surface A_{surface}, and finite-site $A_{\text{finites size}}$, which are given as [30]:

$$A_{\text{bulk}} = -Nk_BT\ln(\lambda_1), \tag{4}$$

$$A_{\text{surface}} = -k_BT\ln\left(\sin^2 2\phi\sinh(K)\left(1-\frac{\lambda_2}{\lambda_1}\right) + e^{-K}\right), \tag{5}$$

$$A_{\text{finite size}} = -k_BT\ln\left(1 + \frac{\sin^2 2\phi\sinh(K)\left(1-\frac{\lambda_1}{\lambda_2}\right) + e^{-K}}{\sin^2 2\phi\sinh(K)\left(1-\frac{\lambda_2}{\lambda_1}\right) + e^{-K}}\left(\frac{\lambda_2}{\lambda_1}\right)^N\right), \tag{6}$$

where Equation (4) is the bulk free energy which scales linearly with the sites numbers of system. Equation (5) is the surface free energy which is independent of site number. Equation (6) (the finite-size free energy) does depend on the site number of the system. If $\lambda_2 < \lambda_1$, then, by defining the parameter a,

$$a = \frac{\sin^2 2\phi\sinh(K)\left(1-\frac{\lambda_1}{\lambda_2}\right) + e^{-K}}{\sin^2 2\phi\sinh(K)\left(1-\frac{\lambda_2}{\lambda_1}\right) + e^{-K}}, \tag{7}$$

the third term on the right-hand side of Equation (5) may be approximately written as:

$$-k_BT\ln\left(1 + \frac{\sin^2 2\phi\sinh(K)\left(1-\frac{\lambda_1}{\lambda_2}\right) + e^{-K}}{\sin^2 2\phi\sinh(K)\left(1-\frac{\lambda_2}{\lambda_1}\right) + e^{-K}}\left(\frac{\lambda_2}{\lambda_1}\right)^N\right) \approx -k_BTa\left(\frac{\lambda_2}{\lambda_1}\right)^N$$

$$= -k_BTa\left(1 - \frac{2\sqrt{\sin^2 h + e^{-4K}}}{\cosh(\text{h}) + \sqrt{\sinh^2(h) + e^{-4K}}}\right)^N = -k_BTa\left(1 - \frac{\frac{2}{\sin(2\phi)}}{e^{2K}\cosh(h) + \frac{1}{\sin(2\phi)}}\right)^N$$

$$= -k_BTa\left(1 - \frac{2}{1 + e^{2K}\cosh(h)\sin(2\phi)}\right)^N.$$

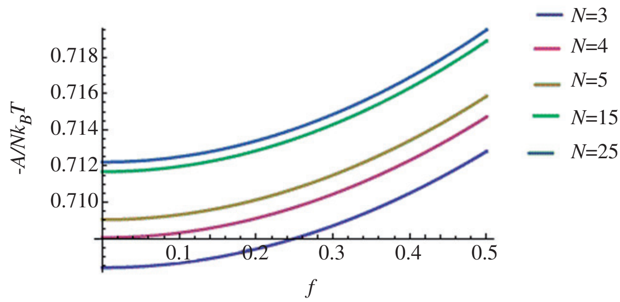

Figure 1. Reduced free energy vs. f (the ratio of magnetic-field/spin interaction energy over the nearest-neighbor spin–spin coupling energy) for $N = 3$, 4, 5, 15, and 25 in the case of NN interactions.

If N is large and also

$$\frac{1}{1 + e^{2K}\cosh(h)\sin(2\phi)} \to 0,$$

then

$$-k_B Ta\left(1 - \frac{2}{1 + e^{2K}\cosh(h)\sin(2\phi)}\right)^N \approx -k_B Ta \exp\left(-\frac{2N}{1 + e^{2K}\cosh(h)\sin(2\phi)}\right).$$

Therefore,

$$A_{\text{finite size}} \approx -k_B Ta \exp\left(-\frac{2N}{1 + e^{2K}\cosh(h)\sin(2\phi)}\right). \tag{8}$$

3. Results and discussion

3.1. *Thermodynamic limit*

The exact reduced Helmholtz free energy of 1-D Ising model in the presence of a magnetic field considering the NN interactions can be obtained for any size of system in terms of the parameter f (ratio of spin magnetic field interaction energy to spin-spin coupling energy). Figure 1 indicates that the reduced free energy increases with N (system size) but the increment slows down for larger values of N. As may be expected, the free energy approaches the bulk value when N becomes large. In the thermodynamic limit, the reduced free energy given by Equation (3) equals ln(λ1). On the other hand, small eigenvalue of transfer matrix is important in free energy calculation of the finite-size system. Such an eigenvalue may give a significant contribution to the free energy; therefore, it could not be ignored in our calculation. We can easily calculate the relative difference in total free energy for the finite model and the bulk (ΔA) which is shown in Figure 2. Relative difference in free energy is defined as $\Delta A = \frac{(A_N - A_{\text{bulk}})}{A_{\text{bulk}}}$ for a finite Ising model. A_N is free energy per site for the finite spin chain. On the basis of Figure 2, the relative difference increases with f, as expected for large values of N. On the basis of Figure 2, relative difference (ΔA) in $f = 0$ for site $N = 25$ is 0.001. It increases by increasing the value of f and

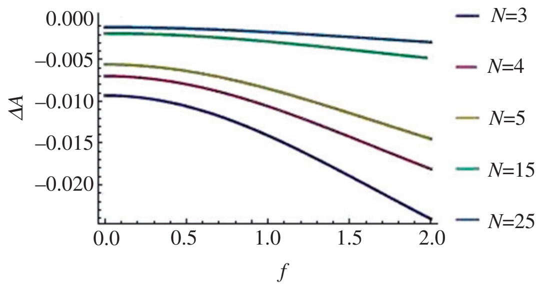

Figure 2. Relative difference in free energy $\Delta A = \frac{(A_N - A_{\text{bulk}})}{A_{\text{bulk}}}$ for a finite Ising model with $N = 3$, 4, 5, 15, and 25 with respect to the bulk as a function of the parameter f in the case of NN interactions.

becomes 0.003 in $f = 2$. When the number of sites constructing the system is limited, finite-size effect seems important as the deviation of the free energy from the bulk value is large. In fact, in such a case, edge effects have a significant impact in deviation of the free energy from the bulk value. For spins at the boundary, the number of spin–spin interactions is not the same as for the other spins in the chain. Increasing the number of sites in the spin chain will result in a decrease of edge effect because of the decrease in the ratio of the number of boundary spins to the total number of spins of the system.

3.2. *Nonextensive thermodynamics*

In spite of multifractal concepts, Tsallis [31] has proposed a generalization of the Boltzmann–Gibbs (BG) statistical mechanics. He introduced an entropic expression characterized by an index q which leads to a nonextensive statistics,

$$S_q = k \frac{1 - \sum_{i=1}^{W} p_i^q}{q - 1}, \tag{9}$$

where p_i are the probabilities associated with the microscopic configurations and W is the total number of the configurations. The value of q is a measure of the nonextensivity of the system; $q = 1$ corresponds to the standard, extensive, BG statistics. Indeed, using $p_i^{q-1} \approx 1 + (q-1)\ln(p_i)$ in the limit $q \to 1$, Equation (9) is converted to

$$S_1 = k \sum_{i=1}^{W} p_i \ln(p_i). \tag{10}$$

The novelty of the statistical entropy (9) is that it does not satisfy additivity, for example for two systems A and B described by independent probability distributions [32]

$$S(A+B) = S(A) + S(B) + (1-q)S(A)S(B), \tag{11}$$

where $(1-q)S(A)S(B)$ is the nonextensivity term of calculation as for BG statistics is zero. Let us consider a magnetic system with N spins following a d-D Ising model with long-range interaction potential Hamiltonian [31]:

$$H = J \sum_{i,j} \frac{J}{r_{i,j}^{\alpha}} s_i s_j, \tag{12}$$

where J is the exchange coupling constant ($J > 0$), $r_{i,j}$ is the distance between the spins i and j, s_i assumes the values ± 1, and α is the range of the interaction ($0 \leq \alpha < \infty$). The internal energy per spin of the system is calculated by integrating Equation (12) over the volume

$$\frac{E}{N} \cong \int_1^{N^{1/d}} \frac{r^{d-1}}{r^\alpha} \mathrm{d}r = \frac{1}{d} \frac{N^{1-\frac{\alpha}{d}} - 1}{1 - \frac{\alpha}{d}}. \tag{13}$$

For large system, the energy per spin is given by (see [31] for more details):

$$\frac{E}{N} \cong \left\{ \begin{array}{l} (\alpha - d), \frac{\alpha}{d} > 1 \\ \ln(N) \frac{\alpha}{d} = 1 \\ N^{1-\frac{\alpha}{d}} \frac{\alpha}{d} < 1 \end{array} \right\}. \tag{14}$$

When $0 \leq \frac{\alpha}{d} \leq 1$ for $N \to \infty$, energy per spin does not converge. Convergence of energy can be achieved by introducing an auxiliary N^* parameter in terms of $\frac{E}{NN^*}$ [31]. For 1-D system, N^* can be defined by the following equation [33]:

$$N^* = \left\{ \begin{array}{ll} \frac{N^{1-\alpha}}{1-\alpha} & 0 \leq \alpha < 1 \\ \ln(N) & \alpha = 1 \\ \frac{1}{\alpha - 1} & \alpha > 1 \end{array} \right\}. \tag{15}$$

To make the internal energy behave extensively and achieve its convergence in the thermodynamic limit, we can write the internal energy as $U(N) = NN^* U_1$. The extensive property imposes observables to be a linear homogeneous function of N and $U(\lambda N) = \lambda U(N)$. However, when long-range interactions are included, this property is violated, and it is easy to show that thermodynamic functions are homogeneous of degree $1 + |\alpha - 1|$ for $\alpha < 1$ [33].

In addition, we expect that the internal energy of a magnetic system with long-range interactions adopts to the following form: $U(S, M, N) = NN^* U_1(S/N, M/N)$, where U_1 is a function per particle of the entropy S and magnetization M [33].

By definition of some extensive quantity such as Gibbs free energy ($G^* = \frac{G}{NN^*} = \frac{U}{NN^*} - \frac{T}{N^*}\frac{S}{N}\frac{H}{N^*}\frac{M}{N}$), internal energy ($U^* = \frac{U}{N^*}$), and some intensive quantities, such as temperature ($T^* = \frac{T}{N^*}$) and magnetic field ($H^* = \frac{H}{N^*}$), thermodynamic quantities will converge [33]. On the other hand, by such definitions, we will have extensive quantities, such as internal energy, and free energy, behave extensively and intensive quantities, such as temperature and magnetic field behave intensively.

The free energy for $N = 3$ shows a large deviation from bulk free energy even in zero magnetic field. In fact, in finite-size system, the free energy per site (see Figure 1) is not the same for each size and all graphs do not coincide. It means that the free energy in finite size is not an extensive quantity with definition of some variables such as f, h, J, and T for Hamiltonian which is defined as

$$H = \sum_i J s_i s_{i+1} + h \sum_i s_i. \tag{16}$$

On the basis of Figure 1, deviation of the free energy from the bulk value in finite size is large. It may be concluded that finite size has a significant effect on system that leads the free energy to behave as a nonextensive quantity. On increasing the number of sites in finite-size system, all the free energy per site coincides with each other and the free energy becomes an extensive quantity. Nonextensivity of the free energy for finite-size Ising model in the presence of a magnetic field comes from some sources, such as the surface free energy and the finite-size free energy. According to Equation (3), the total free energy of finite spin chain has three terms. The first term is linearly scaled by number of spin in the system (N). The second and third terms are the surface free energy and finite-size free energy contributions. If we take into account just the first term of free energy and neglect the other parts, we expect free energy of the system to become extensive. Such a conclusion is on the basis that the first term is linearly scaled by the number of spins in the system (N) and could be considered as the bulk free energy. Since the surface free energy and the finite-size free energy of the system (the second and third parts of Equation (3)) do not scale by number of spin, they give a significant contribution to the nonextensivity of the system's free energy.

3.3. *Surface energy*

Energy is needed to close the ends of 1-D spin chain. In periodic boundary condition, the ends of spin chain are closed but there is no connection between the ends of spin chain in free boundary condition. Difference of the free energy between the two mentioned cases (closed and opened ends) is the surface free energy. The surface free energy depends on magnetic field and spin coupling interaction. The surface energy increases by increasing the magnetic field as well as spin coupling interaction. The reduced surface free energy, $A_{\text{surface}}/k_B T$, is given by Equation (4) for the model in the presence of a magnetic field, considering only the NN interactions. On the basis of Equation (4), $A_{\text{surface}}/k_B T$ is independent of N. Intuitively, the energy that is needed to connect the ends of chain does not depend on the length of the spin chain. In 1-D of spin chain, there is just two spins in the ends of the chain; therefore, the surface energy is completely independent of number of spin in the chain. Figure 3 shows $A_{\text{surface}}/k_B T$ as a function of parameter f. It increases with f and finally approaches a constant value. Increase in f means that interaction energy of ending spins in the spin chain with magnetic field is going to be high; therefore, we can conclude that the system is going to be more stable ($-A_{\text{surface}}/k_B T$ is increasing) and then, it approaches an asymptotic limit. In fact, with increasing f, the surface free energy will finally be saturated. On increasing f, all spins will be aligned parallel to the magnetic field, after which the magnetic field will not change the surface free energy. As a matter of fact, the surface free energy does not depend on the size of system in 1-D of Ising model. In zero magnetic field, $f = 0$, the reduced surface free energy is small and its value is 0.01. On increasing the magnetic field from $f = 10$, the reduced surface free energy becomes 0.20 without major changes after that. As a matter of fact, the surface free energy in high magnetic field has a significant contribution to the nonextensivity of the free energy (see Figure 3).

3.4. *Finite-size free energy*

There is one part in the free energy which depends on the size of the system. Such part is independent of boundary condition and is the so-called finite-size free energy. Solving the transfer matrix of 1-D Ising model, one has two eigenvalues. One of them is bigger than the other one. In the thermodynamic limit, we can neglect the smaller one. But, in finite-size systems, the two eigenvalues should be taken into account for computation of the free energy. The finite-size free energy decreases by the size of system exponentially and neglecting the smaller eigenvalue does not affect the validity of the free energy.

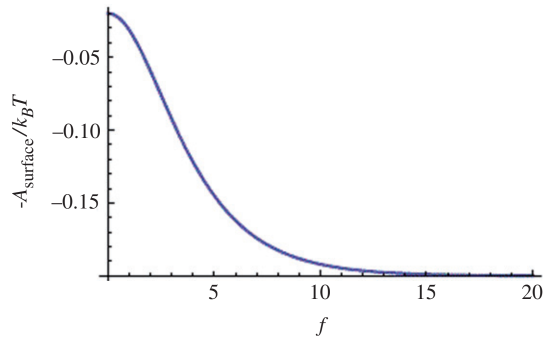

Figure 3. Reduced surface free energy as a function of the parameter f in the case of NN interactions.

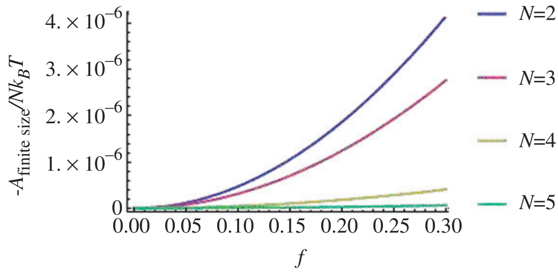

Figure 4. Reduced finite-site free energy versus the parameter f when $N=2$, 3, 4, and 5 in the case of NN interactions. The free energy for $N=6$ is so small that is not shown.

The finite-size free energy is obtained analytically which is given by Equation (6). The finite-size free energy, $A_{\text{finite site}}$, given by Equation (6), versus f is plotted in Figure 4 for $N=3$, 4, 5, and 6. According to Figure 4, $A_{\text{finite site}}$ vanishes for large value of N. For small values of N, the contribution of the finite-size free energy seems to be significant, which becomes especially more important for stronger magnetic field. In fact, interaction energy between spins and magnetic field energy of the system increases; therefore, the free energy in finite size in high magnetic field approaches bulk properties in enormous sites.

4. Conclusion

A finite-size 1-D Ising model is investigated in the presence of a magnetic field. The transfer matrix method is used for obtaining some physical properties, such as the surface free energy, the finite-site free energy, and deviation of the free energy of finite system from the bulk value as a function of size, analytically. The reduced total free energy per site is plotted as a function of f; increasing size of system will result in reduction of differences of free energy plots and let them be close to each other (Figure 1). Relative difference in free energy $\Delta A = \frac{(A_N - A_{\text{bulk}})}{A_{\text{bulk}}}$ for a finite Ising model with $N=3$, 4, 5, 15, and 25 with respect to the bulk value as a function of the parameter f is also obtained (Figure 2).

On the basis of Figure 2, increasing size of the system will lead it to approach bulk value. The surface energy in 1-D Ising model in the presence of a magnetic field is obtained analytically as a function of *f*. Results of surface free energy as a function of the *f* parameter are shown in Figure 3. According to Figure 3, in low magnetic field, the surface free energy has a low value and it increases when the magnetic field increases and finally, it approaches a constant value. Increasing the parameter *f* may lead the finite-size free energy to increase (Figure 4). At small values of the parameter *f*, the finite-size free energy has a low value and it increases by increasing the magnetic field. We have also shown that the surface free energy and the finite-size free energy are the most important parts that lead the free energy to be nonextensive in finite-size systems.

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