Correlation between Curie temperature and system dimension

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Abstract

Curie temperature $T_{\rm C}$ of spin arrangement with arbitrary dimension was considered. We assumed that interaction of a spin with all other spins vary with a power-law decay rate in exchange integral on Heisenberg model. As a result, we found that $T_{\rm C}$, which was obtained from $T_{\rm C} = \lambda C$ (λ: mean-field coefficient and C: Curie constant), significantly depends on fractal dimension of spin arrangements D, the exchange integral and the decay constant. This semi-quantitavely explains how $T_{\rm C}$ depends on D ($1 \leq D \leq 3$) in a universal way and also the finite size effect on $T_{\rm C}$ in low-dimensional spin systems.

Key words: Curie temperature, low-dimensional spin system, finite size effect, fractal dimensions of spin arrangements and lattices, Heisenberg model, mean-field theory

1. Introduction

Magnetic properties of nanoparticles and ultrathin films of ferromagnets and antiferromagnets, i.e., low-dimensional spin systems, have been intensively investigated because of their importance in fundamental physics and applications. Finite size effect on Curie temperature $T_{\rm C}$ and Néel temperature $T_{\rm N}$ is one of the unique magnetic properties of the low-dimensional spin systems [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28]. For example, the shift of $T_{\rm C}$ from ca. 600 K to

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ca. 50 K is observed with the decrease of the thickness of ultrathin Ni films [4, 5, 6, 7, 8], while T_N in CoO layers is suppressed from 300 K to 15 K [21]. The suppressions of T_C and T_N have been discussed in terms of scaling laws of the critical temperatures in bulk samples, correlation length and system size (particle diameter and film thickness) [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25].

On the other hand, the origin of critical temperatures (T_C and T_N) can be understood by Heisenberg and Ising models [29]. In Heisenberg model, the interaction between a spin and its nearest neighbors determines T_C and T_N. In Ising models, T_C can be analytically obtained from series expansion in magnetic susceptibility χ with respect to $\tanh(J/kT)$, where J, k and T are the interaction energy between neighboring two spins, the Boltzmann constant and temperature, respectively, and the series expansion reflects system dimension and lattice type of unit cell. From the series expansions for different models, it has been found that T_C in three-dimensional (3D) spin system is higher than that in two-dimensional (2D) spin system, while there is no T_C in one-dimensional (1D) spin system. This approach is suitable for determination of the precise T_C's for 1D, 2D and 3D systems, however, unsuitable to solve a fundamental problem how T_C directly relates to system dimension through a non-integer ("*fractal*") dimension such as self-similar sponge-like spin arrangements because it is very difficult to obtain the series expansions in χ for non-integer spin systems as dimension-dependent functions [30, 31]. To understand this problem, a semi-quantitative approach would be helpful. It may also give us a general understanding of the finite size effect on the critical temperatures in low-dimensional spin system and related phenomena. Our purpose in this article is to catch an essence of the relation between T_C and system dimension. To solve the problem semi-quantitatively, we adopted Heisenberg model with fractal spin arrangements. Here, "fractal spin arrangement" means the ferromagnetic system with ideal spin distribution described by fractal dimension D.

2. Heisenberg model with fractal spin arrangements

The fractal ferromagnetic system is modeled as follows. We consider two kinds of dimensions, which are independent parameters of each other. The lattice has a dimension d ($d = 1$, 2 and 3), while the spin arrangement has a dimension D ($1 \leq D \leq 3$). The spin-spin interaction is assumed to spread out over the system with a power-law decay rate as discussed later. Figure 1

(a) illustrates an example of the spin arrangement on the square lattice. In Fig. 1 (a), the distance between the origin and a site is defined as a radius j ($j = 1, 2, 3, \ldots$). Figure 1 (b) illustrates the arrangement of the spins $\vec{S}_{jk}$ ($k = 1, 2, 3, \ldots, n_j$) within the zone between the distances $j-1$ and j. The number of $\vec{S}_{jk}$ is defined as n_j, which is formulated later as a function of j and D. In particular, n_1, the number of nearest neighboring site, is denoted by z. Through the model in Fig. 1 (a), we will consider a sponge structure in cubic lattice as shown in Fig. 2 (d) later.

In the mean-field theory, T_C is defined as

$$T_\mathrm{C} = \lambda C \tag{1}$$

where λ is a mean-field coefficient and C is the Curie constant. If we could relate λ to D, we could determine how T_C depends on D. First, let us consider the exchange energy to evaluate λ from the Heisenberg model. The exchange energy between the spin at the origin ($\vec{S}_0$) and all other spins ($\vec{S}_{jk}$), E_ex, is

$$E_\mathrm{ex} = -2\sum_{j=1}^{\infty}\sum_{k=1}^{n_j} J_{0j}\vec{S}_0 \cdot \vec{S}_{jk} = -2\sum_{j}\sum_{k} J_{0j} S_0 \cdot S_{jk} \tag{2}$$

where J_{0j} is the average exchange integral between $\vec{S}_0$ and the spins at the distance j ($\vec{S}_{jk}$'s). S_{jk} in Eq. (2) can be summarized as

$$\sum_{k} S_{jk} = n_j \langle S_{jk} \rangle \tag{3}$$

where n_j is the number of $\vec{S}_{jk}$ and $\langle S_{jk} \rangle$ is the average of S_{jk}. Here, n_j should be considered because it depends on D. The number of sites in the range between the distances $j - \Delta j$ and j, $\Delta N(j)$, is

$$\Delta N(j) = N(j) - N(j - \Delta j) \tag{4}$$

where $N(j)$ and $N(j - \Delta j)$ are the total number of the sites within the distances j and $j - \Delta j$, respectively. Next, $N(j)$ should be formulated in terms of D. In spin systems with $D = 1$, 2 and 3, the number of sites within the radius j from the origin, $N(j)$, is proportional to j^D. Therefore, $N(j)$ in D-dimensional spin system is assumed [32]

$$N(j) = aj^D \tag{5}$$

where a is a proportional constant. Eq. (4) would be approximated as

$$\Delta N(j) = aj^D - a(j-\Delta j)^D \approx aDj^{D-1}\Delta j \tag{6}$$

Therefore, the increasing rate of the site number from the distance $j - \Delta j$ to the distance j, which corresponds to n_j, is

$$n_j = \frac{\Delta N(j)}{\Delta j} = aDj^{D-1} = zj^{D-1} \tag{7}$$

where aD is determined to be z from the initial condition of $n_1 = z$. Therefore, Eq. (2) becomes

$$E_{\rm ex} = -2z \sum_j J_{0j} j^{D-1} S_0 \langle S_{jk} \rangle \tag{8}$$

Next, let us consider J_{0j} to treat the interaction between $\vec{S}_0$ and $\vec{S}_{jk}$'s. Here, we assume that J_{0j} is directly related to the average path length (the number of steps) from $\vec{S}_{1k}$ to $\vec{S}_{jk}$. Let us discuss the path length in two-dimensional lattice first. Fig. 2 (a) shows two-dimensional spin distribution (dimensionality of spin arrangement $D = 2$) on square lattice (dimensionality of lattice $d = 2$). Now we consider the path length from the origin to $\vec{S}_{jk}$. There are different paths. The path lengths with most and least steps from the origin, $L_{0j}^{d=2}$ and $l_{0j}^{d=2}$, are approximately $\sqrt{2}j$ and j, respectively. The average path length from the origin to $\vec{S}_{jk}$, $l_{0j,\rm av}^{d=2}$, is $\approx (1+\sqrt{2})j/2$. The average path length from $\vec{S}_{1k}$ to $\vec{S}_{jk}$, $l_{1j,\rm av}^{d=2}$, is approximated to be $(1+\sqrt{2})j/2-1$. On the other hand, Figure 2 (b) illustrates a schematic representation of a fractal ferromagnet ($D < 2$) on square lattice ($d = 2$). There are still different paths. Similarly, $L_{0j}^{d=2}$, $l_{0j}^{d=2}$, $l_{0j,\rm av}^{d=2}$ and $l_{1j,\rm av}^{d=2}$ are $\sqrt{2}j$, j, $(1+\sqrt{2})j/2$ and $(1+\sqrt{2})j/2-1$, respectively.

Now let us consider the cases of the cubic ($D = 3$) and fractal spin arrangements ($D < 3$) in cubic lattices ($d = 3$) as shown in Figs. 2(c) and 2(d), respectively. Similarly, the path lengths with most and least steps and the average path length from the origin to $\vec{S}_{jk}$, $L_{0j}^{d=3}$, $l_{0j}^{d=3}$ and $l_{0j,\rm av}^{d=3}$, are approximately $\sqrt{3}j$, j and $(1+\sqrt{3})j/2$, respectively. Therefore, average path length from $\vec{S}_{1k}$ to $\vec{S}_{jk}$ $l_{1j,\rm av}^{d=3}$, is

$$l_{1j,\rm av}^{d=3} \approx \frac{(1+\sqrt{3})j}{2} - 1 = 1.37j - 1 \tag{9}$$

Here, let us assume that J_{0j} should be approximated as

$$J_{0j} \approx \alpha^{1.37j-1} J_1 \tag{10}$$

where α is a decay constant ($\alpha < 1$) [33], J_1 is the exchange integral between $\vec{S}_0$ and one of the nearest neighbor sites ($\vec{S}_{1k}$). Accordingly, Eq. (8) becomes

$$E_{\text{ex}} = -2z \sum_j \alpha^{1.37j-1} j^{D-1} J_1 S_0 \langle S_{jk} \rangle \tag{11}$$

On the other hand, the energy arising from molecular magnetic field E_{m} is

$$E_{\text{m}} = -g\mu_{\text{B}} S_0 B_{\text{m}} \tag{12}$$

where g is the g factor and μ_{B} is the Bohr magneton. E_{m} should be equal to E_{ex}. From Eqs. (11) and (12),

$$B_{\text{m}} = \frac{2z \sum_j \alpha^{1.37j-1} j^{D-1} J_1 \langle S_{jk} \rangle}{g\mu_{\text{B}}} \tag{13}$$

Since $B_{\text{m}} = \lambda M$ and $M = Ng\mu_{\text{B}} \langle S_{jk} \rangle$, the mean-field constant λ is obtained as

$$\lambda = \frac{2z(\sum_j \alpha^{1.37j-1} j^{D-1}) J_1}{Ng^2\mu_{\text{B}}^2} \tag{14}$$

On the other hand, C is

$$C = \frac{Ng^2\mu_{\text{B}}^2 S(S+1)}{3k_{\text{B}}} \tag{15}$$

where S is the spin momentum and k_{B} is the Boltzmann constant. T_{C} is obtained from the relation $T_{\text{C}} = \lambda C$ as

$$T_{\text{C}}(D) = \frac{2z(\sum_j \alpha^{1.37j-1} j^{D-1}) J_1 S(S+1)}{3k_{\text{B}}} \tag{16}$$

Here, let us discuss the dependence of $\Sigma_j \alpha^{1.37j-1} j^{D-1}$ on D. If we consider that the interaction is spread out over a large system, it can be described as

$$\sum_j \alpha^{1.37j-1} j^{D-1} \approx \frac{\Gamma(D)}{\alpha(-1.37 \ln \alpha)^D} \tag{17}$$

where $\Gamma(D)$ is the Gamma function. Therefore,

$$T_{\mathrm{C}}(D) = \frac{2z\Gamma(D)}{\alpha(-1.37\ln\alpha)^{D}}\frac{J_1 S(S+1)}{3k_{\mathrm{B}}} \tag{18}$$

Moreover, the normalized Curie temperature $T_{\mathrm{C}}/T_{\mathrm{C}}^{\mathrm{bulk}}$ is useful to discuss the dependence of T_{C} on D in comparison with $T_{\mathrm{C}}^{\mathrm{bulk}}$, where $T_{\mathrm{C}}^{\mathrm{bulk}}$ is the T_{C} of bulk sample (3D).

$$\frac{T_{\mathrm{C}}(D)}{T_{\mathrm{C}}^{\mathrm{bulk}}} = \frac{(-1.37\ln\alpha)^{3-D}\Gamma(D)}{\Gamma(3)} \tag{19}$$

3. Results and discussion

Figure 3 shows the dependence of $T_{\mathrm{C}}/T_{\mathrm{C}}^{\mathrm{bulk}}$ on D with various α. First, note that T_{C} is suppressed from 3D to 1D. T_{C} is significantly suppressed from 3D to 2D, especially. It is also shown that the suppression of T_{C} is remarkable at larger α. For example, on going from 3D to 1D, $T_{\mathrm{C}}/T_{\mathrm{C}}^{\mathrm{bulk}} \sim 0$ at $\alpha \sim 1$. This is consistent with the exact solution in 1D Ising model ($T_{\mathrm{C}} = 0$) [29]. Contrary to this, T_{C} in 1D with $\alpha = 0.6$ is of the order of ca. 25 % of $T_{\mathrm{C}}^{\mathrm{bulk}}$, which contradicts with the exact solution of 1D Ising model. The dependence of T_{C} on α would be interpreted as follows. When α is larger, then the spin-spin interaction is relatively stronger and long-range magnetic order occurs. Therefore, T_{C} with larger α is sensitive to the spin arrangement dominating D. Contrary to this, long-range magnetic order does not occur under smaller α (weak interaction between spins) and the dependence of T_{C} is insensitive to D. Comparing this theory with Ising model, the theory would be reliable at $\alpha \sim 1$.

On the other hand, the theory phenomenologically explains the dependence of T_{C} on D in the experimental results of finite effect on T_{C} in ultrathin ferromagnetic films. Here, it is possible to discuss D in the ultrathin films with respect to a ratio between film thickness and a characteristic length emerged in critical phenomena such as ξ_0, where ξ_0 is defined by $\xi(T) = \xi_0(|T - T_{\mathrm{c}}|/T_c)^{-\nu}$ ($\xi(T)$: the correlation length at T, ξ_0: the correlation length extrapolated to $T = 0$, T_{c}: critical temperature, ν: a critical exponent. $T_{\mathrm{c}} = T_{\mathrm{C}}$ in this case). Thin film would be close to 3D if the film thickness t is significantly larger than ξ_0. On the other hand, it would be close to 2D if t is comparable to ξ_0. In fact, dimensional crossover between

3D ($t > \xi_0$) and 2D ($t \leq \xi_0$) was experimentally reported in ultrathin Ni films [5, 6].

Let us notice the experimental results of finite size effect. Various results have been obtained using Ni, Gd, Co, etc. The experimental results of ξ_0, $T_{\rm C}(\xi_0)$ and $T_{\rm C}^{\rm bulk}$ of typical ferromagnets are summarized in Table 1, where $T_{\rm C}(\xi_0)$ is the $T_{\rm C}$ at $t = \xi_0$. It is obvious that there is a scattering in $T_{\rm C}(\xi_0)$ of same materials because the magnetic properties depend on the growth conditions of the ultrathin films. However, the general tendency between $T_{\rm C}$ and t could be summarized that $T_{\rm C}$ is closer to $T_{\rm C}^{\rm bulk}$ in $t \geq 5\xi_0$ ($D \sim 3$), and reduced to 10 - 50 % in $t \sim \xi_0$ ($D \sim 2$). The theory phenomenologically explains the experimental tendency by varying D and α. For example, $T_{\rm C}$ is suppressed to at least 10 $\sim$ 40 % of $T_{\rm C}^{\rm bulk}$ from 3D to 2D in $\alpha = 0.6 \sim 0.9$ as shown in Fig. 3.

4. Conclusion

In conclusion, we have discussed the fundamental problem on how $T_{\rm C}$ systematically changes with the fractal dimension D of ferromagnets based on the Heisenberg model. If we introduce a decay constant α and use the Curie constant C, $T_{\rm C}$ can be formulated as a function of α, D, J_1 and S. Moreover, the $T_{\rm C}/T_{\rm C}^{\rm bulk}$ obtained from the formula phenomenologically explains the experimental results in low-dimensional ferromagnets, and may be extended to the discussion on $T_{\rm N}$ in antiferromagnets. Recently, we have prepared Menger sponge-like fractal bodies; fractal porous silica with $D =$ 2.5 - 2.7 with a pore size within the range of 50 nm - 30 μm [35, 36, 37]. We are now preparing fractal antiferromagnets of transition metal oxides. Such fractal magnetic samples should be suitable for experimental investigations on the correlation between $T_{\rm C}$, $T_{\rm N}$ and D, and for studying critical phenomena in fractal dimension. Furthermore, $T_{\rm C}$ of fractal spin system and the dependence of $T_{\rm C}$ on D should be precisely determined in further studies.

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[33] Here, α means the spin-spin correlation between neighboring two spins except $\langle \vec{S}_0 \cdot \vec{S}_{1k} \rangle$. It plays a similar role to $\tanh(J/kT)$, the spin-spin correlation in Ising model.

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Table 1: ξ_0, $T_C(\xi_0)/T_C^{bulk}$ and T_C^{bulk} of typical ferromagnets.

Ferromagnet	ξ_0	$T_C(\xi_0)/T_C^{bulk}$(%)	T_C^{bulk} (K) [34]	Ref.
Ni	2 ML*	< 40	672	[6]
	4 ML	50-70		[7]
	4.7 ML	40		[4, 7, 9]
	3.4 ML	35		[5]
	5 ML	50		[4]
Gd	13 $\AA$	17	292	[10]
	4 ML	85		[12]
	22 ML	95		[20]
	8.6 ML	50		[4]
Fe	2.3 ML	30	1043	[4]
Co	2.2 ML	20	1388	[4]
$CoNi_3$	3.8 ML	40	—	[5]

*ML: monolayers

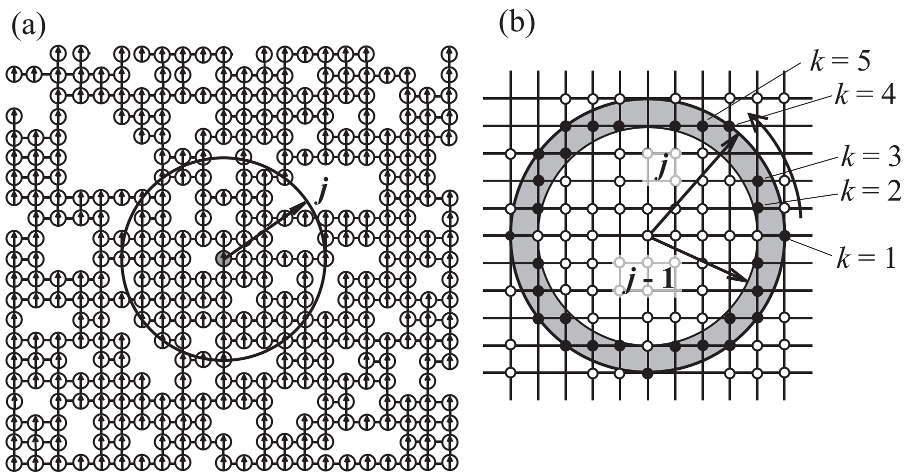

Figure 1: (a) A schematic representation of a fractal ferromagnet, which illustrates a sponge structure. The gray site represents $\vec{S}_0$ at the origin. (b) Spin arrangement of $\vec{S}_{jk}$ in the zone between the distances $j-1$ and j (the closed circles) and the spins out of the area (the opened circles), where the spin arrangement is based on Fig. 1 (a).

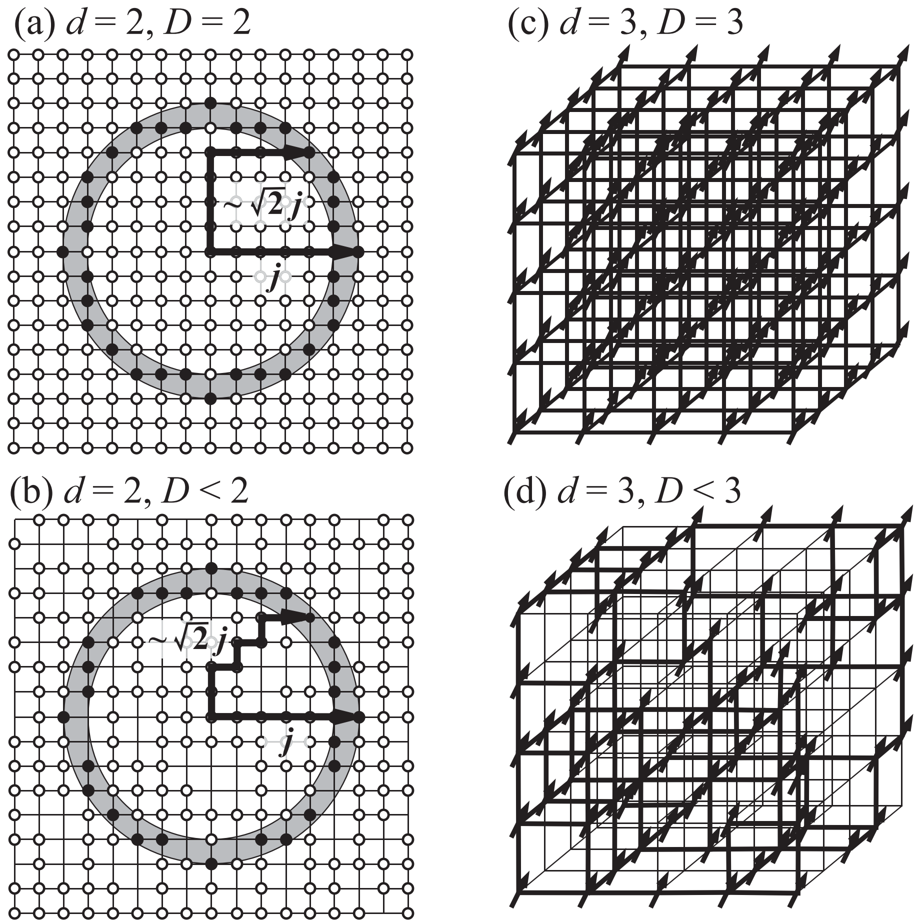

Figure 2: Schematic illustrations of spin arrangements in square spin systems without (a) and with spin defects (some sites have no spins) (b) (a "fractal ferromagnet"). The dots represent spin sites. Dimensionality of lattice d is 2 in both cases, however, dimensionalities of spin arrangements (D) are 2 (a) and < 2 (b), respectively. The closed and opened circles represent the spins in and out of the range between the distances $j-1$ and j, respectively. The arrows indicate paths from the origin to $\vec{S}_{jk}$'s with most and least steps. In (c) and (d), cubic ($D = 3$) and fractal spin arrangements ($D < 3$) in cubic lattices ($d = 3$) are illustrated, respectively.

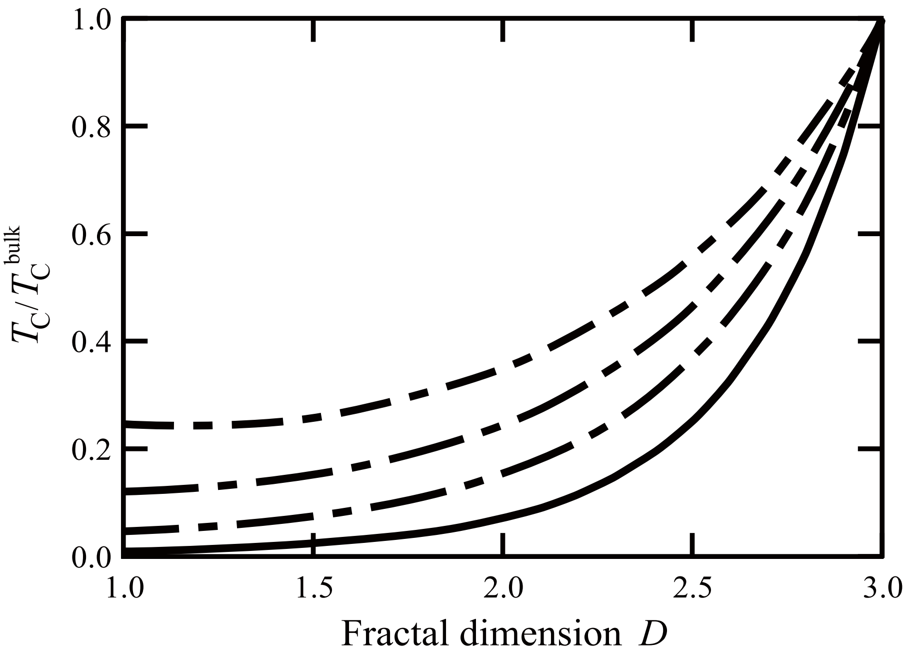

Figure 3: Dependence of normalized Curie temperature $T_{\mathrm{C}}/T_{\mathrm{C}}^{\mathrm{bulk}}$ on fractal dimension D with different α, where $T_{\mathrm{C}}^{\mathrm{bulk}}$ is equal to T_{C} in 3D and α is 0.6, 0.7, 0.8 and 0.9 from top to bottom, respectively.