

Curie Temperature of Low-dimensional Ferromagnetic Material

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ABSTRACT

This paper attempts to develop a unified theoretical model to describe the size dependence of the Curie temperature for materials of different sizes. To this end, the size-dependence model of glass transition temperature was expanded into a size-dependence model of the Curie temperature T_c of uniform low-dimensional ferromagnetic materials, based on energy theory and thermodynamics. The proposed model focuses on the critical size and the material shape, and has no adjustable free parameter. The model was used to predict the effect of material size on the Curie temperature, revealing that the T_c value decreases with the material size. This result agrees well with the experimental results and the prediction by other theoretical models. The research findings make up for the gap in the previous research on the Curie temperature.

1. INTRODUCTION

With reduced symmetry, ferromagnetic nanometals have different properties from bulk metals. The size of these materials has direct impacts on local surface properties like valence band structure and membrane shape. In return, these properties can serve as sensitive functions of material size and growth conditions [1].

In terms of application, a nanocrystal can be divided into two categories, namely, a thin membrane less than 10nm in thickness [2-3] and a bulk material consisting of nanoparticles or nanocrystals [4]. In recent years, much research has been done on the magnetic properties of membranes, especially the temperature of magnetic phase transition, *i.e.* the Curie temperature [2-3], revealing that the Curie temperature changes with the material size. On this basis, some theoretical models have been developed to describe the variation of Curie temperature with sizes [1-3].

One of these models is grounded on magnetic pole coupling. For a thick membrane, the Curie temperature satisfies:

$$T_c/T_{c0} \approx 1 - [(n_0+1)/2n]^\lambda \quad (1)$$

where T_{c0} is the Curie temperature of the bulk crystal; $\lambda \in [1, 1.59]$ [2] is a constant dependent on the magnetic pole coupling; $n=S/h$ is the number of deposited membrane layers (MLs), with S being the membrane thickness and h being the layer spacing (*i.e.* atomic diameter). If the membrane thickness becomes sufficiently small n_0 , there will be a linear relationship between the Curie temperature and the material size:

$$\frac{T_c}{T_{c0}} = \frac{n-1}{2n_0} \quad (2)$$

The above model predicts that Curie temperature decreases with the membrane thickness, which has been validated by experimental results.

Another model, created on the sequence, length and strength of keys [5], can also accurately depict the Curie transition of ferromagnetic membranes.

Meanwhile, the unique physical-chemical properties of ferromagnetic particles have also attracted attention in basic and applied research. For these particles, the Curie transition is essentially the shift of magnetic domains from disordered alignment to ordered alignment. The shift takes place right after the decline in particle size D , *a.k.a.* particle/grain diameter. To our knowledge, there is no theoretical model on the size dependence of the Curie temperature of ferromagnetic particles, which differs from that of membranes [2]. Thus, it is necessary to develop a unified theoretical model for particles with various sizes d .

The previous studies have shown the dependence of Curie transition on the material shape and size [2]. As a result, the physical nature of the transition should be fully considered in both theory and application.

In addition, the Curie transition is a secondary phase transition. Its features can be derived from those of other secondary phase transitions like glass transition. For glass transition, the size-dependence of the transition temperature T_g has been obtained for various low-dimensional materials by extending the size-dependence model of the melting temperature [6-7].

In this paper, the size-dependence model of glass transition temperature is expanded to examine the size dependence of the Curie temperature for magnetic materials in different sizes. Special attention was paid to determining the critical size D_0 , *i.e.* the size of a single-domain particle/domain. The determination requires different methods for different material sizes. Several experiments were carried out on Fe, Co and Ni membranes as well as Ni nanoparticles and Ni nanowires. The prediction of our model was found to be consistent with the experimental results.

2. MODELLING

Replacing the glass transition temperature T_g and bulk material transition temperature T_{g0} with the Curie temperature T_c and bulk material Curie temperature T_{c0} [6-7], the glass transition temperature T_g model can be modified as:

$$\frac{T_c}{T_{c0}} = \exp\left(-\frac{2\Delta C_p}{3R} \frac{1}{\left(\frac{D}{D_0-1}\right)}\right) \quad (3)$$

where, ΔC_p is the specific heat difference of the ferromagnetic body at the Curie temperature T_{c0} ; R is the ideal gas constant; D_0 is the critical size of the material that induces the Curie transition.

The critical size D_0 is usually defined as the diameter of all atoms on the surface of a low-dimensional material. If the dimension k of a spherical particle is zero, then the critical size D_0 equals six times the atomic diameter h ; if $k=1$ (*i.e.* the material is a nanowire), then the critical size equals four times the atomic diameter; if $k=2$ (*i.e.* the material is a membrane), then the critical size equals twice the atomic diameter.

The material size D directly bears on the magnetic transition of low-dimensional magnetic materials. The magnetic transition takes place when the temperature reaches a critical value T_c , turning the ferromagnetic body into paramagnetic body. In other words, the magnetic domains change from ordered alignment to disordered alignment. The magnetic transition is reversible. If the temperature decreases, the paramagnetic body will change back into ferromagnetic body, and the magnetic domains will become more orderly. Here, $2D_0$ is defined as the minimum material size for magnetic domains to remain orderly.

With low magnetic permeability and high coercive force, single-domain particles have a great influence on material properties and critical size estimation. These particles can only be magnetized rotationally, due to the absence of domain walls, and cannot be easily magnetized or demagnetized without external field or external force.

The critical size is the division point between the single-domain and other domain structures. Thus, the energy of the critical size of the single-domain structure is comparable to that of the simplest adjacent multi-domain structure. The two structures will have equal energy if both are at the critical size. Thus, the critical size of a single domain can be identified using the comparability [8].

The domains in ultrafine particles are spherical. For such a spherical domain, the total energy can be calculated as $E_T = E_w + E_H + E_d$, where E_w is the domain wall energy, E_H is the magnetostatic energy, and E_d is the demagnetizing energy [9]. Without external magnetic field, there is naturally no magnetostatic energy. The demagnetizing energy can be neglected because the domain surface only has a weak magnetic pole. Hence, the total energy of a spherical domain without external magnetic field is equivalent to the domain wall energy [9]:

$$E_T = E_w = S\sigma/2 \quad (4)$$

where, $S = \pi D^2$ and σ are the area and the energy density of the domain wall, respectively; D is the diameter of the domain. The energy density varies from domain wall to domain wall.

For the 90° domain wall in cubic crystals, σ equals $\pi\sqrt{A_1K_1}/2$; for the 180° domain wall in uniaxial crystals, σ

equals $4\sqrt{A_1K_1}$ [10]. Here, A_1 is the exchange integral constant and K_1 is the magnetic anisotropy constant.

For single-domain crystals, the magnetocrystalline anisotropy energy is minimized when the magnetic moments are arranged in parallel along the easy magnetization axis. In the absence of external magnetic field and internal stress, neither the magnetostatic energy nor magnetoelastic energy of the external field needs to be considered. Coupled with the lack of energy exchange, it is only necessary to consider the demagnetizing energy. Hence, the total energy of a single-domain crystal can be expressed as [10]:

$$E_T = E_d = \mu_0 V N M_s^2 / 2 \quad (5)$$

where, μ_0 is the magnetic constant or a vacuum permeability; M_s is the saturation magnetization intensity; $V = \pi D^3/6$ is the volume of the domain; N is the demagnetization factor. Here, $N = N_a \alpha_a^2 + N_b \alpha_b^2 + N_c \alpha_c^2$, with α_i ($i=a, b, c$, the three major axes of the crystal) being the direction cosine of the saturation magnetization intensity M_s , and N_a , N_b and N_c being the demagnetization factors along the three axes ($N_a + N_b + N_c = 1$), respectively. For spherical particles, the major axes are equal in length ($a=b=c$) such that $N_a = N_b = N_c$. Thus, the demagnetization factor N of a spherical particle equals $1/3$ [10]. According to formulas (2) and (3), the critical size D_0 can be derived as:

$$D_0 = \frac{9\sigma}{\mu_0 M_s^2} \quad (6)$$

The size dependence of the Curie temperature for magnetic nanoparticles can be obtained from formulas (3) and (6). It can be seen from the formulas that the Curie temperature increases exponentially with the reduction in particle size.

For wires and membranes, the ferromagnetic coupling is sufficiently strong to make the magnetic momentum parallel to the membrane direction [1], when the wire/membrane thickness n (the number of atomic layers) is greater than $20 \sim 30$. Therefore, the size dependence of the Curie temperature T_c for the wire/membrane can be predicted directly by formulas (3) and (6).

If n is smaller than $20 \sim 30$, most ferromagnetic membranes will change from 2-dimension to quasi-1-dimension [11]. In this case, the membranes exist as an island-like deposition on the substrate, and the critical size D_0 should be recalculated. For these membranes, the domain size is no longer the membrane thickness, but the diameter of island-like membrane.

By first-order approximation, the shape of island-like membrane can be viewed as a flat sphere or a spherical crown. Then, we have $S = \pi D' t$, $V = \pi (D')^2 t / 4$, with D' and t being the diameter and thickness of the flat spherical domain, respectively. As discussed above, D'_0 equals $2\sigma / (\mu_0 N M_s^2)$ if $E_w = E_d$. The island-like membrane can be considered as a quasi-1D system, because it is an intermediate form between particle (0D) and thin membrane (2D) [94]. Then, the demagnetization factor N of the island-like membrane equals the arithmetic mean ($2/3$) of the demagnetization factor ($N=1/3$) of the particle and the demagnetization factor ($N=1$) of the membrane.

The relationship between D' and D can be determined according to the Gibbs free energy difference ΔG between the island-like membrane and the normal membrane [94].

Here, a square membrane (side length: L ; thickness: D ; volume: $V=L^2D$) is cited as an example for the calculation of ΔG . Under a certain temperature T , the membrane will transform into a flat spherical particle of the same volume. In this case, the Gibbs free energy difference ΔG can be calculated as $V\Delta P_0+(\gamma_2S_2-\gamma_1S_1)$, where, $\Delta P_0=P_1-P_0$ is the pressure difference between the island-like membrane and the normal membrane; $S_2=\pi(D')^2/4+\pi(D')^2/4+\pi D't$ is the sum of the surface area of the island-like membrane and the area of the interface between the membrane surface and the substrate; $S_1=L^2+L^2+4LD$ is the sum of the surface area and the interface area before the formation of the island-like membrane; γ_1 and γ_2 are the surface energies (interface energies) before and after the transition, respectively. Note that P_1 refers to the pressure difference between the inside and outside of the membrane resulted from the surface curvature of the island-like membrane. According to the Gibbs–Thomson equation, P_1 equals $2f/D'$, with f being the surface stress of the crystal [12]. For the normal membrane, P_0 equals zero as the surface is flat with no curvature. In addition, the first and second terms in S_2 represent the surface area of the upper portion and the interface area between the lower portion and the substrate, respectively.

Thus, it can be derived that $\Delta G=V(2f/D'+\gamma/t+\gamma'/t+4\gamma/D'-\gamma'/D'-\gamma'/D-4\gamma/L)$, where γ is the surface energy of the crystal; γ' is the solid interface energy between the membrane and the substrate. Considering the material difference across the interface between the membrane and the substrate, the value of γ' can be computed by calculating the solid interface energy between the same materials and taking the arithmetic mean as the solid interface energy between two different materials. The values of f and γ_{ss} can be calculated as [13]:

$$f = \pm \frac{7}{2} \frac{T_m}{(T_m+6)} \sqrt{3h^2 S_{vib} H_m / (\kappa R V_m)} \quad (7)$$

$$\gamma_{ss} = \frac{196hT^2 H_m S_{vib}}{3R(T_m+6T)^2 V_m} \quad (8)$$

where T_m is the melting point of the material; h is the atomic diameter; H_m is the melting enthalpy; S_{vib} is the vibrational entropy in the melting entropy S_m ($S_{vib} \approx S_m$ for metals [63]); R is the ideal gas constant; V_m is the molar volume; $\kappa=1/B$ is the compression factor, with B being the bulk modulus.

Once the membrane thickness is reduced to a certain extent, the uniform membrane will spontaneously transform into the island-like membrane if the Gibbs free energy difference ΔG_0 between the two membranes is below zero. Mathematically, the spontaneous transition will take place if $\Delta G \leq 0$. Since $V=L^2D=\pi(D')^2t/4$ and $L=D'$ at the critical point, it is possible to deduce that $D = \pi t/4$. Then, the transition condition can be written as $D'/D \geq 8f/[(4-\pi)(\gamma+\gamma')]$. Assuming that $D'/D=C$, we have:

$$C=8f/[(4-\pi)(\gamma+\gamma')] \quad (9)$$

Substituting $D'/D=C$ into $D'_0=\sigma/(\mu_0 N M_s^2)$, we have:

$$D_0 = \frac{3\sigma}{\mu_0 C M_s^2} \quad (10)$$

3. RESULTS AND DISCUSSION

The T_c curve of Ni particles and wires is illustrated in Figure

1, where the solid line depicts the T_c curve of Ni particles and wires obtained by formulas (3) and (6), the solid dot stands for the experimental results of Ni particles [4], and the hollow dots and plus sign represent the experimental results of Ni wires [14, 15].

Considering the 90° domain walls of Ni particles and wires, the value of σ must be $\pi\sqrt{A_1 K_1}/2$. For large nanowires, the size dependence of the Curie temperature can also be theoretically obtained from formula (3), after redefining D as the diameter of the nanowire [14]. As shown in Figure 1, both the theoretical and experimental results agree that T_c decreases with the particle size D .

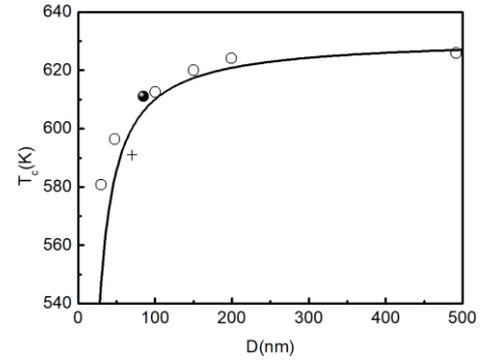


Figure 1. The T_c curve of Ni particles and wires

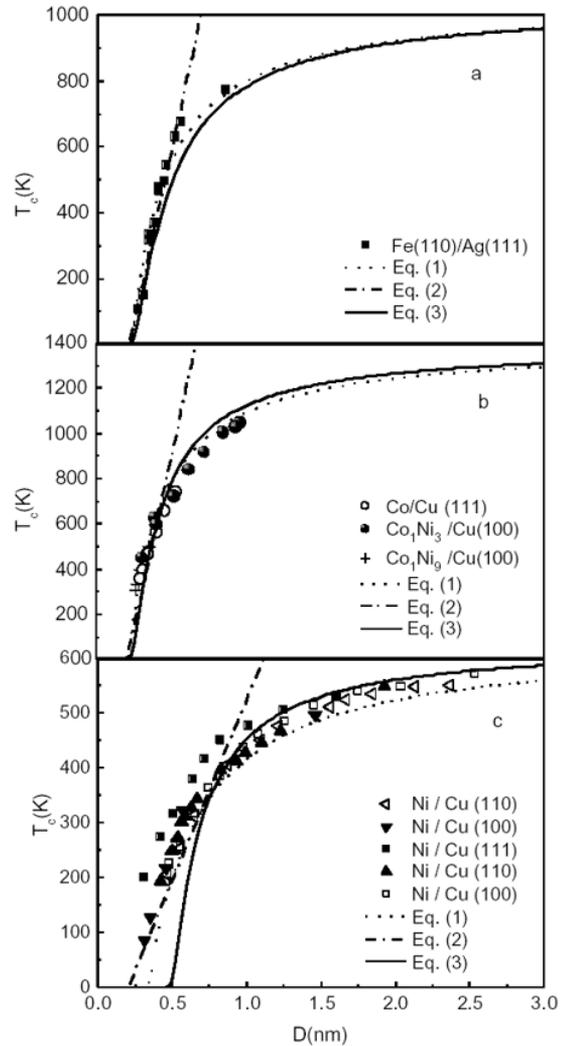


Figure 2. The T_c curves of Fe, Co and Ni membranes

The T_c curves of Fe, Co and Ni membranes deposited on different substrates with coherent interfaces are shown in Figure 2, where the solid lines stand for the results predicted by formulas (3) and (10); the dash lines reflect the results predicted by formulas (1) and (2); the solid boxes represent the experimental results for Fe [23]; the solid dots, hollow dots and plus sign jointly describe the experimental results for Co [24]; the solid up arrows, hollow boxes, solid left arrows, solid down arrows and solid boxes joints represent the experimental results for Ni [25-27].

Considering the 180° domain walls of the membranes, the value of σ must be $4\sqrt{A_1K_1}$. As shown in Figure 2, the theoretical predictions on the T_c of Fe, Co and Ni membranes agree well with the experimental results, indicating that the material size changed from 2-dimension to quasi-1-dimension with the reduction in membrane thickness. Since the magnetic order relies on material size, it is concluded that the dimensional transition has a certain impact on the ordering of magnetic domains. In addition, the T_c of membranes declined much faster than that of large particles and wires. This is because the island-like membrane has a much smaller D_0 than particles.

Compared with the model based on spin interaction (formulas (1) and (2)), formula (3) can predict the theoretical value of T_c excellently using the unified T_c equation only. Moreover, that formula (3) has no free parameter, while both n_0 and λ are fitting parameters in formulas (1) and (2).

Our model also discloses the relationship between membrane shape and membrane thickness from the angle of thermodynamics. The low surface-to-volume ratio was found to be the primary cause for the membrane to change from a uniform 2-dimension material to an island-like membrane. The model can also reveal the size dependence of ferroelectric transitions, after redefining $2D_0$ as the transition limit [28].

Table 1. Parameters and data in Figures 1 and 2

	Fe	Co	Ni
T_{c0} (K) [18]	1042	1395	631
A_1 (10^{-11} J/m) [18]	1.21	1.5	0.67
K_1 (J/m^3) [18]	4.8×10^4	4.3×10^4	-4.5×10^3
M_s (A/m) [16]	1.712×10^6	1.422×10^6	4.85×10^5
ΔC_p ($Jg\text{-atom}^{-1} K^{-1}$) [19]	15.24	10.9	5.06
h (nm) [20]	0.344	0.334	0.324
V_m (cm^3mol^{-1}) [20]	7.1	6.7	6.59
H_m ($KJ mol^{-1}$) [20]	13.80	16.19	17.47
T_m (K) [20]	1809	1768	1726
S_m ($J mol^{-1} K^{-1}$)	7.63	9.16	10.12
κ ($10^{-12}Pa^{-1}$) [21]	5.889	5.510	5.640
f (J/m^2)	3.69	4.56	3.984
γ (J/m^2) [22]	2.43	2.78	2.01
γ_{ss} (J/m^2)	0.275	0.407	0.491
γ' (J/m^2)	0.249	0.364	0.406
C	12.80	13.52	15.39

The relevant parameters in Figures 1 and 2 are listed in Table 1, where the magnetic constant $\mu_0=4\pi \times 10^{-7}$ H/m [16]; For metals, $S_{vib} \approx S_m=H_m/T_m$ [17]; $\kappa=1/B$, with B being the bulk modulus [12]; The f and γ_{ss} were computed by formulas (7) and (8) with $T=298K$; the value of γ' was calculated with the γ_{ss} values of the substrate being $0.222J/m^2$ and $0.320J/m^2$ for Ag and Cu, respectively [12].

4. CONCLUSIONS

Considering the critical size and material shape, this paper puts forward a size-dependence model of the Curie temperature T_c of uniform low-dimensional ferromagnetic materials, based on energy theory and thermodynamics. The proposed model, with no adjustable free parameter, was used to predict the effect of material size on the Curie temperature, revealing that the T_c value decreases with the material size. This result agrees well with the experimental results and the prediction by other theoretical models.

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