# Thickness-Dependent Curie Temperatures of Ultrathin Magnetic Films: Effect of the Range of Spin-Spin Interactions

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(Received 31 October 2000)

We present a simple model of spin-spin coupling which provides insight into the nature of the rapid decrease in the Curie temperature with decreasing thin film thickness $n$ (number of monolayers). The shift of Curie temperature $t(n) = 1 - T_c(n)/T_c(\infty)$ follows the usual power law $t(n) \sim n^{-\lambda}$ in thin films crossing over to linear behavior $t(n) \sim n$ in the ultrathin film thickness limit. Experimental results for ferromagnetic thin films are compared, and shown to follow curves of $t(n)$ with $\lambda$ values dependent on the nature of the spin-spin interactions.

DOI: 10.1103/PhysRevLett.86.2665 PACS numbers: 75.70.Ak, 75.10.Hk, 75.75.+a

The influence of the changing thickness on the magnetization of ultrathin ferromagnetic films is a topic of considerable interest. Our general understanding is strongly influenced by the pioneering theoretical work of Fisher and his co-workers [1,2], which predicts that the critical temperature $T_c$ will shift to lower temperatures than that of the bulk when the spin-spin correlation exceeds the film thickness. A thickness-dependent Curie temperature has been measured in Fe [3], Co [4,5], Ni [5–8], Gd [9,10], and CuMn spin-glass films [11].

The theory of critical behavior [1] informs us that the finite thickness of the film limits the divergence of the spin-spin correlation length $\xi$ at $T_c$. According to the scaling relationship, $\xi = \xi_0 t^{-\nu}$, where $t = (1 - T/T_c)$ is the reduced temperature, $\xi_0$ is a microscopic length, and $\nu$ is a universal critical exponent [12]. This translates into a shifting (and "smearing") of $T_c$ which can be expressed as

$$\delta T = [T_c(\infty) - T_c(n)]/T_c(\infty) = (C_1/n)^{\lambda} \qquad (1)$$

which measures the shift of the critical temperature $T_c(n)$ of a thin film of $n$ monolayers (ML) with respect to the bulk value $T_c(\infty)$. Here $C_1$ is a constant. The shift exponent $\lambda$ ($\lambda \leq 1/\nu$) reflects the appropriate universality class [1,12]. Theoretically, Eq. (1) is only valid in the thick film limit [13]. Alternatively, as advocated, for example, in Refs. [13,14], one can fit the experimental data according to

$$\Delta T = [T_c(\infty) - T_c(n)]/T_c(n) = (C_2/n)^{\lambda'}. \qquad (2)$$

Empirically, using $\Delta T$ rather than $\delta T$ is advantageous since measured values for $T_c(n)$ coincide with a power law over a larger range of orders of $n$ for $\Delta T$ than for $\delta T$. A problem arises, however, from the fact that $\lambda$ and $\lambda'$ values do not agree, or show the $1/\nu$ behavior expected on the basis of universal behavior [15]. A recent field theoretic argument [16] reconciles this discrepancy in terms of an effective shift exponent

$$\lambda_{\text{eff}} = \lambda' = (1 + 2\Delta_1)/\nu\,, \qquad (3)$$

where $\Delta_1$ describes leading terms in corrections to the usual finite-size scaling result. The key point to emerge from this most recent analysis is that the exponent $\lambda_{\text{eff}}$ depends on the nonuniversal value of the actual coupling strength in these ultrathin films [17].

In this Letter, we examine this premise in terms of a simple model of the thickness-dependent behavior of the Curie temperatures in ultrathin nickel films deposited epitaxially on copper single crystal substrates. Based on a simplified picture of the effective range of the spin-spin interactions, the model successfully accounts for the rapid decrease of $T_c$ in the few monolayer thickness limit. Furthermore, it provides insight into the different behavior observed and reported for Gd [9,10] and Ni [5–8] films, characterized by different $\lambda_{\text{eff}}$ values.

Experiments were performed on Ni films grown epitaxially on Cu(100), Cu(110), and Cu(111) substrates at room temperature. The change of the magnetization as a function of temperature and thickness was measured using the surface magneto-optical Kerr effect with a HeNe laser ($\lambda = 632$ nm) incident at 70° from the surface normal. Details of our apparatus and methodology have been reported elsewhere [5,14]. The deposition rate was about 0.5 ML/min for all of the films. Auger spectroscopy showed all the films to be free of contamination, and their structure was monitored with electron diffraction methods (LEED and RHEED), revealing well-ordered fcc structures.

Figure 1(a) shows plots of the thickness-dependent Curie temperatures of each of these Ni films. In each case, $T_c$ shows a rapid decline following a slow decline with decreasing thickness. The difference between the curves appears to reflect a changing symmetry. However, this difference disappears if we normalize the interlayer spacing to that of the Ni(100) lattice [Fig. 1(b)]. The different orientation Ni films have different interlayer spacing: 1.246 Å, Ni(110); 1.762 Å, Ni(100); 2.035 Å, Ni(111), all with different pairwise spin-spin interaction

  

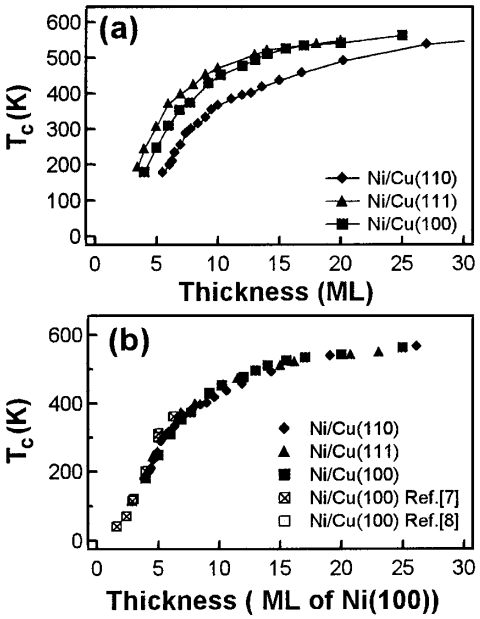


FIG. 1. Plots of thickness-dependent Curie temperatures for Ni films: (a) plot with the unit of monolayers, (b) normalized to the Ni(100) spacing. Low temperature data from other studies is included. (⊠: Ref. [7]; ⊡: Ref. [8].)

strengths. Normalizing the thickness in Angstrom units [or, alternatively, the number of monolayers of Ni(100)] produces the single curve [Fig. 1(b)]. This suggests that spin-spin interaction strength is important in determining the rapid decrease in $T_c$ to zero in the few monolayer thickness limit.

With this in mind, we examine this $T_c(n)$ behavior on the basis of a model of the range of spin-spin interactions in metallic systems. Our starting point is the mean field approximation [18],

$$k_B T_c = \frac{2}{3} \sum_j J_{ij} \times S \times (S + 1) . \tag{4}$$

Here $k_B$ is the Boltzmann constant, $S$ is the spin of the system, and $J_{ij}$ is the exchange coupling coefficient. In this model, the range of coupling between spins extends over many lattice sites $j$, which is appropriate for an itinerant metallic system. For Ni, spin-wave measurements show the mean spin-spin interaction range extending to the fifth nearest neighbors [19]. Thus, we assume a model in which spins on lattice sites are coupled to a cluster of such spins corresponding to a radius of interactions of $N_0$ monolayers (see Fig. 2). With this approximation, we have

$$k_B T_c = N \times E_0 , \tag{5}$$

where $N$ is the number of pairwise interactions and $E_0$ is a constant ("the coupling energy"). Equation (5) informs us that the Curie temperature $T_c$ is directly proportional to the number of pairwise spin-spin interactions in a manifold cluster of spins scaling with $N_0$ monolayers.

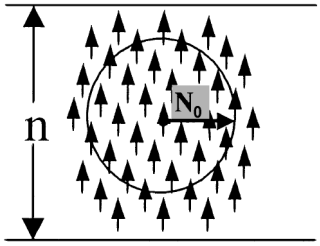


FIG. 2. Simple model showing range of spin-spin interactions, $N_0$ (ML), relative to film thickness $n$ (ML).

For a ferromagnetic material with this coupling range, $N_0$ monolayers, we now consider a slab of material in the bulk of thickness $n$, and compare its behavior to that of a free film of thickness $n$. In the film, spins at the surface have fewer spin interactions than those in the interior. The reduction in the number $N$ of spin interactions is expected to result in a decreased $T_c$, according to Eq. (5), to an extent dependent on the surface-to-volume ratio.

We restrict ourselves to a simple cubic lattice with a cluster size equivalent to a cube of side $2N_0$. Then the number of spins interacting with a particular spin is proportional to the volume of the cube, $(2N_0)^3$, for $N_0 \gg 1$. For a slab that is infinite in plane, we need only count the pairwise interactions in a single column. For the bulk slab of thickness $n$, the number of pairwise interactions normal to the slab plane is simply $n \times (2N_0)^3$.

For a free film, when the film thickness $n > N_0$, the spins at each surface lose $N_0 \times (2N_0)^2$ pairwise interactions, the next spin-in will lose $(N_0 - 1) \times (2N_0)^2$ pairwise interactions, the third spin-in will lose $(N_0 - 2) \times (2N_0)^2$ pairwise interactions, and so on, until the $(N_0 - 1)$th spin-in loses $(2N_0)^2$ pairwise interactions. The total number of pairwise interactions (in each column of spins) lost is $2 \times [N_0 + (N_0 - 1) + (N_0 - 2) + (N_0 - 3) + \cdots + 1] \times (2N_0)^2 = N_0 \times (N_0 + 1) \times (2N_0)^2$, or the number of pairwise spin interactions (in each column of spins) reduces to $n \times (2N_0)^3 - N_0 \times (N_0 + 1) \times (2N_0)^2$. According to Eq. (5), the ratio $T_c(n)/T_c(\infty)$ can now be expressed as

$$T_c(n)/T_c(\infty) = [n \times (2N_0)^3 - N_0 \times (N_0 + 1) \times (2N_0)^2]/[n \times (2N_0)^3]$$

which reduces to

$$T_c(n)/T_c(\infty) = 1 - (N_0 + 1)/2n , \tag{6}$$

such that

$$t(n) = 1 - T_c(n)/T_c(\infty) = (N_0 + 1)/2n \qquad (n > N_0) . \tag{7}$$

When the film thickness reduces to $n < N_0$, the number of pairwise interactions (in each column of spins) reduces to $n \times (2N_0)^3 - n \times (2N_0 + 1 - n) \times (2N_0)^2$ and the ratio

$$T_c(n)/T_c(\infty) = [n \times (2N_0)^3 - 2 \times (2N_0 + 1 - n) \times (2N_0)^2]/[n \times (2N_0)^3] = (n - 1)/2N_0$$

so that

$$t(n) = 1 - T_c(n)/T_c(\infty) = 1 - (n - 1)/2N_0 \quad (n < N_0). \tag{8}$$

Comparing Eq. (7) with the scaling law, Eq. (1), we observe that, for film thickness $n > N_0$, $t(n)$ follows a power law curve with shift exponent $\lambda = 1$. In the ultrathin film range ($n < N_0$), $t(n)$ is linear in $n$ [Eq. (8)]. This behavior is plotted in Fig. 3(a). For a range of spin-spin interactions, $N_0$, we observed that the "thin film" ($n > N_0$) behavior is modified in the "ultrathin film" limit ($n < N_0$). This linear behavior of $T_c(n)$ in ultrathin films has been observed experimentally and reported in earlier works [Fe/Ag(100), Ref. [3]; Co/Cu(100), Ref. [4]; CoNi/Cu(100), Ref. [5,14] ].

Finite-size scaling theory [1,12,13] predicts that the shift exponent $\lambda$ varies from $\lambda = 1$ for the mean field approximation [1] to 1.59 for the 3D Ising model [20] depending on the type of interaction and the boundary conditions used in the calculations [1,13,20,21]. Thus, Eqs. (7) and (8) can take a general form

$$t(n) = 1 - T_c(n)/T_c(\infty) \approx [(N_0 + 1)/2n]^{\lambda} \quad (n > N_0, \text{ thin film}), \tag{9}$$

$$t(n) = 1 - T_c(n)/T_c(\infty) \approx 1 - (n - 1)/2N_0 \quad (n < N_0, \text{ ultrathin film}). \tag{10}$$

In our simple model, the key parameter is $N_0$, the range of the spin-spin coupling. In a mean field approximation, we assume that the pairwise spin-spin interactions sum equally within a range of $N_0$ monolayers. This a reasonable approximation for an itinerant ferromagnet such as Ni, as evidenced by the superposition of experimental data (Fig. 1), shown in Fig. 3(b). We observe that the Ni data follow closely to the $N_0 = 5$ ML curve, which is in reasonable agreement with the experimental data [19] and theoretical calculation [22], showing a spin-spin interaction range about 4 ML. Our data, together with that of prior published work [7,8] on ultrathin nickel films [shown plotted in Fig. 3(b)] shows $t(n)$ varying linearly initially, in agreement with Eq. (8) for $n < N_0$. A recent study of the shift of the Curie temperature with nanometer diameter in nickel nanowire arrays [23] shows a similar power law curve with $\lambda = 0.94$ [using Eq. (1)] which is very close to our thin film result ($\lambda \approx 1$). However, they observed that the rapid drop of the $T_c$ value happened at a much larger thickness than that of the Ni thin films. In our model, this is simply due to the increased surface/volume ratio [i.e., a more rapidly decreasing $N$ in Eq. (5)] in the nanowires compare to the thin films for the same thickness.

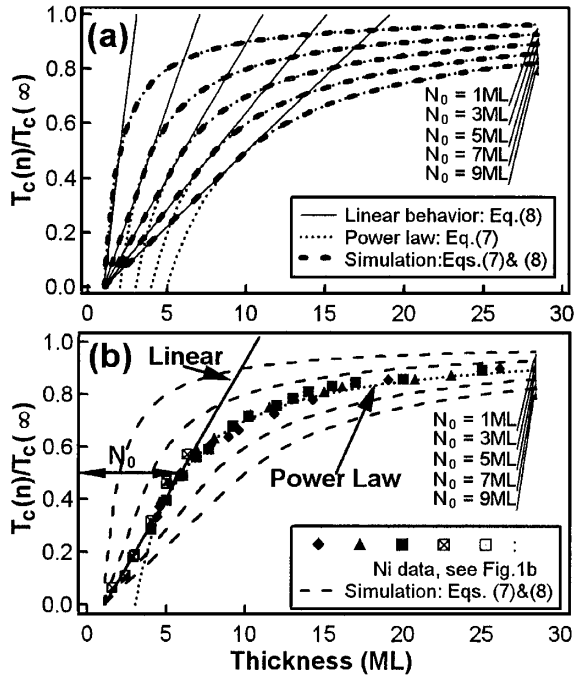


FIG. 3. (a) The reduced thickness-dependent Curie temperature follows a power law dependence ($\lambda = 1, n > N_0$) changing to a linear dependence ($n < N_0$) at a particular range of interactions, $N_0$. (b) The data for Ni follows closely the behavior for $N_0 = 5$ ML.

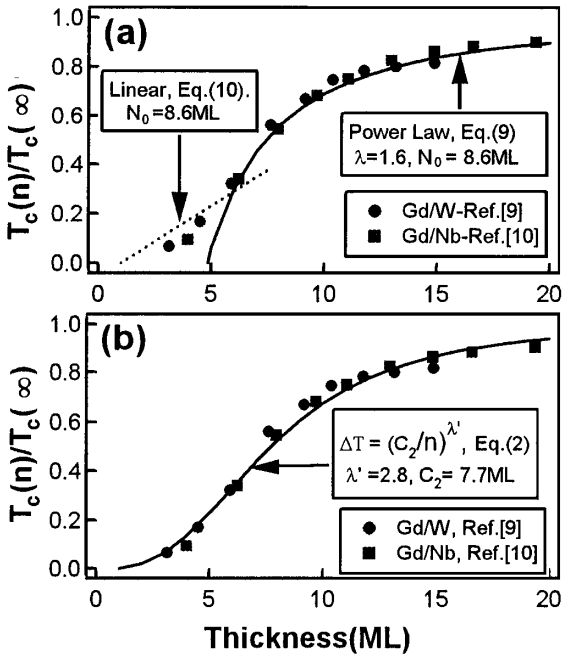


FIG. 4. (a) Plot of data for Gd(0001) thin films (Refs. [9,10]) showing linear ($n < N_0$) and power law ($\lambda = 1.6 \pm 0.2, n > N_0$) behaviors. (b) Data fitted to a single power law curve using Eq. (2), $\lambda' = 2.8 \pm 0.3$.

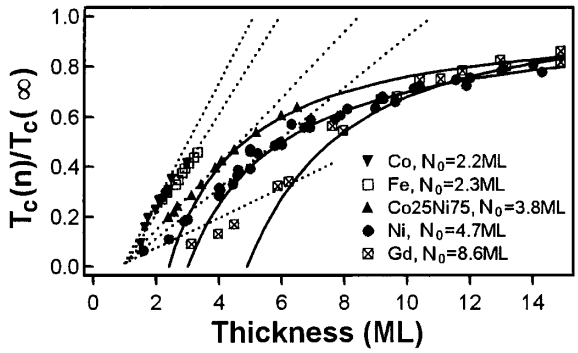


FIG. 5. The fitting of various magnetic thin film data: Fe [3], Co [4], Ni [7,8], Gd [9,10], and $Co_1Ni_3$ [5] alloys, using Eqs. (9) and (10) (solid and dotted lines). The fitting parameter $N_0$ is deduced using Eq. (10).

Our simple model is also able to reproduce the published $T_c(n)$ data for ferromagnetic Gd thin films [9,10] [Fig. 4(a)]. [We plotted the Gd data with $T_c(\infty) = 293$ K, and 1 ML Gd(0001) = 2.89 Å.] In this case, the change from power law ($n > N_0$) to linear ($n < N_0$) behavior is more marked. We attribute this to the larger and more localized nature of the atomic moments. A best fit to the data requires $\lambda = 1.6 \pm 0.2$ and $N_0 = 8.6$ ML [24]. In this case, $\lambda$ accurately reflects a Heisenberg ferromagnet. If we fit a single curve to the data using Eq. (2) [see Fig. 4(b)], the "effective" shift exponent is $\lambda' = 2.80 \pm 0.3$. This discrepancy between $\lambda$ and $\lambda'$ shift exponents is clearly the result of fitting the data to a single curve common to both the linear ($n < N_0$) and the power law ($n > N_0$) regions. Similar measurements of the $T_c(n)$ of the Fe layers on Ir(100) [16] show an effective shift exponent $\lambda' = 3.15 \pm 0.15$. Finite-size scaling corrections applied to a Heisenberg ferromagnet [Eq. (3)] predict a corrected value $\lambda' = 3.0 \pm 0.1$, in agreement with the experimental data fitting to a single thickness-dependent curve [16]. These values are substantially in agreement with those of the Gd(0001) films [Fig. 4(b)].

The crossover from power law to linear behavior has been observed in other magnetic thin film systems. Figure 5 shows this crossover for Gd [9,10], Ni [7,8], and $Co_1Ni_3$ [5] alloy thin films. Fe [3] and Co [4] data show linear behavior over a temperature/thickness range limited by interdiffusion with the substrate. The fitting parameter $N_0$ is indicated in the figure.

In summary, we have analyzed data for the thickness dependence of the Curie temperature of thin ferromagnetic Ni and Gd films in terms of a simple model, predicting a crossover from power law to linear behavior when the film thickness reduces below some mean value of the range of the spin-spin interactions. This dictates the value of the effective shift exponent $\lambda'$. The effective shift exponent relates to the critical shift exponent via corrections which reflect the strength of the coupling between spins, as suggested by our simple model and the field theoretical analysis of Henkel *et al.* [16].

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