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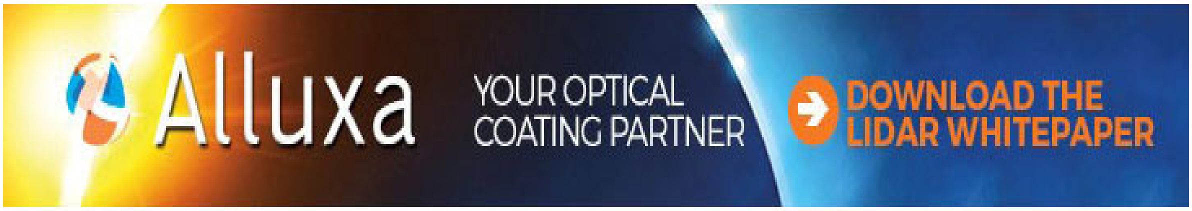

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Finite-size scaling behavior of ferromagnetic thin films

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We have used molecular-beam epitaxy to grow high-quality pseudomorphic Ni and Co_1Ni_9 films on Cu(001). From temperature-dependent surface magneto-optic Kerr effect measurements of these films, we have determined the finite-size scaling behavior of the Curie temperature of ultrathin films for a thickness range of $n=2.5$–16 monolayers (ML). The film thickness dependent Curie temperature for each of these ferromagnetic thin-film systems, $T_C(n)$, is described by a finite-size scaling formula: $[T_C(\infty) - T_C(n)]/T_C(n) = [(n-n')/n_0]^{-1/\nu}$, where $T_C(\infty)$ is the bulk Curie temperature, $n_0=2.5\pm0.5$ ML for Co films and 3.5 ± 0.4 ML for Ni and Co_1Ni_9 films is the microscopic length scale, and $\nu=0.76\pm0.08$ is the bulk correlation length exponent. An interesting result is that $T_C(n)$ extrapolates to zero in the single mononolayer limit, $n'=1$.

I. INTRODUCTION

The question of what happens to the bulk thermodynamic properties of a system when one or more of its dimensions is reduced to atomic size is one of fundamental importance. Statistical mechanical calculations of second-order phase transitions[1] suggest that the position and nature of a critical singularity will be effected in a precisely defined way. Finite-size scaling theory[2] predicts that the Curie temperatures of magnetic thin films will decrease from the bulk value as the film thickness is reduced to values comparable to the bulk microscopic length scale. This has been observed in a variety of thin-film systems. For example, the thickness dependent Curie temperatures $T_C(n)$ in ferromagnetic thin films[3] and the spin-freezing temperature T_g in spin-glass multilayers[4] have been observed to be significantly lower than their bulk values. Recent experiments on ferrimagnetic nanoscale particles have reported an increasing T_C with decreasing particle size.[5]

The experimental data is usually fitted to a finite-size scaling formula of the form[2]

$$[T_C(\infty)-T_C(n)]/T_C(\infty)=(n/n_0)^{-\lambda}, \qquad (1)$$

where the shift in transition temperature with changing length $T_C(n)$ is given by the shift exponent $\lambda=1/\nu$, and n scales with a microscopic length, n_0, characteristic of the particular system. A difficulty arises in the case of ultrathin ferromagnetic films. The above finite-size scaling formula[2] is strictly valid only in the thick-film limit,[6,7] whereas T_C is observed to deviate significantly from its bulk value for films a few atomic layers thick. The data extrapolate to $T_C(n)=0$ for microscopic length scales n_0 of the order of a few angstroms, in contrast to the spin-glass and particulate systems which show T_C varying over a much wider size range. This makes accurate determinations of the critical exponents more possible in these latter systems.

Theoretical plots of the above finite-size scaling formula show strong deviations from $\lambda=1/\nu$ in the limit of a small number of monolayers n.[6] Much better agreement is maintained down to small thicknesses using[7]

$$[T_C(\infty)-T_C(n)]/T_C(n)=(n/n_0)^{-\lambda}. \qquad (2)$$

Other measurements on polycrystalline Ni films[3] and single-crystal Ni films[8–10] have been made. The data were fitted to both relations[8,9] giving values of λ in the range $1.27\pm0.20<\lambda<1.48\pm0.20$. Any discrepancies only become apparent as $n\rightarrow0$. Equation (1) gives $T_C(n)=0$ for finite values of n while Eq. (2) predicts $T_C(n)=0$ in the limit $n=0$. However, the value of λ is strongly dependent on this limit. Moreover, it is in the precise determination of the finite-size scaling parameters that the experiments make contact with the statistical mechanical models and calculations.

We have used molecular-beam epitaxy (MBE) techniques to deposit fcc pseudomorphic layers of ferromagnetic materials on a Cu(001) substrate and to control their thickness and morphology down to the single monolayer limit.[11,12] The magnetic properties can be simultaneously monitored using the surface magneto-optic Kerr effect (SMOKE) which is sensitive to changes in the magnetization density in this limit.[13] Measurements of the Curie temperatures of Co, Ni, and Co_1Ni_9 alloy films show the film thickness dependent Curie temperature in $T_C(n)$ is best described in the ultrathin film limit by a finite-size scaling relation of the form

$$[T_C(\infty)-T_C(n)]/T_C(n)=[(n-n')/n_0]^{-\lambda} \qquad (3)$$

with $T_C(n)=0$ at a finite film thickness n'.

II. THICKNESS DEPENDENT CURIE TEMPERATURES

Ni on Cu(001) represents a lattice-matched epitaxial system which remains stable against interdiffusion to cycling temperatures up to 490 K, even for the thinnest films.[14,15] This is important since the widest possible temperature range (i.e., thickness range) is required to extend the measurements to up to the bulk Curie temperature, $T_C(\infty)=627$ K. Reflection high-energy electron-diffraction (RHEED) oscillations were used to calibrate the film thicknesses, and low-energy electron-diffraction (LEED) measurements showed the layers were pseudomorphic.[15] The Curie temperatures $T_C(n)$ were determined from power-law fits to the data: $M=M_0(1-T/$

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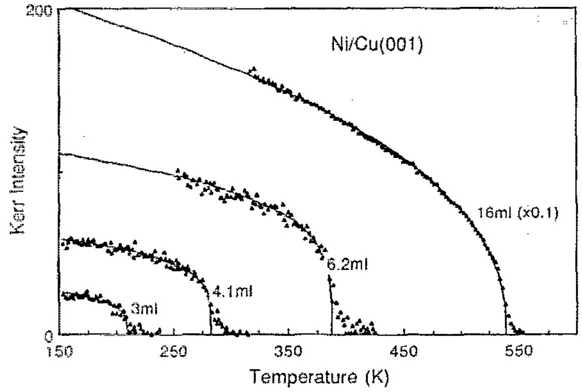

FIG. 1. Magnetization curves for Ni/Cu(001) ferromagnetic thin films, showing the Curie temperature decreasing with decreasing film thickness (1 ML=1.8 Å). The full lines are power-law curves $M=M_0(1-T/T_C)^\beta$.

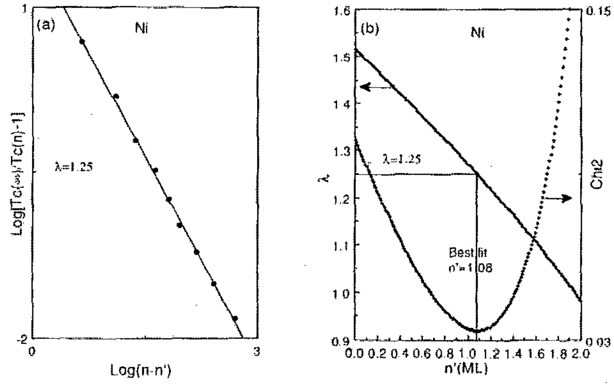

FIG. 2. (a) The log-log plot of the finite-size scaling expression: $[T_C(\infty)/T_C(n)-1]=C_0(n-n')^{-\lambda}$ for the Ni films, and (b) the least-squares fit to $\log[T_C(\infty)/T_C(n)-1]$ vs the $\log(n-n')$ plot for various values of n'. Note the sensitivity of λ to the value of n'.

$T_C)^\beta$, as shown in Fig. 1. The measurements were checked against a second system: a homogeneous, pseudomorphic alloy of $\mathrm{Co_1Ni_9}$ with a higher bulk Curie temperature of $T_C(\infty)=753$ K. Both sets of data show second-order phase transition behavior with the critical temperature decreasing with decreasing film thickness.[3,8,9]

III. FINITE-SIZE SCALING

Log-log plots were used to compare the validity of Eq. (1) vs Eq. (3). The $\log[1-T_C(n)/T_C(\infty)]$ vs $\log(n)$ plots [Eq. (1)] of the experimental data give a nonlinear behavior over the whole thickness range, and the best fit to the data gives values of λ outside the range of theoretical predictions for both the Ni ($\lambda=0.96$) and $\mathrm{Co_1Ni_9}$ ($\lambda=0.89$) films. For the $\log[T_C(\infty)/T_C(n)-1]$ vs $\log(n-n')$ plots [Eq. (3)] shown in Fig. 2 for the Ni films, the experimental data falls on a straight line, and the shift exponents for Ni ($\lambda=1.25$) and $\mathrm{Co_1Ni_2}$ ($\lambda=1.39$) approach theoretically predicted values.[2,6,7] The precise value of λ is sensitive to the extrapolated value of n'. To determine n', we used least-square fits to the $\log[T_C(\infty)/T_C(n)-1]$ vs $\log(n-n')$ plots for various values of n', the minimum in χ^2 deviation giving $n'=1.08$ ML and $\lambda=1.25$ for the Ni films, as shown in Fig. 2. A similar procedure gives $n'=0.87$ ML and $\lambda=1.39$ for the $\mathrm{Co_1Ni_9}$ alloy films.

The finite-size scaling behavior is plotted in Fig. 3, together with published data for Co epilayers[16] using the least-squares fit parameters in Table I. The measurements confirm that the data are best described by a scaling-law curve which extrapolates to $n'\approx 1$ in the limit of $T_C(n)\to 0$. The shift exponent value, $\lambda=1.33\pm0.15$ is close to the theoretically predicted value[6] and within the range of values previously reported for Ni films.[3,8,9] The precise value of the shift exponent is dependent to some extent on surface and interface properties of these ultrathin layers.[7] The microscopic scaling lengths, $n_0=2.5\pm0.5$ ML for Co and $n_0=3.5\pm0.4$ ML for Ni and $\mathrm{Co_1Ni_9}$ are close to the dimension of a single unit cell in these fcc(001) films which gives the appropriate microscopic length scale. The experimentally determined values are summarized in Table I.

IV. DIMENSIONAL CROSSOVER

As noted previously, the scaling relations in Eqs. (1)–(3) strictly hold only in the limit of large n when the film thickness is large compared to the microscopic scaling length. A question exists in the theoretical literature[1,2,6,7] as to the corrections necessary as the thickness approaches this length, $n\approx 3$ ML. The dimensional crossover effect is seen as a sudden change in the power-law exponents plotted in Fig. 4, from $\beta=0.25\pm0.05$ to $\beta=0.38\pm0.05$ near 7 ML (compare the magnetization curves in these two limits, inset Fig. 4). The value for the films thinner than 7 ML agrees with previously reported values for epilayers of Fe/Au(001) (Ref. 17) and Ni/Cu(111),[9] $\beta\approx 0.24$. The value of the power-law exponent above the crossover point is characteristic of a Heisenberg system, as is the value of the

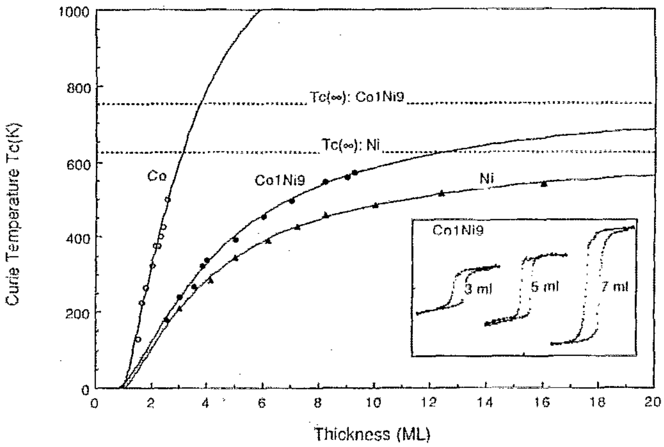

FIG. 3. Finite-size scaling of $T_C(n)$ as a function of decreasing film thickness. Fits to the data using the scaling law relation (3) are shown for Ni ($n'=1.08$, $\lambda=1.25$), $\mathrm{Co_1Ni_9}$ ($n'=0.87$, $\lambda=1.39$) and Co ($n'=1.00$, $\lambda=1.34$). The inset diagram shows typical Kerr intensity hysteresis loop behavior from which the retentivity $M(T)$ is determined (Fig. 1).

TABLE I. Experimental values of the finite-size scaling parameters.

Parameter	Ni films	Co_1Ni_9 films	Co films (Ref. 16)
λ	1.25 ± 0.07	1.39 ± 0.08	1.34
ν	0.80 ± 0.04	0.72 ± 0.04	0.75
n'	1.08 ± 0.20	0.87 ± 0.20	1.00
n_0	3.38 ± 0.14	3.74 ± 0.12	2.42
C_0	4.58 ± 0.24	6.25 ± 0.28	3.27

shift exponent $\lambda = 1.39$ (compared to 1.42).[6] Also, the fact that $T_C(n)$ extrapolates to zero in the limit of a single monolayer argues a case for an isotropic Heisenberg ferromagnet showing zero long-range order in the two-dimensional limit.[18] Measurements of the microscopic domain structure suggest that ultrathin epitaxial Co films on Cu(001) behave like two-dimensional Heisenberg magnets at temperatures close to T_C.[19] However, the crossover to $\beta \approx 0.25$ suggests an alternate case for XY-model behavior in the thinnest films, which is polarized with the easy axis of magnetization within the film plane.[15] This crossover is not apparent in the scaling-law curves (Fig. 3), i.e., no discontinuity in the Curie temperature is observed at $n \approx 7$ ML. This is due to the fact that the finite-size scaling parameter λ is not sensitive to changes in the correlation length critical exponent ν in this thickness range, given the measured microscopic scaling length $n_0 \approx 3$ ML.[20]

V. CONCLUSIONS

Although these films were prepared using state-of-the-art MBE techniques, the experimental magnetization curves show a small tail above T_C, indicative that the thinnest films are not of uniform thickness in the 2D plane (Fig. 1). This signifies "pseudocritical" behavior[6] arising from finite-size rounding and critical temperature shifts in these ultrathin-film systems. However, for three of these ultrathin-film systems, the film thickness dependent Curie temperatures extrapolate to zero in the mololayer limit.

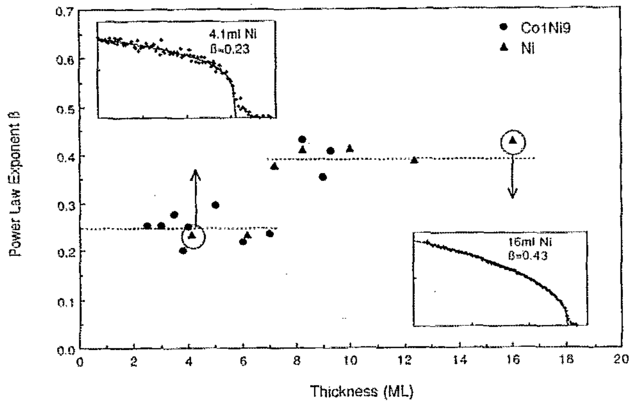

FIG. 4. "Crossover effect" observed as a sudden change in the value of the power-law exponent, β, at a film thickness of $n \approx 7$ ML.

What is significant about these, and previous finite-size scaling measurements,[3,8,9] is that while such effects as magnetic anisotropy[21] and substrate crystalline symmetry strongly influence the shape of the magnetization curves and, hence, the approach to criticality, the shift of the Curie temperature to lower temperatures with decreasing thickness down to the monolayer thickness limit is well described by the finite-size scaling relationship [Eq. (3)] proposed here. This finite-size scaling relationship yields values for the bulk correlation length exponent which are in better agreement with current theoretical models.

ACKNOWLEDGMENT

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[21] The Kerr measurements show the magnetic anisotropy to lie within the film plane. In the case of the Ni films, it switches to normal to this plane for thicknesses $n > 7$ ML.