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Magnetic phase transition in a two-dimensional system: $p(1\times1)$-Ni on Cu(111)

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The magnetic phase transition of ultrathin epitaxial Ni films on Cu(111) is studied using the magneto-optic Kerr effect. Analysis of the thickness and temperature dependence of the spontaneous magnetization yields a shift exponent $\lambda=1.44\pm0.2$, in good agreement with previous experimental studies of thin Ni layers, and a critical exponent $\beta=0.24\pm0.07$ in good agreement with a recent study of $p(1\times1)$-Fe on Au(100). In-plane symmetry does not appear to play an important role in critical behavior of these systems.

Recent advances in spin-sensitive spectroscopic techniques offer important new opportunities to experimentally study magnetic phase transitions. Magnetic thin films represent a particularly important subset of magnetic materials in which to explore the universality hypothesis[1] that underlies theories of phase transitions, and to test specific theoretical predictions of critical exponents[2,3] and scaling parameters.[2,4] These quasi-two-dimensional systems also offer unique opportunities for studying finite-size scaling effects[4] based on the ability to deliberately modify the structure and composition of surfaces and thin films using modern surface-science techniques.

Near the transition temperature T_c that separates the ferromagnetic and nonmagnetic phases, the disappearance of the magnetization M can be accurately characterized by a simple power law $M(T)=C^*(1-T/T_c)^{\beta}$, where β is the critical exponent. Based on theoretical grounds, it is generally accepted that phase transitions can be grouped into universality classes that are characterized by a small number of parameters such as the dimensionality of the system and symmetry of the order parameter. One manifestation of the universality hypothesis is that systems with different dimensionality should have different values of critical exponents depending on the specific universality class they fall into. For example, bulk systems are predicted to generally exhibit values of β near 0.38,[5] and two-dimensional (2D) universality classes are predicted to generally exhibit values of β in a range 0.1–0.15.[6] The value of β for the 2D Ising model is $\frac{1}{8}$.[7]

In ultrathin films, magnetic properties, in particular the transition temperature, become functions of thickness-dependence corrections. Specifically, the critical temperature T_c must be regarded as a thickness-dependent parameter, $T_c(n)$ which approaches the bulk (infinite system) critical temperature $T_c(\infty)$ as the scale factor n (the number of layers) approaches ∞. It has been shown that the approach of $T_c(n)$ to $T_c(\infty)$ can also be described by a simple power law[8,9] characterized by a shift exponent λ defined by

$$\epsilon_n \propto \frac{1}{T_c(n)}-\frac{1}{T_c(\infty)}\approx An^{-\lambda}.$$

The shift exponent λ is predicted to have a value between 1.0 and 2.0, depending on whether free-surface or periodic boundary conditions are assumed.[8,9]

In spite of the significant number of theoretical studies that have focused on the properties of 2D phase transitions, and the variety of specific predictions that are available for critical exponents and related scaling parameters, very few relevant experimental results are available, especially for ultrathin films. There are a few recent experimental studies that have obtained values for the critical exponent β and for the shift exponent λ. Specifically, Rau, Xing, and Robert[10] have obtained $\beta=0.128\pm0.01$ for $p(1\times1)$-V films on Ag(100), and Durr *et al.*[11] have obtained $\beta=0.22\pm0.05$ for $p(1\times1)$-Fe films on Au(100). The result for V films lies very close to the 2D-Ising-model value, while the result for Fe on Au(100) lies outside the range of values expected for two-dimensional universality classes ($0.1\leq\beta\leq0.15$). It is also outside the range of values expected for bulk materials ($\beta\cong0.365$ for a Heisenberg ferromagnet) or surface magnetism of a 3D ferromagnet ($\beta\sim0.8$). There are also limited experimental results for the shift exponent λ. Bergholz and Gradmann[12] have found, using a torsion magnetometer, that epitaxial Ni(111) films on Re(0001) yield $\lambda=1.27\pm0.2$. Lutz *et al.*[13] found, based on electrical-resistance measurements, that (polycrystalline) Ni films grown on quartz substrates yielded $\lambda=1.33\pm0.13$.

This paper reports magneto-optic Kerr-effect studies of the temperature- and thickness-dependent magnetic behavior of ultrathin epitaxial Ni films grown on Cu(111) surfaces. The Kerr-effect method offers the advantage of studying the macroscopic order parameter $M(T)$ over a film thickness range from one monolayer to several hundred angstroms. This feature permits evaluation of the shift exponent λ that describes the thickness-dependent critical temperature $T_c(n)$, as well as the critical exponent β.

Our epitaxial films were prepared and studied *in situ* under ultra-high vacuum conditions (below 1×10^{-10} torr). Conventional techniques were used to prepare the 1-cm-diam Cu(111) crystals: The crystals were aligned by x-ray Laue techniques to an accuracy of approximately $\pm\frac{1}{2}°$ and mechanically polished to obtain a mirror surface. After repeated Ne^+ sputtering and annealing, analysis by Auger spectroscopy and low-energy electron diffraction (LEED) were used to verify that the substrate surface was clean and well ordered. Based on our visual observation of LEED spot size and using well-established

analysis methods,[14] the average terrace width of our surface and epitaxial films was ~75 Å. Thin magnetic films were grown by electron-beam evaporation (at a rate of ~0.1 Å/min) from the tip of a 99.95% pure 0.08-in.-diam Ni wire. The Cu(111) crystal was held at temperatures ranging from 370 to 400 K during sample preparation. This temperature range and growth rate was found to yield very good epitaxy[15–18] while avoiding interdiffusion effects. Recent positron tunneling measurements[18] show that strained coherent growth occurs for $p(1\times1)$-Ni on Cu(111) up to 15 Å. We explored interdiffusion effects (changing of Auger ratios, deterioration of LEED patterns, and dramatic changes in hysteresis curves) by temperature-dependent studies above 600 K. All of our results and the recent photoemission and positron studies of $p(1\times1)$-Ni on Cu(111) are consistent with good film integrity over the temperature and thickness range required by our experiments. Film thickness was calibrated using a technique based on a pair of quartz microbalances. Our thickness calibrations are consistent with breaks in the slope of Auger peak intensities plotted as a function of time during film growth (also documented by Ref. 17) which generally indicate the completion of successive layers.

We use the magneto-optic Kerr effect to probe the magnetization of the sample. Figure 1 displays a typical set of Kerr-effect data (magnetic hysteresis loops M vs H) for a 2.8 layer film at several selected temperatures. The sharp corners of the loop and the low coercive force are properties that we have found to be characteristic of high-quality epitaxial films. Such films exhibit a single magnetic domain at low applied field, and abrupt domain reversal at low values of applied field (low coercive force). The saturated regions of the hysteresis curve provide a means of averaging statistics for $\pm M_{sat}$ to obtain high accuracy in the measurement of M. A previous report[19] contains additional details related to magneto-optic Kerr-effect techniques applied to thin magnetic films. When comparing the present results (for Ni) with our previous result (for Fe) it should be noted that the Fe films of equivalent thickness yield Kerr-effect signals which are roughly 5 times larger in the longitudinal configuration and at least 30 times larger in the polar configuration than the longitudinal configuration signals reported in this article (accounting for the poor signal-to-noise ratio).

Figure 2 displays a set of M_{sat} (T) vs T curves for $p(1\times1)$-Ni films on Cu(111) for thicknesses from $n=1$ to $n=8$ layers. These curves show the striking dependence of the Curie temperature $T_c(n)$ on film thickness, and the temperature dependence of M near the phase transition. We assume that the magnetization near the transition temperature is described by $M=C[1-T/T_c(n)]^{\beta}$ and adopt a curve-fitting procedure to determine both β and $T_c(n)$ that favors points near $T_c(n)$ subject to certain constraints. The average terrace width of our epitaxial layers (~75 Å) is assumed to also represent the maximum distance in the film over which a magnetic fluctuation maintains coherency. This correlation length is given by[4] $\xi=a(1-T^*/T_c)^{-\nu}$, where a is the lattice constant and T^* represents the "rounding" temperature at which M departs from the power law. Using for the critical exponent ν the 2D Ising value ($\nu=1.0$), we find that it is ac-

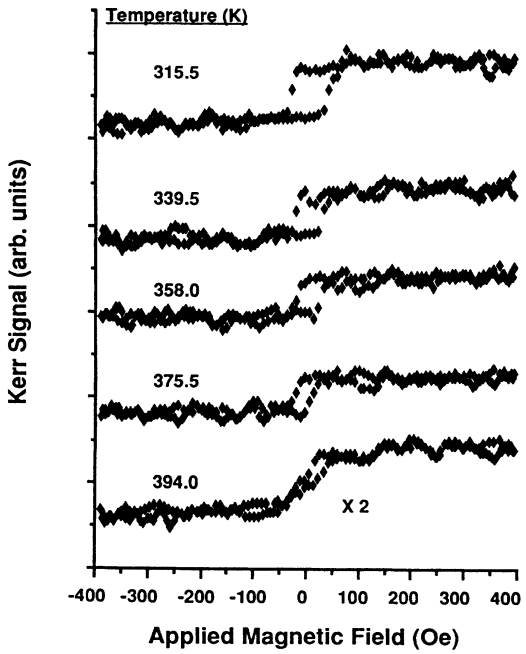

FIG. 1. Hysteresis curves for a 2.8 monolayer $p(1\times1)$-Ni film on Cu(111) as a function of temperature.

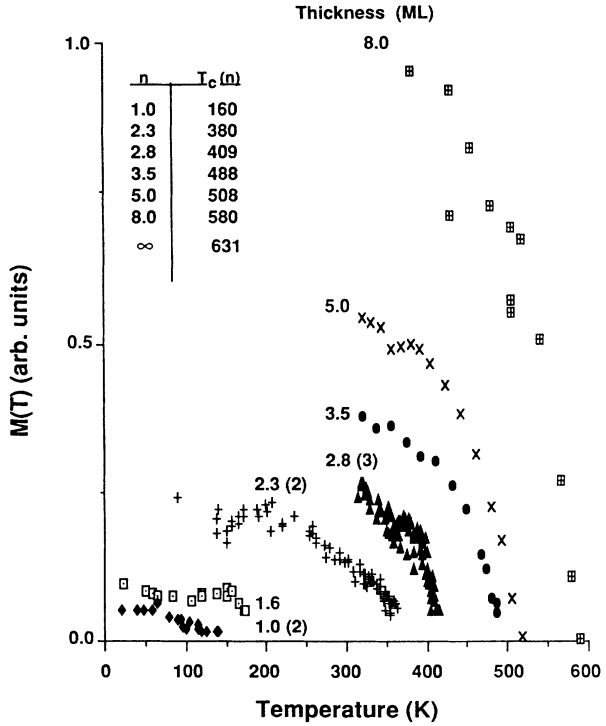

FIG. 2. Magnetization vs temperature for $p(1\times1)$-Ni films of several thicknesses on Cu(111). The numbers in parenthesis after the film thickness represent the number of films plotted at that thickness. This data shows the reproducibility of our film thickness and magnetization measurements. Inset: layer-dependent transition temperatures.

ceptable to model the phase transition using the above power law to within 1% of $T_c(n)$ in units of $T/T_c(n)$.

Figure 3 displays a magnetization versus temperature (top panel) and a log-log representation of the magnetization versus $[1-T/T_c(n)]$ (bottom panel) for a 2.6 layer film. In the log-log form, the slope of the line is equal to β. We stress that this procedure for obtaining T_c and β is subject to well-known uncertainties based on the limited range of our data.[20] In addition to the length ξ which defines the correlation distances in the film plane, a second characteristic distance (the film thickness Δx) corresponding to the maximum allowed correlation distances perpendicular to the film surface can be defined. Since our films and those of Durr *et al.*[11] have thicknesses satisfying $\Delta x \ll \xi$ and exhibit in-plane magnetization, we may expect that they will belong to the same universality class and thus have the same critical exponent β provided there is no in-plane anisotropy resulting from the different crystal symmetry.[21]

For two different epitaxial films (in the range $2 \leq n \leq 3$), we obtain $\beta = 0.24 \pm 0.07$ by fitting our data over a reduced temperature range $-2 \leq \log_{10}[1-T/T_c(n)] \leq -0.5$. This value is significantly larger than the result obtained by Rau *et al.* [$\beta = 0.128$ for a V monolayer on Ag(100)], but agrees very well with the value (0.22 ± 0.05) obtained by Durr *et al.* for an Fe monolayer on Au(100). Our error bars are generous and take into account the limited reduced temperature range over which our data are fitted, which is slightly over one decade. We have also obtained $\beta = 0.32 \pm 0.09$ for an eight-monolayer film. Although this value still agrees within the error bars with the value for 2.6 layers, the higher value may indicate a departure from 2D behavior as Δx approaches ξ. Hence, it is possible that an "effective" exponent is determined by our experiment even at $n = 2.6$; even if this is the case, the actual critical exponent would likely lie within the error bars we place on β for $n = 2.6$.

TABLE I. Predicted and measured (experimental) values of the shift parameter λ. When $h = 0$ (refer to text), the best fit occurs for $\lambda = 1.44$, which is near the Ising result and the Heisenberg result for free surface boundary conditions. When h is allowed to vary, optimum fit values of $h = 0.6$ and $\lambda = 1.01$ agree well with the Ising prediction with $h \neq 0$. It has been suggested that λ should equal $1/\nu$, where ν is the bulk correlation length exponent (Ref. 22). For the Heisenberg model this results in $\lambda = 1.4$. The meaning of C_0, λ, and h are explained in the text.

	C_0	λ	h
	0.116	1.00 ± 0.00	0.60
Free surface [a]	0.196	1.27 ± 0.05	0.0
Cyclic [a]	0.196	2.00 ± 0.08	0.0
Free surface [b]		1.24	0.0
Free surface [c]		1.00	0.0
Periodic [d]		2.00	0.0
Experimental	2.30	1.44 ± 0.2	0.0
	1.03	1.01 ± 0.1	0.60
	1.08	1.04 ± 0.1	−0.40

[a] $n = 1, 3, 4, 5, \infty$, Ising.
[b] $2 \leq n \leq 6$, Heisenberg (good fit).
[c] $1 \leq n \leq 6$, Heisenberg (reasonable fit).
[d] $n = 1, 2, 3$, and 4, Heisenberg (reasonable fit).

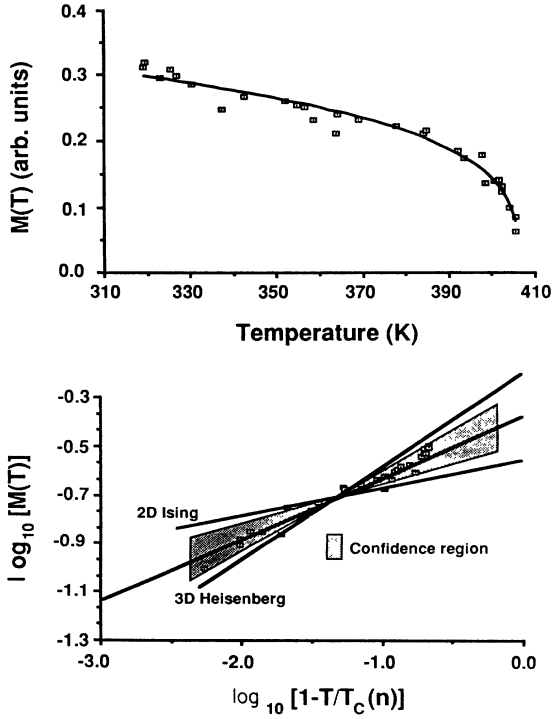

FIG. 3. (Top panel) magnetization vs temperature for a 2.6 layer $p(1\times1)$-Ni film on Cu(111). (Bottom panel) $\log_{10}M$ vs $\log_{10}(1-T/T_c)$ for the same film as in the top panel.

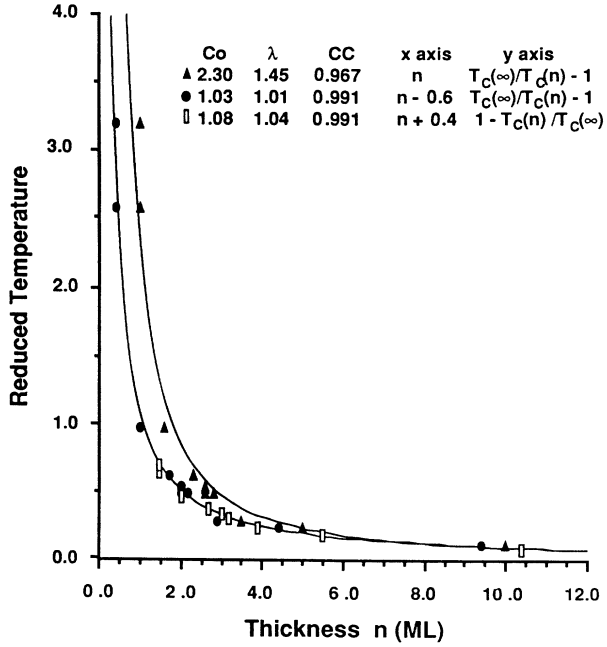

FIG. 4. Reduced critical temperature vs thickness for $p(1\times1)$-Ni films on Cu(111). In the table, CC stands for the correlation coefficient as described in the text.

We now discuss the thickness dependence of the transition temperature $T_c(n)$. For large n, ϵ_n can be expressed in two equivalent forms: $\epsilon_n \equiv T_c(\infty)/T_c(n) - 1 \simeq An^{-\lambda}$ or $\epsilon_n \equiv 1 - T_c(n)/T_c(\infty) \simeq C_0 n^{-\lambda}$. Allan[8] has suggested that higher-order corrections to this expression can be simply accounted for by introducing an additional parameter h [$\epsilon_n \approx C_0(n-h)^{-\lambda}$].

Table I summarizes theoretical results for C_0 λ and h obtained by Ritchie and Fisher[9] and by Allan[8] by analysis of cubic symmetry films having $n = 1,2 \ldots 6$. These theoretical values are based on various assumptions (specified in the table) and also the assumption that the asymptotic form of ϵ_n remains accurate at small n (excellent theoretical fits are obtained even for $n = 1$ and $n = 2$).

Based on these results, we have also fit our experimental data for $T_c(n)$ vs n using the above expressions for ϵ_n to obtain best-fit parameters for λ with various values of h. Figure 4 displays our experimental data and curves that fit the points for several sets of parameters. For $h = 0$, good fits result for $C_0 = 2.3$ and $\lambda = 1.45$ including $n = 1$ as a point to be fit. As we vary h toward larger positive values we obtain improved fits up to $h = 0.6$ where we obtain the best fit (as measured by a linear correlation coefficient) with parameters $C_0 = 1.0$, $\lambda = 1.01$. By varying h toward negative values, we obtain improved fits up to $h \approx -0.4$ where we get the parameters $C_0 = 1.1$ and $\lambda = 1.04$ and the same quality of fit (based on the correlation coefficient) as for the $h = 0.6$ case.

In summary, $p(1\times1)$-Ni films on Cu(111) yield a critical exponent $\beta = 0.24 \pm 0.07$ in excellent agreement with the result obtained for $p(1\times1)$-Fe films on Au(100). Both systems exhibit in-plane magnetization. If the nearly identical exponent for these systems, which have different 2D symmetry (fourfold versus sixfold), is an indication that they are members of the same universality class, then the in-plane symmetry differences of the films are apparently unimportant. The shift exponent λ obtained from measurements of $T_c(n)$ appears to be consistent with corresponding results obtained using polycrystalline films, also suggesting in-plane symmetry does not play a dominant role in determining critical behavior.

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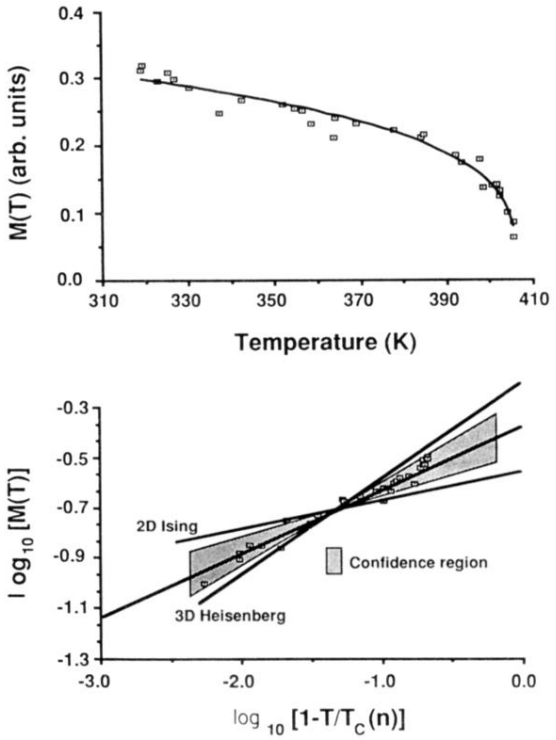
[1]K. Wilson, Sci. Am. **241**, No. 2, 140 (1979).
[2]M. E. Fisher, Rep. Prog. Phys. **30**, 615 (1967); Rev. Mod. Phys. **46**, 597 (1974).
[3]K. Binder, in *Phase Transitions and Critical Phenomena,* edited by C. Domb and J. L. Lebowitz (Academic, New York, 1983), Vol. 8.
[4]M. N. Barber, in *Phase Transitions and Critical Phenomena,* edited by C. Domb and J. L. Lebowitz (Academic, New York, 1983), Vol. 8.
[5]R. M. Suter and C. Hohenemser, J. Appl. Phys. **50**, 1814 (1979).
[6]F. Y. Wu, Rev. Mod. Phys. **54**, 235 (1982).
[7]D. C. Mattis, *The Theory of Magnetism II* (Springer-Verlag, Berlin, 1985); C. N. Yang, Phys. Rev. **85**, 808 (1952).
[8]G. A. T. Allan, Phys. Rev. B **1**, 352 (1970).
[9]D. S. Ritchie and M. E. Fisher, Phys. Rev. B **7**, 480 (1973).
[10]C. Rau, G. Xing, and M. Robert, J. Vac. Sci. Technol. A **6**, 579 (1988).
[11]W. Durr, M. Taborelli, O. Paul, R. German, W. Gudat, D. Pescia, and M. Landolt, Phys. Rev. Lett. **62**, 206 (1989).
[12]R. Bergholz and U. Gradmann, J. Magn. Magn. Mater. **45**, 389 (1984).
[13]H. Lutz, J. D. Gunton, H. K. Schurmann, J. E. Crow, and T. Mihalisin, Solid State Commun. **14**, 1075 (1974).
[14]M. Henzler, Surf. Sci. **73**, 240 (1978).
[15]S. P. Tear and K. Röll, J. Phys. C **15**, 5521 (1982).
[16]U. Gradmann, Ann. Phys. (Leipzig) **17**, 91 (1966).
[17]K. H. Frank, R. Dudde, H. J. Saguer, and W. Eberhardt, Phys. Rev. B **39**, 940 (1989).
[18]D. W. Gidley, Phys. Rev. Lett. **62**, 811 (1989).
[19]J. Araya-Pochet, C. A. Ballentine, and J. L. Erskine, Phys. Rev. B **38**, 7846 (1988).
[20]R. M. Suter and C. Hohenemser, J. Appl. Phys. **50**, 1814 (1979).
[21]M. Schick, Prog. Surf. Sci. **11**, 245 (1981).
[22]C. Domb, J. Phys. A **6**, 1296 (1973).



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