Dimensional Crossover in Ultrathin Ni(111) Films on W(110)

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In situ magnetic resonance measurements of ultrathin Ni(111) on W(110) prepared in ultrahigh vacuum have been carried out to determine the thickness dependence of the critical exponent β and the Curie temperature T_C. We find that β changes drastically from 0.30 to 0.13 between 7 and 5 monolayers. We attribute this behavior to a crossover from a three- to a two-dimensional magnetic behavior. The observed T_C's and linewidths also exhibit the effect of the dimensional crossover. Ni films ≤ 4 layers thick show a truly two-dimensional magnetic phase transition with $\beta = 0.13$.

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The investigation of the magnetic properties of thin films has recently attracted much attention. Intensive research has focused on the magnetic phase transition [1-5], the magnetic moment [6-8], and the magnetic anisotropy energy [9-11]. Our understanding of the magnetic phase transition has advanced considerably through theoretical and experimental effort. Theoretically, one predicts that in a magnetic system the long-range order parameter, the magnetization M, as a function of temperature T disappears at the Curie temperature T_C according to $M \propto (1-T/T_C)^{\beta}$. For three-dimensional (3D) Heisenberg and Ising systems β should be 0.365 and 0.325 [12]. For two-dimensional (2D) systems β is expected to be in the 0.1-0.15 range, in particular $\beta = 0.125$ for a 2D Ising system [13,14]. Experimentally, the 2D behavior of the magnetization of magnetic thin films has been studied recently with improved experimental techniques [1-3,15]. β has been found to range from 0.127 to 0.24 for 2D Fe, Ni, and V films. However, for a given temperature range the thickness dependence of β has never been investigated over a large thickness range (3D $\rightarrow$ 2D), since experiments at high temperatures close to T_C are complicated and are faced with the danger of interdiffusion.

Magnetic lattices of dimensionality 1, 2, and 3 were investigated in bulk for "simple magnetic model systems" a long time ago [16]. A crossover as a function of T from 3D to 2D for diverging correlation length $\xi(T)$ close to T_C has been detected many times. However, only the UHV ultrathin film technique allows the preparation of layer-by-layer films. In this Letter we present the first study of the critical behavior and its crossover from 3D to 2D for a fixed temperature range. The Ni(111)/W(110) system was selected, because this system is one of the few cases where layer-by-layer growth up to 30 and more monolayers (ML) is well known [5,17] and interdiffusion for ≤ 600 K is negligible [17]. Ni films on W(110) are stable and magnetic measurements from 300 to 600 K were always reversible [5]. In comparison with bulk Fe and Co, Ni has a low Curie temperature $T_C = 630$ K. This permits one to perform the experiments over a very wide range of temperatures even above T_C.

Ni films ranging in thickness from 20 to 2 ML were grown epitaxially in UHV below 5×10^{-11} mbar. The W substrate was prepared by the usual Ar^+ sputtering and annealing cycles. No contaminants were found within the detection limit of our Auger system ($<1\%$ of an oxygen monolayer). The Ni films were evaporated from a 99.999%-pure Ni ingot onto W(110) at room temperature. Epitaxy was checked by Auger spectroscopy, low-energy electron diffraction (LEED), and a quartz microbalance. We find the same layer-by-layer growth [5] as was determined by Bauer and co-workers [17]. Up to a coverage of 0.75 monolayer, the Ni film grows pseudomorphically as bcc (110). Upon further evaporation a strained fcc (111) Ni monolayer forms, which exhibits a 7×1 superstructure in the LEED pattern [a superposition of a bcc (110) and a fcc (111) pattern]. A LEED picture taken at 6 ML exhibits the usual hexagonal Ni(111) symmetry only. In agreement with [17] we conclude that from 2 ML on, Ni grows in a fcc (111) structure layer by layer. Magnetic resonance at 9 GHz was performed *in situ* on freshly evaporated films. The external magnetic field was parallel to the film plane and oriented along the [001] direction of the bcc (110) substrate. This is identical to the [$1\bar{1}0$] direction of the fcc (111) Ni film.

For transition metals like Ni and Fe, the ferromagnetic resonance (FMR) which occurs below T_C has a linewidth $\Delta\tilde{H}$ that can be written as [5,18,19]

$$\Delta\tilde{H}(T) = \Delta H_{pp} - \Delta H_0 = K(1-T/T_C)^{-\beta}, \quad (1)$$

where ΔH_{pp} is the measured peak-to-peak resonance linewidth and ΔH_0 is the residual linewidth, which is frequency and temperature independent and is caused by magnetic inhomogeneities [18] and an exchange contribution of the conduction electrons [19]. The constant K is temperature independent [5]. According to Eq. (1), the FMR linewidth is a divergent quantity at T_C^-. Our experimental data agree well with this expectation. It may be surprising that one deduces the static critical exponent β from the FMR linewidth. But this is just a consequence of the dissipation-fluctuation theorem.

Figure 1 shows the temperature dependence of the resonance linewidth $\Delta\tilde{H}$ for Ni films ranging in thickness from 20 to 2 ML. For every film a peaklike broadening of $\Delta\tilde{H}$ as a function of temperature was observed. This

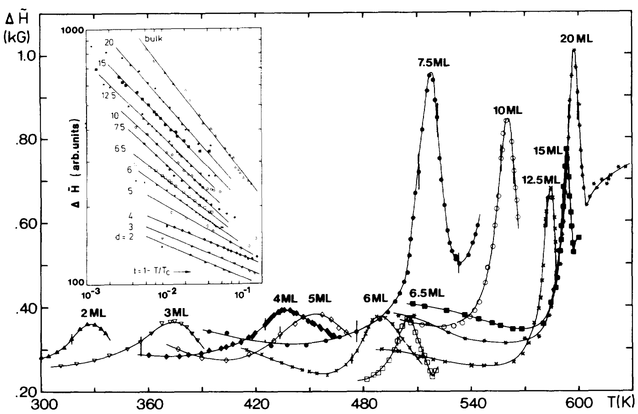

FIG. 1. Linewidth $\Delta\tilde{H}$ as a function of T. For Ni(111) 1 ML corresponds to a layer spacing of 2.035 Å. The solid lines are guide to the eyes. ΔH_0 in the thinner films is larger than in the thicker ones. A similar effect was found in Fe films [30]. Inset: A log-log plot of $\Delta\tilde{H}$ as a function of the reduced temperature t. The bulk data are taken from [20]. The "bending off" from the power law at low t may be attributed to a crossover as a function of T. But it is also likely and known from experiments in the bulk that dipolar interaction and other effects limit the increase of ΔH very close to T_C. The data do not allow one to distinguish between these possibilities.

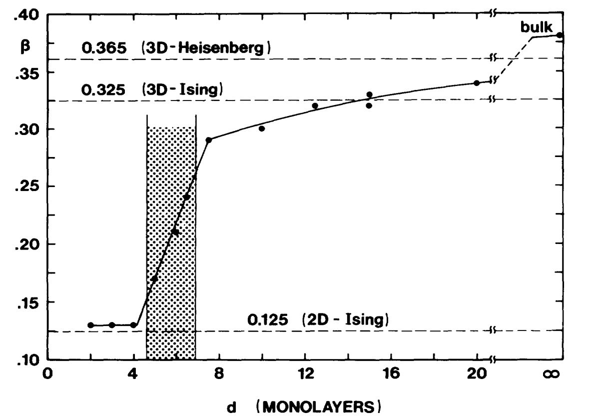

FIG. 2. Critical exponent β as a function of film thickness (error bars are given in Table I). The dashed lines show the theoretical values for a 3D Heisenberg, 3D Ising, and 2D Ising system. The shaded regime marks the crossover from 3D to 2D.

result is consistent with an earlier observation in bulk Ni [20]. It is attributed to critical spin fluctuations in the region of the phase transition and it is to our knowledge one of the most direct techniques determining the phase transition. It occurs at T_{max} (corresponding to $\Delta\tilde{H}_{max}$) or slightly below as discussed in [5]. Doing a least-squares fit as described in [5] we determined β and T_C for Ni films from Eq. (1). It is worthwhile to mention that T_C is not a free parameter; T_C is given first by visual inspection as T_{max}. Only in a second-order refinement of the fitting procedure do we allow T_C to vary over a few degrees. The best fits are shown in the inset in Fig. 1 as solid lines. The decreasing slope for thinner films corresponds to a decrease of β. The experimental values obtained for β and T_C as well as β for theoretical models are given in Table I.

TABLE I. Values of the critical exponent β, and the Curie temperature T_C for different Ni samples. The error bar on T_C is <2 K. For 15 ML two samples were prepared: (a) on a smooth W substrate and (b) on a rough substrate. The anisotropy field changes [21], but β and T_C show no effect within error bars.

d (ML)	β	$T_C(d)$ (K)	Reference
∞	0.365		3D Heisenberg
∞	0.325		3D Ising
Bulk	0.38(4)	630	This work and Ref. [20]
20	0.34(4)	596	This work
15(a)	0.33(4)	592	This work
15(b)	0.32(4)	593	This work
12.5	0.32(4)	583	This work
10	0.30(5)	557	This work
7.5	0.29(6)	512	This work
6.5	0.24(6)	504	This work
6	0.21(6)	487	This work
5	0.17(6)	451	This work
4	0.13(6)	435	This work
3	0.13(6)	371	This work
2	0.13(6)	325	This work
...	0.125		2D Ising

The interesting point to note from Table I is the drastic change of the critical exponent β. This can be further clearly seen in Fig. 2, where β is plotted as a function of thickness. Figure 2 indicates a discontinuity in the change of β with the thickness of the Ni films. The thickness dependence of β can be divided into three regions: a monotonic decrease of β with a thickness decrease from bulk to 7.5 ML, a drastic reduction of β between 7 and 5 ML, and a flat region for thicknesses ≤ 4 ML. In the first region β shows a monotonic change from 0.38 to 0.29. The decrease of β may be interpreted as a crossover from 3D Heisenberg to 3D Ising, due to an increasing anisotropy energy [21,22]. The value of β in the second region exhibits a sudden change between 7 and 5 ML. This area clearly indicates that there is a significant change from 3D to 2D Ising [23]. The behavior of the peak in $\Delta\tilde{H}$ (Fig. 1) is consistent with this. In 2D the fluctuations are smeared out over a larger temperature range. Additional evidence for the change of the dimensionality of the system is the behavior of T_C (Fig. 3), which deviates from the values expected of a 3D system below 7 ML (to be discussed in the next paragraph). In the third region $\beta=0.13$ is independent of the film thickness. This independence shows that Ni films ≤ 4 ML thick exhibit a behavior like 2D Ising systems in the temperature range of $10^{-3}<t<10^{-1}$, which was also observed in 1- to 3-ML Fe films [1,3]. The value of β in this region agrees

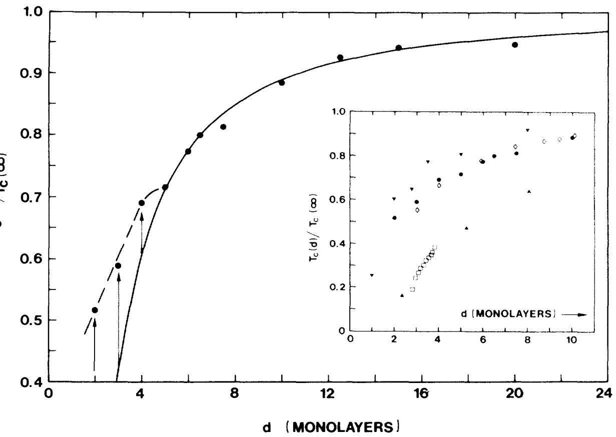

FIG. 3. $T_C(d)$ plotted as a function of film thickness (for error bars see Table I). $T_C(d)$'s are normalized to bulk Ni [$T_C(\infty)=630$ K]: The solid line is the result of a fit obtained by using Eq. (2) with $\lambda=1.4$ and $C_0=2.7$ (note $T_{C,\text{bulk}}$ is included in the fit). The dashed line is a guide to the eyes. The arrows indicate the T_C enhancement (see text). Inset: Several systems are compared; Ni(111)/W(110) from this work, Ni(111)/Cu(111) from Ref. [2] (▼), Ni(111)/Re(0001) from Ref. [11] (◇), Au/Ni/Au(111) from Ref. [28] (□), and Cu/Ni/Cu(100) from Ref. [29] (▲).

very well with the theoretical 3D Ising value. From the above analysis we conclude that a change from a 3D to a 2D system in Ni films happens between 7 and 5 ML. The magnetic coherence distance between the magnetic moments in Ni is about 3 ML (in the temperature range of $10^{-3}<t<10^{-1}$).

Now we discuss the thickness dependence of T_C. According to Refs. [24,25] $T_C(d)$ can be described by a power law

$$[(T(\infty)-T_C(d)]/T_C(\infty)=C_0 d^{-\lambda}. \quad (2)$$

The constant C_0 ranges from 1 to 10 in various experiments. The shift exponent λ is related to the critical exponent ν [correlation length $\xi=\xi_0(|1-T/T_C|)^{-\nu}$] by $\lambda=1/\nu$. The thickness dependence of T_C is shown in Fig. 3. Using Eq. (2) we fitted our experimental T_C as a function of C_0 and λ. The solid line in Fig. 3 uses $C_0=2.7(5)$ and $\lambda=1.4(1)$ ($\nu=1/\lambda=0.71$), which agrees perfectly with the theoretical value for a 3D Heisenberg system. The finite-size scaling of Eq. (2) is an asymptotic limit for a 3D sample with finite thickness. Therefore one expects Eq. (2) to hold only in a 3D regime, which is indeed the case. It is worthwhile to mention that in the literature Eq. (2) is occasionally used to scale down to 1 ML or zero thickness. This is not meaningful as stated above. Depending on the values of C_0 and λ Eq. (2) already yields unphysical results at ≈ 2 ML. Our experimental data depart from the behavior (solid line in Fig. 3) at exactly the same thickness $d=4$ ML at which β indicates 2D behavior. The experimentally determined T_C's for 4 and 3 ML are enhanced by approximately 20% and 40%, respectively, and the calculated $T_C(2\text{ ML})\to 0$ from Eq. (2). In other words, Eq. (2) predicts $T_C(3\text{ ML})/T_C(\text{bulk})=0.43$, but one measures 0.59. This enhancement may be due to a different asymptotic value of a 2D Ising system, an enhancement of the magnetic moment of the outermost Ni layers, or interface effects [8,26,27].

In the inset in Fig. 3 we compare our results with other Ni films given in the literature. The results for Ni(111)/Re(0001) [11] (◇) agree very well with Ni/W. Both systems are prototypes for no interdiffusion. However, from the Ni/Re data the 3D→2D transition cannot be deduced (missing experimental data). The data of Ni(111)/Cu(111) [2] (▼) show large scatter and were not reproducible after thermal treatment. Au/Ni/Au(111) and Cu/Ni/Cu(100) thin films [28,29] (□ and ▲) show systematically lower T_C's with respect to the nominal thickness. Renard and Beauvillain [28] argue that the Au and Cu cover and substrate sheets change the Ni band structure and as a consequence reduce the magnetic moment per Ni atom. From the present study we conclude that there is not just one dead layer on each side.

Finally we turn back to the peaklike broadening of $\Delta\tilde{H}$ at T_C. It can be seen from Fig. 1 that $\Delta T_{1/2}$, the width of this broadening (FWHM), increases for thinner films. A quantitative analysis yields that $\Delta T_{1/2}\approx 10$ K for the 3D regime and increases steplike to ≈ 40 K for the 2D regime. It is interesting to note that $\Delta T_{1/2}$ for the bulk equals only 7 K. That means the spin fluctuations appear in a temperature range of 10^{-2} only, whereas in the 2D regime (films of 2 to 4 ML) the fluctuations extend over $\Delta T_{1/2}\approx 40$ K, that is, 10^{-1}. The change happens at the same thickness at which β changes. This gives us one more independent argument for the dimensional crossover. For quantitative analyses a more rigorous theory of the FMR and ESR linewidth at the phase transition is needed.

In conclusion, we have shown a crossover in the magnetic properties of Ni films as a function of thickness. In principle this should be detectable also for the thicker films at temperatures $t\ll 10^{-3}$; however, other effects suppress it. The present investigation is the only one which in practice determines the layer thickness d below which Ni behaves 2D-like. This crossover is surprisingly sharp between 5 and 7 ML (10→14 Å). Ni films thinner than 4 ML exhibit a 2D phase transition with a constant β, which shows that the universality hypothesis applies to the magnetic transition of Ni films. The critical exponents determined in our experiment agree very well with the known exponents for a 2D Ising system and bulk Ni.

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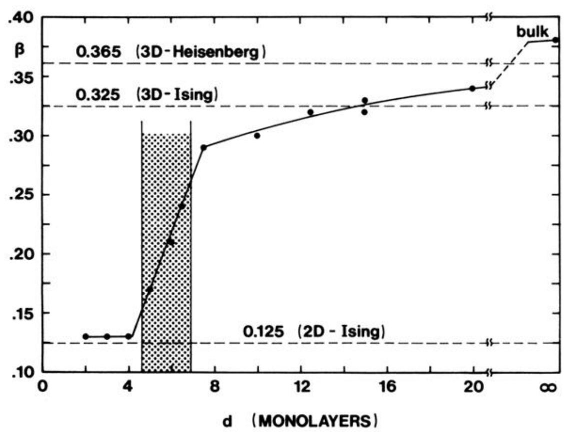

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