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Critical spin fluctuations and Curie temperatures of ultrathin Ni(111)/W(110): A magnetic-resonance study in ultrahigh vacuum

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Ferro- and paramagnetic resonance is performed *in situ* in ultrahigh vacuum on 15- to 40-Å thin Ni(111) films from 300 to 630 K. As a function of temperature a huge peaklike broadening of the linewidth ΔH_{pp} is observed. From the thickness dependence of this peak we deduce the Curie temperatures $T_c(d)$ of the respective films with thickness d, e.g., T_c (15 Å) = 512 K = $0.81T_c$(bulk). The shift exponent λ of the thickness dependence of T_c is determined to be 1.42 ± 0.3. The temperature dependence of ΔH_{pp} below T_c is analyzed in terms of $\Delta H_{pp} \propto (1-T/T_c)^{-\beta}$, with β the critical exponent of the spontaneous magnetization. β ranges from 0.38 for bulk Ni to 0.29 for a 15-Å film.

Recently it has been demonstrated that paramagnetic (EPR) (Ref. 1) and ferromagnetic (FMR) resonance[2] can be performed *in situ* on well-characterized magnetic monolayers in UHV. The magnetic properties of Fe and Ni monolayers and multilayers have been studied deep in the ferromagnetic phase[2–4] and valuable information on the thickness dependence of the magnetic anisotropy in these films was obtained. In the present Rapid Communication, however, we want to focus on the magnetic properties near the paramagnetic-to-ferromagnetic phase transition. In this regime magnetic quantities, such as the static susceptibility χ_0 and the spontaneous magnetization M_{sp}, follow a power law with respect to the reduced temperature $|T-T_c|/T_c = t$. For Gd monolayers and/or multilayers on W(110), we were able to determine the Curie temperatures $T_c(d)$ and the critical exponent γ of $\chi_0(T>T_c)$ as a function of film thickness d.[1] Critical broadening of the EPR linewidth has been observed in bulk ferromagnets near T_c.[5,6] In the case of bulk Ni only one[6] of numerous magnetic resonance experiments showed a resonancelike broadening when approaching T_c (630 K) from below and also from above. In all other resonance experiments[7–10] a broadening was found only for $T \to T_c^-$, but no subsequent decrease in the linewidth above T_c. The result of the present work is to show that epitaxial films grown under controlled conditions in UHV are "more perfect" solids than previously[7–10] used bulk materials. Biller and co-workers[6,11] have demonstrated that it is possible to obtain the FMR linewidth ΔH_{pp} of only 102 G for bulk Ni at room temperature by very careful annealing in vacuum and strain-free mounting. For thin Ni films, ΔH_{pp} ranges from 300 to 500 G, depending on film quality.[12]

Another point of interest is the study of the thickness dependence of the Curie temperature. Bergholz and Gradmann[13] and recently Ballentine *et al.*[14] studied $T_c(d)$ of uncovered Ni(111)/Re(0001) and $p(1\times1)$ Ni/Cu(111) by magnetometry and the magneto-optic Kerr effect in UHV. Allan has shown[15] that the phase-transition temperature T_c is reduced from the bulk value, as soon as the diverging correlation length $\xi = \xi_0 t^{-\nu}$ reaches geometrical dimensions of the sample. This is a purely geometrical argument, it does not depend on the type of interaction. A depression of $T_c(d)$ starts already for 50- to 20-layers thin films (as has been shown in Ref. 13) far in the 3D regime. According to[15,16] this $T_c(d)$ can be described by a power law

$$[T_c(\infty) - T_c(d)]/T_c(\infty) = c_0 d^{-\lambda}. \qquad (1)$$

The constant c_0 ranges from 1 to 10 in various experiments. The shift exponent λ is related to ν by $\lambda = 1/\nu$.[16] With $\nu = 0.705$ for a three-dimensional (3D) Heisenberg system one obtains $\lambda = 1.42$. The experimental shift exponent λ for Ni films is found to be between 1.01 ± 0.1 and 1.44 ± 0.2 in Ref. 14. Bergholz and Gradmann[13] obtain $\lambda = 1.27 \pm 0.2$.

In the present communication we will present experimental results on the thickness dependence of T_c and on the FMR and/or EPR linewidth near T_c of fcc Ni(111) films on W(110). The ferromagnetic resonance in metallic samples is described by the Landau-Lifshitz equation of motion,[17]

$$\frac{d\mathbf{M}}{dt} = -\gamma(\mathbf{M}\times\mathbf{H}_{\text{eff}}) + \frac{G}{\gamma M(H,T)^2}\left[\mathbf{M}\times\frac{d\mathbf{M}}{dt}\right], \qquad (2)$$

with the Gilbert form of damping. This leads to[12,17]

$$\Delta H_{pp} = \Delta H_0 + 1.16\frac{\omega G}{\gamma^2}\frac{1}{M(H,T)}, \qquad (3)$$

where the frequency independent part ΔH_0 is caused by magnetic inhomogeneities[2] and an exchange contribution of the conduction electrons.[9] ω is the microwave frequency, γ the gyromagnetic ratio, and G the Gilbert damping parameter. The latter has been shown[7] to be frequency independent for Ni in the vicinity of T_c. $M(H,T)$ is the magnetization in an applied field H at a temperature T.[12] Since a rigorous theory of the FMR linewidth does not exist and because $M(H,T)$ in small fields and for reduced temperatures of $t \approx 10^{-2}$ differs only very little from the spontaneous magnetization $M_{sp}(T)$, we replace $M(H,T)$ by $M_{sp}(T)$ in Eq. (3). This leads to a new scaling of the

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FMR linewidth by the inverse of the order parameter M_{sp},

$$\Delta\tilde{H} \equiv \Delta H_{pp} - \Delta H_0 = K t^{-\beta} , \quad (4a)$$

$$M_{sp} = M(T = 0\ \mathrm{K}) t^{\beta} . \quad (4b)$$

According to this, the inverse of the spontaneous magnetization and the FMR linewidth are divergent quantities at T_c^- [Eq. (4a)]. We will present some experimental evidence which shows that β, determined from our data and Eq. (4), agrees well with the bulk value $\beta = 0.38(2)$ for Ni. Moreover we see a decreasing β for thinner films, which is discussed at the end.

Our films are sublimed from 99.999% clean Ni bulk material onto W(110) in a vacuum $< 2\times10^{-10}$ mbar during evaporation (base pressure $< 5\times10^{-11}$ mbar). The substrate is at room temperature. The samples are clean within the detection limit of our Auger system ($< 1\%$ of an oxygen monolayer). Epitaxy is checked by low-energy electron diffraction (LEED). We find the same layerwise growth modus, as has been determined earlier by Kolaczkiewicz and Bauer.[18] The amplitude of the Ni (61 eV) Auger peak is plotted in Fig. 1, as function of evaporation time. Four distinct slope changes are seen. Up to a coverage of $0.75\Theta_1$ the Ni monolayer grows pseudomorphic, that is "bcc (110)." Upon further evaporation a strained more densely packed fcc(111) Ni monolayer forms which is complete at Θ_1. A LEED picture taken at a coverage $6\Theta_1$ (i.e., six monolayers) shows an undistorted hexagonal Ni(111) surface. The layer spacing equals $d = 2.035$ Å.

Magnetic resonance at 9.0 GHz is performed *in situ* on freshly evaporated films with the static magnetic fields in the film plane.[19] In Fig. 2(a) we show typical experimental FMR spectra of a 15-Å (curve *a*) and a 40-Å (curves *b* and *c*) Ni film near T_c. The high sensitivity of our *in situ* UHV-FMR apparatus is evident from the excellent signal-to-noise ratio of the 15-Å spectrum at $T/T_c(15\ \text{Å}) = 0.85$. The line shape is symmetric since d is smaller than the skin depth. In Fig. 2(b) we show the peak-to-peak linewidth ΔH_{pp} as function of temperature. A huge and narrowly peaked line broadening as function of temperature is observed. This is attributed to critical fluctuations in the thin film. It is well known[20] that approaching T_c from the paramagnetic side a critical "speeding up" of the exchange narrowed relaxation rate sets in near T_c. This is followed by a so-called thermodynamically "slowing down" of the critical fluctuations at and below T_c. This behavior has also been observed in insulating ferromagnets. The only experiment on Ni where this has been seen is the work of Spörel and Biller.[6] In the present experiment [Fig. 2(b)] the narrow peak in ΔH_{pp} is only achieved when using freshly prepared W(110) crystal surfaces.

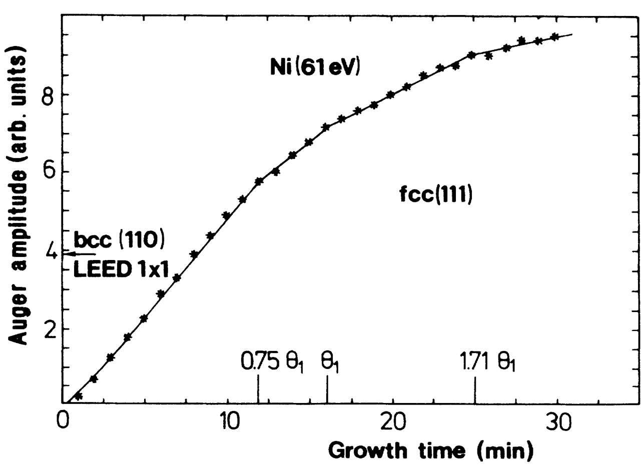

FIG. 1. The Auger peak amplitude of Ni(61 eV) as a function of evaporation time. The completion of a Ni monolayer is denoted by Θ_1. Clear changes in the slope are observed at the indicated times. Above a coverage of $0.71\Theta_1$ a (7×2) overstructure is seen in LEED which changes continuously to the hexagonal spot pattern of a fcc(111) surface (for details see Ref. 18).

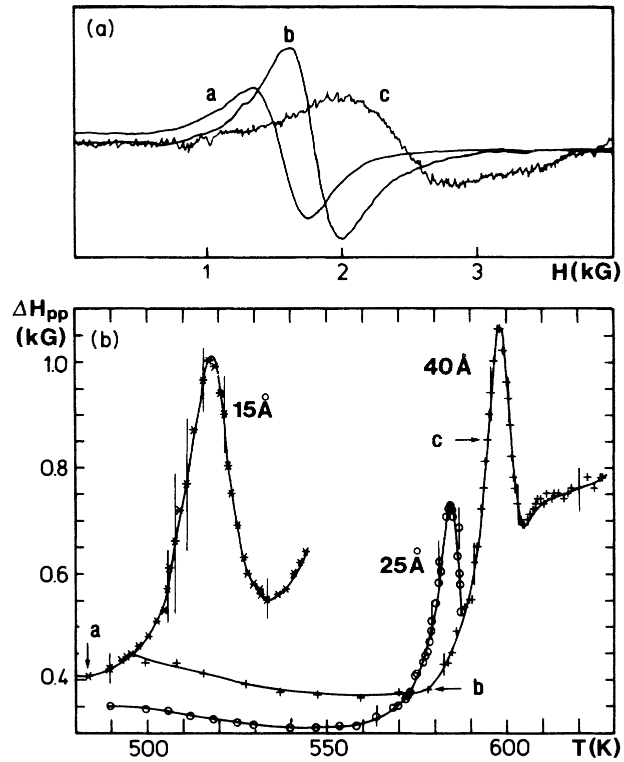

FIG. 2. (a) Typical experimental spectra of 15 Å (curve *a*) and 40 Å (curves *b* and *c*) Ni(111) on W(110) recorded at 9 GHz. (b) Peak to peak linewidth as function of temperature. ΔH_{pp} of spectra in Fig. 2(a) are indicated.

The phase transition occurs at T_{max} (corresponding to ΔH_{max}) or slightly below.[5] An exact quantitative theoretical expression is not known. Using Eq. (4a) with $K = 1.16\omega G/\gamma^2 M$ ($T = 0$ K), we try to determine β having ΔH_0 and T_c as parameters. Bhagat and co-workers[9] gave an estimate for ΔH_0, stating that ΔH_0 should be lower than 10% of the linewidth measured at room temperature, that is $\Delta H_0 \lesssim 35$ G in our case. Heinrich, Cochran, and Hasegawa[12] have calculated ΔH_0 for disordered ferromagnetic metals. They find values for ΔH_0 which are well below 100 G. The parameter K is assumed temperature independent. This is satisfied according to Refs. 7-9 in the vicinity of T_c. The upper limit of T_c is given by T_{max}. Varying ΔH_0 from 20 to 80 G and allowing T_c to be $(T_{max}-10\text{ K}) < T_c \leq T_{max}$ we obtain as best fits the curves shown in Fig. 3. As best fits we define a fit to a theoretical temperature dependence with the largest possible reduced temperature interval and the lowest least-squares deviation of the experimental data. For K we obtain 94.5 G which is within 10% equal to a theoretical K computed with the known Gilbert damping $G=2.45\times10^8$ s^{-1},[21] the microwave frequency 9.0 GHz and with a saturation magnetization $M(T=0\text{K})$ taken from Ref. 13. In addition we analyzed the FMR of bulk Ni (x in Fig. 3) from Ref. 6. We obtain the critical exponent $\beta=0.38(4)$ in agreement with Ref. 22. The results of our fitting procedure are listed in Table I. For the least-squares-fitted analysis β, ΔH_0 and T_c are correlated. A change of T_c within the 10-K limit effects β very little. The error bar for β corresponds to a change of ΔH_0 from 20 to 80 G. The best T_c value appears to be 2–5 K below T_{max}. The solid lines (fits) in Fig. 3 show a decreasing slope for thinner films, which is in agreement with other work (see Table I). It is worthwhile to note that the fitted reduced temperature interval approaches T_c down to $t\approx3\times10^{-3}$, which is almost one decade smaller than in Ref. 14. For completeness we have also included the theoretical β of different magnetic models in Table I. The decrease of β for thinner films may be interpreted in two ways. First, the effect of lowering the dimensions towards 2D behavior causes a quasicontinuous decrease of the effective β determined in our experiment (3D Heisenberg to 2D anisotropic Heisenberg). Second, comparing $\beta=0.29$ of our 15-Å (7.5 layers) film to the 3D Ising system, one sees that the smaller β may be also due to an increase of the magnetic anisotropy in the plane of the thinner film, which then corresponds to a change from a 3D Heisenberg to a 3D Ising system in the limit of infinitely strong uniaxial anisotropy. We have experimental evidence for this; a detailed analysis of the magnetic anisotropy as function of film thickness will be published elsewhere.

The thickness dependent Curie temperatures $T_c(d)$ from Refs. 13 and 14 as well as from the present work are plotted in Fig. 4. To extract the second critical exponent ν from our measurements we fit $T_c(d)$ according to Eq. (1). We obtain $\lambda=1.42\pm0.3$ which agrees with the expected value for a 3D Heisenberg system. The constant

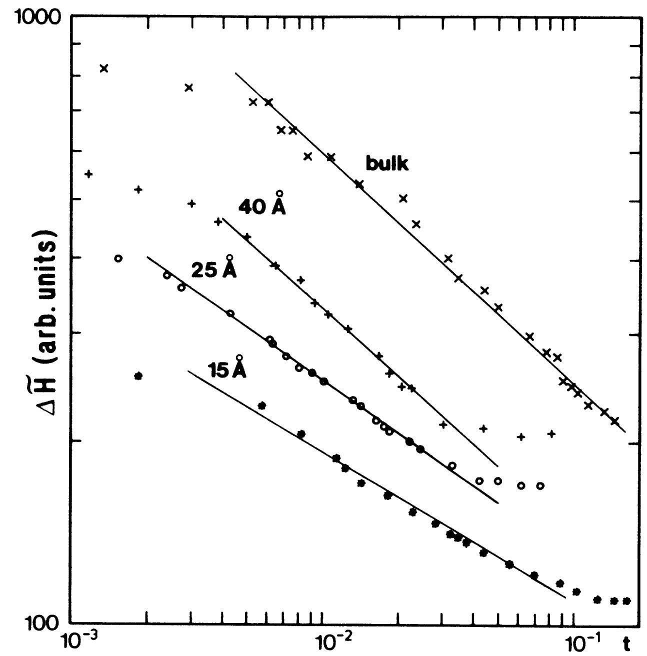

FIG. 3. Double logarithmic plot of the experimental peak to peak linewidths as function of reduced temperature for the bulk Ni sample of Ref. 6 and our data in Fig. 2(b). Curves have arbitrary vertical offset for clarity. The same residual linewidths $\Delta H_0=50$ G are subtracted from all experimental values. Straight lines are best fits as outlined in the text. The parameters T_c and β are summarized in Table I.

$c_0=2.8\pm1$ is within the known limits.[13,14]

Most of the experiments exploring critical phenomena in thin films limit themselves to determine *one* power law and *one* critical exponent. Strictly speaking critical phenomena need to fulfill scaling relations as

$$2\beta+\gamma=\nu D\,, \tag{5}$$

where D is the spatial dimension. The present experiment gives some hint to determine at least two exponents (β,ν) for a 3D Heisenberg system. γ is expected to range from 1.241 to 1.386 depending on the type of interaction. With the β value of the bulk sample (Table I) and $\nu=0.704$ from our experiment we calculate from Eq. (5) a $\gamma=1.35$.

TABLE I. Curie temperatures, critical exponents β, and the reduced temperature interval Δt of different Ni samples. Thickness d is given in number of layers. The error bar in T_c is < 2 K.

$T_c(d)$ (K)	β	Δt	d	Reference
	0.365	· · ·	∞	3D Heisenberg
	0.302	· · ·	∞	3D Ising
630	0.38 ± 0.04	0.005–0.1	Bulk	This work, and Ref. 6
596	0.34 ± 0.04	0.003–0.04	19.7	This work
583	0.32 ± 0.04	0.003–0.04	12.3	This work
580	0.32 ± 0.09	Not given	8.0	Ref. 14
512	0.29 ± 0.06	0.005–0.05	7.5	This work
380	0.24 ± 0.07	0.01–0.31	2.6	Ref. 14
	0.125	· · ·	1	2D Ising

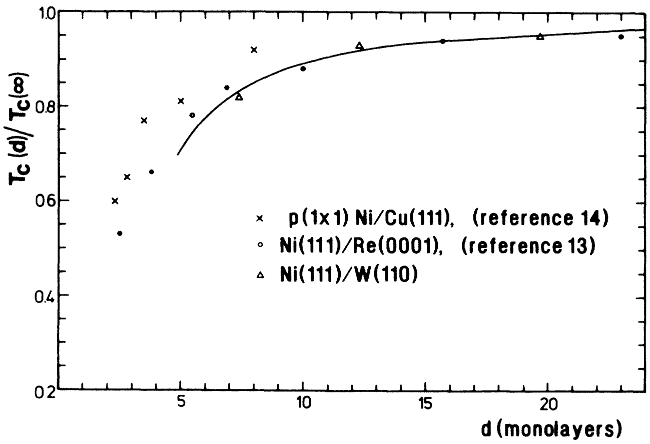

FIG. 4. The Curie temperature $T_c(d)$ of uncovered Ni films in UHV from Refs. 13 and 14 is plotted together with our data as function of d. $T_c(d)$ values are normalized with respect to bulk Ni $T_c(\infty)=630$ K. A fit according to Eq. (1) yields for $p(1\times1)$Ni/Cu(111): $\lambda=1.44\pm0.2$, $c_0=2.3$; for Ni(111)/Re(0001): $\lambda=1.27\pm0.13$, $c_0=1.9\pm0.5$. We obtain $\lambda=1.42\pm0.3$, $c_0=2.8\pm1$ (solid line).

We like to mention that recently it became possible to measure the *ac* (initial) susceptibility of ultrathin films in a direct way *in situ* in UHV (Ref. 23).

In conclusion we have demonstrated that it is possible to observe critical broadening of the FMR linewidth in ultrathin Ni films on W(110), which is due to magnetic fluctuations. From this we determine the Curie temperature $T_c(d)$ and confirm the findings of Bergholz and Gradmann for Ni(111)/Re(0001).[13] The well-known relation (3) for the FMR linewidth is used for the first time to determine the critical exponent β. We find critical exponents β which agree very well with known exponents in the bulk and also in thin Ni films. A decrease of β with decreasing film thickness is found. The usefulness of magnetic resonance to obtain information on the critical behavior of magnetic ultrathin films is demonstrated once again.

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