

# Magnetic anisotropy of Ni modified by extreme lattice expansion

Y. Manzhur<sup>1</sup>, P.M. Imielski<sup>2</sup>, K. Potzger<sup>1</sup>, W.D. Brewer<sup>2</sup>, M. Dietrich<sup>3</sup>, M.J. Prandolini<sup>1,2,a</sup>, and H.H. Bertschat<sup>1</sup>

<sup>1</sup> Bereich Strukturforschung, Hahn-Meitner-Institut Berlin GmbH, 14109 Berlin, Germany

<sup>2</sup> Institut für Experimentalphysik (WE1), Freie Universität Berlin, 14195 Berlin, Germany

<sup>3</sup> Technische Physik, Universität des Saarlandes, 66041 Saarbrücken, Germany and The ISOLDE-Collaboration, CERN, 1211 Genève 23, Switzerland

Received 26 January 2005 / Received in final form 9 May 2005

Published online 7 September 2005 – © EDP Sciences, Società Italiana di Fisica, Springer-Verlag 2005

**Abstract.** The induced magnetism of isolated (Cd) adatoms, soft-landed onto ultrathin Ni layers grown pseudo-morphologically on Pd(001) ( $a_{Pd} = 3.8907 \text{ \AA}$ ), was studied using the perturbed angular correlation (PAC) technique. The magnitude of the induced magnetic response (magnetic hyperfine field of  $|5.2| \text{ T}$ ) was found to be ca. 30% smaller than on bulk Ni ( $a_{Ni} = 3.524 \text{ \AA}$ ) surfaces. This result is compared to *ab initio* calculations. Additionally, the magnetic anisotropy of the induced response was found to be modified as compared to bulk Ni surfaces and the induced hyperfine field was observed to emerge from the plane at an angle of  $50(5)^\circ$  to the surface normal. This canted magnetic anisotropy is attributed to the large lattice expansion of the ultrathin Ni film on Pd(001).

**PACS.** 75.30.Gw Magnetic anisotropy – 75.70.-i Magnetic properties of thin films, surfaces, and interfaces – 31.30.Gs Hyperfine Interactions

## 1 Introduction

An important new property provided by quasi two-dimensional ultrathin magnetic films is enhanced *perpendicular magnetic anisotropy* (PMA). At present, almost all magnetic recording of digital data uses conventional (parallel or in-plane) magnetic materials. One method for achieving ultrahigh density magnetic recording (in the Tbit/inch<sup>2</sup> range) is the use of a medium which is magnetised perpendicular to the plane; see reference [1]. Thus it is of technological importance to find new materials with PMA. In terms of fundamental studies, magnetic anisotropy has proved to be a challenge to first-principle calculations. For example, in bulk Ni, even the sign of the magnetic anisotropy constant cannot be predicted [2]. Ultrathin Ni films have been shown to exhibit PMA, through in-plane lattice expansion, by growing Ni on substrates with larger lattice constants: for example, Ni on Cu(001) ( $(a_{Cu} - a_{Ni})/a_{Ni} = 2.5\%$ ) [3], and Ni on Cu<sub>3</sub>Au(001) (6.3%) [4]. The aim of the present study is to investigate the surface of an ultrathin Ni film grown on Pd(001), which exhibits an extreme in-plane expansion of 10.4%. The experiments were conducted using isolated, non-magnetic radioactive probe atoms which were soft-landed onto the surface and then investigated by PAC (perturbed angular correlation) spectroscopy. This method is uniquely sensitive to the magnetic surface properties, achieving atomic resolution.

In previous work, isolated non-magnetic probes were soft-landed onto bulk ferromagnetic surfaces and their induced magnetic properties were determined using PAC spectroscopy (e.g. Se on Ni(001) and Ni(111) [5], and Cd on Ni(001) and Ni(111) single crystals [6–8]). In particular, using Cd probes on Ni surfaces, it was possible to distinguish between probe atoms at different surface lattice sites, i.e., in/on terraces, in/at atomic steps, and at atomic corners. These studies showed that the strength of the induced magnetic polarisation of the Cd atoms is largely determined by their coordination number ( $NN$  – the number of nearest Ni neighbours) [6]; and secondly, that the induced *s*-electron moment on Cd is relatively independent of the local symmetry and surface orientation [7,8]. Recently, these measurements were complemented by theoretical work which supports the experimental conclusions [9,10].

In addition to the induced magnetic responses of Cd probes at various Ni surface sites, such probes, especially adatoms atop the terraces, can provide a sensitive measure of the magnetic alignment of the surrounding nearest-neighbour Ni moments. The Cd electronic magnetic moment, and therefore its hyperfine field, are dominated by the spin polarisation of the 5*s*-electrons; spin-orbital contributions to the electronic magnetic moment and magnetic hyperfine field are very small [9,10]. Thus the direct induced spin polarisation resulting from the *sd*-hybridisation with the Ni nearest neighbours is expected to be collinear, i.e., the alignment of the induced Cd *s*-moment is parallel to that of the magnetic moments of the Ni nearest neighbours. Similar arguments have been theoretically

<sup>a</sup> e-mail: prandoli@physik.fu-berlin.de

tested for the induced polarisation of Ag due to neighbouring Fe surfaces [11]. In previous experimental studies, the surface magnetisation of both Ni(001) and Ni(111) disk-like single crystals was always found to be in the plane. In the present work, we use PAC of the Cd probe adatoms to study the different situation of an ultrathin and highly stressed Ni film grown on a Pd(001) single-crystal substrate.

Generally, as observed for a number of Fe and Co thin films grown on Cu, Ag, Au, Pd, or Pt substrates, the surface anisotropy ( $K_s$ ) orients the magnetisation of the magnetic film perpendicular to the plane below a critical thickness, while the surface anisotropy of Ni is in contrast found to favour in-plane magnetisation [12]. However, the bulk magnetostriction constant ( $\lambda_{001}$ ) of Ni is negative, compared to the positive values found for Co or Fe [13]. Thus, perpendicular magnetism would be observable for ultrathin Ni(001) films if it were possible to apply a compressive stress to the film, resulting in a strain ( $\epsilon_2$ ) in the [001] direction. Assuming coherent epitaxial growth of Ni(001) films on surfaces with larger lattice constants such as Cu or Pd, it would be possible to artificially induce strain and therefore to change the magnetic anisotropy energy balance from one favouring an in-plane to an out-of-plane magnetisation.

The important energy terms which describe the effective magnetic anisotropy  $K_{eff}$  of a magnetic thin film of thickness  $d$  and angle  $\theta$  between the surface normal and magnetisation direction are

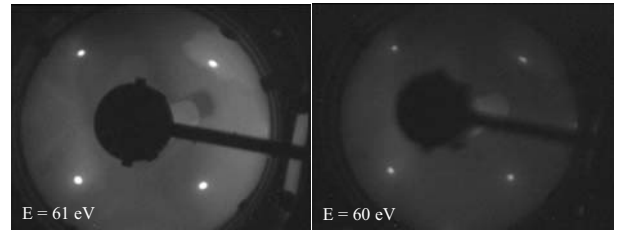
$$E = -K_{eff} \cos^2 \theta = -(K_v + K_{me} + 2K_s/d) \cos^2 \theta, \quad (1)$$

where  $K_v$  ( $= -\mu_0 M_s^2/2$ ; for bulk Ni  $M_s = 522$  kA/m) is the shape anisotropy term and  $K_{me}$  is the magnetoelastic anisotropy

$$K_{me} = \frac{3}{2} \lambda_{001} (c_{11} - c_{12}) (\epsilon_2 - \epsilon_1), \quad (2)$$

where  $\epsilon_{1/2}$  are the in-plane/out-of-plane strains and  $c_{11}$  and  $c_{12}$  are the cubic elastic stiffness constants. This interplay between competing anisotropies has been demonstrated on ultrathin Ni films grown on Cu [3]. Below a critical thickness  $d^* = 7$  monolayers (MLs), the ultrathin Ni films were found to be magnetised in-plane; here the (in-plane) surface anisotropy ( $K_s$ ) dominates at small  $d$ . Above this critical thickness, the magnetic anisotropy was found to be governed by the magnetoelastic term ( $K_{me}$ ) and the film is therefore magnetised perpendicularly. It should be noted that the lattice mismatch between Ni and the slightly larger Cu lattice is only 2.5%. Finally, for much thicker Ni films, where the lattice constant relaxes back to the value of bulk Ni, the in-plane shape anisotropy ( $K_v$ ) dominates.

The epitaxial growth of ultrathin Ni films on Pd(001) was studied recently using angle-scanned photoelectron diffraction and LEED [14]. Up to 6 MLs, a clear  $(1 \times 1)$  LEED pattern was observed, indicating epitaxial pseudomorphic growth of Ni on Pd(001). However, a large out-of-plane tetragonal distortion was predicted by simple linear elastic theory [15] and has been confirmed experimentally, even though the lattice mismatch for the



**Fig. 1.** LEED pattern of a Pd(001) single crystal (right) and 2 MLs of Ni grown on Pd(001) (left).

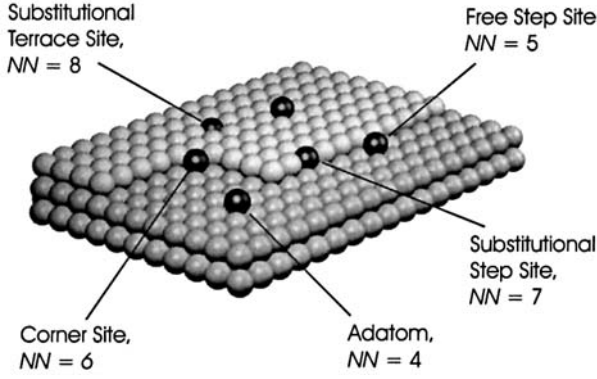
Ni/Pd system is extremely large, at 10.4%. Between 6 and 12 MLs, a  $(10 \times 10)$  LEED pattern was observed, demonstrating the formation of an over-structure. Finally, at thicknesses of more than 12 MLs, the LEED pattern reverts to a simple  $(1 \times 1)$  structure with lattice parameters similar to those of bulk Ni. It can be expected that the large tetragonal strain, especially in the first 6 MLs, would considerably change the magnetic properties of the ultrathin Ni film.

In this work, we report the first measurement of an out-of-plane magnetisation of *ultrathin* (2 MLs) Ni grown on Pd(001) using PAC spectroscopy. Furthermore, we critically re-examine some of the published claims –both experimental [6–8] and theoretical [9,10,16]– concerning the role of the coordination number and symmetry, by carefully measuring the magnetic and electric hyperfine response of Cd adatoms on the lattice-expanded 2 ML Ni film. The measurements are also compared to previous experiments with Cd on bulk Ni(001) single-crystal substrates [17] and Pd(001) single crystals [18].

## 2 Experimental procedures

The sample preparation and measurement were performed *in situ* in the UHV chamber “ASPIC” (Apparatus for Surface Physics and Interfaces at CERN) described in reference [19] (base pressure  $1.5 \times 10^{-10}$  mbar). A Ni film was grown on a Pd(001) single crystal previously cleaned by repeated cycles of sputtering and annealing to 980 K. The surface purity and structure of the substrate were controlled by Auger and LEED spectroscopy. A final  $(1 \times 1)$  LEED pattern of the Pd(001) surface as prepared is shown in Figure 1. After 2 MLs of Ni were MBE-evaporated onto the Pd surface at room temperature, a similar  $(1 \times 1)$  LEED pattern was observed, confirming that the Ni over-layer is pseudomorphic with the substrate – and thus subjected to a large lattice expansion of 10.4%, as was previously experimentally demonstrated in reference [14]. Thereafter, the sample was cooled to 40 K and a small amount of radioactive  $^{111m}\text{Cd}$  probe atoms were soft-landed onto its surface.

From previous studies [6,17], no diffusion of the Cd atoms can be expected at this low temperature of 40 K. Furthermore, at this temperature we expect the radioactive  $^{111m}\text{Cd}$  probes to occupy predominantly only a single site on a (001) surface, i.e., they will be adatoms with 4 nearest neighbours; cf. Figure 2. Additionally, Figure 2



**Fig. 2.** Idealised surface of a Ni single crystal with (001) orientation. All possible regular lattice sites for impurity atoms as local probes are indicated. The probe atoms can be differentiated by the number of their Ni nearest neighbours, the coordination number  $NN$ ; e.g., the adatom has the coordination number  $NN = 4$ .

shows all the possible sites that might be occupied at higher temperatures. A similar diagram can be produced for a (111) surface, with different coordination numbers for similar sites. At these regular sites, the probe nuclei experience combined hyperfine interactions due to electric-field gradients (EFG) and magnetic hyperfine fields ( $B_{HF}$ ), resulting in discrete PAC frequencies. From the EFG, which is a measure of the electron-density distribution around the nucleus, we acquire a characteristic “fingerprint” for the exact position of the probe atoms, since probes at interfaces, as adatoms, in terraces, etc., exhibit different and unique EFG signatures.

The EFG is characterised by the following principal parameters:  $V_{zz}$  is the largest component of the diagonalised EFG tensor, the asymmetry parameter  $\eta$  is defined by  $\eta = |V_{xx} - V_{yy}|/V_{zz}$ , and the angle  $\beta$  gives the deviation of the EFG symmetry axis from the surface normal. For adatoms on the (001) surface of a Ni bulk crystal,  $\eta = 0 = \beta$  is required by symmetry. The frequency of the interaction between the EFG and the quadrupole moment  $Q_N$  of the probe nucleus is described by the quadrupole coupling constant

$$\nu_Q = eQ_N|V_{zz}|/\hbar. \quad (3)$$

Because of the spin polarisation of the Ni atoms, the  $s$ -electrons of the Cd probe atoms are also polarised, resulting in a magnetic hyperfine field ( $B_{HF}$ ) at the Cd nuclei.  $B_{HF}$  is obtained from the measured magnetic Larmor frequency:

$$\omega_L = -g_N\mu_N|B_{HF}|/\hbar. \quad (4)$$

The nuclear properties are taken from reference [20]. Both interaction frequencies are measured simultaneously in terms of the combined interaction frequency  $\omega_c = \omega_c(\omega_L, \omega_Q, \eta, \beta)$ , deduced from the ratio function  $R(t)$  measured by a 4-detector array:

$$R(t) = \frac{2[C(180^\circ) - C(90^\circ)]}{[C(180^\circ) + C(90^\circ)]}, \quad (5)$$

where  $C(\Omega)$  ( $\Omega$  is the angle between the detectors) are the counting rates in the respective detectors.

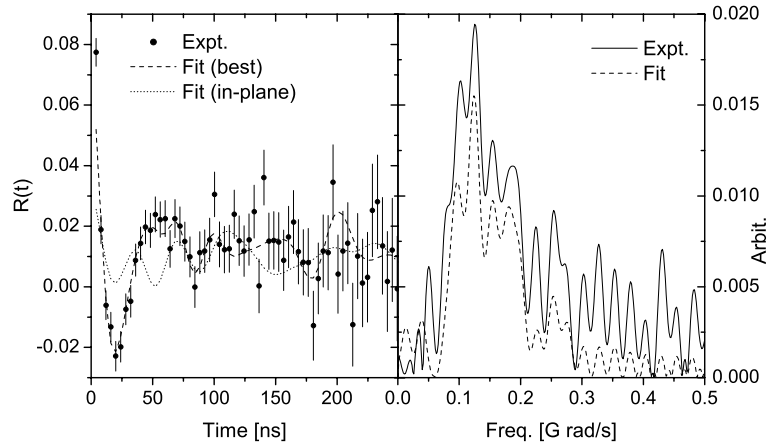
In Figure 3, the PAC time spectrum  $R(t)$  together with its corresponding Fourier transform are shown. On a real surface, a large fraction of the probes occupies surface sites related to various defects, including contamination. In particular, where the 2 MLs of Ni are strongly expanded, the surface contains many defects. These fractions experience a broad distribution of EFGs and related  $B_{HF}$ s which causes a reduction (in Fig. 3: 70%) of the nuclear  $\gamma$ -ray anisotropy within the first 2–4 ns of the measurement. The essential part of the time spectrum represents discrete frequencies from probe atoms at well-defined sites; i.e., PAC spectroscopy, observing on a local, atomic scale, filters out those probes which are in disturbed positions. This part of the spectrum is fitted by the general expression describing PAC for static electric and magnetic interactions on single crystals [21]. The relevant parameters ( $V_{zz}$ ,  $B_{HF}$  and the deduced canting angle  $\theta$  between  $B_{HF}$  and the surface normal) obtained from the fit of eq. (5) are compiled in Table 1 together with a comparison with Cd parameters measured previously on Ni(001) [17] and Pd(001) [18] single crystals.

The spectrum was fitted by a single fraction (dashed line) with an amplitude of 30%, see Figure 3. A second fit to the spectrum was performed in order to show the sensitivity of the PAC time distribution to changes in  $\theta$  (dotted line). In this second fit,  $\theta$  was constrained to  $90^\circ$ , allowing all other parameters to vary. Constraining  $\theta$  not only changes the overall shape of the time distribution, but also changes the frequencies. This confirms the sensitivity of the data and our analysis to the canting angle  $\theta$  (see below). The significant values derived from only the best fit are given in Table 1.

The value of  $V_{zz}$  obtained is considerably larger than for a Cd adatom on a pure Ni(001) surface; however, it lies surprisingly close –within the error bars– to the value of  $V_{zz}$  from previously-measured adatom sites on pure Pd(001) (see Tab. 1). Additionally, from symmetry, the other EFG parameters,  $\eta = 0.00(5)$  and  $\beta = 5(5)$ , were found to be zero within error bars; they are not given in the table. This is clear evidence that we are observing a Cd adatom on the (001) surface. The magnetic hyperfine field of this adatom is found to be ca. 30% smaller than the hyperfine field of a Cd adatom on a pure Ni(001) surface; cf. the first and second rows of Table 1. Another more important difference relative to the measurements on a pure Ni(001) single crystal surface is that the direction of the magnetic hyperfine field is canted at an angle of  $\theta = 50(5)^\circ$  to the surface normal; thus the magnetic anisotropy of 2 MLs of Ni(001) is modified as compared to that at the surface of a bulk Ni crystal. This angle was measured in zero applied field.

### 3 Discussion

Table 1 compares the Cd adatom hyperfine field parameters measured on 2 MLs of Ni grown on Pd(001) with previous measurements of Cd adatoms on pure Ni(001) [17]



**Fig. 3.** The PAC time spectrum  $R(t)$  (left) and its Fourier transform (right) are shown together with the corresponding best fit (dashed line) and the Fourier transform of the fit (dashed line, right panel) for  $^{111}\text{Cd}$  probes on 2 MLs of Ni grown on a Pd(001) single crystal, measured at 40 K. In addition, a fit keeping  $\theta = 90^\circ$  and allowing all other parameters to vary was performed (dotted line, left panel), in order to demonstrate the sensitivity of the PAC time spectrum to changes in  $\theta$ .

**Table 1.** The experimental results for the Cd adatom measurements on Ni(2ML)/Pd(001) (this work), on Ni(001) [17] and on Pd(001) [18] surfaces for the hyperfine field parameters  $V_{zz}$ ,  $B_{HF}$  and  $\theta$ , where  $\theta$  is the angle between the surface normal and  $B_{HF}$ . In addition, results of first-principles calculations are shown for the parameters  $V_{zz}$  [16] and  $B_{HF}$  [10] in the relaxed and unrelaxed lattice configurations, obtained using the WIEN2k code. The unrelaxed calculation used the ideal fcc Ni crystallographic site for the Cd adatom. In the relaxed calculation, the Cd atom was shifted outwards  $0.2 \text{ \AA}$  away from the ideal surface site.

Adatom Position	$V_{zz}$ <sup>1</sup>			$B_{HF}$ <sup>2</sup>			$\theta$
	Unrelax.	Relax.	Expt.	Unrelax.	Relax.	Expt.	
Ni(2ML)/Pd(001)			2.4(3)			5.2(5)	50(5) <sup>o</sup>
Ni(001)	4.0	1.9	0.27(4)	2.6	22.9	7.3(2)	80(5) <sup>o</sup>
Pd(001)	2.8	3.4	2.8(5)				

<sup>1</sup>Units: [ $\times 10^{21} \text{ V/m}^2$ ], with  $|Q_N(^{111}\text{Cd})| = 0.83 \text{ b}$  [20].

<sup>2</sup>Units: [Tesla], with  $g_N = -0.306$  [20].

and pure Pd(001) [18] surfaces, together with ab initio calculations [10,16]. We notice that the value of  $V_{zz}$  for the Cd adatom on 2 MLs of Ni grown on Pd(001) is approximately 9 times greater than for an adatom on a bulk Ni(001) surface, whereas the values of the magnetic hyperfine fields are similar (only ca. 30% smaller on Ni(2ML)/Pd(001), see Tab. 1). The different responses of the magnetic and electric hyperfine fields reflect the fact that the EFG tensor at the Cd nuclear site depends sensitively on the relative population distributions of its  $5p$ -electrons ( $5p_x$ ,  $5p_y$  and  $5p_z$ ; see Ref. [16] and references therein), while the magnetic hyperfine field,  $B_{HF}$ , is generated essentially by the  $5s$ -electron polarisation on the Cd ion [9,10].

The much larger value of  $V_{zz}$  for the Cd adatom on the surface of expanded Ni indicates that the Cd adatom is located relatively closer to the surface than on a bulk Ni(001) surface for the following reasons: experimentally, on bulk Ni(001) surfaces, the  $V_{zz}$  values of Cd probes are found to progressively increase with increasing  $NN$ ; for example, Cd probes located *on* the terrace with  $NN = 4$  (adatoms) have  $V_{zz} = 0.27(3)$  [ $10^{21} \text{ V/m}^2$ ], while for Cd probes *in* the terrace with  $NN = 8$ ,  $V_{zz} = 8.2(2)$  [ $10^{21} \text{ V/m}^2$ ] is observed. This general progression is

supported by theory [16,22]. Smaller increases in  $V_{zz}$  have also been predicted when the adatom position is changed by small amounts relative to the surface, cf. the calculated relaxed and unrelaxed  $V_{zz}$  for adatom sites on Ni(001) in Table 1. Similar results were found experimentally for Se adatoms on Ni surfaces, where small changes in  $V_{zz}$  were observed on changing the temperature. These changes could be theoretically understood only if the adatom position moved away from the surface with increasing temperature [23]. Finally, assuming that the ionic radii remain relatively constant, it would be expected that on an expanded Ni lattice, the equilibrium Cd adatom position is located closer to the surface than on a relaxed Ni surface.

Before comparing the magnetic hyperfine field of the Cd adatom on bulk Ni(001) with adatoms on an expanded Ni(001) layer, we must establish whether both systems were measured at magnetic saturation. The Curie temperature ( $T_C$ ) of bulk Ni is  $\sim 630 \text{ K}$ , thus the measured hyperfine field on bulk Ni surfaces at 77 K is comparable to the case of magnetic saturation at 0 K [17]. Thin Ni films have been shown to have a thickness-dependent  $T_C$  [24]. The Curie temperature of a 2 ML Ni film on Cu(001) is estimated to be  $\sim 60 \text{ K}$ . However, both a 2.5 ML Ni film on Re(0001) [25], and a 2 ML Ni film on W(110) [24] were

found to have a  $T_C$  of 320 K. Both W(110) and Re(0001) have similar surface lattice parameters to Pd(001). Furthermore, Ni films of  $\sim 1$  ML were found to be ferromagnetic, when grown on thick ( $\geq 10$  ML) Pd(111) films on a Re(0001) substrate. This is to be expected, since Pd is a strongly exchange-enhanced metal. Therefore we could expect that at our measurement temperature of 40 K, the 2 ML Ni film is close to magnetic saturation and the measured hyperfine field for  $NN = 4$  at 40 K can be compared to the magnetic saturation value at 0 K.

In previous experiments, it was found that the magnetic hyperfine field of Cd on Ni surfaces is (i) dominated by the number of nearest neighbours,  $NN$  [6], and (ii) is practically independent of the local site symmetry [7]. Therefore, it is not surprising that in spite of the large changes in the lattice parameter, the Cd magnetic hyperfine fields on 2 MLs of Ni on Pd(001) and on bulk Ni(001) surfaces are similar, because in both cases  $NN$  and the local site symmetry are the same. Unfortunately, there are no theoretical predications for the Cd hyperfine fields on expanded Ni surfaces; however, Cd hyperfine fields for adatoms have been calculated for the relaxed and unrelaxed lattice sites on bulk Ni(001) (see Tab. 1). These results appear to be highly sensitive to the Cd ion position; a shift of only 0.2 Å away from the ideal fcc Ni position corresponds to a change in the calculated hyperfine field from 2.6 T in the unrelaxed to 22.9 T in the relaxed lattice. This large sensitivity of the magnetic hyperfine field to the Ni-Cd distance appears to be due to a peak in the density of states for Cd atoms on the surface [10]. However, if the general trends are to be believed, the small drop (ca. 30%) in the value of the Cd magnetic hyperfine field on an expanded Ni surface compared to a bulk Ni(001) surface could well be caused by a relative shift towards the surface compared to Cd on bulk Ni(001).

The measured canting angle of the magnetic hyperfine field of the Cd adatom away from the surface normal on ultrathin Ni grown on Pd(001) is  $50(5)^\circ$ , see Table 1. This is to be compared to the quoted value of  $80(5)^\circ$  for the Cd adatom on a Ni(001) single-crystal surface [17]; the latter value is only two standard deviations away from being perfectly in-plane. Generally, it is not expected that the surface magnetisation of a Ni(001) surface of a single crystal would emerge from the plane, as confirmed by Cd probes at other surface sites [5]. Thus there is a significant change in the magnetic anisotropy for ultrathin (lattice expanded) Ni films as compared to the surfaces of bulk Ni crystals.

As described in the Introduction, ultrathin Ni films, below a critical thickness, are dominated by in-plane magnetic anisotropy; for the case of Ni films grown on Cu(001), with 2.5% lattice expansion, this critical thickness is  $d^* = 7$  ML [3]. In the case of Ni grown on  $\text{Cu}_3\text{Au}(001)$ , the original lattice strain of 6.3% is reduced to 4% above 5 ML. The critical thickness in this case was found to be around 5.5 ML. Since the surface anisotropy scales as  $1/d$ , 2 MLs of Ni could also be expected to be dominated by in-plane surface anisotropy; however, Ni with a lattice expansion of 10.4% on Pd would have a greatly

enhanced magnetoelastic term, giving rise to an out-of-plane magnetisation. An estimate for the corresponding energy in equation (2) is  $+1470$  kJ/m<sup>2</sup>, which is about 4 times larger than measured for Ni films grown on Cu [3]. To obtain this estimate, we have assumed a simple linear elastic model [15], where the out-of-plane elastic strain,  $\epsilon_2$ , is given by  $\epsilon_2 = -2c_{12}\epsilon_1/c_{11} = -13.3\%$ , with  $\epsilon_1 = 10.4\%$  being the in-plane strain due to the lattice mismatch, and the values for  $\lambda_{001}$  and  $c_{11}$  and  $c_{12}$  have been taken from reference [13]. As a result, this anisotropy is dominant, decreasing the critical thickness as compared to Ni grown on Cu(001) or  $\text{Cu}_3\text{Au}(001)$ .

The fact that the adatom exhibits a canting angle (i.e., neither  $0^\circ$  nor  $90^\circ$ , corresponding to perpendicular or parallel magnetisation) suggests a contribution from a fourth-order term (usually of the form  $K_{4\perp} \cos^4 \theta$ ), because equation (1) allows only two stable energy states ( $\theta = 0^\circ$  or  $90^\circ$ ) [24]. Perpendicular fourth-order anisotropy terms are generally small and are important only near the critical thickness  $d^*$ , where the magnetisation crosses over from in-plane to out-of-plane. In the case of Ni on Cu(001), this cross-over region occurs over a small range of Ni thicknesses  $d$  in the interval 6.8–7.0 ML. Also in this regime, the second order  $K_{eff}$  term is small, thus indicating that the critical thickness for Ni grown on Pd(001) must be near 2 MLs. Additionally, by modifying equation (1) to include a fourth-order term and by minimising the energy with respect to  $\theta$ , the ratio  $K_{4\perp}/K_{eff} = 0.826$  can be shown to yield our measured canting angle of  $50^\circ$  for 2 ML of Ni.

There are a number of theoretical calculations of the strain-induced magnetocrystalline anisotropy for the Ni/Cu(001) and Cu/Ni/Cu(001) systems [26–28]. The predicted critical thickness of 10–11 ML was calculated for the Ni/Cu(001) bilayer using the *full potential linearised augmented plane wave* (FLAWP) method [26]. However, using a *tight-binding* calculation, no magnetisation re-orientation was predicted [28]. In the case of bulk Ni, estimates of the magneto-crystalline anisotropy (MCA) are usually quite poor, even predicting the wrong easy axis [2,26]. Because of the high symmetry in cubic systems, the MCA energies are very small (1  $\mu\text{eV}/\text{atom}$ ) and are at the limit of precision for total energy calculations. However, estimates for the magnetostriction constant ( $\lambda_{001}$ ) are usually in good agreement with theory [26,27]. In the case of extreme tetragonal distortion, for example Ni grown on Pd(001), whether equation (2) remains linear in the strain with the same constants has not been yet been tested. However, if we assume that the constants in equation (2) remain valid and use the same surface anisotropy as for the Ni/Cu system, a critical thickness of about 1 ML can be estimated for the case of Ni/Pd(001).

Clearly, at such low Ni coverages these ideal models (e.g. Eq. (1)) become invalid, because surface roughness, diffusion, and fluctuations in film thickness become critical factors in determining the magnetic behaviour. Using PAC, we have achieved atomic resolution within the plane, distinguishing between probe atoms on flat surfaces and steps; however, our results represent an average over Ni

thickness fluctuations. We hope that this work inspires detailed theoretical calculations on systems with extreme lattice distortions.

## 4 Conclusions

The magnetic properties of highly-strained Ni films on Pd(001) have been observed using  $^{111m}\text{Cd}$  adatoms on their surface investigated by PAC. The coherent pseudomorphic epitaxial growth of 2 MLs of Ni on Pd(001) was confirmed using LEED. The determination of the Cd hyperfine field was used to test a number of previous experimental and theoretical assumptions about the induced hyperfine field of Cd, here on a greatly expanded Ni lattice. From previous experiments, the dominant factors contributing to the magnetic hyperfine field were found to be the coordination number [6] and to a much lesser extent the local surface symmetry [7]. Our results have shown that the magnetic hyperfine field of a Cd adatom on an expanded Ni ultrathin film is similar to that for adatoms on bulk Ni (only ca. 30% smaller); in both cases the Cd adatom has the same coordination number and the same local site symmetry. The changes in  $V_{zz}$  caused by the lattice expansion are much more pronounced as compared to bulk Ni surfaces. DFT predicts the general trends of these results; however, it poorly matches the absolute values. Clearly, these results represent an exacting test for DFT.

Finally, the magnetic anisotropy of highly-strained 2 ML of Ni(001) is drastically modified compared to the Ni(001) surface of a bulk crystal. The hyperfine field of Cd adatoms was found to be canted at an angle of  $50(5)^\circ$  to the surface normal. Usually, for 2 MLs of Ni, in-plane shape and surface anisotropy dominate; however, due to the large degree of strain for ultrathin Ni films on Pd(001), the out-of-plane magnetoelastic anisotropy is predominant, even at these low Ni thicknesses.

We thank the ISOLDE staff and the HMI for support of the experiments. M.J.P. also wishes to thank the DFG for a fellowship during this work.

## References

1. A.S. Hoagland, IEEE Transactions on Magnetism **39**, 1871 (2003)
2. M.D. Stiles, S.V. Halilov, R.A. Hyman, A. Zangwill, Phys. Rev. B **64**, 104430 (2001)
3. B. Schulz, K. Baberschke, Phys. Rev. B **50**, 13467 (1994)
4. A. Braun, B. Feldmann, M. Wuttig, J. Magn. Magn. Mat. **171**, 16 (1997)
5. H. Granzer, H.H. Bertschat, H. Haas, W.-D. Zeitz, J. Lohmüller, G. Schatz, Phys. Rev. Lett. **77**, 4261 (1996)
6. K. Potzger, A. Weber, H.H. Bertschat, W.-D. Zeitz, M. Dietrich, Phys. Rev. Lett. **88**, 247201 (2002)
7. M.J. Prandolini, Y. Manzhur, A. Weber, K. Potzger, H.H. Bertschat, H. Ueno, H. Miyoshi, M. Dietrich, Appl. Phys. Lett. **85**, 76 (2004)
8. K. Potzger, A. Weber, Y. Manzhur, M.J. Prandolini, H.H. Bertschat, W.-D. Zeitz, M. Dietrich, Phys. Stat. Sol. (c) **1**, 3275 (2004)
9. P. Mavropoulos, J. Phys.: Condens. Matter **15**, 8115 (2003)
10. V. Bellini, S. Cottenier, M. Çakmak, F. Manghi, M. Rots, Phys. Rev. B **70**, 155419 (2004)
11. T. Phalet, M.J. Prandolini, W.D. Brewer, P. Schuurmans, N. Severijs, B.G. Turrell, B. Vereecke, S. Versyck, Phys. Rev. B **71**, 144431 (2005)
12. W.J.M. de Jonge, P.J.H. Bloemen, F.J.A. den Broeder, in *Ultrathin Magnetic Structures 1*, edited by J.A.C. Bland, B. Heinrich (Springer-Verlag Berlin, 1994), Chap. 2.3, p. 65
13. S. Chikazumi, S.H. Charap, *Physics of Magnetism* (John Wiley & Sons New York, 1964)
14. G.A. Rizzi, M. Petukhov, M. Sambì, G. Granozzi, Surf. Sci. **522**, 1 (2003)
15. P.M. Marcus, F. Jona, Surf. Rev. Lett. **1**, 15 (1994)
16. S. Cottenier, V. Bellini, M. Çakmak, F. Manghi, M. Rots, Phys. Rev. B **70**, 155418 (2004)
17. J. Voigt, Ph.D. thesis, Universität Konstanz, Konstanz, Germany, (1990, unpublished)
18. R. Fink, B.-U. Runge, K. Jacobs, G. Krausch, J. Lohmüller, B. Luckscheiter, U. Wöhrmann, G. Schatz, J. Phys.: Condens. Matter **5**, 3837 (1993)
19. H.H. Bertschat, K. Potzger, A. Weber, W.-D. Zeitz, Eur. Phys. J. A **13**, 233 (2002)
20. *Table of Isotopes*, 8th edn., edited by R.B. Firestone, V.S. Shirley (Wiley & Sons: New York, 1996)
21. B. Lindgren, Hyp. Int. (C) **1**, 613 (1996)
22. B. Lindgren, Z. Naturforsch. a **57**, 544 (2002)
23. A. Weber, K. Potzger, H. Granzer, H.H. Bertschat, W.-D. Zeitz, M. Dietrich, B. Lindgren, Phys. Rev. B **64**, 081404(R) (2001)
24. K. Baberschke, Appl. Phys. A **62**, 417 (1996)
25. U. Gradmann, R. Bergholz, Phys. Rev. Lett. **52**, 771 (1984)
26. R. Wu, A.J. Freeman, J. Magn. Magn. Mater. **200**, 498 (1999)
27. G.Y. Guo, J. Magn. Magn. Mater. **176**, 97 (1997)
28. A. Lessard, T.H. Moos, W. Hübner, Phys. Rev. B **56**, 2594 (1997)