Layer-Resolved Magnetic Moments in Ni/Pt Multilayers

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(Received 23 November 1999)

The magnetic moments in Ni/Pt multilayers are thoroughly studied by combining experimental and *ab initio* theoretical techniques. SQUID magnetometry probes the samples' magnetizations. X-ray magnetic circular dichroism separates the contribution of Ni and Pt and provides a layer-resolved magnetic moment profile for the whole system. The results are compared to band-structure calculations. Induced Pt magnetic moments localized mostly at the interface are revealed. No magnetically "dead" Ni layers are found. The magnetization per Ni volume is slightly enhanced compared to bulk NiPt alloys.

PACS numbers: 75.70.Cn, 75.25.+z, 75.30.Cr

Magnetic multilayers constitute a new class of materials exhibiting a rich variety of novel effects related to the artificial structure, the large number of interfaces, and the confinement of electrons in ultrathin layers [1–3]. Understanding the mechanisms governing these and related effects is crucial for designing materials with desirable properties. However, in the past the lack of experimental sensitivity in the monolayer (ML) limit was restricting our insight. It is only now that experiments on magnetic thin films and multilayers can provide complete and detailed information to be compared with calculations [4]. In parallel, *ab initio* calculations allow one nowadays to study magnetism on an atomic scale. Satisfactory agreement between theory and experiment can be achieved even for the magnetic anisotropy energy which is only a small fraction (10^{-6}) of the total energy in solids; see, e.g., [5,6].

In this work we combine powerful experimental and *ab initio* theoretical techniques to give a complete, layer-resolved, picture of the magnetic moments in multilayers. As a prototype system we select the Ni/Pt one because (i) the interfaces between Ni and Pt in evaporated Ni/Pt multilayers are sharp in the ML limit, as was shown by structural characterization via x-ray diffraction (XRD) and electron microscopy [7,8]. As evidence we show in Fig. 1 the θ-2θ XRD spectrum for a Ni_2/Pt_2 multilayer. The indices are numbers of ML in one multilayer period. The multilayer diffractions prove that the multilayer structure is present even for films with extremely thin (2 ML) Ni and Pt layers. That is, interdiffusion, if any, is strictly limited to the interface. (ii) Ni/Pt multilayers are candidates for magneto-optic (MO) recording, since they show perpendicular magnetization at room temperature and a pronounced Kerr rotation maximum at the blue wavelength [7]. Both effects are strongly related to strain and hybridization at the Ni/Pt interface. Although the properties of the Ni/Pt

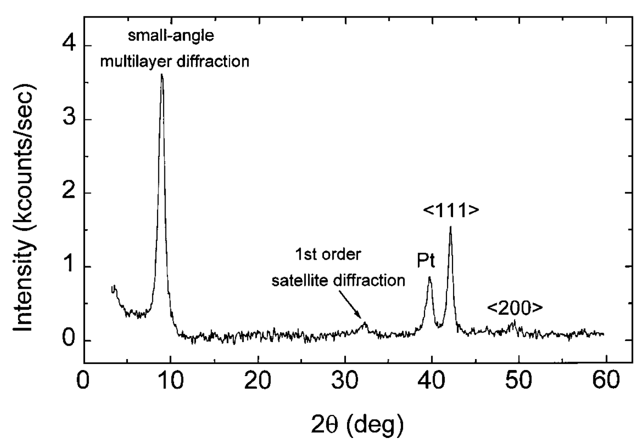

FIG. 1. θ-2θ XRD spectrum for a Ni_2/Pt_2 multilayer. Despite the very thin Ni and Pt layers a small-angle multilayer and a high-angle first-order satellite diffraction are shown indicative of sharp interfaces. The Pt-buffer-layer diffraction and the solid solution diffraction from $\langle 111 \rangle$ and $\langle 200 \rangle$ are indicated.

interface are crucial for applications, the role of Pt at this interface is not completely understood. Studies using magnetometry techniques report either a slightly enhanced [9] or strongly reduced magnetization [10] for Ni/Pt multilayers. They attribute these effects to possible Pt polarization [9] or, based on an analysis of the room temperature magnetization with the inverse Ni thickness, to “dead” Ni layers at the Ni/Pt interface [10]. While puzzling results have been reported for multilayers, diffuse neutron scattering measurements for NiPt alloys revealed that Ni has a smaller magnetic moment in these alloys than the pure Ni, while Pt shows induced moments [11]. No increased magnetization per Ni volume was deduced [11]. Our work shows that Pt in Ni/Pt multilayers acquires a relatively large moment up to 0.29 μ_B/atom, like the induced Pt moment in Co/Pt based systems [12]. By comparing the results of a non-element-specific magnetometry technique to element-specific ones, we demonstrate that the commonly used inverse-thickness analysis of the magnetization is not proper for multilayers with strongly polarizable paramagnetic constituents. We show that, contrary to alloys, it is possible to find enhanced magnetization per Ni volume by a suitable choice of the individual Ni and Pt thicknesses. We unambiguously exclude the existence of magnetically dead Ni layers. A magnetic-moment profile coming from the element-specific data is provided for the first time, to our knowledge, for *both* constituents of a multilayer and it is compared to theory. The profile shows that hybridization effects are located mainly at the interface.

Ni_n/Pt_m multilayers with n and m ranged between 2 and 13 ML were prepared and characterized with respect to their structural and MO properties [7]. SQUID magnetometry measurements were carried out at a temperature of 10 K with a maximum applied field of 2 T. The x-ray magnetic circular dichroism (XMCD) experiments were performed at the European Synchrotron Radiation Facility (ESRF) in Grenoble (France) on the ID12A and ID12B beam lines [13] at the Ni and Pt $L_{2,3}$ edges using the total electron and fluorescence yield detection modes, respectively. The degree of polarization of the circular light was 85%. Large magnetic fields (2–5 T) were applied along the x-ray beam direction for magnetic saturation of the samples at 10 K. The determination of Ni and Pt magnetic moments was done by application of the sum rules [14]. A 20 nm thick Ni sample was used as reference. The Pt moments were determined via the same process as the one in Refs. [12,15].

The electronic structure of the multilayers was calculated self-consistently by means of the tight-binding linearized muffin-tin orbital (TB-LMTO) method in the combined correction atomic-sphere approximation mode [16]. Exchange and correlation effects were treated within the framework of local density functional theory, using the parametrization of von Barth and Hedin [17]. Relativistic effects were included through the well-known scalar-relativistic approximation, i.e., the effects of the spin-orbit coupling were not taken into account. For the calculations an fcc(111) layered structure was assumed. The lattice parameters used in the unit cell and the interlayer distances were estimated using elastic theory and experimental data for the pure elements.

In Fig. 2 we show the total magnetization of Ni_n/Pt_m multilayers normalized with respect to the Ni volume as a function of n (top) or $1/n$ (bottom). An excellent agreement is shown between SQUID (squares) and XMCD (circles) measurements. Two groups of samples may be seen: The first, group A, for $4 < n < 12$ ML shows no dependence of the total magnetization on n. If we would not have the possibility to separate the Ni and Pt moments we could ascribe this finding to a purely bulklike behavior. The second, group B, for $n < 4$ ML shows a decreasing magnetization with m. Interestingly, samples with very thin Pt layers ($m = 2$ ML) show a small increase in the total magnetization which was not found in NiPt alloys. Contrary to [10], no evidence for a strongly reduced magnetization is found.

By means of the element-specific XMCD measurements we may separate the contributions of Ni and Pt to the total magnetization of the samples. The results for five multilayers of Fig. 2 are summarized in Table I. For thicker Ni layers ($n > 2$) the average Ni magnetic moment approaches the bulk value [18]. No dead Ni layers are found. Note that if about 2 ML of Ni at each interface were nonmagnetic [10] our Ni_2/Pt_m samples would not be magnetic at all. All samples, even those with thinner Ni and thicker Pt layers show induced magnetic moments at the Pt sites. This is demonstrated by the XMCD spectra at the $L_{2,3}$ Pt-edges for three multilayers (Fig. 3). The larger induced moment of the $0.29\mu_B$/atom is, within the error bars, equal to the highest one reported for Ni-rich NiPt alloys [11]. Figures 2 and 3 and Table I show that the

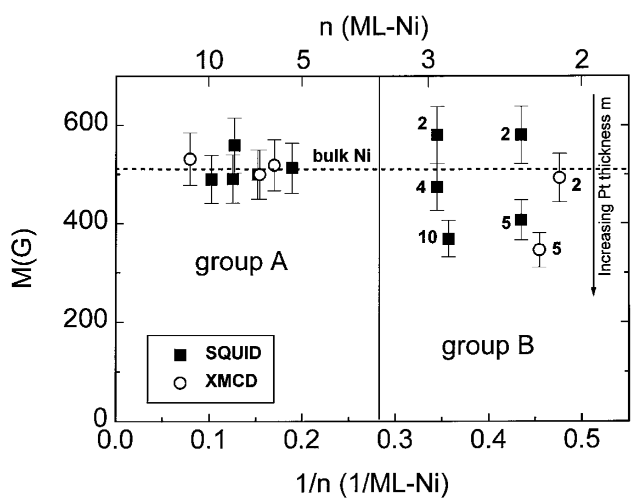

FIG. 2. Total magnetization of Ni/Pt multilayers (normalized to the Ni volume) with n (top) or $1/n$ (bottom). Two data sets from SQUID (squares) and XMCD (circles) are shown. The dashed line stands for the magnetization of bulk Ni [18]. The films of group B show a decrease of the magnetization with increasing Pt thickness m (arrow). The m values for each sample are also indicated. $T = 10$ K.

TABLE I. XMCD data for the average total magnetic moment of Ni and Pt. Induced Pt moments are found for all the samples. The induced moments are larger for thin Pt and thick Ni layers. The Ni magnetic moment is always reduced compared to the bulk (see the last row). The effect is stronger for thin Ni separated by thick Pt layers. The error bars are typically of about 10%.

n (ML)	m (ML)	μ_{Ni} (μ_B/atom)	μ_{Pt} (μ_B/atom)
2	5	0.24	0.09
2	2	0.39	0.17
6	5	0.47	0.17
6	2	0.49	0.29
13	5	0.54	0.21
Bulk Ni		0.61 [18]	

common $1/n$ analysis of the total magnetization coming from non-element-specific measurements is not appropriate for Ni/Pt multilayers: Groups A and B have noncomparable $1/n$ magnetization dependence and Pt has a sizable induced magnetization.

The XMCD data allow us to construct a layer-dependent magnetic-moment profile for a Ni_6/Pt_5 multilayer, shown in Fig. 4(a). Details of construction are given in Ref. [15]. For this profile two series of Ni_n/Pt_m samples, one with constant $m = 5$ and $n = 2, 4, 6$ and another with constant $n = 6$ and $m = 2, 5$ were used. Contrary to alloys where the atomic distances are strongly dependent on their composition, for thick multilayers with large lattice misfit (~10%) between their constituents, like ours, it has been shown that the constituents retain almost their bulk lattice parameters and the residual strain is very small (0%–2%); see, e.g., [8,19]. Thus, one may not expect sizable changes of the volume of Pt and Ni in our samples due to their composition which could have affected the spin magnetic moment [20] and, consequently, the determination of the magnetic profile by using various samples [21]. While similar profiles have been published for the nonmagnetic polarizable constituent [15,22], this is, to our knowledge, the first time that such a profile for both constituents of the multilayer is constructed. Figure 4(a) reveals that Pt acquires a large moment $\approx 0.29\mu_B$/atom at the interface, while the Ni moment at the interface is strongly reduced. These effects may be attributed to hybridization at the Ni/Pt interface. They are much less pronounced in the second from the interface Ni and Pt layers indicating sharp interfaces in agreement with the structural studies [7–9].

Here we turn to the theoretical attitude: In Fig. 4(b) we show the theoretically predicted profile of the spin magnetic moments for the Ni_6/Pt_5 sample. This shows only a small (≈8%) reduction of the interface Ni moment compared to the bulk. Note that the experimental profile of Fig. 4(a) shows that this reduction should be of about 50%. The predicted interface Pt magnetic moment of about $0.11\mu_B$/atom is of about 2–3 times smaller than the experimental one. Nevertheless, the trend predicted by the calculations is in full accordance with that deduced from the XMCD data.

In Table II we present theoretical results for a series of multilayers with $n = 2$ ML and variable m. We find increased Ni moment for very small m values. This

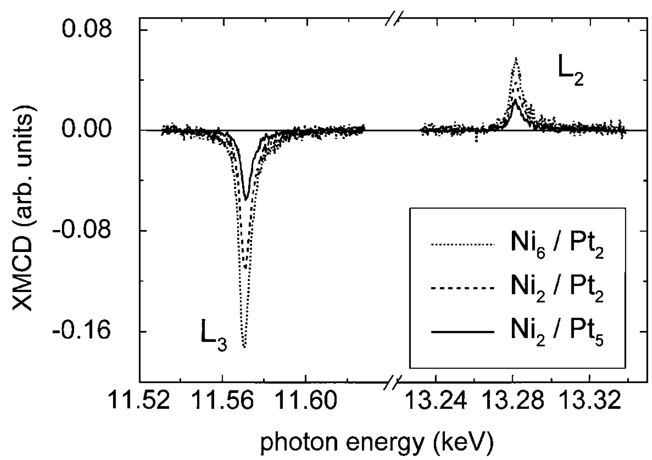

FIG. 3. XMCD spectra at the $L_{2,3}$ edges of Pt for three Ni/Pt multilayers as indicated. The Pt polarization is stronger for thicker Ni and thinner Pt layers and it decreases with increasing m.

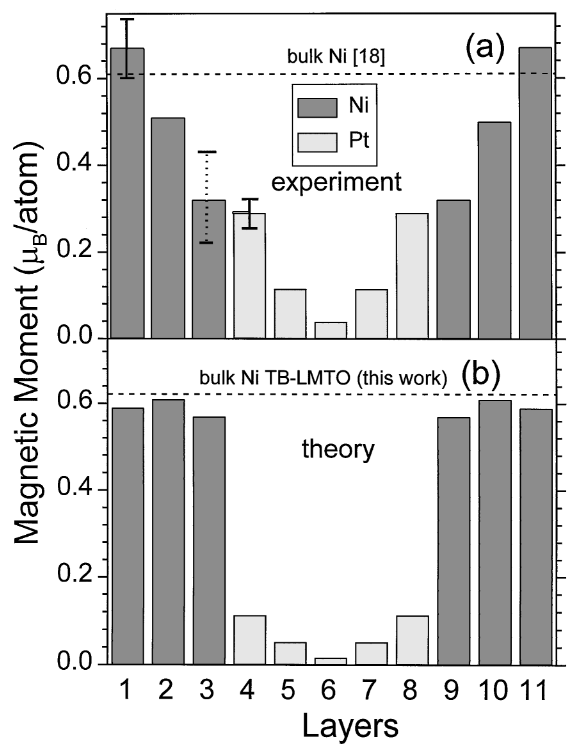

FIG. 4. (a) Experimental magnetic moment profile for a Ni_6/Pt_5 multilayer. The magnetization per Ni volume is equal to the bulk (Fig. 2). Only via an element-specific method like the XMCD layer-resolved information may be obtained. The error bars indicated with solid lines (±10%) are typical for all the moments except the one of Ni at the interface (dotted line) [21]. For the separation of the contribution of the second- to the third-from-the-interface Pt layer we used the ratio of the Pt moments by the TB-LMTO. (b) Theoretical result for the spin magnetic moments. The trends between experiment and theory are the same, but the calculated effects at the interface are smaller than the experimental ones.

TABLE II. Theoretical results for the Ni and Pt spin magnetic moment of three multilayers with $n = 2$ and $m = 1, 4, 6$ and for bulk Ni. Ni magnetic moments larger than the bulk one are predicted for very thin Pt layers. The Ni moment decreases with m justifying the data of Fig. 2 (group B).

m (ML)	μ_{Ni} (μ_B/atom)	μ_{Pt} (μ_B/atom) At interface	μ_{Pt} (μ_B/atom) At interface-1	μ_{Pt} (μ_B/atom) At interface-2	μ_{Pt} (μ_B/atom) Average
1	0.66	0.16	⋯	⋯	0.16
4	0.51	0.12	0.05	⋯	0.09
6	0.42	0.10	0.02	0.02	0.05
Bulk Ni:	0.62				

decreases below the bulk-Ni value with m. The calculated Ni moments are larger than the experimental ones of Table I. On the other hand, the average Pt moments are in very good agreement with the ones for the $n = 2$ series of Table I. Note that the calculated moments should result in an enhanced magnetization per Ni volume for very thin Ni and Pt layers. This effect was not observed for bulk NiPt alloys [11]. However, in contrast to the alloys, multilayers with $n = 2$ have all Ni atoms at the interface where a narrowing of the d-band width and enhanced magnetic moments are expected [23]. In addition, when m is very small (1–2 ML) interlayer exchange coupling and proximity effects become stronger, as shown recently for Co/Cu/Ni trilayers [24].

Theory and experiment show a similar variation of the magnetic moments as a function of m and n while giving different absolute values. However, note that (a) slightly different lattice constants compared to the real ones may have been used in the calculations. These might have resulted in small changes in the volume (~1%–5%) and affected the magnetic moments (up to about 10% for fcc Ni [20]). (b) In real samples interface exchange processes between Pt and Ni could occur. These would form some alloyed regions at the interface. Analysis of our data with respect to the ones recorded for alloys [11] shows that the Ni moment is reduced by alloying, while the Pt moment is almost unaffected [25].

In conclusion, we present a complete, layer-resolved, profile of the magnetic moments in Ni/Pt multilayers. We show that only a combination of techniques: SQUID magnetometry for the total magnetization, x-ray magnetic circular dichroism for separation of Ni and Pt contributions, and *ab initio* theoretical calculations may give a meaningful picture of the magnetic moments in magnetic multilayers. An $(1/n)$ analysis of the magnetization is not suitable for multilayers with strongly polarizable paramagnetic constituents. Experiment and theory show the same trends for the magnetic moments: Hybridization effects between Ni and Pt are mostly localized at the interface. They result in a reduction of Ni moment (stronger for thick Pt layers) and spin polarization of Pt (stronger for thicker Ni layers). Enhanced magnetizations, not observed for bulk alloys, are reported.

This work was supported by BMBF 05 SC8 KEA3, BMBF 05 SC8 WMA7, DFG Sfb290, and the ESRF.

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