Influence of interface mixing on the magnetic properties of Ni/Pt multilayers

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Motivated by existing experimental data, we study here the influence of interface mixing on the magnetic behavior of $Ni_6/Pt_5(111)$ multilayers. In the present *ab initio* calculations the mixing, restricted to the interface layers, was simulated by ordered two-dimensional Ni-Pt lattices. Two different degrees of mixing of the components at the interface were considered—namely, 25% and 50%. The perfect interface was also calculated and for some of the systems orbital moments were obtained. We find that interface mixing explains rather well the observed magnetic moment profile for Ni sites. But even with the inclusion of orbital contributions, the theoretical results tend to underestimate the induced moment at the Pt sites found experimentally.

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Magnetic multilayers have attracted attention in recent years due to their interesting physical behavior and potential applications. With the techniques now available, multilayers with a few atomic layers of each component can be produced and their magnetic properties studied by several methods.[1–3] In good quality multilayers the interfaces can be very well defined, but they are rarely ideal: some intermixing of the components, in the form of roughness or alloying, is usually present. Interface mixing can affect the behavior of the systems. In Fe/Cr(001) multilayers, for example, the moments of Cr sites close to the interface are enhanced for ideal interfaces, but drastically reduced in the presence of mixing.[4,5]

Ni-Pt multilayers have been studied using several types of magnetometry techniques, yielding conflicting results which ranged from slightly enhanced to reduced Ni moments in the multilayer. Enhanced moments were attributed to induced magnetism on the Pt atoms,[2] while the reduced ones were explained by the presence of dead Ni layers.[3] Recently, the x-ray magnetic circular dichroism (XMCD) technique was applied to study the magnetic behavior of Ni and Pt sites in $Ni_m/Pt_n(111)$ multilayers[1,6,7] and the presence of magnetically dead Ni layers was unambiguously ruled out. In one of these papers,[1] a complete set of theoretical and experimental values for local moments is presented for $Ni_6/Pt_5(111)$ multilayers. The theoretical values were obtained from *ab initio* calculations, assuming ideal interfaces and neglecting the orbital contributions to the local moment. The calculated magnetic moment profile and that inferred from experiment are shown there and discussed extensively. In spite of the reasonable qualitative agreement between theory and experiment there are some noticeable differences: (i) experiment shows a steady increase of the Ni moments as one goes from the Ni sites at the Ni-Pt interface to central ones in the Ni layers, a behavior which is not reproduced by the calculations, (ii) the calculated moment for Ni sites at the Ni-Pt interface is roughly twice as large as that inferred from experiment, and (iii) the calculated moments at Pt sites are much smaller than those obtained in the experiments. In the present work, we try to understand the origin of these differences using the *ab initio* real-space linear muffin-tin orbital method within the atomic sphere approximation (RS-LMTO-ASA) to study the effect of interface mixing on the magnetic behavior of $Ni_6/Pt_5(111)$ multilayers. The role of orbital moments and their possible relevance to the magnetic moment profiles is also discussed.

The RS-LMTO-ASA is an order-N method (the computational cost grows linearly with the number of inequivalent sites) and is therefore suitable to treat complex metallic structures with a large number of atoms in the unit cell. Being implemented in real space, it can also be used to treat systems which lack translational symmetry. The RS-LMTO-ASA has been employed with success to study crystalline systems, multilayers,[5] substitutional and interstitial impurities in metallic hosts, and surfaces, as well as impurities and defects in metals and metallic surfaces.[8,9] It follows the steps of the well-known LMTO-ASA formalism, but uses the recursion method[10] to solve the eigenvalue problem directly in real space. It is a linear method and the solutions are accurate near a given energy E_ν, usually taken at the center of gravity of the occupied bands. We work in the orthogonal representation of the LMTO-ASA formalism and expand the Hamiltonian in terms of tight-binding (TB) parameters, neglecting terms of order $(E-E_\nu)^3$ and higher. The orthogonal Hamiltonian can then be written as[11]

$$H = E_\nu + \overline{h} - \overline{hoh}, \tag{1}$$

where

$$\overline{h} = \overline{C} - E_\nu + \overline{\Delta}^{1/2}\overline{S}\overline{\Delta}^{1/2}. \tag{2}$$

Here $\overline{h}$ is a Hermitian matrix, $\overline{C}$, $\overline{\Delta}$, and $\overline{o}$ are potential parameters of the tight-binding LMTO-ASA representation, and $\overline{S}$ is the structure constant in this representation. To solve the eigenvalue problem in real space we consider a large cluster to simulate the system and use the recursion method[10] with the Beer-Pettifor terminator[12] to complete the recursion chain.

The present calculations were performed using clusters of $\approx$9000 atoms to simulate the $Ni_6/Pt_5(111)$ multilayer. In all cases a 44-atom supercell, representing the 11 atomic planes, was used. Since the lattice parameters of Ni (3.52 Å) and Pt

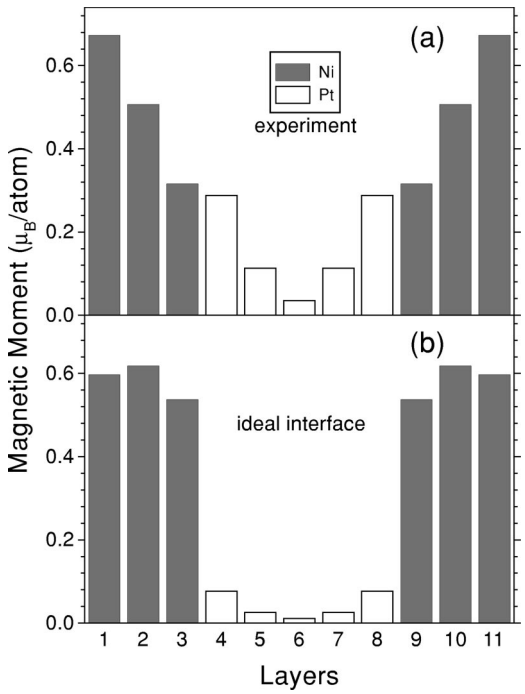

FIG. 1. The magnetic moment profile for Ni and Pt layers in $Ni_6/Pt_5(111)$ multilayers. (a) XMCD profile (Ref. 1) and (b) RS-LMTO-ASA results for the case of ideal interfaces.

(3.92 Å) are considerably different, some distortions from the regular fcc lattice are to be expected. Here we took the in-plane atomic distances at all layers to be an average between those of fcc Ni and fcc Pt. The distance between the (111) planes varied in the cluster, being that of (111) planes in Ni between Ni-Ni planes, that of (111) planes in Pt between Pt-Pt planes, and an average between these distances for the Ni-Pt interface. The mixing was simulated by exchanging Ni and Pt atoms at the interface, keeping the lattice sites fixed. We employed a basis with nine orbitals per site (s, p, and d electrons) and used 25 levels of recursion. We performed scalar relativistic (SR) calculations within the local spin density approximation, with the exchange and correlation potential of von Barth and Hedin.[13] The orbital moment was obtained by introducing the spin-orbit coupling $\mathbf{L}\cdot\mathbf{S}$ in the LMTO Hamiltonian.[14] Orbital polarization corrections[15] (OP) were also included.

In Fig. 1 we present results for the local magnetic moment profile for Ni and Pt layers in $Ni_6/Pt_5(111)$ multilayers. Figure 1(a) shows the profile inferred from XMCD experiments, which has error bars of $\pm 10\%$.[1] Our theoretical results, obtained within the SR approach for the case of ideal interfaces, are shown in Fig. 1(b). The present RS-LMTO-ASA results are in excellent agreement with the k-space TB-LMTO calculations in the literature,[1] showing the same shortcomings when compared to the experiment. Since interfaces are rarely ideal, we proceed to investigate the effect of interface mixing on the magnetic moment profile of $Ni_6/Pt_5(111)$ multilayers. Two degrees of mixing were considered: 25% and 50%. The 25% mixing was introduced by substituting the Ni (or Pt) layer at the interface by an ordered binary two-dimensional structure with three Ni (or Pt) and one Pt (or Ni) atom per unit cell. For the 50% mixing, ordered layers with equal amounts of Ni and Pt atoms were used. In the case of the 25% mixing, two inequivalent ordered structures obeying the above requirements could be constructed: a nondegenerate one yielding 12 inequivalent local moments and a threefold-degenerate one with 18 inequivalent local moments in the $Ni_6/Pt_5(111)$ multilayer. To investigate whether the results were sensitive to the particular kind of ordering, both arrangements were considered. The averaged moments for Ni and Pt atoms in each of the layers for ideal interfaces and in the presence of mixing are shown in Table I. As in Fig. 1, the layers are numbered from 1 to 6. In the presence of mixing, layer Nos. 3 and 4 have both Ni and Pt sites and both averaged moments are presented.

TABLE I. Results (in μ_B) for the averaged magnetic moment of Ni and Pt in $Ni_6/Pt_5(111)$ multilayers with ideal and mixed interfaces. For 25% mixing, moments for the threefold-degenerate (3-deg) and nondegenerate (nondeg) configurations are given. The layers are numbered as in Fig. 1.

Interface	Site	Layer number					
		1	2	3	4	5	6
Ideal	Ni	0.60	0.62	0.54	-	-	-
	Pt	-	-	-	0.077	0.026	0.011
25%	Ni	0.61	0.57	0.40	0.146	-	-
3-deg	Pt	-	-	0.17	0.078	0.024	0.010
25%	Ni	0.59	0.58	0.41	0.206	-	-
Nondeg	Pt	-	-	0.14	0.070	0.027	0.018
50%	Ni	0.62	0.55	0.27	0.135	-	-
	Pt	-	-	0.14	0.061	0.017	0.009

The local moment of Ni is rather sensitive to the number of Pt neighbors around the site. This tendency is clearly seen in Fig. 2, where the moments of all calculated Ni sites are plotted as a function of the corresponding number of Pt neighbors. In the configurations with 25% mixing, for example, the Ni site in layer No. 4 has ten Pt neighbors in the threefold-degenerate configuration and nine Pt neighbors in the nondegenerate one, both with small moments of $0.15\mu_B$ and $0.21\mu_B$, respectively. We find that the Ni impurity in Pt is nonmagnetic. As for Co in Co-Cu systems,[9] the moment of Ni in Ni-Pt systems seems to be strongly influenced by first-

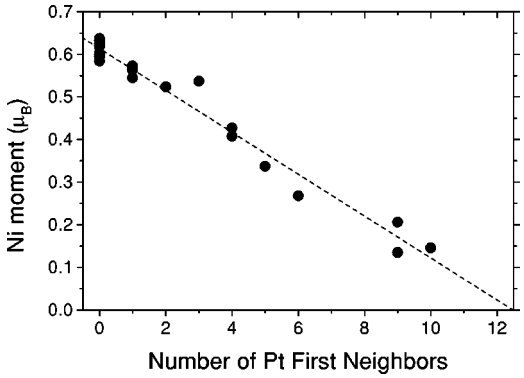

FIG. 2. Local moment of Ni (in μ_B) as a function of the number of Pt neighbors, for $Ni_6/Pt_5(111)$ multilayers.

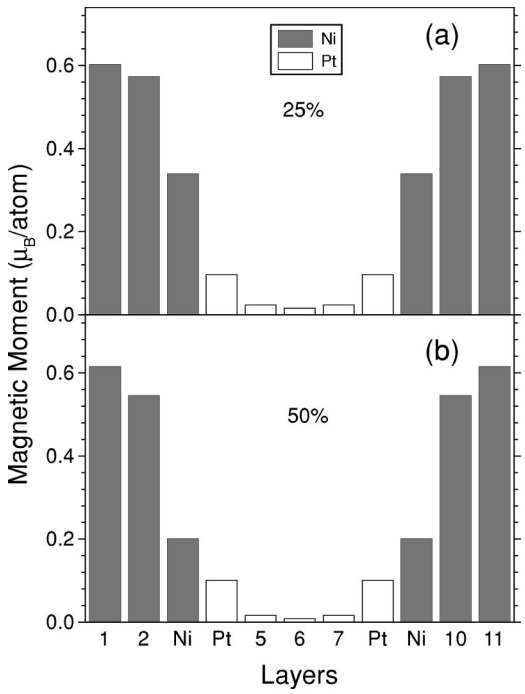

FIG. 3. The magnetic moment profile for $Ni_6/Pt_5(111)$ multilayers, in the presence of interface mixing of 25% (a) and 50% (b).

neighbor interactions, justifying a procedure which has been used to evaluate moment profiles in Ni-Pt multilayers.[1]

In Fig. 3 we show the magnetic profile for interface mixings of 25% [Fig. 3(a)] and 50% [Fig. 3(b)]. The difference between values for the threefold-degenerate and nondegenerate configurations in Table I was shown to be small, and in Fig. 3(a) a weighted average of these two values is presented. Our objective is to compare the calculated magnetic profiles with the experimental XMCD vales of Fig. 1(a), which are probe specific. Therefore our results in Fig. 3 were arranged according to probe type (Ni or Pt) and not by layer. In the third column (labeled Ni) in Fig. 3, we give the averaged moment for all the Ni sites at the interface (those located in layers 3 and 4 of Table I), while in the fourth column the averaged moment for Pt sites at the interface (again layers 3 and 4) is shown. The magnetic profiles of multilayers in the presence of interface mixing (Fig. 3) can now be compared with those obtained for ideal interfaces [Fig. 1(b)]. The Ni moment at the interface decreases significantly with mixing, being much smaller than the one calculated for ideal interfaces. The trends for Ni moments in the presence of mixing also differ significantly from that obtained for ideal interfaces, being much closer to what is observed experimentally [Fig. 1(a)]. Since XMCD experiments directly measure the averaged moments of all Ni (or all Pt) atoms in the sample, it is interesting to compare theoretical and experimental values of this quantity. The averaged Ni moments obtained for multilayers with ideal interfaces, 25% mixing, and 50% mixing were $0.59\mu_B$, $0.51\mu_B$, and $0.44\mu_B$, respectively, compared with an experimental value of around $0.50\mu_B$. Considering that the experiment has an accuracy of $\approx 10\%$, we conclude that both the averaged Ni moment and the magnetic profile of the Ni layers, for systems with an interface mixing of 25% [see Fig. 3(a)], are in excellent agreement with the XMCD experiments. On the other hand, our results indicate (see Table I) that interface mixing does not significantly change the induced moments at Pt sites. The averaged moment for Pt in $Ni_6/Pt_5(111)$ multilayers varies between $0.043\mu_B$ and $0.052\mu_B$, being in all cases about a factor of 3 smaller than the averaged induced moment of $0.168\mu_B$ obtained from XMCD measurements.[6]

Experiments indicate that the orbital contribution to the magnetic moment can be significant, with measured ratios of orbital moment (μ_L) to spin moment (μ_S) of the order of 0.10–0.11 for Ni and 0.2–0.3 for Pt.[6] To evaluate the importance of the orbital contribution, we have included the $\mathbf{L}\cdot\mathbf{S}$ term in the Hamiltonian and obtained both μ_S and μ_L for two of the systems: (i) the multilayer with ideal interfaces and (ii) the nondegenerate configuration with a mixing of 25%. When OP is included, the calculated μ_L/μ_S ratios vary between 0.08 and 0.14 for Ni sites and between 0.12 and 0.34 for Pt sites. But we find that, in spite of the large μ_L/μ_S ratios, the total moments at Pt sites are not larger than those obtained in the SR calculations. The inclusion of $\mathbf{L}\cdot\mathbf{S}$ leads to a small reduction of the spin moment of Pt in these systems, inhibiting the enhancement of the total moment at Pt sites. The results also suggest that a large number of neighbors of the opposite kind can significantly change the μ_L/μ_S ratio of Ni and Pt in Ni-Pt systems. For Ni surrounded by nine Pt neighbors, μ_L is very small, and the μ_L/μ_S ratio is close to zero. Small values of orbital moments at $3d$ sites have been also obtained for other systems, e.g., Fe impurities in Ag and Mn impurities in Au.[16] The largest μ_L/μ_S ratio in Pt (0.34) was obtained for a site surrounded by nine Ni neighbors. To verify that a large number of Ni neighbors tends to favor large μ_L/μ_S ratios at Pt sites, we have calculated both μ_L and μ_S for a Pt impurity in fcc Ni, taking as lattice constant an average between that of Ni and that of Pt. We find $\mu_L=0.138\mu_B$ and $\mu_S=0.340\mu_B$ at the impurity site, yielding a high μ_L/μ_S ratio of order 0.40 and confirming our expectations.

XMCD measurements give a large value, of the order of $0.28\mu_B$, for the induced moment of Pt at the interface in $Ni_6/Pt_5(111)$ multilayers. It was noted[6] that these moments are similar to the induced Pt moments observed for Ni-rich NiPt alloys.[17] Considering that the results fail to reproduce the observed moments for Pt in $Ni_6/Pt_5(111)$ multilayers, it is interesting to investigate whether we can, using our theoretical approach, reproduce the large induced moments which should be present at Pt sites in Ni-Pt systems with high concentrations of Ni. Therefore we have calculated the local moments in a Ni_3Pt ordered fcc structure with three Ni and one Pt atom in the cubic unit cell. This system, due to its composition, is expected to have moments comparable to those observed in Ni-rich NiPt alloys of similar Ni concentration. In Table II we show the results of our calculations of Ni_3Pt both with (Yes) and without (No) the self-consistent inclusion of orbital polarization. The total moments of $0.30\mu_B$ for Pt and $0.57\mu_B$ for Ni are comparable to the experimental values ($0.28\mu_B$ for Pt and $0.52\mu_B$ for Ni) cited in the literature[17] for Ni-rich $Ni_{80}Pt_{20}$ ferromagnetic alloys. The large induced moments and μ_L/μ_S ratios obtained for Pt in

TABLE II. Moments (in μ_B) for Ni and Pt sites in Ni_3Pt obtained with (Yes) or without (No) the inclusion of orbital polarization (OP). Spin, orbital and total moments are given.

OP	No			Yes		
	Spin	Orb	Total	Spin	Orb	Total
Ni	0.523	0.036	0.559	0.522	0.045	0.568
Pt	0.228	0.066	0.294	0.230	0.074	0.304

Ni-rich Ni-Pt alloys and in our calculations for Ni_3Pt are of the same order as those inferred from XMCD measurements for Pt sites at the interface layer in $Ni_6/Pt_5(111)$ multilayers. But one should note that the local environment around the Pt site in Ni_3Pt is rather different from that assumed for Pt in our $Ni_6/Pt_5(111)$ supercell. While in Ni_3Pt the Pt site is surrounded by 12 Ni sites with a considerable moment $(0.52\mu_B)$, there are only three Ni neighbors around Pt in the interface layer in the case of $Ni_6/Pt_5(111)$ multilayers with ideal interfaces. In the mixed configurations the number of Ni neighbors at Pt sites at the interface may be larger, but these Ni neighbors are often surrounded by a large number of Pt neighbors and, according to the trends of Fig. 2, their moments are small. Our calculations on Ni_3Pt and Pt impurities in Ni, as well as the experiments in Ni rich Ni-Pt alloys,[17] indicate that high induced moments, of the order of $0.3\mu_B$, are expected to be present at Pt sites surrounded by a large number of Ni neighbors in Ni-Pt systems. The high induced moments obtained by XMCD could indicate that Pt atoms may be diffusing deep into the Ni layers.

Summarizing, we have studied using a first-principles approach the influence of interface mixing on the moment profile in $Ni_6/Pt_5(111)$ multilayers. The importance of orbital contributions to the local moment in these systems was also evaluated. While the presence of interface mixing explains the observed moment profile of Ni in $Ni_6/Pt_5(111)$ multilayers, more research is needed to understand the origin of the discrepancy between theory and experiment regarding the induced moments of Pt.

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