Electronic and magnetic structure of idealized metallic multilayers: Ni_3Fe-FeMn system

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We have studied the magnetic structure of three different superlattices composed of alternating layers of Ni_3Fe and FeMn, arranged in real as well as hypothetical ordered phases. Since the constituent compounds have the fcc structure and nearly the same lattice constant, we consider alternating lattice-matched slabs as idealizations of multilayered systems. By carrying out spin-polarized self-consistent linear muffin-tin orbital band-structure calculations, we have determined the magnetic and electronic properties of superlattices composed of Fe, Ni, and Mn atoms located on a common fcc sublattice. For reasons of computational economy, we limited ourselves to heterostructures modulated along the [001] direction and to very thin slabs. For each of the structures studied, the superlattice unit cell contains eight atoms, with two atoms each on four successive atomic planes. The essential results are as follows: First, the individual atomic moments are not very different from their corresponding values in bulk Ni_3Fe and FeMn compounds. Second, the spins in the atomic planes parallel to the interfaces are ferromagnetically coupled to one other, with, for instance " up" spins for the Ni_3Fe planes and alternating "up" and "down" spins for the FeMn planes. In view of these results, we can expect exchange anisotropy to occur in such multilayers. This theoretical conclusion is consistent with experimental observations of unidirectional anisotropy in two-phase systems composed of Permalloy and FeMn alloys.

I. INTRODUCTION

Metallic layered structures with good structural properties can now be prepared by deposition techniques. For instance, recent experiments indicate that long-range structural order exists in the direction perpendicular to the layers in ≈ 1-μm-thick heterostructures, in which the layers can be as thin as 8–10 Å.[1,2] In the long run, multilayer materials with specific properties will be produced. As a result, various fields of pure and applied physics (superconductivity, magnetism, etc.) are concerned with the actual development of the technique. Up until now, as far as magnetic investigation is concerned, experimental analyses[2–4] as well as theoretical band-structure calculations[5–7] have mainly been devoted to modulated or multilayered structures of ferromagnetic/nonmagnetic materials. In contrast, multilayers formed from ferromagnetic/antiferromagnetic metals have not attracted as much interest. However, special magnetic properties of these systems can be expected due to exchange coupling between the layers, as indicated by experiments on Ni/Mn and Co/Mn layered films.[8]

The present paper deals with such ferromagnetic/antiferromagnetic multilayers composed of alternating slabs of Ni_3Fe and FeMn, both in real or hypothetical ordered structures. This system has been selected because both Ni_3Fe and FeMn have fcc structure at room temperature, with a small mismatch between their lattice constants (2.0%, comparable with that of the easily produced Cu/Ni superlattice). Consequently, we do not introduce strong constraints by assuming that the atoms in Ni_3Fe/FeMn multilayers are located on a common sublattice. In addition, the Néel temperature of the $Fe_{0.5}Mn_{0.5}$ alloy, for which we will assume an ordered structure, is well above room temperature (220 °C). This explains why there has been considerable experimental work devoted to the study of exchange anisotropy in two-layer systems formed from FeNi and FeMn alloys, as well as in single-phase FeNiMn alloys, reinforcing the interest of the present study.

The aim of this paper is to investigate the magnetic coupling between ferromagnetic (Ni_3Fe) and antiferromagnetic (FeMn) layers in an idealized multilayer system. To avoid the theoretical complications associated with disordered alloys and random interfaces, the following approximations are made: First, the atoms are located on a common fcc sublattice. Next, interfacial diffusion is neglected. Finally, existence of chemical and magnetic order is assumed within each layer. These approximations provide us with the possibility of performing standard and reliable self-consistent linear muffin-tin orbital (LMTO) band-structure calculations with spin polarization. Furthermore, owing to computing-time considerations and convergence problems, we limit ourselves to very thin layers. The present computations, however, shed some light on the spin arrangement in the ferromagnetic/antiferromagnetic multilayers. Indeed, despite the fact that the Ni_3Fe and FeMn layers are composed of only two atomic planes in our calculations, we find that the magnetic moments in the multilayers are not dramatically different from their bulk values in Ni_3Fe and FeMn. Consequently, we infer that the same conclusion holds true, *a fortiori*, in modulated structures formed from thicker layers. Furthermore, we find that the interfacial planes are not compensation planes for the moments of the antiferromagnetic material. Thus, unidirectional anisotropy is expected to occur in Ni_3Fe/FeMn multilayers, in agreement with experimental observation of exchange anisotropy in similar two-layer systems.

The paper is organized as follows. In Sec. II we summarize the observations of exchange anisotropy in FeNi/FeMn two-layer systems and in γ-FeNiMn bulk alloys. This section reviews the experimental data closely related to our multilayer systems, as mentioned above. In Sec. III, results of band-structure calculations are given, first in Ni_3Fe, next in ordered FeMn, and finally in Ni_3Fe/FeMn multilayers with different atomic configurations. Discussion of the results is presented in Sec. IV.

II. EXCHANGE ANISOTROPY IN FeNiMn ALLOYS

The concept of exchange anisotropy applies to systems of coexisting ferromagnetic and antiferromagnetic materials.[9] As a result of the exchange coupling between the two spin systems, unidirectional anisotropy can appear, the first effects of which were discovered in Co/CoO systems in 1956.[10] Since that time, evidence of exchange anisotropy has been reported in both single-phase and two-phase systems. Manifestations of exchange anisotropy occur at temperatures such that $T \lesssim T_N \ll T_C$, where T_N and T_C denote the Néel temperature of the antiferromagnet and the Curie temperature of the ferromagnetic material, respectively.[11,12] Other requirements are (i) an intimate contact between the two materials, and (ii) a uniform magnetization of the ferromagnet, which implies particle dimensions or film thickness smaller than the width of domain walls ($\lesssim 0.1$ μm). In this section the salient features of ferromagnetic and antiferromagnetic interactions in FeNiMn alloys are briefly reviewed.

The early experiments of Massenet *et al.*[13] revealed a shift of the hysteresis loop obtained by evaporating a (100—1000)-Å-thick Mn layer on a 1800-Å-thick Permalloy ($Fe_{0.19}Ni_{0.81}$) substrate in the presence of a steady magnetic field. This shift is a manifestation of the exchange anisotropy. In these experiments the samples were prepared by thermal evaporation of the constituents onto glass substrates heated at 300 °C. It was shown that subsequent annealing followed by cooling in a magnetic field modified the hysteresis loop, including the possibility of reversing the sign of the shift. The same kinds of experiments were conducted on Permalloy/$Permalloy_{0.5}Mn_{0.5}$ with comparable results.[14]

Glazer and co-workers[15] and Salanskii *et al.*[16] carried out similar experiments on $Fe_{0.18}Ni_{0.82}$/Mn. In addition to the shift of the hysteresis loop, they found a $\sin\theta$ component in the torque curve and nonvanishing rotational hysteresis losses at high magnetic fields, which are other well-known manifestations of the exchange anisotropy.[9] Since these phenomena occur at temperatures well above the Néel temperature of α-Mn ($T_N = 95.6$ K) and because they are strongly dependent on the annealing time, with important magnetic aftereffects, it was suggested[14,15] that atomic interdiffusion plays an important role in this system. In view of the results of Massenet *et al.*,[13] who showed that the loop shift is maximum at a temperature of about 100 °C, the antiferromangetic material responsible for the exchange anisotropy should be characterized by a Néel temperature $T_N \gtrsim 100$ °C. In this connection, we note that such a Néel temperature is realized in the ternary series $(Fe_{0.2}Ni_{0.8})_{1-x}Mn_x$ when $x \approx 0.6$.

Exchange anisotropy has also been observed at room temperature in $Fe_{0.2}Ni_{0.8}$/Fe_xMn_{1-x}.[17] It was found that the hysteresis-loop shift versus FeMn composition is maximum at the equiconcentration ($x = 0.5$). We compare this observation with the fact that the Néel temperature of FeMn alloys is maximum at the equicomposition ($T_N = 220$ °C; refer to Sec. III B). Conducting similar experiments, Tsang *et al.*[18] emphasized the differences in exchange anisotropy between FeMn sputter-deposited on Permalloy and a Permalloy deposition on FeMn alloys. In the latter case, a subsequent annealing strongly enhanced the loop shift. At the present time, this behavior has no explanation. Finally, it was shown[18] that the loop shift in Permalloy/FeMn systems decreases when the thickness of one of the components increases, indicating that exchange anisotropy is an interfacial property in these two-phase films.

We also report on experiment performed by Gradmann and Salewski[19] in which a $Fe_{0.4}Ni_{0.6}$ (111) film was coated with Mn at 300 K. The layers were extremely thin (from 5 to 150 Å), and interdiffusion was shown to play minor a role, if any, in this system. Magnetization measurements indicated an exchange anisotropy, the effects of which rapidly vanished when the temperature was raised above 100 K. In addition, a ferromagnetic moment reduction was observed indicating that the Mn moments coupled antiferromagnetically with the moments of the NiFe alloy. The moment reduction increased with the thickness of both of the layers and saturated at about $2.2\mu_B$ per surface atom when the thickness of the layers exceeded 10 Å.

We now consider bulk FeNiMn alloys in which exchange anisotropy has been reported. Kouvel[20] found that samples with composition $(Fe_xNi_{1-x})_{0.75}Mn_{0.25}$, when cooled to helium temperature in a magnetic field, had their hysteresis loop shifted in the opposite of the direction of the field used during cooling. For Fe concentrations larger than 50 at. % ($x > \frac{2}{3}$), where the alloy is antiferromagnetic (Fig. 1), this manifestation of unidirectional anisotropy disappears. Similar results have been report-

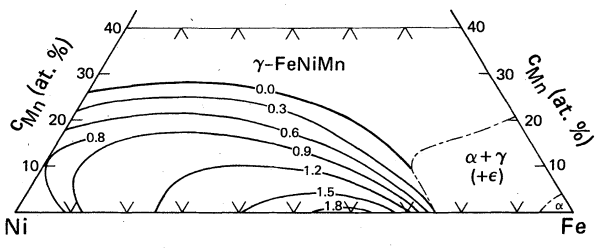

FIG. 1. Magnetic phase diagram of FeNiMn alloys at 0 K. The solid curves represent contour plots of the average ferromagnetic moment (in μ_B per atom) compiled from experimental data (Ref. 51). In the outer region of the curve labeled 0.0 γ-FeNiMn alloys are antiferromagnetic, except for a thin portion of the diagram (not represented in the figure) along the 0.0 curve, where spin-glass behavior is suspected. The dashed curves indicate equilibrium phase boundaries computed at high temperature (Ref. 52) (800 K), from which the magnetic samples were quenched.

ed[21] in the alloy sequence $Fe_{0.65}(Mn_xNi_{1-x})_{0.35}$ for $x \leq 0.4$. In addition, a $\sin\theta$ component in torque curves and high-field rotational hysteresis losses have been detected at temperatures below 5 to 120 K, depending on the actual composition x of the alloy.[21]

Existence of exchange anisotropy in bulk γ-FeNiMn alloys [as well as in NiMn (Ref. 22) and FeNi (Ref. 23) binary alloys] can be understood by assuming the coexistence of ferromagnetic and antiferromagnetic regions.[24] It is not clear,[25] however, that exchange anisotropy in disordered bulk alloys can be interpreted in terms of the classical exchange coupling between macroscopic ferromagnetic and antiferromagnetic materials. We note, however, that the Kouvel experiments[20] indicate that unidirectional anisotropy in FeNiMn alloys is maximum for compositions close to the ferromagnetic-antiferromagnetic border (refer to the curve labeled 0.0 in Fig. 1), where spin-glass behavior is suspected.[26] Ferromagnetic and antiferromagnetic short-range order coexists at low temperature in this region,[27] and small-angle neutron-scattering data indicate the existence of magnetic inhomogeneities, with effective sizes of about 10—15 Å, and maximum effect in the spin-glass region.[26] The presence of a magnetic field during cooling modifies the low-temperature magnetization curves of the samples[28] and probably contributes to the formation of stable ferromagnetic and antiferromagnetic clusters, favoring exchange anisotropy in γ-FeNiMn alloys.

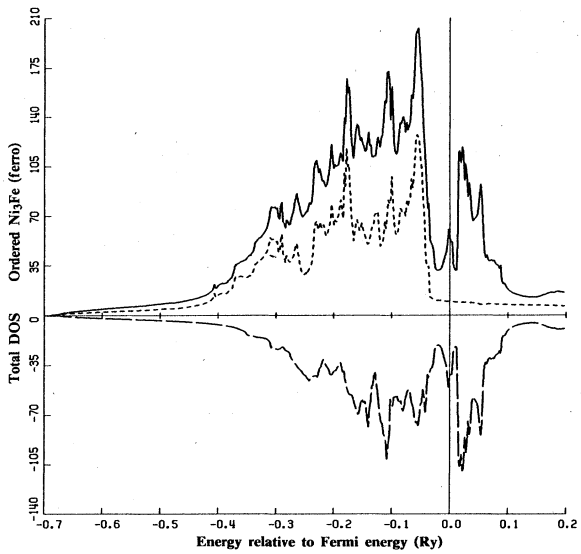

FIG. 2. Spin-polarized densities of states in Ni_3Fe ordered alloy. Here, and in the following figures, the spin-down density of states is represented by negative ordinates. The spin-up density of states is given by the dashed curve and the solid curve is the total density of states (spin down plus spin up). The peak at the Fermi level has mainly Ni character, while the strong double peak above the Fermi level has mainly Fe character. We used 286 $\vec{k}$ points in the irreducible part of the Brillouin zone of the simple-cubic lattice.

III. ELECTRONIC BAND CALCULATIONS FOR THE Ni_3Fe/FeMn SYSTEM

In this section results of band-structure calculations are presented. In these computations an idealized, ordered multilayer system is considered, the atoms being located on a common fcc lattice. Self-consistent spin-polarized LMTO band-structure programs[29] have been used in which exchange and correlation are approximated by the von Barth—Hedin[30] local potential. Immediately below, the results of Ni_3Fe are presented; next, an ordered FeMn compound is considered, and finally, the magnetic properties of the Ni_3Fe/FeMn system are analyzed.

A. Ni_3Fe

Long-range chemical order has been shown to have small effects ($\approx 5\%$) on the magnetic properties of $Ni_{0.75}Fe_{0.25}$ according to neutron experiments[31] and magnetization measurements.[32] This fact, confirmed by coherent-potential-approximation (CPA) calculations,[33] provides us with the possibility of studying the magnetic properties of the above disordered alloy by considering its Cu_3Au ordered phase. Experimental atomic moments determined by polarized-neutron experiments in ordered Ni_3Fe are $\mu_{Fe}=(3.10\pm0.01)\mu_B$ and $\mu_{Ni}=(0.682\pm0.005)\mu_B$.[34]

Figure 2 shows the spin-polarized density of states we computed using the experimental lattice parameter $a=3.556$ Å.[35] The results indicate that Ni_3Fe is a strong ferromagnet (both Fe and Ni majority d bands are filled) with an average magnetic moment equal to $1.19\mu_B$/atom, in agreement with equivalent calculations performed by Williams *et al.*[36] The atomic moments are $\mu_{Fe}=2.79\mu_B$ and $\mu_{Ni}=0.68\mu_B$, not fully consistent with the above experimental values. Among the various approximations which lead to these results (atomic-sphere approximation, linearized energy dependence of the muffin-tin orbitals, etc.), a local approximation to the exchange-correlation potential probably has the most important effects in the present calculations.[37] Indeed, the magnetic moment varies by a factor ≈ 5 between Ni and Fe atoms, and the magnetization density revealed by polarized-neutron experiments[34] is far from being a smooth function in Ni_3Fe. Nevertheless, the results of this computation are satisfactory.

B. Ordered FeMn

In this section we present the results of band-structure calculations which have been worked out on an ordered $Fe_{0.5}Mn_{0.5}$ compound. The existence of this ordered alloy is still hypothetical.[38] Nevertheless, the present calculations shed some light on the magnetic coupling between Fe and Mn atoms embedded in a common lattice, and will be useful when interpreting our results on the magnetic interface in Ni_3Fe/FeMn. Before proceeding with the details of the electronic calculations, we summarize below the main properties of FeMn disordered alloys.

γ-FeMn alloys are antiferromagnetic over the entire range of solid solutions.[39—41] Near the equicomposition, the average magnetic moment shows a "V-shaped curve" as a function of composition, with its minimum of about $1\mu_B$ at equiconcentration, while the Néel temperature

takes its maximum value at that composition.[40] This peculiar behavior has been discussed by Asano and Yamashita[42] in terms of an energy-band-gap mechanism which would be optimum in the 50 at. %—50 at. % alloy.

The spin configuration of γ-FeMn alloys is closely related to the crystallographic structure of these alloys. The Mn-rich alloys (whose γ phase is stabilized by small additions of Cu or C) have face-centered-tetragonal (fct) structure, with a ratio c/a of the lattice parameters smaller than 1, and their spin configuration is collinear.[40] In the collinear model, each (002) atomic plane is ferromagnetic in itself, but the coupling between two successive planes is antiferromagnetic: The magnetic moments are alternately up and down with separation distances equal to half the lattice parameter c. In fact, band-structure calculations[43] indicate that the collinear spin model is the magnetic ground state of fct γ-Mn when $c/a<1$.

When the Fe concentration exceeds ≈ 15 at. %, γ-FeMn alloys have fcc structure—at least at low temperature; Frolov *et al.*[41] have detected a phase transition ($T_t \approx 200$ K, varying slightly with the composition) which they interpreted in terms of a fcc→fct transformation with $c/a>1$. Neutron data indicate that the spin structure of fcc γ-FeMn alloys is presumably noncollinear,[40] the four spins in a cubic cell being assumed parallel to the four diagonals of the cube. Further calculations indicate that the collinear spin model is unstable with respect to multiple spin-density waves in fcc γ-Mn.[44] Furthermore, the noncollinear model agrees with neutron-scattering measurements of spin-wave excitations in γ-FeMn alloys.[45]

The noncollinear spin structure is intimately connected with the cubic symmetry and can be thought of as the superposition of three linear spin-density waves,[46]

$$\vec{P}=\frac{1}{\sqrt{3}}[\vec{x}\cos(Qx)+\vec{y}\cos(Qy)+\vec{z}\cos(Qz)]\ ,$$

where $\vec{P}$ is the modulation of the spin polarization, $Q=2\pi/a$, and $\vec{x}$, $\vec{y}$, and $\vec{z}$ are unit vectors along the three cubic axes. The (average) cubic symmetry existing in the disordered $Fe_{0.5}Mn_{0.5}$ alloy is no longer present in the hypothetical ordered FeMn compound for which we assume the CuAu structure ($L1_0$), in which the (002) planes of the fcc sublattice are alternatively occupied by Fe and Mn atoms. Owing to the tetragonal symmetry of the CuAu structure, we believe that a collinear spin arrangement is the magnetic ground state of FeMn, although the existence of two-component spin-density wave cannot be excluded in tetragonal systems.[43]

In our calculations, we have adopted the lattice parameter $a=3.620$ Å for the fcc sublattice of ordered FeMn, a value slightly smaller than that (3.692 Å) reported for the disordered γ-$Fe_{0.5}Mn_{0.5}$ alloy.[40] The simplest magnetic cell in FeMn is the crystalline tetragonal unit cell, with lattice parameters $a'=a/\sqrt{2}$ and $c'=a$, which contains two atoms, one Fe atom (at the corners) and one Mn atom (at the center). This unit cell yields ferromagnetic Fe and Mn planes perpendicular to the [001] direction. We found that the Fe and Mn planes are antiferromagnetically coupled; the atomic moments obtained are $\mu_{Fe}=+1.24\mu_B$ and $\mu_{Mn}=-1.19\mu_B$, and they lend this system a weak ferrimagnetic character. The density of states of this system is shown in Fig. 3.

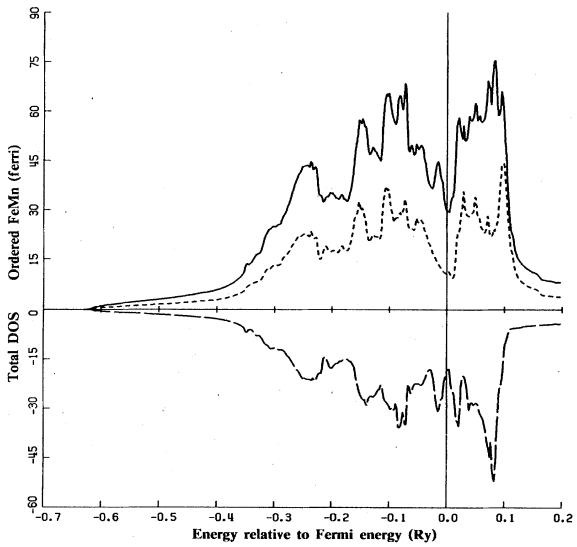

FIG. 3. Spin-polarized density of states of ordered FeMn compound with tetragonal magnetic cell. Calculations, performed using 315 $\vec{k}$ points in the irreducible sector of the Brillouin zone of the tetragonal lattice, show that the Fe moments do not exactly compensate for the Mn moments. Therefore, the spin structure corresponds to a weak ferrimagnetic character of FeMn.

Calculations have shown that a true antiferromagnetic state in FeMn is obtained when considering a simple-cubic magnetic cell which contains four spins. In this structure, discussed by Kasper and Kouvel[47] in their neutron experiments on ordered NiMn, each Fe and Mn plane is antiferromagnetic in itself; the spin arrangement of the (002) planes resembles a checkerboard. The computed sublattice moments are $\mu_{Fe}=0.85\mu_B$ and $\mu_{Mn}=1.55\mu_B$. Figure 4 shows the density of states obtained.

The two spin structures considered above both yield an average sublattice magnetization of about $1.2\mu_B$/atom. This result can be compared with the average antiferromagnetic moment ($1.0\mu_B$/atom) determined experimentally in the disordered γ-$Fe_{0.5}Mn_{0.5}$ alloy.[40] Since we did not perform total-energy calculations, we do not know which one of the two spin configurations corresponds to the magnetic ground state of our ordered FeMn compound. As we show in the following section, the occurrence of the first or the second arrangement of the FeMn layer in the Ni_3Fe/FeMn multilayer depends on the atomic configuration.

C. Ni_3Fe/FeMn multilayer system

We now consider idealized Ni_3Fe/FeMn multilayers as models of the Permalloy/$Fe_{0.5}Mn_{0.5}$ system. In our model, which was designed to be handled by classical band-structure programs, the atoms are located on a common fcc sublattice. The thickness of the Ni_3Fe and FeMn layers is restricted to a few atomic planes because of computation-time and convergence considerations.

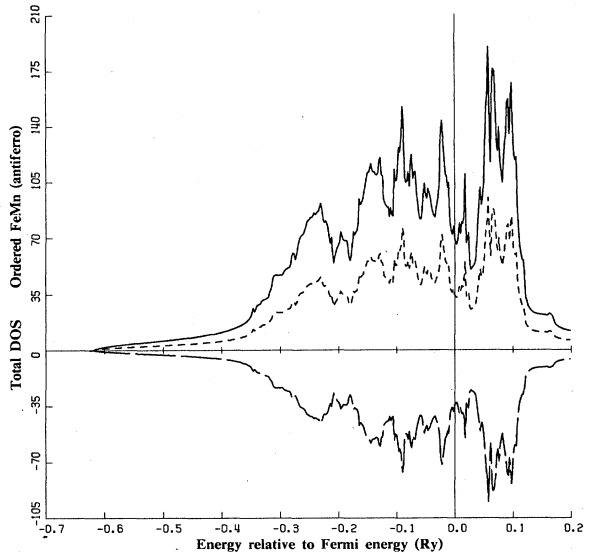

FIG. 4. Spin-polarized density of states of ordered FeMn alloy with cubic magnetic cell. In this system the magnetic structure can be broken down into four cubic sublattices: two Fe sublattices with opposite spins, and two similar Mn sublattices, with a zero net magnetization. In this computation, 286 $\vec{k}$ points were used in the irreducible part of the Brillouin zone of the simple-cubic lattice.

Figure 5 shows the unit cell of three multilayer systems we have considered. There are eight atoms in the tetragonal cell ($c/a=2$). We decided not to determine the lattice parameter of the fcc sublattice by minimizing the total energy of the systems, because this procedure systematically yields lattice parameters which are too small by a few percent. Instead, the lattice parameter of the multilayers was taken to be equal to the arithmetic average of the fcc constants of Ni_3Fe and FeMn ordered compounds, that is, 3.59 Å. Besides, in the language of the atomic-sphere approximation, the lattice parameter of the systems represented in Fig. 5 is related to the average atomic-sphere radius $\bar{S}$ through the formula

$$\bar{S}=\left[\frac{3}{4\pi}\frac{2a^3}{8}\right]^{1/3}.$$

By setting $\bar{S}$ equal to the average of the Fe, Ni, and Mn atomic-sphere radii of the related pure metals, this latter procedure gives exactly the lattice parameter we have determined above.

As an illustration, Fig. 6 shows the self-consistent density of states of the system represented in Fig. 5(a), which is referred to as FeNi/NiNi/FeFe/MnMn. Notice the position of the Fermi level in a valley. The atomic composition of our multilayer systems is $Fe_{0.375}Ni_{0.375}Mn_{0.250}$. Although the disordered FeNiMn alloy with identical composition lies on the border between the ferromagnetic and antiferromagnetic regions (Fig. 1), calculations show that FeNi/NiNi/FeFe/MnMn possesses a net magnetization ($0.70\mu_B$/atom). The atomic moments are given in Table I. We note that the spins on both Mn and Fe [(Fe_2,Fe_3) in Fig. 5(a)] planes are ferromagnetically coupled. In fact, the magnetic structure of the FeMn layer corresponds to the simplest spin arrangement of this system, as explained in Sec. III B. Furthermore, we note that the Mn moments are coupled antiferromagnetically with the moments of the other layers, in agreement with the finding of Gradmann and Salewski[19] that the Mn coating of a FeNi alloy reduces the ferromagnetic moment of the substrate, as discussed in Sec. II. Finally, we note that the atomic moments in FeNi/NiNi/FeFe/MnMn do not differ dramatically from their values in Ni_3Fe and FeMn.

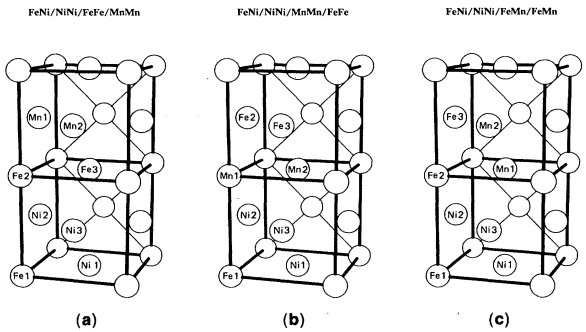

FIG. 5. Tetragonal unit cell of idealized Ni_3Fe/FeMn multilayer systems. The atoms are located on a fcc sublattice, with a lattice parameter $a=3.59$ Å. The unit cell is reproduced periodically in three directions, leading to multilayers modulated along the [001] direction. In these multilayers, the ferromagnetic and antiferromagnetic layers consist of two atomic planes each. Three different atomic configurations of the FeMn layer are shown in panels (a)—(c)

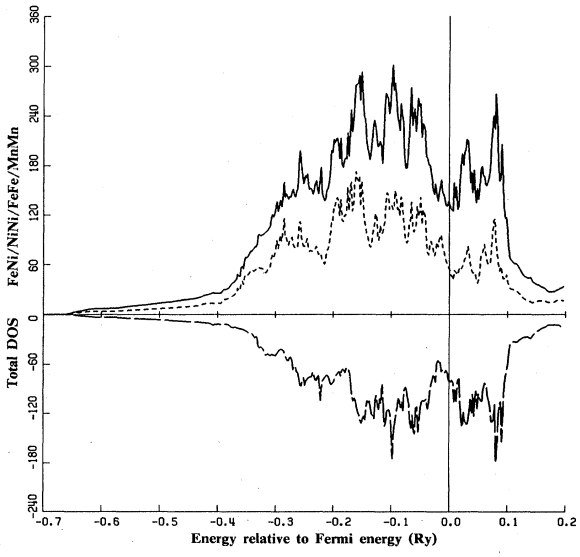

FIG. 6. Spin-polarized density of states of FeNi/NiNi/FeFe/MnMn multilayer [Fig. 5(a)]. The self-consistent band structure was computed at 225 $\vec{k}$ points in the irreducible sector of the first Brillouin zone of the tetragonal lattice.

TABLE I. Atomic moments (in μ_B/atom) in the FeNi/NiNi/FeFe/MnMn system. The atom labels refer to Fig. 5(a).

Atom	Fe_1	Ni_1	Ni_2	Ni_3	Fe_2	Fe_3	Mn_1	Mn_2
Moment	2.34	0.34	0.63	0.57	2.09	1.83	−1.17	−1.03

Of course, the iron moments on Fe_2 and Fe_3 are enhanced with respect to their value in FeMn (Sec. III B) due to the Ni atoms. In addition, the reduction of the Ni_1 moment is attributed to the presence of the underlying Mn plane, for a small Ni moment ($0.3\mu_B$) has been reported in NiMn.[47]

In order not to exclude the possibility of an antiferromagnetic spin arrangement in the FeMn (002) planes, we did not consider the atoms Mn_1 and Mn_2, nor the atoms Ni_2 and Ni_3, as equivalent atoms [refer to Fig. 5(a)]. In fact, the calculations show that these pairs of atoms have parallel moments, as stated above, and should be equivalent. Consequently, the discrepancy between their magnetic moments (see Table I) is a measure of the precision of our calculations. Finally, we mention that the charge transfers were found to be small in this system, as well as in the superlattices considered hereafter. More precisely, the maximum charge transfer has been found in Fe_1 atomic sphere, with a value of nearly −0.04 electron.

Calculations have also been performed for another superlattice [Fig. 5(b)], referred to as FeNi/NiNi/MnMn/FeFe, which can be deduced from Fig. 5(a) by interchanging the Mn and Fe planes of the FeMn layer. The atomic moments given in Table II still confer a net magnetization ($0.55\mu_B$/atom) to the system. By comparison with the previous system (Table II), we note the reduction of the Ni_2 and Ni_3 moments, which take values similar to the Ni moment in NiMn. Furthermore, the moments on the (Fe_2,Fe_3) plane are smaller than in FeNi/NiNi/FeFe/MnMn. This plane may be thought of as belonging to FeMn and Fe_3Ni systems. As a result, it is not surprising that the Fe_2 and Fe_3 moments are intermediate between the values of Fe moment in FeMn ($1.24\mu_B$; see Sec. III B) and Fe_3Ni ($2.0\mu_B$).[36] By contrast, the Mn moments are very close to the values obtained for the first superlattice.

The last atomic configuration we have considered is shown in Fig. 5(c). In this case the (002) planes of the antiferromagnetic layer contain both Fe and Mn atoms, contrary to the previous geometries, for which these planes were Fe or Mn planes. This multilayer system is referred to as FeNi/NiNi/FeMn/FeMn. The magnetic structure (Table III) of this system is similar to that of the previous calculations. We still have a net magnetization ($0.55\mu_B$/atom). However, we emphasize that, in the present situation, the Mn_1 moment is parallel to the host magnetization, while the moment on the Mn atoms was found to be antiparallel in the other configurations (Tables I and II). This behavior is discussed in the next section. Finally, we note that the Ni_2 and Ni_3 moments in FeNi/NiNi/FeMn/FeMn are intermediate between their values reported in Tables I (Ni plane in contact with Fe plane) and II (Ni plane in contact with Mn plane).

IV. DISCUSSION

We begin this section by discussing the orientation of the Mn moments with respect to the host magnetization in the Ni_3Fe/FeMn multilayers we have considered. As pointed out above, the Mn moments are found to be antiparallel, except for Mn_1 in FeNi/NiNi/FeMn/FeMn. In order to understand this behavior, we remark that Mn_1 in this multilayer system has a local environment which is richer in Ni than in Mn_2. In terms of average number $\bar{n}_d$ of d electrons in the first-neighbor atoms, our band-structure calculations indicate $\bar{n}_d = 7.0$ for the mean environment of Mn_1 and $\bar{n}_d = 6.7$ in the case of Mn_2. By contrast, $\bar{n}_d$ for both Mn_1 and Mn_2 is equal to 6.5 in FeNi/NiNi/FeFe/MnMn and 6.8 in FeNi/NiNi/MnMn/FeFe, and both have an antiparallel moment. In relation to Moriya's analysis of exchange interaction in transition metals,[48] we interpret the above results by saying that an antiparallel Mn moment is obtained for a low value of $\bar{n}_d$, while a parallel moment is realized when $\bar{n}_d$ is large, the transition between the two orientations occurring near $\bar{n}_d = 6.9$. This value is close to that found by Moriya for a vanishing exchange energy between Mn (5.5 d electrons) and a transition-metal atom with $\bar{n}_d$ d electrons. In this connection let us add that, in disordered ferromagnetic alloys containing Mn, both parallel and antiparallel orientations of the Mn can occur, depending on local environment, as proved by theory[49] as well as experiment.[50] This peculiarity has been invoked as a possible mechanism responsible for exchange anisotropy in these single-phase alloys.[25]

In connection with the observations of exchange anisotropy in Permalloy/FeMn two-layer systems (Sec. II), we also note that our calculations indicate a ferromagnetic

TABLE II. Atomic moments (in μ_B/atom) in the FeNi/NiNi/MnMn/FeFe system. The atom labels refer to Fig. 5(b).

Atom	Fe_1	Ni_1	Ni_2	Ni_3	Mn_1	Mn_2	Fe_2	Fe_3
Moment	2.41	0.46	0.28	0.24	−1.00	−1.16	1.54	1.63

TABLE III. Atomic moments (in μ_B/atom) in the FeNi/NiNi/FeMn/FeMn system. The atom labels refer to Fig. 5(c).

Atom	Fe_1	Ni_1	Ni_2	Ni_3	Fe_2	Mn_1	Fe_3	Mn_2
Moment	2.36	0.30	0.53	0.42	1.92	1.26	−0.84	−1.54

arrangement of the moments in each atomic plane parallel to the interfaces of the multilayers we have considered. This remark is important because exchange anisotropy would not occur in these systems if the contact plane between layers were a compensation plane of the antiferromagnetic material,[12] as we show below. Furthermore, in the case of FeNi/NiNi/FeFe/MnMn, for instance, any attempt to realize a checkboard-like spin structure in the Fe and Mn planes failed. On the contrary, the self-consistent magnetic structure is that reported above, lending to the FeMn layer the simplest spin arrangement discussed in Sec. III B. In the case of FeNi/NiNi/FeMn/FeMn, the (002) atomic planes still have a ferromagnetic spin arrangement. However, owing to the particularity of this atomic configuration [Fig. 5(c)], the Fe moments as well as the Mn moments in the FeMn layer are antiferromagnetically coupled.

Finally, it is worth noting that the atomic moments given in Tables I—III do not differ dramatically from the corresponding moments in bulk Ni_3Fe and FeMn systems. Presumably, if the thickness of the layers were increased, the moments on the atoms far away from the interfaces would be very close to those in pure ferromagnetic and antiferromagnetic materials, as observed in calculations devoted to multilayers formed from magnetic/nonmagnetic materials.[5–7]

We now show that exchange anisotropy would not occur if the plane in contact with the ferromagnetic layer were a compensation plane for the antiferromagnetic material. As stated in Sec. II, exchange anisotropy is due to exchange coupling between the ferromagnetic and antiferromagnetic layers. According to the Heisenberg Hamiltonian, the exchange-coupling interaction is

$$H_K = -2 \sum_i \sum_j J_{ij} \vec{S}_a^{\,i} \cdot \vec{S}_f^{\,j} ,$$

where i and j denote atomic sites in the antiferromagnetic and ferromagnetic layers, respectively, and $\vec{S}_a^{\,i}, \vec{S}_f^{\,i}$ are the corresponding spins. Owing to the short range of the exchange interaction, we restrict the summations to over the contact planes only (that is, one frontier plane of each material). Assuming a compensation plane for the contact plane of the antiferromagnetic layer, with the further simplifying assumption that the spin structure of this plane corresponds to two sublattices with opposite spins $\vec{S}_a$ and $-\vec{S}_a$, we obtain

$$H_K = -N \left[\sum_j J_{i_1 j} \vec{S}_f^{\,j} - \sum_j J_{i_2 j} \vec{S}_f^{\,j} \right] \cdot \vec{S}_a ,$$

where i_1 (i_2) denotes a site of the sublattice 1 (2) of the antiferromagnetic frontier plane, and N is the number of spins in this plane. The above interaction is usually small (zero in idealized situations) due to the cancellation of the two summations over ferromagnetically ordered spins. By contrast, the exchange-coupling interaction is a maximum when the spins of the antiferromagnetic frontier plane are ferromagnetically ordered, a situation which occurs in our theoretical model of the Permalloy/FeMn interface.

To summarize, using a "first-principles" band-structure calculation, we have analyzed the magnetic structure of multilayers formed from ferromagnetic and antiferromagnetic materials. Within the idealized framework in which our calculations were performed, we find that the spin arrangement does not yield magnetic moment compensation for the interfacial planes of the antiferromagnet. On the contrary, the spins are ferromagnetically ordered on these planes, a situation which favors exchange anisotropy. This result is consistent with the experimental observations of unidirectional anisotropy in Permalloy/FeMn two-layer systems. In the present paper we limited ourselves to (001) interfaces, although epitaxial layers should be parallel to a (111) plane We are currently investigating (111) interfaces in idealized Ni_3Fe/FeMn multilayers, dealing with a large number of atoms per unit cell than in the present calculation. The results will be published in a planned forthcoming paper.

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