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Theoretical study of the onset of magnetism in $(\mathrm{Fe})_x/(\mathrm{Ru})_x$ superlattices

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The electronic and magnetic structure of $(\mathrm{Fe})_x/(\mathrm{Ru})_x$ (x being the number of atomic planes in each layer) hexagonal-close-packed superlattices has been investigated by self-consistent spin-polarized linear-muffin-tin-orbital–atomic-sphere-approximation calculations for x ranging from 1 to 5. It is shown that for $x=1$ no magnetic order exists due to the strong hybridization between the Fe and Ru atoms. For larger thicknesses several magnetic phases appear, with a ferromagnetic, antiferromagnetic, or ferrimagnetic ordering within each layer. The total energies of these phases are very close to each other, and the ground state depends upon x. Comparison is made with the available experimental results on $(\mathrm{Fe})_x/(\mathrm{Ru})_y$ samples grown by molecular-beam epitaxy.

I. INTRODUCTION

Recently artificial materials such as metallic multilayers and superlattices have attracted considerable attention due to their particular magnetic properties. Samples constituted by alternating layers with very well-defined interfaces can now be synthetized by molecular-beam epitaxy (MBE) techniques.[1–3] By these techniques a very large variety of well-ordered periodic structures can be obtained even from two components which do not form spontaneously ordered compounds. This opens a very wide field of research, especially in the domain of magnetism, due to the great sensitivity of the magnetic state of atoms to their chemical environment.

Fe and Ru are miscible in a large interval of concentration, but only form disordered solid solutions.[4] Maurer *et al.*[2] have recently obtained by MBE new well-ordered $(\mathrm{Fe})_x/(\mathrm{Ru})_y$ superlattices (x and y being, respectively, the number of atomic planes in the alternating Fe and Ru layers, ranging from 2 to 7 monolayers for x and 2 to about 40 monolayers for y). The samples have a full hexagonal close packed (hcp) structure with nearly no interdiffusion and sharp interfaces. The lattice parameters have been measured by x-ray techniques and show that Fe is constrained in these materials to adopt an artificially expanded structure which modifies strongly its magnetic properties with respect to those of pure Fe. Bulk magnetic measurements and Mössbauer spectroscopy[2] have put in evidence the existence of a ferromagnetic order in the Fe layers, with about two nonmagnetic atomic planes at either side of each Fe layer. Beyond four Fe atomic planes, the global moment of the Fe layers increases by about $2\mu_B$ per supplementary Fe plane.

As the crystal parameters are well defined, a theoretical study of the onset of magnetism in these multilayers is possible. This is the aim of this paper, and in order to focus attention on the role played by the thickness of the layers, the series $(\mathrm{Fe})_x/(\mathrm{Ru})_x$ ($x=1$ to 5) has been chosen: the relative concentration of the two constituents is maintained constant, so that to a good approximation the dilatation of the lattice for $x=1$ to 5 can be supposed negligible.[2]

We have performed self-consistent band-structure calculations by the "linear muffin-tin orbitals" method (LMTO), in order to obtain all the possible stable magnetic configurations of the atomic planes within each Fe layer. All the Fe layers are supposed to be equivalent, i.e., we do not consider in this first step the possibility of an antiferromagnetic coupling between the Fe layers.

Numerical aspects of the calculations are presented in the second paragraph. In order to distinguish, in the magnetic properties of Fe atoms in superlattices, the role of the Fe volume expansion from the one played by the presence of nonmagnetic Ru neighbors, we have first carried out the electronic structure of a hypothetical pure Fe in the $(\mathrm{Fe})_x/(\mathrm{Ru})_y$ lattice at different volumes (Sec. III). The results obtained for the $(\mathrm{Fe})_x/(\mathrm{Ru})_x$ series are given in Sec. IV for x varying from 1 to 5. They are compared in Sec. V to other theoretical as well as experimental studies of magnetic metallic multilayers.

II. METHOD

The experimental studies of $(\mathrm{Fe})_x/(\mathrm{Ru})_y$ superlattices performed by Maurer *et al.* have shown that these compounds have a hcp structure with a and c parameters depending only upon the Fe and Ru concentrations.[2] Their values follow a "Vegard law" to a very good approximation:

$$c(x,y)=(xc_{\mathrm{Fe}}+yc_{\mathrm{Ru}})/(x+y)\ ,$$

$$a(x,y)=(xa_{\mathrm{Fe}}+ya_{\mathrm{Ru}})/(x+y)\ ,$$

with $c_{\mathrm{Ru}}=4.28$ Å, $c_{\mathrm{Fe}}=4.15$ Å, $a_{\mathrm{Ru}}=2.71$ Å, and $a_{\mathrm{Fe}}=2.63$ Å.

This has been deduced from crystallographic studies of samples with layer thicknesses ranging, respectively, from 4 to 12 Å for Fe and from 4 to 52 Å for Ru. In this paper we restrict ourselves to materials with thin alternate layers of the same thicknesses ($x=y$) in order to study the onset of magnetism independently of the effect of parameters variations: we assume that for these superlattices the Vegard law is still valid, though the layers are thinner than the experimental ones, and that the inter-

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plane distance is the same in the Fe and Ru layers. Then the values of the parameters of the hexagonal cell in $(\mathrm{Fe})_x/(\mathrm{Ru})_x$ are $a=2.67$ Å and $c=4.21$ Å.

In all calculations the crystal is assumed to be perfect. This neglects the possible displacements and local rearrangements of the atoms, with respect to the ideal hcp positions, which certainly exist in the samples because of the very large interatomic distances imposed to Fe by Ru.

We use the self-consistent LMTO method in the frozen-core approximation: the core charge density of each atom is obtained from self-consistent relativistic atomic calculations[5] and is kept unchanged during the iterative scalar relativistic calculation of the band structure (s, p, and d orbitals). The atomic radii (S) for each component of the compounds are chosen to be equal, in the atomic-sphere-approximation (ASA); the departure from neutrality in each Wigner-Seitz sphere gives an estimate of the electronic redistribution in the compound.

The Brillouin zone (BZ) integrations for the densities of states are performed on a grid of 196 **k** points in the irreducible zone. The c lattice parameter of the unit cell in $(\mathrm{Fe})_2/(\mathrm{Ru})_2$, $(\mathrm{Fe})_3/(\mathrm{Ru})_3$, $(\mathrm{Fe})_4/(\mathrm{Ru})_4$, and $(\mathrm{Fe})_5/(\mathrm{Ru})_5$ being, respectively, 2, 3, 4, and 5 times larger than the one in $(\mathrm{Fe})_1/(\mathrm{Ru})_1$, their Brillouin zones are reduced by the same factor along the k_z axis in the reciprocal space with respect to that of $(\mathrm{Fe})_1/(\mathrm{Ru})_1$. Since the number of **k** points is the same in the five compounds, the density of **k** points, and therefore the fineness of the mesh, increases with the number x of monolayers. This is necessary in order to maintain good accuracy in the density-of-states calculations for an increasing number of bands in the same energy interval, resulting from the increase of the number of atoms per unit cell. The reliability of the numerical results has been tested by considering a pure hcp Fe sample as a layered compound $(\mathrm{Fe})_x/(\mathrm{Ru})_x$ with increasing values of x, and has been controlled by a band-structure calculation of $(\mathrm{Fe})_2/(\mathrm{Ru})_2$ with a mesh of 936 **k** points. Herman *et al.*[6] have studied the convergence of spin distribution in Co/Cr bcc superlattices as a function of the number of mesh points in the reduced zone, within the LMTO method. According to their results, it seems again that a grid of about 200 points is a good compromise between accuracy and computing time.

The relative stability of the different phases in each superlattice is obtained by comparison of the total energies of the electronic distributions. In the frozen-core approximation the intrasite core contribution E_c to the total energy can be separated from all the other terms.[7] E_c stays unchanged during the iterative process and is independent of the nuclear positions. It can be omitted as long as absolute values of the energies are not required.

III. PURE EXPANDED hcp Fe

Under normal conditions of temperature and pressure pure iron is stable in the bcc phase ($a=2.866$ Å). This phase is ferromagnetic with a moment of 2.2 μ_B per atom. At pressures in excess of ≈ 130 kbar one obtains the hcp ϵ phase ($a=2.46$ Å, $c=3.94$ Å at 145 kbar) which is paramagnetic.[4] These two phases correspond to structures which are more compact than that of ruthenium ($a=2.706$ Å, $c=4.281$ Å).

In Fe/Ru superlattices Ru imposes its expanded hcp structure on Fe. The only influence of Fe is a slight contraction of the lattice when its concentration increases. For equal concentrations of the two components Fe undergoes a volume expansion of 10.4% with respect to the α phase and 22.0% with respect to the ϵ phase.[2] The magnetic properties of a compound depending strongly upon the structure and the interatomic distances, one can expect important modifications of Fe properties in $(\mathrm{Fe})_x/(\mathrm{Ru})_x$ as compared to those in regular α and ϵ phases. For this reason it is interesting to study theoretically in a first step a hypothetical pure expanded hcp iron.

We have calculated the band structure of hcp Fe in its paramagnetic (PM), ferromagnetic (FM), and antiferromagnetic (AF) phases with lattice parameters corresponding to the experimental values[2] of a and c for superlattices with Fe concentration equal to 100% ($S=2.716$ a.u., $c/a=1.578$), 75% ($S=2.733$ a.u.), 50% ($S=2.756$ a.u., $c/a=1.580$), 25% ($S=2.778$ a.u.), and 0% ($S=2.796$ a.u., $c/a=1.582$). The comparison of the total energies of these three phases shows that the ground state is AF up to $S\approx 2.78$ a.u. and FM beyond [Fig. 1(a)]. The evolution of the FM and AF moments versus S are given in Fig. 1(b). These calculations have been performed with a fine grid of 936 **k** points in the irreducible BZ.

Figures 2(a) and 2(b) compare the densities of states of expanded hcp Fe ($S=2.756$ a.u.) in FM state with those of hcp Co in its regular structure. These curves present similar shapes which are typical of a hcp lattice and consist mainly in two peaks A and B separated by a large zone of weak density. In this phase the majority and minority conduction bands are simply shifted one with respect to the other with nearly no deformation. As the majority d band is almost filled, the electronic properties are controlled by the filling of the minority d band. This places the Fermi level in the weak density zone in Co [Fig. 2(b)] with a resulting moment of 1.58 μ_B, and in peak A in Fe [Fig. 2(a)].

Figures 3(a) and 3(b) show the densities of states of Fe and Co, respectively, in the AF phase. This phase is metastable for Co ($\mu=0.76\mu_B$). Contrary to the FM case, there is almost no shift between the majority and minority conduction bands, but an important deformation of the first one compared to the second one. The characteristic peaks A and B of the hcp structure exist in the two spin directions, but they are strongly reduced in the majority band, especially in Fe where the moment is greater than in Co. The situation is reversed here compared to Fig. 2: in the AF phase the Fermi level is in peak B for Co whereas it lies in a weak density zone for Fe.

Pure hcp Co is of course FM and the AF phase is about 25 mRy higher in energy. At first sight one could try to justify this result by an argument similar to the one giving the Stoner criterion for the instability of PM phases: a Fermi level in a low and rather flat density region might be more energetically favorable and more stable with respect to small shifts of the up and down densities than when it lies in a peak, as it is the case for AF Co. However, this argument is not valid for hcp Fe. For $S=2.756$ a.u. the FM phase presents a peak at the Fermi level and is indeed 5 mRy above the AF phase. But for $S=2.796$ a.u. the FM phase is 1.3 mRy below the AF phase though the densities of states in the two phases and the position of E_F are nearly the same as in Figs. 2(a) and 3(a). This shows that for these artificial structures presenting several magnetic phases with very close total energies, a complete total-energy calculation is necessary to find the ground state, since it results from very small rearrangements of the occupied states in each of the possible configurations. Theoretically a great deal of study has been devoted to the problem of relative stabilities of bcc and fcc iron in magnetic and nonmagnetic phases,[8] but in the case of hcp Fe there are few results. Recently, Kübler[9] has calculated the total energy of pure hcp Fe as a function of S in PM, FM, and AF phases by the ASW method: for small atomic volumes the ground state is nonmagnetic and becomes AF at $S=2.625$ a.u. At $S\simeq2.63$ a.u. a metastable high spin FM phase appears which becomes the stablest one for large values of

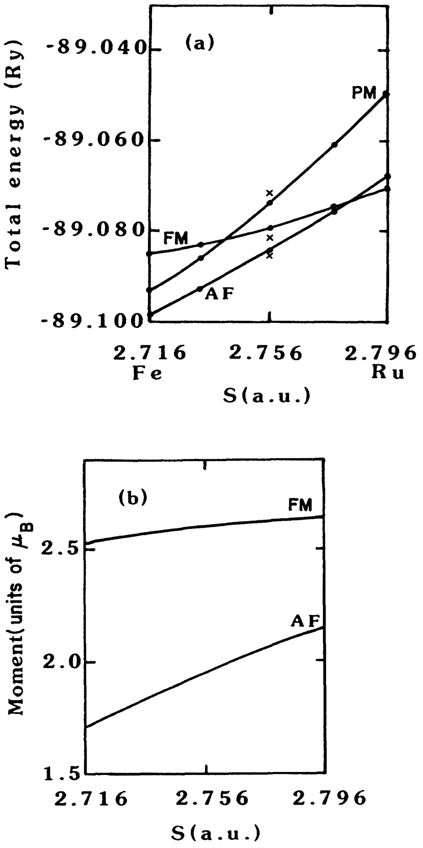

FIG. 1. (a) Total energy per atom (intra-atomic core contribution substracted) for hcp Fe vs the Wigner-Seitz radius S. PM paramagnetic; FM ferromagnetic; AF antiferromagnetic. ●: $c/a\simeq1.58$ (see text), ×: $c/a=1.633$. (b) Corresponding magnetic moments.

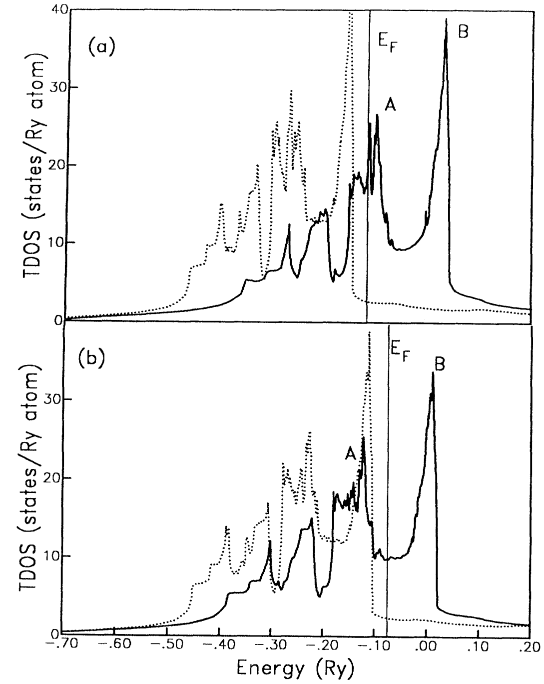

FIG. 2. Total densities of states in a (a) pure ferromagnetic hcp Fe ($S=2.756$ a.u.) and in (b) Co. (——) minority spin electrons; (· · · ·) majority spin electrons.

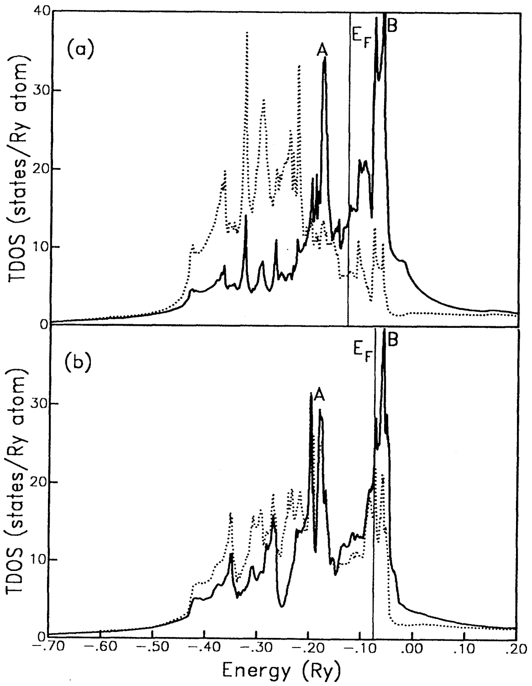

FIG. 3. Total densities of states in (a) pure antiferromagnetic hcp Fe ($s=2.756$ a.u.) and in (b) antiferromagnetic Co. (——) minority spin electrons; (· · · ·) majority spin electrons.

S. This general behavior, together with the value of the moments in these two magnetic states are in good agreement with our results though the two calculations were not performed with the same values of the parameters: $c/a = 1.633$ a.u. in Ref. 9, which is the standard value for close packed structures, whereas in Fig. 1(a) c/a varies slightly from 1.578 for $S = 2.716$ a.u. to 1.582 for $S = 2.796$ a.u.

The effect of a small variation of c/a is visualized in Kübler's paper for the FM phase. In the present calculation, the crosses in Fig. 1(a) for $S = 2.756$ a.u. represent the total energies for the three phases for $c/a = 1.633$. The tendency in both results is the same: a smaller value of c/a shifts the occurrence of the transitions to higher S values. This could partly explain the differences in the values of S for the onset of the FM phase, though for $S = 2.756$ a.u. and $c/a = 1.633$ it is still not the stablest one in our calculation. Other sources of discrepancies, which concern very small energy differences, may be found in the details of the two numerical methods.

The conclusion of this calculation is that in the major part of the parameter interval of the Fe/Ru superlattices, the tendency of iron is to adopt an AF configuration between consecutive planes of the hcp structure. The FM phase becomes more stable only for the largest values of S.

IV. $(\mathrm{Fe})_x/(\mathrm{Ru})_x$ SUPERLATTICES

We present in this section the results of the calculation of the band structure of $(\mathrm{Fe})_x/(\mathrm{Ru})_x$ superlattices, x ranging from 1 to 5. As Fe and Ru concentrations are identical in the five cases, S is supposed to be always equal to 2.756 a.u. The c/a ratio of the unit cell is proportional to x: it is equal to 1.580 in $(\mathrm{Fe})_1/(\mathrm{Ru})_1$, 3.160 in $(\mathrm{Fe})_2/(\mathrm{Ru})_2$, 4.740 in $(\mathrm{Fe})_3/(\mathrm{Ru})_3$, 6.320 in $(\mathrm{Fe})_4/(\mathrm{Ru})_4$, and 7.900 in $(\mathrm{Fe})_5/(\mathrm{Ru})_5$. For parameters corresponding to those of $(\mathrm{Fe})_x/(\mathrm{Ru})_x$ superlattices, pure iron is AF with a moment of 1.97 μ_B and has a moment of 2.60 μ_B in the metastable FM phase. Ruthenium is paramagnetic. Its density of states also presents the characteristics of the hcp structure: two peaks A and B separated by a large zone of weak density (Fig. 4), but the bandwidth is much larger than for iron.

A. $(\mathrm{Fe})_1/(\mathrm{Ru})_1$

We found no stable magnetic state in $(\mathrm{Fe})_1/(\mathrm{Ru})_1$. Figure 5 shows the partial densities of states for the two components of the compound in the paramagnetic phase. The characteristic peaks of the hcp structure are conserved on each atom, but because of the strong hybridization between Fe and Ru the peaks A and B of Fe are at the same energies than those of Ru, which induces an expansion of about 30% of the conduction bandwidth of Fe in $(\mathrm{Fe})_1/(\mathrm{Ru})_1$ with respect to that of the hypothetical pure Fe.

The departure from neutrality in the Wigner-Seitz sphere of each components is weak ($\simeq 0.2$ electron), but contrary to the general tendency in metallic ordered compounds, this slight electron displacement goes from the heaviest atom (Ru) toward the lightest atom (Fe).

B. $(\mathrm{Fe})_2/(\mathrm{Ru})_2$

In this compound the Fe planes are no more sandwiched on the two sides by the Ru layers, therefore the hybridization between Fe and nonmagnetic Ru is weaker than in $(\mathrm{Fe})_1/(\mathrm{Ru})_1$. This explains the existence, in addition to the paramagnetic state, of two stable magnetic phases: a ferromagnetic one and an antiferromagnetic one. In these two phases, a weak moment appears by hybridization on the Ru atoms, which at the interfaces is aligned antiparallel to that of the Fe nearest neighbors (Fig. 6). The ground state is AF (Table I), and in this phase the total moment for the Fe layer is zero.

Figures 7(a) and 7(b) show the densities of states of Fe in the FM and AF states. The characteristic peaks A and B of the hcp structure are strongly deformed because of the hybridization between the two chemical species, but the bandwidth of Fe for each spin direction is now comparable to that of pure Fe in the same lattice. The departure from neutrality in the Fe and Ru spheres is weaker in $(\mathrm{Fe})_2/(\mathrm{Ru})_2$ than in $(\mathrm{Fe})_1/(\mathrm{Ru})_1$ and is almost the same in the two magnetic phases (Table I). The decomposition of the Fe moment in s, p, and d symmetries shows that the d contribution is the dominant term. The calculation on a grid of 936 **k** points leads to

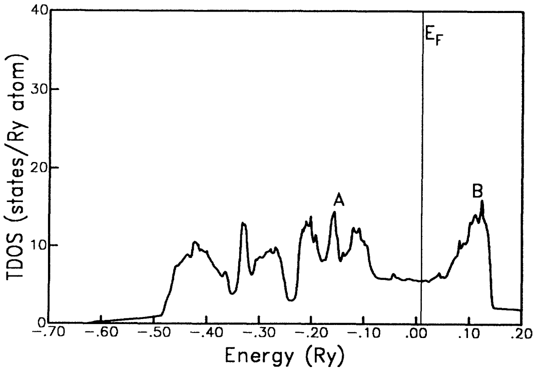

FIG. 4. Total density of states per spin of pure hcp Ru ($S = 2.756$ a.u.).

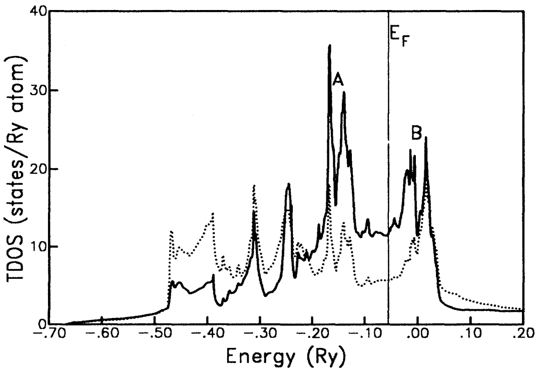

FIG. 5. Total densities of states per spin on Fe atoms (——) and Ru atoms (· · · ·) in $(\mathrm{Fe})_1/(\mathrm{Ru})_1$.

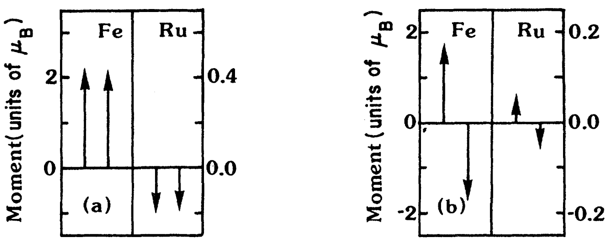

FIG. 6. Fe and Ru moments on each atomic plane in $(Fe)_2/(Ru)_2$ in (a) ferromagnetic and (b) antiferromagnetic phases.

variations of the order of 0.01 electron for the partial numbers of electrons, of 0.01 μ_B for the Fe moments, and of 0.001 μ_B for the Ru moments. The weak differences between the results obtained with this mesh and those obtained with a grid of 196 points show the good numerical stability of the results.

C. $(Fe)_3/(Ru)_3$

As in $(Fe)_2/(Ru)_2$, we found in $(Fe)_3/(Ru)_3$ three phases. The PM and FM states are also metastable. The negative coupling between Fe planes within each layer, which corresponds to an AF phase in $(Fe)_2/(Ru)_2$, gives in $(Fe)_3/(Ru)_3$ a ferrimagnetic (FI) phase, which is the ground state (Table II). The moments on each Fe and Ru plane for the FM and FI phases are given in Fig. 8. In $(Fe)_3/(Ru)_3$ the sites at the interface of a layer (type I: Fe I and Ru I) and those in the central plane (type II: Fe II and Ru II) are not equivalent. On Fe I atoms, in the two phases, the moment is weaker than in pure expanded hcp because of the hybridization with nonmagnetic Ru. For Fe II atoms the hybridization with Ru is weak and the moment (FM: 2.67 μ_B; FI: 2.17 μ_B) is reinforced by the charge transfer (Fe II loses 0.05 electrons from its minority band) (Table II) and is therefore slightly greater than in pure Fe. The number of Fe monolayers is odd and the positive and negative Fe moments of the successive monolayers do not cancel each other. The resulting total moment for the Fe layer is -0.83 μ_B (the moment on the central plane being supposed to be positive).

The densities of states in $(Fe)_3/(Ru)_3$ present the same characteristics as those obtained for $(Fe)_2/(Ru)_2$, but due to the larger thicknesses of the Fe and Ru layers peaks *A* and *B* are less deformed, especially on sites of type II. The departures from neutrality in each Wigner-Seitz sphere in $(Fe)_3/(Ru)_3$ are of the same magnitude as those in $(Fe)_2/(Ru)_2$ (Table II).

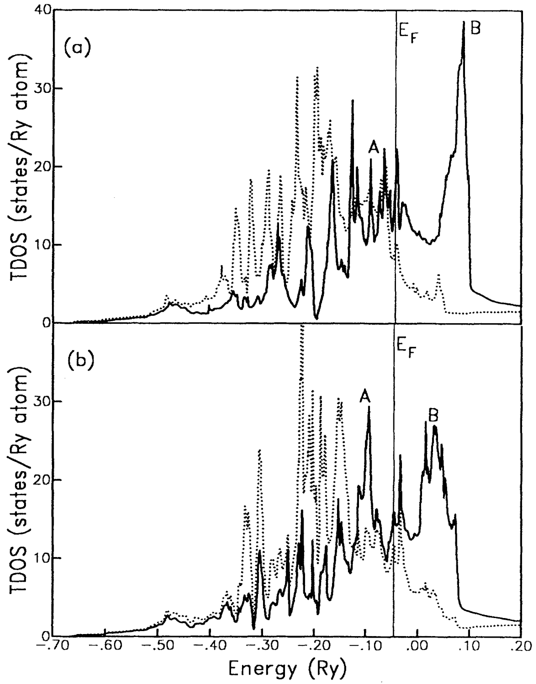

FIG. 7. Total densities of states of Fe in $(Fe)_2/(Ru)_2$ in (a) ferromagnetic state and in (b) antiferromagnetic state. (——) minority spin electrons; (· · · · ·), majority spin electrons.

D. $(Fe)_4/(Ru)_4$

In $(Fe)_4/(Ru)_4$ four stable phases exist. The PM and FM [Fig. 9(a)] states are still metastable and there are in addition two FI states. The first one, with parallel moments on the two central planes of the Fe layer, is metastable [Fig. 9(b)]. The second one, which is the ground state (Table III), corresponds to an AF coupling with moments of opposite directions on the successive Fe planes [Fig. 9(c)] (this state is therefore noted "AF"). In the ground state the total moment of the Fe layer is equal to zero. The charge transfers in each Wigner-Seitz sphere are given in Table III: the Fe II spheres lose less electrons in $(Fe)_4/(Ru)_4$ than in $(Fe)_3/(Ru)_3$ and their moments (FM: 2.62 μ_B; "AF": 2.11 μ_B) are closer to those in pure expanded hcp Fe. At the interface the Fe I moments in the FM and "AF" states, are slightly smaller than in $(Fe)_3/(Ru)_3$ because of the larger thickness of the Ru layer.

E. $(Fe)_5/(Ru)_5$

In $(Fe)_5/(Ru)_5$ we found no self-consistent solution with antiparallel moments corresponding to an AF coupling

TABLE I. $E-E_{PM}$: difference between the total energy of the ferromagnetic (FM) or antiferromagnetic (AF) state and the paramagnetic one in $(Fe)_2/(Ru)_2$. ΔQ: excess number of electrons in each Fe Wigner-Seitz sphere.

	$E-E_{PM}$ (mRy)	ΔQ Fe	Partial Fe moment (μ_B) s	p	d
FM	-3.09	0.065	-0.007	-0.025	$+2.261$
AF	-5.73	0.067	$+0.003$	$+0.005$	$+1.737$

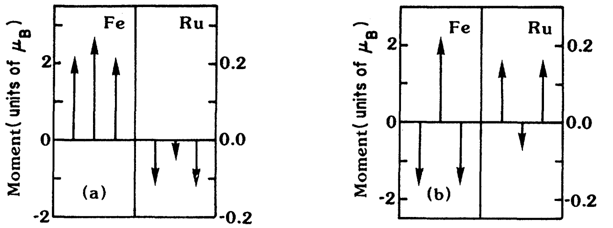

FIG. 8. Fe and Ru moments in $(Fe)_3/(Ru)_3$ in (a) ferromagnetic and (b) ferrimagnetic phases.

between Fe planes in the layers. The PM and FM [Fig. 10(a)] states are metastable and the ground state is the FI phase [Fig. 10(b)] with parallel moments on the three central planes of the Fe layer (Table IV). This gives a total moment for the Fe layer of 4.4 μ_B. In the FM phase the central atoms Fe III of the Fe layer have the same moment as those in pure HCP Fe (2.60 μ_B). On the second planes, Fe II atoms have a moment slightly higher than 2.60 μ_B because of the charge transfer (Table IV).

In the FI phase the moment of the central atoms (Fe II and Fe III) increase with respect to that of Fe II in $(Fe)_3/(Ru)_3$. As in the FM phase, the magnitude of the Fe moment at the interface decreases with increasing Ru thickness.

V. DISCUSSION

The magnetic properties of iron are very sensitive to the local environment of its atoms. hcp pure iron is only obtained at high pressure and is thus nonmagnetic, due to the compression of the lattice, as it is now well understood from theoretical calculations (Refs. 9 and 10 and this work). Pearson and Williams[4] succeeded in elaborating hcp iron at normal pressure by alloying it with 30% Ru. The lattice is then slightly expanded ($\approx 5\%$) compared to the α phase volume and the sample exhibits weak antiferromagnetism ($\mu_{Fe} \approx 0.1 \mu_B$) at low temperature.

In the new magnetic $(Fe)_x/(Ru)_y$ multilayers synthetized by Maurer *et al.*,[2] the iron layers are still in a more expanded hcp structure. Then, in view of the theoretical results for pure hcp iron versus volume expansion, one would expect either an AF or, for the largest values of S, a FM phase, both with large values of the moments on each Fe atom. This FM ordering is observed for thick Ru layers and Fe layers thicker than 4 atomic planes, but with nearly two dead magnetic planes on either side of each Fe layer. The presence of these nonmagnetic iron sites has been confirmed recently by Liu and Bader[11] for thin films of Fe grown on (0001) Ru. Similar results have been obtained previously for bcc Fe/*V* superlattices,[12] but the existence of dead layers is in this case controversial and seems to depend upon the sharpness of the interfaces.

Comparative studies have been performed on other similar multilayers obtained by MBE or sputtering techniques, such as hcp Co/Ru and Co/Cr or bcc Fe/Cr.[13,14,3] For this last superlattice as well as for magnetic double layers[15] the magnetic moments of successive Fe layers on either side of the Cr spacer layer are reported to be aligned antiparallel.

Recent self-consistent calculations of the electronic structure of (110) Fe/Cr superlattices have been performed by Xu *et al.*[16] by the LMTO method. In these calculations, two Cr atoms in the same atomic layer are supposed to be of different types, all the iron sites in one plane being found to be equivalent except for Fe_1/Cr_1. The moments on the Fe sites are not very different from the bulk values, even for very thin layers. No dead layers are obtained here, neither in the case of bcc Co/Cr studied by Herman *et al.*[6] In hcp Fe/Ru, the in-plane structure is hexagonal. It seems then more reasonable than in a cubic structure to assume that all the sites within one plane of given chemical species are equivalent, in order to avoid frustration. In our calculations we considered all the possible magnetic configurations of the different atomic planes, the successive Fe layers being supposed to be identical.

Except for $x=1$, several magnetic phases have been obtained in $(Fe)_x/(Ru)_x$, with very close total energies. Figure 11 shows the values of the total-energy differences between these phases and the nonmagnetic one versus x (these energies have been divided by x for comparison and it has been verified that the uncertainty on the total-energy values due to the choice of the number of **k** points in the BZ does not exceed 0.5 mRy). The difference between two magnetic phases is of the order of 2 mRy (times x), which is smaller than in pure hcp iron and typically one order of magnitude smaller than in pure bcc iron. Hence, these total-energy calculations give a good indication on the different magnetic orders one can expect in these multilayers, but their relative stabilities may

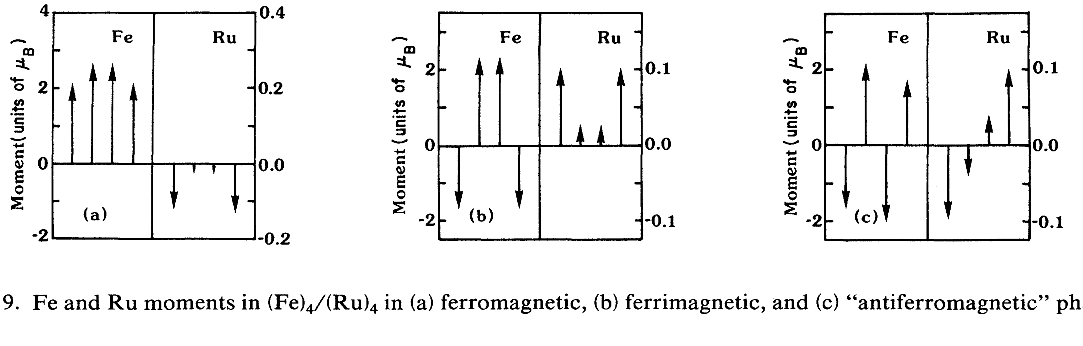

FIG. 9. Fe and Ru moments in $(Fe)_4/(Ru)_4$ in (a) ferromagnetic, (b) ferrimagnetic, and (c) "antiferromagnetic" phases.

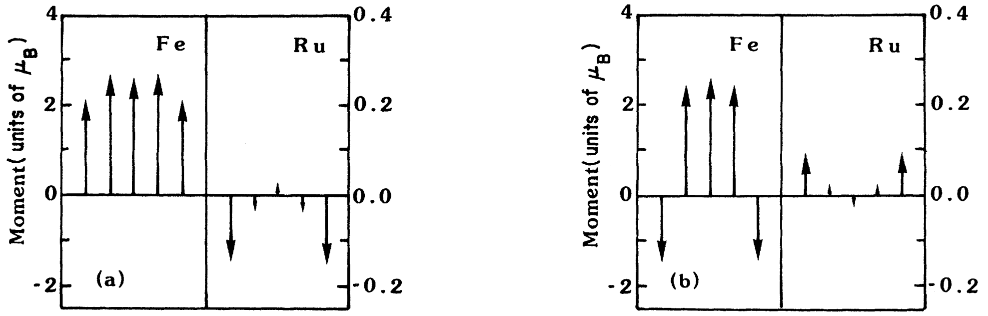

FIG. 10. Fe and Ru moments in $(Fe)_5/(Ru)_5$ in (a) ferromagnetic and (b) ferrimagnetic phases.

TABLE II. $E-E_{PM}$: difference between the total energy of the ferromagnetic (FM) or ferrimagnetic (FI) state and the paramagnetic one in Fe_3/Ru_3; ΔQ: excess number of electrons in each Wigner-Seitz sphere (Fe I and Ru I: Fe and Ru at the interfaces; Fe II and Ru II: central Fe and Ru).

	$E-E_{PM}$ (mRy)	ΔQ (electron) Fe I	Fe II	Ru I	Ru II
FM	-5.97	$+0.10$	-0.05	-0.09	$+0.03$
FI	-13.84				

TABLE III. $E-E_{PM}$: difference between the total energy of the ferromagnetic (FM) or ferrimagnetic (FI) or "antiferromagnetic" (AF) state and the paramagnetic one in $(Fe)_4/(Ru)_4$. ΔQ: excess of electrons in each Wigner-Seitz (Fe I and Ru I: Fe and Ru at the interfaces; Fe II and Ru II: central Fe and Ru).

	$E-E_{PM}$ (mRy)	ΔQ (electron) Fe I	Fe II	Ru I	Ru II
FM	-7.79	$+0.09$	-0.02	-0.08	$+0.01$
FI	-15.61				
AF	-19.87				

TABLE IV. $E-E_{PM}$: difference between the total energy of the ferromagnetic (FM) or ferrimagnetic (FI) state and the paramagnetic one in $(Fe)_5/(Ru)_5$. ΔQ: excess number of electrons in each Wigner-Seitz sphere (Fe I and Ru I: Fe and Ru at the interfaces; Fe II and Ru II: Fe and Ru second planes; Fe III and Ru III: central Fe and Ru).

	$E-E_{PM}$ (mRY)	ΔQ (electron) Fe I	Fe II	Fe III	Ru I	Ru II	Ru III
RM	-12	$+0.071$	-0.010	~ 0	-0.066	$+0.006$	-0.001
FI	-20	$+0.056$	-0.006	-0.001	-0.052	$+0.001$	$+0.002$

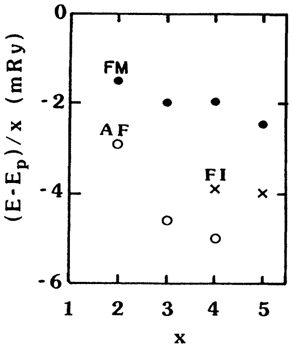

FIG. 11. Differences between the total energies of $(\mathrm{Fe})_x/(\mathrm{Ru})_x$ superlattices in magnetic states and nonmagnetic state vs the number x of atomic planes in each layer (FM: ferromagnetic; FI: ferrimagnetic; AF: "antiferromagnetic". (These energies have been divided by x.)

of course easily be reversed by very small perturbations, so that the ground state of the real sample cannot be predicted without ambiguity.

The general tendency, when the thickness of the layers is increased, is to stabilize the magnetic phases compared to the nonmagnetic one. For $x = 5$ no antiferromagnetic order was obtained. The stable phase is the FI one, with the three central Fe atoms aligned parallel and a large global moment in the Fe layer. For $x \leq 4$ the theoretical global moment remains negligible or small in the AF ground state, depending upon the parity of x. This could be consistent with the bulk magnetic measurements[2] showing ferromagnetic ordering for $x > 4$, except for the fact that we did not obtain dead layers at the interfaces. On the first Fe plane the moments are reduced compared to their values in the center of the layer [or even killed for $(\mathrm{Fe})_1/(\mathrm{Ru})_1$], due to hybridization with the atoms of the spacer layer, as it was also found in Fe/Cr.[16] For the same reason the Ru atoms are slightly polarized, the moments at the interfaces on Fe and Ru neighbors being always antiparallel.

The reasons why we did not obtain nonmagnetic iron sites can be manyfold. The magnetic measurements[2] have been performed on samples with large nonmagnetic spacer layers: $(\mathrm{Fe})_x/(\mathrm{Ru})_{13}$. This spacing could be large enough to considerably reduce the magnetic coupling between the Fe layers. These samples are therefore not directly comparable to the $(\mathrm{Fe})_x/(\mathrm{Ru})_x$ series, besides the fact that their lattice is more expanded. This could explain why on the contrary dead layers were obtained by Selme and Pecheur[17] for large slabs within the tight-binding approximation. But the most probable reason is that we considered here ideal crystals with perfect interfaces, equidistant atomic planes, and atoms at the exact hcp standard positions. In the experimental samples the interfaces are defined within 1 Å; this does not rule out imperfections such as steps or antistructure defects, which could reduce considerably the atomic moments on the first Fe plane. Moreover, the large lattice mismatch between Fe and Ru has to be compensated by stacking faults or relaxations. de Andres *et al.*[18] have shown from measurements of the intensities of the diffraction satellites that the distance between two Fe planes is approximately 0.5% shorter than between two Ru planes; this could be taken into account in the calculation. The value of the moments at the interfaces should also be very sensitive to the distance Fe-Ru which has not been measured precisely up to now.

In summary, we have shown in this work that for the series $(\mathrm{Fe})_x/(\mathrm{Ru})_x$ several different magnetic orders are possible within each Fe layer, with very close total energies. The next step will be to study the effect of the dilatation of the lattice with increasing width of the Ru layers for a given Fe width,[19] which should stabilize a ferromagnetic order, and to examine the possibility of an AF interlayer coupling between the Fe layers, such as the one reported for Co/Ru or Fe/Cr.[13]

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