Magnetic polarization of the Pd spacer and interlayer magnetic couplings in Fe/Pd (001) superlattices: First principles calculations

D. Stoeffler and K. Ounadjela
Institut de Physique et de Chimie des Matériaux de Strasbourg-Gemme, Université Louis Pasteur, 4 rue Blaise Pascal, 67070 Strasbourg, France

J. Sticht*
Technische Hochschule, D-6100 Darmstadt, Germany

F. Gautier
Institut de Physique et de Chimie des Matériaux de Strasbourg-Gemme, Université Louis Pasteur, 4 rue Blaise Pascal, 67070 Strasbourg, France
(Received 3 August 1993)

This paper is devoted to a theoretical study of the magnetic properties of Fe/Pd superlattices. We mostly focus our attention on the determination of both the interlayer magnetic couplings (IMC) and the magnetic-moment distributions as a function of the Pd spacer's thickness. We use an *ab initio* method (augmented spherical wave) to determine self-consistently the electronic structure, the magnetic-moment distributions, and the total energies for the considered systems. We consider two model structures for the Pd spacers (1) a fct structure for which the Pd atoms keep their bulk atomic volume and (2) a fcc structure for which the Pd atomic volume is expanded. For the first structure, the magnetic polarization in the palladium spacer is limited mostly to the first three monolayers near the Fe-Pd interfaces and the IMC are similar to the ones obtained for nonmagnetic spacers. The IMC decrease rapidly with an oscillating behavior. On the contrary, for the second atomic structure, the whole Pd spacer is polarized with a moment of about $0.15\mu_B$/atom for $n \leq 14$ (n being the number of Pd atomic layers) and the IMC are ferromagnetic in a large range of Pd thicknesses. We present a detailed study of (i) Fe_3Pd_1 superlattices whose ground state is shown to be antiferromagnetic and (ii) the polarization induced in the Pd spacer in relation with the current theoretical models.

I. INTRODUCTION

The recent discovery of oscillating interlayer magnetic couplings (IMC) between ferromagnetic (F) layers A_m (A=Fe, Co, Ni) separated by a nonmagnetic or antiferromagnetic (AF) metallic spacer B_n (Ref. 1) (m and n correspond, respectively, to the number of atomic layers) and of the related giant magnetoresistance[2] (GMR) has stimulated a lot of experimental and theoretical activity. The purposes of such studies are both to understand the physical origins of these effects and to design magnetoresistive devices for various applications. Up to now, the experimental data on sandwiches and superlattices based on noble- and transition-metal spacers allow one to determine general trends as far as the magnitudes of the IMC and the relations between GMR and IMC are concerned. Moreover, the magnitude of the IMC and of their oscillating behavior have been found to be very sensitive to the quality of the interfaces.[3]

From a theoretical point of view, the physical origin of the oscillating behavior of the IMC is qualitatively understood[4-6] from an extension of the classical theory of pair interactions either between magnetic impurities in nonmagnetic hosts[7] or between planar defects in metals.[8] The physical picture for such a coupling can be understood in a perturbation scheme as follows: (1) the first F layer induces in the spacer an oscillating magnetic polarization, (2) this polarization interacts with the second layer and gives rise to the IMC. Therefore, the magnetic-moment distributions in the spacer near an interface separating two semi-infinite crystals is strongly related to the IMC for *large* thicknesses of the metallic spacer. In this asymptotic regime, both quantities present an oscillating behavior whose periods are deduced from the spacer's Fermi-surface topology.[6] Several assumptions have been used to describe semi-qualitatively these effects: Ruderman-Kittel-Kasuya-Yosida (RKKY) theory,[6] hole confinement model,[9] "*s-d*" mixing models based on Anderson's model for impurities in metals.[10] The comparison of theoretical predictions for the period of these oscillations and of experiments has been successful for systems with noble-metal spacers such as Cu/Co.[11] However, for transition-metal spacers the oscillations are multiperiodic from the complicated topology of the Fermi surface and are difficult to relate to experiments.[12] Moreover, the amplitudes and the phases of each of these oscillations are strongly sensitive to the velocities and curvatures of the Fermi surface and to the scattering properties of the F layers.[6] Finally, the range of validity of the asymptotic form for the IMC is difficult to access so that a satisfactory understanding of the amplitudes of the IMC, of their variations along the transition-metal series and of their dependence on the interfaces rough-

0163-1829/94/49(1)/299(11)/$06.00

ness is still lacking.

The IMC can also be determined by calculating directly the total-energy difference ΔE between two magnetic configurations of the A_m layers (F and AF for example) and by using either *ab initio*[13–15] or semiempirical tight-binding (TB) methods.[16] For transition-metal spacers, some calculations have been carried out by TB and *ab initio* methods, both methods agreeing in the considered range of thicknesses and for all investigated superlattices.[13,16,17] In contrast to other models, these calculations do not depend on the assumptions concerning either the spacer's thicknesses ($n \rightarrow \infty$) or the scattering properties of the F layers. They can be performed only for small n values ($\approx 6-8$) from limitations related to (i) the numerical accuracy required for very small IMC (resolution about $\Delta E < 1$ meV/crystallographic cell) and (ii) from the necessary computation time. Nevertheless, for superlattices with noble-metal spacers, the period of the calculated IMC ($n \leq 8$) have been qualitatively related to the shape of the Fermi surface.[15] For superlattices with AF spacers the situation is quite different, the IMC oscillating with a period of two monolayers[13,16] for the systems presenting the best crystalline quality. This oscillation has been related to the existence of a magnetic defect in the AF layer induced by the strong antiparallel interfacial *A-B* coupling.[16,18] Therefore, the corresponding perturbating potential is extended in the whole spacer so that such a situation cannot be described by the previous approaches.

The strong sensitivity of the amplitude and of the periodicity of such oscillations to interfacial roughness has been shown in such superlattices both experimentally and theoretically.[3,16] However, the theoretical values of magnitude of the IMC for all the investigated superlattices are one order of magnitude larger than experimentally observed. The origin of this discrepancy is not clearly understood. Up to now, it has been related to interfacial diffusion or roughness but a systematic study of such effects is lacking.

In this paper, we present a detailed theoretical *ab initio* study of the Fe/Pd IMC and of the related magnetic-moment distributions. Palladium is nearly ferromagnetic so that the magnetic superlattices $A_m\mathrm{Pd}_n$ (where A is a magnetic metal such as Fe, Co or Ni) can *a priori* exhibit new and interesting magnetic features. For example, in pure fcc palladium, the existence of such a large and long-range ferromagnetic Pd polarization is well established around A impurities.[19] Therefore, we can expect a large and long-range palladium polarization near A/Pd interface as resulting from the superposition of the effect of all interfacial A atoms. However, the physical situation is different in $A_m\mathrm{Pd}_n$ superlattices, the magnetic properties being modified by (i) the confinement of the spacer's slab between iron layers and (ii) the strains induced in the A and B slabs by the epitaxial growth of two element with different equilibrium atomic volumes. Therefore, it is essential to take these effects into account and to determine their roles on the magnetic order and on the polarization of $\mathrm{Fe}_3\mathrm{Pd}_n$ superlattices.

Co/Pd and Fe/Pd multilayers have been synthesized experimentally during the last few years. The first system has been intensively studied and exhibits perpendicular anisotropy (the Co magnetization is found perpendicular to the plane of the interfaces for small Co thicknesses).[20] The Fe/Pd sandwiches and multilayers have been studied more recently. Fe(001)/Pd_n(001)/Fe(001) sandwiches have been synthesized by Celinski and co-workers[21,22] with a molecular beam epitaxy technique. They obtain samples of high crystalline quality up to 12 Pd monolayers on Fe(001) with an in-plane Pd lattice parameter's expansion of 5.1% with respect to its equilibrium value. Although the growth of Fe on the Pd spacer present some roughness, the Fe(001)/Pd(001)/Fe(001) trilayer have been found to be well suited for magnetic studies.[21] Ferromagnetic resonance, Brillouin-light-scattering, and surface-magneto-optical Kerr effect techniques have been used in order to measure the exchange coupling. The authors found that (i) the whole Pd spacer is not entirely polarized for Pd thicknesses n larger than 4 monolayers and (ii) the IMC are ferromagnetic, behave nonmonotonously for $n \leq 12$ monolayers, show oscillations with a period of 4 monolayers for $n < 12$, and are strongly sensitive to temperature variations. Fe/Pd(001) multilayers have been realized more recently in the Thomson group[23–26] to study their magnetotransport properties. The superlattices were grown directly on a MgO substrate or with a Pd buffer: Similar epitaxial growths have been obtained in the two cases in agreement with the results of Celinski *et al.* The authors obtained hysteresis loops consistent with an antiferromagnetic (AF) coupling between the Fe layers in the same thickness range[23] as the one observed by Celinski *et al.*[22] However, by a more detailed analysis of the hysteresis loops, they found no evidence for isotropic AF couplings. Moreover, by Kerr optical microscopy, they have recently obtained a strong evidence of a F coupling for $5 \leq n \leq 25$.[25]

Some other experimental works have been done[27,28] on Pd/Fe superlattices but they were mostly focused on the polarization of the interfacial Pd layers—the interfaces corresponding to the Fe(110)/Pd(111) stacking. These works have shown evidence of a strong polarization of the interfacial Pd atoms by Mössbauer analysis[27] and by spin-resolved photoemission.[28]

Theoretically, the Fe/Pd interfaces have been mostly studied[29–31] when ultrathin Pd overlayers are deposited onto a Fe substrate. These studies have shown that the Pd overlayer exhibits a strong polarization with an interfacial magnetic moment of $0.3-0.4\mu_B$. However, it is not possible to split the origin of the polarization into an Fe/Pd interfacial and a surface contribution, so that these results cannot be transposed to a multilayer situation. More recently, $\mathrm{Fe}_5/\mathrm{Pd}_n/\mathrm{Fe}_5$(001) sandwiches and $\mathrm{Fe}_5\mathrm{Pd}_n$(001) superlattices have been studied using a TB description of the electronic structure.[32,33] In these papers, the authors are mainly interested in the polarization induced in the Pd layers by the interfacial hybridization with Fe. They report also magnitudes for the IMC which are very strong as compared to the experimental data, to the results of similar calculations for $\mathrm{Co}_4\mathrm{Pd}_n$ superlattices[16] and to those obtained in this work.

In this paper, our aim is to determine the main features of the magnetic-moment distributions and of the IMC in

the local-spin-density approximation (LSDA) framework. In Sec. II, we describe the method used in this paper and the atomic structures we will consider; we present in Sec. III the magnetic-moment distributions and the IMC obtained for perfect superlattices for $n \leq 6$ monolayers (ML); in Sec. IV, we detail the densities of states in Fe_3Pd_1 and the role of the interfacial Fe-Pd hybridization. Finally, in Sec. V we study the polarization induced in the Pd layers for $n \leq 14$ ML as a function of the structure of the palladium spacer.

II. METHOD

The aim of this work is to study the electronic structure of Fe/Pd superlattices with a method sufficiently precise to exhibit the essential features which allow the physical understanding of the experiments. Here, we have chosen the augmented spherical wave (ASW) method[34] and the LSDA formalism for treating exchange and correlation of a many-electron system which allow one to determine the electronic structure of the superlattices. It has been shown recently, that this method is well suited to study such systems as Fe/Cr,[13] Fe/Mn,[35] Co/Cu,[36] Fe/Nb,[37] Co/Ru,[17] Fe/Si,[38]. . . superlattices.

One essential problem in such calculations is to get self-consistent magnetic-moment distributions and IMC with a sufficient accuracy. Here, the numerical accuracy is mostly determined by the choice of the k points in the Brillouin zone. The IMC are obtained from the energy difference of F and AF interlayer arrangements, where the number of **k** points (N_k) remains unchanged for both calculations. Large N_k values allow one to get a fine resolution of the band structure and of the densities of states but the computer time increases linearly with this number. Moreover, to increase significantly the resolution of the structures from one calculation to the next, the number of k points would be at least multiplied by a factor of 2 and consequently the computer time by the same factor. When the number N_A of atoms in the elementary cell is small (less than 10), some thousands of k points in the irreducible part of the Brillouin zone can easily be used, but when N_A becomes very large (in this work we will have up to $N_A = 24$), smaller values of N_k must be used to limit the computing time to reasonable values. In order to optimize the number of k points, we study the convergence of the IMC (< 1 meV/crystallographic cell) and of the magnetic moments as a function of N_k starting from small ($N_k = 12$) to large values (up to $N_k = 816$). We assume that self-consistent solutions are obtained with a good accuracy when the variations of the energies and the magnetic moments for the two largest values of N_k are small as compared to their absolute values.

We have found that to achieve self-consistent solutions, the calculation needs more and more iterations when (i) the number of Pd atoms is increased for given values of N_k and N_A because of the Pd magnetic instability and (ii) when N_k is increased.

As discussed previously, the crystalline structure of the sandwiches and of the superlattices have been found to be very similar.[21,22] However, only the in-plane parameter is experimentally known and is found to be nearly equal to the bulk Fe one. If we assume that the (001) plane of fcc Pd when rotated by 45° matches the (001) plane of bcc Fe, 5% in-plane expansion of the Pd parameter is obtained. Consequently, the fcc cell is *a priori* tetragonally deformed giving a fct structure for the Pd layers (Fig. 1). Such a behavior has already been observed in platinum on (001) iron for which in-plane lattice expansion is accompanied by an out-of-plane lattice compression.[39]

The precise value of the tetragonality, i.e., of the interplanar distances being unknown, we will consider two extreme model structures: (i) the first one for which the atomic volume of the Pd atoms is taken to be equal to the bulk one [constant atomic volume (CAV)] and consequently induces a tetragonal deformation of the Pd fcc structure (fct) and (ii) the second one for which the structure of the Pd layers is assumed to be fcc [expanded atomic volume (EAV)]. In the first structure the distance, $d_\perp$(Pd-Pd), between Pd planes is equal to $0.625 \times a_{Fe}$, $a_{Fe} = 5.42$ atomic units (a.u.) being the experimental value of the bcc iron lattice parameter. The Wigner-Seitz radius taken for Pd and Fe are, respectively, $s_{Fe} = 2.668\,66$ a.u., and $s_{Pd} = 2.875\,49$ a.u. For the second structure, $d_\perp$(Pd-Pd) is equal to $(\sqrt{2}/2) \times a_{Fe}$ so that the palladium atomic volume increase $\Delta\Omega$ from its bulk value Ω, $\Delta\Omega/\Omega$ is equal to 13%. The Wigner-Seitz radius taken for Pd and Fe are, respectively, $s_{Fe} = 2.668\,66$ a.u., and $s_{Pd} = 2.995\,47$ a.u. We have verified that, for these two structures, the bulk Pd crystal is not magnetic. Finally, for the distance $d_\perp$(Pd-Fe) between a Pd plane and a Fe plane, we use the average $[d_\perp(\text{Pd-Pd}) + d_\perp(\text{Fe-Fe})]/2$ value with $d_\perp(\text{Fe-Fe}) = a_{Fe}/2$.

These calculations allow one to determine the sensitivity of the superlattice's magnetic properties to the structure of the palladium layers. However, although the experimental structure is thought to be intermediate between the previous ones, we cannot compare the theoreti-

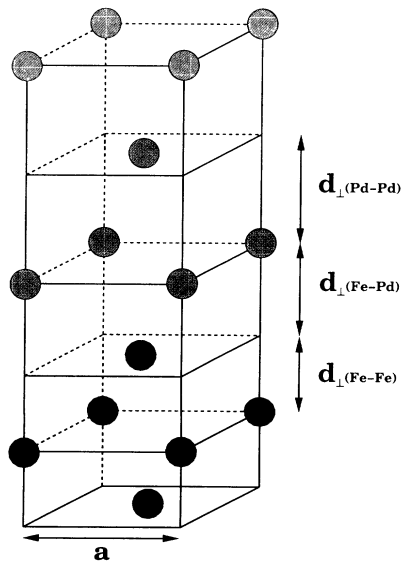

FIG. 1. Schematic representation of a Fe-Pd interface showing the interplanar distances.

cal values of the IMC with the experimental ones without doing an energy minimization with respect to the atomic displacement. Up to now, such a calculation seems to be too difficult to be performed with the required accuracy without limiting the number of degrees of freedom. For example, we can expect that the main features are obtained by varying $d_{\perp}$(Pd-Pd) assuming that it is the same in the whole Pd layer. Such a study is now under progress.

III. INTERLAYER MAGNETIC COUPLINGS IN Fe_3Pd_n SUPERLATTICES

We determine the IMC by calculating the difference $\Delta E_{\text{F-AF}} = E_{\text{F}} - E_{\text{AF}}$ between the total energies obtained for the two opposite interlayer magnetic arrangements F and AF. The F (AF) interlayer magnetic arrangement corresponds (respectively) to parallel (antiparallel) magnetizations of successive Fe layers (Fig. 2). The calculated range of the antiferromagnetic IMC can then be directly compared to the ones deduced from magnetometry measurements.

To determine the accuracy obtained for the IMC, we studied the variation of the IMC as a function of N_k. We have used $N_k = 12$, 30, 84, 114, 330 for the CAV structure [Fig. 3(a)] and the same set plus $N_k = 816$ for the EAV structure [Fig. 3(b)]. This last value of N_k is really necessary to achieve a good convergence for large thicknesses as shown in Fig. 3. From this study, the following remarks can be made:

(i) The curves obtained for $N_k = 12$ allow one to get a qualitative idea of the variation of the IMC with the number of Pd atomic layers.

(ii) The convergence of the IMC is rapidly achieved for the CAV structure, the variations of the IMC for the largest N_k values being relatively small [Fig. 3(a)]. However, it is more difficult to achieve a good convergence for the EAV structure [Fig. 3(b)] due to the fact that the Pd is closer from the ferromagnetic instability.

(iii) If we consider the variations of the IMC with N_k, we estimate that the error induced by the restricted number of k points is equal to 2 meV/crystallographic cell.

Figure 4 is a plot of the IMC versus the spacer thickness n. The IMC obtained for the CAV structure oscillate from AF ($n = 1,2$) to F ($n = 3,4,5$) and to AF ($n = 6$) values. These oscillations can be roughly assimilated to the ones obtained with the RKKY-like theories[6,9] but they cannot be quantitatively related to the Fermi-surface topology. The range of the n values is too small to determine the periodicity of such oscillations and to relate them to an asymptotic regime valid for large spacer thicknesses ($n \to \infty$). On the contrary, the IMC obtained for the EAV structure are F except for $n = 1$. The comparison between both curves shows interesting features:

(i) For $n = 1$, the IMC are positive for both CAV and EAV structures, i.e., the AF arrangement is the most stable one (Fig. 4). This result is *a priori* in contradiction

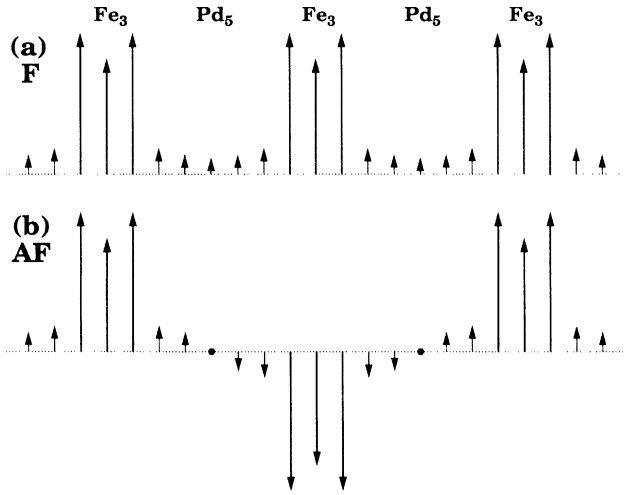

FIG. 2. Schematic representation of the magnetic-moment distribution in Fe_3Pd_5 for (a) the F and (b) The AF interlayer magnetic arrangement.

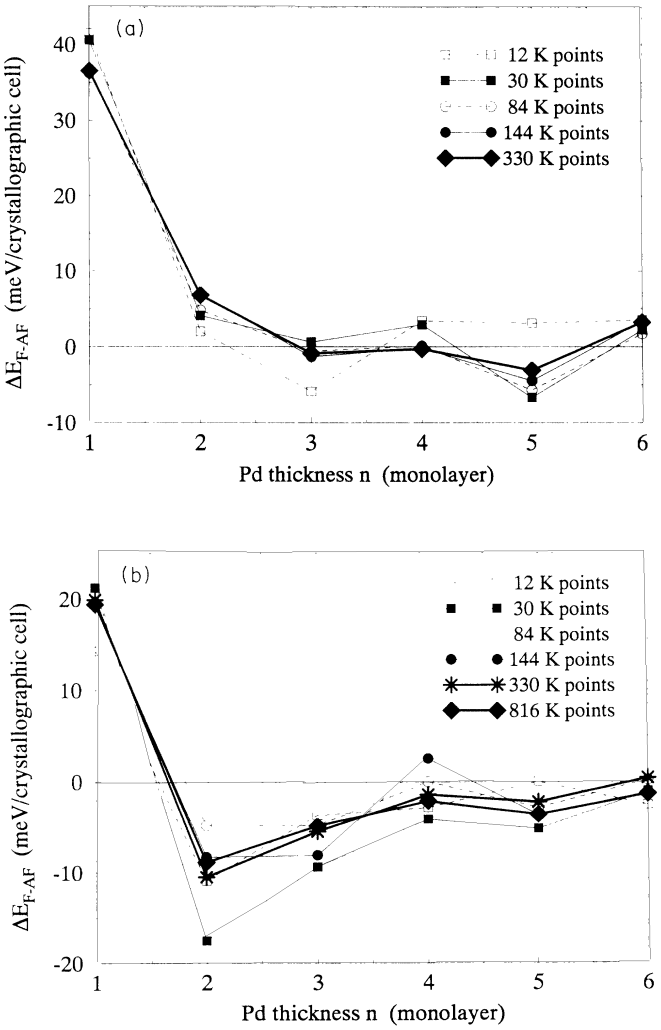

FIG. 3. Interlayer magnetic couplings $\Delta E_{\text{F-AF}} = E_{\text{F}} - E_{\text{AF}}$ in Fe_3Pd_n superlattices as a function of n for (a) the CAV and (b) the EAV structure showing the variation of the behavior with the number N_k of k points used for the calculation.

with the idea that the ground state is obtained for palladium layers coupled ferromagnetically to their Fe neighboring layers, i.e., for a F arrangement. Due to the iron polarization, the Pd moments are expected to be large in the F arrangement. Indeed, they are found to be equal to $0.4\mu_B$ and are a small bit larger for the CAV structure than for the EAV one [Figs. 5(a), and Table I]. On the contrary, in the AF arrangement, which is the ground state, the magnetic moment of the atoms in the single Pd layer is equal to zero from symmetry requirements [Figs. 5(a), 5(b), and Table II]. This suggests that, for the F arrangement, the polarization of the Pd spacer costs some magnetic-moment formation energy which can be larger than the energy gain coming from the ferromagnetic interfacial coupling between Fe and Pd layers, these two contributions vanishing for the AF arrangement because the Pd layer is nonmagnetic. This conjecture is consistent with the fact that the lowest antiferromagnetic IMC is obtained for the EAV structure, i.e., for large Fe-Pd distances so that a given polarization of the Pd atoms costs less energy, the Pd atoms being closer from the Stoner instability.

(ii) For $n=2$, the magnetic moment distributions of the CAV and EAV structures for each magnetic configuration are very similar too (Tables I and II) but for this thickness the magnetic moment is slightly larger for the EAV structure. In the CAV structure the IMC remains AF but its magnitude is strongly reduced (by a factor of 5) with respect to its value for $n=1$: This result can be correlated with the fact that the differences in the polarization of the Pd spacer are less important than for $n=1$, the magnetic moments in the F and AF cases being nearly equal. In the EAV structure, the IMC is nearly opposite to the ones obtained for the CAV structure. The contribution due to polarization of the Pd layers

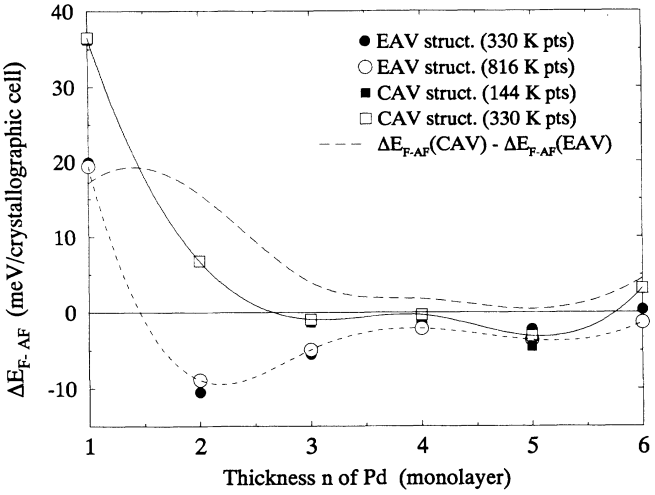

FIG. 4. Interlayer magnetic couplings $\Delta E_{\text{F-AF}}=E_{\text{F}}-E_{\text{AF}}$ in Fe_3Pd_n superlattices as a function of n for the CAV (full line with square symbols) and for the EAV (dashed line with circle symbols) structure obtained with the two largest N_k. The long dashed line corresponds to the difference $\Delta E_{\text{F-AF}}$ (CAV) $-\Delta E_{\text{F-AF}}$ (EAV).

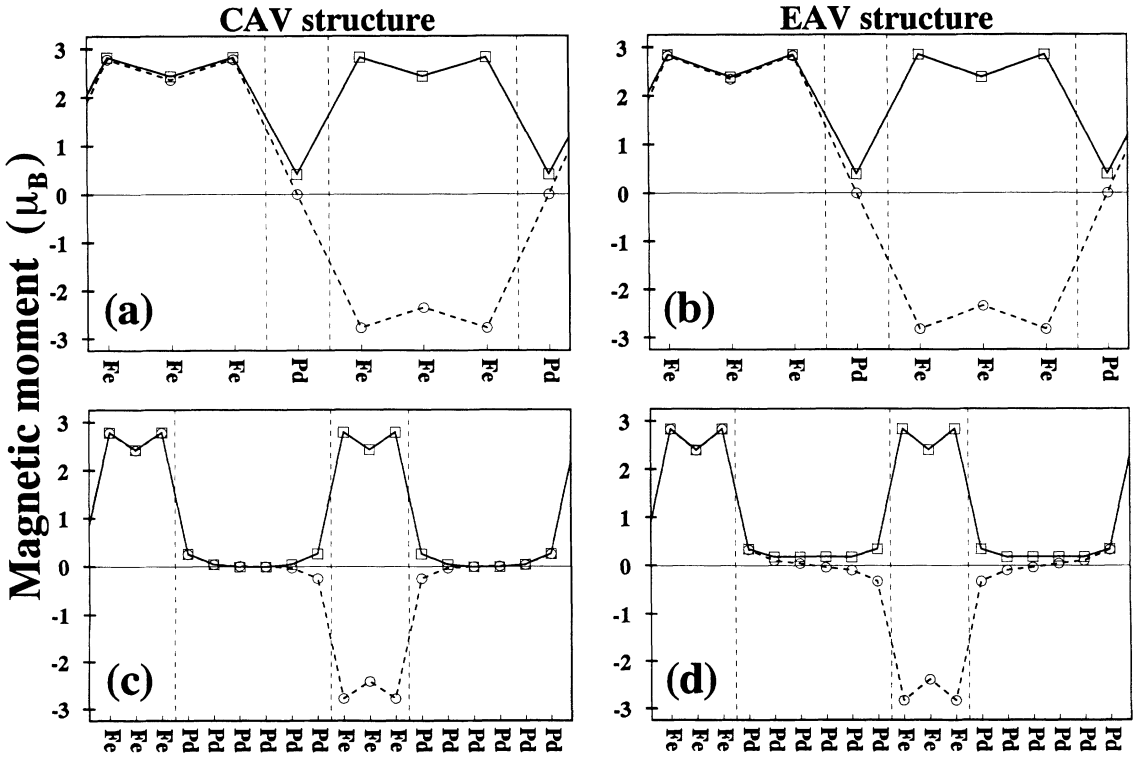

FIG. 5. Magnetic-moment distributions in (a) Fe_3Pd_1 for the CAV structure, (b) Fe_3Pd_1 for the EAV structure, (c) Fe_3Pd_6 for the CAV structure, and (d) Fe_3Pd_6 for the EAV structure.

TABLE I. Values of the magnetic moments on all inequivalents sites in Fe_mPd_n superlattices for $n=1-14$ and for the F interlayer magnetic arrangement. C, I and $I+k$ correspond, respectively, to an atom in the inner Fe layer, in the interfacial layers, and in the kth Pd layer from the interface. For large Pd thicknesses ($n>6$), we obtain an artificial dependence of the Pd magnetic moments with the parity of n, the number of Fe atomic layers being successively equal to $m=3$ or 4. However, these slight differences do not affect the general features of the magnetic moments distributions.

CAV struct.	$M_{Fe}(C)$	$M_{Fe}(I)$	$M_{Pd}(I)$	$M_{Pd}(I+1)$	$M_{Pd}(I+2)$	$M_{Pd}(I+3)$	$M_{Pd}(I+4)$	$M_{Pd}(I+5)$	$M_{Pd}(I+6)$
$m=3,n=1$	2.429	2.819	0.413						
$m=3,n=2$	2.443	2.806	0.307						
$m=3,n=3$	2.442	2.796	0.289	0.129					
$m=3,n=4$	2.428	2.785	0.256	0.038					
$m=3,n=5$	2.426	2.782	0.253	0.013	−0.025				
$m=3,n=6$	2.424	2.786	0.262	0.040	−0.010				
$m=3,n=7$	2.428	2.789	0.267	0.059	0.021	0.006			
$m=4,n=8$	2.469	2.794	0.279	0.064	0.024	0.017			
$m=3,n=9$	2.421	2.784	0.262	0.048	0.010	0.006	0.013		
$m=4,n=10$	2.465	2.790	0.276	0.052	0.009	0.002	0.014		
$m=3,n=11$	2.423	2.784	0.263	0.049	0.008	0.001	0.008	0.010	
$m=4,n=12$	2.466	2.792	0.277	0.057	0.015	0.006	0.013	0.013	
$m=3,n=13$	2.421	2.783	0.262	0.048	0.007	0.001	0.009	0.006	0.003
$m=4,n=14$	2.467	2.791	0.277	0.061	0.020	0.010	0.015	0.011	0.009
EAV struct.									
$m=3,n=1$	2.383	2.849	0.397						
$m=3,n=2$	2.401	2.821	0.322						
$m=3,n=3$	2.414	2.841	0.339	0.171					
$m=3,n=4$	2.409	2.843	0.346	0.184					
$m=3,n=5$	2.405	2.839	0.340	0.171	0.179				
$m=3,n=6$	2.403	2.839	0.342	0.177	0.179				
$m=3,n=7$	2.402	2.839	0.341	0.160	0.165	0.158			
$m=4,n=8$	2.471	2.842	0.353	0.175	0.181	0.190			
$m=3,n=9$	2.401	2.839	0.344	0.156	0.151	0.145	0.153		
$m=4,n=10$	2.468	2.841	0.352	0.172	0.167	0.155	0.149		
$m=3,n=11$	2.399	2.838	0.340	0.158	0.155	0.150	0.150	0.144	
$m=4,n=12$	2.468	2.840	0.350	0.169	0.163	0.153	0.146	0.133	
$m=3,n=13$	2.398	2.837	0.341	0.158	0.156	0.149	0.149	0.139	0.136
$m=4,n=14$	2.466	2.841	0.350	0.177	0.180	0.180	0.181	0.172	0.171

favors the F state due to the fact that it costs less energy in an expanded structure to create a magnetic moment on a given atom.

(iii) For $n>2$, the IMC become small (a few meV/crystallographic cell). In the EAV structure, the IMC are always F due to the ferromagnetic polarization of the whole Pd layer [Figs. 5(c), 5(d), Tables I and II]. On the contrary, in the CAV structure the IMC oscillate and change of sign, the polarization being limited to the first Pd layer: The interfacial magnetic moment is equal to $0.26\mu_B$ (Tables I and II) whereas the moment is much smaller ($\approx 0.04\mu_B$) on the second layer.

TABLE II. Values of the magnetic moments on all inequivalents sites in Fe_3Pd_n superlattices for $n=1-6$ and for the AF interlayer magnetic arrangement.

CAV struct.	$M_{Fe}(C)$	$M_{Fe}(I)$	$M_{Pd}(I)$	$M_{Pd}(I+1)$	$M_{Pd}(I+2)$
$n=1$	2.356	2.779	0.000		
$n=2$	2.430	2.785	0.223		
$n=3$	2.430	2.790	0.258	0.000	
$n=4$	2.422	2.777	0.261	0.041	
$n=5$	2.414	2.774	0.253	0.046	0.000
$n=6$	2.413	2.776	0.254	0.036	0.005
EAV struct.					
$n=1$	2.341	2.822	0.000		
$n=2$	2.380	2.813	0.245		
$n=3$	2.395	2.832	0.295	0.000	
$n=4$	2.395	2.834	0.324	0.055	
$n=5$	2.391	2.830	0.322	0.091	0.000
$n=6$	2.392	2.831	0.326	0.098	0.040

(iv) The difference $\Delta E_{\text{F-AF}}(\text{CAV}) - \Delta E_{\text{F-AF}}(\text{EAV})$ between the IMC obtained for the two structures is always positive (Fig. 4). The interplanar Pd expansion introduces then a contribution which stabilizes always the F state. However, from Fig. 4, we deduce that this effect is short ranged (three monolayers).

The comparison with the experimental results of Celinski and co-workers[21,22] can be summarized as follows:

(i) The mean magnetic moment per Pd atom is estimated by the authors to be equal to $0.25\mu_B$ in $Fe_5/Pd_4/Fe_{10}$.[21] This result is in agreement with the one we obtain for the Fe_3Pd_4 superlattices in the EAV structure. The calculated mean magnetic moment is found equal to $0.265\mu_B$. This suggests that the experimental structure is expanded for such small thicknesses.

(ii) The authors have also deduced that the maximum number of Pd atomic layers for which the whole Pd layers is ferromagnetically ordered is $n=4$.[21] This is nonconsistent with our calculations: we have shown that the Pd polarization for the CAV structure is limited to the interfacial atomic layer, while the Pd spacer is entirely polarized up to $n=14$ for the EAV structure. However, the present results are consistent with experiment if we assume that a structural change (from the EAV to the CAV structure) occurs for $n=5$. Such a structural transition—which has not been observed—for which the Pd recovers its equilibrium volume, occurs usually in this range of thicknesses as shown in several epitaxial systems.[39–42]

IV. ELECTRONIC STRUCTURE OF Fe_3Pd_1: INTERFACIAL HYBRIDIZATION AND LOCAL DENSITIES OF STATES

In this section, we focus our attention on the hybridization between the interfacial Fe and Pd atoms. Figures 6 and 7 represent the local densities of states (LDOS) for all the inequivalent sites in Fe_3Pd_1 superlattices for the two interlayer magnetic arrangements and for both the CAV (Fig. 6) and the EAV (Fig. 7) structures. We compare these results to the LDOS of (i) the bulk pure metals [Fig. 8(a), (b), and (c)], respectively, bcc Fe, fct Pd (CAV structure) and fcc Pd (EAV structure), and (ii) the Pd atoms which are the furthest from the Fe layers in Fe_3Pd_5 superlattices [(Figs. 8(d) and 8(e)] in the AF interlayer arrangement in order to study the influence of the interfacial perturbation on the inner Pd layers.

Let us discuss separately the main features of the majority and the minority spin bands for the inequivalent atoms in the Fe_3Pd_1 superlattices, i.e., for (i) the inner iron atoms, (ii) the interfacial Fe atoms, and (iii) the palladium atoms.

Inner Fe atoms

The majority spin LDOS resemble a typical bcc DOS [Fig. 8(a)] in all four cases—CAV and EAV structures, respectively, in the F [Figs. 6(a) and 7(a)] and AF [Figs. 6(b) and 7(b)] orderings—but the Fermi level falls above the "d" bands. This is related to (i) the increase of the magnetic moment (from $2.2\mu_B$ to $2.4\mu_B$) which shifts the majority band to lower energies and (ii) to the small charge transfer equal to $+0.07(+0.1)$ electron in the CAV (EAV) structure. The minority-spin LDOS are strongly perturbed as compared to the bulk ones [Fig. 8(a)]. They present different features according to the two magnetic arrangement. For the F state, the Fermi level falls in a peak in the LDOS whereas for the AF state it falls nearly in a minimum of the LDOS although the magnetic moments are found to be nearly equal.

Interfacial Fe atoms

The LDOS are similar to those of the inner Fe atoms but both spin bands are now strongly modified by the Fe-Pd hybridization [Figs. 6(c), 6(d), 7(c), and 7(d)]. The majority spin "d" bands are filled as for the inner Fe

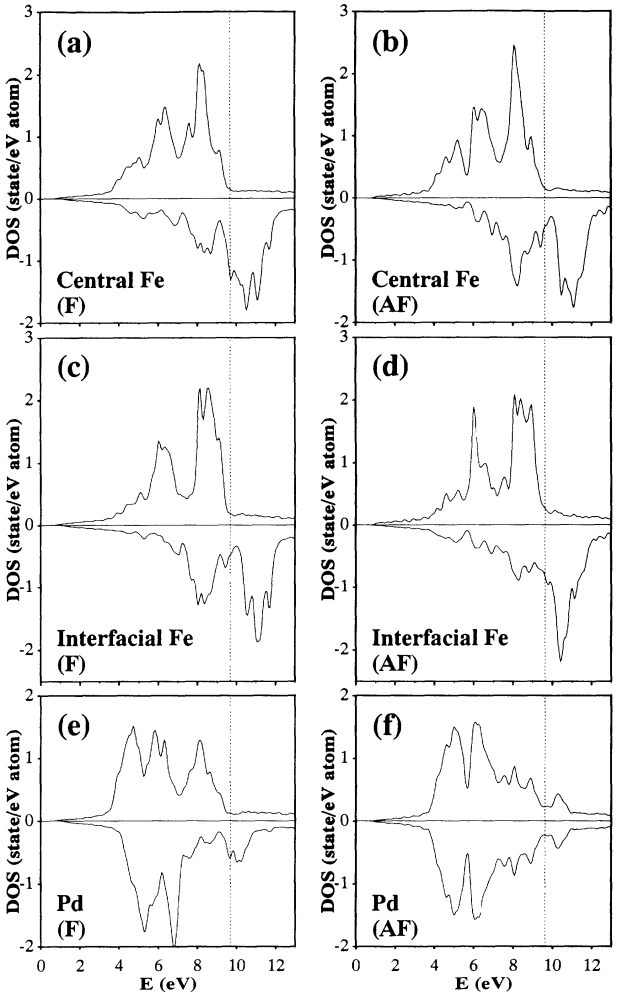

FIG. 6. Local densities of states on all inequivalent atoms in Fe_3Pd_1 superlattices for the CAV structure: (a) inner Fe layer, (c) interfacial Fe layer, (e) Pd layer in the F interlayer magnetic arrangement and (b) inner Fe layer, (d) interfacial Fe layer, (f) Pd layer in the AF interlayer magnetic arrangement. The majority (minority) LDOS $n^{\uparrow}(\varepsilon)[n^{\downarrow}(\varepsilon)]$ correspond (respectively) to the positive (negative) values. The Fermi level is given by the vertical dashed line. The curves have been smoothed by convolution with a Gaussian whose width is taken equal to 0.1 eV.

atoms, the corresponding magnetic moment being equal to $2.8\mu_B$. The minority-spin bands are slightly shifted to higher energies than the corresponding LDOS for the inner atoms, the charge transfer corresponding to a loss equal to 0.12 (0.21) electrons in the CAV (EAV) structure. For this site, the differences between the F and AF states are much more pronounced in the minority-spin bands than in the majority ones: In the F state, there is a valley in the minority-spin LDOS around the Fermi level whereas, in the AF state, the minority-spin LDOS is less structured below the Fermi level.

These results suggest that the hybridization of the "*d*" bands at the interfaces occurs up to the second Fe layer. We have found that, in the Fe_5Pd_1 superlattice, the LDOS of the inner Fe layer have almost recovered the bulk characteristics.[43] In conclusion, the electronic structure of iron and more especially of the inner Fe atoms in Fe_3Pd_1 superlattices is very different from the bulk Fe. As a consequence, the Fe becomes a strong ferromagnet for which the majority-spin "*d*" bands are saturated. The IMC in Fe_mPd_1 superlattices are then strongly dependent on the iron thickness m for $m \leq 3$.

Pd atoms

The palladium LDOS are strongly modified by the hybridization with Fe interfacial atoms in both CAV and EAV structures [Figs. 6(e), 6(f), 7(e), and 7(f)]. However, this hybridization is much larger in the majority- than in the minority-spin bands, the differences between the Pd and Fe "*d*" energy levels being much smaller in the majority- than in the minority-spin bands. This is shown, for example, in Figs. 6 and 7, by the following features: (i) in the energy domain (4–7 eV) corresponding to large peaks in the bottom of the Pd minority-spin "*d*" LDOS, the Fe minority-spin LDOS are very small, (ii) on the

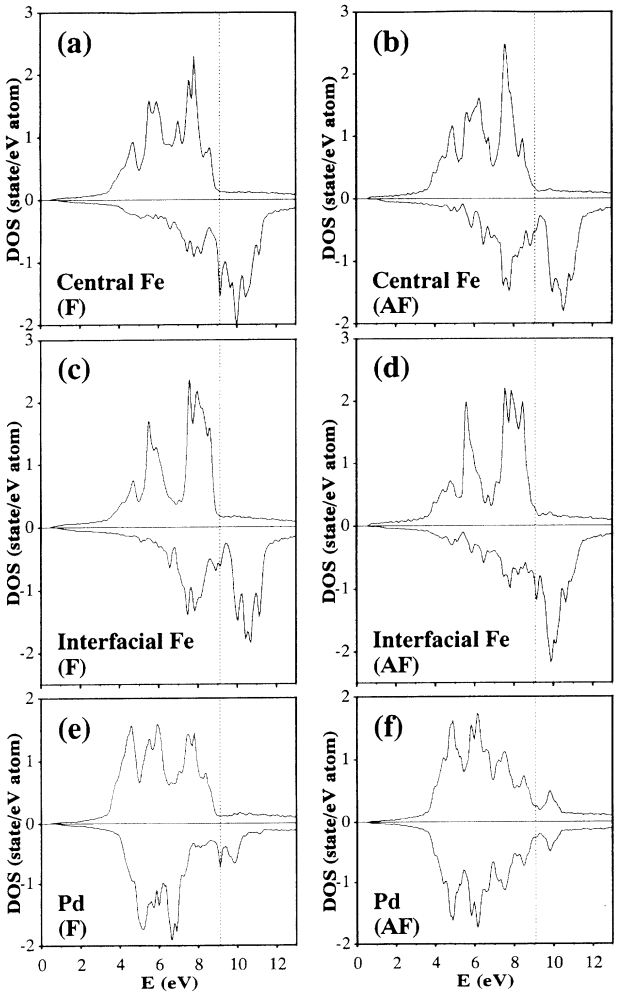

FIG. 7. Local densities of states on all inequivalent atoms in Fe_3Pd_1 superlattices for the EAV structure with the same plot disposition as that in Fig. 6. The curves have been smoothed by convolution with a Gaussian whose width is taken equal to 0.07 eV because of the larger number of k points than in the CAV structure, for which the LDOS are more structured.

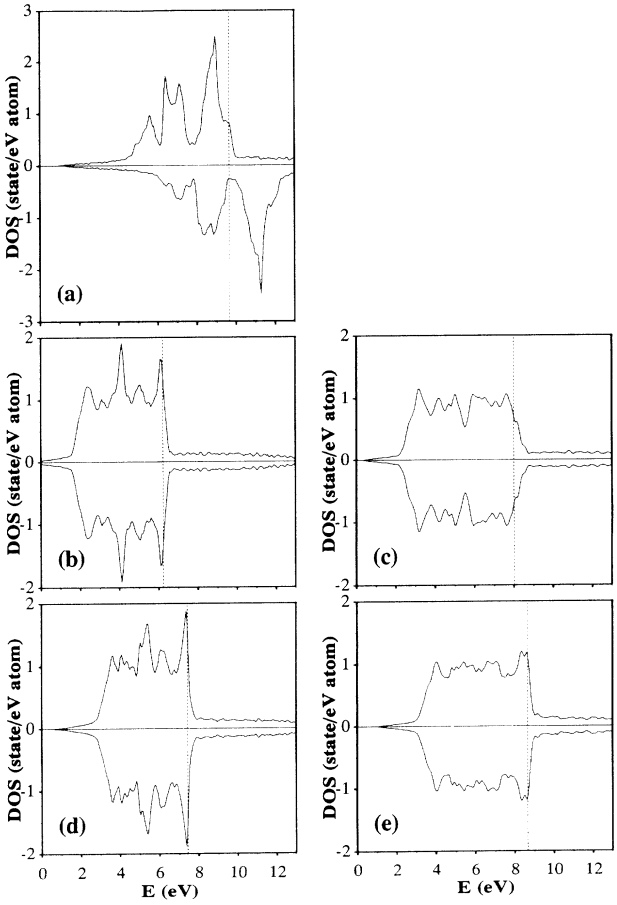

FIG. 8. Local densities of states of (a) a bulk Fe atom, (b) a bulk Pd atom in the EAV structure, (c) a bulk Pd atom in the CAV structure, (d) a central Pd atom in Fe_3Pd_5 superlattices in the EAV structure for the AF interlayer magnetic arrangement, and (e) a central Pd atom in Fe_3Pd_5 superlattices in the CAV structure for the AF interlayer magnetic arrangement. The majority (minority) LDOS $n^{\uparrow}(\varepsilon)$ $[n^{\downarrow}(\varepsilon)]$ correspond (respectively) to the positive (negative) values. The Fermi level is given by the vertical dashed line. The curves have been smoothed by convolution with a Gaussian whose width is taken equal to 0.07 eV.

contrary, the energy ranges (7.5–10 eV) for which the Fe minority-spin LDOS are large corresponding to small Pd LDOS. This small Fe-Pd hybridization in the minority-spin bands is illustrated by the presence of a new structure in the Pd LDOS above the Fermi level (9–11 eV), which corresponds to maxima in the Fe LDOS. This structure in the Pd LDOS is characteristic of the hybridization with the Fe minority-spin states. In consequence, it is obtained only in the Pd minority-spin LDOS for the F state, while this structure exists in both majority- and minority-spin LDOS in the AF state.

For Fe_3Pd_n superlattices and for larger Pd spacer thicknesses, the features we described previously remain valid for the Fe layers and the Pd interfacial sites. However, the structure above the Fermi level in the Pd interfacial LDOS is less pronounced than in Fe_3Pd_1 superlattices, each interfacial Pd atom having four Fe neighbors only as compared to eight in Fe_3Pd_1 superlattices. Because interfacial Pd sites have Fe on just one side, this new structure is revealed only in one-spin LDOS (minority or majority) due to symmetry considerations. For the other Pd sites, the LDOS are similar to the bulk one as shown in Figs. 8(d) and 8(e) suggesting that the hybridization is limited to the first Pd layer.

V. POLARIZATION OF THE Pd SPACER

The experimental ferromagnetic IMC as measured by Celinski and Heinrich[22] show a nonmonotonic behavior with n. The authors have obtained local minima for $n=6,9$ and local maxima for $n=7,11$. Unfortunately, it is not possible to determine theoretically the IMC for such large Pd thicknesses because they are certainly too small to be accurately determined and would need a too large computer time. However, it is possible to determine the magnetic-moment distributions for larger n values; since these distributions require less computer time, this study allows one to obtain information on the magnetic Pd polarization. Therefore, we have determined the polarization in the Pd spacer for large thicknesses (up to $n=14$). For such calculations, we need to use an elementary cell as small as possible. Since the AF magnetic arrangement requires double crystallographic cells, we restrict our study to the F arrangement. Moreover, we consider only superlattices with an even number of atomic planes in the elementary cell to reduce by symmetry the size of the cell by a factor of 2. This leads us to study Fe_3Pd_n superlattices for odd n values and F_4Pd_n superlattices for even n values. The magnetic moments being less sensitive to the number of k points used, we have only calculated the self-consistent solutions with $N_k=12$, 30, 63, 108 for the CAV structure and with the same set plus $N_k=312$ for the EAV structure.

In Fig. 9 we have plotted the magnetic moment distributions in the Pd spacer for thicknesses $n=11-14$ monolayers for both structures and for a F interlayer magnetic arrangement. All magnetic moments for $n=1$ to 14 are also given in Table I. The magnetic moment distributions for $n=1,\ldots,6$ have been determined in the double cell with 330 (816) k points (see Sec. III) and the others ($n=7-14$) have been determined in the single crystallographic cell with 108 (312) k points in the CAV (EAV)

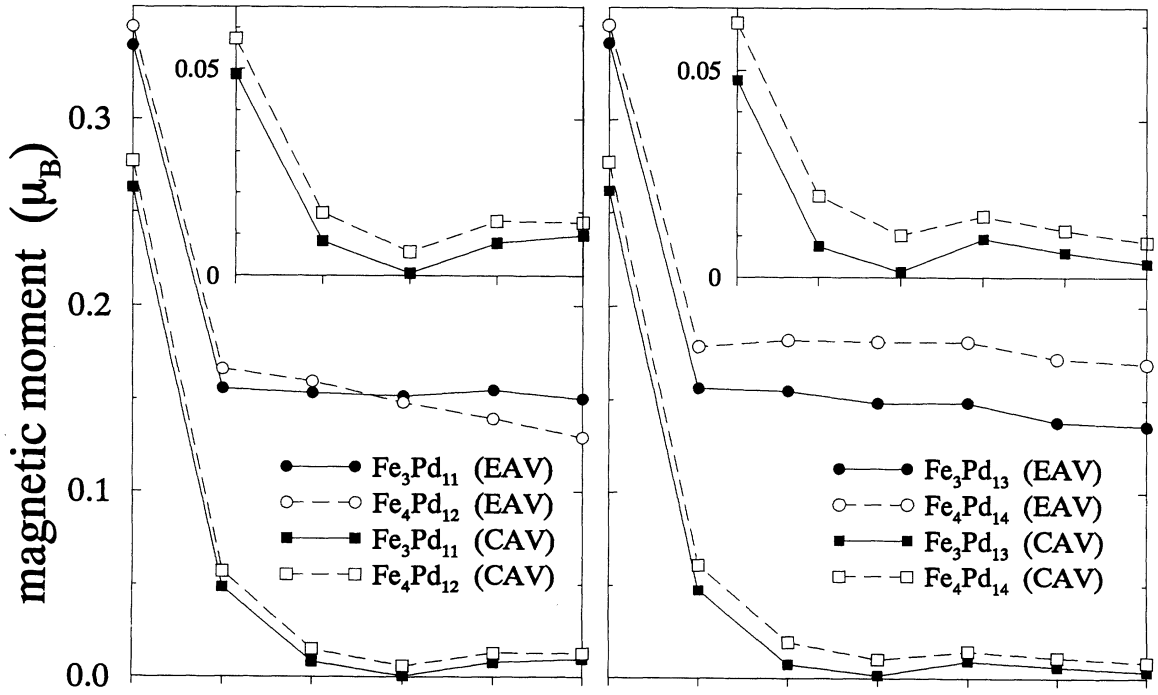

FIG. 9. Polarization of the Pd spacer in Fe_3Pd_{11} and Fe_3Pd_{12} superlattices (left) and in Fe_3Pd_{13} and Fe_3Pd_{14} superlattices (right). The values are only given for half of the sites. The others can be deduced by symmetry. The inserts represent the same values for the CAV structure at a larger scale.

structure as explained above. Let us now comment separately on the results we obtained for the CAV and EAV structures.

CAV structure

Our results show that we are far from reaching the asymptotic regime for small Pd thicknesses ($n \leq 6$). Most striking is that we get negative Pd magnetic moments in the center of the Pd layer for $n = 5,6$ which suggest that larger Pd thicknesses are needed to describe the polarization decay.

The magnetic polarization of the palladium layers by the Fe interfacial layers is limited to the first three neighboring layers, the total induced magnetic moment being equal to about $0.35\mu_B$. If both interfaces are independent, we can expect an exponential decay of the induced Pd polarization.[44] However, even for $n = 14$ (Fig. 9), the magnetic polarizations induced in the Pd layer by each of the two interfaces with Fe show a nonmonotonous decay and suggest that both polarizations are interacting. More especially, a minimum is always obtained in the magnitude of the magnetic moments on the fourth Pd layers from the interface.

EAV structure

In the EAV structure, the whole Pd spacer remains polarized with large magnetic moments on the inner layers ($0.13-0.19\mu_B$) up to large thicknesses (14 monolayers). This is due to the fact that, for this atomic volume, the Pd is close to the ferromagnetic state. This explains why the interfacial polarization gives rise to long-range and slowly decreasing tails whose superposition leads to the polarization we obtain. No periodic modulation of the magnitude of the magnetic moments is observed in contrast with the case of the CAV structure. However, if we consider the central magnetic moment versus the spacer thickness, we see that it varies nonmonotonously and presents a minimum for $n = 12$. This can be considered as a possible origin of the oscillations in the F couplings obtained by Celinski and Heinrich.[22] Calculations for larger Pd thicknesses are needed to confirm this result and to determine the period of these oscillations.

VI. CONCLUSION

In this paper we have studied the Pd polarization and the IMC in $Fe_3Pd_n(001)$ superlattices. The electronic structure of the superlattices has been determined with the LSDA formalism and the ASW method to treat accurately the polarization of the Pd spacer. In order to study the role of tetragonal deformations of the Pd layers, we have considered two different extreme structures (CAV and EAV) for the Pd spacer. Several features have been clearly identified:

(i) For large n values ($n > 4$), we have shown that the polarization of the Pd spacer is completely different for the two structures: It is limited to the interfacial atoms for the CAV structure whereas the whole Pd spacer is polarized with a magnetic moment of approximately $0.15\mu_B$/atom for the inner layers in the EAV structure. Consequently, the IMC are found to oscillate with a RKKY-like behavior in the CAV structure whereas they are F in the EAV structure.

(ii) The palladium thicknesses for which we have determined the IMC being too small, the asymptotic regime characteristic of large n values is far from being obtained so that it is not possible to directly compare the periods of the IMC and of the polarization. However, for the CAV structure, we have obtained a single modulation of the magnitude of the Pd magnetic moments with a period of four monolayers, a result which we can tentatively relate to the observed experimental period.[22]

(iii) For $n = 1,2$ we have shown that the electronic structure of the palladium atoms is strongly modified by the Fe-Pd hybridization whereas for $n > 2$ the inner Pd layers have recovered their bulk characteristics. The ground state of Fe_3Pd_1 in both structures and for Fe_3Pd_2 in the CAV structure is antiferromagnetic.

It has been shown recently that the polarization induced in Pd crystals by an interstitial Fe monolayer decays exponentially with the distance to the interface.[44] We have not observed such an exponential behavior for the two structures we considered. Moreover, we have obtained nonmonotonous magnetic-moment distributions as a function of the Pd position with respect to the interface for large n. Therefore, these results show that the polarization of a Pd slab surrounded by Fe layers in a superlattice does not behave as the one resulting from a simple superposition of the polarizations induced by each interface. Up to now, we do not know the respective roles of the superlattice periodicity and of the tetragonality of the Pd structure. However, the comparison between the IMC obtained for the two structures (CAV and EAV) and the experimental data[21,22] suggests that the c/a ratio varies with the Pd thickness. Such a variation has never been measured experimentally in Fe/Pd superlattices. This shows the evidence to perform very specific experiments, such as surface-extended x-ray-absorption fine structure, for samples having varying Pd thicknesses.

In a forthcoming paper, we develop these studies (i) by increasing the thickness of the Pd spacer in the CAV structure to determine the periodicity of the magnetic moment distribution, (ii) by introducing interfacial ordered compounds to study the effect of interdiffusion, and (iii) by varying the structure of the Pd layer for some representative cases.

ACKNOWLEDGMENT

The Institut de Physique et de Chimie des Matériaux de Strasbourg is Unité Mixte de Recherche du CNRS-ULP-EHICS, No. 46.

*Present address: BIOSYM Technologies, Inc., 9685 Scranton Rd., San Diego, CA 92121.

[1] S. S. P. Parkin, N. More, and K. P. More, Phys. Rev. Lett. **64**, 2304 (1990).

[2] M. N. Baibich, J. M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Etienne, G. Creuzet, A. Friederich, and J. Chazelas, Phys. Rev. Lett. **61**, 2472 (1988).

[3] J. Unguris, R. J. Cellotta, and D. T. Pierce, Phys. Rev. Lett.

67, 140 (1991).
[4]C. Chappert and J. P. Renard, Europhys. Lett. **15**, 553 (1991).
[5]P. Bruno and C. Chappert, Phys. Rev. Lett. **67**, 1602 (1991).
[6]P. Bruno and C. Chappert, Phys. Rev. B **46**, 261 (1992).
[7]B. Caroli, J. Phys. Chem. Solids **28**, 1427 (1967).
[8]A. Blandin, J. Friedel, and G. Saada, J. Phys. **27-C3**, 128 (1966).
[9]D. M. Edwards, J. Mathon, R. B. Muniz, and M. S. Phan, Phys. Rev. Lett. **67**, 493 (1991).
[10]P. Bruno, J. Magn. Magn. Mater. **116**, L13 (1992).
[11]M. T. Johnson, S. T. Purcell, N. W. E. McGee, R. Coehoorn, J. aan de Stegge, and W. Hoving, Phys. Rev. Lett. **68**, 2688 (1992).
[12]S. S. P. Parkin, Phys. Rev. Lett. **67**, 3598 (1991).
[13]F. Herman, J. Sticht, and M. van Schilfgaarde, J. Appl. Phys. **69**, 4786 (1991).
[14]K. Ounadjela, C. B. Sommers, A. Fert, D. Stoeffler, F. Gautier, and V. L. Moruzzi, Europhys. Lett. **15**, 875 (1991).
[15]F. Herman, J. Sticht, and N. van Schilfgaarde, in *Magnetic Thin Films, Multilayers and Surfaces,* edited by S. S. P. Parkin, MRS Symposia Proceedings No. 231 (Materials Research Society, Pittsburgh, 1991).
[16]D. Stoeffler and F. Gautier, Prog. Theor. Phys. Suppl. **101**, 139 (1990); Phys. Rev. B **44**, 10 389 (1991).
[17]D. Stoeffler and F. Gautier (unpublished).
[18]D. Stoeffler and F. Gautier, J. Magn. Magn. Mater. **121**, 259 (1993); in *Magnetism and Structure in Systems of Reduced Dimension,* Proceedings of the NATO Advanced Research Workshop, Corsica, 1992, edited by B. Dieny and M. Donath (Plenum, New York, 1993).
[19]T. Moriya, in *Theory of Magnetism in Transition Metals,* Proceedings of the International School of Physics "Enrico Fermi," Course XXXVII, Varenna, 1966, edited by W. Marshall (Academic, New York and London, 1967).
[20]F. J. A. M. Greidanus, W. B. Zeper, J. J. A. den Broeder, W. F. Godlieb, and P. F. Garcia, Appl. Phys. Lett. **54**, 2481 (1989).
[21]Z. Celinski, B. Heinrich, J. F. Cochran, W. B. Muir, A. S. Arrott, and J. Kirshner, Phys. Rev. Lett. **65**, 1156 (1900).
[22]Z. Celinski and B. Heinrich, J. Magn. Magn. Mater. **99**, L25 (1991).
[23]A. Schuhl, J. R. Childress, J.-M. George, P. Galtier, O. Durand, A. Barthelemy, and A. Fert, J. Magn. Magn. Mater. **121**, 275 (1993).
[24]J. R. Childress, A. Schuhl, J.-M. George, O. Durand, P. Galtier, V. Cros, K. Ounadjela, R. Kergoat, and A. Fert, in *Magnetism and Structure in Systems of Reduced Dimension* (Ref. 18).
[25]J. R. Childress, R. Kergoat, O. Durand, J.-M. George, P. Galtier, L. Miltat, and A. Schuhl (unpublished).
[26]J. R. Childress, O. Durand, J.-M. George, and A. Schuhl (unpublished).
[27]A. Boufelfel, R. M. Emrick, and C. M. Falco, Phys. Rev. B **43**, 13 152 (1991).
[28]W. Weber, D. A. Wesner, G. Güntherodt, and U. Linke, Phys. Rev. Lett. **66**, 942 (1991).
[29]H. Huang, J. Hermanson, J. G. Gay, R. Richter, and J. R. Smith, Surf. Sci. **172**, 363 (1986).
[30]S. Blügel, B. Drittler, R. Zeller, and P. H. Dederichs, Appl. Phys. A **49**, 547 (1989).
[31]C. Li, A. J. Freeman, H. J. Hansen, and C. L. Fu, Phys. Rev. B **42**, 5433 (1990).
[32]H. Nait-Laziz, S. Bouarab, C. Demangeat, A. Mokrani, and H. Dreyssé, J. Magn. Magn. Mater. **118**, 365 (1993).
[33]H. Nait-Laziz, C. Demangeat, S. Ouannasser, and H. Dreyssé, J. Magn. Magn. Mater. (to be published).
[34]A. R. Williams, J. Küber, and C. D. Gelatt, Jr., Phys. Rev. B **19**, 6094 (1979).
[35]S. T. Purcell, M. T. Johnson, N. W. E. McGee, R. Coehorn, and W. Hoving, Phys. Rev. B **45**, 13 064 (1992).
[36]F. Herman, M. van Schilfgaarde, and J. Sticht, in *Physics of Transition Metals,* Proceedings of the International Conference on the Physics of Transition Metals, Darmstadt, 1992, edited by P.M. Oppeneer and J. Kübler (World Scientific, Singapore, 1993).
[37]J. Sticht (unpublished).
[38]J. Sticht, F. Herman, and J. Kübler, in *Physics of Transition Metals* (Ref. 36).
[39]G. W. R. Leibbrandt, R. van Wijk, and F. H. P. Habraken, Phys. Rev. B **47**, 6630 (1993).
[40]K. Ounadjela, P. Vennegues, Y. Henry, A. Michel, V. Pierron-Bohnes, and J. Arabski (unpublished).
[41]D. Muller, K. Ounadjela, P. Vennegues, V. Pierron-Bohnes, A. Arbaoui, J. P. Jay, A. Dinia, and P. Panissod, J. Magn. Magn. Mater. **104**, 1873 (1992).
[42]S. Pizzini, F. Baudelet, A. Fontaine, M. Galtier, D. Renard, and C. Marlière, Phys. Rev. B **47**, 8754 (1993).
[43]D. Stoeffler, K. Ounadjela, J. Sticht, and F. Gautier (unpublished).
[44]P. Lang, R. Zeller, and P. H. Dederichs (unpublished).

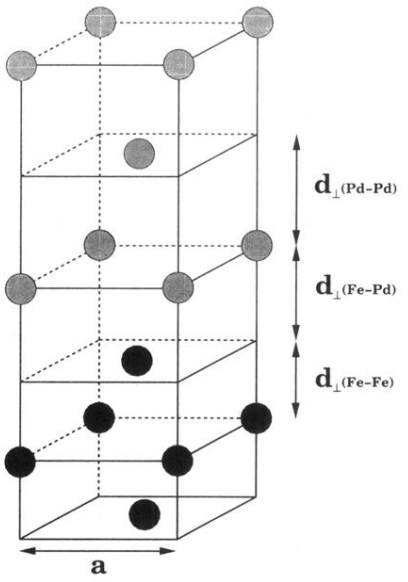

FIG. 1. Schematic representation of a Fe-Pd interface showing the interplanar distances.