Orbital and dipolar contributions to the hyperfine fields in bulk bcc Fe, hcp Co, and at the Fe/Ag(100) interface: The inclusion of orbital polarization

C. O. Rodriguez,[1] M. V. Ganduglia-Pirovano,[2] E. L. Peltzer y Blancá,[1] M. Petersen,[2] and P. Novák[3]
[1]*IFLYSIB, Grupo de Física del Sólido, C.C. 565, La Plata 1900, Argentina*
[2]*Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, D-14195, Berlin, Germany*
[3]*Institute of Physics, Academy of Sciences of the Czech Republic, Cukrovanická 10, 16253 Praha 6, Czech Republic*
(Received 3 October 2000; published 18 April 2001)

First-principles calculations can make quantitative predictions of many properties of solids including magnetic hyperfine fields (B_{hf}). The inclusion of relativistic effects becomes important in this case. We have selected to study three systems in order to assess the importance of orbital and dipolar contributions to the B_{hf} and its anisotropies: bulk bcc Fe, hcp Co, and the Fe/Ag(100) interface. For the last two, in-plane (parallel to the hexagonal planes and to the interface, respectively) and perpendicular magnetizations were considered. The influence of different exchange-correlation potentials (local density and generalized gradient approximation) and the inclusion of the orbital polarization term (known to improve the evaluation of orbital moments) in the Hamiltonian is reported. A comparison is made with other theoretical studies and to experiment when possible.

DOI: 10.1103/PhysRevB.63.184413 PACS number(s): 75.70.Cn, 76.80.+y

I. INTRODUCTION

Electrons in a solid produce a magnetic field at the site of the atomic nucleus: the magnetic hyperfine field of an atom or ion, B_{hf}. For an observer at the nucleus, the hyperfine interaction is caused by the interaction between this magnetic field produced by the electron spin and orbital currents and the magnetic moment of the nucleus. Different nuclear methods [Mössbauer-effect spectroscopy (MES) and nuclear magnetic resonance (NMR)] can be used to measure it.

Magnetic hyperfine fields (HFF's) can give information about the electronic structure and magnetic properties of a solid as they probe the electron spin density close to the nuclei.[1] Of interest is the relation of the local magnetic moment on the atom and the hyperfine field. Hyperfine field measurements are element and site selective and thus allow us to probe the local environment and the coordination number of the atoms studied, i.e., have atomic scale resolution. In particular, hyperfine techniques have become important in the study of magnetism in multilayers and thin films. Hyperfine studies for example have been used to determine the magnetic anisotropy in ultrathin Fe/Ag(100) films[2] and to study the induced magnetization of nonmagnetic metallic spacers of, e.g., Cu in Fe/Cu multilayers[3] and Ag in the Fe/Ag(100) interface.[4–6]

Generally the interpretation and understanding of measured values of B_{hf} are difficult since the origin of B_{hf} comes from the behavior of electrons in the inner region of the atoms close to the nucleus. Their values are sensitive to details such as interlayer distances and reconstructions at the interface.

All electron *ab initio* theoretical studies based on the local spin density (LSDA) and the generalized gradient approximations (GGA) to (spin) density-functional theory (DFT) have shown to provide a practical tool for the investigations of many properties of solids including magnetism. Within LSDA (or GGA), spin-orbit (SO) coupling is the only effect considered responsible for orbital magnetization and to be included in the evaluation of the orbital and magnetic dipole contributions to B_{hf}. However, the theoretically calculated orbital magnetic moments underestimate the experimental values by approximately 50%. Other effects, namely noncentral field contributions to the electron-electron interaction[7] and orbital current polarization,[8] are neglected.

According to Brooks,[9–11] an extension to LSDA can be made to account for the spin-orbit induced orbital polarization (OP), which gives magnetic moments in fairly good agreement with the experimentally observed values. Recently, Ebert and Battocletti[12] showed that formally the orbital polarization correction of Brooks has the form to be expected from current density-functional theory (CDFT), which should provide a sound and rigorous basis for the investigation of properties related to orbital magnetism.[13] We want to point out that Brooks's OP term is quite different from the exchange-correlation vector potential which appears in CDFT.[12]

This paper presents results on spin-density functional calculations within LSDA and GGA including spin-orbit coupling in order to study magnetic moments and the influence of orbital and dipolar contributions to the hyperfine field. We also address the issue of how sensible these two contributions are to a self-consistent inclusion of the OP mechanism in the calculation (as proposed by Brooks[9–11]). Three systems are investigated: bulk bcc Fe with $\mathbf{M}\|[001]$, bulk hcp Co (with $\mathbf{M}\|[0001]$ or $[11\bar{2}0]$) and Fe/Ag(100) with $\mathbf{M}$ either perpendicular or parallel to the interface. We want to stress the fact that we study the anisotropy in the hyperfine interaction and the relative influence of the orbital and dipolar terms when going from bulk Fe to the Fe/Ag interface and establish the degree of prediction that *ab initio* determinations have for these quantities by comparing to experimental data.

II. DETAILS OF THE CALCULATIONS

In the present study the full potential LAPW method has been used as implemented in the WIEN97 code.[14,15] This has

proven to be one of the most accurate schemes for electronic structure calculations of solids. All electrons are treated self-consistently, the core fully relativistic and the valence electrons scalar relativistically. We have included local orbitals[16] (in addition to the usual LAPW basis) for the high lying core states—the so-called semicore states—to increase the flexibility of the basis set.

Within this method the unit cell is divided into nonoverlapping atomic spheres and an interstitial space. The potential is expanded into spherical harmonics inside the atomic spheres and in plane waves in the interstitial space. A similar expansion is used for the basis functions. Spin-orbit effects are treated within a second variational approach.[17] This means that the Hamiltonian without the spin-orbit coupling is diagonalized first. Then an energy cutoff is chosen and only the states under this spin-orbit cutoff energy are used as a basis for diagonalization of the Hamiltonian including the spin-orbit coupling. This procedure is faster compared to the direct inclusion of the spin-orbit coupling into the Hamiltonian. The accuracy of this approach can be systematically controlled by the energy cutoff.

All calculations have been performed using density-functional theory and both the LSDA approximation of Perdew and Wang[18] and the PBE-GGA approximation for the exchange-correlation functional.[19]

We use identical muffin-tin sphere radii for Fe, Co, and Ag of $R_{\mathrm{MT}}=2.2$ a.u. Fe and Co have semicore $3p$ and valence $3d$, and $4s$ states, and Ag has semicore $4p$ and valence $4d$, and $5s$ states. The energy cutoff for the LAPW wave functions was set to $E_{\mathrm{cut}}=13.2$ Ry. We have used a sufficiently fine **k** points sampling. In the present calculations, for the bulk systems the experimental lattice constants ($a=5.405$ a.u. for bcc Fe and $a=4.743$ a.u., $c=7.691$ a.u. for hcp Co) were used in order to evaluate the accuracy of the present approach while comparing to previous theoretical analyses within a fully relativistic approach at the same lattice parameters.[20,21] Moreover, we have tested the results of the LSDA and GGA approximations to the HFF's in bcc Fe at the calculated structural parameters within each approximation ($a=5.205, 5.351$ a.u. for LSDA and GGA, respectively). The Fe/Ag(100) interface is modeled using the supercell approach, where we use a five-layer (100) Ag slab with the theoretically determined lattice constant of fcc Ag as predicted by GGA, which deviates by 1.13% from the experimental value. Additional five (100) layers of bcc Fe assume a lattice constant fixed by the Ag substrate in the (100) plane. The interlayer spacings in Fe correspond to the DFT-GGA calculated lattice constant. The interface separation d_i was determined from energy minimization.

As already mentioned, it is found that the use of either LSDA or GGA leads to a considerable underestimation of the orbital magnetic moments.[22,23] The orbital moment may be enhanced by the inclusion of the orbital polarization term,[24] which mimics the second Hund's rule:

$$\hat{H}_{\mathrm{OP}}=H_{\mathrm{orb}}\hat{l}_z, \tag{1}$$

$$H_{\mathrm{orb}}=-I_{\mathrm{OP}}L_z, \tag{2}$$

where $\hat{l}_z$ is the z component of the angular moment operator, the z axis being parallel to the magnetization, and L_z is the projection of the orbital moment on the z axis:

$$L_z=\sum_{i,\mathbf{k}} n_{i,\mathbf{k}}\langle\psi_{i,\mathbf{k}}|\hat{l}_z|\psi_{i,\mathbf{k}}\rangle \tag{3}$$

$\psi_{i,\mathbf{k}}$ is the eigenstate of an energy band i with a wave vector $\mathbf{k}$; $n_{i,\mathbf{k}}$ are the occupation numbers. Finally the parameter I_{OP} is equal to the Racah parameter B for the d electrons and E^3 for the f electrons. Racah parameters are easily calculated once the wave functions inside the atomic spheres are known (see, e.g., Ref. 25).

In our computational scheme $\hat{H}_{\mathrm{OP}}$ is treated using the second-variational method,[17] i.e., in the same way as the spin-orbit coupling. In the framework of this approach $\hat{H}_{\mathrm{OP}}$ is proportional to the orbital momentum operator $\hat{l}_z$. At the same time $\hat{H}_{\mathrm{OP}}$ and H_{orb} are coupled via Eq. (1). Therefore an additional self-consistency loop must be added into the computational scheme. In the first step L_z is calculated using the eigenfunctions of the Hamiltonian $\hat{H}_0$, which does not contain the orbital polarization term (but the spin-orbit coupling). $\hat{H}_{\mathrm{OP}}$ is then determined using the expression above and the eigenfunctions of $\hat{H}_0+\hat{H}_{\mathrm{OP}}$ are found. L_z is calculated in the new basis and the new $\hat{H}_{\mathrm{OP}}$ is constructed. The procedure is repeated until self-consistency is reached. We found that for the systems discussed here the procedure converges fast and no mixing of the intermediate results is necessary.

The relativistic expressions for the B_{hf} in a perturbative approach have been derived by several groups.[26–28] In the lowest order, B_{hf} is given by

$$B_{\mathrm{hf}}=B_{\mathrm{hf}}^{\mathrm{ct}}+B_{\mathrm{hf}}^{\mathrm{o}}+B_{\mathrm{hf}}^{\mathrm{d}} \tag{4}$$

which consists of the conventional contact term $B_{\mathrm{hf}}^{\mathrm{ct}}$, and the orbital and dipolar contributions $B_{\mathrm{hf}}^{\mathrm{o}}+B_{\mathrm{hf}}^{\mathrm{d}}$. The latter are usually called the non-s contribution to the hyperfine field, $B_{\mathrm{hf}}^{\mathrm{ns}}$. Further decomposition of $B_{\mathrm{hf}}^{\mathrm{ct}}$ is made as a sum of $B_{\mathrm{hf}}^{\mathrm{cv}}$ (contact valence) and $B_{\mathrm{hf}}^{\mathrm{cc}}$ (contact core). In our scalar-relativistic approach an expression derived by Blügel[28] for $B_{\mathrm{hf}}^{\mathrm{ct}}$ has been used. Simple relations exist between the spin, orbital and magnetic dipole moments and the contact, orbital, and dipolar contributions to the hyperfine fields.[20]

In all results here presented we have only considered the intra-atomic hyperfine fields and, for simplicity, have neglected those contributions from the spin moments on the other atoms. That interatomic field comes only from the magnetic dipole and orbital terms, and therefore is small. For cubic systems, this effect is zero and for hcp Co has been estimated to be less than 0.05 kG.[20] Its detailed evaluation for the Fe/Ag interface problem is outside the purpose of our study but we expect it to be small compared to the intra-atomic contributions.

TABLE I. Spin and orbital magnetic moments (M,M_o) and hyperfine field (B_{hf}) for bulk bcc Fe for $\mathbf{M}\|[001]$. B_{hf}^{cv}, B_{hf}^{cc}, B_{hf}^{ct}, B_{hf}^{o}, B_{hf}^{d} are the contact-valence, contact-core, contact-total, orbital, and dipolar contributions to the hyperfine field, respectively. SO means that spin-orbit has been included and SO+OP indicates that the orbital polarization contribution has been added (see text). The indices *expt* and *calc* mean that the calculations have been performed at the experimental or DFT-LSDA (GGA) theoretical lattice constant, respectively. A comparison is also made with previous LSDA calculations (at the experimental lattice constant) by Guo and Ebert (Ref. 20), and experiment (Ref. 29). vBH means von Barth-Hedin local density potential and VWN Vosko-Wilk-Nusair potential (see Ref. 20) for details. Magnetic moments are in μ_B and hyperfine fields in kG.

bcc Fe				SO				
	M	B_{hf}^{cv}	B_{hf}^{cc}	B_{hf}^{ct}	M_o	B_{hf}^{o}	B_{hf}^{d}	B_{hf}
Present work (LSDA)^{expt}	2.23	−46.3	−258.1	−304.4	0.048	26.0	0.0	−278.0
Present work (LSDA)^{calc}	2.03	−41.4	−231.2	−272.7	0.038	19.4	0.0	−253.2
Present work (GGA)^{expt}	2.23	−34.2	−287.9	−322.1	0.045	24.1	0.0	−298.0
Present work (GGA)^{calc}	2.20	−36.8	−283.3	−320.1	0.041	20.6	0.0	−299.5
Guo-Ebert (vBH)		−65.9	−237.5	−303.4		15.2	0.0	−288.2
Guo-Ebert (VWN)	2.175	−72.8	−250.2	−323.0	0.042	15.2	0.0	−307.9
Experiment	2.13				0.080			−339.0
				SO+OP				
					M_o	B_{hf}^{o}	B_{hf}^{d}	B_{hf}
Present work (LSDA)^{expt}					0.086	51.3	0.0	−253.1
Present work (LSDA)^{calc}					0.060	33.6	0.0	−239.0
Present work (GGA)^{expt}					0.078	45.7	0.0	−276.4
Present work (GGA)^{calc}					0.065	36.2	0.0	−283.9
Experiment					0.080			−339.0

III. RESULTS

Our results are summarized in Tables I–III. Table I presents our results for bulk bcc Fe with $\mathbf{M}\|[001]$, Table II for bulk hcp Co (with $\mathbf{M}\|[0001]$ and $[11\bar{2}0]$), and Table III for the Ag/Fe(001) interface (with **M** perpendicular and parallel to the interface). In the upper panel of Tables I–III, the results with the inclusion of the spin-orbit term (SO) are shown, and in the lower panel results which include orbital polarization (SO+OP) effects are listed.

For bulk bcc Fe with $\mathbf{M}\|[001]$ (see Table I) we report values calculated with both LSDA and GGA potentials, evaluated at the experimental equilibrium structural parameters (expt), as well as at the calculated LSDA or GGA lattice parameter (calc), respectively. The differences in the calculated B_{hf} (at the experimental lattice constant) due to the use of different exchange correlation potentials amount up to 10% (see upper panel of Table I). The DFT-GGA calculated B_{hf} at the GGA determined lattice parameter differs by 0.5% from the value at the experimental lattice constant. This is mainly because of the small 1% underestimation of the DFT-GGA parameter. On the other hand, LSDA predicts a lattice constant much smaller than the experimental one (−3.7%) and the resulting B_{hf} becomes quite dissimilar to the other theoretical values and much smaller (−25%) than the experiment. The (LSDA)^{expt} result is the one to be compared with the work by Guo and Ebert.[20,21] The difference between these three LSDA results can be traced to the different parametrizations used for the LSDA exchange correlation potential. From this comparison, it is clear that the larger differences are mainly caused by the different lattice constants.

Interestingly, the DFT-GGA predicted spin magnetic moment M differs by less than 5% from the experimental value and from other DFT-LSDA results (at the experimental lattice parameter).[20] On the contrary, M_o, the orbital magnetic moment, as predicted with SO, is found to be 50% too small when compared to the experimental value. This can be partially compensated by the inclusion of Brook's OP term as shown in the lower panel of Table I. Orbital contributions to B_{hf} are of the order of 8% of the total value. Dipolar contributions vanish due to the cubic symmetry of the surrounding.

Comparing now the magnitude of the different contributions to B_{hf} of Fe bcc bulk one sees that already the contact term B_{hf}^{ct} is close to the experimental value (see upper panel of Table I) and the addition of the positive orbital contribution results in a 10% disagreement with the experimental value. If SO+OP is included, the (positive) orbital contribution is even more expressed. Although in this case the orbital magnetic moment agrees very well with the experimental value, the disagreement in the contact term is even larger (about 25%).

The case of bulk hcp Co (Table II) is similar. The SO calculation predicts a spin magnetic moment close to the experimental value. But the orbital moment (M_o) is a factor of 2 too small. The contact term overestimates B_{hf} by 10% but adding the orbital contribution makes it 10% underestimated (see upper panel of Table II). The orbital moment M_o is close to experiment when SO+OP is used but as in the case of bulk Fe, the increased positive contribution of the

TABLE II. Spin and orbital magnetic moments (M,M_o) and hyperfine field (B_{hf}) for bulk hcp Co for $\mathbf{M}\|[0001]$ or $[11\bar{2}0]$ directions. B_{hf}^{cv}, B_{hf}^{cc}, B_{hf}^{ct}, B_{hf}^{o}, B_{hf}^{d} are, respectively the contact-valence, contact-core, contact-total, orbital, and dipolar contributions to the hyperfine field. SO means that spin orbit has been included and SO+OP indicates that the orbital polarization contribution has been added. The index *expt* mean that the calculations have been performed at the experimental lattice constant. A comparison is also made with previous LSDA calculations (at the same lattice constant) by Guo and Ebert (Ref. 20), and experiment (Ref. 29). Magnetic moments in μ_B and hyperfine fields in kG.

hcp Co					SO			
	M	B_{hf}^{cv}	B_{hf}^{cc}	B_{hf}^{ct}	M_o	B_{hf}^{o}	B_{hf}^{d}	B_{hf}
$\mathbf{M}\|[0001]$								
Present work (LSDA)^{expt}	1.58	−71.2	−193.9	−265.2	0.079	59.0	−0.59	−206.8
Guo-Ebert (VWN)	1.60	−85.2	−190.5	−275.7	0.077	48.60	1.89	−225.2
Experiment	1.52				0.140			−219.0
$\mathbf{M}\|[11\bar{2}0]$								
Present work (LSDA)^{expt}	1.59	−71.4	−193.9	−265.9	0.075	55.9	0.57	−209.5
Guo-Ebert (VWN)	1.59	−85.2	−190.5	−275.7	0.072	45.77	−1.27	−231.1
Experiment	1.52				0.140			−227.0
					SO+OP			
					M_o	B_{hf}^{o}	B_{hf}^{d}	B_{hf}
$\mathbf{M}\|[0001]$								
Present work (LSDA)^{expt}					0.13	100.9	−0.31	−164.6
Experiment					0.14			−219.0
$\mathbf{M}\|[11\bar{2}0]$								
Present work (LSDA)^{expt}					0.13	93.9	0.87	−171.1
Experiment					0.14			−227.0

orbital term makes the disagreement of the hyperfine field to be of the order of 25% (see lower panel of Table II). It is interesting to notice that M_o decreases when going from $\mathbf{M}\|[0001]$ to $\mathbf{M}\|[11\bar{2}0]$. The anisotropy is almost completely caused by the anisotropy in the non-*s* hyperfine field, which in turn is correlated with the anisotropy in the magnetic dipole and orbital moments. Therefore spin-orbit coupling is essential in order to study the anisotropy in the hyperfine field. We do reproduce a change of B_{hf} in the right direction. The experimentally determined 8-kG anisotropy of the B_{hf} is reproduced in our calculations only when SO+OP is included.

Finally we analyze the case of the Fe/Ag(100) interface (Table III). We use supercells with five layers of Fe and an equal number of Ag layers (5+5) as already described. Atoms Ag_1, Ag_2, and Ag_3 in the table denote the Ag atoms. Ag_3 is the Ag atom at the interface and Ag_1 can be considered as representing an Ag bulklike atom. Atoms Fe_1, Fe_2, and Fe_3 are Fe atoms and Fe_3 is the Fe atom at the interface. We note that because of the occurrence of interface induced Friedel oscillations of magnetic moments and hyperfine fields in multilayers, the result for the magnetic moment of Fe_1 atoms, in particular, is at variance with that of bulk Fe (see Tables I and III). To obtain a better agreement the use of a much larger supercell would have been necessary. Nevertheless, test calculations for a 7+7 supercell indicated that the conclusions made in this work are not affected by the size of the supercell used. We have relaxed the structure (interface distance d_i) within the GGA approximation and a 7+7 supercell, obtaining a value of $d_i=3.5$ a.u. Importantly, the magnitude $|B_{hf}^{ct}|$ is quite sensitive to the value of d_i. For example, at the Ag side ranges from zero at $d_i=\infty$ to 400 kG at $d_i=2.5$ bohr and the difference between its value for atoms Fe_3 and Ag_3, i.e., across the Fe/Ag(100) interface, can vary from 140 kG to 0 when d_i varies from 2.5 to 3.7 a.u. This suggests that it is important to allow for this relaxation before comparing theoretical results to experiments, and that hyperfine-field measurements in combination with theoretical calculations can be used as a probe of the structural and chemical environment of the considered atom in magnetic multilayers.

Very recent experiments using the low temperature nuclear orientation (LTNO) technique for the Fe/Ag(100) interface[30] have complemented previous Mössbauer studies by Liu *et al.*[5] and Schurer *et al.*[6] and shown that the induced Ag hyperfine field of the Ag at the Fe/Ag interface is directed out of the plane of the multilayer, i.e., perpendicular to the magnetization of the Fe which lies in the plane of the multilayer. The calculations we have performed assumed that the induced magnetic hyperfine field of the Ag is parallel to that of Fe, thus the results may not be directly compared to the experiments. Nevertheless, the relative influence on magnetic moments and hyperfine fields of orbital and dipolar effects at the Fe atom of the interface, where larger effects are expected due to the reduced symmetry compared to the bulk case, can be discussed. A straightforward comparison

TABLE III. Spin and orbital magnetic moments (M,M_o) and hyperfine field (B_{hf}) for Fe/Ag interface for $\mathbf{M}\|$ to the (001) and (100) directions. A 5+5 supercell was used. Atoms Ag_1 and Fe_1 would represent bulk Ag and Fe, respectively. Atoms Ag_3 and Fe_3, Ag and Fe at the relaxed interface. B_{hf}^{cv}, B_{hf}^{cc}, B_{hf}^{ct}, B_{hf}^{o}, B_{hf}^{d} are, respectively the contact-valence, contact-core, contact-total, orbital, and dipolar contributions to the hyperfine field. SO means that spin-orbit has been included and SO+OP indicates that the orbital polarization contribution has been added. Magnetic moments in μ_B and hyperfine fields in kG.

Fe/**Ag**(001)					SO				
	Atom	M	B_{hf}^{cv}	B_{hf}^{cc}	B_{hf}^{ct}	M_o	B_{hf}^{o}	B_{hf}^{d}	B_{hf}
$\mathbf{M}\|(001)$									
Present work (GGA)	Ag_1	0.00	8.1	−0.31	7.8	0.2×10^{-3}	0.85	1.4	10.5
	Ag_2	0.00	−2.1	−0.07	−2.1	0.4×10^{-4}	−0.25	4.8	2.45
	Ag_3	0.04	−293.3	−14.6	−308.0	0.3×10^{-2}	5.9	9.4	−292.7
	Fe_3	2.82	59.8	−352.9	−293.1	0.078	45.0	−8.7	−256.8
	Fe_2	2.49	−46.3	−314.6	−361.0	0.053	29.0	1.6	−330.4
	Fe_1	2.52	−8.6	−318.0	−326.6	0.050	27.0	2.6	−297.0
$\mathbf{M}\|(100)$									
Present work (GGA)	Ag_1	0.00	8.3	−0.1	8.2	0.2×10^{-3}	0.4	−0.6	8.0
	Ag_2	0.00	−1.9	0.12	−1.81	-0.1×10^{-3}	−0.4	−2.4	−4.6
	Ag_3	0.04	−292.9	−14.5	−307.4	0.4×10^{-2}	6.5	−4.6	−305.5
	Fe_3	2.82	60.0	−352.2	−292.2	0.062	34.9	4.1	−253.2
	Fe_2	2.49	−46.2	−314.0	−360.2	0.052	28.3	−0.9	−332.8
	Fe_1	2.52	−8.2	−317.1	−325.3	0.050	28.0	−1.4	−298.7
					SO+OP				
	Atom					M_o	B_{hf}^{o}	B_{hf}^{d}	B_{hf}
$\mathbf{M}\|(001)$									
Present work (GGA)	Ag_1					0.19×10^{-3}	0.8	1.4	10.0
	Ag_2					0.15×10^{-3}	−0.05	4.8	2.6
	Ag_3					0.3×10^{-2}	3.8	9.4	−306.8
	Fe_3					0.149	90.0	−8.9	−211.9
	Fe_2					0.094	56.0	1.4	−303.6
	Fe_1					0.083	49.0	2.5	−275.1
$\mathbf{M}\|(100)$									
Present work (GGA)	Ag_1					0.2×10^{-3}	0.5	−0.7	8.0
	Ag_2					-0.9×10^{-4}	−0.5	−2.4	−4.7
	Ag_3					0.3×10^{-2}	5.5	−4.6	−306.5
	Fe_3					0.104	61.9	3.9	−226.4
	Fe_2					0.084	49.9	−1.0	−311.3
	Fe_1					0.082	48.6	−1.5	−278.2

with the theoretical work of Guo and Ebert[20,21] becomes difficult since they only studied monolayers of Fe on fcc Ag where many effects such as anisotropy become much more exaggerated. Some qualitative observations (also extracted from their Fe/Au bilayers studies) are nevertheless similar. Our main observations are: (i) Spin and orbital moments increase as one approaches the interface from the Fe side. They are negligible at the Ag side. Yet, small induced moments are observed at the Ag interface layer. (ii) Interface-induced Friedel oscillations at the Fe side are found in qualitative agreement with experiments by Liu and Gradmann[5] and theoretical pioneering work of Ohnishi *et al.*[31] as well as in more recent theoretical studies (in this case for the Fe/Au interface).[20] (iii) Both the orbital and magnetic dipole contributions to B_{hf} in the multilayers are of comparable magnitudes in the Ag side of the interface and considerable smaller than the contact term. Whereas at the Fe side, the orbital term represents 20–30% of the total hyperfine field. When approaching the interface from the Fe side, the orbital term increases due to the enhanced orbital moment. (iv) The calculated B_{hf} for Fe at the interface is reduced compared to that of the corresponding bulk metal. This becomes mostly because the valence *s*-electron hyperfine field B_{hf}^{cv} on the Fe at the interface changes sign and its magnitude overcompensates the increased B_{hf}^{cc} due to the enhancement of M. This reduction is further increased by the enhanced orbital term. This is in agreement with monolayer-probe ^{57}Fe Mössbauer-effect measurements on bcc Fe/fcc Ag(100) bilayers where several satellites were observed for the ^{57}Fe at the interface. An average value of −283 kG between these satellites is close to our calculated values for the different magnetization orientations obtained with SO. Similarly as for the Fe and Co

bulk studies, the inclusion of OP reduces the total B_{hf} even further, while increasing the orbital magnetic moment. (v) The calculated magnetic anisotropy in B_{hf} from in-plane and perpendicular magnetizations amounts up to 4 kG and stems almost completely from the non-*s* electron contributions B_{hf}^{o} and B_{hf}^{d}. (vi) As already discussed, the results are sensitive to the interface separation.

IV. CONCLUSIONS

We have performed an *ab initio* all electron study using both LSDA and GGA approximations to DFT which included spin-orbit effects, and obtained hyperfine fields and magnetic moments (spin and orbital) for three test cases: bulk bcc Fe, hcp Co, and Fe/Ag(100) interface. In the bcc Fe case we studied $\mathbf{M} \| [001]$. In the other cases **M** was taken parallel and perpendicular to the interface for the Fe/Ag(100) interface and in-plane and perpendicular to the hexagonal planes of the Co hcp structure.

The use of LSDA or GGA gives a reasonable approximation to the spin moment. The inclusion of spin-orbit coupling allows to obtain the orbital moment. But this is a factor 2 smaller than the experimental value. Orbital polarization (OP), which has the effect of enhancing the spin-orbit induced orbital moments has been considered by the OP approach, as proposed by Brooks. The enhancement of the orbital moments nearly removes the discrepancy between theory and experiment, but the predicted B_{hf} disagrees much more than when OP was not included.

In the case of Fe/Ag(100) interface the hyperfine fields are very sensitive to the interface separation d_i. And, d_i in turn to the exchange correlation energy used. If the minimized d_i is used, LSDA or GGA predictions are quantitatively different but the qualitative behavior of B_{hf} at the interface of Fe/Ag(001) remained: it decreases at the interface layer of Fe and increases as an induced hyperfine field on the first monolayer of Ag. Unlike the bulk metals both the orbital and magnetic dipole contributions to B_{hf} become important. A magnetic anisotropy was found for the B_{hf} which can be linked to the anisotropy observed in the orbital and dipolar terms.

The *ab initio* determination of B_{hf} and its link to spin and orbital moments helps establish a qualitative picture. Further work is needed in order to improve on a quantitative one. In particular, the noncollinearity observed in recent experiments should be taken into account.

ACKNOWLEDGMENTS

We acknowledge helpful communications with B. U. Runge, G. Schatz, M. Prandolini, E. K. U. Gross, W. D. Brewer, and M. Scheffler during the course of this work. This work was supported by a DRL-SECYT (Germany-Argentina) International Collaboration, Project No. ARG AD 45P. The implementation of the orbital polarization in the WIEN97 code was supported by Grant No. A1010715 of the AS CR.

[1] See, e.g., C.E. Johnson, Hyperfine Interact. **90**, 24 (1994), and references therein.
[2] N.C. Koon, B.T. Jonker, F.A. Volkening, J.J. Krebs, and G.A. Prinz, Phys. Rev. Lett. **59**, 2463 (1987).
[3] Q.Y. Jin, Y.B. Xu, H.R. Zhai, C. Hu, M. Lu, Q.S. Bie, Y. Zhai, G.L. Dunifer, R. Naik, and M. Ahmad, Phys. Rev. Lett. **72**, 768 (1994).
[4] B.U. Runge, M. Dippel, G. Fillebock, K. Jacobs, U. Kohl, and G. Schatz, Phys. Rev. Lett. **79**, 3054 (1997).
[5] G. Liu and U. Gradmann, J. Magn. Magn. Mater. **118**, 99 (1993).
[6] P.J. Schurer, Z. Celinski, and B. Heinrich, Phys. Rev. B **51**, 2506 (1995).
[7] I.V. Solovyev, A.I. Liechtenstein, and K. Terakura, Phys. Rev. Lett. **80**, 5758 (1998).
[8] K. Capelle and E.K.U. Gross, Phys. Rev. Lett. **78**, 1872 (1997).
[9] M.S.S. Brooks, Physica B **130**, 6 (1985).
[10] O. Eriksson, M.S.S. Brooks, and B. Johansson, Phys. Rev. B **41**, 7311 (1990).
[11] H. Ebert and M. Battocletti, Solid State Commun. **98**, 785 (1996).
[12] H. Ebert, M. Battocletti, and E.K.U. Gross, Europhys. Lett. **40**, 545 (1997).
[13] G. Vignale, in *Current Density Functional Theory and Orbital Magnetism*, Vol. 337 of *Nato Advanced Study Institute, Series B: Physics*, edited by E.K.U. Gross and R.M. Dreizler (Plenum Press, New York, 1995), p. 485.
[14] P. Blaha, K. Schwarz, P. Dufek, and R. Augustyn, WIEN97, Technical University of Vienna, 1997 [improved and updated Unix version of the original copyrighted WIEN code, which was published by P. Blaha, K. Schwarz, P. Sorantin, and S. B. Trickey, in Comput. Phys. Commun. **59**, 399 (1990)]; P. Blaha, K. Schwarz, and P.H. Dederichs, Phys. Rev. B **37**, 2792 (1988).
[15] J. Kunes and P. Novak, J. Phys.: Condens. Matter **11**, 6301 (1999).
[16] D.J. Singh, Phys. Rev. B **43**, 6388 (1991).
[17] D.J. Singh, *Plane Waves, Pseudopotentials and the LAPW Method* (Kluwer Academic, Assinippi Park, Norwell, MA, 1994).
[18] J.P. Perdew and Y. Wang, Phys. Rev. B **45**, 13 244 (1992).
[19] J.P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. **77**, 3865 (1996); **78**, 1396 (1997).
[20] G.Y. Guo and H. Ebert, Phys. Rev. B **53**, 2492 (1996).
[21] G.Y. Guo and H. Ebert, Phys. Rev. B **50**, 10 377 (1994).
[22] P. Söderlind, O. Eriksson, B. Johansson, R.C. Albers, and A.M. Boring, Phys. Rev. B **45**, 12 911 (1992).
[23] T. Huhne, C. Zecha, H. Ebert, P.H. Dederichs, and R. Zeller, Phys. Rev. B **58**, 10 236 (1998).
[24] O. Eriksson, B. Johansson, and M.S.S. Brooks, J. Phys. C **1**, 4005 (1989).
[25] J.S. Griffith, *The Theory of Transition-Metal Ions* (Cambridge University Press, Cambridge, 1961).
[26] G. Breit, Phys. Rev. **35**, 1447 (1930).
[27] L. Terlikis, S.D. Mahanti, and T.P. Das, Phys. Rev. **176**, 10 (1968).

[28] S. Blügel, H. Akai, R. Zeller, and P.H. Dederichs, Phys. Rev. B **35**, 3271 (1987).
[29] M. Kawakami, T. Hihara, Y. Koi, and T. Wakiyama, J. Phys. Soc. Jpn. **33**, 159 (1972).
[30] T. Phalet, J. Camps, P. De Moor, P. Schuurmans, N. Severijns, M. Trhlik, A. Van Geert, L. Vanneste, B. Vereecke, and W.D. Brewer, J. Magn. Magn. Mater. **165**, 234 (1997).
[31] S. Ohnishi, M. Weinert, and A.J. Freeman, Phys. Rev. B **30**, 36 (1984).