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SPIN-POLARIZED FIRST-PRINCIPLES LMTO-ASA  
CALCULATIONS IN REAL-SPACE

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"Spin-polarized First-Principles LMTO-ASA Calculations in Real-Space"

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ABSTRACT

We have developed an approach, based on the LMTO-ASA formalism and the recursion method, which allows us to perform first principles spin-polarized self-consistent calculations of electronic structure in real space. To illustrate the method we obtain the electronic structure of ferromagnetic  $\text{FeNi}_3$  and antiferromagnetic  $\text{FeMn}$ . The results compare well with those obtained by other methods. The scheme described here can be applied to non-periodic systems and is very useful to obtain local magnetic moments in complex metallic systems.

I. INTRODUCTION

Real space methods such as the recursion method<sup>1</sup> do not require symmetry and their cost when solving an eigenvalue problem grows linearly with the number of inequivalent atoms being considered. For these reasons, real space methods are very useful to describe the electronic properties of complex systems, for which the usual k-space methods are inapplicable or extremely costly. Real-space methods are not very practical in general, but they are extremely efficient when the system in consideration can be well described by a tight-binding (TB) Hamiltonian. Because localized d-bands play a central role in the electronic structure of transition metal alloys for a long time parametrized TB Hamiltonians and real space methods have been used to study the magnetic properties of these systems. A lot of progress in understanding the magnetic properties of metallic systems was made by using this approach and simple d-band parametrized model Hamiltonians. Usually the parameters are obtained from a LCAO fit to more exact k-space calculations or adjusted to fit experimental results and it is assumed that they can be transferred to describe the more complex systems one wants to study<sup>2</sup>. In the case of magnetic systems the Stoner parameter is also needed. This is an additional problem, because the results for the magnetic moments are sensitive to the choice of  $I$  and often the results can be made to agree with experiment through small variations of this quantity. The LCAO parametrization has often been extended, with encouraging results, to treat s and p electrons but the lack of a sound theoretical background to justify the procedure leaves some fundamental questions unanswered. Which are the approximations being used when one forces the Hamiltonian to be tightly bound through

a fit? Should the usually extended s-p electrons be treated within the TB scheme? How do we treat the wave function and quantities which which depend on it? Do the parameters in binary alloys change as a function of the magnetic moment?

A major progress towards obtaining a tight-binding Hamiltonian based on a solid theoretical understanding of the problem, came in 1984 with the advent of the LMTO-ASA-TB formalism<sup>3</sup>. The LMTO-ASA is a linear method implemented around an energy  $E_V$  that treats s-p and d electrons in the same manner. In this formalism, the Hamiltonian can be expressed in terms of different sets of basis functions<sup>4</sup>. One can always choose an appropriate set for which the relevant part of the Hamiltonian is tightly bound for s-p and d electrons. The sound theoretical framework of the LMTO-ASA formalism allows us to evaluate wave functions, and to know exactly which approximations being made. Within the LMTO-ASA theory, simple parametrized Hamiltonians can be built without the need of fits to more exact calculations or experiment<sup>5-6</sup>. The lack of adjustable parameters make the results more reliable. Parametrized LMTO-ASA calculations have been used with success to study the electronic structure of several systems. In the case of magnetic systems, it has been used in conjunction with the Stoner criterion to obtain the electronic structure of  $\text{FeNi}_3$ <sup>6</sup>. But in some cases, such as that of antiferromagnetic  $\text{FeMn}$ , the parametrized LMTO-ASA approach fails. This suggests that a more rigorous first principles self-consistent approach may be needed, to obtain reliable results for the magnetic properties of complex metallic systems. The possibility of using the LMTO-ASA TB formalism in conjunction with the recursion method in order to perform self-consistent calculations

in real space was first pointed out by Fujiwara<sup>7</sup>. At that time some technical aspects regarding the determination of reliable LMTO-ASA TB structure constants were still unknown. They were interested in a very complex amorphous Fe-B alloy, and therefore never attempted a site by site self-consistency or a spin-polarized calculation. They made a self-consistent calculation for the average Fe and B in the cluster, ignoring for sake of simplicity, the local variations of the potential.

In this paper we develop a spin-polarized, first-principles, self-consistent real space calculation. The procedure is very similar to the regular k-space LMTO-ASA formalism, but the solution of the eigenvalue problem is implemented in real-space with the help of the recursion method<sup>1</sup>. This approach has been tested in non-magnetic  $\text{Zr}_2\text{Fe}$ , giving occupation numbers for s-p and d-bands which agree within one hundredth of electrons with those obtained in k-space<sup>8</sup>. The paper is organized in the following way. In Sec. II we give a description of the LMTO-ASA and of its several representations. In Sec. III we describe the real-space self-consistent approach and present some results for  $\text{FeNi}_3$  and  $\text{FeMn}$ . Finally, in Sec. V. we present our conclusions.

## II. THE LMTO-ASA-TB FORMALISM

The LMTO-ASA-TB formalism is a well known first-principles method and has been described in several papers<sup>3,4,9,10</sup>. Therefore we will be as brief as possible in our discussions of the method. The LMTO is a linear method and its solutions are valid around a given energy  $E_V$ . Here, as in most of the literature,  $E_V$  is chosen at the center of gravity of the occupied part of the given (s,p or d) band. We use a first order TB Hamiltonian where terms of order of  $(E-E_V)^2$  and higher are neglected. We also work in the atomic sphere approximation (ASA),

where the space is divided into Wigner-Seitz cells, which are then approximated by Wigner-Seitz (WS) spheres of same volume. The function  $\varphi_{\nu}^{\circ}(r)$ , defined as the radial part of the solution of the Schrödinger equation for a spherical potential inside a given sphere at energy  $E_{\nu}$ , and its energy derivative  $\dot{\varphi}_{\nu}(r)$  defined at energy  $E_{\nu}$ , are very fundamental quantities in the LMTO-ASA formalism. To understand the LMTO-ASA formalism from a real space point of view we should start by describing the choice of the LMTO-ASA basis functions. Instead of obtaining the basis set from solutions of an isolated atom (as in the LCAO), we consider the solutions for an isolated muffin-tin sphere of radius  $s$ , with a given spherical potential for  $r < s$  and a flat potential outside. It is assumed that the kinetic energy for one electron outside the muffin-tin sphere is approximately zero and the solution of the Schrödinger equation outside the muffin-tin sphere reduces to the non-divergent solution of Laplace's equation  $r^{-\ell-1}$  where  $\ell=0,1,2$  for s, p and d orbitals respectively. The solution inside the sphere should match the one outside at the sphere boundary. This set of muffin-tin orbitals will be used as an envelope in order to force the LMTO-ASA basis set to be continuous and differentiable in all space. To build the LMTO-ASA basis functions from given muffin-tin orbitals we use a procedure involving  $\varphi_{\nu}(r)$  and  $\dot{\varphi}_{\nu}(r)$  which is described below. First we consider the orbital centered at a site  $R$ . The tail goes as  $|r - R|^{-\ell-1}$  outside the central sphere and is a regular function within every other sphere centered around any  $R' \neq R$ . Around the site  $R'$ , the tail can be expanded in series using regular solutions  $|r - R'|^{\ell}$  of the Laplace equation. If we use a scale  $a$  and define  $r_{\frac{R}{R'}} \equiv |r - R|$ , the tail of a muffin-tin orbital centered at  $R$  can be expressed around any other site

$R'$  by the expansion<sup>9</sup>:

$$\left[ \frac{r_R}{a} \right]^{-\ell-1} Y_L(\hat{r}_R) = - \sum_{L'} \left[ \frac{r_{R'}}{a} \right]^{\ell'} \frac{Y_{L'}(\hat{r}_{R'})}{2(2\ell'+1)} S_{R'L',RL}^0 \quad (1)$$

where  $L=(\ell,m)$  is a collective angular momentum index and  $S_{R'L',RL}^0$  are the well known coefficients of the expansion<sup>9</sup>. These coefficients depend on the position of the sites on the given structure, but not on the type of atoms being considered. Now that the envelope function is written in a convenient form, to build the corresponding LMTO-ASA orbital, we substitute the solutions of the envelope inside every WS sphere by a linear combination of  $\varphi_{\nu}(r)$  and  $\dot{\varphi}_{\nu}(r)$ , chosen in order to preserve the value of the function and its derivative at the sphere boundary. When built in this way the LMTO-ASA basis is nearly orthogonal to the core levels and provides a much better basis for the actual solutions than the original muffin-tin orbitals. Using the LMTO-ASA basis set  $\{X_{RL}^0\}$  we can build the Hamiltonian  $H^0$  and the overlap matrix  $O^0$  in the usual way. These quantities can be expressed in terms of  $S^0$  and of potential parameters which depend on the values of the functions  $\varphi_{\nu}(r)$  and  $\dot{\varphi}_{\nu}(r)$  substituted at the WS sphere boundary.

Until now we have described the standard LMTO-ASA formalism<sup>10</sup>, which does not give rise to a TB Hamiltonian. The structure matrix  $S^0$  entering the Hamiltonian decays as  $r^{-2\ell-1}$  with distance and is very long ranged for s( $\ell=0$ ) and p( $\ell=1$ ) orbitals. Andersen and Jepsen<sup>3</sup> have shown that one of the characteristics of the LMTO-ASA formalism is that that the choice of basis set can be changed to suit ones' purpose. A controlled mixing of the original basis set can yield a new basis, built to have a particularly desirable property. For a general basis

$\{\bar{\chi}_{R,L}\}$  the amount of mixing is determined by a set of parameters  $\bar{Q}_\ell$ . These parameters define the basis and can be adjusted to produce a set with the desired property. Because the sets are related through mixing, they can be obtained from each other. There are three very important LMTO-ASA representations. The first is the standard representation with no mixing ( $\bar{Q}_\ell=0$ ) which we have described. The second is the nearly orthogonal representation where  $\bar{Q}_\ell$  is chosen to make the overlap matrix close to unit. Finally we have the TB or most localized representation, with a mixing chosen to make the interactions between neighboring sites as short ranged as possible. Here, following the literature<sup>4</sup>, we use quantities without bars to denote the potential parameters  $Q_\ell$ ,  $C_\ell$ ,  $\Delta_\ell$  in the nearly orthogonal representation. The mixing  $Q_\ell$  and the other potential parameters  $C_\ell$  and  $\Delta_\ell$  in the orthogonal representation are given in terms of the solutions at the boundary of each WS sphere, being different for every inequivalent atom in the system. From now we will use quantities with a bar to designate quantities in the most localized representation. The structure constant matrix  $\bar{S}$  for a basis set defined by a mixing  $\bar{Q}_\ell$  is written in terms of the original canonical structure matrix  $S^0$  of eq.1 as<sup>4</sup>:

$$\bar{S} = S^0(I - \bar{Q})^{-1} \quad (2)$$

Here  $I$  is the unit matrix and  $\bar{Q}$  is a diagonal matrix with elements  $\bar{Q}_\ell$ . Using this expression, the mixing that gives the TB or most localized representation can be found by trial and error<sup>3</sup>, choosing the values of  $\bar{Q}$  in expression (2) in order to obtain a localized structure constant matrix  $\bar{S}$ . The values of mixing were found to be approximately independent of the structure and are given for s,p and d electrons by

$$\bar{Q}_s = 0.3485, \quad \bar{Q}_p = 0.05293 \text{ and } \bar{Q}_d = 0.010714^{-3-4}$$

In the self-consistent real-space approach described in this paper, we will work on the orthogonal representation<sup>9</sup>, but will express the orthogonal Hamiltonian in terms of localized parameters of the TB representation. Because the basis functions the several representations are not independent, the orthogonal parameters  $C_\ell$ ,  $\Delta_\ell$ ,  $Q_\ell$  are related to the potential parameters  $\bar{C}$  and  $\bar{\Delta}$  of the TB representation. For a given energy  $E_\nu$  we have<sup>4</sup>:

$$\frac{\bar{C}_\ell - E_\nu}{C_\ell - E_\nu} = \frac{\bar{\Delta}^{1/2}}{\Delta^{1/2}} = 1 - (Q_\ell - \bar{Q}_\ell) \frac{C_\ell - E_\nu}{\Delta_\ell} \quad (3)$$

Finally, to first order in  $E - E_\nu$  we can express the Hamiltonian  $H$  of the orthogonal representation, in terms of TB parameters<sup>3,4</sup> as:

$$H = \bar{C} + \bar{\Delta}^{1/2} \bar{S} \bar{\Delta}^{-1/2} \quad (4)$$

In the orthogonal representation the overlap matrix is close to unit and we have to solve a simple eigenvalue problem of the form:

$$(H - E)C = 0$$

$$\psi_E = \sum_{RL} \left\{ \phi_\nu(r_R) + (E - E_\nu) \phi_\nu(r_R) \right\} Y_L(\hat{r}_R) C_{LR}(E) \quad (5)$$

It is interesting to note that the LMTO-ASA basis functions, when written in this form, can be seen as a Taylor series expansion of an energy dependent partial wave.

### 3. SELF-CONSISTENT REAL-SPACE SCHEME

The present LMTO-ASA recursion scheme, solves the eigenvalue problem given above in a self-consistent manner. As in k-space, the problem can be divided in two independent parts. First we find the

structure constant matrix  $\bar{S}$  for the given system. It has been shown that  $\bar{S}$  decreases exponentially with distance and that to find the  $9 \times 9$  matrices connecting each inequivalent atom to its neighbors it is sufficient to invert a cluster of about 20 atoms around each of the atoms. Because the values of  $\bar{Q}$  are given by constants that do not depend on the potential,  $\bar{S}$  does not change during the self-consistent process. Given  $\bar{S}$ , to build the Hamiltonian we should find the potential parameters  $\bar{C}$  and  $\bar{A}$ . They can be found from the orthogonal potential parameters  $C$ ,  $A$  and  $Q$  using Eq. (3). But to obtain  $C$ ,  $A$  and  $Q$  we have to solve the Schrödinger equation inside each non-equivalent WS sphere. This part of the problem is often called "the atomic part" and is treated in the same manner as in  $k$ -space. Actually we use regular LMTO-ASA codes when solving for the "atomic part" in real-space. This part gives all the non trivial information about the potential. Therefore the approximations for the exchange and correlation terms used in real-space are exactly the same as the ones used in regular  $k$ -space LMTO-ASA formalism. The potential inside a WS sphere and therefore the potential parameters, are uniquely determined if we give the occupation for each local ( $s$ ,  $p$  and  $d$ ) band at the site, the first and second moments of the local density of states relative to  $E_\nu$  and the logarithmic derivative of  $\varphi_\nu(r)$  at the sphere boundary<sup>11</sup>. This is so because the spherical average of the charge density inside the sphere is given in terms of the radial part of the solutions of the Schrödinger equation inside the sphere, and the moments of the local density of states (LDOS). With given moments and a guess for the solutions at  $E=E_\nu$  inside the sphere, we can find the charge density. Using Poisson's equation we can find the electrostatic potential. If we

summ the exchange and correlation terms and use the given boundary conditions we can obtain better solutions for the Schrödinger equation inside the sphere and better values for the spherical charge density. We proceed until the potential reaches a final self-consistent value for the given moments and logarithmic derivatives. Here we choose  $E_\nu$  in to keep the first moment of the density of states for the occupied part of the band always zero. To start a self consistent process, we give reasonable guesses for the occupation, second moment and logarithmic derivatives for each inequivalent WS sphere. With this starting guess we find the orthogonal potential parameters. Using Eq. (3) we find  $\bar{C}$  and  $\bar{A}$  and build the real space TB Hamiltonian of Eq. (6). We use the recursion method and a large cluster to obtain the LDOS for  $s$ - $p$  and  $d$  electrons at each inequivalent site. With this knowledge we find the new energy  $E_\nu$  and the new moments for each band at each inequivalent site. As in  $k$ -space, the new logarithmic derivatives are given in terms of new values of  $E_\nu$  and the old potential parameters<sup>8</sup>. We use the new values of the moments and logarithmic derivatives to obtain new values for the orthogonal parameters and new TB parameters  $\bar{C}$  and  $\bar{A}$ . We build a new real space Hamiltonian using expression (5) for fixed  $\bar{S}$  and use the recursion method to obtain the local density of states which will be used for the next iteration. The results will be converged when the moments and logarithmic derivatives obtained by solving the eigenvalue problem differ by less than a previously established amount from the ones which have generated the Hamiltonian. We should note that when we solve for the "atomic part", we choose the potential to be zero at the at the boundary. When building the Hamiltonian we should correct the relative energy scale of each WS sphere by the Madelung energy due to

charged WS spheres of other sites at the given sphere and also take into account, the electrostatic contribution of the sphere itself.

For magnetic systems the procedure is similar, but we have to treat up and down bands separately, when solving for the eigenvalue problem. These bands are generated by Hamiltonians with the same structure constant, but different potential parameters and therefore have different moments and logarithmic derivatives associated with them. When solving for the "atomic part" we use these moments to obtain the spherical charge density associated up and down spins. We use the total charge density when solving Poisson's equation, but the up and down contributions are needed to obtain the exchange and correlation term. In the present work a exchange and correlation term of the form proposed by Barth Heidin<sup>12</sup> is used.

#### 4. RESULTS AND DISCUSSION

To illustrate the procedure we have obtained spin polarized results for ferromagnetic FeNi<sub>3</sub> and antiferromagnetic FeMn. For the FeNi<sub>3</sub> structure we have Fe in the corner and Ni in the faces of a FCC unit cell with lattice constant of 6.620 a.u. For FeMn we have a basic FCC unit, with a lattice constant of 6.850 a.u., and two Fe and two Mn atoms in the basis. We performed self-consistent LMTO-ASA calculations in real-space (with the scheme developed here) and in reciprocal space for the two compounds. In reciprocal space we did two calculations: one using for the Hamiltonian the same approximations that we have used in real-space and the second using the standard LMTO-ASA approach. For calculations in real-space we have used, for both FeMn and FeNi<sub>3</sub>, a large cluster of 1372 atoms. To avoid surface effects, the LDOS was obtained via the recursion method, for an atom close to the center of

the cluster. Here we have used a cutoff parameter  $LL=20$  in the recursion chain<sup>1</sup>. A Beer and Petifor<sup>13</sup> terminator was used to obtain the LDOS and its moments. We note that the precision can be increased by using a larger cutoff parameter  $LL$ .

We have mentioned that the potential within the sphere is governed by the moments of the local density of states and the logarithmic derivatives for up and down bands. We find that the occupation (moment of order zero) is the most sensitive of these quantities and the one that differs the most from corresponding  $k$ -space values<sup>4</sup>. Therefore, to give an idea of the efficiency of the real-space approach, we show in tables I and II, the results for the occupations of up and down bands around Fe and Ni in FeNi<sub>3</sub> and around Fe and Mn in FeMn. We show results from three different calculations: self-consistent real-space results (RS), self-consistent  $k$ -space results with the first-order (FO) Hamiltonian and regular LMTO-ASA  $k$ -space results (FH). We also show the local magnetic moment for Fe and Ni in FeNi<sub>3</sub> and for Fe and Mn in FeMn. Comparing the results of FO and FH we see that second order effects and the inclusion of combined corrections are not very important. Therefore our real-space Hamiltonian should represent the systems well.

For FeNi<sub>3</sub> there are some results the magnetic moment<sup>14,15</sup>. The spin-polarized parametrized LMTO-ASA approach, which uses potential parameters for the pure metals and the Stoner criterion to obtain the electronic structure, works very well for FeNi<sub>3</sub><sup>6</sup>. Our present results are shown in Table I. We can see that the real-space self-consistent approach compares very well with the  $k$ -space results, for which the same FO approximation was used. Occupations of  $s$  and  $p$  electrons differ by a negligible amount and even for  $d$ -bands the differences in

occupation are of order of hundredths of electron. This is the kind of agreement that we expect from our experience with nonmagnetic systems<sup>8</sup>. The magnetic moments compare very well with those obtained in k-space. The parametrized LMTO-ASA approach gives  $2.5\mu_B$  for Fe and  $.76\mu_B$  for Ni. A DVM cluster calculation<sup>15</sup> with 19 atoms gives  $3.1\mu_B$  for Fe and  $.82\mu_B$  for Ni. All these values are rather close and it is clear that the present real-space approach is quite competitive.

FeMn is more interesting. While the magnetic moments in  $FeNi_3$  is quite stable, the moments in FeMn are very sensitive to small variations of the lattice constant<sup>14</sup>. The parametrized LMTO-ASA approach in this case, clearly indicates that the system will be antiferromagnetic, if any magnetism should be present. But the parametrized calculation slowly converges to a non-magnetic state. The FeMn moment is very sensitive to details of the calculation and should be an interesting test the method. If we compare the RE and FO occupation values we see that s and p bands are in very good agreement, but the d-bands differ by 0.1 electron. This discrepancy is small, but is larger than the one expected on basis of previous results for nonmagnetic systems. It leads to values of magnetic moments which are less accurate than in the case of  $FeNi_3$ . Even so the results are quite good considering the difficulty of the problem. We have investigated the reasons for the failure of the parametrized approach in FeMn. In the parametrized approach<sup>6</sup> we use the Stoner criterion where the d-band shift should be equal to the magnetic moment times the Stoner parameter (extracted from pure metals). Using the d-band shifts and d-band magnetic moments of the RS calculations we would need a decrease of 8% on the Stoner parameter of Fe and a decrease of 17% on the Stoner

parameter of Mn to stabilize the moments. We also noticed that the parameter  $\bar{A}$  which regulates the band width is different for the up and down bands. In the parametrized approach, a single value of  $\bar{A}$  is used for both bands. We note that if we use in the converged calculations an average value of  $\bar{A}$ , between the up and down values, the magnetic moments decrease. The case of FeMn illustrates why methods involving parameters are not reliable. Small differences in these parameters can determine whether the system is magnetic or not. Here we have shown that even when the system is very sensitive to parameters, the self-consistent real-space approach is reliable.

#### V. CONCLUSIONS

We have developed a first-principles self-consistent real-space method that can be used to study the electronic structure of complex metallic systems. The method was tested for simple crystalline  $FeNi_3$  and FeMn, where k-space results can be obtained. But it does not make use of symmetry and can be applied to non-periodic systems. To illustrate the advantage of the present approach let us consider the problem of a magnetic substitutional impurity on a non magnetic host. The cost of the real-space method grows linearly with the number of non-equivalent atoms. But only the density of states is used, atoms with same LDOS are considered equivalent. Therefore to study the influence of the impurity over four shells of neighbors we only need to consider seven non-equivalent atoms, and the cost of the calculation is just seven times that of a mono atomic crystal! It is clear that we can also easily treat magnetic interaction between impurities.

In conclusion, we have presented a new approach to the study of spin-polarized electronic structure in complex metallic systems. The



method is competitive and its potential should be further investigated.

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They have performed spin-polarized LMTO band structure calculations as a function of lattice parameter to a series of ternary FCC Mn-Fe-Ni systems, including FeNi<sub>3</sub> and FeMn. Our k-space results agree well with theirs for the given lattice parameter.

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FeNi <sub>3</sub>		Fe			Ni		
		RS	FO	FH	RS	FO	FH
↑	s	0.325	0.327	0.320	0.333	0.332	0.325
	p	0.368	0.376	0.377	0.355	0.356	0.366
	d	4.664	4.671	4.656	4.667	4.631	4.602
↓	s	0.320	0.321	0.311	0.354	0.354	0.342
	p	0.378	0.382	0.383	0.416	0.416	0.416
	d	1.782	1.750	1.783	3.929	3.969	4.006
mag. mom.		2.877	2.921	2.876	0.656	0.580	0.529

TABLE I: Results for the occupation of up (↑) and down (↓) spins for the real-space method (RS), k-space with first order Hamiltonian (FO) and with the full LMTO-ASA Hamiltonian (FH). Magnetic moments in  $\mu_B$  are also shown.

FeMn		Fe			Mn		
		RS	FO	FH	RS	FO	FH
↑	s	0.350	0.352	0.341	0.319	0.315	0.307
	p	0.421	0.431	0.428	0.378	0.376	0.374
	d	3.929	4.068	4.028	2.071	1.961	2.013
↓	s	0.332	0.330	0.320	0.339	0.337	0.325
	p	0.394	0.400	0.400	0.424	0.428	0.418
	d	2.588	2.449	2.515	3.453	3.553	3.530
mag. mom.		1.386	1.672	1.562	-1.448	-1.666	-1.579

TABLE II: Results for the occupation of up (↑) and down (↓) spins for the real-space method (RS), k-space with first order Hamiltonian (FO) and with the full LMTO-ASA Hamiltonian (FH). Magnetic moments in  $\mu_B$  are also shown.