Implementation of the LDA+U method using the full potential linearized augmented plane wave basis

A. B. Shick\textsuperscript{a}, A. I. Liechtenstein\textsuperscript{b} and W.E. Pickett\textsuperscript{a}

\textsuperscript{a}Department of Physics, University of California, Davis, CA 95616

\textsuperscript{b}IFF, Forschungszentrum Jülich, Germany

We provide a straightforward and efficient procedure to combine LDA+U total energy functional with the full potential linearized augmented plane wave method. A detailed derivation of the LDA+U Kohn-Sham type equations is presented for the augmented plane wave basis set, and a simple “second-variation” based procedure for self-consistent LDA+U calculations is given. The method is applied to calculate electronic structure and magnetic properties of NiO and Gd. The magnetic moments and band eigenvalues obtained are in very good quantitative agreement with previous full potential LMTO calculations. We point out that LDA+U reduces the total d charge on Ni by 0.1 in NiO.

I. INTRODUCTION

The limitations of the local density approximation (LDA) in calculations of electronic and magnetic ground state properties of strongly correlated materials are well known. One of the simplest methods for going beyond LDA is provided by so-called “LDA+U” total energy functional which has proven to give large but reasonable corrections for a number of magnetic insulators. (Throughout this paper we understand LDA to include its generalization to spin polarized systems.) The method presumes that an appropriate set of local orbitals \{\phi_m\} (such as 3d or 4f) can be identified. Then a strong intra-atomic interaction is introduced and treated in a Hartree-Fock-like manner; in particular, the intra-atomic exchange term is treated without any local density approximation. A primary result is the splitting apart of occupied and unoccupied states within the \{\phi_m\} shell.

Technically, the explicit use of local orbitals and an orbitally dependent correction to the energy functional and to the LDA effective potential makes it most convenient to implement the LDA+U within a method using atomic-like orbitals as basis functions. This is true for the linearized muffin-tin orbital method (LMTO) in atomic sphere approximation (ASA) or with its full potential version, both of which are formulated in terms of atom-based orbitals, and most LDA+U work has been done with the LMTO-ASA method. Recently the implementation of LDA+U in the pseudopotential plane wave method was presented by Sawada by projecting the plane waves onto an atomic orbital.

The LDA+U method has not yet been implemented within an all-electron, full potential method whose basis set is not based explicitly on local orbitals. The linearized augmented plane wave (LAPW) method, which makes no shape approximation and is acknowledged to be state-of-the-art in accuracy, uses a basis set of plane waves that are matched onto a linear combination of all radial solutions (and their energy derivative) inside a sphere centered on each atom. Hence the basis functions are nothing like individual atomic orbitals. It is important to establish that the LDA+U method is not limited to certain basis sets, but rather is more widely applicable. The full potential aspect may also become important in delicate situations, such as at the onset of orbital ordering.

In this paper we present a simple and numerically efficient procedure to combine the LDA+U total energy functional with the full potential linearized augmented plane wave (FLAPW) method. As an example of its efficacy we apply our implementation to study the effects of strong on-site repulsion on the magnetic insulator NiO and the ferromagnetic metal Gd and compare with previous work on these materials.

The paper is organized as follows. For the sake of completeness, in Sec. II we recall the basic equations of LDA+U method. Then we address the fact that the LAPW method is not a local orbital basis set approach. We show that we can define the on-site orbital occupation matrix and formulate a variational procedure to obtain Kohn-Sham single particle equations. We also describe a “second variation” procedure that may provide additional insight in analyzing the results. In Sec. III we present the results of self-consistent total energy LDA+U FLAPW calculations for the antiferromagnetic insulator NiO and for Gd metal. These results are compared with previous work, and additional features are pointed out.

II. COMPUTATIONAL METHOD

The essence of the LDA+U method is to identify atomic-like orbitals \{\phi_m\} (m is the magnetic quantum number, we will often suppress other indices) and to treat interactions of electrons in these orbitals in a non-LDA manner. The LDA+U variational total energy functional takes the form:

$$E^{\text{tot}}(\rho, \hat{n}) = E^{LDA}(\rho) + E^{ee}(\hat{n}) - E^{dc}(\hat{n}),$$

where, $E^{LDA}(\rho)$ is usual local spin density functional of the total electron spin densities $\rho^\sigma(\mathbf{r})$ ($\sigma = \uparrow, \downarrow$). $E^{ee}$ is an electron-electron interaction energy and $E^{dc}$ is a “double-counting” term which accounts approximately for an electron-electron interaction energy already included in $E^{LDA}$. Both are functions of the local orbital occupation matrix $\hat{n}^\sigma = n^\sigma_{m,m'}$ of the orbitals \{\phi_m\}.

A particular form of $E^{ee}$ is taken in accordance to the multi-band Hubbard model for d(f) electrons:

$$E^{ee} = \frac{1}{2} \sum_{m_1,m_2,m_3,m_4} n^\sigma_{m_1,m_2} \left( < m_1,m_3 | V^{ee} | m_2,m_4 > - < m_1,m_3 | V^{ee} | m_4,m_2 > \delta_{\sigma,\sigma'} n^\sigma_{m_3,m_4} \right).$$


where the $V^{ee}$ is an effective on-site “Coulomb” interaction and $< | >$ indicates an angular integral.

The electron-electron interaction potential in the atomic limit is given by

$$< m_1, m_3 | V^{ee} | m_2, m_4 > = \sum_k a_k (m_1, m_3, m_2, m_4) F_k$$  \hspace{1cm} (3)

$$a_k (m_1, m_3, m_2, m_4) = \frac{4\pi}{2k + 1} \sum_{\gamma = -k}^{k} < lm_1 | Y_{k\gamma} | lm_2 > \times < lm_3 | Y_{k\gamma}^* | lm_4 >$$

where $F_k$ are the Slater integrals, $|lm>$ is d(f)-spherical harmonic, and $n_{m_1, m_2}^\sigma$ is the on-site d(f) occupation matrix in the spin-orbital space, which has to be defined with respect to the chosen localized orbital basis set. Note that in $E^{ee}$ in Eq.(2) the self-interaction in the first term $(m_1 = m_2 = m_3 = m_4$ and $\sigma = \sigma')$ is cancelled exactly by the exchange interaction in the second term. This is one of the important features of the LDA+U approach. The “double-counting” term is taken to satisfy an “atomic-like” limit of the LDA total energy

$$E^{dc} (\hat{n}) = \frac{U}{2} (n - 1) - \frac{J}{2} \sum_{\sigma} n^\sigma (n^\sigma - 1)$$  \hspace{1cm} (4)

where the Hubbard $U$ and exchange $J$ constants are given by

$$U = \frac{1}{(2l + 1)^2} \sum_{m_1, m_3} < m_1, m_3 | V^{ee} | m_1, m_3 > ,$$

$$J = U - \frac{1}{2(2l + 1)} \sum_{m_1, m_3} < m_1, m_3 | V^{ee} | m_1, m_3 > - < m_1, m_3 | V^{ee} | m_3, m_1 > .$$

In Eq.(4), $n^\sigma = Tr \hat{n}^\sigma$, and $n = \sum_\sigma n^\sigma$ is a total d(f) on-site occupation numbers. The energy Eq.(1) is a functional with respect to the spin density

$$\rho^\sigma (r) = \sum_i f_i \phi_i^\sigma (r) \phi_i^\sigma (r)$$  \hspace{1cm} (6)

and the local orbital occupation matrices $\hat{n}^\sigma$.

### A. Density matrix definition within the LAPW basis

It is rather straightforward to choose an orbital basis for Eq.(3) in the case of the LMTO method as an orthogonal atomic-type LMTO basis set. In the case of the LAPW method there are no naturally defined atomic-like basis orbitals. Therefore, we will use a projection of augmented plane waves onto radial functions inside muffin-tin spheres to define the density matrix for Eq.(3).

Let us recall some basic features of LAPW method (we will use the notations of Singh). The LAPW basis function $\phi_{k+G}^\sigma (r)$ is a plane wave $\exp (i(k + G) \cdot r)$ outside MT sphere, and inside sphere $i$ $(r_i = r - R_i)$ it is a linear combination of all radial functions

$$\phi_{k+G}^\sigma (r) = \sum_{l,m} [a_{lm}^{k+G} u_l^\sigma (r_i) + b_{lm}^{k+G} u_l^\sigma (r_i)] Y_{lm}(\hat{r}_i)$$  \hspace{1cm} (7)

where $u_l^\sigma$, $u_l$ are the radial wavefunction and its energy derivative at an appropriate energy $E_l$ for angular quantum number $l$ (recall $< u_l | u_l > = 1$, $< u_l | u_l > = 0$), $k$ is a k-point in BZ, and $G$ is any reciprocal lattice vector.

The solution of the Kohn-Sham equations is a set of eigenfunctions

$$\Phi_k^{l,\sigma} (r) = \sum_G b_{lm}^{k+G} \phi_{k+G}^{l,\sigma} (r)$$  \hspace{1cm} (8)

where $b$ stands for band index and the sum runs over reciprocal lattice vectors. The charge density is

$$\rho^\sigma (r) = \sum_{k,b} f_{kb} | \Phi_k^{b,\sigma} (r) |^2$$  \hspace{1cm} (9)

where $f_{kb}$ is the occupation of the state. In the $i$-th sphere, from Eqs. (8) and (9) the spin density

$$\rho^\sigma (r) = \sum_{k,b} f_{kb} \sum_{G,G'} c_{k+G,G,b,\sigma}^{b,\sigma} \times \sum_{l,l',\sigma} [a_{lm}^{k+G} u_l^\sigma (r_i) + b_{lm}^{k+G} u_l^\sigma (r_i)] [a_{l'm}^{k+G} u_{l'}^\sigma (r_i) + b_{l'm}^{k+G} u_{l'}^\sigma (r_i)] Y_{lm}(\hat{r}_i) Y_{l'm}(\hat{r}_i).$$

Taking the elements of the density matrix from the projection of the wavefunctions onto the $Y_{lm}$ subspace gives

$$n_{lm,l'm'} = \sum_{k,b} f_{kb} \sum_{G,G'} c_{k+G,G,b,\sigma}^{b,\sigma} \times \int d\tau \tau^2 [a_{k+G,G,b}^{l,m} u_{l'}^\sigma (r_i) + b_{k+G,G}^{l,m} u_{l'}^\sigma (r_i)] [a_{k+G,G}^{l,m} u_{l'}^\sigma (r_i) + b_{k+G,G}^{l,m} u_{l'}^\sigma (r_i)]$$  \hspace{1cm} (10)

Such a projection gives the desired result (i.e. in agreement with other implementations) in the atomic limit, and its diagonal elements give what are commonly thought of as local orbital occupation factors.

This expression indicates that the density matrix is not necessarily diagonal in $l$ but this detail is not expected to be important. Keeping only the $l' = l$ part and suppressing the $l$ index on the density matrix, after radial integration Eq.(11) becomes

$$n_{m,m'}^{\sigma} = \sum_{k,b} f_{kb} \sum_{G,G'} c_{k+G,G,b,\sigma}^{b,\sigma} \times \int d\tau [a_{k+G,G,b}^{l,m} a_{k+G,G}^{l,m} + b_{k+G,G}^{l,m} b_{k+G,G}^{l,m}] < \hat{u} | \hat{u} >$$  \hspace{1cm} (12)

Finally, one can rewrite Eq. (12) as

$$n_{m,m'}^{\sigma} = \sum_{k,b} f_{kb} < \hat{u}^\sigma Y_{lm} | \Phi_k^{b,\sigma} > < \Phi_k^{b,\sigma} | a_{k+G,G}^{l,m} Y_{lm} > +$$  \hspace{1cm} (13)

$$< \hat{u}^\sigma Y_{lm} | \Phi_k^{b,\sigma} > < \Phi_k^{b,\sigma} | \hat{u}^\sigma Y_{lm} > / < \hat{u} | \hat{u} >$$

where $\hat{u}$ is a radial wavefunction and its energy derivative at an appropriate energy $E_l$ for respective radial quantum number $l$. $k$ is a k-point in BZ, and $G$ is any reciprocal lattice vector.
B. Variational principle.

Using Eq.(13) and orthonormality of the Φ’s one can minimize Eq.(1) with respect to Φ_i^{a,\alpha}:

\[ \frac{\delta E}{\delta \Phi_i^{a,\alpha}} = \delta \sum_j c_{ij} d_j^a <\Phi_j^{a,\alpha} | \Phi_i^{a,\alpha}> = 0 \]  

(14)

It gives the set of Kohn-Sham equations, which can be written in the form

\[ \left( -\nabla^2 + V^{\sigma}_{LDA}(r) \right) \Phi_i^{\sigma}(r) + \sum_{m,m'} V^{\sigma}_{m,m'} \frac{\delta n^{\sigma}_{m,m'}}{\delta \Phi_i^{\sigma}} = \epsilon_i^{\sigma} \Phi_i^{\sigma}(r) \]  

(15)

where an effective potential acting on the Y_{im} subspace (the d(f-states)) is

\[ V^{\sigma}_{m,m'} = \sum_{p,q,\sigma'} \left( <m,p|V^{\text{ee}}|m',q> - <m,p|V^{\text{xc}}|q,m'> \delta_{\sigma,\sigma'} \right) \delta_{\sigma'} + \delta_{m,m'} U(n - \frac{1}{2}) + \delta_{m,m'} J(n^\sigma - \frac{1}{2}) \]  

and from Eq.(13)

\[ \frac{\delta n^{\sigma}_{m,m'}}{\delta \Phi_i^{\sigma}} = <u_i^{\sigma} Y_{im}|\Phi_i^{\sigma}> u_i(r) Y_{im}(r) + <u_i^{\sigma} Y_{im}|\Phi_i^{\sigma}> u_i(r) Y_{im}(r)/ <\Phi_i^{\sigma}|\Phi_i^{\sigma}> = \left[ u_i^{\sigma} Y_{im} > u_i^{\sigma} Y_{im} + \frac{1}{2} \right] \]  

(16)

(17)

Euqns (15)-(17) indicate specifically how the additional potential that arises in the LDA+U method acts on the projections of the wavefunctions Φ onto the Y_{im} subspace.

C. Use of “second variation” procedure.

The Eq.(15) can be solved directly using the LAPW basis set Eq.(7). For large systems it can be solved more efficiently using a “second variation” procedure. We define an auxiliary orthogonal basis set Ψ_k^{b,\sigma}(r) as the solutions of LDA band Hamiltonian

\[ \left( -\nabla^2 + V^{b}_{LDA}(r) \right) \Psi_k^{b,\sigma}(r) = \epsilon_k^{b,\sigma} \Psi_k^{b,\sigma}(r) \]  

(18)

The functions Ψ_k are somewhat like the LDA eigenfunctions, but they are not identical because it is not the LDA density that goes into V^{b}_{LDA}. Expanding the functions Φ via Ψ (suppressing k and shortening notation)

\[ |\Phi^i> = \sum_j d_j^i |\Psi^i> \]  

(19)

Eq.(15) is then transformed to the following,

\[ \sum_j \epsilon_j d_j^i |\Psi^i> + \sum_{j} d_j^i \sum_{m,m'} V^{\sigma}_{m,m'} <u_i^{\sigma} Y_{im}|\Psi^i> \times \]  

(20)

\[ u_j^{\sigma}(r) Y_{im}(r) + <u_j^{\sigma} Y_{im}|\Psi^i> u_j^{\sigma}(r) Y_{im}(r)/ <\Psi^i|\Psi^i> \]  

\[ = \epsilon_i d_j^i |\Psi^i> \]

and using <Ψ_i|Ψ_j> = δ_ij we obtain the “second variation” Hamiltonian \[ \sum_j H_{ij}^\sigma d_j^i = \epsilon_i d_j^i \] of the form:

\[ H_{ij}^\sigma = \epsilon_j \delta_{ij} + \sum_{m,m'} <\Psi_i^\sigma|u_i^{\sigma} Y_{im}> V^{\sigma}_{m,m'} <u_i^{\sigma} Y_{im}|\Psi_j^\sigma> + \]  

(21)

\[ <\Psi_i^\sigma|u_i^{\sigma} Y_{im} > V^{\sigma}_{m,m'} <u_i^{\sigma} Y_{im}|\Psi_j^\sigma> / <\Psi_i^\sigma|\Psi_i^\sigma> \]

Now, given some plane wave cutoff as usual, the self-consistent solution of Eq.(15) is performed as follows. (i) Solving Eq.(18) for the given LDA effective potential we obtain the entire orthogonal basis set \{Φ\}; (ii) within this basis set we solve Eq.(21) to obtain the coefficients \( d_j^i \) of the LDA+U wavefunctions in terms of the auxiliary wavefunctions in Eq.(19); (iii) the new LDA+U wavefunction is then projected back to LAPW basis set (cf., Eq.(7)) and the usual procedure to calculate charge and spin densities Eq.(10) and occupation matrices Eq.(12) is then used to achieve self-consistent solution.

D. Total energy evaluation

After the self-consistent solution of Eq. (15) the LDA+U total energy can be calculated from Eq. (1), using:

\[ E^{\text{LDA}}(\rho) = T_s(\rho) + \int dv_{\text{ext}}(r)\rho(r) + E_{\text{H}}(\rho) + E_{\text{xc}}(\rho) \]  

(22)

where \( T_s \) is the kinetic energy for non-interacting electrons with density \( \rho \), \( v_{\text{ext}} \) is an external potential (including the interaction with nuclei), \( E_{\text{H}} \) is the Hartree energy and \( E_{\text{xc}} \) is an exchange-correlation energy. It is convenient to use, as usual, the eigenvalue sum, which includes \( T_s \) and the interaction with nuclei exactly but miscounts other terms. A so-called “double-counting” correction \( E^{\text{dc}}_{\text{LDA}} \) leads to the correct energy.

For our LDA+U calculations, the eigenvalue sum contains additional terms (involving U and J) whose contributions to the energy are miscounted. When these terms are corrected, the expression for the energy is:

\[ E^{\text{tot}}(\rho,\tilde{n}) = \sum_i \epsilon_i - E^{\text{dc}}_{\text{LDA}}(\rho) - E^{\text{dc}}_{\text{LDA}+U}(\tilde{n}) \]  

(23)

where, \( E^{\text{dc}}_{\text{LDA}} \) is a “double-counting” LDA correction, and \( E^{\text{dc}}_{\text{LDA}+U} \) is a “double-counting” correction to the eigenvalue sum in LDA+U calculations. This correction is expressed in terms of electron-electron interaction energy Eq. (2) and “double-counting” term Eq.(4) as

\[ E^{\text{dc}}_{\text{LDA}+U}(\rho,\tilde{n}) = E^{\text{xc}}(\tilde{n}) - E^{\text{dc}}(\tilde{n}) - \frac{1}{2} (U - J) \tilde{n} \]  

(24)

where, the last term appears due to a specific form of the LDA+U “double counting” energy Eq. (4) with an explicit removal of self-interaction.
The LDA+U expression for the energy (Eq. (1)) has been used very sparingly except to generate the Kohn-Sham equations which lead to the self-consistent density and orbital occupation matrices. Partly this has been due to the implementation of the LDA+U in the LMTO-ASA method, which makes severe approximations to the shape of the potential and density and for the variational freedom of the wavefunctions, which translates into questionable ability to resolve energy differences. Another feature of the LDA+U energy functional that has not been emphasized is that it explores a regime of $E_{LDA}[\rho]$ which is not understood, by evaluating $E_{LDA}$ for densities that do not minimize it. This procedure deserves scrutiny, so we will give attention to the energy differences in the following section.

III. RESULTS

As representative systems to illustrate the numerical procedure we have proposed, we choose the antiferromagnetic insulator NiO and Gd metal, which has a ferromagnetic ground state. For both of them LDA fails to provide the correct electronic and magnetic ground states due to significant electron correlation effects.

A. NiO

LDA-ASA calculations of antiferromagnetic (AFII) NiO gives the value of the band gap and magnetic moment in disagreement with experimental data (cf., Table I). While the band gap requires an energy dependent self-energy to obtain rigorously and therefore is not a true test of LDA+U corrections, the magnetic moment is a fundamental test. Generally, it is expected that an improved energy functional will also give a band gap in closer agreement with experiment than the very poor LDA value. Our FLAPW LDA calculations give somewhat larger values of both moment and gap than does the ASA calculation (Table II), reflecting the approximations in the shape of the density and potential, and the less flexible basis functions in the ASA. For NiO, we choose the lattice constant as 7.927 a.u. in accordance to the experimental data. For self-consistency, 110 special k-points in the irreducible part of the AFII BZ were used, with Gaussian smearing of 2 mRy of the Fermi energy to promote convergence. The sphere radii $R_{Ni}^{MT} = 2.0$ a.u. and $R_{MT}^{MT} = 1.8$ a.u. were used, and $R_{MT}^{MT} \times K_{max} = 6.6$ determined the basis set size. Literature values $U = 8.0$ eV and $J = 0.95$ eV were used to obtain the values of Slater integrals in Eq.(3) ($F_0 = 8.00$ eV, $F_2 = 8.19$ eV, $F_4 = 5.11$ eV).

The calculated values of spin magnetic moment and band gap are shown in Table III and are within about 5% of recent full-potential LMTO based LDA+U calculations. The calculated projected densities of states (PDOS) for Ni d states and O p states are shown in Fig. 1 and are very similar to these of Ref. The $t_{2g}$ and $e_{g}$ PDOS for Ni d states for LDA+U are compared with those for LDA in Fig. 2. The LDA+U calculations give a charge transfer type band gap ($\approx 3.4$ eV) between mainly O p states at the top of valence band and primarily Ni $e_g$ states in the conduction band as opposed to the LDA calculations where a small band gap (0.4 eV) is formed between mainly Ni d states. There is a large downward shift of majority Ni $e_g$ states (cf. Fig. 2) and the resulting energy difference between $e_g^R$ and $e_g^J$ states ($\approx 4.8$ eV) is not far from model GW calculations ($\approx 9$ eV). For the symmetry of the Ni ion, the occupation matrix is specified by two numbers (for each spin), the $e_g$ and $t_{2g}$ occupancies. The LDA+U correction results in a decrease in the Ni d occupation by 0.12 electrons, with an increase in spin majority charge of 0.2 electrons (to essentially 100%) more than compensated by a decrease by 0.3 electrons in spin minority charge (nearly all of it $e_g$). This difference of d charge is potentially important for the understanding of correlation in magnetic insulators.

Since we take into account only the muffin-tin part of LDA+U correction, it is important to analyze numerically the stability of our results with respect to the choice of the MT radii. We have varied the sphere radii for both Ni and O atoms in the unit cell and found that these changes have little effect on the calculated values of magnetic moment and band gap (cf. Table III), due to the fact that the sphere contains all but a few percent of the Ni d states.

B. Gd

A detailed analysis of ground state and magnetic properties of ferromagnetic Gd has been presented by Singh. It was shown that Gd 4f-electrons have to be considered as "band" electrons instead of "core" to obtain at least qualitative agreement with experimentally observed Fermi-surface. (The "core" treatment consists of treating the 4f states as corelike, nondispersive bands that do not hybridize with the valence bands, and removing the 4f character from the valence electron basis set.) Heinemann and Temmerman found with the LMTO-ASA method that using GGA rather than LDA led to a ferromagnetic state being favored over the AF state. The more recent full potential LMTO calculations of Harmon et al. did not confirm this conclusion. Instead, it was shown that only LDA+U gives a correct ferromagnetic ground state for Gd.

For Gd, we chose the lattice constant as 6.8662 a.u. and $c/a$ ratio as 1.557 in accordance with the experimental crystal structure data. Here, 150 special k-points in irreducible part of the hexagonal BZ were used, again with Gaussian smearing. The values $R_{MT} = 3.2$ a.u. and $R_{MT} \times K_{max} = 9.6$ were used. Literature values of Ref. $U = 6.7$ eV and $J = 0.7$ eV were used to obtain the values of Slater integrals in Eq.(3) ($F_0 = 6.70$ eV, $F_2 = 8.34$ eV, $F_4 = 5.57$ eV, $F_6 = 4.13$ eV).

The spin magnetic moment for FM and AFM Gd are shown in Table III. The LDA result was in nearly perfect agreement with experiment, and the 2% increase given by LDA+U does not degrade the agreement much. The energy difference $E(AFM) - E(FM)$ is negative in LDA calculations and positive in LDA+U calculations. Our results therefore confirm quantitatively the conclusion of Ref. that the correct FM result is only predicted by LDA+U. The LDA and LDA+U partial f-densities of states are shown in Fig. 3. There is a large (4.5 eV) shift of spin-majority f-states below the Fermi level and re-
The removal of the unoccupied spin-minority f-states from close vicinity of the Fermi level due to an upward shift of 1.5 eV. The enhancement of an exchange splitting can be understood as an increase of an effective exchange splitting parameter $I = (U + 6J)/7 = 1.5$ eV in mean-field Hubbard model in comparison with LDA ($I = J = 0.7$ eV) due to Hubbard $U$. The exchange splitting $(IM)$ for f-states is then increases from 5 eV with LDA to 11 eV with LDA+U (cf. Fig.3). A more complete discussion of the LDA+U description of Gd will be given elsewhere.

IV. CONCLUSION

To summarize, we have presented a straightforward and numerically efficient procedure to combine the LDA+U total energy functional with the FLAPW method. This implementation is all-electron and includes no shape approximations. In spite of a modest limitation – we use the only “muffin-tin” part of the LDA+U correction – the method works well for representative systems NiO and Gd for the electronic spectrum and predicts the correct magnetic order. It also can be easily extended to incorporate spin-orbit interaction. Moreover, a similar numerical procedure can be used to implement the “dynamic mean field” parametrization within the FLAPW method as a more realistic model for the self-energy of metallic strongly correlated systems.

We have emphasized the eigenvalue shifts in NiO and in Gd that result from the strong repulsion in the LDA+U method. These shifts are probably important in obtaining an improved description. However, it should be kept in mind that the LDA+U method is based on the energy functional for the electronic ground state, and that eigenvalues are not the most fundamental quantities for judging the success of the LDA+U method. It is satisfying that the LDA+U energies for Gd lead to the observed magnetic ground state.

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TABLE I. The spin magnetic moments ($M_s$ in $\mu_B$) and band gap ($E_g$ in eV) for NiO. The increases in both quantities are about the same in the FLAPW and LMTO-ASA methods, although the individual value differ due to approximations made in the LMTO-ASA method.

| Method     | FLAPW | LMTO-ASA | FLMTO | Exp. |
|------------|-------|----------|-------|------|
| LDA        | 1.186 | 1.687    | 1.0   | 1.56 |
| LDA+U      | 0.41  | 3.38     | 0.2   | 3.1  |

$M_s$ 1.0 - 1.70

TABLE II. The spin magnetic moments ($M_s$ in $\mu_B$) and band gap ($E_g$ in eV) for different “muffin-tin” radii (a.u.) for Ni and O atoms in NiO. The small differences are entirely unimportant.

| Atom | FLAPW | LMTO-ASA | FLMTO | Exp. |
|------|-------|----------|-------|------|
| Ni   | 2.0   | 2.1      | 2.1   |      |
| O    | 1.8   | 1.8      | 1.7   |      |

$M_s$ 1.687 - 1.700

$E_g$ 3.38 - 3.38

TABLE III. The spin moments (in $\mu_B$) and the total energy difference (in meV) between antiferromagnetic (AFM) and ferromagnetic (FM) phases for the bulk Gd.

| Method     | LDA | LDA+U | Exp. |
|------------|-----|-------|------|
| FM         | 7.659 | 7.818 | 7.63 |
| AFM        | 7.246 | 7.437 |      |

$E(AFM-FM)$ per atom, meV 63 - 82

GGA-ASA +1.4

FIG. 1. The partial DOS for Ni d-states (filled) and for O p-states for NiO. Majority is plotted upwards; minority is plotted downward.
FIG. 2. The partial DOS for Ni $t_{2g}$ (filled) and $e_g$ states for LDA+U (a) and LDA (b) for NiO. The increase in the exchange splitting of the Ni d states due to LDA+U is dramatic.

FIG. 3. The total DOS (full line), the partial DOS for Gd f-states for LDA+U (filled) and LDA (dotted) for the ferromagnetic Gd. The 4f exchange splitting is increased from 5 eV to 11 eV by LDA+U. Majority is plotted upwards; minority is plotted downward.