# I. ASA CONSTRUCTION OF THE LDA+U POTENTIAL

Implementation of the LDA+U hamiltonian has some ambiguities. Usualy implementations add a spin-dependent, on-site potential at site $R$, for a given $l$ partial wave. The potential need not be diagonal in $m$. This potential is not explicit; instead its matrix element $V^U_{Rl\sigma;m,m'}$ are computed from the density-matrix and $U$ parameters.

There is some ambiguity about how this parametrized potential is to be folded into the ASA hamiltonian. The dominant part of $V^U_{Rl\sigma;m,m'}$ is usually the diagonal part, $V^U_{Rlm\sigma} \equiv V^U_{Rl\sigma;m,m}$. If we assume $V^U_{Rl\sigma;m,m}$. If we assume $V^U_{Rlm\sigma}$ simply adds constant potential shift for a particular $Rlm\sigma$, the modification of the ASA hamiltonian to incorporate $U$ becomes well defined. It means that the shape of the radial wave function does not change; only its energy is shifted by a constant $V^U_{Rlm\sigma}$.

In the diagonal-only approximation, the modification of the ASA hamiltonian reduces to a modification of the potential parameters. We can write a modificatin of the ASA-LDA hamiltonian that is independent of representation. For a given spherical potential, the ASA-LDA potential is parameterized in terms of the five "potential parameters," the linearization energy $\varepsilon_{\nu RL}$, the band center $C_{RL}$, the bandwidth parameter $\Delta_{RL}$, $\gamma_{RL}$ which specifies the transformation to the orthogonal basis, and the "small" parameter $p_{RL} = \left\langle \dot{\phi}_{RL} | \dot{\phi}_{RL} \right\rangle$. Moreover, these parameters are independent of $m$. In the second-order ASA form, it is (suppressing indices)

$$H = C + \sqrt{\Delta} S^\gamma \sqrt{\Delta} \tag{1}$$

$$S^{\gamma-1} = S^{0-1} - \gamma^{-1} \tag{2}$$

According to Andersen's conventions, the ASA hamiltonian can be written in a general screened representation parameterized by screening parameters $\alpha_{RL}$. A 3-center version of above equations, which apply for $\alpha_{RL} = \gamma_{RL}$, can be used for any $\alpha_{RL}$. It has the same formal structure, provided $C$, $\Delta$, and $p$ are redefined. $C$, $\Delta$, and $p$ correspond to the $\gamma$, or "orthogonal" representation, where in the 2-center ASA form of it, the overlap matrix is unity. In a general $\alpha$ representation, they become $C^\alpha$, $\Delta^\alpha$, and $p^\alpha$ . In the Varenna notes, p88, Anderson shows the following relations:

$$\frac{\sqrt{\Delta^\alpha}}{\sqrt{\Delta}} = 1 - (\gamma - \alpha)\frac{C - \varepsilon_\nu}{\Delta} \tag{3}$$

$$= \frac{C^\alpha - \varepsilon_\nu}{C - \varepsilon_\nu} \tag{4}$$

$$\frac{1}{o^\alpha} = C - \varepsilon_\nu - \frac{\Delta}{\gamma - \alpha} \tag{5}$$

$$= -(C^\alpha - \varepsilon_\nu) + \frac{\Delta^\alpha}{\alpha - \gamma} \tag{6}$$

$$p^\alpha = p + (o^\alpha)^2 \tag{7}$$

Through Eq. (3) $\Delta^\alpha$ is generated in terms of $C$, $\varepsilon_\nu$ and $\Delta$; Eq. (4) generates $C^\alpha$. A key point of the full 3-center hamiltonian is that the spectrum of $H$ is exactly independent of the choice for $\alpha_{RL}$.

Ideally we would like to construct an LDA+U hamiltonian that is independent of representation. As a practical matter, it is valuable for connecting Green's function LDA+U and hamiltonian LDA+U, and also means we can move away from the $\gamma$-representation, which is slow and doesn't permit downfolding. It provides an unambiguous prescription for incorporating third-order contributions into the hamiltonian.

It turns out that this can be accomplished, but under certain restrictions. Program lm has two ways computing energy moments of the output density, controlled by input parameter `QASA`. In the

Methfessel style (`QASA=0`), the output density is accumulated in terms of true power moments. This way relies on a connection between the (linearized) KKR and LMTO methods, that is the relation between the energy-dependent KKR and energy-independent LMTO basis sets. Alternatively, once the LMTO basis is defined, the power moments can be defined independently of any relation to the KKR method (`QASA=0`). This is the preferred method, but the disadvantage is that the relation between GF and LMTO methods is lost. The GF method can only accumulate the output density in terms of true power moments.

There is no difficulty in the special case when $V^U_{R\sigma;m,m'}$ is diagonal in $m$. If we assume that $V^U_{RL\sigma} = V^U_{Rl\sigma;m,m'}\delta_{m,m'}$ is spatially constant, but spin-and $m$-dependent potential shift in channel $RL$, it means that the band center and linearization energy $C_{RL}$ and $\varepsilon_{\nu RL}$ should shift by $V^U_{RL\sigma}$. The other parameters $\Delta_{RL}$, $\gamma_{RL}$ and $p_{RL}$, and also $C_{RL} - \varepsilon_{\nu RL}$ should not change, since they are calculated from the wave function $\phi$ and its derivatives on the MT boundary ($p$ is calculated from an integral of wave functions). Eq. (3) shows that $\Delta^\alpha$ is independent of the addition of $V^U_{Rlm\sigma}$ to both $C$ and $\varepsilon_\nu$; Eqs. (3) and (4) together show that $C^\alpha$ also does not change. Finally, Eq. (6) shows explicitly that $p^\alpha$, which is computed from $p$ and $o^\alpha$, is not affected by the addition of $V^U_{Rlm\sigma}$.

Thus, by merely adding $V^U_{RL\sigma}$ to $C_{RL}$ and $\varepsilon_{\nu RL}$, the diagonal-only part of the LDA+U hamiltonian can be incorporated, under the assumption that $V^U_{RL\sigma}$ is spatially constant. If there is no off-diagonal part, $H$ is independent of representation (actually only nearly so—there is a slight dependence through the interplay of combined correction terms and the $U$ part of $H$).

In the general case, we fold the diagonal part $V^U_{RL\sigma}$ of $V^U_{Rl\sigma;m,m}$ into the potential parameters, and add the difference $V^U_{Rl\sigma;m,m'}(1-\delta_{mm'})$ as a perturbation. If the output density is accumulatd by the definition of the LMTO basis functions (`QASA=3`), the result is essentially independent of represention. However, when the density is accumulated from true power moments, as is necessary in our GF formalism, there is a problem in defining the "band center," and thus ambiguity in how first and second energy power moments of the output density are defined (there is no difficulty withzeroth moment, the density). In principle, it would be possible to rectify this by diagonalizing the $m \times m$ matrix of $V^U_{Rl\sigma;m,m}$ and accumulating the power moments in that representation, but this is a rather painful exercise and has not been done so far.

## II. IMPLEMENTATION

Programs `lmf` and `lm` each have a "rotationally invariant" implementation of the LDA+U hamiltonian. When the rotationally invariant form is used together with `QASA=3`, `lm` can also be used in any screening representation (e.g. $\alpha$ or $\gamma$). There is a slight, but not identically zero dependence of the results on screening representation; however, the differences should be very small. It also matters little whether real or spherical harmonics are used. You can see how diagonal the density matrix is by inspecting file `dmats`, or looking at the output file when LDA+U is turned on.

A test case verifies the preceding remarks for hcp Er. This case can be run with

```
testing/test.lm 9
```

LDA+U in the noncollinear case is illustrated for hcp Er through

```
nc/test/test.nc 7
```

`lm` can also be invoked in a diagonal-only approximation: i.e. $V^U_{Rl\sigma;m,m'}$ is discarded for $m \neq m'$. (It is implementation by adding $V^U_{Rl\sigma;m,m}$ to band parameters `C` and $\varepsilon_\nu$). In this approximation, `QASA=0` may be used. Without rotational invariance, it now matters whether real or spherical

harmonics are used. The density matrix of the $d$ block in cubic transition is diagonal in real harmonics. The $f$ states of Er are atomic-like; they follow Hund's rules and are nearly diagonal in spherical harmonics. There is no implementation of a general prescription to make $V^U_{Rl\sigma;m,m'}$ diagonal.

`lmgf` is implemented in a "rotationally invariant form;" however, since only `QASA=0` is implemented in this program, self-consistent calculations are not particularly meaningful. The magnetic exchange interactions can use off-diagonal parts of $V^U$; however, the Lichtentstein expressions for magnetic exchange interactions are assumed not to change relative to the LDA case. This is only correct when $V^U$ is diagonal in $m$. The LDA+U implementation in `lmgf` is illustrated by

```
gf/test/test.gf eras
```

Tests 1 and 2 compare $2^{nd}$ and $3^{rd}$ order potential functions to corresponding calculations by `lm`. Test 4 shows a self-consistent calculation; test 7 demonstrates a noncollinear implementation. Magnetic exchange interactions are illustrated by

```
gf/test/test.gf co 5
gf/test/test.gf fe 5
```

`lmpg` has been implemented in a diagonal-only approximation. Test case

```
pgf/test/test.pgf co
```

offers two demonstrations or checks: test 2 for Co shows that it can generate energy bands equivalent to those generated by `lm`. (`kmap` option, `PGF MODE=3`). Also, test 8 illustraes the transport mode (`PGF MODE=5`) in LDA+U.

NOTE: LDA+U has not yet been implemented in conjunction with the fully relativistic mode (`LREL=2`).

In summary, the LDA+U is only "rotationally invariant" for program `lm`, when (`QASA=3`) is used, or in the full-potential program (`lmf`. How diagonal $V^U_{Rl\sigma;m,m'}$ is depends on the case, and also on other choices, such as whether spherical or real harmonics are used.