



ATOMIC FORCES IN FLEUR and a bit on what to do with them

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Part I: Definition of Forces



DEFINITION OF FORCES IN FLEUR

- Forces = Forces on each atom

→ Derivative of the total energy with respect to the atomic position

$$\mathbf{F}_\alpha = -\frac{dE_{tot}}{d\tau_\alpha}; \quad \frac{\partial E(\lambda)}{\partial \lambda} \stackrel{HF}{=} \langle \Psi | \frac{\partial H}{\partial \lambda} | \Psi \rangle$$

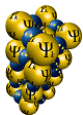
- *Highly* nontrivial to calculate in FLAPW
- Implementation in Fleur based on the work of Yu et al.¹ and split into contributions based on the formulae in the paper

¹ All-electron and pseudopotential force calculations using the linearized-augmented-plane-wave method, <https://doi.org/10.1103/PhysRevB.43.6411>





Part II: Calculating Forces in FLAPW



GENERAL CONCEPT

- Most energy contributions are expressed as an integral, which has two separate contributions

$$\mathbf{F}_\alpha = -\frac{d}{d\tau_\alpha} \int_\Omega d\mathbf{r} g(\mathbf{r}) \approx -\sum_{\beta \neq \alpha} \int_{MT_\beta} d\mathbf{r} \frac{dg(\mathbf{r})}{d\tau_\alpha} - \int_{IR} d\mathbf{r} \frac{dg(\mathbf{r})}{d\tau_\alpha}$$

- Other important factor: Pulay contributions from basis dependency on the atom positions and incompleteness of the set

$$\frac{d\epsilon_{\nu\mathbf{k}}}{d\tau_\alpha} = \langle \psi_{\nu\mathbf{k}} | \frac{dH}{d\tau_\alpha} | \psi_{\nu\mathbf{k}} \rangle + \langle \frac{d\psi_{\nu\mathbf{k}}}{d\tau_\alpha} | H - \epsilon_{\nu\mathbf{k}} | \psi_{\nu\mathbf{k}} \rangle + \text{c.c.}$$



FORCE CONTRIBUTIONS IN FLEUR

- Each term is based on the respective equation in the paper¹

- A3 Hellmann-Feynman force contribution from differentiating the Coulomb energy (valence density; Equation A3)
- A4 The same for core electron density (Equation A4)
- A8 First valence Pulay contribution (Equation A8)
- A12 Second valence Pulay contribution (Equation A12)
- A21 Last valence Pulay contribution (Equation A17 and A20)

¹ *All-electron and pseudopotential force calculations using the linearized-augmented-plane-wave method*, <https://doi.org/10.1103/PhysRevB.43.6411>



ADDITIONAL FORCE CONTRIBUTIONS

- Added to reduce drift forces (non-vanishing sum of all atomic forces)
- Labelled as 'Force levels' 1 to 3; 2 and 3 contain surface terms:

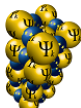
$$\mathbf{F}_\alpha = - \sum_{\beta \neq \alpha} \int_{MT_\beta} d\mathbf{r} \frac{dg(\mathbf{r})}{d\tau_\alpha} - \int_{IR} d\mathbf{r} \frac{dg(\mathbf{r})}{d\tau_\alpha} \\ - \oint_{\partial MT_\alpha} dS \hat{\mathbf{e}}_{MT_\alpha} (g_{MT}(\mathbf{r}) - g_{IR}(\mathbf{r}))$$



FORCE LEVELS

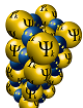
- IS/MT addition to A4: Additional Pulay terms for the core states (level 1; Equation 20 in reference paper by Klüppelberg et al.²)
- A12 is modified with surface contributions of the kinetic energy and eigenenergies (level 2; Equation 22)
- Surface correction: Remaining surface terms of integral contributions (level 3; Equation 14)
- Caveat: Level 1 doesn't extend to the vacuum region for film calculations and the levels are only explicitly tested for LDA functionals

²Atomic force calculations within the all-electron FLAPW method: Treatment of core states and discontinuities at the muffin-tin sphere boundary,
<https://doi.org/10.1103/PhysRevB.91.035105>



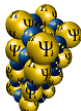
INPUT SWITCHES

- First tag in <calculationSetup> (**red**: not generated by default)
- <geometryOptimization l_f="F" **f_level="0"**
forcealpha="1.00" forcemix="BFGS" epsdisp=".00001"
epsforce=".00001" **force_converged="0.00001"/>**
- **L_f** activates the force calculation/relaxation; the force level, mixing constant, mixing scheme, convergence criteria for atomic displacements/forces and the force distance required in the scf loop can be set additionally.
- Additional tags for each atom species:
- <force calculate="T" relaxXYZ="TTT"/>
- Calculate this particular force yes/no and do we relax in each respective direction?



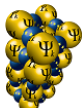
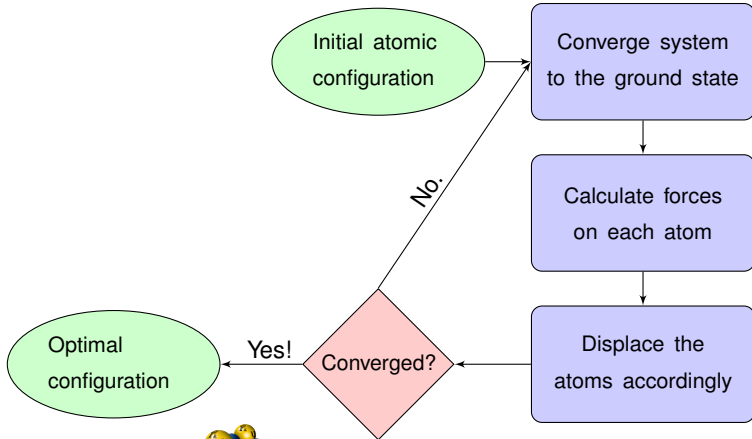


Part III: Using Forces (Fleur Applications)



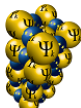
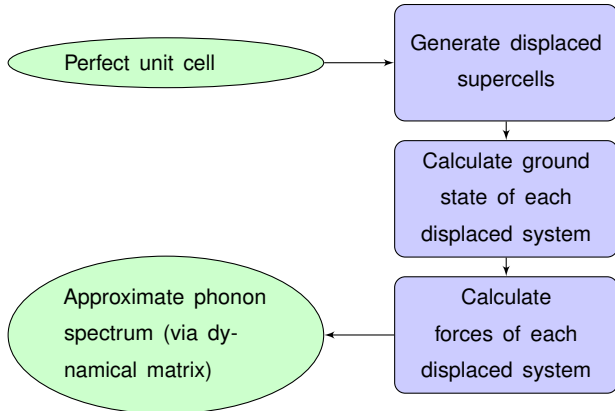
STRUCTURAL RELAXATIONS

- Find the energetically optimal configuration for a given system with respect to its atoms



PHONONS FROM SUPERCCELLS

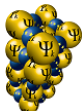
- From an optimized unit cell, construct supercells with displacements to simulate frozen phonons (FD method)



ADDITIONAL INFORMATION

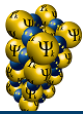
- For relaxations, the MT radii should be reduced to 95%, so the MTs do not crash into each other
- The relaxation workflow can be automated in Aiida-Fleur³
- We do *not* calculate and use stress tensors in a similar fashion; lattice optimization has to be done by e.g. a Birch-Murnaghan fit.
- Fleur is *not* a code for Molecular Dynamics simulations.
- The idea of energy derivatives reappears in the context of Density-Functional Perturbation Theory, which is currently in development

³<https://aiida-fleur.readthedocs.io/en/v1.1.1/index.html>





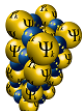
Part IV: A small Test System



EXAMPLE SYSTEM

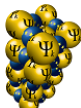
- fcc Aluminium: Displace atom at origin by a hundredth of the unit cell size in z-direction
- Calculate atomic forces and drift force
- Relax back into origin
- 4x4x4 k-points, $a = 7.646 a_0$, $k_{max} = 4.2/a_0$, $G_{max} = 14.5/a_0$, $l_{max} = 10$, LDA functional by Vosko/Wilk/Nusair ⁴

⁴Accurate spin-dependent electron liquid correlation energies for local spin density calculations: a critical analysis, <https://doi.org/10.1139/p80-159>

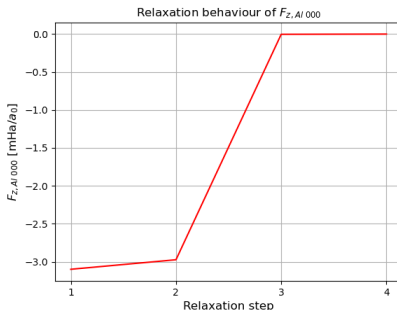
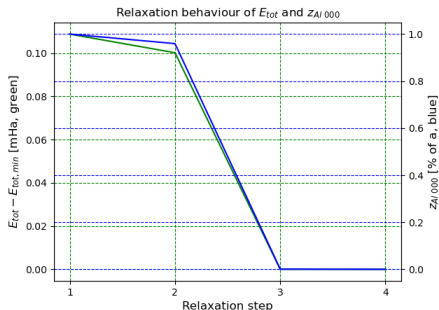


FIRST ITERATION FORCES

- Al in [0,0,0]: $-3.11037 \text{ mHa}/a_0$
 - Al in [0,1/2,1/2]: $2.16279 \text{ mHa}/a_0$
 - Al in [1/2,0,1/2]: $2.16279 \text{ mHa}/a_0$
 - Al in [1/2,1/2,0]: $-1.21521 \text{ mHa}/a_0$
-
- Drift force: $-1.45776 \text{ nHa}/a_0$



RELAXATION INTO THE ORIGIN



- Rapidly converges back into symmetric order (as it should); more interesting stuff in the tutorial!

THAT'S IT

Thank you for your attention!

