

Atomistix ToolKit Tutorial

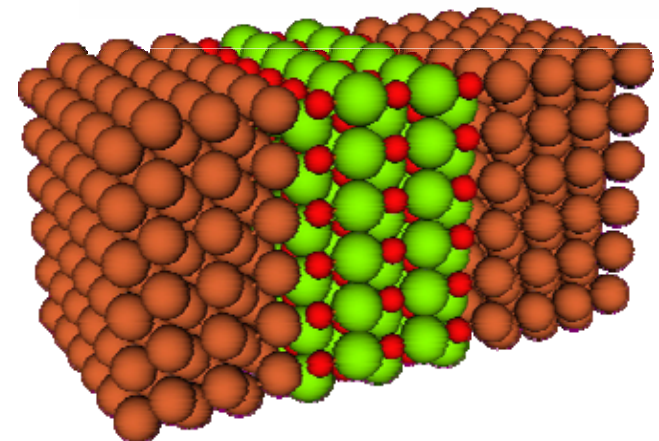
Part 1:
Two-probe geometry
& convergence parameters

Quantum
Wise

Outline

❖ Part 1:

- » Fundamental concepts of a two-probe system
- » Solving convergence problems
- » Based on a specific example which is hard to converge (FeMgO MTJ)



❖ Part 2: Analysis

Fundamental but complex topics in ATK

- ❖ Understanding the fundamental concepts of a two-probe system is crucial to establish confidence in the simulations and their results
- ❖ Most calculations in ATK converge with default parameters
 - » But some do not... (for a variety of reasons)
- ❖ Geometrical aspects
 - » Related to certain “hidden” approximations and/or simplifications in ATK
- ❖ Numerical parameters
 - » Accuracy
 - » Solving convergence problems
- ❖ Both aspects are related to convergence, but may also compromise quality of results if not chosen appropriately (even if the calculation converges!)



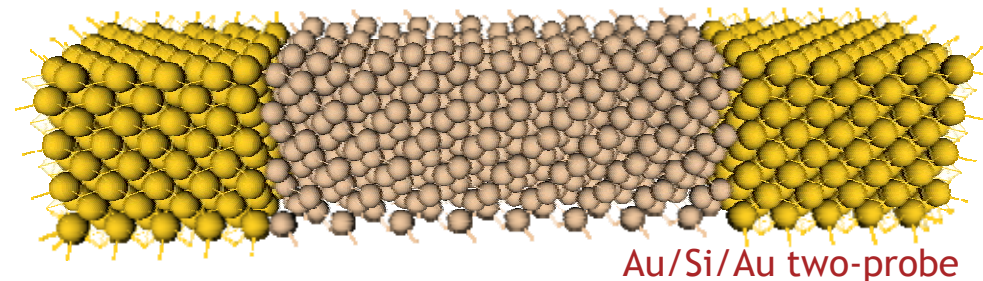
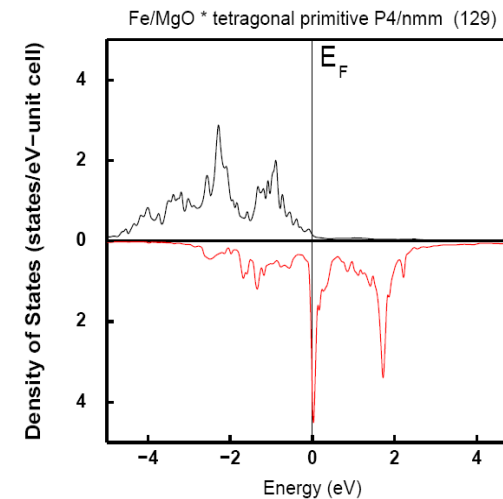
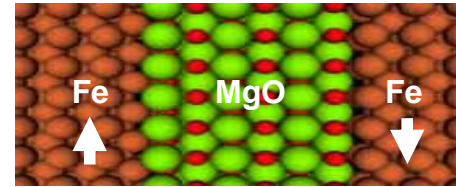
Three ways to improve performance

- ❖ Total calculation time =
time/iteration \times number of iterations
- ❖ Reduce time/iteration
 - » Parallelization (scaling depends on system & parameters)
 - » Code and algorithm improvements (ATK 2008.10)
- ❖ Reduce number of iterations
 - » Algorithm improvements
(new convergence criterion in ATK 2008.02)
 - » Parameter tuning
- ❖ Third way?
 - » Avoid running end-less calculations which do not converge!
 - » Avoid re-running calculations because of poor quality results



Our guinea pig

- ❖ Fe/MgO/Fe magnetic tunnel junction (MTJ)
- ❖ Anti-parallel electrode spin polarization
 - » Among the most difficult systems to converge
 - » Strong peak in the minority DOS at the Fermi level
- ❖ Many other interface systems exhibit similar convergence issues

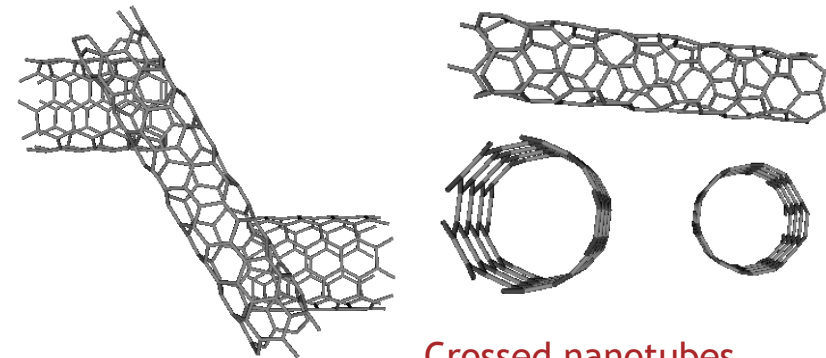


Electrodes, the simple stuff

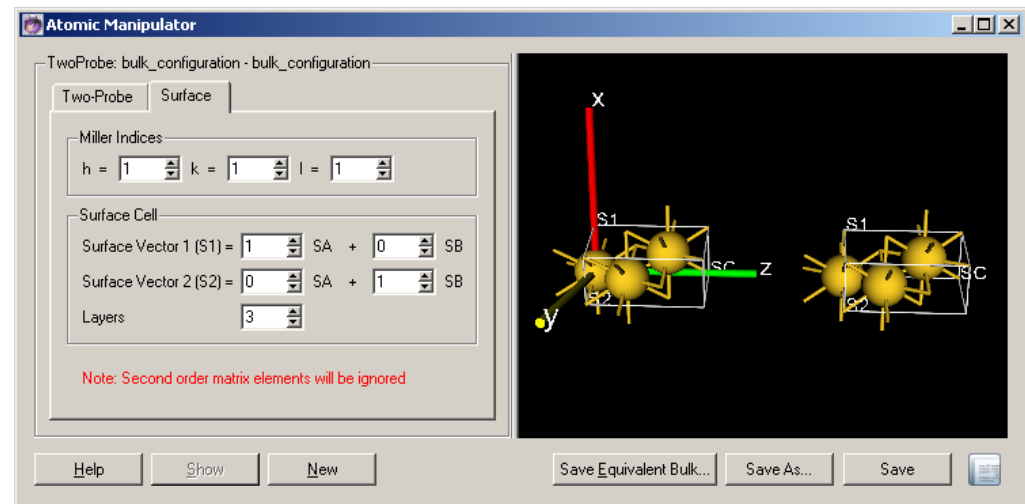
- ❖ Transport direction must be perpendicular to the interface plane
 - » Still possible to consider transport at an angle

- ❖ Electrodes must not feel each other
 - » Atoms in the left electrode may not have any basis set overlap with atoms in the right electrode

- ❖ Electrode cell must be periodic in the transport direction
 - » Example: fcc [111] → 3, 6, 9, ... layers



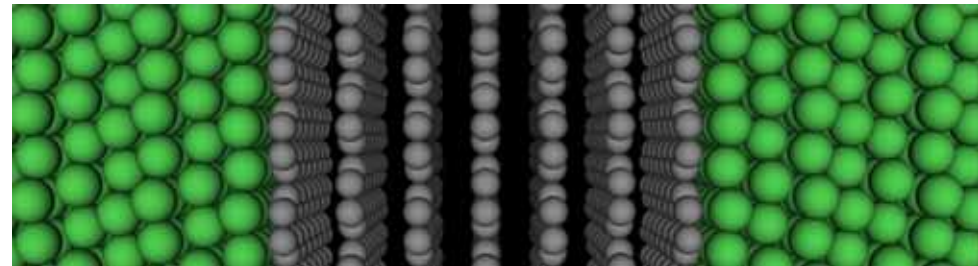
Crossed nanotubes



Electrodes, more simple stuff

❖ Periodic boundary conditions in the interface plane (x/y)

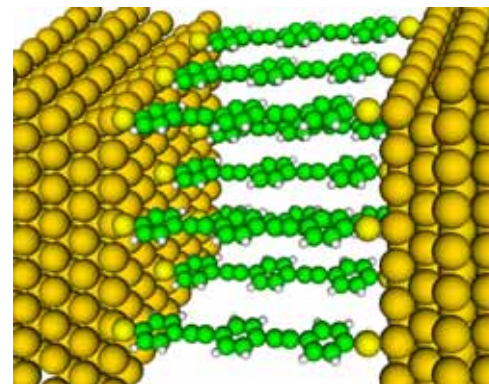
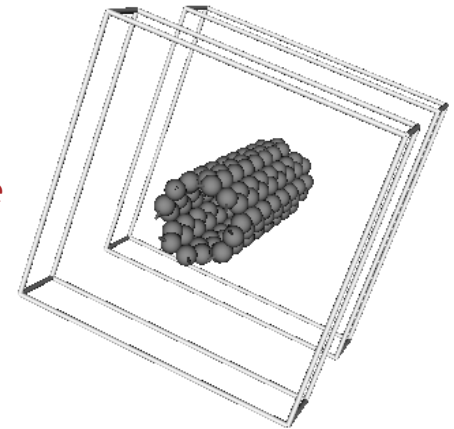
- » Allows study of true interfaces
- » Vacuum padding needed to allow electrostatic interactions to decay for 1D/2D systems
- » Sufficiently large metal surface cell needed for “broad” molecules



Nickel - Graphene - Nickel spin filter

» Vacuum padding needed to allow electrostatic interactions to decay for 1D/2D systems

Nanotube two-probe



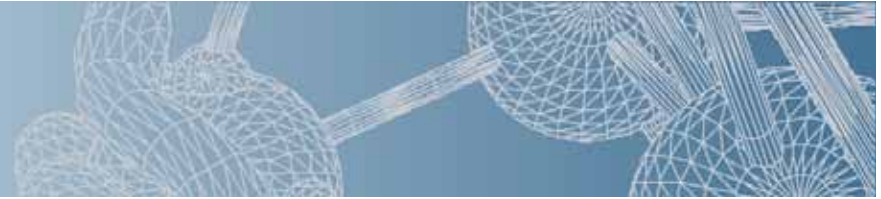
Au [111] 3x3

Alignment

- ❖ Input geometries are relative (L/C/R)
 - » Alignment is done by specially appointed alignment pairs (atom indices)
- ❖ Pros:
 - » Homogeneous electrodes only need to be defined once
 - » Easy to change the internal geometries of electrodes or central region
 - » No need to think about absolute alignment
- ❖ Cons:
 - » Difficult to assign alignment atoms by hand
 - » Atom indices may change if the geometry is modified → must remember to update alignment
 - » Easy to make non-obvious mistakes



Always inspect two-probe geometries in VNL (Nanoscope) before the calculation to ensure proper alignment!



Alignment, the details

- ❖ Appoint 2 atom pairs to align the 3 components
 - » (left electrode, central region) = (L,C_L)
 - » (right electrode, central region) = (R,C_R)
 - » NanoLanguage input is [(L,C_L), (R,C_R)]
- ❖ Convenient to use relative index (-1) for last atom in list

```
twoprobe_configuration = TwoProbeConfiguration(
    (left_electrode_configuration, right_electrode_configuration),
    scattering_elements,
    scattering_coordinates,
    equivalent_atoms = ([0,0],[3,5]) )
```

1. Central region is aligned to left electrode such that

$$\mathbf{r}_C(0) = \mathbf{r}_L(3) + \mathbf{u}L_3 \quad (\mathbf{u}L_3 = 3\text{rd unit cell vector for left electrode})$$
2. Right electrode is aligned such that

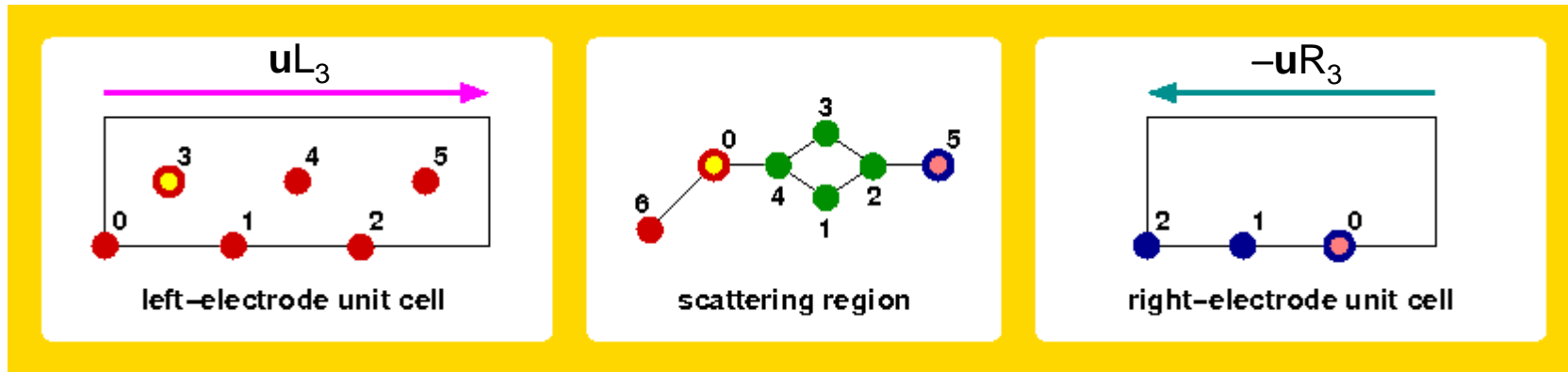
$$\mathbf{r}_R(0) = \mathbf{r}_C(5) + \mathbf{u}R_3 \quad (\mathbf{u}R_3 = 3\text{rd unit cell vector for right electrode})$$

Concrete alignment example

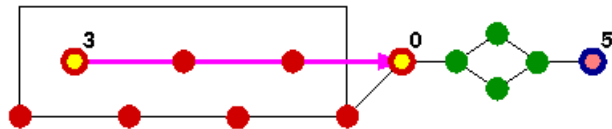
Alignment atoms: $[(3,0), (0,5)]$



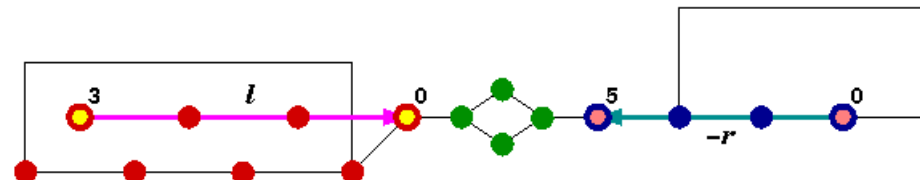
See the ATK manual on *TwoProbeConfiguration* for more details!



1. Left + central



2. Right

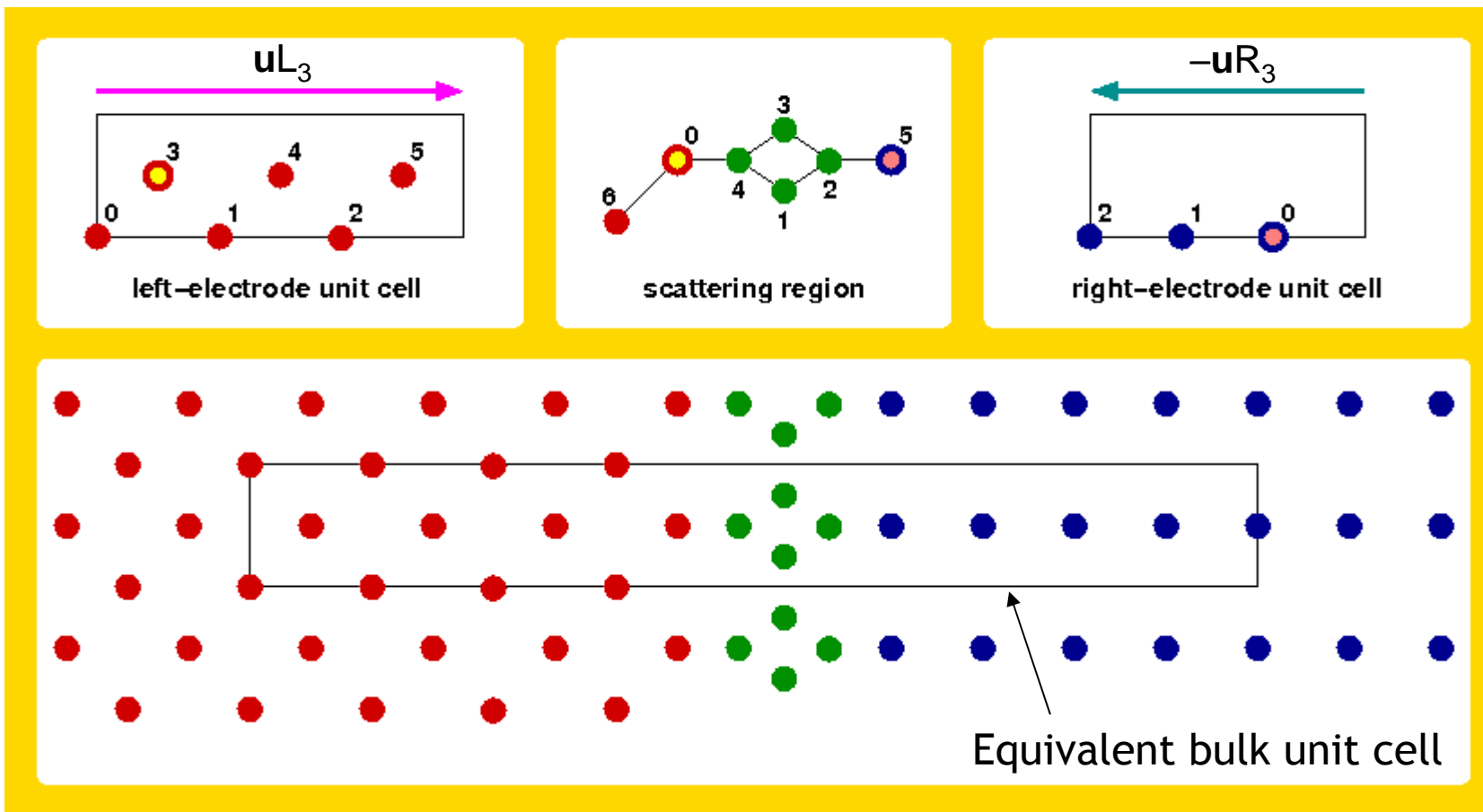


Concrete alignment example

Alignment atoms: $[(3,0), (0,5)]$

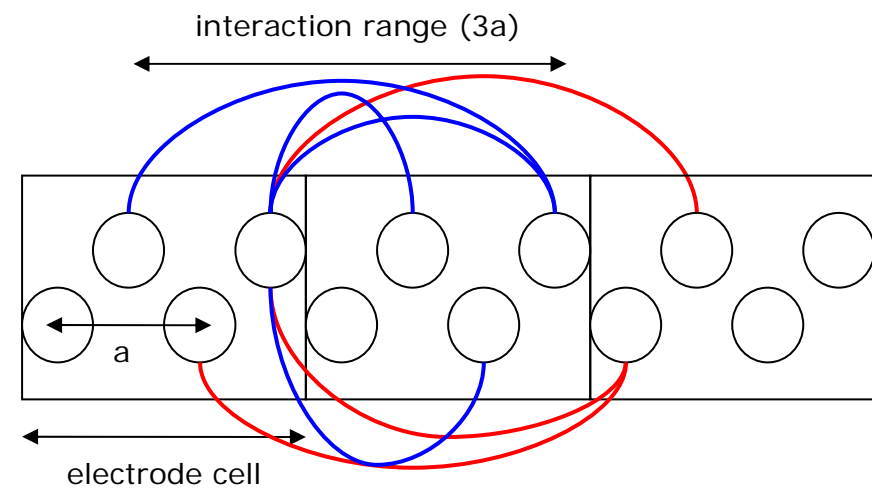


See the ATK manual on *TwoProbeConfiguration* for more details!



Electrodes, the subtleties

- ❖ **Electrode calculation:**
electrode is treated as a bulk system
 - » All interactions included
- ❖ **Two-probe calculation:**
interactions extending beyond the nearest-neighbor cell (**red**) are truncated from the Hamiltonian
 - » May shift the Fermi level
 - » Excluded interactions constitute an **approximation**



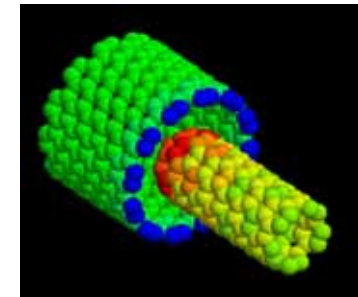
Always ensure that
the electrodes are
deep enough!

Heterogeneous two-probes

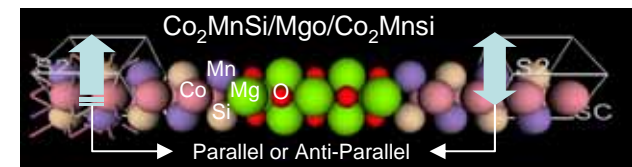
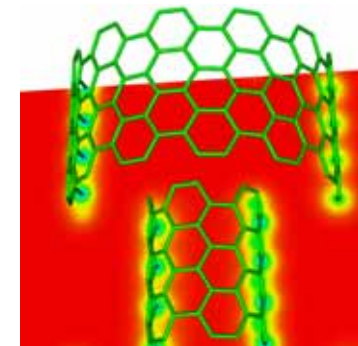
- ❖ Two-probe systems can be
 - » **Homogeneous** (identical electrodes)
 - » **Heterogeneous** (different electrodes)

- ❖ Electrostatics (Poisson equation)
 - » Homogeneous: **FFT**
 - » Heterogeneous: **Multigrid** (in z, FFT in x/y)

- ❖ Geometrically identical electrodes with different **numerical parameters** is a **heterogeneous** system
 - » Example: MTJ anti-parallel case



Nanotube heterojunction



Heusler/MgO MTJ

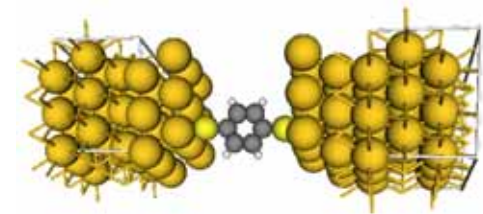
Homogeneous vs. heterogeneous

- ❖ Deciding factor: two-probe system and method constructions in NanoLanguage
 - » L/R electrode = same variable → homogeneous
 - » L/R electrode = different variables → heterogeneous

```
homogeneous_twoprobe = TwoProbeConfiguration(  
    (electrode_configuration, electrode_configuration),  
    ...)  
  
heterogeneous_twoprobe = TwoProbeConfiguration(  
    (left_electrode_configuration, right_electrode_configuration),  
    ...)  
  
homogeneous_twoprobe_method = TwoProbeMethod(  
    electrode_parameters = (electrode_parameters, electrode_parameters),  
    ...)  
  
heterogeneous_twoprobe_method = TwoProbeMethod(  
    electrode_parameters = (left_electrode_parameters, right_electrode_parameters),  
    ...)
```

Initial density

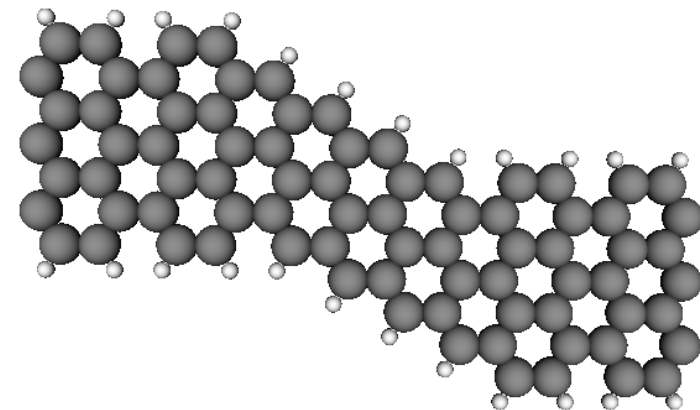
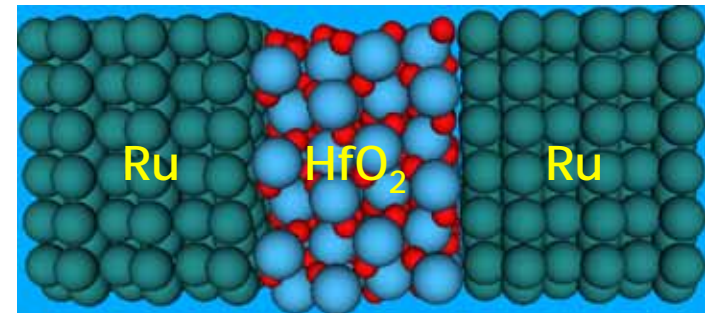
- ❖ Two-probe systems can be hard to converge for many reasons
 - » One reason is that the bulk density matrix from the converged electrodes is far from the “neutral atom” density matrix
- ❖ An initial `EquivalentBulk` calculation can help in many cases
 - » ATK will automatically create an equivalent bulk system from L+C+R and perform a bulk calculation for this system
 - » The converged density matrix is then used as a starting guess for the two-probe calculation
 - » `InitialDensityType.EquivalentBulk` is default
- ❖ **Requirement:** the entire system L+C+R should be periodic, or at least without “bad” stacking faults
 - » Examples, fcc [111]:
 - [ABC]AB-...-BC[ABC] (no mirror, no stacking fault)
 - [ABC]AB-...-BA[CBA] (mirror, bad stacking fault BAAB)
 - [ABC]AB-...-BAC[BAC] (mirror, minor stacking fault ACAB)
- ❖ **Homogeneous** systems should always fulfill the requirement
 - » Otherwise the electrode itself is not periodic!
- ❖ **Heterogeneous** systems may or may not
 - » If left/right electrodes are really different (physically or with different stacking), change to `InitialDensityType.NeutralAtoms`



Au-DTB-Au

Transverse unit cell

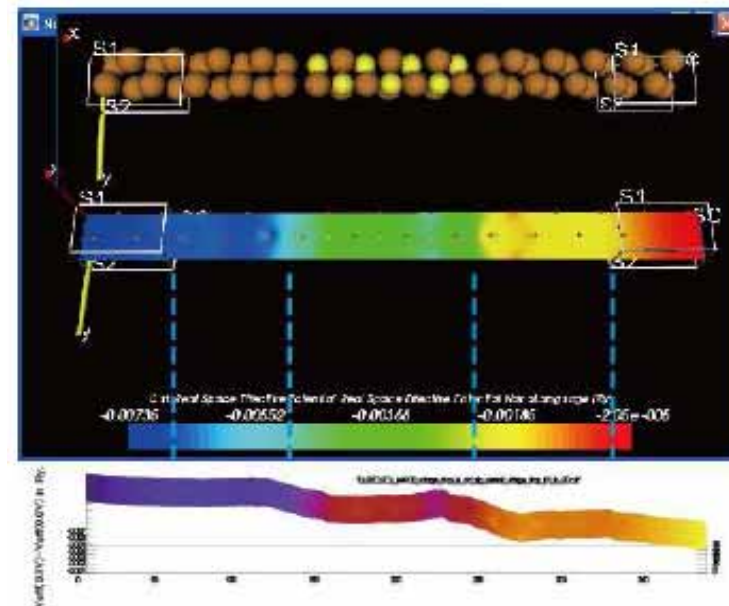
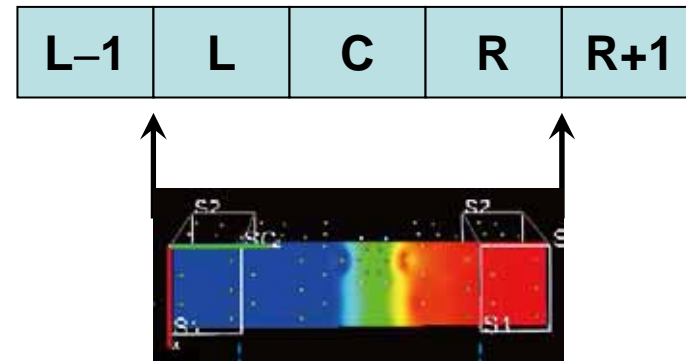
- ❖ For **heterogeneous** systems, X/Y unit cell **commensurability** must be carefully ensured
 - » Trivial for 1D systems like CNTs and graphene (vacuum padding)
 - » Challenge for interfaces which are not lattice matched (large supercell)
- ❖ Too small X/Y unit cell leads to electrostatic "cross-talk"
 - » May occur both for true 1D system and quasi-1D wires (molecular or metallic) between extended interfaces
 - » Typical signal of residual electrostatic interactions is broken degeneracies, e.g. in high symmetry point of the band structure

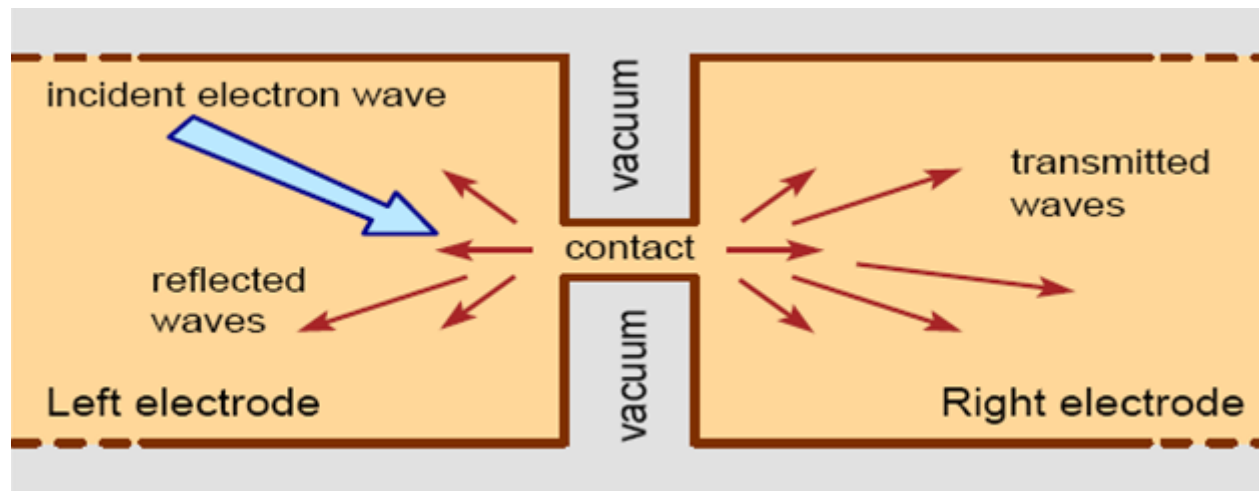
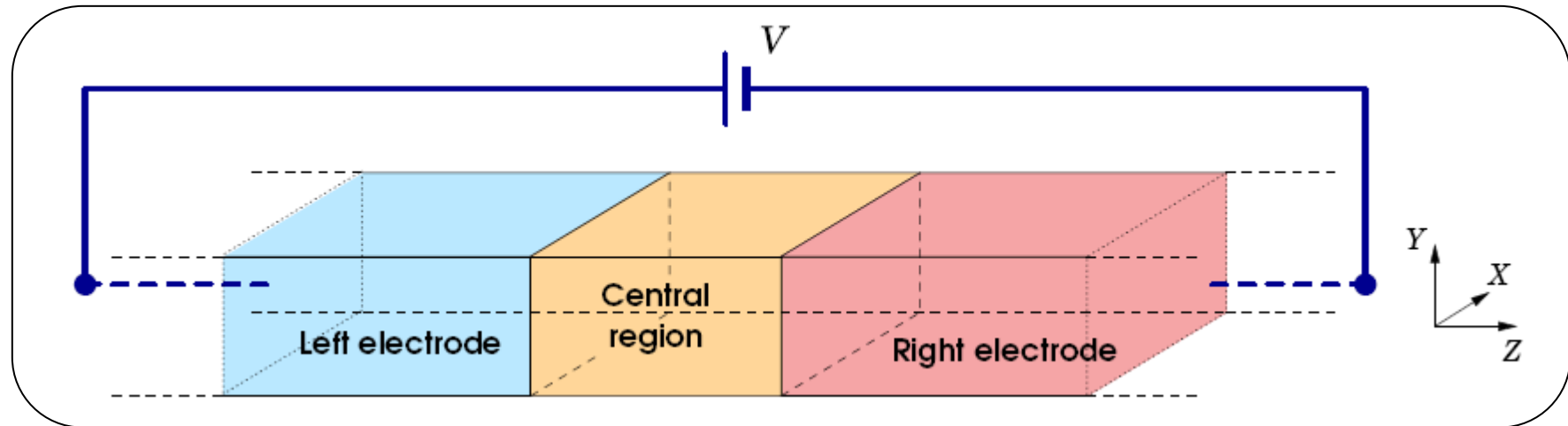


Z-shaped graphene transistor

Screening approximation

- ❖ Why is it necessary to have a lot of “electrode atoms” in the central region?
 - » These atoms form the **screening layers**
- ❖ **Two-probe boundary condition:** match the bulk electrode effective potential at the outer electrode cell boundaries (arrows)
 - » In the region in between (L/C/R), the **effective potential** and **electron density** are calculated self-consistently
- ❖ **Surface layers** provide **screening** so that the electrode region is close to bulk-like
 - » The surface layers are part of the central region config!
 - » Too few screening layers constitutes an approximation

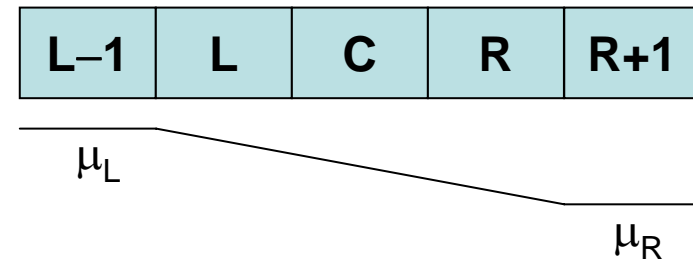




Voltage bias

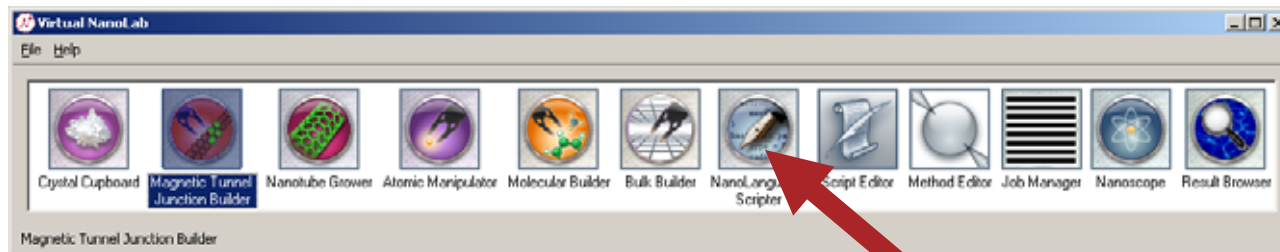
- ❖ Bias is applied across entire two-probe region (L+C+R)

- » FFT (homogeneous): linear ramp added to effective potential
- » Multigrid (heterogeneous): included in the boundary conditions

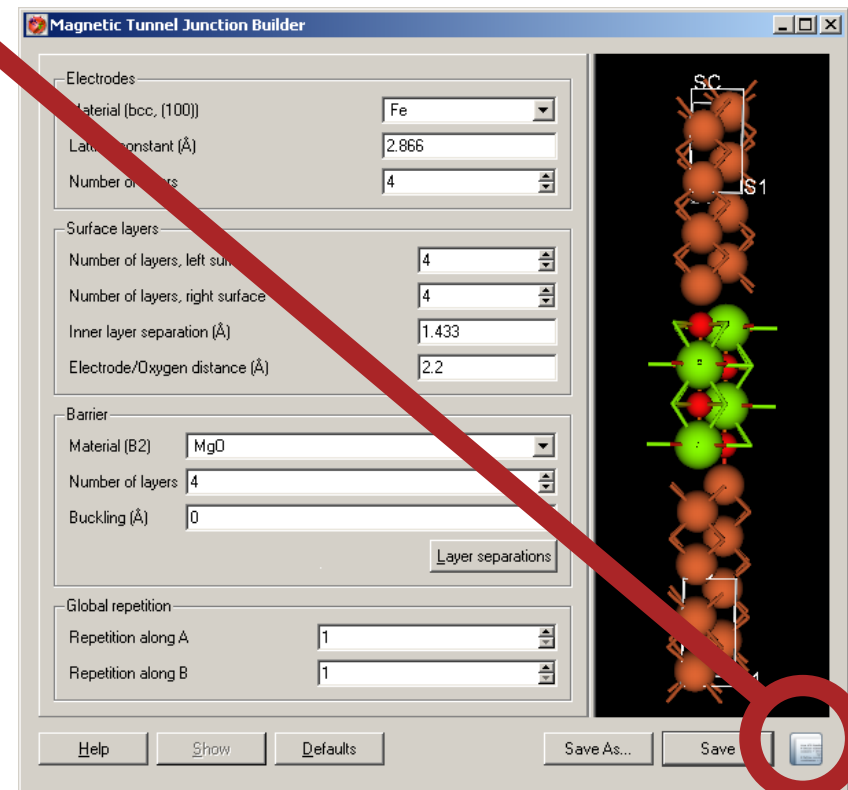


- ❖ Bias is relative
 - » Can be applied symmetrically ($\pm V/2$) or asymmetrically ($V,0$)
 - » Bias direction determines direction of current (diode characteristics)
- ❖ Convergence under bias is more difficult
 - » Converge with zero bias first, then use zero-bias calculation to initialize density matrix for finite bias
 - » Up to about 2 V bias is currently possible in ATK (above this, convergence becomes very difficult)
- ❖ Especially important to have enough screening surface layers under bias

Back to our guinea pig: Geometry

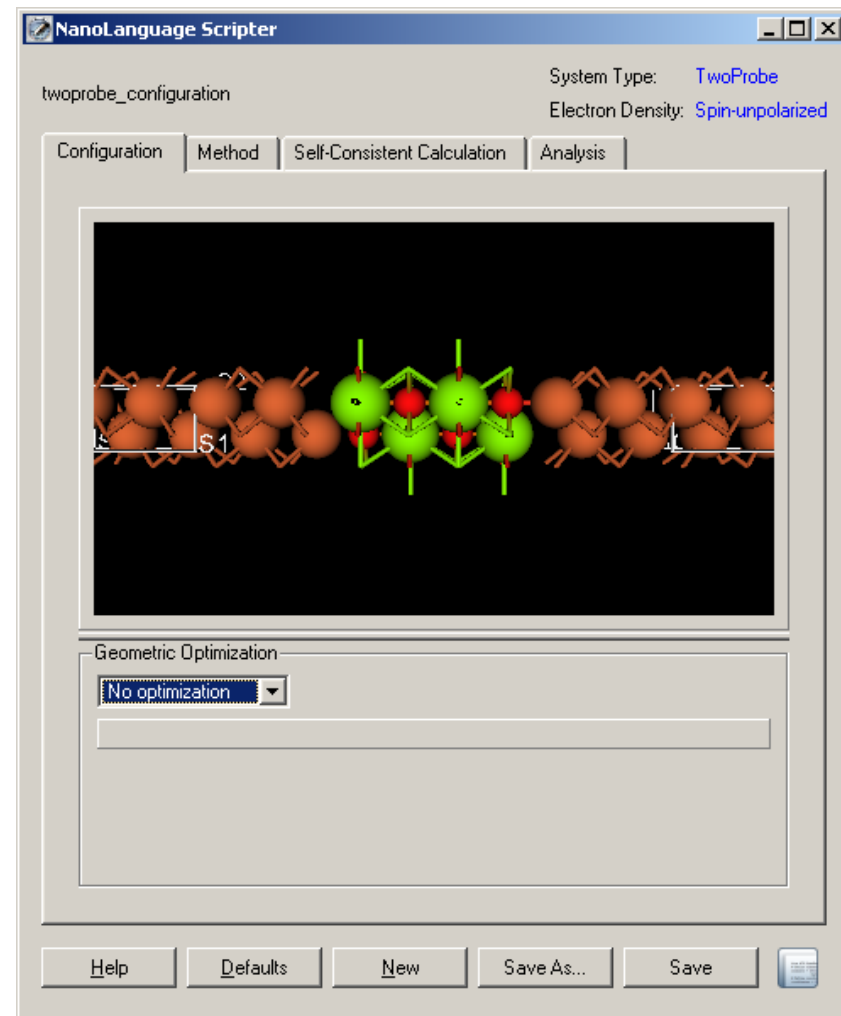


- ❖ Use MTJ Builder in Virtual NanoLab (new in 2008.02)
- ❖ Default geometry works for our demonstration
 - » 4 electrode layers and 4 surface layers
 - » 6 would be better, costs more time & memory
- ❖ Drop system on NanoLanguage Scriptor



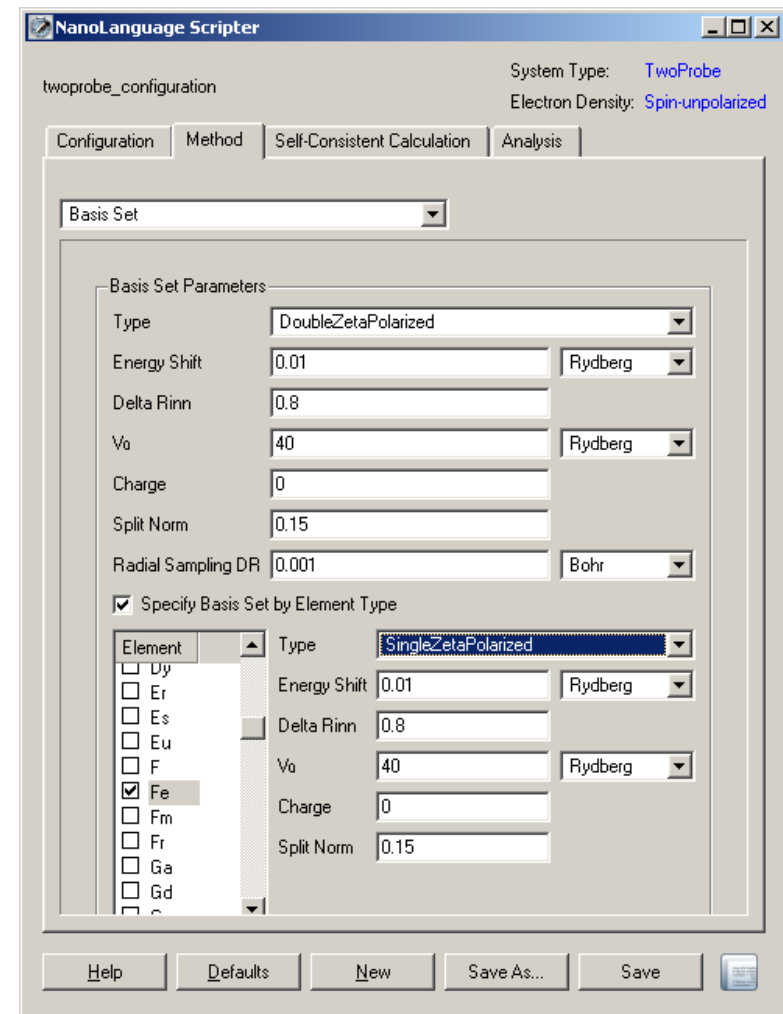
NanoLanguage Scripter

- ❖ Powerful tool for generating NanoLanguage code
 - » Even QuantumWise experts use it!
 - » Can also be used for post-SCF analysis
- ❖ Generated code is
 - » MPI safe
 - » Explicit (all parameters visible)
 - » Verbose (prints details to console)
- ❖ Non-standard things can always be added to the script by hand afterwards



Basis set

- ❖ ATK uses localized atomic orbitals basis sets with finite range (SIESTA)
- ❖ Most parameters are “advanced” (keep default), except:
 - » Type (basis set size)
 - » Energy shift (lower to increase basis function range)
- ❖ SingleZetaPolarized or DoubleZetaPolarized are usually optimal
 - » Balance time & memory vs. accuracy
- ❖ Possible to use different parameters for each element

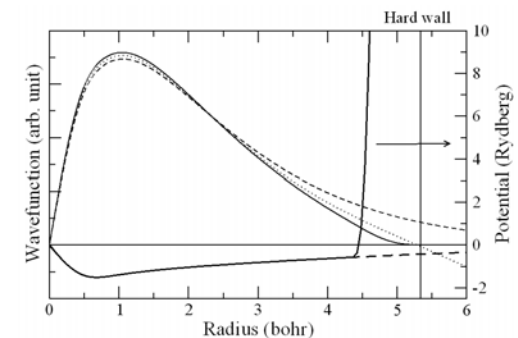


Basis set, details

- ❖ Norm-conserving pseudopotentials describe the core electrons
- ❖ Valence orbitals are expanded in localized basis functions with a finite range
 - » A list of valence electrons for each element can be found in the manual
- ❖ The range of the cut-off is determined implicitly by the "Energy shift"
 - » Energy shift = 0 means infinite range, basis orbital = atomic orbital (cannot be used)
 - » Decrease with caution from default to increase range (to improve accuracy)
 - » Range increases exponentially and thus the number of interacting pairs goes up fast
- ❖ Basis sets (ordered least to most accurate):
 - » SingleZeta; 1 basis orbital for each valence orbital
 - » SingleZetaPolarized; SingleZeta + 1 basis orbital for the first unoccupied shell
 - » DoubleZeta; 2 basis orbitals for each valence orbital
 - » DoubleZetaPolarized; DoubleZeta + 1 basis orbital for the first unoccupied shell
 - » DoubleZetaDoublePolarized; DoubleZeta + 2 basis orbitals for the first unoccupied shell
- ❖ For details, see the manual for "AtomicOrbitals"

$$V_{\text{conf}}(r) = \begin{cases} 0 & \text{if } r < r_{\text{inn}} \\ V_0 \frac{\exp[-1/(r-r_{\text{inn}})]}{(r_c-r)} & \text{if } r_{\text{inn}} < r < r_c \\ \infty & \text{if } r_c < r \end{cases}$$

$$\phi_l^{2\zeta}(r) = \begin{cases} r^l(a_l - b_l r^2) & \text{if } r < r_l^{\text{split}} \\ \phi_l^{1\zeta}(r) & \text{if } r \geq r_l^{\text{split}} \end{cases}$$

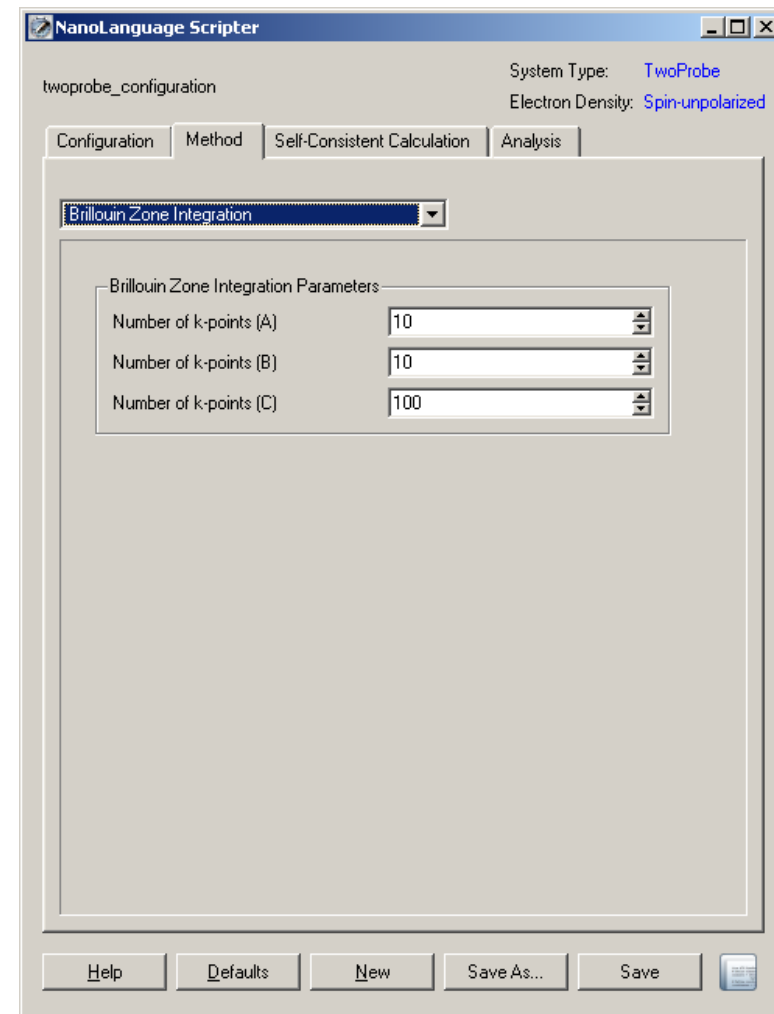


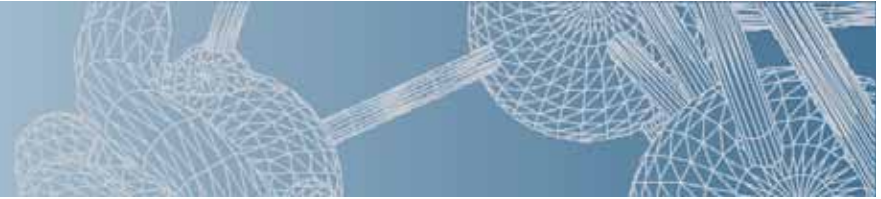
Brillouin zone integration

ATK is parallelized
over the k-points

- ❖ K-points are crucial for good accuracy and convergence
 - » Especially important for FeMgO due to sharp DOS peak at Fermi level
- ❖ As always, larger cell → fewer k-points needed
- ❖ C-direction k-points only used in electrode calculation
- ❖ Memory/time increases with k-points, but slower (N) than with number of atoms (N^2)
- ❖ Both electrodes must use the same number of k-points

For details see next 2 pages

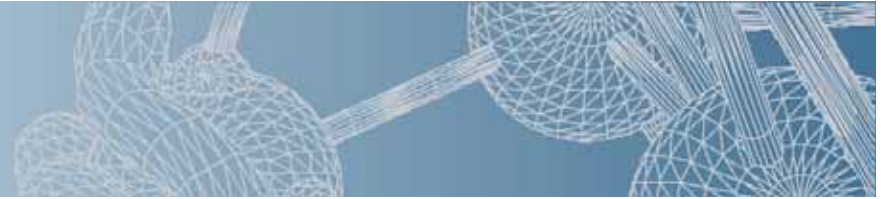




Details on k-points A/B

- ❖ A/B k-points used both in electrodes, equivalent bulk, and two-probe calculation
- ❖ Number required depends on system size (wider electrode in A/B, fewer points)
- ❖ In directions without dispersion/periodicity (vacuum padding), only 1 point needed
 - » Examples: nanotubes, wires, graphene (perpendicular to the sheet)
- ❖ Some systems, like FeMgO, require more points to get accurate Fermi level
- ❖ Increased temperature can be used to reduce need for many k-points, at the expense of somewhat reduced accuracy in the results



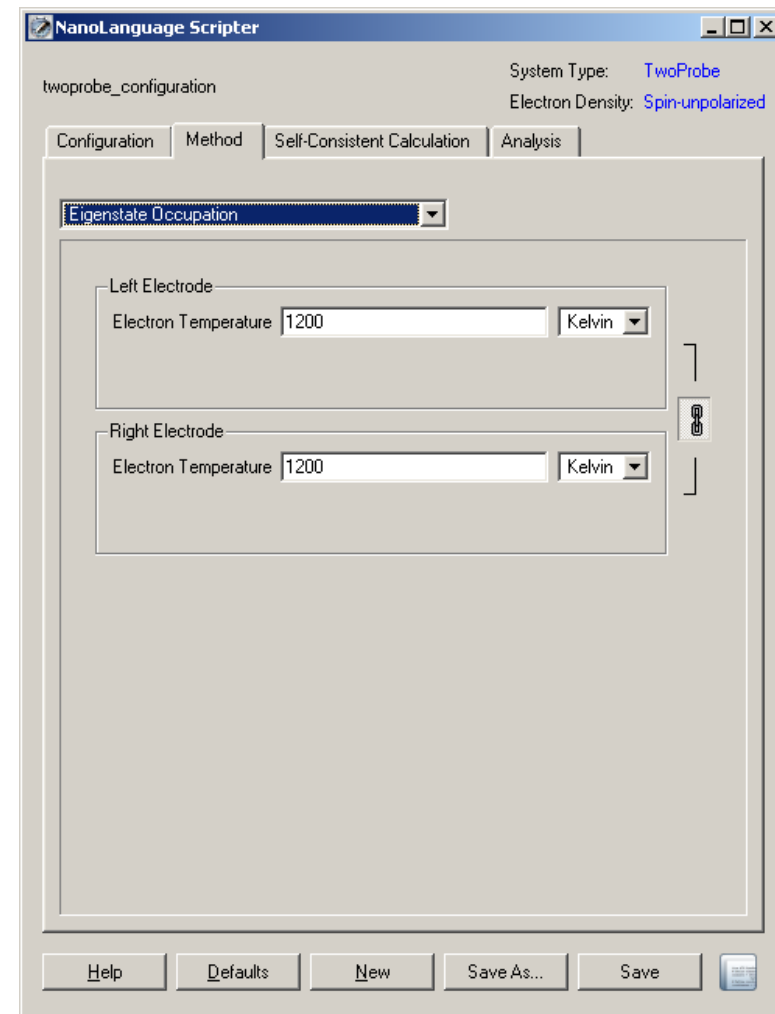


Details on k-points C

- ❖ Why k-points in the C (Z) direction?
 - » Only used in the electrodes bulk calculation
 - » **NOT** used in the two-probe calculation - no periodicity!!!
 - » $N_A \times N_B \times 1$ used in the equivalent bulk calculation
 - If $N_A = N_B = 1$ the equivalent bulk calculation is performed with real matrices (faster, less memory)
 - Also applies to regular bulk calculations, if $N_A = N_B = N_C = 1$
- ❖ Why so many k-points in the C direction?
 - » Crucial to get an accurate determination of the Fermi level
 - » Two-probe calculation assumes semi-infinite electrode, while electrode calculation is finite (different boundary conditions)
 - » Thus, two-probe calculation corresponds, effectively, to **INFINITELY** many k-points
 - » 25, 50, 100 points are usual;
longer electrode, fewer points needed

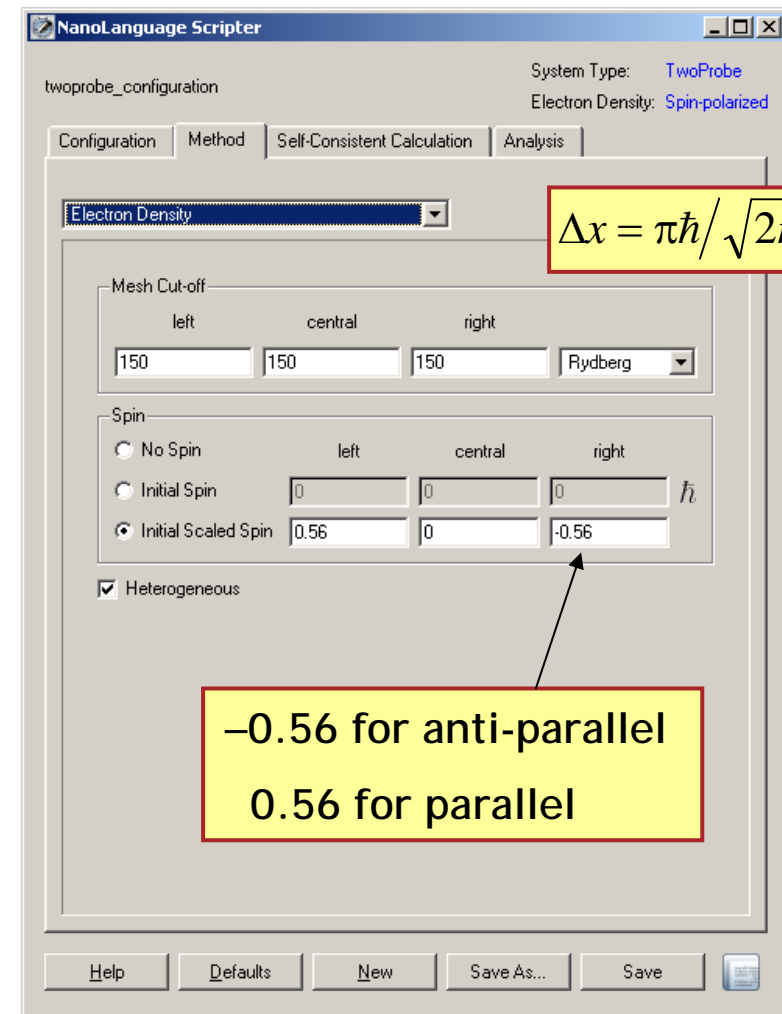
Eigenstate occupation

- ❖ Electron temperature
 - » Used in the electrode Fermi distribution
 - » No phonons...!
- ❖ Increase to cure convergence problems
 - » Smoother Fermi function
 - » 1000–1300 K usually does the trick
- ❖ **NOTE: Changing the temperature may change physical results (like the current) slightly!**
 - » Converge at high temperature, then anneal, to be safe



Electron density

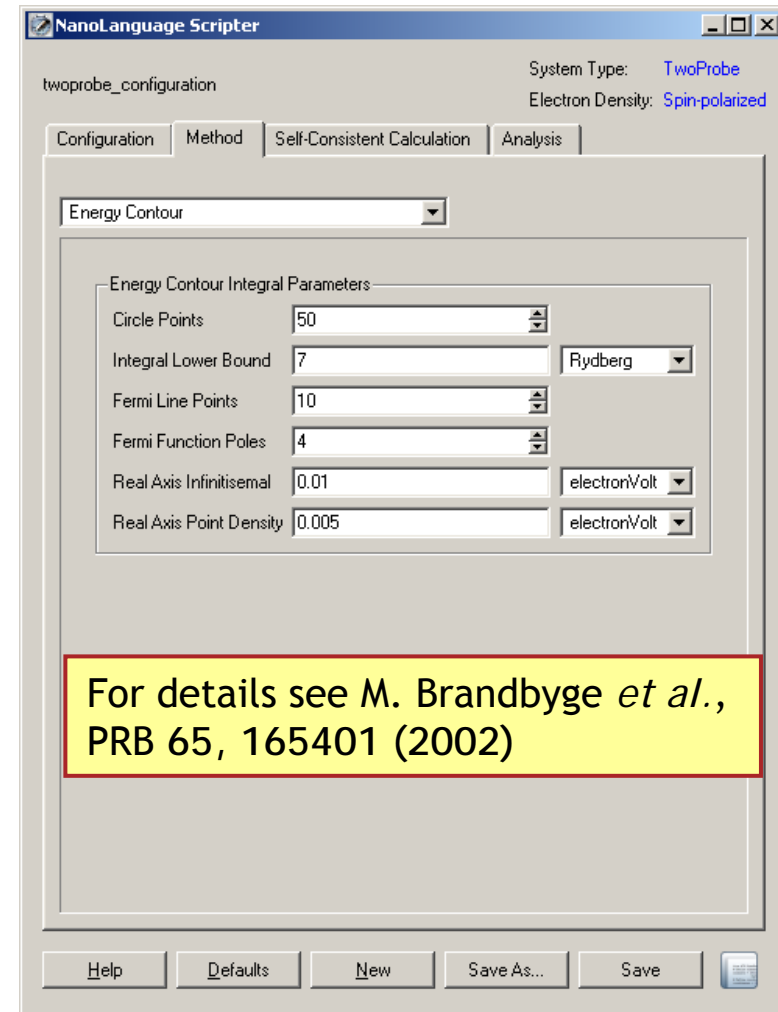
- ❖ Mesh cut-off
 - » Controls electrostatic mesh for Poisson equation
 - » Default usually fine for electron structure calculations
 - » d-elements like Fe need higher cut-off for good geometries (in optimizations)
- ❖ Spin
 - » Initial = absolute, in \hbar
 - » Scaled = fraction of max spin polarization for isolated atom $-1 \leq s \leq 1$
 - » To set spin for individual atoms, edit the script
- ❖ Each can be given for left/central/right independently
 - » Forces the system to be treated as heterogeneous



Contour integration

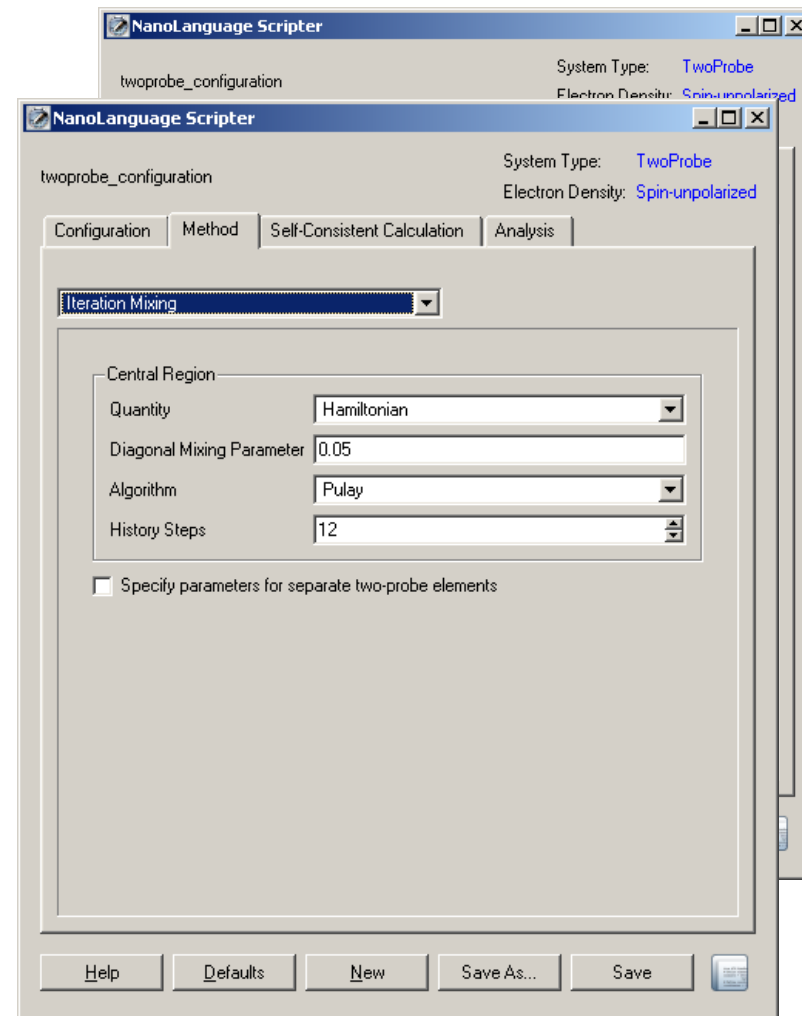
ATK is parallelized over the contour integration points

- ❖ Integral lower bound
 - » Increase (5-7 Ry) avoid poles below the contour, which lead to charge run-away (calculation converges to $q=0$)
- ❖ Circle points
 - » Increase (50-70) not to lose accuracy when increasing the length of the contour
- ❖ Real axis infinitesimal
 - » Can be increased in really difficult cases
 - » **Changes the results - anneal!**
- ❖ Real axis point density
 - » Lower to cure convergence problems
 - » Only relevant for finite bias



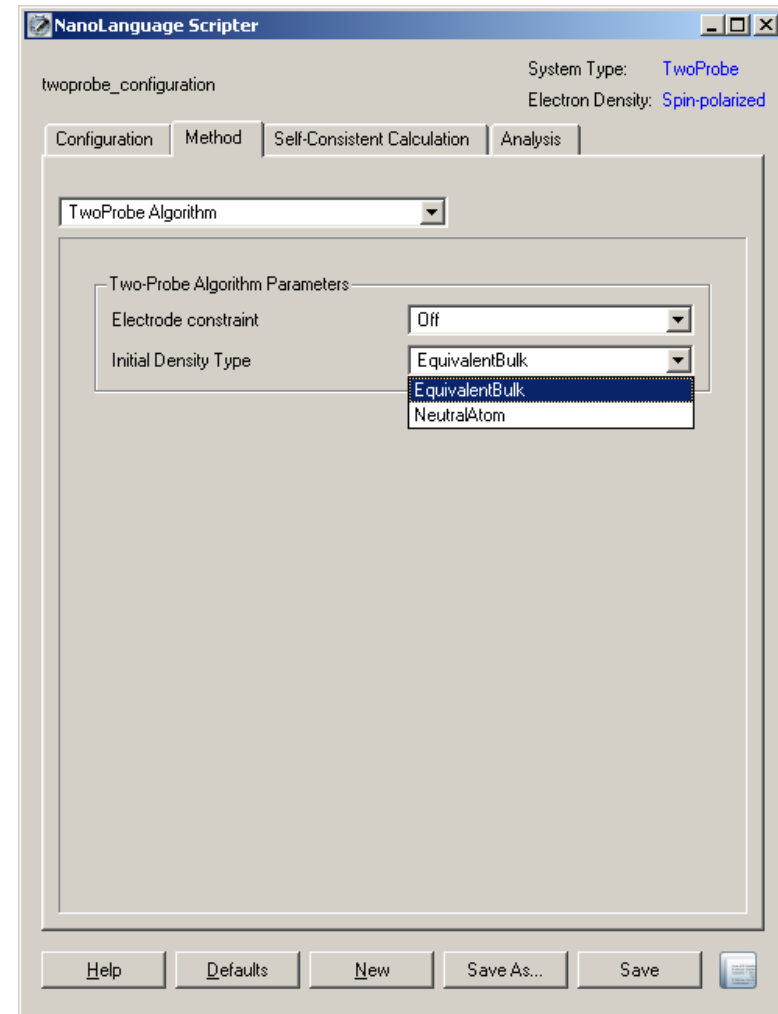
Mixing

- ❖ **Total energy** criterion can reduce iterations by a factor 2 without compromising the results
- ❖ Always use **Hamiltonian** mixing for two-probe systems
- ❖ **History Steps**
 - » Experiment with increasing to improve convergence (doesn't always work, sometimes fewer history steps are better...)
 - » Costs memory (but not a lot)
- ❖ **Diagonal mixing parameter (β)**
 - » Reduce β to stabilize convergence
 - » Increase β to speed up convergence (works well in simple systems like carbon nanotubes)
- ❖ **Advanced strategy for difficult cases:**
 - » Run with small β for some iterations (say, 5 or 20), then break
 - » Restart with larger β and run to convergence



Two-probe algorithm

- ❖ Electrode constraint
 - » Complex issue
 - » See separate slides for details
 - » Default = "Off", best workhorse for transmission and current
 - » Undocumented option "DensityMatrix" best, but can be hard to converge; only option for good voltage drop
- ❖ Initial density type
 - » EquivalentBulk
 - Bulk calculation used to initialize the two-probe density matrix
 - Often the best option!
 - » NeutralAtom
 - Heterogeneous systems



Checkpoint file

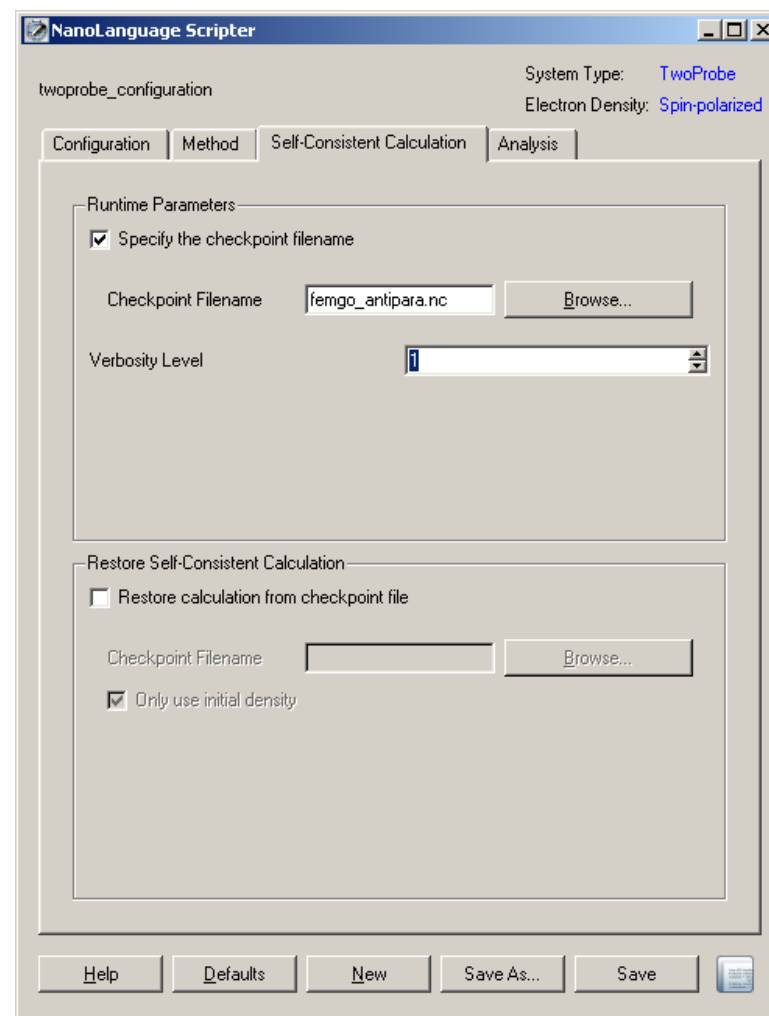
- ❖ NetCDF file stores all details of the calculation
 - » Restore for analysis
 - » Initialize density matrix for other calculations



WARNING: By default, no NetCDF file is created!



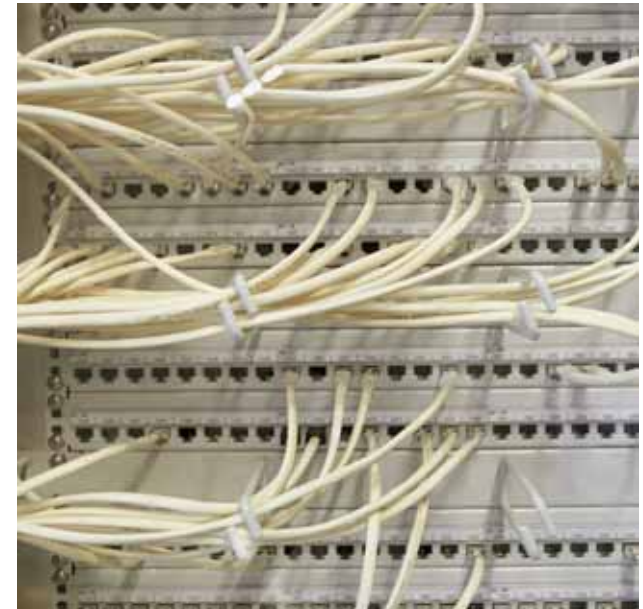
- ❖ Verbosity level
 - » Default in ATK = 0 (quiet)
 - » Default in VNL = 10 (all info)
 - » Level 1 is often best



Run the calculations!

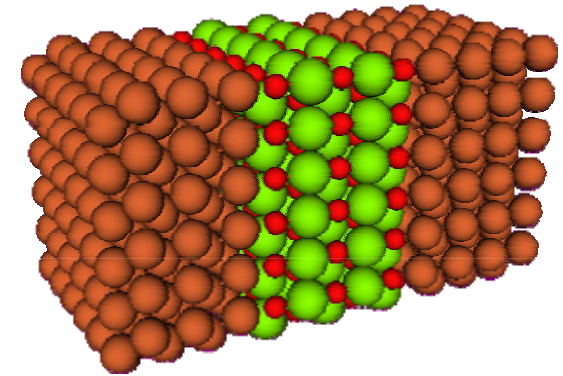
- ❖ Two calculations (special for MTJs)
 - » **Parallel:** 0.56 initial spin (right electrode/surface atoms)
 - » **Anti-parallel:** -0.56 initial spin (right electrode/surface atoms)
- ❖ Different filenames for the checkpoint files!

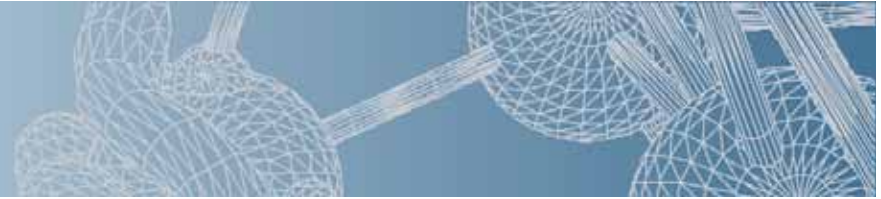
- ❖ Possible to run in serial
 - » Calculation uses < 1 Gb of RAM
 - » About 12 hours in serial (dual-core)
- ❖ Run in parallel!
 - » Use as many nodes as possible
 - » Excellent scaling because of many k-points and contour integration points
 - » Can cut the calculation time by order of magnitude



Summary

- ❖ To converge the FeMgO system, modify the following parameters:
 - » Basis Set
 - Type (for Fe) = SingleZetaPolarized
 - » Brillouin Zone Integration
 - Number of k-points (A/B/C) = 10/10/100
 - » Eigenstate Occupation
 - Electron Temperature = 1200 Kelvin
 - » Electron Density
 - Heterogeneous (check)
 - Initial Scaled Spin = 0.56 (left) and -0.56 (right)
 - » Energy Contour
 - Circle Points = 50
 - Integral Lower Bound = 7 Rydberg
 - Real Axis Points Density = 0.005 eV (for finite bias)
 - » Iteration Mixing
 - Diagonal mixing parameter = 0.05
 - » TwoProbe Algorithm
 - Initial Density = NeutralAtom
 - » Remaining parameters are left at default
- ❖ Geometry = default from MTJ Builder
- ❖ Set initial spin per atom in editor (parallel / anti-parallel)
- ❖ NetCDF file!





Outlook

❖ Part 2: Analysis

- » K-point resolved transmission coefficients
- » Transmission spectrum
- » Current & conductance
- » Tunneling magneto-resistance (TMR)
- » Scattering states