

# Tutorial: ART nouveau

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ART nouveau [1–4] is an open-ended search method for finding local transition states. It directly computes the direction of lowest curvature using the Lanczos algorithm, a very efficient iterative method that offers the fastest and most stable way to follow a local eigendirection. In this tutorial you'll use ART nouveau to find saddle points and neighbouring states in systems of crystalline silicon and iron.

## 1 Introduction

The tutorial was first run at the IAS vacation school on *Molecular and Materials Modelling* at the University of Warwick in September 2016. [Everything specific to the setup at the Centre for Scientific Computing at the University of Warwick is shown in blue.](#)

## 2 Setting up

The tutorial requires a LAMMPS-enabled ART nouveau (ARTn) binary. Instructions on how to compile are included in the official ART nouveau Guide (located in the DOC directory of the ARTn distribution). By coupling ARTn to LAMMPS[5], the wide variety of force fields accessible to the LAMMPS package become available for ARTn. This comes at a cost of a slightly more complicated setup. [The tutorial package is stored in phslay/ias\\_school/ on the CSC system. A CSC-compiled binary called artlammeps is located in the bin directory of the distribution. This binary requires that the intel compiler modules are loaded using the command](#)

```
module load intel/13.1 ompi/1.6.4/intel/13.1
```

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Please note that although the binary is MPI-enabled, the parallel functionality has not been tested and it is recommended that it is run in serial.

ART nouveau is controlled via input files and a startup script that communicates parameters via environment variables. Details are explained in Section IV B of the Guide.

The different section of this tutorial use the test calculations set out in the `example` directory of the distribution.

## 3 Vacancy in c-Si

The files necessary for this tutorial are in the `Si-vac-LAMMPS` subdirectory.

### 3.1 The system

Silicon has a face-centred diamond cubic crystal structure with 2 atoms in the fcc unit cell (and 8 atoms in the cubic cell). In this tutorial, we are looking at a  $4 \times 4 \times 4$  supercell of the cubic cell with periodic boundary conditions, from which one atom at the centre has been removed, for a total of 511 atoms.

### 3.2 Configuration files

The atom positions are reproduced twice: Once for ARTn in the file `refconfig`, once for LAMMPS in the `conf.sw` file. The latter is only used to initialise LAMMPS as a library, and only cell size and number of atoms are relevant from that file. Kinetic ART is controlled using the `bart.sh` shell script, which contains a number of variable-value pairs. The following variables are most relevant:

`NATOMS` Number of atoms, here 511.

`ENERGY_CALC` LAM for using the LAMMPS interface.

`REFCONFIG` Filename of atomic configuration. *Note:* This file is overwritten and always contains the current minimum.

`INPUT_LAMMPS_FILE` `in.lammps` is the name of the LAMMPS configuration file.

The final line contains a link to the binary, [artlammps](#), in the `bin` directory.

The atomic interaction is controlled using the file given as `INPUT_LAMMPS_FILE` in the main ARTn control script. LAMMPS reads its configuration from the file specified in the `read_data` line (here `conf.sw`). The interaction is given as a Stillinger-Weber potential (`pair_style sw`)[6], read from the `Si.sw` file (`pair_coeff`). This can easily be replaced by any other potential for which there exists a LAMMPS compatible implementation.

### 3.3 Running the Si-monovacancy

The simulation is started by executing the `bart.sh` shell script. This produces a considerable amount of output which can safely be stored away. The most important information is written to the log file `log.file.N`, where  $N$  is incremented for each run of ARTn. Typical output of an event creation looks like this:

```
-----
- Simulation          : 1
- Attempt            : 8
- Starting from minconf : 1001
- Reference Energy (eV) : -2.2133458330E+03
- Temperature        : -0.500000

- That atom          : 131
      E-Eref  m_perp  ftot   fpar  fperp   eigen   delr  npart  evalf  a1
      ( eV )                ( eV/Ang ) ( eV/Ang**2 )
0  K=  0  0.0117  2  2   0.5807  -0.5594  0.1558  0.0000  0.061  0  1974  0.00
1  K=  1  0.3979  2  2   3.3307  -3.0770  1.2751  0.0000  0.372  4  1977  0.00
2  K=  2  1.4167  2  3   6.5783  -5.3099  3.8832  0.6458  0.717  9  1998  0.00
3  K=  3  2.4954  2  3   6.9707  -6.5457  2.3970  0.5665  1.024  12 2019  0.85
4  K=  4  3.9188  2  3   7.4241  -7.0875  2.2103  0.2142  1.365  14 2040  0.30
5  K=  5  5.6666  2  2   9.4164  -8.3046  4.4386 -0.0417  1.692  19 2060  0.94
6  K=  6  7.3354  2  3   9.9064  -8.6056  4.9071 -0.4800  2.007  22 2081  0.89
7  K=  7  8.8148  2  3   8.8463  -8.2094  3.2957 -2.5164  2.298  23 2102  0.74
13 L=  1  9.1057  0  0   8.8024  -0.9560  8.7443 -4.4255  2.315  22 2126  0.97
24 L=  2  8.3931  0  0   7.2756  -0.5579  7.2752 -3.5342  2.129  23 2155  0.91
40 L=  3  6.5462  0  0   4.4553  -0.6841  4.4094 -2.3484  1.822  22 2189  0.95
61 L=  4  4.2090  0  0   2.7841  -0.0866  2.7752 -1.2234  1.674  15 2228  0.98
87 L=  5  3.0632  0  0   0.8334  -0.0170  0.8331 -0.6060  1.752  19 2272  0.99
118 L=  6  2.9657  0  0   0.2045  -0.0024  0.2045 -0.3908  1.768  18 2321  1.00
154 L=  7  2.9594  0  0   0.0323  -0.0004  0.0323 -0.3709  1.774  19 2375  1.00

SADDLE 1001 CONVERGED |ret 20154 |delta energy= 2.9592 |...
...force_(tot,par,perp)= 0.0323 -0.0004 0.0323 |eigenval= -0.3709 |...
...npart= 19 |delr= 1.774 |evalf= 2375 |
- Configuration stored in file : sad1001
- Total energy Saddle (eV) : -2.2103866708E+03

RELAXATION
159 M= 5 2.9592 0 0 0.0323 -0.0004 0.0323 -0.3709 0.001 0 2381 0.00
- Configuration stored in file : min1001
- Total energy Minimum (eV) : -2.2103882477E+03
MINIMUM 1001 REJECTED |E(fin-ini)= 2.9576 |E(fin-sad)= -0.0016 |...
...npart= 20 |delr= 1.869 |evalf= 2385 |
Starting Lanczos
Em= *****
Iter   Ep-Em (eV)   Eigenvalue  a1
  1    9.51E-05    0.614790   0.0000
  2    9.48E-05    0.173168   0.4452
Done Lanczos
```

After some introductory banter, kART first attempts to leave the harmonic well (K=), for an activation centred on atom 131. As soon as a sufficiently negative eigenvalue (eigen column) is found, it switches to Lanczos (L=), which is then performed for

7 steps, until the parallel and perpendicular forces (`fpar` and `fperp`) drop below a threshold. This succeeds, and the saddle point is accepted at an energy of 2.652 eV above the ground state. The system is then pushed across the saddle point and minimised to an energy of just 0.0016 below the saddle point. ARTn also wrote minimum and saddle point configurations to file.

In this case ARTn found an event that is essentially the creation of an interstitial-vacancy pair, with a precariously metastable final state. All events are recorded in the file specified as the `EVENTSLIST` option (here: `events.list`). This file only contains the labels of the minima and saddle points involved. To produce more insightful information the `awk` script `create_events.awk` is provided in the `utils` directory. This can create xyz format files of events (i.e. containing snapshots of initial, saddle and final state) and provide additional information about the barriers ARTn found so far. The resulting `event_nnnnn.xyz` files can be visualised with `jmol` (`jmol event_nnnnn.xyz`).

The output from `create_events.awk` also helps identify actual vacancy migration events. They will have barriers that are more or less symmetric, and only a small difference in energy between initial and final state.

## 3.4 Further suggestions

### 3.4.1 Vacancy aggregation

Remove more atoms from the unit cell (nearest neighbours, second nearest neighbours) and attempt to find out how the states are connected.[7]

### 3.4.2 Convergence failures

Looking at the log file, you will notice that some of the attempts to find a saddle point failed. The reason of failure is encoded in the `ret` value. Values over 60000 indicate a loss of negative eigenvalue. Values over 50000 indicate that the system ran out of activation steps (`Activation_MaxIter`) before reaching the saddle point. How could you influence the success rate of the search? At what cost?

## 4 Vacancy in bcc Fe

### 4.1 The system

Iron in the  $\alpha$  phase forms a bcc crystal (2 atoms in the cubic cell). Here we are looking at a  $5 \times 5 \times 5$  supercell of the cubic cell from which one atom near the centre has been removed, for a total of 249 atoms.

### 4.2 Configuration files

The setup is very similar to the one described for Si – main changes are the initial file and the `lammps pair_style` options. This tutorial actually contains two effective EAM potentials for iron, `Fe_2.eam.fs` [8] and `Fe-P.eam.fs` [9] (source information contained in the files), which can easily be swapped in the `in.lammps` file.

### 4.3 Running the Fe-monovacancy

Run the simulation with either of the potentials. How do the results compare? What do you note about the lowest barrier events that are found?

Both sets of potentials have an artefact: There is a metastable state at the split vacancy position (i.e. an atom straddles two neighbouring empty lattice sites). This is remedied in some other potentials, for which, alas, a LAMMPS compatible potential file could not be located in time.

The iron simulations have a much lower probability to succeed, as the interstitial-vacancy creation event is not accessible to ARTn (why?). This means that event searches can only be successful in the vicinity of the vacancy. To limit your searches to those centred a certain atom, you can use the `Central_Atom` parameter, for example to search only for an atom nearest to the vacancy.

Further reading: [10]

### 4.4 Further suggestions

#### 4.4.1 Fe interstitials

Interstitials show a rich configuration space. [11, 12] To have a look at this, first add the removed atom back to the system (at position 6.424425 6.424425 6.424425). Then add another atom anywhere not overlapping with another atom. ARTn will first minimise the system, so the exact location is not that important.

## 5 MC simulation

ARTn allows for crude Monte Carlo simulations, by accepting a converged event with a Metropolis Monte Carlo criterion. This is enabled by setting a positive `Temperature` (in electronvolt) in the `bart.sh` file. While this is a far call from a full KMC simulation, it allows for sampling of configuration space.

## 6 Summary

This tutorial provides a brief taste of ART nouveau. ART nouveau is used as a saddle point finding method in kinetic ART [12, 13], an adaptive Kinetic Monte Carlo code.

## References

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