Molecular Dynamics Simulation of MoS_2 Exfoliation with Water/Iso-proponal

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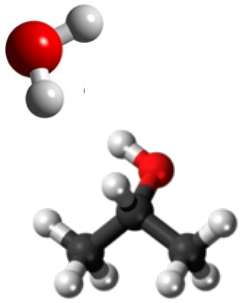
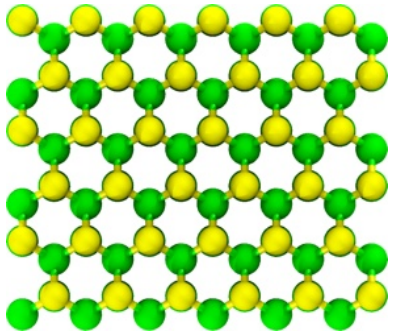
Simulation Outline

1) Create and relax the mixture (Water/IPA) and MoS_2 separately
2) Combine them together and relax
3) Create bubbles and apply shock
4) Analysis

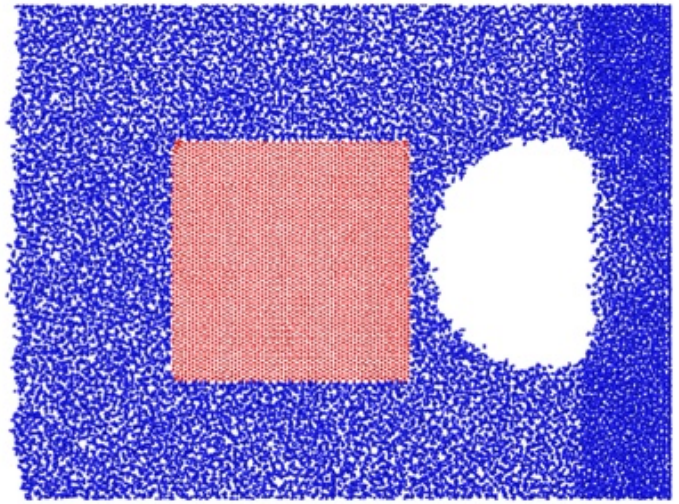
Water



Iso-proponal



MoS_2 Crystal



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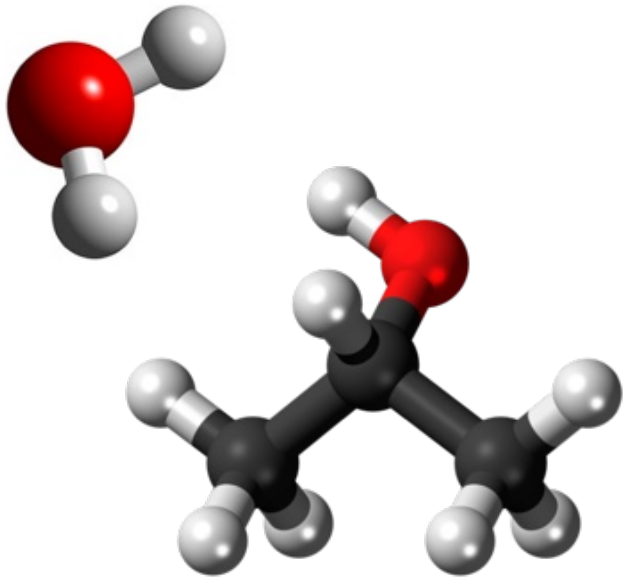
Create the mixture

Water/IPA mixture



Water: TIP4P/2005

3 atoms, 2 types of atoms
2 bonds, 1 type of bond
1 angle, 1 type of angle

IPA: OPLS-AA

12 atoms, 5 types of atoms
11 bonds, 5 types of bond
19 angles, 7 types of angles
21 dihedrals, 5 types of dihedrals

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Create the mixture

- **Create the mixture system with no overlapped molecules, and shrink the system to normal density**

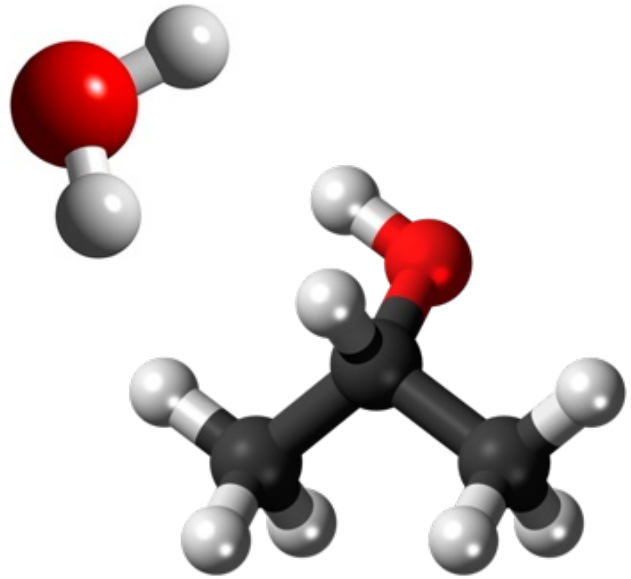
Atom read format

```
Atoms # full

     1      1  1    0.00     1.550     1.550     1.500
     2      1  2    0.00     1.550     2.307     2.086
     3      1  2    0.00     1.550     0.793     2.086
     4      2  3    0.00     6.033     2.429     1.580
     5      2  7    0.00     6.665     1.124     1.090
```

```
Bonds

1 1 1 2
2 1 1 3
3 2 4 8
4 2 4 9
```

```
Angles

1 1 2 1 3
2 2 5 7 15
3 3 4 5 7
4 3 6 5 7
```

```
Dihedrals

1 1 4 5 7 15
2 1 6 5 7 15
3 2 11 5 7 15
4 3 7 5 4 8
```



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Create the mixture

- **Relax the mixture to normal density using NPT ensemble**

```
1   units real
2   atom_style full
3   boundary p p p
4   processors * * *
5   region mybox block 0.0 100.0 0.0 100.0 0.0 100.0 units box
6   create_box 9 mybox bond/types 6 angle/types 8 dihedral/types 5 &
7                      improper/types 0 extra/bond/per/atom 4 &
8                      extra/angle/per/atom 13 extra/dihedral/per/atom 21
9   read_data ./data.solvent
10  include system.ff
11  include system.in.charges
12  fix constrain all shake 1.0e-4 100 0 b 10 a 23
```

Define the box

Force Field

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Create the mixture

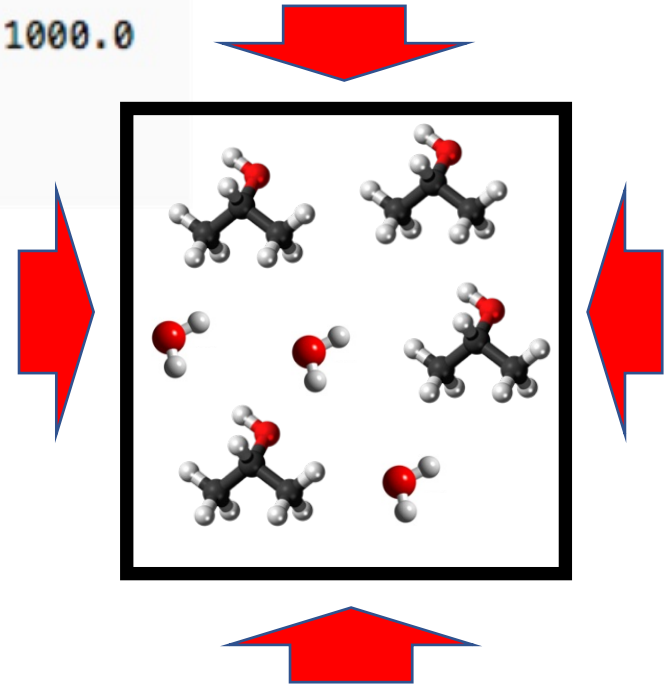
```
14  neighbor 2.0 bin
15  neigh_modify every 10 delay 0 check yes
16  timestep 2.0
17
18  thermo_style custom step temp pe ke etotal press vol density
19  thermo_modify flush yes
20  thermo 100
21
22  variable Text equal 300.0
23  variable Pext equal 0.0
24  fix npt all npt temp ${Text} ${Text} 100.0 iso ${Pext} ${Pext} 1000.0
25  run 10000
26
27  write_restart restart.solvent
```

Neighbor Lists
Timestep

Monitor global info

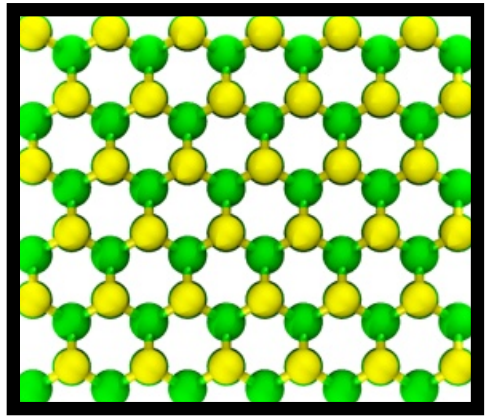
NPT ensemble

Save state

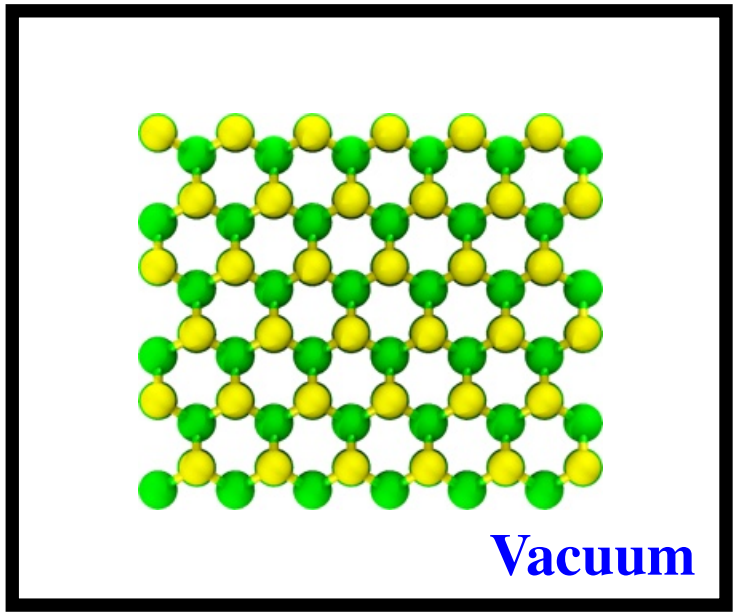


Create MoS_2

- Create MoS2 crystal
- Introduce vacuum
- Apply conjugate gradient to relax the system
- Heat the system to desired temperature and thermalize the system



MoS_2 Crystal



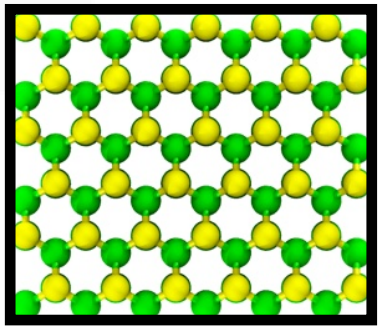
Create MoS_2

Define the crystal unit cell

```
12 lattice custom 1.0 &
13 orient x 1 0 0 &
14 orient y 0 0 -1 &
15 orient z 0 1 0 &
16 a1 3.17 0.0      0.0 &
17 a2 0.0  5.490601 0.0 &
18 a3 0.0  0.0      12.288 &
19 basis 0.00000    0.00000    0.25000 &
20 basis 0.50000    0.50000    0.25000 &
21 basis 0.50000    0.16670    0.75000 &
22 basis 0.00000    0.66670    0.75000 &
23 basis 0.00000    0.00000    0.62750 &
24 basis 0.50000    0.50000    0.62750 &
25 basis 0.50000    0.16670    0.37250 &
26 basis 0.00000    0.66670    0.37250 &
27 basis 0.50000    0.16670    0.12750 &
28 basis 0.00000    0.66670    0.12750 &
29 basis 0.00000    0.00000    0.87250 &
30 basis 0.50000    0.50000    0.87250
```

Create MoS_2

```
48 create_atoms 9 box &
49 basis 1 8 &
50 basis 2 8 &
51 basis 3 8 &
52 basis 4 8 &
53 basis 5 9 &
54 basis 6 9 &
55 basis 7 9 &
56 basis 8 9 &
57 basis 9 9 &
58 basis 10 9 &
59 basis 11 9 &
60 basis 12 9
```



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Create MoS_2

```
62  variable x1 equal xlo-10.0
63  variable x2 equal xhi+10.0
64  variable y1 equal ylo-10.0
65  variable y2 equal yhi+10.0
66  variable z1 equal zlo-10.0
67  variable z2 equal zhi+10.0
68
69  change_box all x final ${x1} ${x2} y final ${y1} ${y2} z final ${z1} ${z2} units box
```

Introduce Vacuum

```
77  thermo_style one
78  min_style cg
79  thermo 2
80  minimize 1.0e-4 1.0e-6 1000 1000
81
82  reset_timestep 0
83  velocity        all create 10.0 156467 mom yes rot yes dist gaussian
84  #dump dfiles all custom 1000 MoS2.lammpstrj id type x y z vx vy vz
85  timestep 1.0
86
87  fix nvt all nvt temp 10.0 300.0 100.0
88  run 200000
89  unfix nvt
90  write_data data.MoS2 nocoeff
```

Conjugate Gradient to relax the system

NVT ensemble to heat the system up to 300K

Simulation Outline

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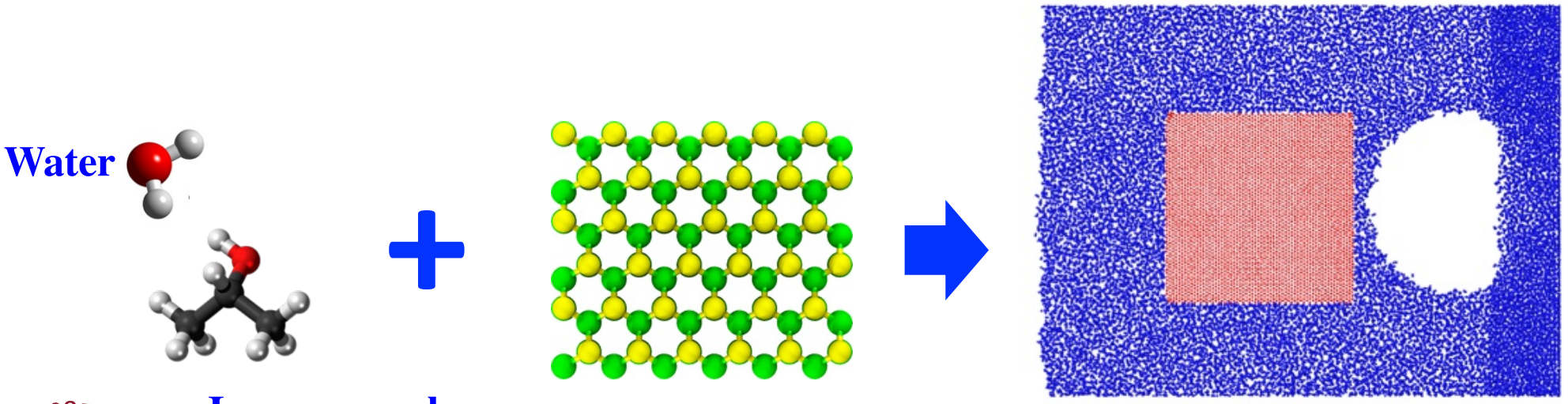

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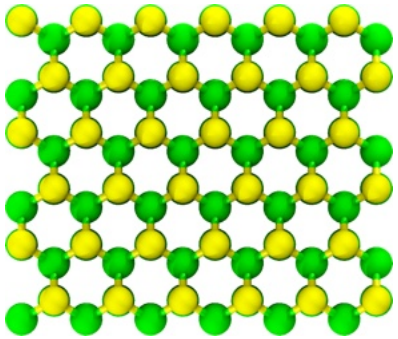
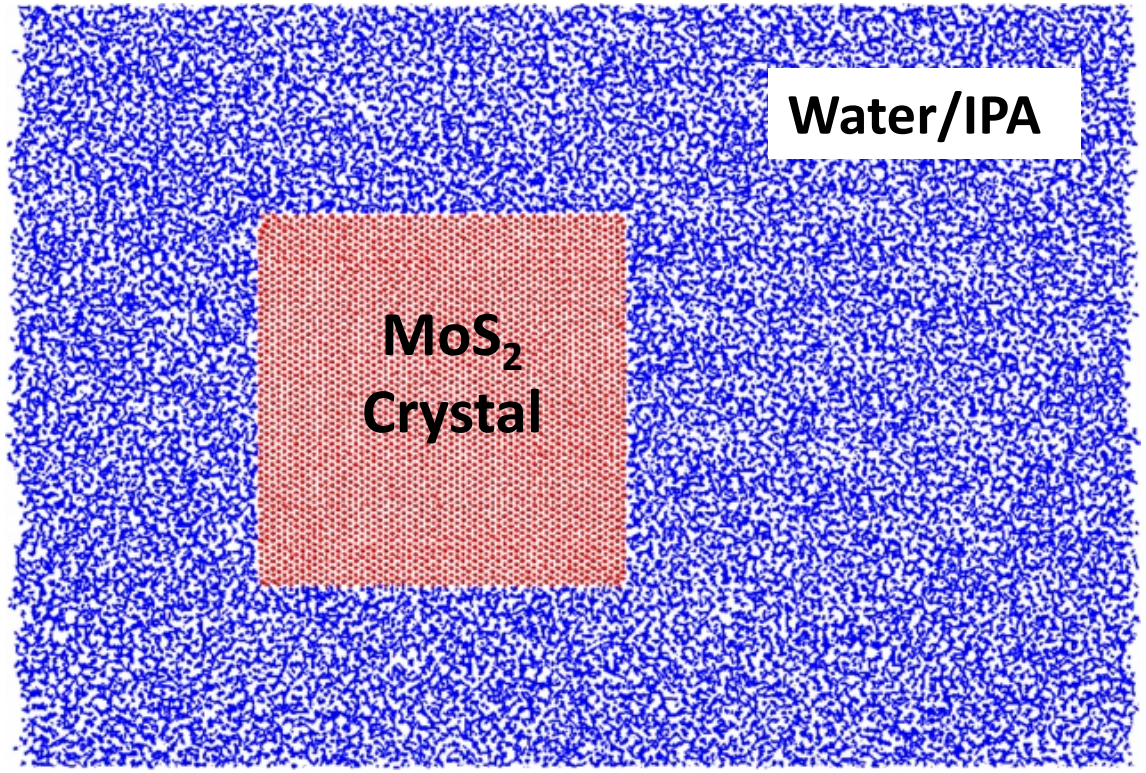
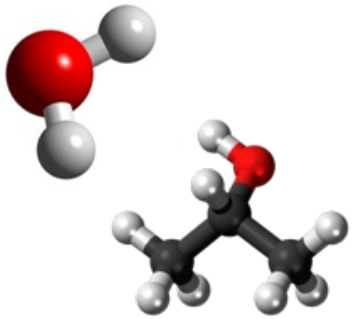
Combine mixture and MoS_2

- Combine the two systems together
- Remove overlapping liquid molecules
- Relax and thermalize the combined system







Combine mixture and MoS_2

```
30  neigh_modify every 8 delay 0 check yes one 5000
31  read_data ../MoS2/data.MoS2 add append shift 63.5 63.5 50.0
32
33  reset_timestep 0
34  set group MoS2 mol 0
35  group solvent type 1 2 3 4 5 6 7
36  group MoS2 type 8 9
37
38  delete_atoms overlap 4.0 solvent MoS2 mol yes
39  timestep 1.0
40  neigh_modify every 8 delay 0 check yes one 3000
```

Remove overlapping molecules

```
51  fix freeze solvent setforce 0.0 0.0 0.0
52  thermo 1
53  thermo_style one
54  min_style cg
55  minimize 1.0e-2 1.0e-2 10 10
56  unfix freeze
57  reset_timestep  0
```

Conjugate Gradient to relax MoS_2

Combine mixture and MoS_2

```
52  velocity solvent scale 300.0
53  velocity MoS2 scale 300.0
54
55  compute T_s solvent temp
56  fix Temp_s solvent ave/time 10 10 100 c_T_s
57  compute T_mos2 MoS2 temp
58  fix Temp_MoS2 MoS2 ave/time 10 10 100 c_T_mos2
59
60  thermo_style custom step temp f_Temp_s f_Temp_MoS2 pe etotal press pxx pyy pzz
61  thermo_modify flush yes
62  thermo 100
63
64  fix   nvt1 solvent  nvt temp 300.0 300.0 50.0
65  fix   nvt2 MoS2     nvt temp 300.0 300.0 50.0
66  run 1000
```

Monitor system

NVT ensemble
two thermostats

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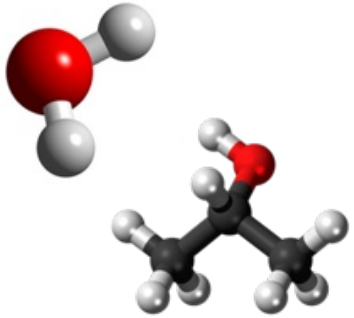
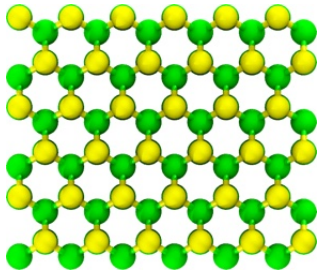
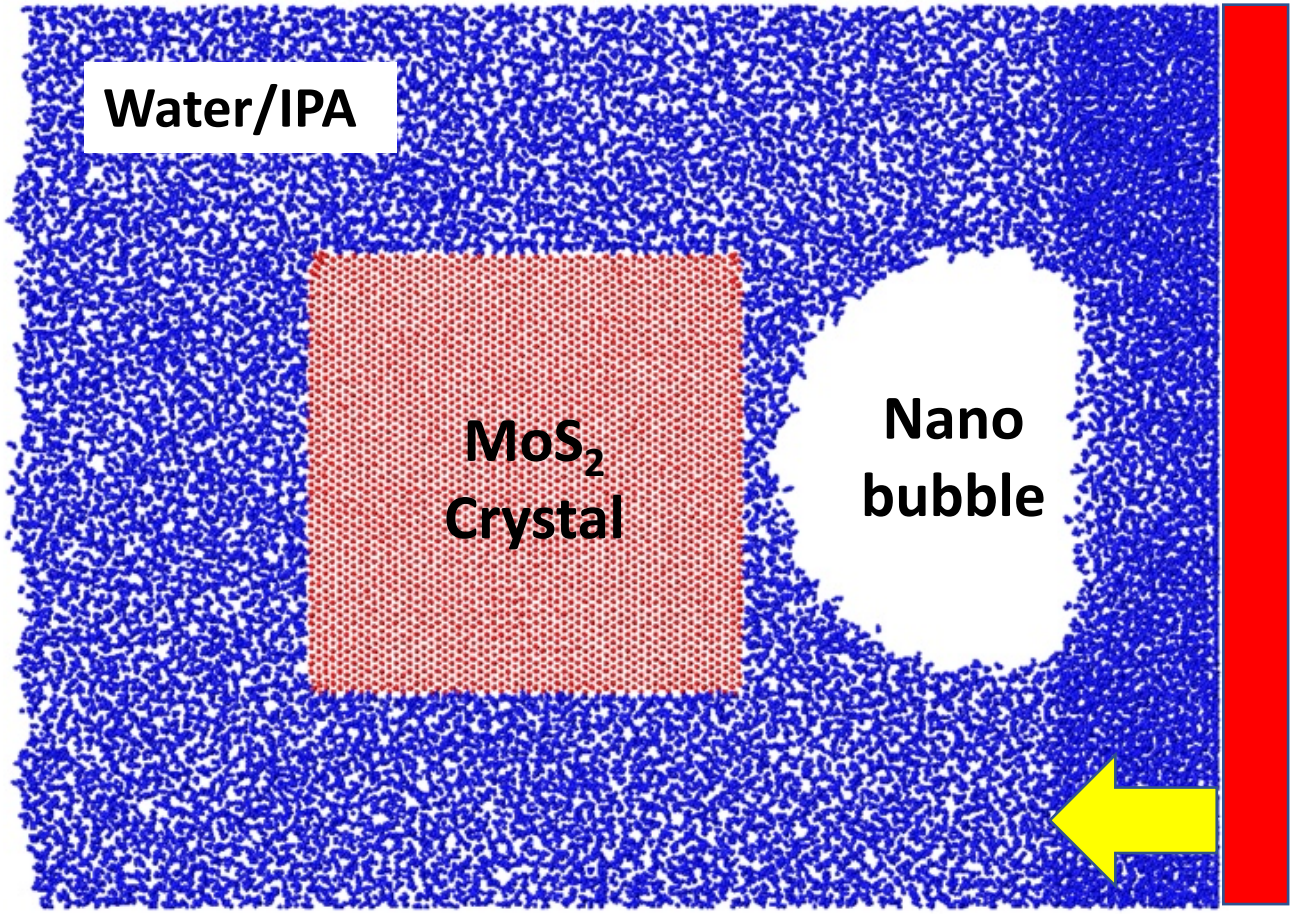
Create bubble then apply shock







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Create bubble then apply shock

```
9  variable z1 equal 2.0
10 variable z2 equal zhi-2.0
11 region left block INF INF INF INF ${z1} ${z2} units box
12 group left region left
13 group del subtract all left
14 delete_atoms group del mol yes
15
16 change_box all boundary p p f
17 kspace_modify slab 3.0
18 fix ceil all wall/reflect zhi EDGE zlo EDGE
```

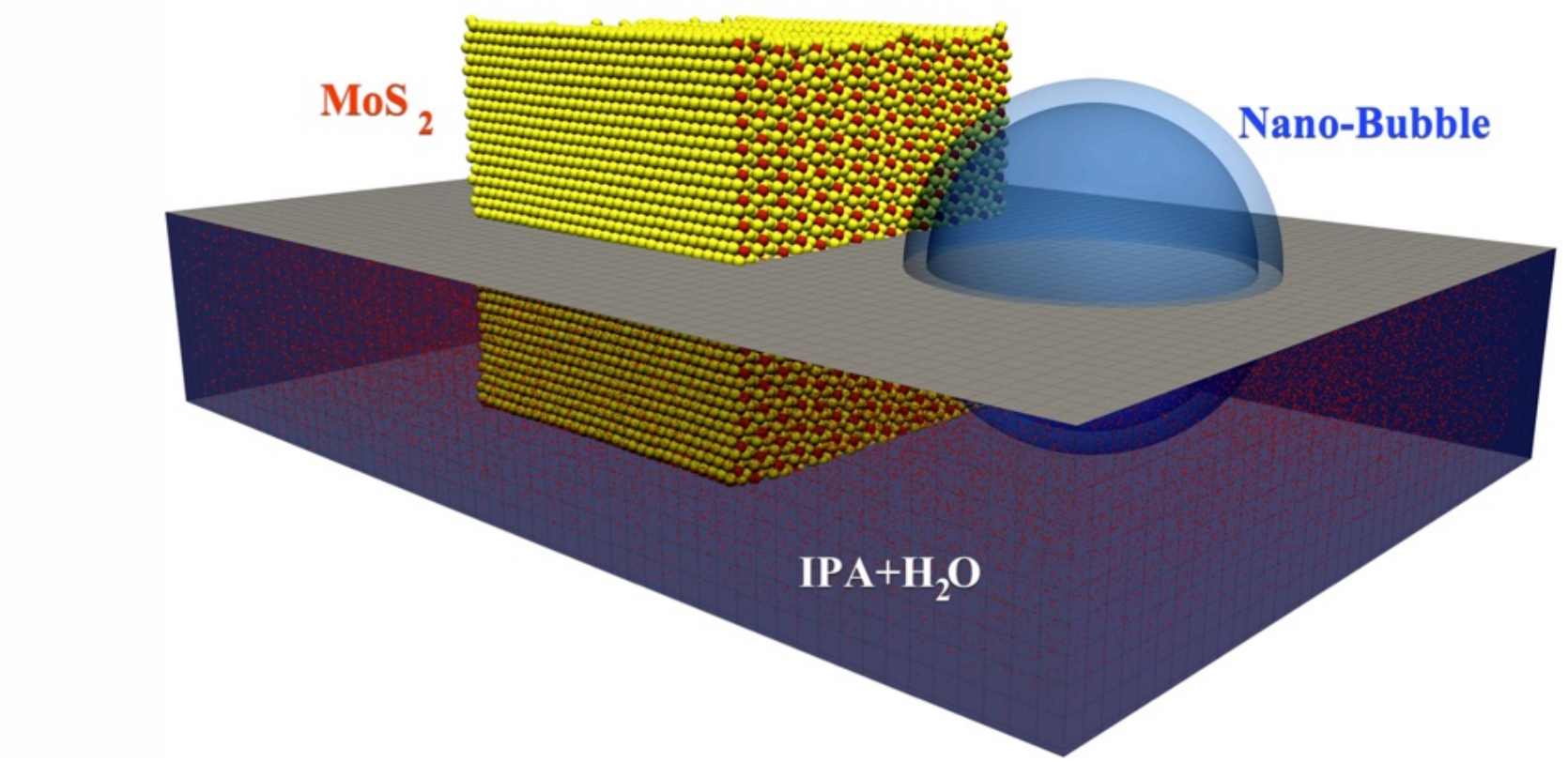
Remove the water/IPA molecules crossing periodic boundary in the shock direction

```
38 velocity all set NULL NULL ${vs} sum yes units box
```

```
30 region bubble sphere ${bx} ${by} ${bz} 40.0 units box
31 delete_atoms region bubble  mol yes
```

Create bubble and apply shock

Results: Exfoliation Movie



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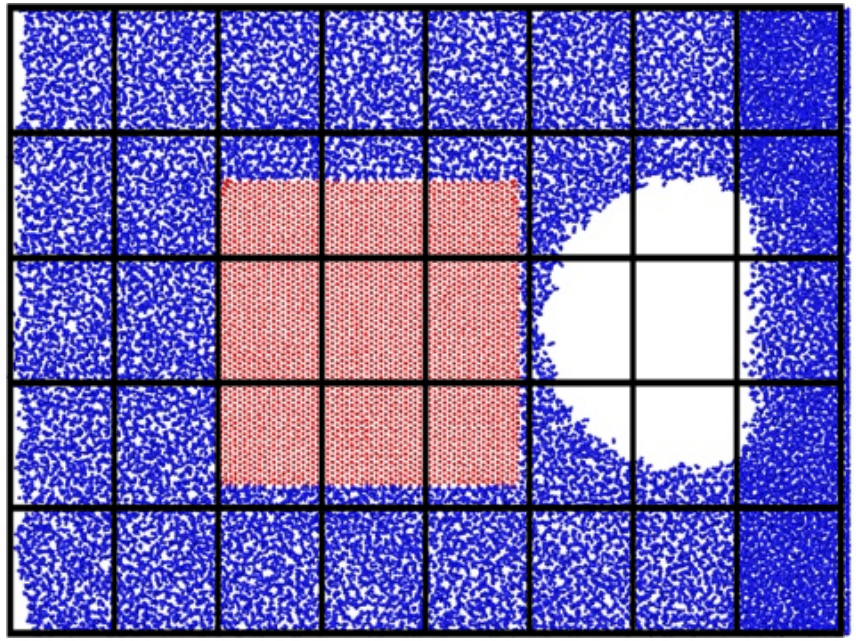
Analysis

- Local temperature, pressure, shear stress in MoS_2 to find exfoliation mechanism
- Surface area, volume, Convex hull's volume/area to determine exfoliation yield



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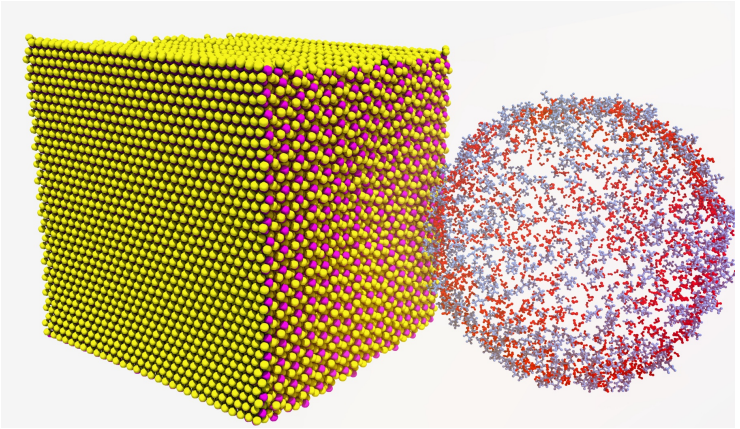
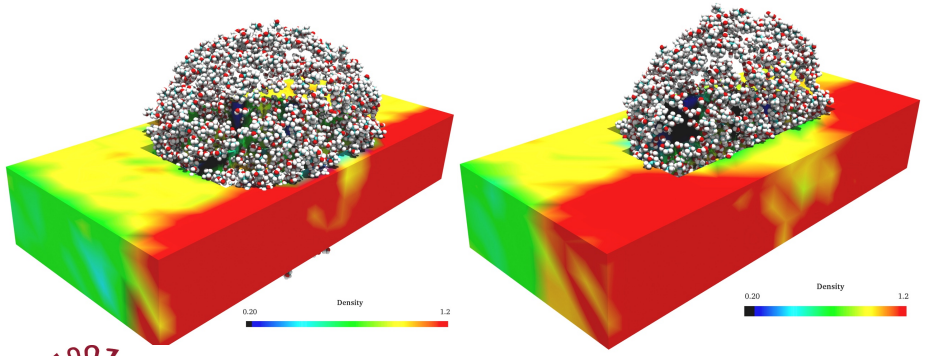
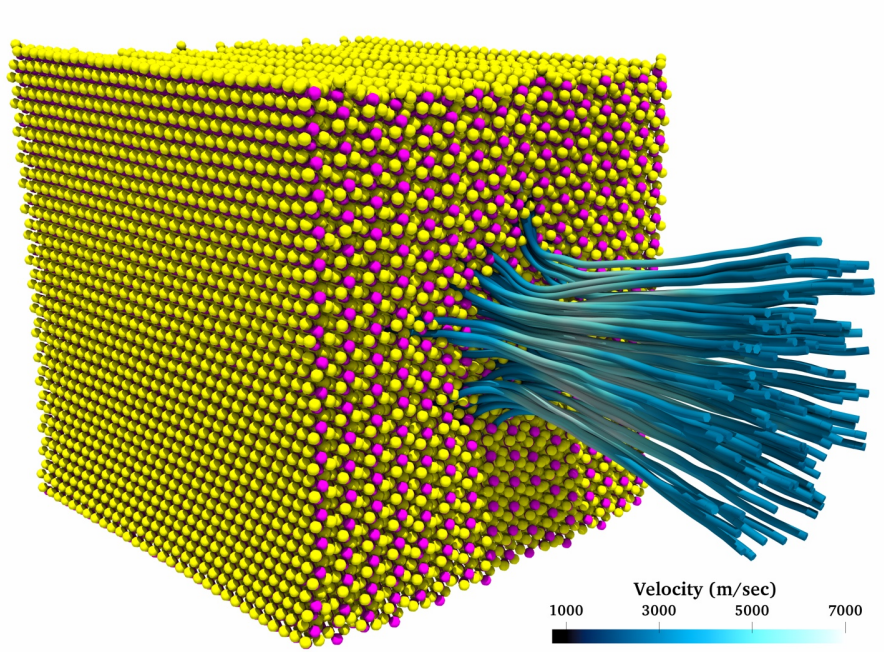
Bubble Collapse and Nanojet Formation

Bubble collapse





Nanojet formation by bubble collapse



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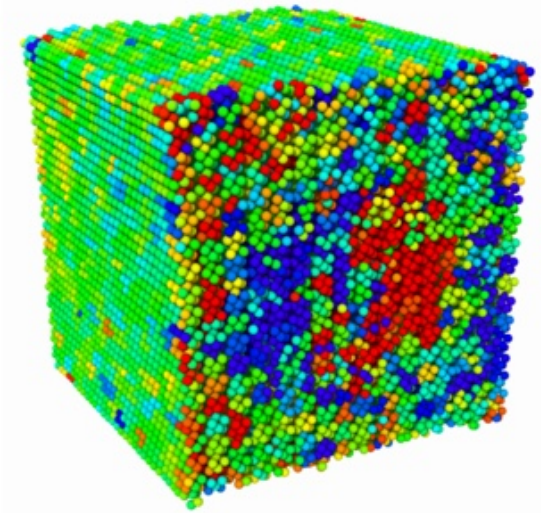
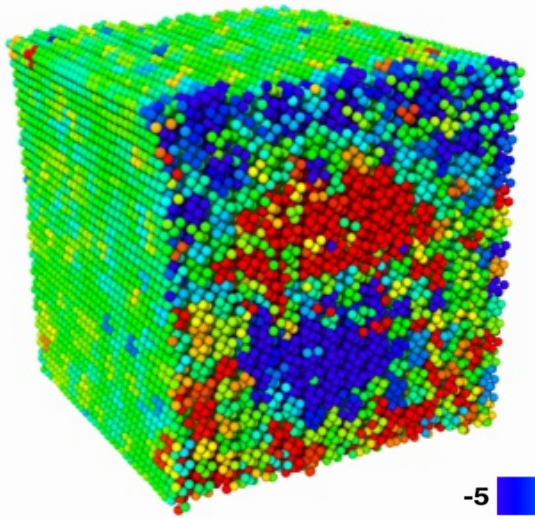
Local Temperature and Stresses

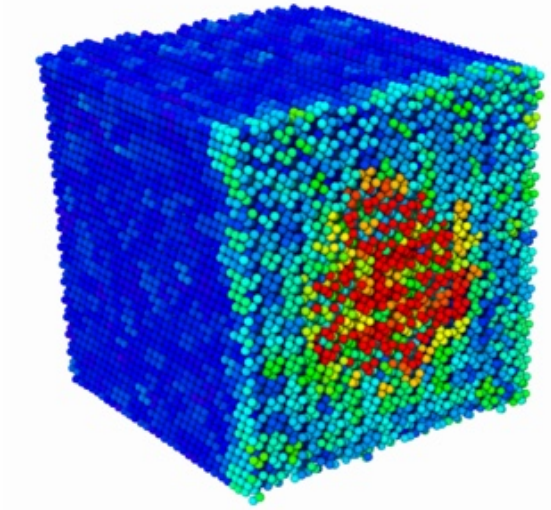
π_{xz}

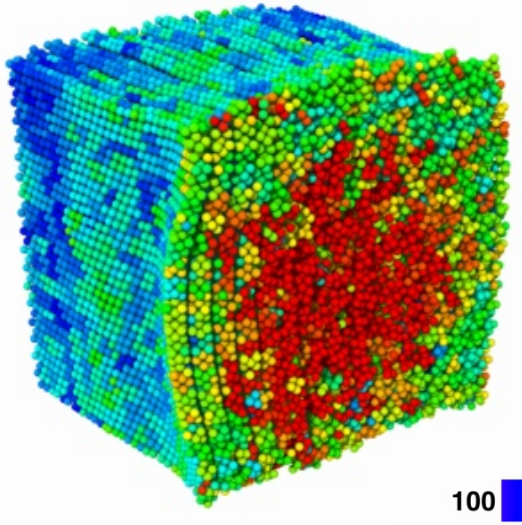


π_{yz}

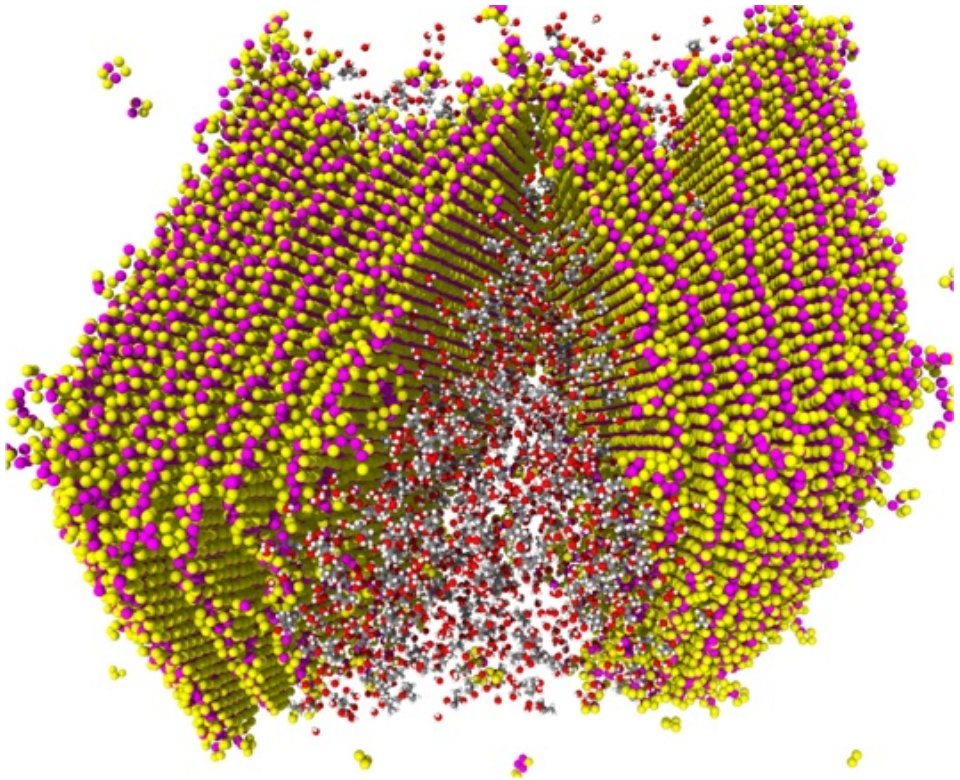


Temperature



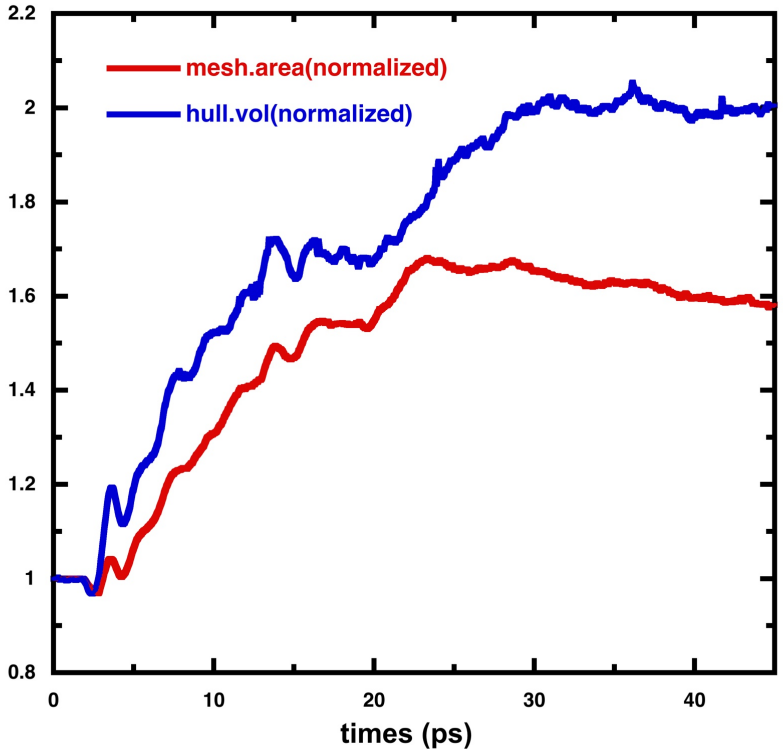


Exfoliation Yield



Exfoliated MoS_2

MoS_2 surface area and volume after exfoliation



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Exfoliation : Hands-on

Copy tar ball of the exfoliation hands-on to your staging directory and untar it.

```
$ cp ~magics35/magics/exfoliation.tar.gz .
$ tar xvfz exfoliation.tar.gz
```

It will create **exfoliation** directory that contains three subdirectories (**create**, **bubble_collapse**, **jet**) and custom LAMMPS executable (**lmp_mpi**)

```
$ ls -F exfoliation
bubble_collapse/  create/  jet/  lmp_mpi*
```

Go to **exfoliation/jet/** and submit job

```
$ cd exfoliation/jet/
$ qsub job.pbs
```

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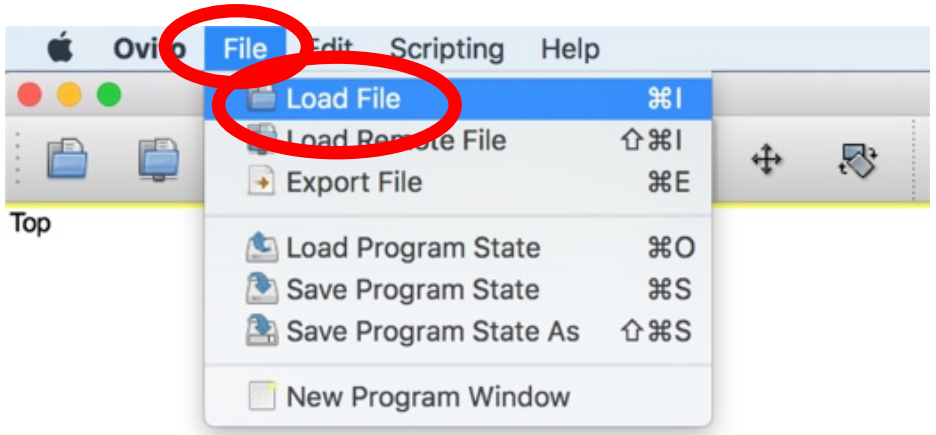
Exfoliation : Hands-on

After the job finishes, you will have several LAMMPS trajectory files.

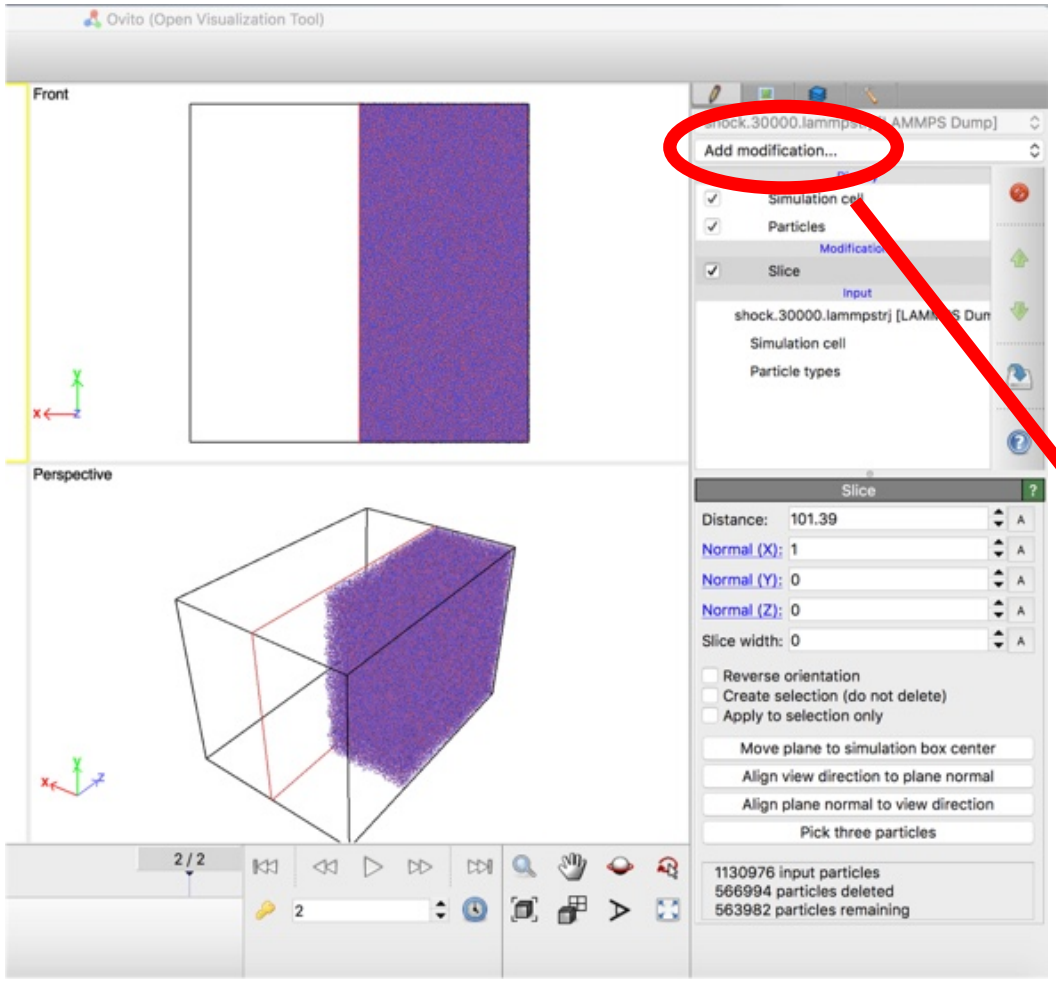
```
$ ls data/*.lammpstrj
$ data/shock.30000.lammpstrj  data/shock.30100.lammpstrj
```

Copy the LAMMPS trajectory files to your laptop and visualize the trajectories using OVITO.



1. File
2. Load File
3. Select **shock.30100.lammpstrj**

Exfoliation : Hands-on



1. Click **Add modification..**
2. Go down the list and select **Slice**