MD Setup

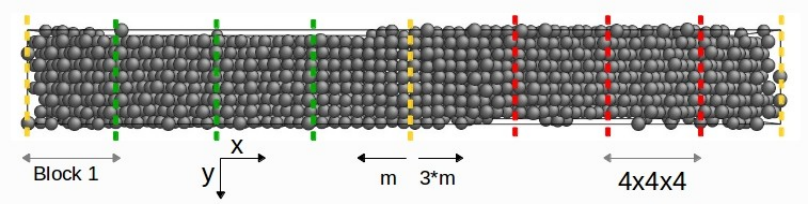


Figure 1: MD domain for interface study of fcc Argon at 20K with 2048 atoms

```
##########MEASURE SED#################################################################
label loop_seed
variable iseed loop 5
variable seed index 11111 22222 33333 44444 55555
clear

#------------READ STRUCTURE---------------------------------------------------------
units lj
atom_style atomic
read_data interface.data
#group                  Ar1 type = 1
#group                  Ar3 type = 3
group c1 id <= 256
group c2 id <> 257 512
group c3 id <> 513 768
group c4 id <> 769 1024
group c5 id <> 1025 1280
group c6 id <> 1281 1536
group c7 id <> 1537 1792
group c8 id >= 1793
#------------LJ Potentials----------------------------------------------------------------------
pair_style lj/cut 2.5
#pair_coeff 1 1 1.0 1.0
pair_coeff * * 1.0 1.0
pair_modify             shift yes
#------------Variables--------------------------------------------------------------------------
#------------LJ Parameters
variable     kB  equal 1.3806504e-23  # [J/K] Boltzmann
variable sigma_Ar  equal 3.4e-10  # m
variable eps_Ar equal 1.67e-21 # J
variable mass_Ar         equal 6.63e-26 # kg
variable tau_Ar equal 2.1423e-12 # s
#------------thermo Parameters
variable     T_melt          equal 300*(${kB}/${eps_Ar})
variable T_0K equal 0.001

variable    dt  equal 0.002
variable quench_rate equal 1.0
variable quench_length   equal 10000
#------------kappa parameters
```

```
variable     p  equal 100000  # correlation length
variable     s  equal 10   # sample interval
variable     d  equal $p*$s  # dump interval
variable scaleJ equal ${eps_Ar}*${sigma_Ar}/${tau_Ar}

#-------------SED parameters
variable w_step equal 2^5 #32 steps
variable w_total         equal 2^16 #65 some 000 steps
variable t_total         equal 2^20 #1048576 steps
variable num_ffts equal ${t_total}/${w_total}

#label loop2
#variable b loop 3
#variable T_run index ${T_1} ${T_2} ${T_3}

variable  T_run  equal 20*(${kB}/${eps_Ar})
variable alat equal 1.5635

#log  log_quench_$a_$b.lammps
log  log_heat_${iseed}.lammps

#------------ NVE rescale -----------------------------------------------------------------

velocity  all create ${T_run} ${seed} rot yes dist gaussian

fix  1 all nve
fix  2 all temp/rescale 1 ${T_run} ${T_run} 0.01 1.0
timestep ${dt}
thermo_style   custom step temp press etotal vol
thermo 1000
        dump 2 all cfg 2000 pos.*.cfg id type xs ys zs # Visualize with AtomEye
run              500000
# run              1000
unfix  1
unfix  2

#------------ NVE -------------------------------------------------------------------------

fix  1 all nve
timestep ${dt}
thermo_style   custom step temp press etotal vol
thermo 1000
run              500000
# run              10000
unfix  1

#------SED--------------------------------------------------------------------------

label loop_fft
variable ifft loop ${num_ffts}

log  log_SED_${iseed}_${ifft}.lammps
```

```
reset_timestep   0
fix  1 all nve
dump  vel1 c1 custom ${w_step} dump_c1_${iseed}_${ifft}.vel vx vy vz
dump_modify  vel1 sort id
dump  vel2 c2 custom ${w_step} dump_c2_${iseed}_${ifft}.vel vx vy vz
dump_modify  vel2 sort id
dump  vel3 c3 custom ${w_step} dump_c3_${iseed}_${ifft}.vel vx vy vz
dump_modify  vel3 sort id
dump  vel4 c4 custom ${w_step} dump_c4_${iseed}_${ifft}.vel vx vy vz
dump_modify  vel4 sort id
dump  vel5 c5 custom ${w_step} dump_c5_${iseed}_${ifft}.vel vx vy vz
dump_modify  vel5 sort id
dump  vel6 c6 custom ${w_step} dump_c6_${iseed}_${ifft}.vel vx vy vz
dump_modify  vel6 sort id
dump  vel7 c7 custom ${w_step} dump_c7_${iseed}_${ifft}.vel vx vy vz
dump_modify  vel7 sort id
dump  vel8 c8 custom ${w_step} dump_c8_${iseed}_${ifft}.vel vx vy vz
dump_modify  vel8 sort id
thermo_style  custom step temp press vol
thermo 5000
timestep ${dt}
run ${w_total}
unfix 1
undump vel1
undump vel2
undump vel3
undump vel4
undump vel5
undump vel6
undump vel7
undump vel8

next ifft
jump in.LJArint.SED loop_fft
next seed
next iseed
jump in.LJArint.SED loop_seed
```

Post-processing

Using the frequencies and eigenvectors obtained from lattice dynamics calculations using GULP, the normal mode coordinates can be determined

$$\dot{Q}(\boldsymbol{\kappa},\nu,t) = \frac{1}{\sqrt{N}} \sum_{jl} \sqrt{m_j} exp(-i\boldsymbol{\kappa}\cdot\boldsymbol{r}(jl))\boldsymbol{e}^*(j,\boldsymbol{\kappa},\nu)\cdot\dot{\boldsymbol{u}}(jl,t) \tag{1}$$

where atom j is in unit cell l of a system with a total number of atoms N. $\boldsymbol{\kappa},\nu$ correspond to the phonon mode of wavevector $\boldsymbol{\kappa}$ and dispersion branch ν. $\boldsymbol{e}^*(j,\boldsymbol{\kappa},\nu)$ is the complex conjugate of the eigenvectors obtained from GULP. $\boldsymbol{u}(jl,t)$ is the velocity of atom jl. By taking a series of velocity samples from the MD simulation of time interval (in signal processing terminology, this is known as

lag which is symbolically represented here by t) an order of magnitude shorter than inverse of the highest frequency present in the system and using Equation 1 to project the sampled velocities onto the eigenvectors, the autocorrelation of the normal modes can calculated by

$$C(\boldsymbol{\kappa},\nu,t) = \lim_{T->\infty} \frac{1}{T}\int_0^T Q(\boldsymbol{\kappa},\nu,t+t')Q(\boldsymbol{\kappa},\nu,t')dt' \tag{2}$$

The spectral energy density (SED), from the Wiener-Khintchine theorem, is thus

$$C(\boldsymbol{\kappa},\nu,\omega) = \int_{-\infty}^{\infty} C(\boldsymbol{\kappa},\nu,t)e^{-i\omega t}dt \tag{3}$$

which is a Lorentzian curve centered at $\omega_0(\boldsymbol{\kappa},\nu)$

$$C(\boldsymbol{\kappa},\nu,\omega) = \frac{C_0(\boldsymbol{\kappa},\nu)}{2}\frac{\Gamma(\boldsymbol{\kappa},\nu)/\pi}{(\omega_0(\boldsymbol{\kappa},\nu)-\omega)^2+\Gamma(\boldsymbol{\kappa},\nu)} \tag{4}$$

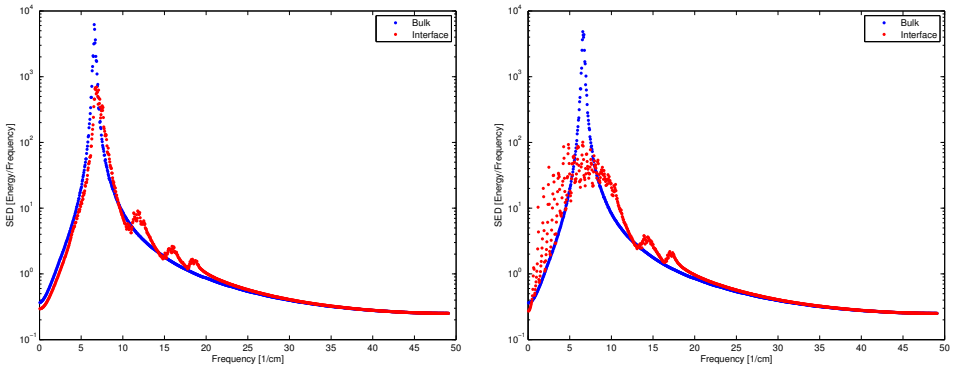
The half width at half maximum of the Lorentzian is related by anharmonic lattice dynamic theory to phonon lifetime by

$$\tau_{p-p}(\boldsymbol{\kappa},\nu) = \frac{1}{2\Gamma(\boldsymbol{\kappa},\nu)} \tag{5}$$

This procedure, from the velocity projection to the Lorentzian curve fitting, was automated using MATLAB.

Sample results

Using the output from the above LAMMPS script for the interface (a more basic one is used for the bulk case), the post-processing, performed on block 1, produced the following sample SED plots:



(a) $\boldsymbol{\kappa} = [0,1,0]$,$\nu$ =TA (b) $\boldsymbol{\kappa} = [1,0,0]$,$\nu$ =TA

Future Steps

The challenge, at this point, is to explain the trend observed in these figures from a fundamental perspective. What is causing the secondary bumps in these curves? For certain phonon modes, why does the presence of the interface destroy the Lorentzian form? Perhaps changing the number of

blocks (changing the periodicity of the superlattice) from the interface will provide insight into these questions.