2024 School on Many-Body Calculations using EPW and BerkeleyGW

Mobility including ionized-impurity and grain-boundary scatterings

Hands-on session (Wed. 7)

Hands-on based on QE-v7.3 and EPW-v5.9a

Introduction

This tutorial shows how to calculate carrier mobility in presence of other scattering mechanisms beside electron-phonon scattering. Here we add ionized impurity scattering and grain boundary scattering. The calculations will be also performed for polar cubic semiconductor BN (c-BN). The calculations for phonon-limited mobility in "Exercise 1 of Hands-on Wed.5" is the prerequiste. The submission file should be prepared as the template below, e. g. `run.sh`. We also provide submission files for all calculations, thus you can use these files if you do not want to create your own files.

run.ph

```
---
#!/bin/bash
#SBATCH -J epw4              # Job name
#SBATCH -o jobout.%j         # Name of stdout output file
#SBATCH -e joberr.%j         # Name of stderr error file
#SBATCH -p skx               # Queue (partition) name
#SBATCH -N 1                 # Total # of nodes
#SBATCH --ntasks-per-node 48 # Sufficiently descriptive
#SBATCH -t 01:00:00          # Run time (hh:mm:ss)
#SBATCH -A DMR23030
#SBATCH --reservation=NSF_Summer_School_Wed
#
module purge
module load TACC
module load intel/23.1
module list
export PATH="/work2/05193/sabyadk/stampede3/EPWSchool2024/q-e_school/bin:$PATH"

pwd
date
ibrun -n 48 epw.x -nk 48 < epw4.in > epw4.out
date
```

For the description of all input flags please follow the EPW documentation via the link below:
https://epwdoc.gitlab.io/source/doc/Inputs.html

Exercise 1

1.1 Theory

Ionized impurity scattering is an extrinsic mechanism that is very important in carrier transport alongside with electron phonon scattering. In doped semiconductors or materials with native ionized point defects, this type of scattering can reduce carrier mobility. In EPW, ionized-impurity scattering is added together with electron phonon scattering

$$\tau_{n\mathbf{k}}^{-1} = \tau_{n\mathbf{k}}^{-1,\mathrm{ph}} + \tau_{n\mathbf{k}}^{-1,\mathrm{ii}}, \tag{1}$$

where electron phonon scattering $\tau_{n\mathbf{k}}^{-1,\mathrm{ph}}$ is calculated as equation (3) of "Exercise 1 of Hands-on tutorial: Calculations of mobility" and $\tau_{n\mathbf{k}}^{-1,\mathrm{ii}}$ is ionized impurity scattering. For bulk crystals, this

quantity is *ab initio* calculated as

$$\tau_{n\mathbf{k}}^{-1,\mathrm{ii}} = \frac{1}{N_{\mathbf{q}}} \sum_{m\mathbf{q}} n_{\mathrm{ii}} \frac{2\pi}{\hbar} \left[\frac{e^2}{4\pi\varepsilon_0} \frac{4\pi Z}{\Omega_{\mathrm{uc}}} \right]^2 \sum_{\mathbf{G}\neq -\mathbf{q}} \frac{|\langle u_{m\mathbf{k}+\mathbf{q}} | e^{i\mathbf{G}.\mathbf{r}} | u_{n\mathbf{k}} \rangle_{\mathrm{uc}}|^2}{|(\mathbf{q}+\mathbf{G}).\varepsilon^0.(\mathbf{q}+\mathbf{G})|^2} \delta(\epsilon_{n\mathbf{k}} - \epsilon_{m\mathbf{k}+\mathbf{q}}). \tag{2}$$

Here, n_{ii} is the concentration of ionized impurities in the system, Z is the charge of the ionized impurities, ε^0 is the low-frequency dielectric tensor which includes screening effects from both electrons and ions. Subscript "u.c" refers to "unit cell". The formula (2) is obtained within the assumption that ionized impurities are randomly distributed in host materials and the Kohn-Luttinger ensemble average is exploited. More details can be found in the article Phys. Rev. B **107**, 125207 (2023).

1.2 Mobility calculations including ionized impurity scattering

▶ First copy the exercise files:

```
$ cd $SCRATCH
$ cp /work2/05193/sabyadk/stampede3/EPWSchool2024/tutorials/Wed.7.Ha.tar .
$ tar -xvf Wed.7.Ha.tar; cd Wed.7.Ha/exercise1
```

▶ Pre-calculations: You can EITHER redo all calculations in 1.2 and 1.3 of "Exercise 1 of Hands-on Wed.5" step by step as follows (you should wait for each job done sequentially)

```
$ sbatch run_scf_ph.sh
$ python3 pp.py
Enter the prefix used for PH calculations (e.g. diam)
bn
$ sbatch run_nscf.sh
$ sbatch run_epw1.sh
```

OR you are able to save time by copying necessary files from the previous directory where you have done "Exercise 1 of Hands-on Wed.5" to your current space

```
$ cp -r path_to_previous_exercise/* .
$ rm restart.fmt
```

▶ Prepare EPW input file `epw4.in`

```
--                                                                                    epw4.in
&inputepw
 prefix = 'bn'
 outdir = './'

 elph = .true.
 epwwrite = .false.
 epwread = .true.
 etf_mem = 0 ! generate k-points within fsthick
 lpolar = .true.
 vme = 'dipole'
 mp_mesh_k = .true.

 nbndsub = 3
 bands_skipped = 'exclude_bands = 1, 5-20'

 scattering = .true.
 scattering_serta = .true.
 int_mob = .false.
 carrier = .true.
 ncarrier = -1E17
```

```
 iterative_bte = .true.
 epmatkqread = .false.
 mob_maxiter = 300
 broyden_beta= 1.0
 bfieldx = 0.0d0
 bfieldy = 0.0d0
 bfieldz = 1.0d-10 ! Apply a magnetic field along Cart. z

 nstemp = 1
 temps = 300
 restart = .true.
 restart_step = 1000

 wannierize = .false.
 num_iter = 50000
 iprint = 2
 dis_win_max = 12.0
 dis_win_min = -1.0

 proj(1) = 'N:p'

 elecselfen = .false.
 phonselfen = .false.
 a2f = .false.

 fsthick = 0.4 ! 0.3 eV
 degaussw = 0.0

 efermi_read = .true
 fermi_energy = 11.246840

 dvscf_dir = './save'

 ii_g = .true.
 ii_charge = 1.0d0
 ii_n = 1.0d17      ! must match ncarrier, keep positive for both holes and electrons
 ii_scattering = .true.
 ii_only = .false.

 nkf1 = 40
 nkf2 = 40
 nkf3 = 40
 nqf1 = 40
 nqf2 = 40
 nqf3 = 40

 nk1 = 4
 nk2 = 4
 nk3 = 4
 nq1 = 4
 nq2 = 4
 nq3 = 4
/
```

Notes:

- `ii_g = .true.` tells EPW to calculate ionized impurity matrix elements.
- `ii_charge = 1.0d0` is the charge of the ionized impurities in the unit of the elementary charge.
- `ii_n = 1.0d17` is the density of ionized impurities in unit of cm^{-3}. For bulk, typical doping values range from 10^{15} to 10^{19} cm^{-3}. Higher concentration leads to degenrate doping.
- `ii_scattering = .true.` requires EPW to calculate the ionized impurity scattering rates.
- `ii_only = .false.` if `.true.`, only ionized impurity scatterings is considered.

- The presence of `quadrupole.fmt` file will enable interpolation of electron phonon matrix with quadrupole contribution

▶ Run `EPW` calculation for hole mobility of c-BN including ionized impurity scattering

```
$ sbatch run_epw4.sh
```

This calculation should be completed after around 1 minute with all 48 cores in 1 node. In the output file `epw4.out` you can see the low-frequency (static) dielectric tensor is calculated and printed out

```
 Low-frequency (static) dielectric tensor for ionized-impurity scattering
          6.6296745525    0.0000000000    0.0000000000
          0.0000000000    6.6296745525    0.0000000000
          0.0000000000    0.0000000000    6.6296745525
```

The experimental value of static dielectric constant in c-BN is about 7.1 [Phys. Rev. **155**, 1039 (1967)] which is higher than the our calculated value of 6.6, however, this calculation is performed via low kinetic energy cutoff `ecutwfc = 40` Ry. Calculations with converged `ecutwfc = 100` Ry in combination with spin-orbit coupling effect give a better result of 6.9 which finds an excellent agreement with experimental measurement.

The drift hole mobility at room-temperature with the presence of ionized impurity density `ii_n = 1.0d17` cm^{-3} is obtained around 95 cm^2/Vs

```
 Iteration number:        28

 ============================================================================================
   Temp     Fermi    Hole density  Population SR            Drift Hole mobility
    [K]      [eV]      [cm^-3]      [h per cell]                    [cm^2/Vs]
 ============================================================================================

 300.000  11.4498   0.10000E+18   0.29646E-18    0.948822E+02    0.893258E-18   -0.178652E-18
                                  0.32526E-18   -0.476404E-18    0.948822E+02   -0.609797E-16
                                 -0.81289E-19    0.815842E-17   -0.476404E-18    0.948822E+02

                                                 0.608537E-06    Max error
```

You can vary the carrier density `ncarrier` and ionized impurity density `ii_n = absolute(ncarrier)` in the input file `epw4.in` from 10^{15} to 10^{19} cm^{-3} and save data to the 3rd column in file `mobilities.dat` for later comparison.

```
# carrier (cm-3)  mob_ph (cm2/Vs)  mob_ph_ii (cm2/Vs)  mob_ph_ii_gb (cm2/Vs)
1e15              162.313          xxx                 xxx
1e16              162.314          xxx                 xxx
1e17              162.321          xxx                 xxx
1e18              162.390          xxx                 xxx
1e19              162.071          xxx                 xxx
```

DO NOT forget to delete `restart.fmt` file after each calculation. After filling up the 3rd column, you can plot and compare phonon-limited mobility with that if ionized-impurity included

```
$ gnuplot mobilities.gnu
```

You should obtain the results as seen in Fig. 1**a**. The convergent results and needed input parameters are shown in Fig. 1**b**

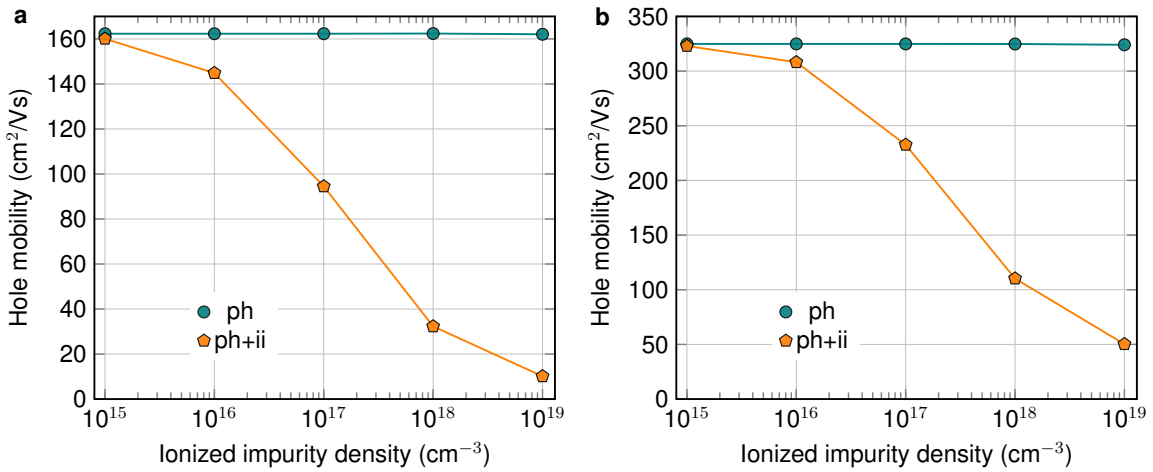

Fig. 1 Drift hole mobilities of c-BN at room-temperature as a function of ionized impurity density. **a** nk = nq = 4, nkf = nqf = 40 and ecutwfc = 40 Ry; **b** Convergent results with nk = 16, nq = 8, nkf = nqf = 80 and ecutwfc = 100 Ry.

Notes: Ionized impurity scattering and Hall mobilities have not been tested together.

Exercise 2

2.1 Theory

For multicrystalline materials, e.g. thin film morphology, there exists the interfaces between crystalline domains. The carriers are scattered at these boundaries, that is named as 'grain boundary scatterings'. We can take into account grain-boundary scattering using the simplest possible model, whereby the relaxation time is given by the time needed for the carrier to traverse the entire grain of size L. For a given carrier with velocity $\mathbf{v}_{n\mathbf{k}}$, the scattering rate in this model is $\tau_{n\mathbf{k}}^{-1,\mathrm{gb}} = |\mathbf{v}_{n\mathbf{k}}|/L$. This model had been popular in the study of thermal transport as it brought excellent success Nano Lett. 5, 1221 (2005). The electron velocities are computed via Wannier-Fourier interpolation within the `EPW` code. The grain size L is an external parameter of the model. The total scattering rates contributed by three different mechanisms are

$$\tau_{n\mathbf{k}}^{-1} = \tau_{n\mathbf{k}}^{-1,\mathrm{ph}} + \tau_{n\mathbf{k}}^{-1,\mathrm{ii}} + \tau_{n\mathbf{k}}^{-1,\mathrm{gb}} \tag{3}$$

2.2 Mobility calculations including grain boundary scattering

Here we will investigate how 100 nm grain size impact the hole mobility in c-BN.
▶ Pre-calculations: You can copy everything from Exercise 1

```
$ cd ../exercise2
$ cp -r ../exercise1/* .
$ rm restart.fmt
```

OR either redo all pre-calculations as in Exercise 1

```
$ sbatch run_scf_ph.sh
$ python3 pp.py
Enter the prefix used for PH calculations (e.g. diam)
bn
$ sbatch run_nscf.sh
$ sbatch run_epw1.sh
```

▶ Prepare EPW input file epw5.in
The input file for this calculation is created from epw4.in by adding three additional flags as follows

```
--                                                                                    epw5.in
&inputepw
...
 gb_scattering = .true.
 gb_size = 100 ! in nanometer
 gb_only = .false.
...
/
```

Notes:

- gb_scattering = .true. requires EPW to calculated grain boundary scattering rates.
- gb_size = 100 the grain size in nm unit.
- gb_only = .false. if .true., only grain boundary scattering is calculated.

▶ Run EPW calculation for hole mobility of c-BN including grain boundary scattering

```
$  sbatch run_epw5.sh
```

Under the impact of grain boundary scattering, the drift hole mobility of c-BN at room-temperature reduces from 95 to 76 cm^2/Vs.

```
   Iteration number:        24

   ================================================================================================
     Temp      Fermi    Hole density  Population SR            Drift Hole mobility
      [K]       [eV]      [cm^-3]      [h per cell]                    [cm^2/Vs]
   ================================================================================================

    300.000  11.4498   0.10000E+18   0.14558E-19    0.755951E+02    0.178652E-17    0.000000E+00
                                     -0.52940E-22    0.476404E-16    0.755951E+02    0.295371E-16
                                     -0.13235E-21   -0.595505E-19    0.304303E-16    0.755951E+02

                                                     0.783232E-06    Max error
```

```
$  cp ../exercise1/mobilities.dat .
$  cp ../exercise1/mobilities.gnu .
```

Similarly, you can keep gb_size = 100 and vary carrier and ionized impurity densities as in Excercise 1 then fill up results into 4th column of mobilities.dat file. Gnuplot file mobilities.gnu is then modified at the bottom as follows

```
plot 'mobilities.dat'u 1:2 w lp pt 4 title 'ph',\
     'mobilities.dat'u 1:3 w lp pt 5 title 'ph+ii',\
     'mobilities.dat'u 1:4 w lp pt 7 title 'ph+ii+gb'
```

If you rerun 'gnuplot mobilities.gnu', you will see results as presented in Fig. 2**a**. The convergent results and needed input parameters are shown in Fig. 2**b**
Notes: There are still other factors which can enhance the accuracy of these transport calculations, e.g. spin-orbit coupling effect, many-body GW band renormalization,...

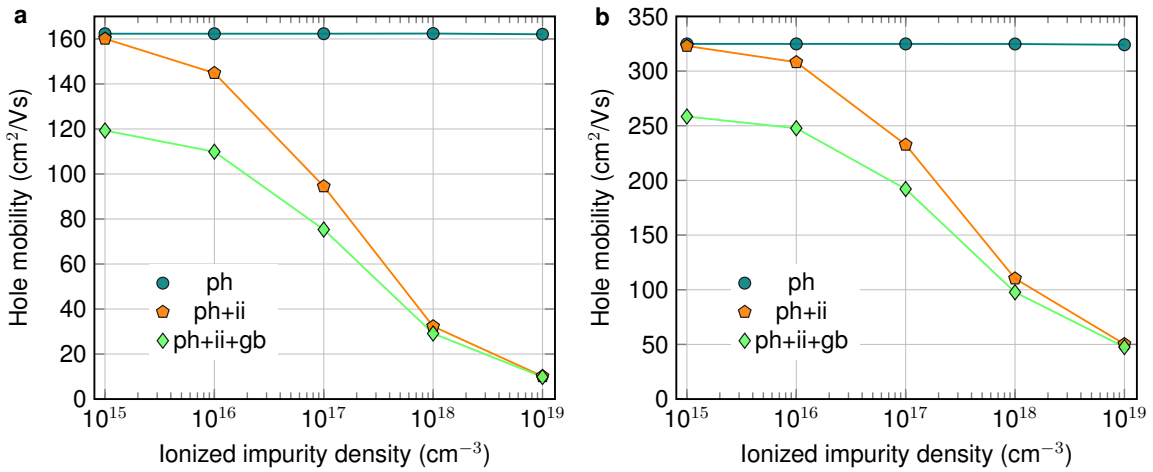

Fig. 2 Drift hole mobilities of c-BN at room-temperature with grain boundary scattering is considered for grain-size of 100 nm. **a** nk = nq = 4, nkf = nqf = 40 and ecutwfc = 40 Ry; **b** Convergent results with nk = 16, nq = 8, nkf = nqf = 80 and ecutwfc = 100 Ry.