UCLouvain
Institute of Condensed Matter
and Nanosciences

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Hall and Drift mobility in EPW - wannier90 school 2022

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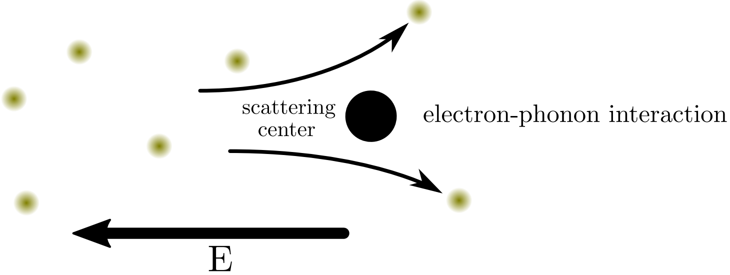
Phonon-limited carrier mobilities

Mobility $\mu \propto \frac{\partial}{\partial E} \int d\mathbf{k} f_{\mathbf{k}} v_{\mathbf{k}}$



- **lattice (phonon) scattering**
- ionized impurity scattering
- alloy scattering
- defects scattering

Berryology @ Ivo Souza

Using Ivo's notation from Tuesday:

$$
\begin{aligned}
j_\alpha &= -e \int_{\mathbf{k}} \dot{r}_a f(\varepsilon) \\
&= -e \int_{\mathbf{k}} [\underbrace{v_a}_{\text{band}} + \underbrace{(e/\hbar)\Omega_{ab} E_b}_{\text{anomalous}} + \ldots][f_0 + \tau e v_c E_c f_0' + \ldots] \\
&= C + \sigma_{ab} E_b + \sigma_{abc} E_b E_c + \ldots
\end{aligned}
$$

$$
\sigma_{ab} = -e^2 \tau \int_{\mathbf{k}} v_a v_b f_0' - \frac{e^2}{\hbar} \int_{\mathbf{k}} \Omega_{ab} f_0
$$

Linear Ohmic + Hall

In system with TR symmetry: $\int_{\mathbf{k}} \Omega_{ab} f_0 = 0$

Linearized Boltzmann transport equation

Drift mobility:
$$\mu^{\mathrm{d}}_{\alpha\beta} = \frac{-1}{V_{\mathrm{uc}} n_{\mathrm{c}}} \sum_n \int \frac{\mathrm{d}^3 k}{\Omega_{\mathrm{BZ}}} v^{\alpha}_{n\mathbf{k}} \partial_{E_\beta} f_{n\mathbf{k}}$$

```
iterative_bte = .true.
```

$$\partial_{E_\beta} f_{n\mathbf{k}} = e v^{\beta}_{n\mathbf{k}} \frac{\partial f^0_{n\mathbf{k}}}{\partial \varepsilon_{n\mathbf{k}}} \tau_{n\mathbf{k}} + \frac{2\pi \tau_{n\mathbf{k}}}{\hbar} \sum_{m\nu} \int \frac{\mathrm{d}^3 q}{\Omega_{\mathrm{BZ}}} |g_{mn\nu}(\mathbf{k},\mathbf{q})|^2 \\ \times \Big[(n_{\mathbf{q}\nu} + 1 - f^0_{n\mathbf{k}}) \delta(\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}+\mathbf{q}} + \hbar\omega_{\mathbf{q}\nu}) \\ + (n_{\mathbf{q}\nu} + f^0_{n\mathbf{k}}) \delta(\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}+\mathbf{q}} - \hbar\omega_{\mathbf{q}\nu}) \Big] \partial_{E_\beta} f_{m\mathbf{k}+\mathbf{q}}$$

where

$$\tau^{-1}_{n\mathbf{k}} \equiv \frac{2\pi}{\hbar} \sum_{m\nu} \int \frac{d^3 q}{\Omega_{\mathrm{BZ}}} |g_{mn\nu}(\mathbf{k},\mathbf{q})|^2 \big[(n_{\mathbf{q}\nu} + 1 - f^0_{m\mathbf{k}+\mathbf{q}}) \\ \times \delta(\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}+\mathbf{q}} - \hbar\omega_{\mathbf{q}\nu}) + (n_{\mathbf{q}\nu} + f^0_{m\mathbf{k}+\mathbf{q}}) \delta(\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}+\mathbf{q}} + \hbar\omega_{\mathbf{q}\nu}) \big]$$

Linearized Boltzmann transport equation

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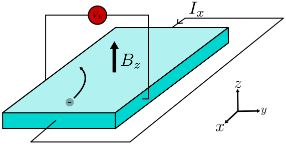
Drift mobility

$$\mu^{\mathrm{d}}_{\alpha\beta} = \frac{-1}{V_{\mathrm{uc}} n_{\mathrm{c}}} \sum_n \int \frac{\mathrm{d}^3 k}{\Omega_{\mathrm{BZ}}} \, v^{\alpha}_{n\mathbf{k}} \partial_{E_\beta} f_{n\mathbf{k}}$$

Self-energy relaxation time approximation (SERTA)

$$\partial_{E_\beta} f_{n\mathbf{k}} = e v^{\beta}_{n\mathbf{k}} \frac{\partial f^0_{n\mathbf{k}}}{\partial \varepsilon_{n\mathbf{k}}} \tau_{n\mathbf{k}}$$

Linearized Boltzmann transport equation



$$\mu^{\mathrm{Hall}}_{\alpha\beta}(\hat{\mathbf{B}}) = \sum_{\gamma} \mu^{\mathrm{drift}}_{\alpha\gamma} r_{\gamma\beta}(\hat{\mathbf{B}})$$

$$r_{\alpha\beta}(\hat{\mathbf{B}}) \equiv \lim_{\mathbf{B}\to 0} \sum_{\delta\epsilon} \frac{[\mu^{\mathrm{drift}}_{\alpha\delta}]^{-1} \mu_{\delta\epsilon}(\mathbf{B}) [\mu^{\mathrm{drift}}_{\epsilon\beta}]^{-1}}{|\mathbf{B}|}$$

$$\mu_{\alpha\beta}(B_\gamma) = \frac{-1}{S_{\mathrm{uc}} n_{\mathrm{c}}} \sum_n \int \frac{\mathrm{d}^3 k}{S_{\mathrm{BZ}}} v_{n\mathbf{k}\alpha} \Big[\partial_{E_\beta} f_{n\mathbf{k}}(B_\gamma) - \partial_{E_\beta} f_{n\mathbf{k}} \Big]$$

$$\mu^{\mathrm{drift}}_{\alpha\beta} = \frac{-1}{S_{\mathrm{uc}} n_{\mathrm{c}}} \sum_n \int \frac{\mathrm{d}^3 k}{S_{\mathrm{BZ}}} v_{n\mathbf{k}\alpha} \partial_{E_\beta} f_{n\mathbf{k}}$$

$\rightarrow$ We need the out-of-equilibrium carrier population

F. Macheda and N. Bonini, Phys. Rev. B **98**, 201201R (2018)
SP *et al.*, Rep. Prog. Phys. **83**, 036501 (2020)
SP *et al.*, Phys. Rev. Research **3**, 043022 (2021)

Linearized Boltzmann transport equation

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```
bfieldz = 1.0d-10
```

$$\left[1 - \frac{e}{\hbar}\tau_{n\mathbf{k}}(\mathbf{v}_{n\mathbf{k}} \times \mathbf{B}) \cdot \nabla_{\mathbf{k}}\right] \partial_{E_\beta} f_{n\mathbf{k}}(\mathbf{B}) = e v_{n\mathbf{k}\beta} \frac{\partial f^0_{n\mathbf{k}}}{\partial \varepsilon_{n\mathbf{k}}} \tau_{n\mathbf{k}}$$
$$+ \frac{2\pi \tau_{n\mathbf{k}}}{\hbar} \sum_{m\nu} \int \frac{\mathrm{d}^3 q}{S_{\mathrm{BZ}}} |g_{mn\nu}(\mathbf{k},\mathbf{q})|^2 \Big[(n_{\mathbf{q}\nu} + 1 - f^0_{n\mathbf{k}})\delta(\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}+\mathbf{q}} + \hbar\omega_{\mathbf{q}\nu})$$
$$+ (n_{\mathbf{q}\nu} + f^0_{n\mathbf{k}})\delta(\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}+\mathbf{q}} - \hbar\omega_{\mathbf{q}\nu})\Big] \partial_{E_\beta} f_{m\mathbf{k}+\mathbf{q}}(\mathbf{B}),$$

where the scattering rate is

$$\tau^{-1}_{n\mathbf{k}} \equiv \frac{2\pi}{\hbar} \sum_{m\nu} \int \frac{d^3 q}{\Omega_{\mathrm{BZ}}} |g_{mn\nu}(\mathbf{k},\mathbf{q})|^2 \big[(n_{\mathbf{q}\nu} + 1 - f^0_{m\mathbf{k}+\mathbf{q}})$$
$$\times \delta(\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}+\mathbf{q}} - \hbar\omega_{\mathbf{q}\nu}) + (n_{\mathbf{q}\nu} + f^0_{m\mathbf{k}+\mathbf{q}})\delta(\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}+\mathbf{q}} + \hbar\omega_{\mathbf{q}\nu})\big]$$

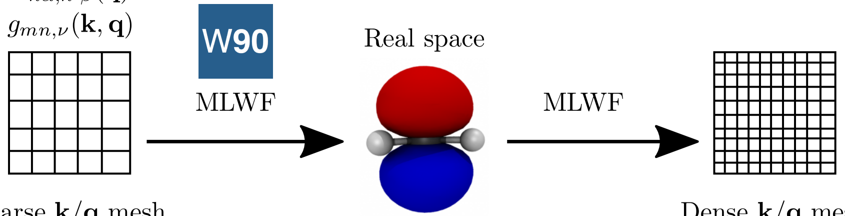
F. Macheda and N. Bonini, Phys. Rev. B **98**, 201201R (2018)
SP *et al.*, Rep. Prog. Phys. **83**, 036501 (2020)
SP *et al.*, Phys. Rev. Research **3**, 043022 (2021)

Electron-phonon Wannier (EPW)

EPW relies on MLWF to interpolate electron-phonon matrix elements.

$C_{\kappa\alpha,\kappa'\beta}(\mathbf{q})$

$g_{mn,\nu}(\mathbf{k},\mathbf{q})$



Coarse $\mathbf{k}/\mathbf{q}$ mesh

Dense $\mathbf{k}/\mathbf{q}$ mesh

SP *et al.*, Comput. Phys. Commun. **209**, 116 (2016)

Electron-phonon Wannier (EPW)

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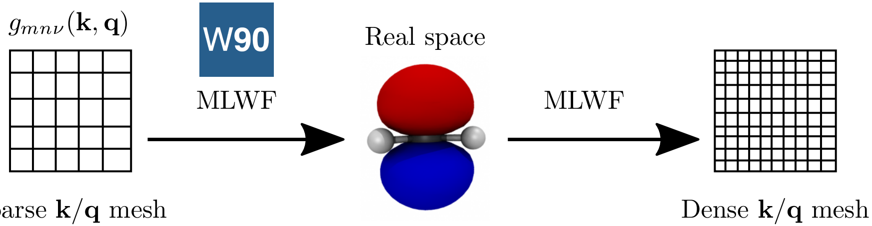
EPW relies on MLWF to interpolate electron-phonon matrix elements.

$$g_{mn\nu}(\mathbf{k},\mathbf{q}) - g^{\mathcal{L}}_{mn\nu}(\mathbf{k},\mathbf{q}) \qquad\qquad g^{\mathcal{S}}_{mn\nu}(\mathbf{k},\mathbf{q}) + g^{\mathcal{L}}_{mn\nu}(\mathbf{k},\mathbf{q})$$



SP *et al.*, Comput. Phys. Commun. **209**, 116 (2016)

Dynamical quadrupoles: Si

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$$g_{mn\nu}(\mathbf{k},\mathbf{q}) = g^{\mathcal{S}}_{mn\nu}(\mathbf{k},\mathbf{q}) + g^{\mathcal{L}}_{mn\nu}(\mathbf{k},\mathbf{q})$$
$$g^{\mathcal{L}}_{mn\nu}(\mathbf{k},\mathbf{q}) = {\color{blue}g^{\mathcal{L},\mathrm{D}}_{mn\nu}(\mathbf{k},\mathbf{q})} + {\color{red}g^{\mathcal{L},\mathrm{Q}}_{mn\nu}(\mathbf{k},\mathbf{q})} + g^{\mathcal{L},\mathrm{O}}_{mn\nu}(\mathbf{k},\mathbf{q}) + \cdots$$

```
lpolar = .true.
```

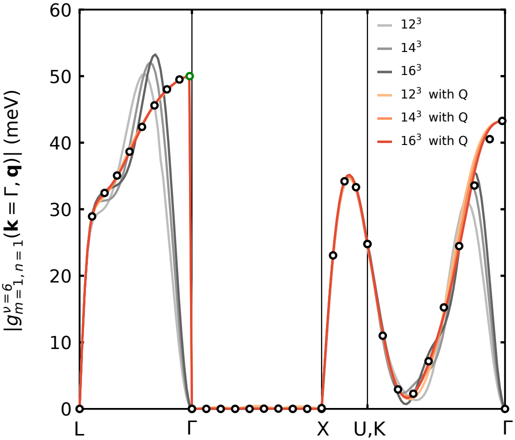
If `quadrupole.fmt` file is present in the directory

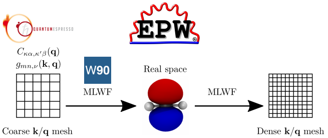
$$g^{\mathcal{L}}_{mn\nu}(\mathbf{k},\mathbf{q}) = \sum_{\kappa\alpha}\left[\frac{\hbar}{2NM_\kappa\omega_\nu(\mathbf{q})}\right]^{\frac{1}{2}} \frac{4\pi e^2 e^{-\frac{|\mathbf{q}|^2}{4\Lambda^2}}}{\Omega\sum_{\delta\delta'} q_\delta \epsilon^\infty_{\delta\delta'} q_{\delta'}}$$
$$\times\, e^{-i\mathbf{q}\cdot\boldsymbol{\tau}_\kappa}\left[\sum_\beta {\color{blue}iq_\beta Z_{\kappa\alpha\beta}} + {\color{red}\sum_\gamma \frac{q_\beta q_\gamma}{2} Q_{\kappa\alpha\beta\gamma}}\right] e_{\kappa\alpha\nu}(\mathbf{q})$$
$$\langle\Psi_{m\mathbf{k}+\mathbf{q}}|\, e^{i\mathbf{q}\cdot\mathbf{r}}[1 + {\color{red}iq_\alpha v^{\mathrm{Hxc},\mathcal{E}_\alpha}(\mathbf{r})}]\,|\Psi_{n\mathbf{k}}\rangle\,.$$

C. Verdi and F. Giustino, Phys. Rev. Lett. **119**, 176401 (2015)
G. Brunin *et al.*, Phys. Rev. Lett. **125**, 136601 (2020)

Dynamical quadrupoles: Si

$$g_{mn\nu}(\mathbf{k},\mathbf{q}) = g^{\mathcal{S}}_{mn\nu}(\mathbf{k},\mathbf{q}) + g^{\mathcal{L}}_{mn\nu}(\mathbf{k},\mathbf{q})$$
$$g^{\mathcal{L}}_{mn\nu}(\mathbf{k},\mathbf{q}) = g^{\mathcal{L},\mathrm{D}}_{mn\nu}(\mathbf{k},\mathbf{q}) + g^{\mathcal{L},\mathrm{Q}}_{mn\nu}(\mathbf{k},\mathbf{q}) + g^{\mathcal{L},\mathrm{O}}_{mn\nu}(\mathbf{k},\mathbf{q}) + \cdots$$





G. Brunin *et al.*, PRL **125**, 136601 (2020)
V. A. Jhalani *et al.*, PRL **125**, 136602 (2020)

How to converge transport calculations + tip

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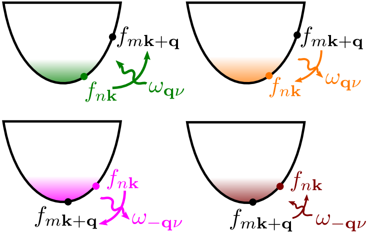
- Treat electron and hole separately
- Choose the smallest `fsthick` window as possible

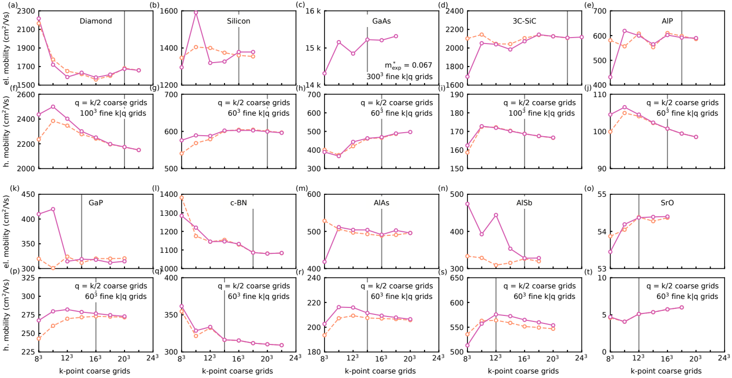
```
etf_mem = 3

mp_mesh_k = .true.
```



$$
\begin{aligned}
-e\mathbf{E}\cdot\frac{1}{\hbar}\frac{\partial f_{n\mathbf{k}}}{\partial \mathbf{k}} = \frac{2\pi}{\hbar}\sum_{m,\nu}\int\frac{\mathrm{d}^3q}{\Omega_{\mathrm{BZ}}}\,|g_{mn\nu}(\mathbf{k},\mathbf{q})|^2 \\
\times\Big[f_{n\mathbf{k}}(1-f_{m\mathbf{k}+\mathbf{q}})\delta(\varepsilon_{n\mathbf{k}}-\varepsilon_{m\mathbf{k}+\mathbf{q}}+\hbar\omega_{\mathbf{q}\nu})n_{\mathbf{q}\nu} \\
+f_{n\mathbf{k}}(1-f_{m\mathbf{k}+\mathbf{q}})\delta(\varepsilon_{n\mathbf{k}}-\varepsilon_{m\mathbf{k}+\mathbf{q}}-\hbar\omega_{\mathbf{q}\nu})(n_{\mathbf{q}\nu}+1) \\
-(1-f_{n\mathbf{k}})f_{m\mathbf{k}+\mathbf{q}}\delta(\varepsilon_{m\mathbf{k}+\mathbf{q}}-\varepsilon_{n\mathbf{k}}+\hbar\omega_{\mathbf{q}\nu})n_{\mathbf{q}\nu} \\
-(1-f_{n\mathbf{k}})f_{m\mathbf{k}+\mathbf{q}}\delta(\varepsilon_{m\mathbf{k}+\mathbf{q}}-\varepsilon_{n\mathbf{k}}-\hbar\omega_{\mathbf{q}\nu})(n_{\mathbf{q}\nu}+1)\Big]
\end{aligned}
$$

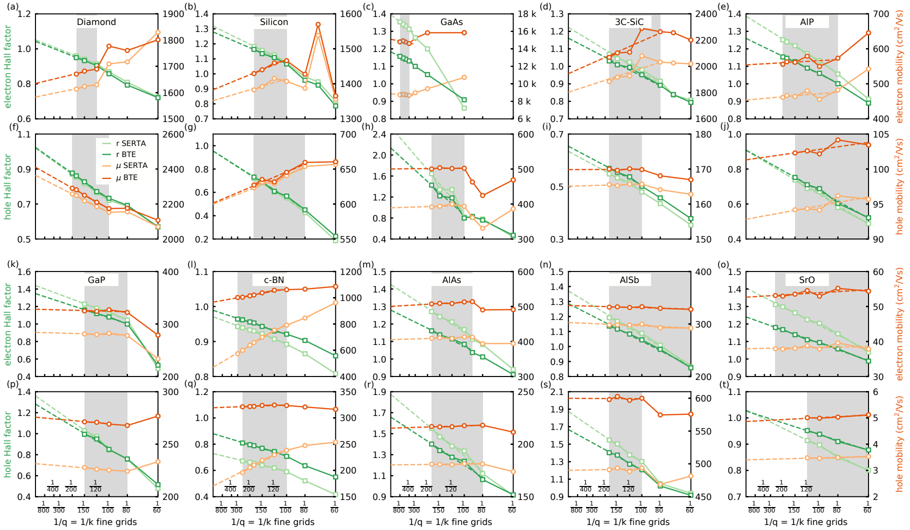
How to converge transport calculations + tip

- Choose a small enough B-field (`bfieldz = 1.0d-10`) for Hall factor
- `vme = 'dipole'` should be more stable than `vme = 'wannier'`
- Use `degaussw = 0.0` for adaptive broadening
- Use the same k-point and q-point coarse grid (more efficient)
- Converge coarse k/q grids.



How to converge transport calculations + tip

- Converge fine k/q grids and Hall factor (extrapolate).

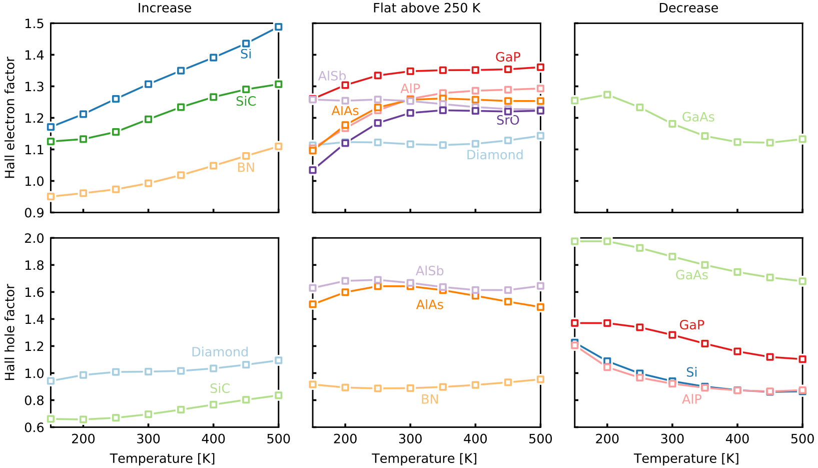


Results

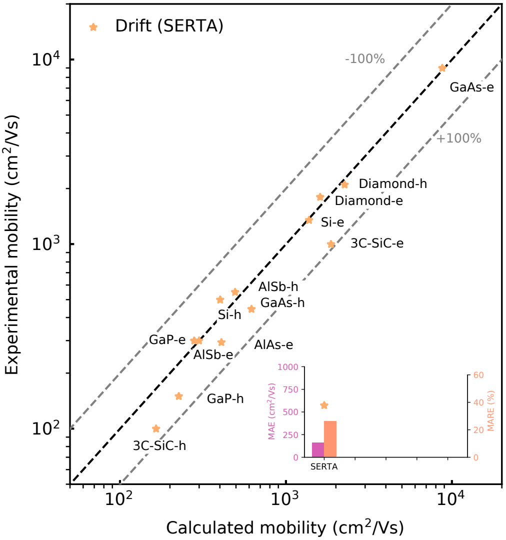
Some results of drift and Hall mobility on 10 simple bulk semiconductors

Do we reproduce experimental mobility ?

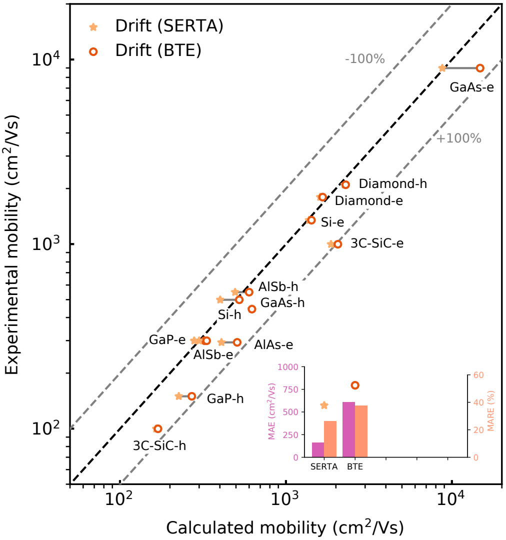
The Hall factor is not 1 !



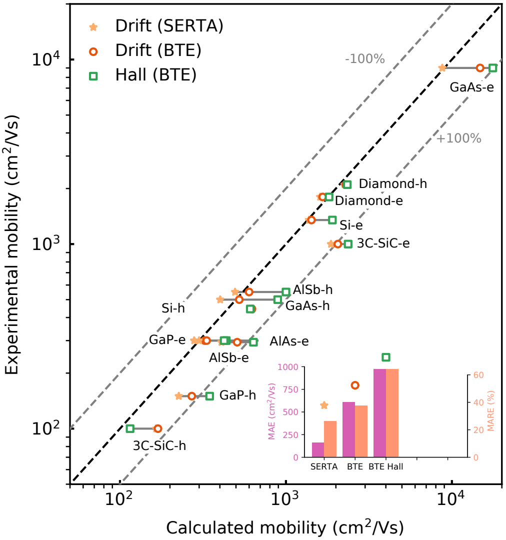
Experimental comparison



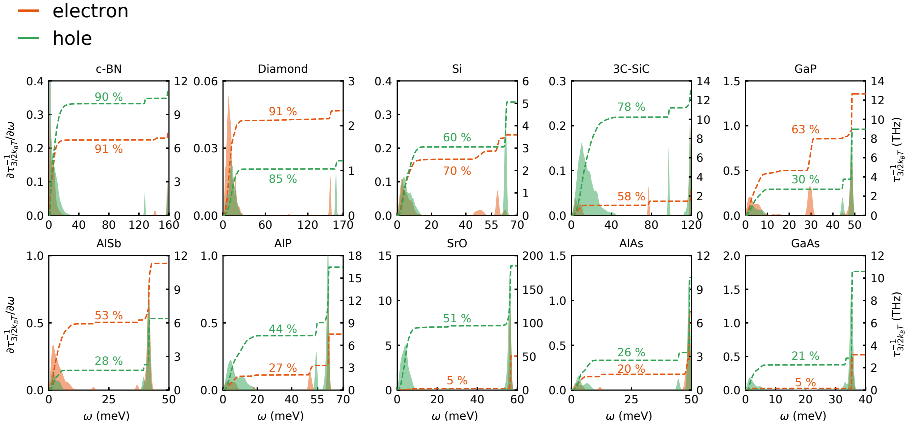
Experimental comparison



Experimental comparison



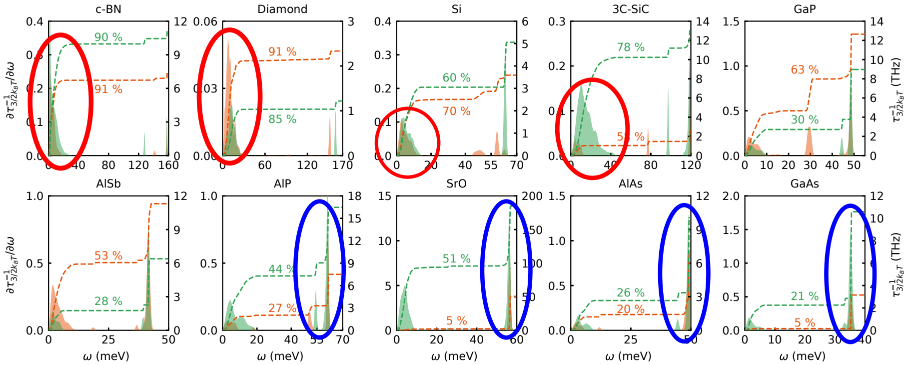
Spectral decomposition: dominant scattering



Spectral decomposition: dominant scattering

— electron
— hole

Acoustic scattering dominates



Optical scattering dominates

Conclusions

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- General framework for drift and Hall mobility based on the BTE
- Quantified the role of various approximations
    - Local velocity approximation
    - Neglect of quadrupoles
    - SOC for hole
    - Self energy relaxation time approximation
- Hall factor is temperature dependent and between 0.7 to 2.0 at RT

Acknowledgments

The current EPW team:

- Feliciano Giustino - 2007
- Roxana Margine - 2011
- Samuel Poncé - 2015
- Hari Paudyal - 2018
- Francesco Macheda - 2018
- Emmanouil Kioupakis - 2019
- Xiao Zhang - 2019
- Hyungjun Lee - 2019
- Chao Lian - 2020
- Joshua Leveillee - 2020
- Jon Lafuente Bartolome - 2021
- YOU ?

More info

UCLouvain

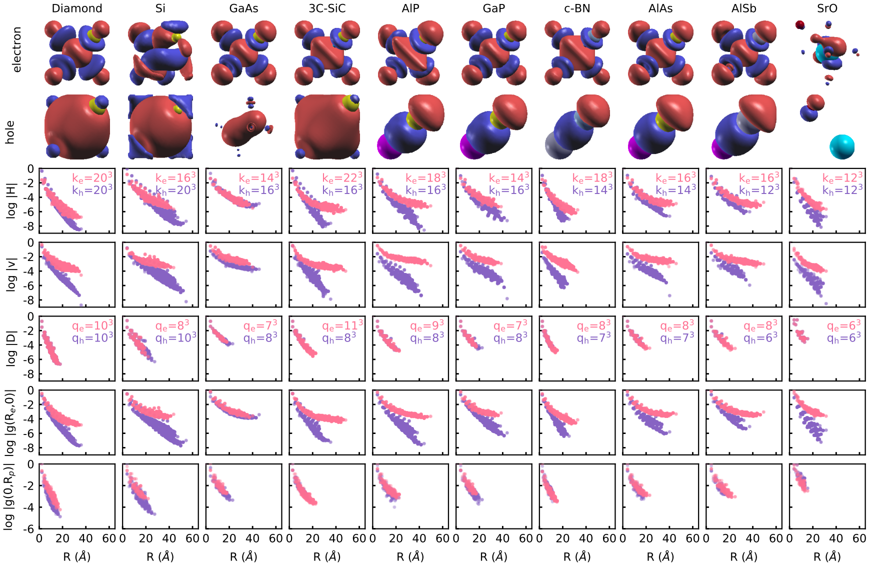
https://epw-code.org

https://forum.epw-code.org

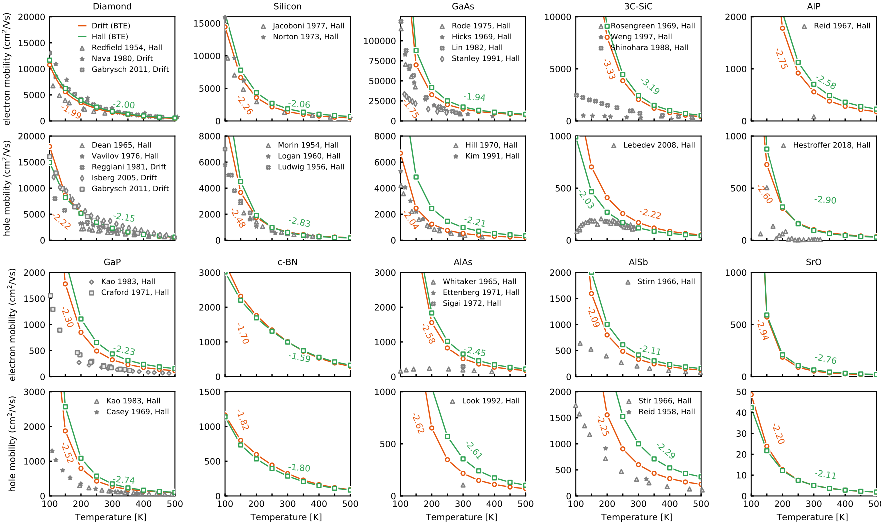
https://gitlab.com/QEF/q-e

SP, W. Li, S. Reichardt, F. Giustino, Reports on Progress in Physics (2020)
SP, E.R. Margine, C. Verdi, F. Giustino, Comput. Phys. Commun. **209**, 116 (2016)
SP, E.R. Margine, F. Giustino, Phys. Rev. B **97**, 121201 (2018)
F. Macheda, SP, F. Giustino, N. Bonini, Nano Lett. **20**, 8861 (2020)
SP, M. Schlipf, F. Giustino, ACS Energy Lett. **4**, 456 (2019)
SP, D. Jena, F. Giustino, Phys. Rev. B **100**, 085204 (2019)
SP, D. Jena, F. Giustino, Phys. Rev. Lett. **123**, 096602 (2019)
SP, F. Giustino, Phys. Rev. Res. **2**, 0333102 (2020)

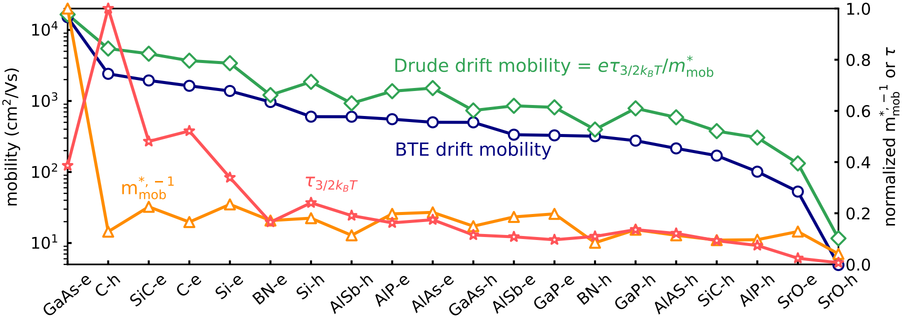
10 simple semiconductors



Temperature dependence mobility

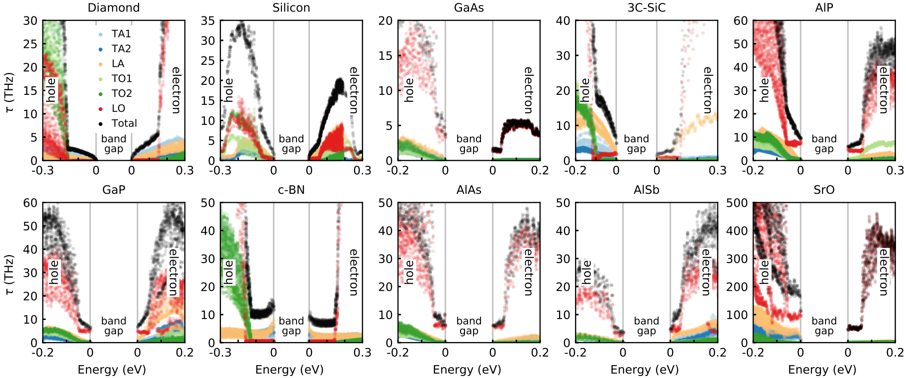


Drude overestimates BTE mobility



$$\frac{1}{m^*_{\mathrm{mob}}} = \frac{-1}{3V_{\mathrm{uc}}n_{\mathrm{c}}}\sum_{\alpha n}\int \frac{\mathrm{d}^3k}{\Omega_{\mathrm{BZ}}}\frac{\partial f^0_{n\mathbf{k}}}{\partial \varepsilon_{n\mathbf{k}}} v^2_{n\mathbf{k}\alpha}.$$

Dominant scattering mode



GaAs, AlP, GaP, AlAs, AlSb, and SrO → LO

diamond, silicon, 3C-SiC and c-BN → LA and TA

Strongest approximations

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- Local velocity approximation
- Neglect of quadrupoles
- SOC for hole mobility
- Self energy relaxation time approximation