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Institute of Condensed Matter
and Nanosciences

LA LIBERTÉ DE CHERCHER

Ab initio calculation of carrier mobility in bulk and low-dimensional materials

Samuel Poncé
Université catholique de Louvain, Belgium

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Acknowledgments

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European Commission
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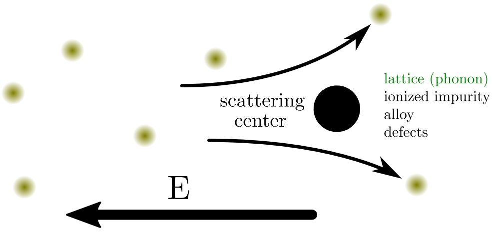
THEOS
THEORY AND SIMULATION OF MATERIALS

PRACE

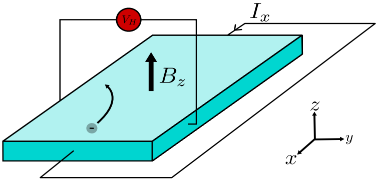
Phonon-limited carrier mobilities

Drift



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

Hall



Drift Boltzmann transport equation (BTE)

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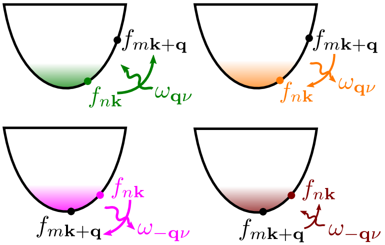
$$
\begin{aligned}
-e\mathbf{E}\cdot\frac{1}{\hbar}\frac{\partial f_{n\mathbf{k}}}{\partial \mathbf{k}} = \frac{2\pi}{\hbar}\sum_{m,\nu}\int\frac{\mathrm{d}^3q}{\Omega_{\mathrm{BZ}}}\,|g_{mn\nu}(\mathbf{k},\mathbf{q})|^2 \\
\times\Big[f_{n\mathbf{k}}(1-f_{m\mathbf{k}+\mathbf{q}})\delta(\varepsilon_{n\mathbf{k}}-\varepsilon_{m\mathbf{k}+\mathbf{q}}+\hbar\omega_{\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



SP *et al.*, Rep. Prog. Phys. **83**, 036501 (2020)

Linearized drift BTE

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

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

where

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

SP *et al.*, Rep. Prog. Phys. **83**, 036501 (2020)

Hall BTE

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

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

where

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

SP *et al.*, Rep. Prog. Phys. **83**, 036501 (2020)

Electron-phonon Wannier (EPW)

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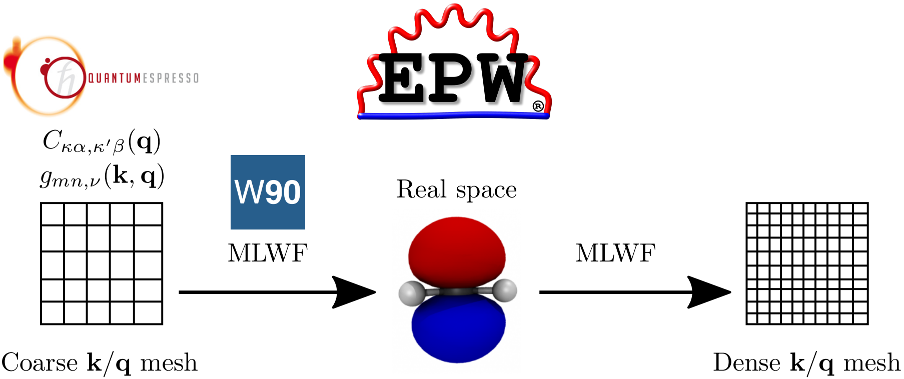
EPW relies on MLWF to interpolate electron-phonon matrix elements.



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

Dynamical quadrupoles in el-ph

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

$$g^{\mathcal{L}}_{mn\nu}(\mathbf{k},\mathbf{q}) = g^{\mathcal{L},\mathrm{eD}}_{mn\nu}(\mathbf{k},\mathbf{q}) + g^{\mathcal{L},\mathrm{eQ}}_{mn\nu}(\mathbf{k},\mathbf{q}) + \cdots$$

$$g^{\mathcal{L},\mathrm{eD}}_{mn\nu}(\mathbf{k},\mathbf{q}) = i\frac{4\pi}{V_{\mathrm{uc}}}\frac{e^2}{4\pi\varepsilon^0}\sum_{\kappa}\left[\frac{\hbar}{2N_{p'}M_\kappa\omega_{\mathbf{q}\nu}}\right]^{\frac{1}{2}}\sum_{\mathbf{G}\neq-\mathbf{q}} e^{-i(\mathbf{G}+\mathbf{q})\cdot\boldsymbol{\tau}_\kappa} \times \frac{(\mathbf{G}+\mathbf{q})\cdot\mathbf{Z}^*_\kappa\cdot\mathbf{e}_{\kappa\mathbf{q}\nu}}{(\mathbf{G}+\mathbf{q})\cdot\boldsymbol{\varepsilon}^\infty\cdot(\mathbf{G}+\mathbf{q})}\langle\Psi_{m\mathbf{k}+\mathbf{q}}|e^{i(\mathbf{q}+\mathbf{G})\cdot\mathbf{r}}|\Psi_{n\mathbf{k}}\rangle$$

$$g^{\mathcal{L},\mathrm{eQ}}_{mn\nu}(\mathbf{k},\mathbf{q}) = \frac{4\pi}{V_{\mathrm{uc}}}\frac{e^2}{4\pi\varepsilon^0}\sum_{\kappa}\left[\frac{\hbar}{2N_{p'}M_\kappa\omega_{\mathbf{q}\nu}}\right]^{\frac{1}{2}}\sum_{\mathbf{G}\neq-\mathbf{q}} \times \frac{(\mathbf{G}+\mathbf{q})\cdot(\mathbf{G}+\mathbf{q})\cdot\mathbf{e}_{\kappa\mathbf{q}\nu}\cdot\tilde{\mathbf{Q}}_{mn\kappa}(\mathbf{k},\mathbf{G}+\mathbf{q})}{(\mathbf{G}+\mathbf{q})\cdot\boldsymbol{\varepsilon}^\infty\cdot(\mathbf{G}+\mathbf{q})} e^{-i(\mathbf{G}+\mathbf{q})\cdot\boldsymbol{\tau}_\kappa},$$

C. Verdi *et al.*, Phys. Rev. Lett. **115**, 176401 (2015)
G. Brunin *et al.*, Phys. Rev. Lett. **125**, 136601 (2020)
V.A. Jhalani *et al.*, Phys. Rev. Lett. **125**, 136602 (2020)

Dynamical quadrupoles in D

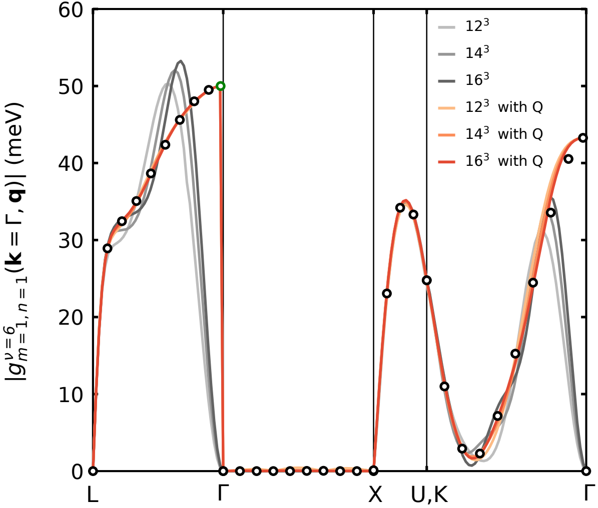
$$D_{\kappa\alpha,\kappa'\beta}(\mathbf{q}) = D^{\mathcal{S}}_{\kappa\alpha,\kappa'\beta}(\mathbf{q}) + \boxed{D^{\mathcal{L}}_{\kappa\alpha,\kappa'\beta}(\mathbf{q})}$$

$$\boxed{D^{\mathcal{L}}_{\kappa\alpha,\kappa'\beta}(\mathbf{q})} = \frac{4\pi e^2}{V_{\mathrm{uc}}}\Bigg[\sum_{\mathbf{G}\neq-\mathbf{q}} \boxed{D^{\mathcal{L},\mathrm{DD+DQ+QQ}}_{\kappa\alpha,\kappa'\beta}(\mathbf{G}+\mathbf{q})} - \delta_{\kappa\kappa'}\sum_{\kappa''}\sum_{\mathbf{G}\neq\mathbf{0}} \boxed{D^{\mathcal{L},\mathrm{DD+DQ+QQ}}_{\kappa\alpha,\kappa''\beta}(\mathbf{G})}\Bigg]$$

$$\boxed{D^{\mathcal{L},\mathrm{DD+DQ+QQ}}_{\kappa\alpha,\kappa'\beta}(\mathbf{q})} = \frac{e^{i\mathbf{q}\cdot(\boldsymbol{\tau}_{\kappa}-\boldsymbol{\tau}_{\kappa'})}e^{\frac{-\mathbf{q}\cdot\boldsymbol{\varepsilon}^{\infty}\cdot\mathbf{q}}{4\Lambda^2}}}{\mathbf{q}\cdot\boldsymbol{\varepsilon}^{\infty}\cdot\mathbf{q}}\Bigg[\mathbf{q}\cdot\mathbf{Z}^{*}_{\kappa\alpha}\cdot\mathbf{q}\cdot\mathbf{Z}^{*}_{\kappa'\beta} + \frac{1}{4}\mathbf{q}\cdot\mathbf{q}\cdot\mathbf{Q}_{\kappa\alpha}\cdot\mathbf{q}\cdot\mathbf{q}\cdot\mathbf{Q}_{\kappa'\beta} + \frac{i}{2}\mathbf{q}\cdot\mathbf{Z}^{*}_{\kappa\alpha}\cdot\mathbf{q}\cdot\mathbf{q}\cdot\mathbf{Q}_{\kappa'\beta} - \frac{i}{2}\mathbf{q}\cdot\mathbf{q}\cdot\mathbf{Q}_{\kappa\alpha}\cdot\mathbf{q}\cdot\mathbf{Z}^{*}_{\kappa'\beta}\Bigg].$$

X. Gonze *et al.*, Phys. Rev. B **55**, 10355 (1997)
M. Royo *et al.*, Phys. Rev. Lett. **125**, 217602 (2020)

Silicon el-ph



Deformation potential of c-BN

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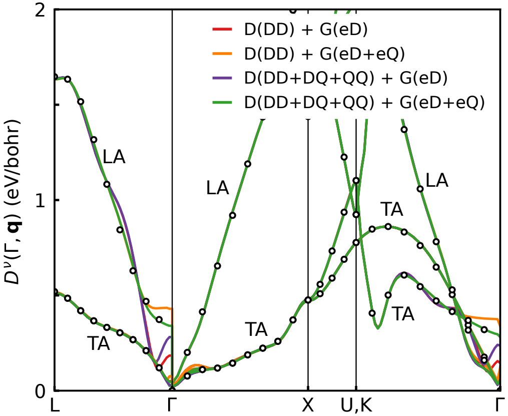
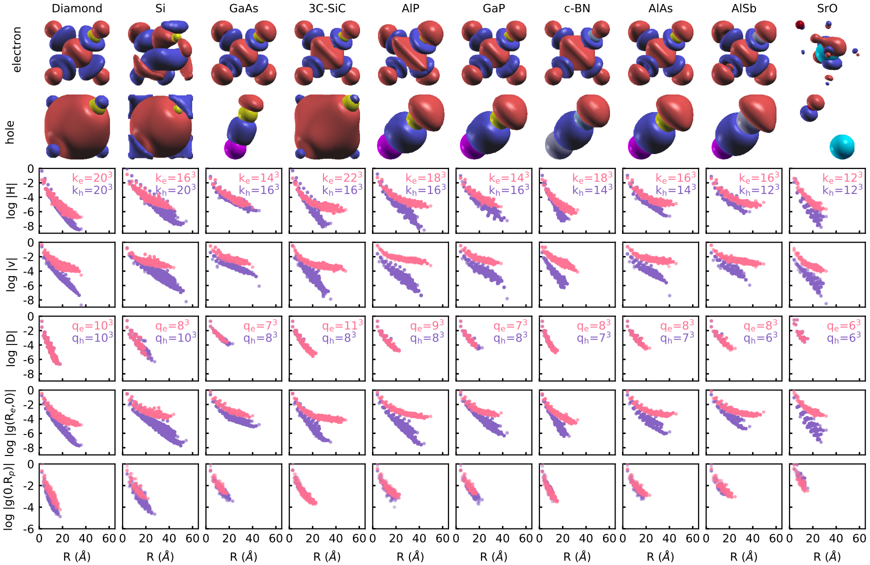
$$D^{\nu}(\Gamma, \mathbf{q}) = \frac{1}{\hbar N_{\mathrm{w}}}\left[2\rho V_{\mathrm{uc}}\hbar\omega_{\mathbf{q}\nu}\sum_{nm}|g_{mn\nu}(\Gamma, \mathbf{q})|^2\right]^{1/2}$$



SP *et al.*, Phys. Rev. Research **3**, 043022 (2021)

10 simple bulk semiconductors



Hall factor

$$r^{\mathrm{H}}_{\alpha\beta\gamma} \equiv \frac{\sigma^{\mathrm{H}}_{\alpha\beta\gamma}(\sigma_{\beta\alpha})^{-1}}{|B_\gamma|} = \frac{\mu^{\mathrm{H}}_{\alpha\beta\gamma}(\mu_{\beta\alpha})^{-1}}{|B_\gamma|}$$

Isotropic approximation:

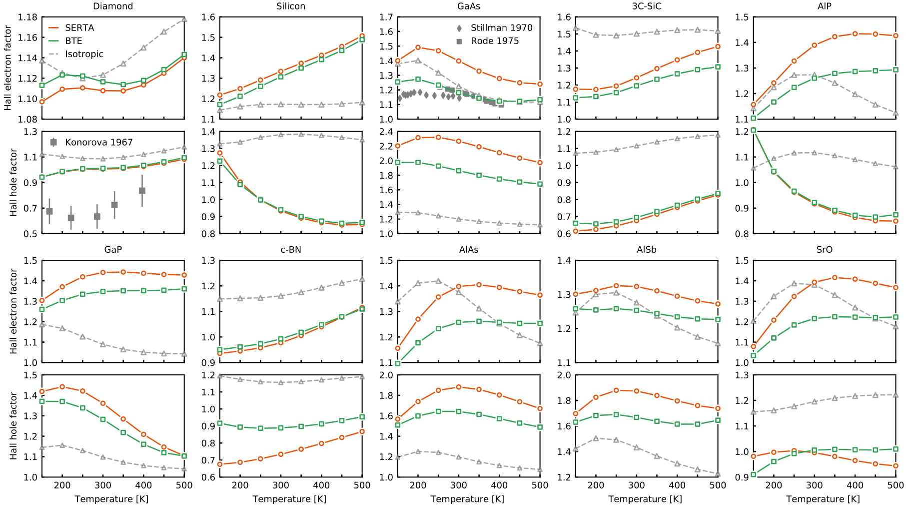
$$\begin{aligned} r^{\mathrm{iso}} &= \frac{\langle\tau^2\rangle}{\langle\tau\rangle^2} \\ \langle\tau^n\rangle &\equiv \frac{\int_0^\infty \tau^n(xk_{\mathrm{B}}T)x^{3/2}e^{-x}\mathrm{d}x}{\int_0^\infty x^{3/2}e^{-x}\mathrm{d}x}. \end{aligned}$$

Here, $x = \varepsilon/(k_{\mathrm{B}}T)$ and

$$\tau(\varepsilon) = \sum_n \int \frac{\mathrm{d}^3k}{\Omega_{\mathrm{BZ}}}\delta(\varepsilon - \varepsilon_{n\mathbf{k}})\tau_{n\mathbf{k}}$$

F. Macheda *et al.*, Nano Letters **20**, 8861 (2020)

Hall factor



SP *et al.*, Phys. Rev. Research **3**, 043022 (2021)

Temperature dependence mobility

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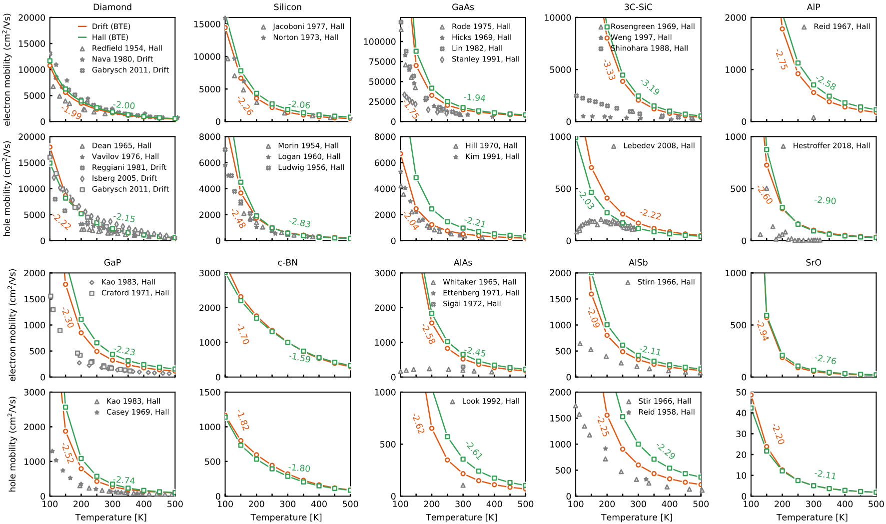
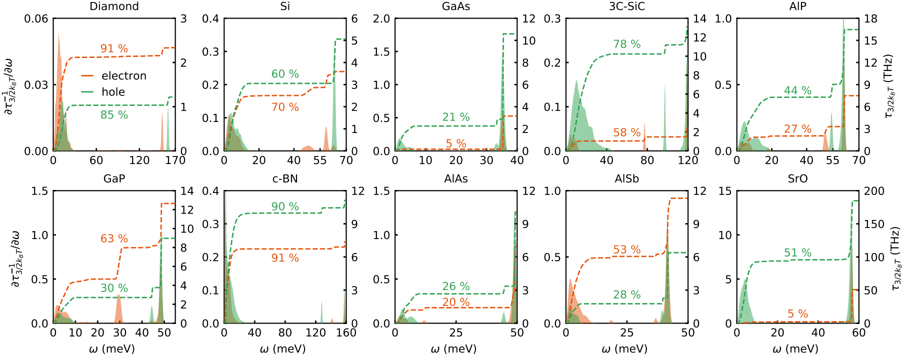


SP *et al.*, Phys. Rev. Research **3**, 043022 (2021)

Spectral decomposition



Optical scattering in GaAs-e, SrO-e, AlAs-e, GaAs-h, AlAs-h, AlP-e, AlSb-h, GaP-h, AlP-h.

SP *et al.*, Phys. Rev. Research **3**, 043022 (2021)

Mobility comparison with experiment

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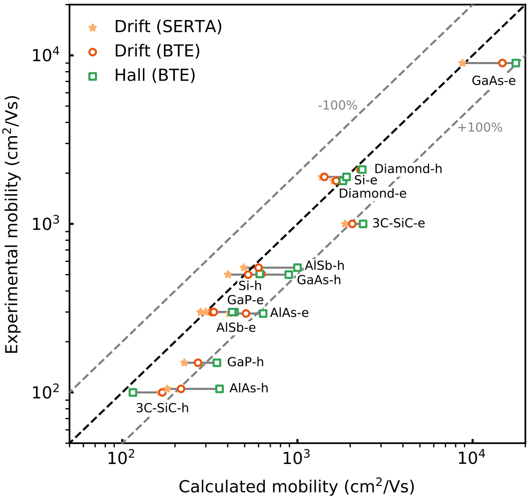

SP *et al.*, Phys. Rev. Research **3**, 043022 (2021)

2D materials - dynamical matrix

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$$D^{\mathrm{2D}}_{\kappa\alpha,\kappa'\beta}(\mathbf{q}) = D^{\mathcal{S},\mathrm{2D}}_{\kappa\alpha,\kappa'\beta}(\mathbf{q}) + D^{\mathcal{L},\mathrm{2D}}_{\kappa\alpha,\kappa'\beta}(\mathbf{q})$$

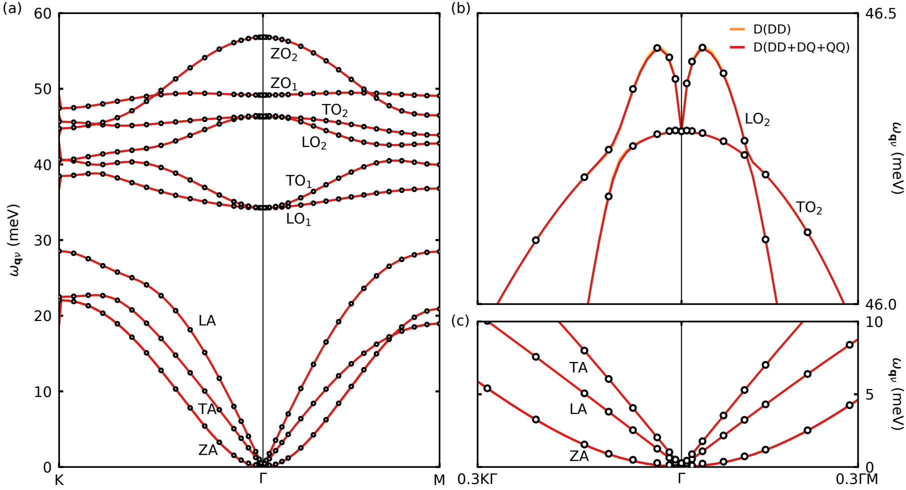
$$D^{\mathcal{L},2D}_{\kappa\alpha,\kappa'\beta}(\mathbf{G}+\mathbf{q}) = \frac{2\pi e^2 f(G+q)}{\frac{\Omega}{c}|G+q|}\left[\frac{Z^{\parallel,*}_{\kappa\alpha}(\mathbf{G}+\mathbf{q})Z^{\parallel}_{\kappa'\beta}(\mathbf{G}+\mathbf{q})}{1+\frac{2\pi f(G+q)}{|G+q|}\alpha^{\parallel}(\mathbf{G}+\mathbf{q})} - \frac{Z^{\perp,*}_{\kappa\alpha}(\mathbf{G}+\mathbf{q})Z^{\perp}_{\kappa'\beta}(\mathbf{G}+\mathbf{q})}{1-2\pi|G+q|f(G+q)\alpha^{\perp}}\right]e^{-i(\mathbf{G}+\mathbf{q})\cdot(\tau_{\kappa'\beta}-\tau_{\kappa\alpha})a}$$

$$Z^{\parallel}_{\kappa\alpha}(\mathbf{G}+\mathbf{q}) = \sum_{\beta}(G_\beta+q_\beta)Z^{\beta}_{\kappa\alpha} - \frac{i}{2}\sum_{\gamma}(G_\beta+q_\beta)(G_\gamma+q_\gamma)Q^{\beta\gamma}_{\kappa\alpha} - \frac{i}{2}|G+q|^2 Q^{zz}_{\kappa\alpha}$$

$$Z^{\perp}_{\kappa\alpha}(\mathbf{G}+\mathbf{q}) = |G+q|Z^{z}_{\kappa\alpha} - i\sum_{\beta}|G+q|(G_\beta+q_\beta)Q^{z\beta}_{\kappa\alpha},$$

M. Royo *et al.*, Phys. Rev. X **11**, 041027 (2021)

Phonon frequency of MoS_2 monolayer



Deformation potential of MoS_2 monolayer

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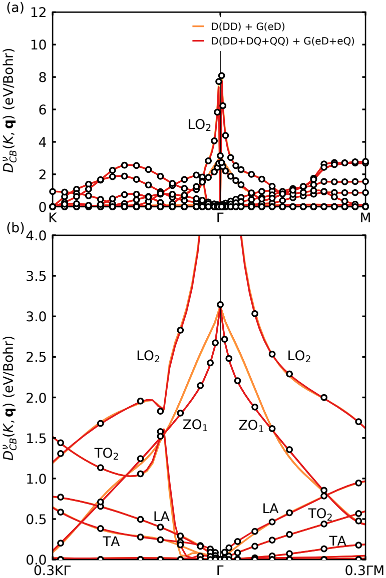
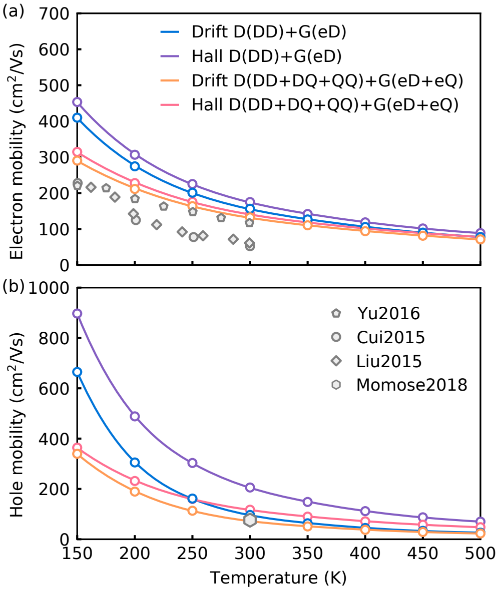


Mobility of MoS_2 monolayer



Conclusion

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- Efficient implementation of drift and Hall mobility in 3D and 2D materials
- Dynamical quadrupoles are important for accurate interpolation
- Phonon-limited mobility overestimates experimental mobility

Open / Discussion

- 1D materials
- Higher level of theory - spectral functions ... - systematic benchmark needed
- Effect of anharmonicity on carrier mobility
- Transport with electron localization

Electron-Phonon School 2022 - Austin (US)

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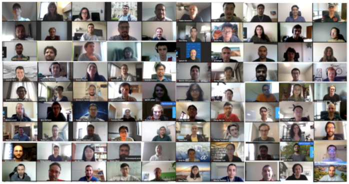
ICTP School 2018

Virtual School 2021



Electron-Phonon School 2022

13-19 June 2022, Austin, TX

NSF

photo by Florence Jones on Unsplash

More info

UCLouvain

https://epw-code.org

https://forum.epw-code.org

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

SP, F. Macheda, E.R. Margine, N. Marzari, N. Bonini, F. Giustino, Phys. Rev. Research **3**, 043022 (2021)

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F. Macheda, SP, F. Giustino, N. Bonini, Nano Lett. **20**, 8861 (2020)

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Quadrupole

$$g_{mn\nu}^{\mathcal{L},\mathrm{eQ}}(\mathbf{k},\mathbf{q}) = \frac{4\pi}{V_{\mathrm{uc}}}\frac{e^2}{4\pi\varepsilon^0}\sum_{\kappa}\left[\frac{\hbar}{2N_{p'}M_\kappa\omega_{\mathbf{q}\nu}}\right]^{\frac{1}{2}}\sum_{\mathbf{G}\neq-\mathbf{q}} \\ \times \frac{(\mathbf{G}+\mathbf{q})\cdot(\mathbf{G}+\mathbf{q})\cdot\mathbf{e}_{\kappa\mathbf{q}\nu}\cdot\tilde{\mathbf{Q}}_{mn\kappa}(\mathbf{k},\mathbf{G}+\mathbf{q})}{(\mathbf{G}+\mathbf{q})\cdot\boldsymbol{\varepsilon}^\infty\cdot(\mathbf{G}+\mathbf{q})}e^{-i(\mathbf{G}+\mathbf{q})\cdot\boldsymbol{\tau}_\kappa},$$

where $\tilde{\mathbf{Q}}_{mn\kappa}(\mathbf{k},\mathbf{G}+\mathbf{q})$ is a third-rank effective quadrupole tensor defined as:

$$\tilde{Q}_{mn\kappa\alpha\beta\gamma}(\mathbf{k},\mathbf{G}+\mathbf{q}) = \frac{1}{2}Q_{\kappa\alpha\beta\gamma}\langle\Psi_{m\mathbf{k}+\mathbf{q}}|e^{i(\mathbf{q}+\mathbf{G})\cdot\mathbf{r}}|\Psi_{n\mathbf{k}}\rangle \\ - eZ^*_{\kappa\alpha,\beta}\langle\Psi_{m\mathbf{k}+\mathbf{q}}|e^{i(\mathbf{q}+\mathbf{G})\cdot\mathbf{r}}V^{\mathrm{Hxc},\mathcal{E}_\gamma}(\mathbf{r})|\Psi_{n\mathbf{k}}\rangle,$$

and $Q_{\kappa\alpha\beta\gamma}$ is the dynamic quadrupole tensor.

Effects

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