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Institute of Condensed Matter
and Nanosciences

Efficient transport calculations in 2D materials.

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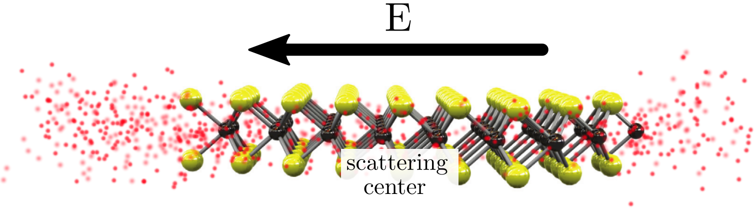
Phonon-limited carrier mobilities

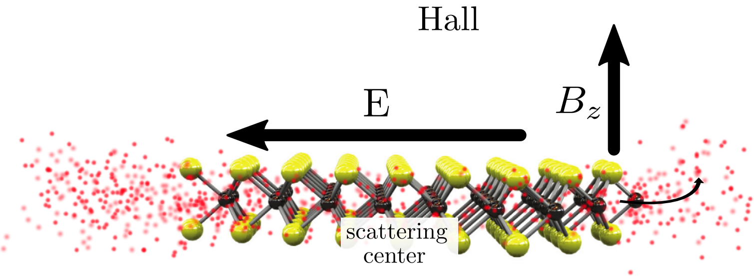
Drift



lattice (phonon)
ionized impurity
alloy
defects

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

Phonon-limited carrier mobilities



lattice (phonon)
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alloy
defects

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

Drift and Hall mobility

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

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

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

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

$\rightarrow$ 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)

Linearlized Hall Boltzmann transport eq.

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$$\left[1 - \frac{e}{\hbar}\tau_{n\mathbf{k}}(\mathbf{v}_{n\mathbf{k}} \times \mathbf{B}) \cdot \nabla_{\mathbf{k}}\right] \partial_{E_\beta} f_{n\mathbf{k}}(\mathbf{B}) = e v_{n\mathbf{k}\beta} \frac{\partial f^0_{n\mathbf{k}}}{\partial \varepsilon_{n\mathbf{k}}} \tau_{n\mathbf{k}}$$

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

$$+ (n_{\mathbf{q}\nu} + f^0_{n\mathbf{k}})\delta(\varepsilon_{n\mathbf{k}} - \varepsilon_{m\mathbf{k}+\mathbf{q}} - \hbar\omega_{\mathbf{q}\nu})\Big] \partial_{E_\beta} f_{m\mathbf{k}+\mathbf{q}}(\mathbf{B}),$$

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)
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Electron-phonon Wannier (EPW)

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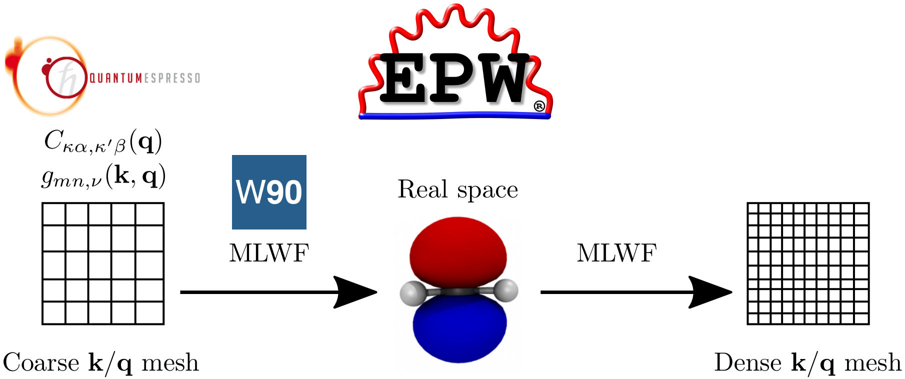
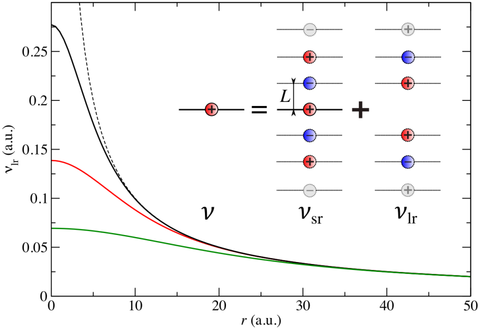
EPW relies on MLWF to interpolate electron-phonon matrix elements.



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

Long-range separable representation

Image-charge decomposition



Range-separation function $f(|\mathbf{q}|) = 1 - \tanh(|\mathbf{q}|L/2)$

M. Royo and M. Stengel, Phys. Rev. X **11**, 041027 (2021)

Long-range el-ph in 2D materials

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$$g_{mn\nu}^{\mathrm{2D}}(\mathbf{k},\mathbf{q}) = g_{mn\nu}^{\mathcal{S},\mathrm{2D}}(\mathbf{k},\mathbf{q}) + \boxed{g_{mn\nu}^{\mathcal{L},\mathrm{2D}}(\mathbf{k},\mathbf{q})}$$

$$\boxed{g_{mn\nu}^{\mathcal{L},\mathrm{2D}}(\mathbf{k},\mathbf{q})} = \sum_{\kappa\alpha}\left[\frac{\hbar}{2NM_\kappa S\omega_\nu(\mathbf{q})}\right]^{\frac{1}{2}}\sum_{l} \times \frac{(-1)^{l+1}2\pi f(|\mathbf{q}|)/|\mathbf{q}|}{1-(-1)^{l+1}\frac{2\pi f(|\mathbf{q}|)}{|\mathbf{q}|}\tilde{\chi}_{l\mathbf{0}\mathbf{0}}^{\mathcal{S},\mathrm{2D}}}(\mathbf{q})\tilde{\rho}_{l\kappa\alpha\mathbf{0}}^{\mathcal{S},\mathrm{2D}}(\mathbf{q})e_{\kappa\alpha\nu}(\mathbf{q}) \times \langle\Psi_{m\mathbf{k}+\mathbf{q}}|\, e^{i\mathbf{q}\cdot\mathbf{r}}[\phi_{l\mathbf{0}}^{\mathrm{2D}}(\mathbf{q},z) + v_{l\mathbf{0}}^{\mathcal{S},\mathrm{2D}}(\mathbf{q},\mathbf{r})]\,|\Psi_{n\mathbf{k}}\rangle\,.$$

- $l = 2 \rightarrow 0$ in materials where the modes are inversion odd
- **approximation:** we neglect the induced short-range potential $v_{l\mathbf{0}}^{\mathcal{S},\mathrm{2D}}(\mathbf{q},\mathbf{r})$ [small in bulk materials - G. Brunin et al., Phys. Rev. Lett. 125, 136601 (2020)]

Long-range el-ph in 2D materials

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$$
\begin{aligned}
g^{2\mathrm{D}}_{mn\nu}(\mathbf{k},\mathbf{q}) =& g^{\mathcal{S},2\mathrm{D}}_{mn\nu}(\mathbf{k},\mathbf{q}) + \boxed{g^{\mathcal{L},2\mathrm{D}}_{mn\nu}(\mathbf{k},\mathbf{q})} \\
\boxed{g^{\mathcal{L},2\mathrm{D}}_{mn\nu}(\mathbf{k},\mathbf{q})} =& \sum_{\kappa\alpha}\left[\frac{\hbar}{2NM_\kappa S\omega_\nu(\mathbf{q})}\right]^{\frac{1}{2}}\sum_l \\
& \times \frac{(-1)^{l+1}2\pi f(|\mathbf{q}|)/|\mathbf{q}|}{1-(-1)^{l+1}\frac{2\pi f(|\mathbf{q}|)}{|\mathbf{q}|}\tilde{\chi}^{\mathcal{S},2\mathrm{D}}_{l\mathbf{0}\mathbf{0}}}(\mathbf{q})\tilde{\rho}^{\mathcal{S},2\mathrm{D}}_{l\kappa\alpha\mathbf{0}}(\mathbf{q})e_{\kappa\alpha\nu}(\mathbf{q}) \\
& \times \langle\Psi_{m\mathbf{k}+\mathbf{q}}|\, e^{i\mathbf{q}\cdot\mathbf{r}}[\phi^{2\mathrm{D}}_{l\mathbf{0}}(\mathbf{q},z) + v^{\mathcal{S},2\mathrm{D}}_{l\mathbf{0}}(\mathbf{q},\mathbf{r})]\,|\Psi_{n\mathbf{k}}\rangle\,.
\end{aligned}
$$

- $l = 2 \rightarrow 0$ in materials where the modes are inversion odd
- **approximation:** we neglect the induced short-range potential $v^{\mathcal{S},2\mathrm{D}}_{l\mathbf{0}}(\mathbf{q},\mathbf{r})$ [small in bulk materials - G. Brunin et al., Phys. Rev. Lett. 125, 136601 (2020)]

Long-range el-ph in 2D materials

Monopole-dipole:

$$g_{mn\nu}^{\mathcal{L},\mathrm{2D},\mathrm{eD}}(\mathbf{k},\mathbf{q}) = i\sum_{\kappa\alpha}\left[\frac{\hbar}{2NM_\kappa\omega_{\mathbf{q}\nu}}\right]^{\frac{1}{2}}\sum_{\mathbf{G}\neq-\mathbf{q}}\frac{2\pi f(G+q)}{S|G+q|}e_{\kappa\alpha\mathbf{q}\nu}e^{-ia(\mathbf{G}+\mathbf{q})\cdot\boldsymbol{\tau}_\kappa}$$
$$\times\frac{(\mathbf{G}+\mathbf{q})\cdot\mathbf{Z}_{\kappa\alpha}\langle\Psi_{m\mathbf{k}+\mathbf{q}}|e^{i(\mathbf{G}+\mathbf{q})\cdot\mathbf{r}}|\Psi_{n\mathbf{k}}\rangle}{1+\frac{2\pi f(G+q)}{|G+q|}\alpha^{\parallel}(\mathbf{G}+\mathbf{q})},$$

and the monopole-quadrupole:

$$g_{mn\nu}^{\mathcal{L},\mathrm{2D},\mathrm{eQ}}(\mathbf{k},\mathbf{q}) = \frac{1}{2}\sum_{\kappa\alpha}\left[\frac{\hbar}{2NM_\kappa\omega_{\mathbf{q}\nu}}\right]^{\frac{1}{2}}\sum_{\mathbf{G}\neq-\mathbf{q}}\frac{2\pi f(G+q)}{S|G+q|}e_{\kappa\alpha\mathbf{q}\nu}e^{-ia(\mathbf{G}+\mathbf{q})\cdot\boldsymbol{\tau}_\kappa}$$
$$\times\frac{(\mathbf{G}+\mathbf{q})\cdot(\mathbf{G}+\mathbf{q})\cdot\mathbf{Q}_{\kappa\alpha}+|G+q|^2Q_{\kappa\alpha}^{zz}}{1+\frac{2\pi f(G+q)}{|G+q|}\alpha^{\parallel}(\mathbf{G}+\mathbf{q})}\langle\Psi_{m\mathbf{k}+\mathbf{q}}|e^{i(\mathbf{G}+\mathbf{q})\cdot\mathbf{r}}|\Psi_{n\mathbf{k}}\rangle.$$

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

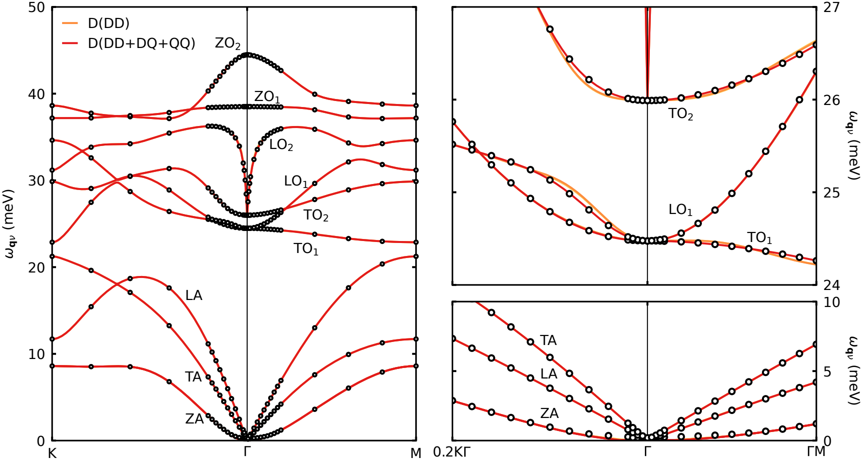
$$\boxed{D^{\mathcal{L},2D}_{\kappa\alpha,\kappa'\beta}(\mathbf{G}+\mathbf{q})} = \frac{2\pi e^2 f(G+q)}{\frac{\Omega}{c}|G+q|}\left[\frac{\boxed{Z^{\parallel,*}_{\kappa\alpha}(\mathbf{G}+\mathbf{q})Z^{\parallel}_{\kappa'\beta}(\mathbf{G}+\mathbf{q})}}{1+\frac{2\pi f(G+q)}{|G+q|}\alpha^{\parallel}(\mathbf{G}+\mathbf{q})}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

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

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

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

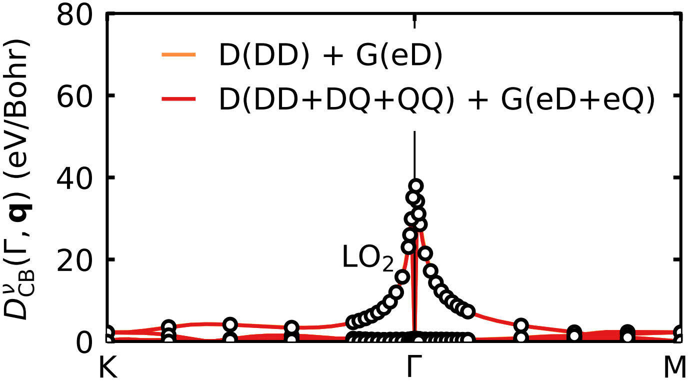
SnS_2 phonon



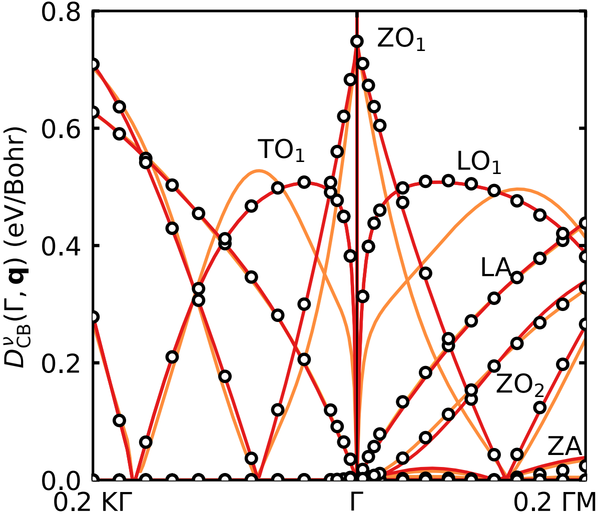
D(DD)
D(DD+DQ+QQ)
ZO2
ZO1
LO2
LO1
TO2
TO1
LA
TA
ZA
ω_qν (meV)
K
Γ
M
0.2KΓ
ΓM
TO2
LO1
TO1
TA
LA
ZA

SnS_2 deformation potential

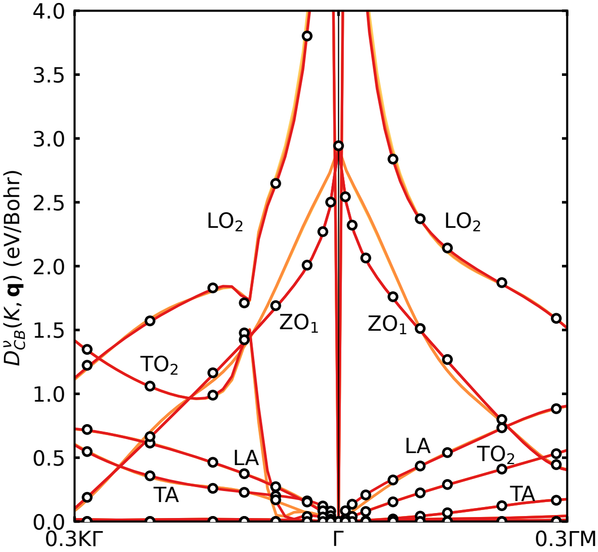
$$D^{\nu}(\Gamma,\mathbf{q}) = \frac{1}{\hbar N_{\mathrm{w}}}\left[2\rho S_{\mathrm{uc}}\hbar\omega_{\mathbf{q}\nu}\sum_{nm}|g_{mn\nu}(\Gamma,\mathbf{q})|^2\right]^{1/2}$$



SnS_2 deformation potential



MoS_2 deformation potential



MoS_2 mobility

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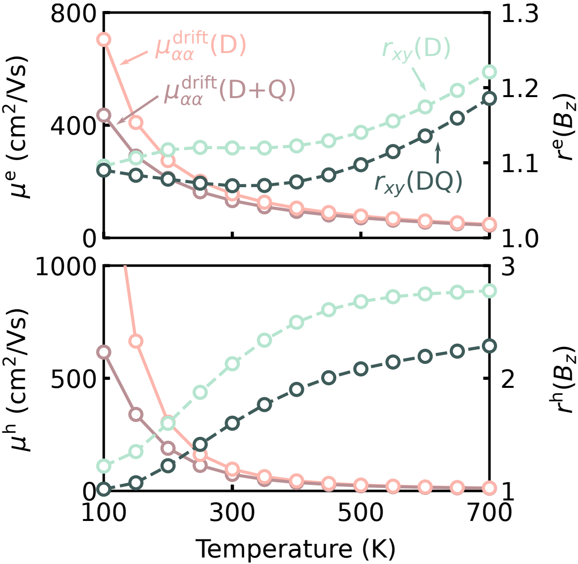
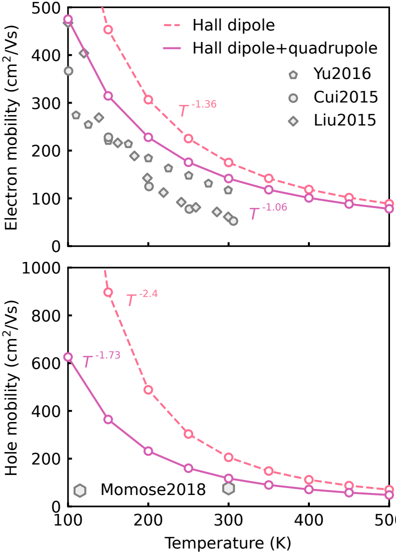
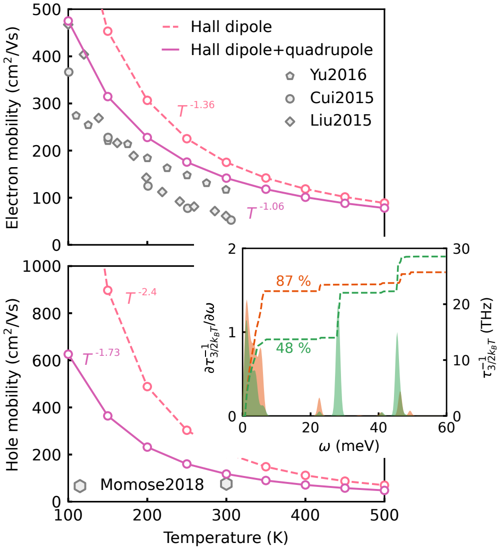


MoS_2 mobility



MoS_2 mobility



Conclusion

- Efficient implementation of drift and Hall mobility in 2D materials
- Dynamical quadrupoles are important for accurate interpolation

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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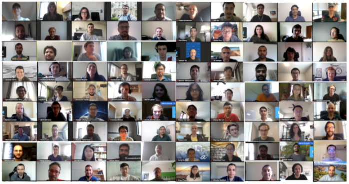
ICTP School 2018

Virtual School 2021



Electron-Phonon School 2022

13-19 June 2022, Austin, TX

More info

https://epw-code.org

https://forum.epw-code.org

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

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Range separation

We minimize

$$d(L) = \frac{1}{N} \sum_{\kappa\kappa'}^{\prime} \sum_{l\alpha\beta} |D^{\mathcal{S}}_{\kappa\alpha,\kappa'\beta}(l, l')|$$