Finding and engineering high-conductivity 2D semiconductors from first principles

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GDR IAMAT - Annual Meeting - June 1st 2022

Discovery and Design

High-mobility quest

~100,000
Experimentally known 3D compounds

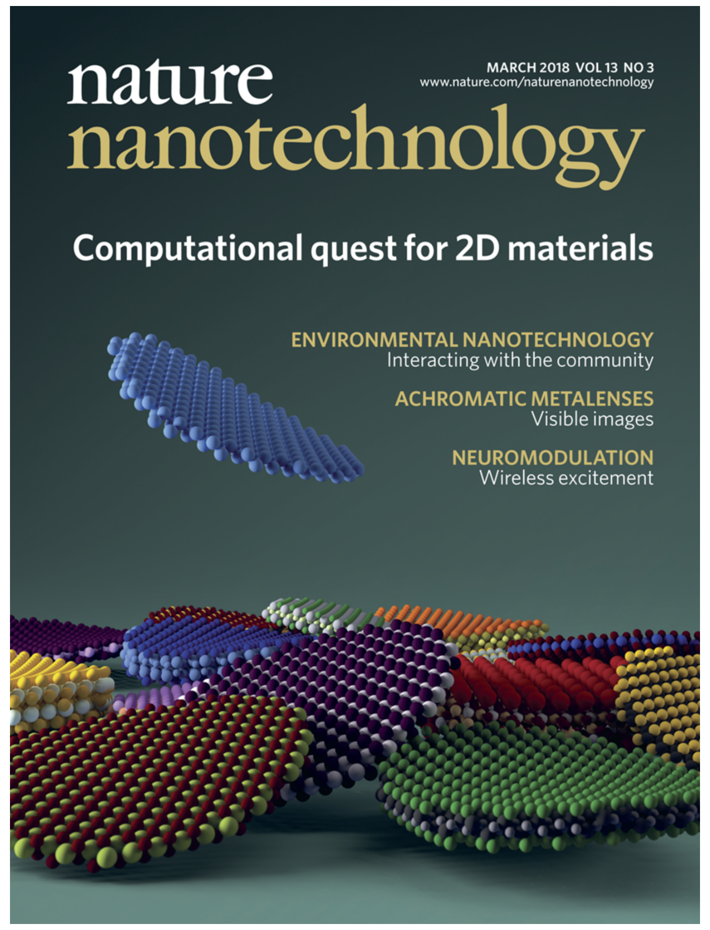
~1000
Easily Exfoliable materials

~250
Small unit cells
($\leq$ 6 atoms)

AiiDA

MATERIALSCLOUD
https://www.materialscloud.org/

From 2D team @ THEOS:



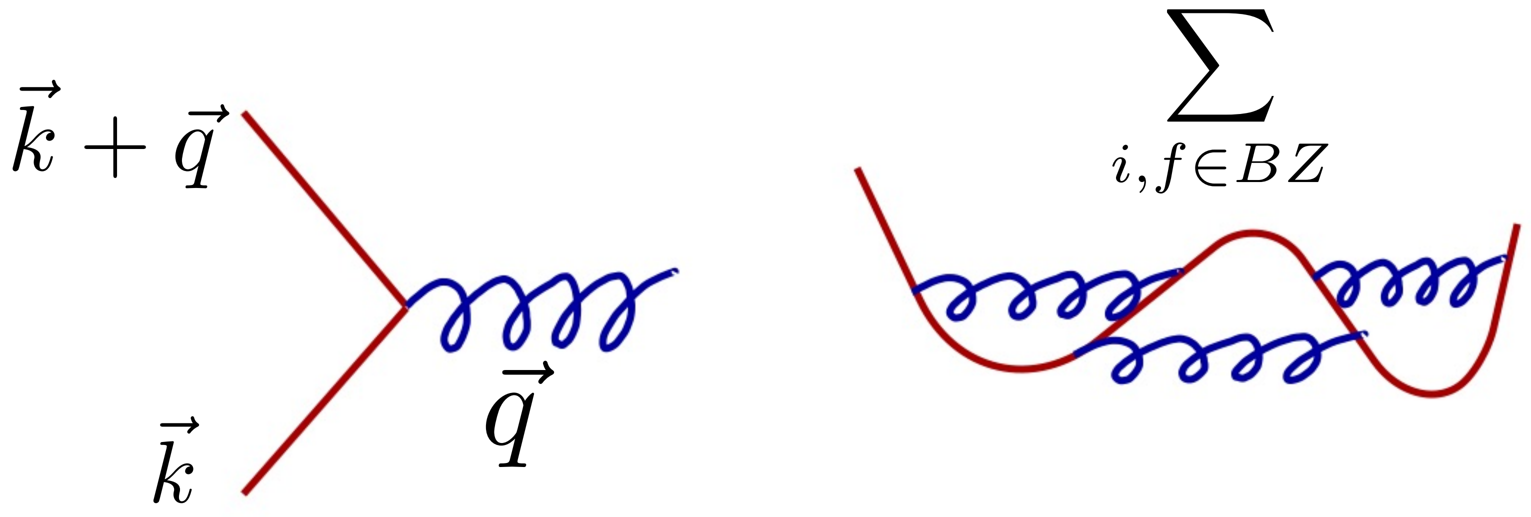
[Mounet et al. Nature Nanotechnology **13**, 246–252 (2018)]

How to find high conductivity semiconductors?

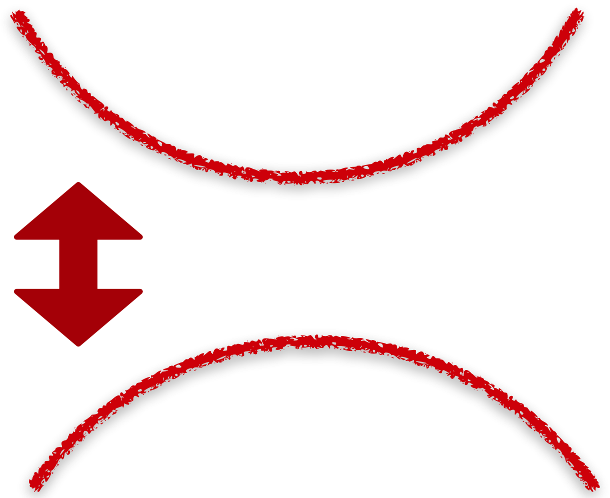
High-mobility 2D materials
Context

Phonon-limited charge transport in gated 2D semiconductors

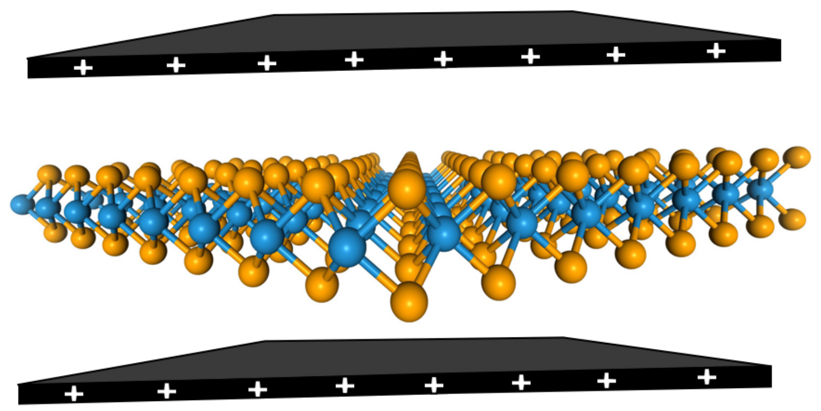
- Only electron-phonon scattering



- Materials with a gap



- Electrostatic doping



Objective: High conductivity / mobility

Why?
- Applications: transistors, sensors, optoelectronics…
- Physics of electron-phonon scattering

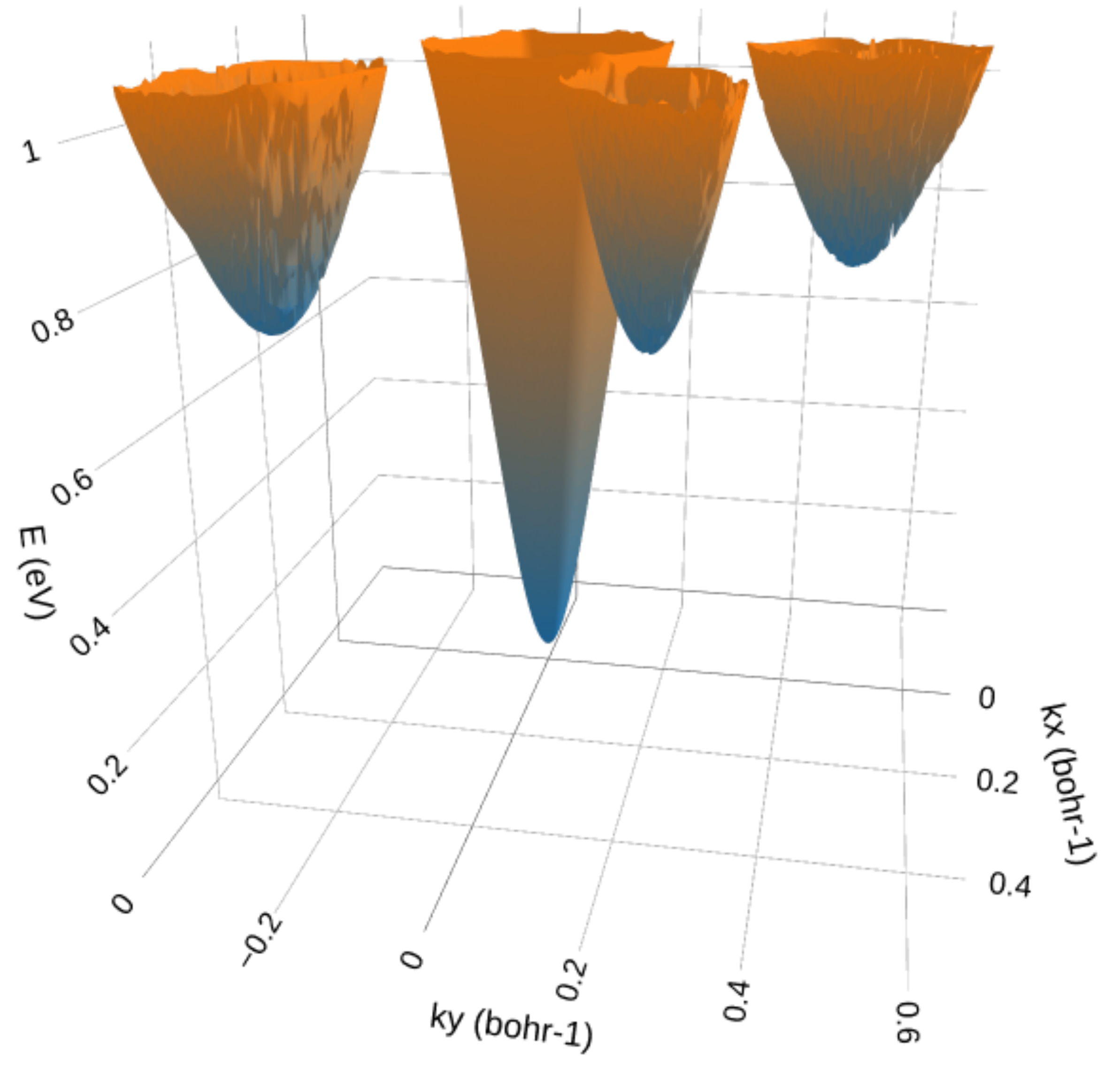
How?
- Intrinsic (discovery) and/or engineered (design)

Profiling high-conductivity materials

Learn from the bests

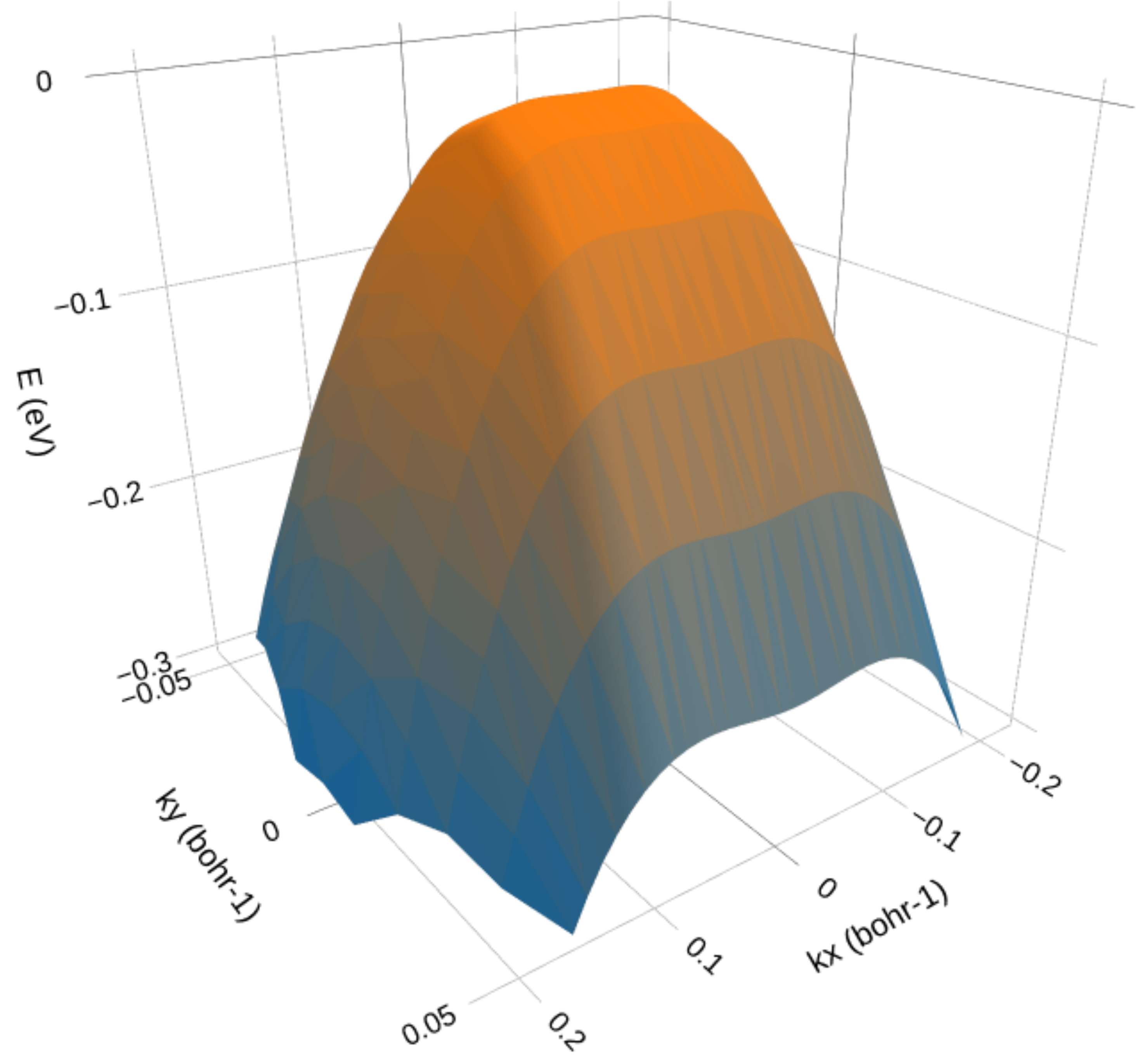
(Electronic) structure-property relation

InSe (electrons)



Steep and deep single valley

P4 (holes)

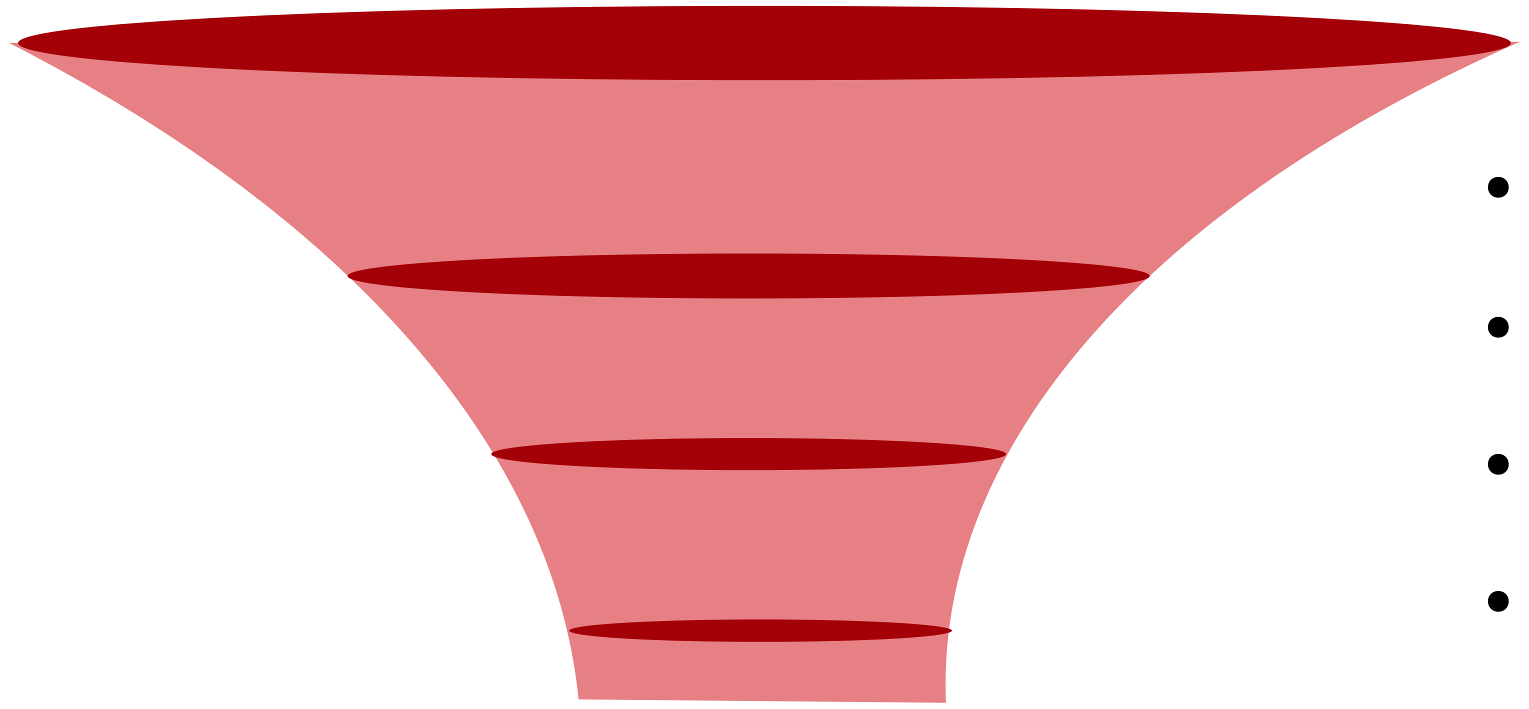


Anisotropic single valley

[T. Sohier, M. Gibertini, N. Marzari, 2D Mater. **8** 015025 (2021)]

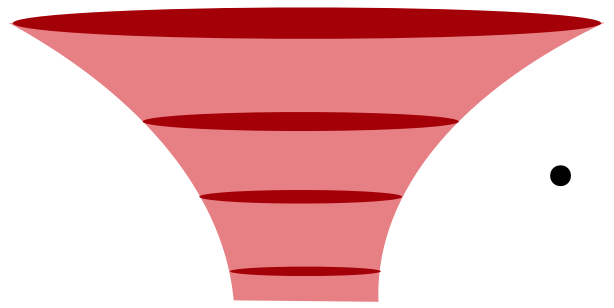
Profiling high-conductivity materials

Screening procedure



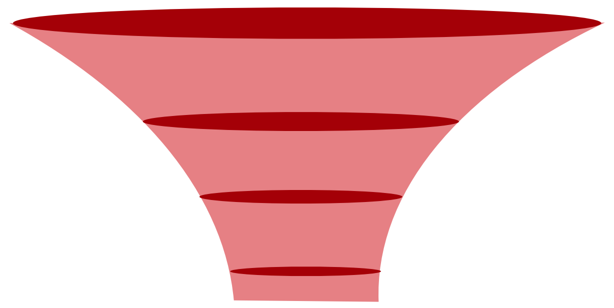
- 256 $\leq$6 atoms per unit-cell
- 150 semiconductors
- 110 with positive phonons
- 44 (e) + 13 (h) with 1 valley and gap < 2.5 eV

Like InSe



- v_F > 6 ARU

Like Phosphorene



- v_F > 2 ARU
- v ratio > 1.7

- Sb_2SeTe_2
- Bi_2SeTe_2
- GaSe
- $BiSe_3$
- $AlLiTe_2$
- BiClTe

- AuI
- ClGaTe

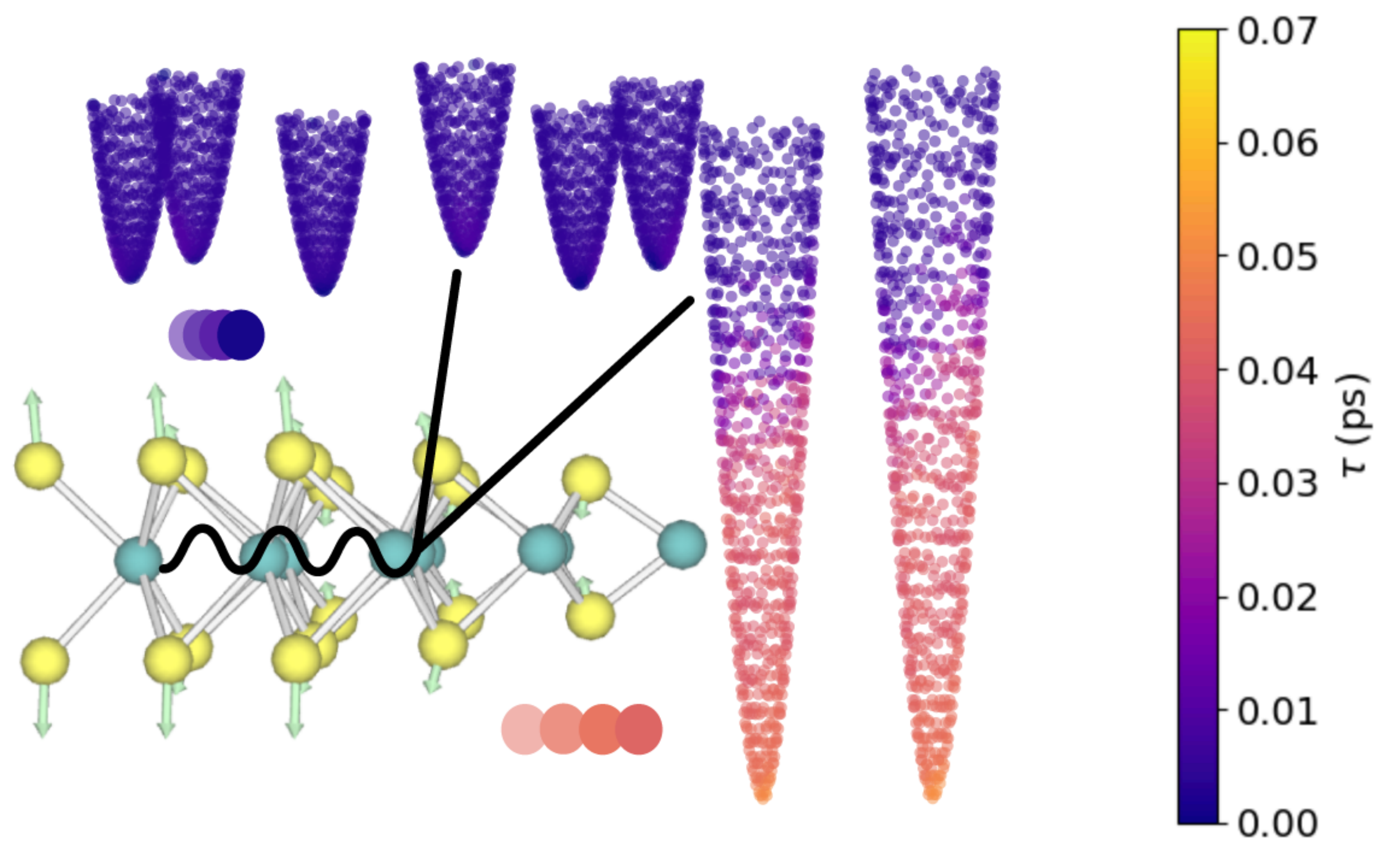
[T. Sohier, M. Gibertini, N. Marzari, 2D Mater. **8** 015025 (2021)]

Profiling high-conductivity materials

A dozen excellent candidates

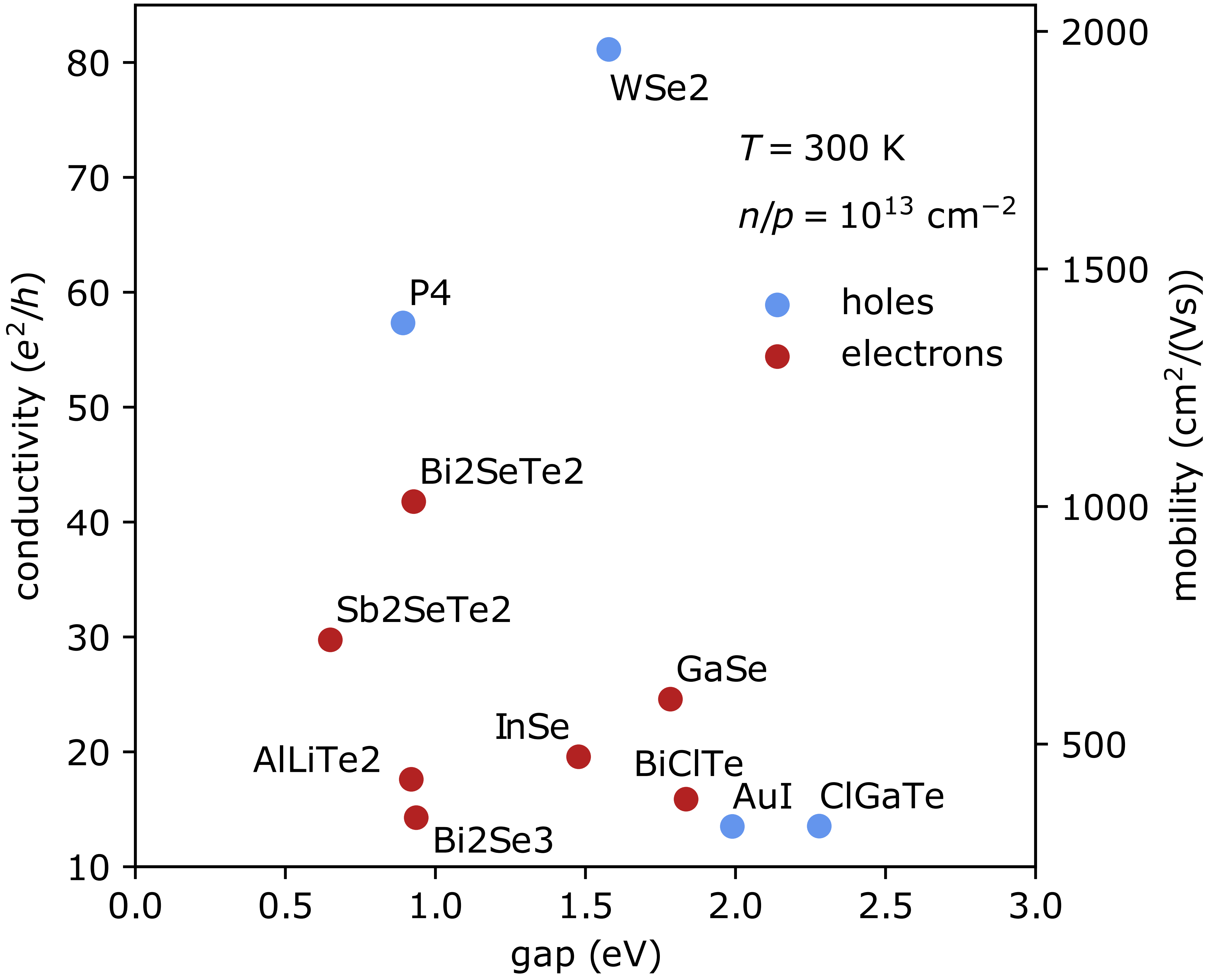
AiiDA

Predictive and automated transport workflow



Scatt. time

$$\sigma = 2e^2 \int_{\mathbf{k}} \frac{d\mathbf{k}}{(2\pi)^2} \left(\mathbf{v}(\mathbf{k}) \cdot \mathbf{u_E}\right)^2 \tau(\mathbf{k}) \frac{\partial f^0}{\partial \varepsilon}$$



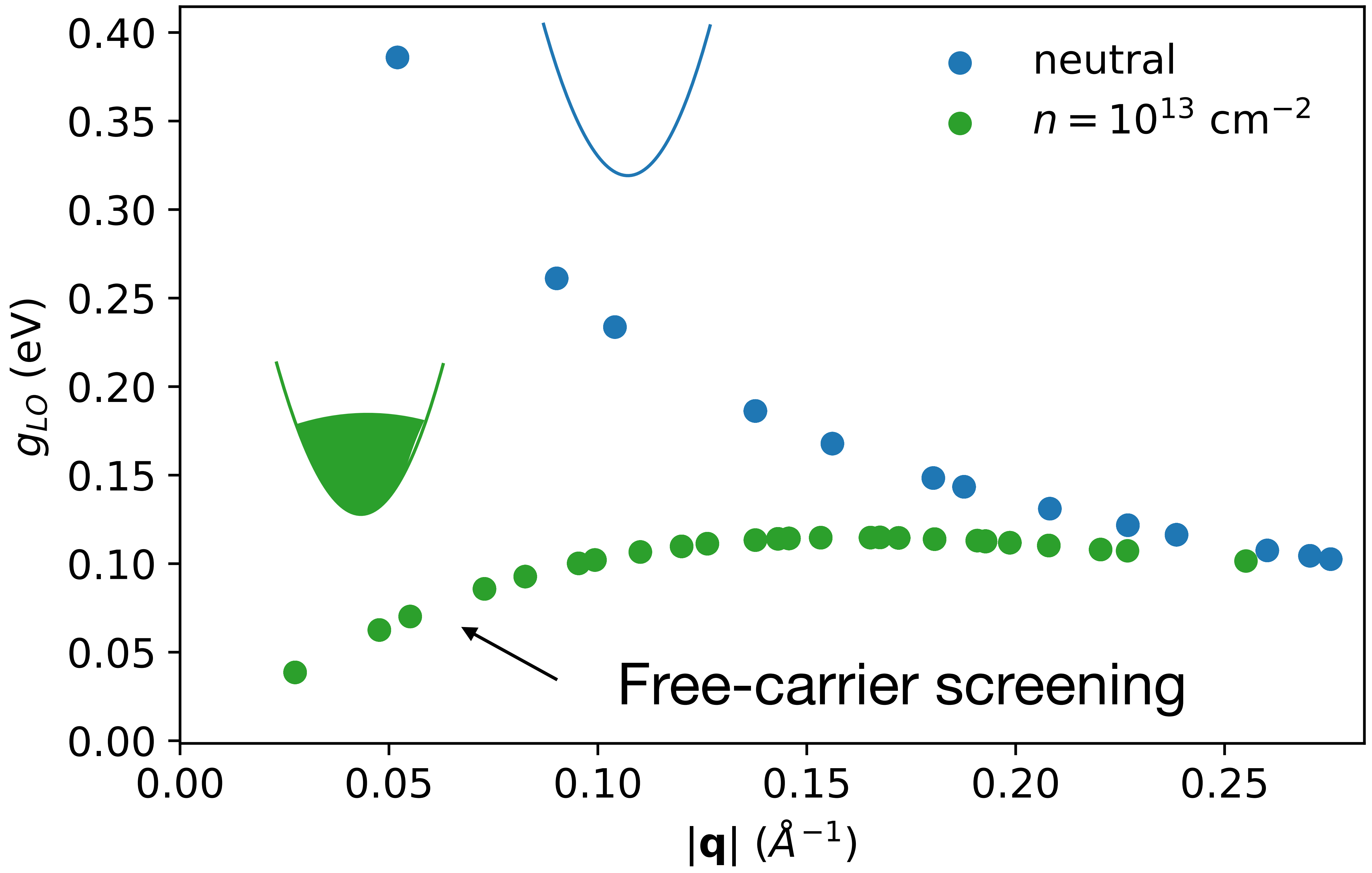
Full provenance available on Materials Cloud archive

Most electron-doped monolayers are limited by Fröhlich interactions…

Remote screening

GaSe, a prototypical Fröhlich-limited material

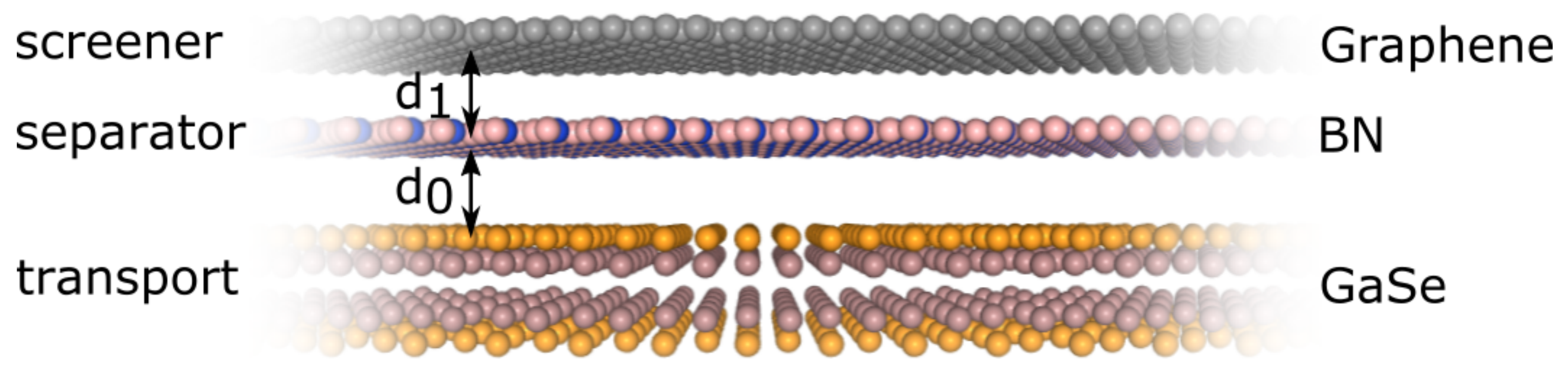
n (cm^{-2})	10^{13}	10^{11} (~ neutral)
μ (cm^2/(Vs))	600	174

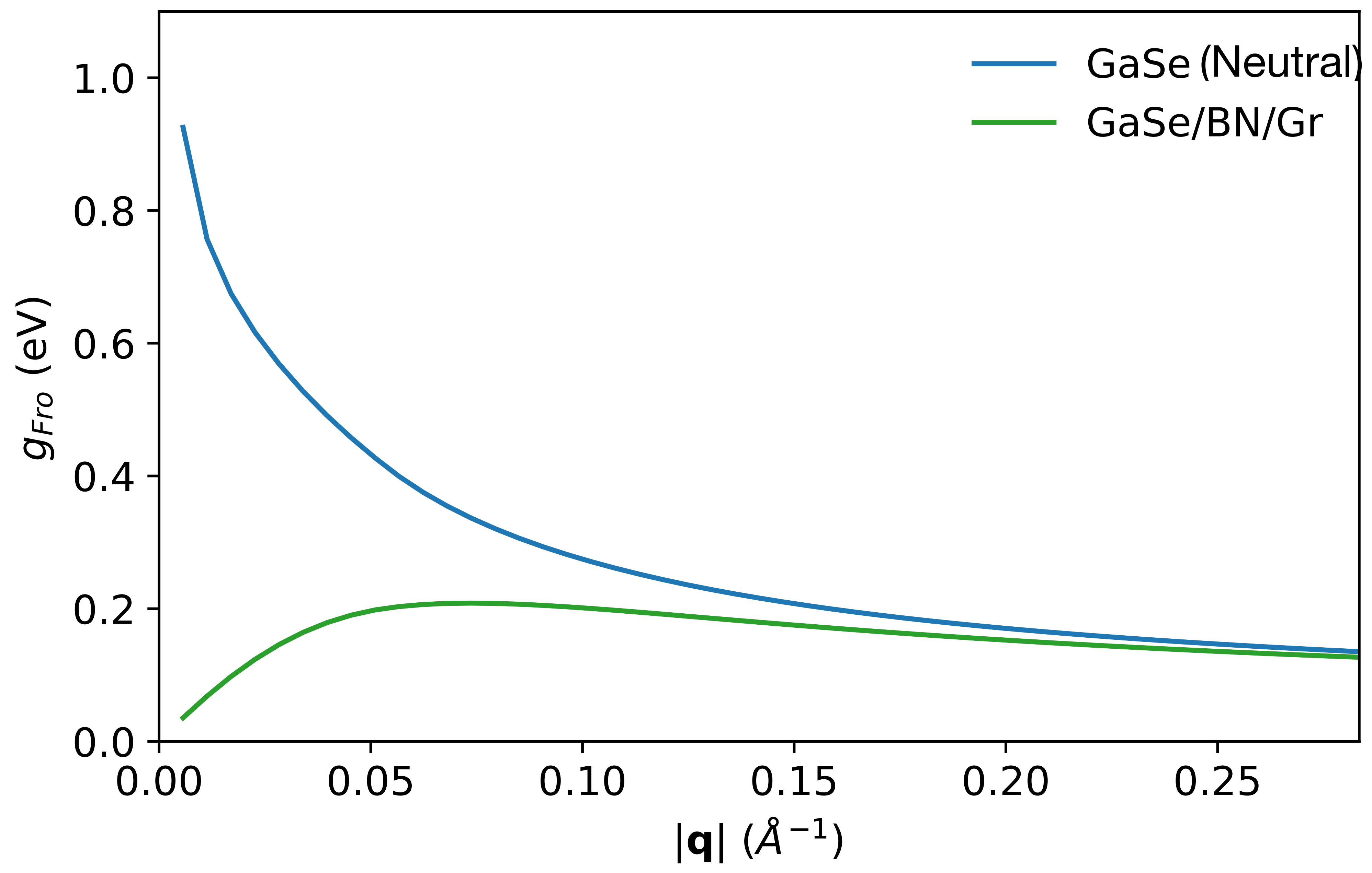


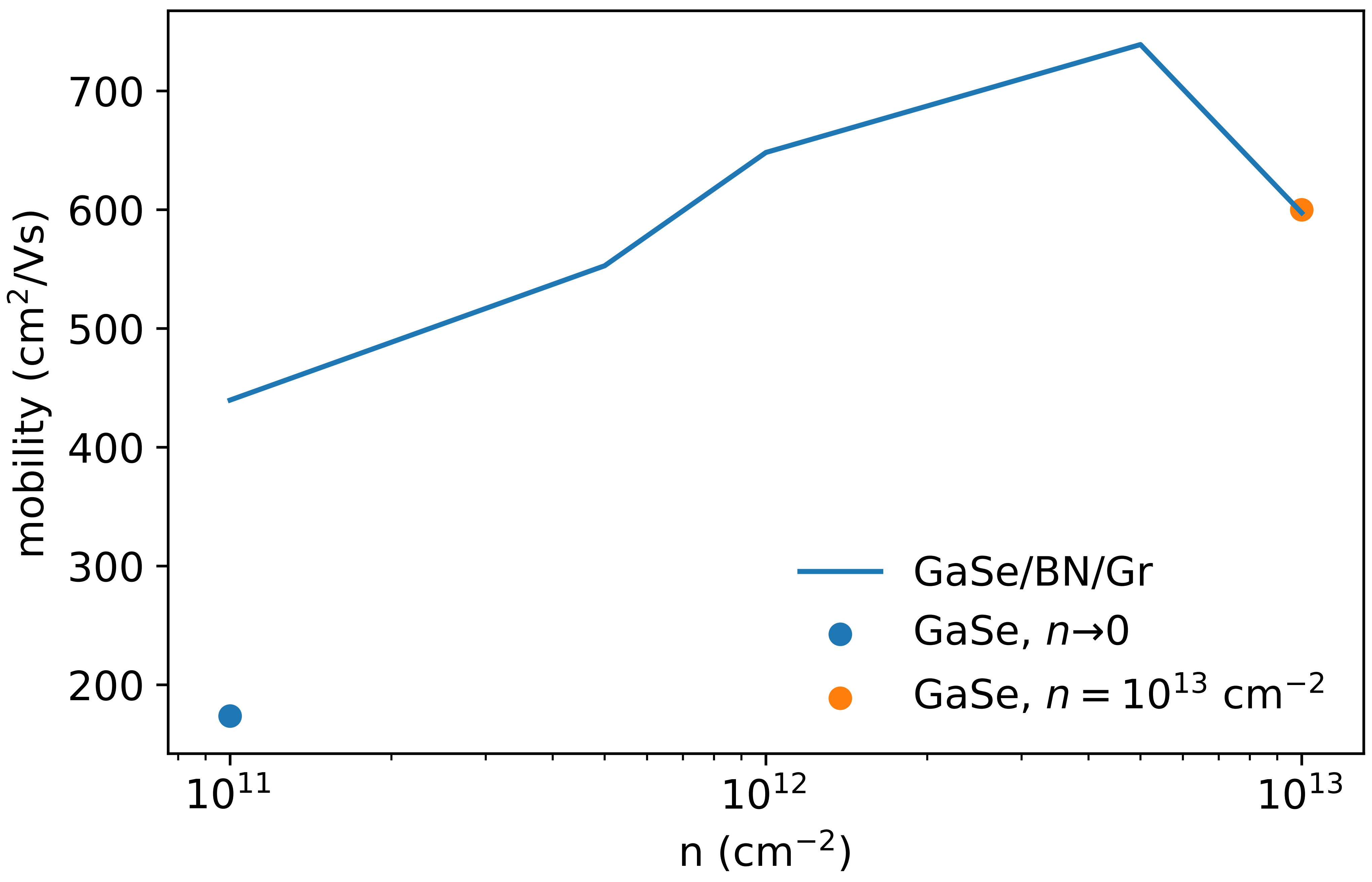
[T. Sohier, M. Gibertini, M. Verstraete, PRM 5, 024004 (2021)]

Remote screening

Mobility enhancement at low carrier density







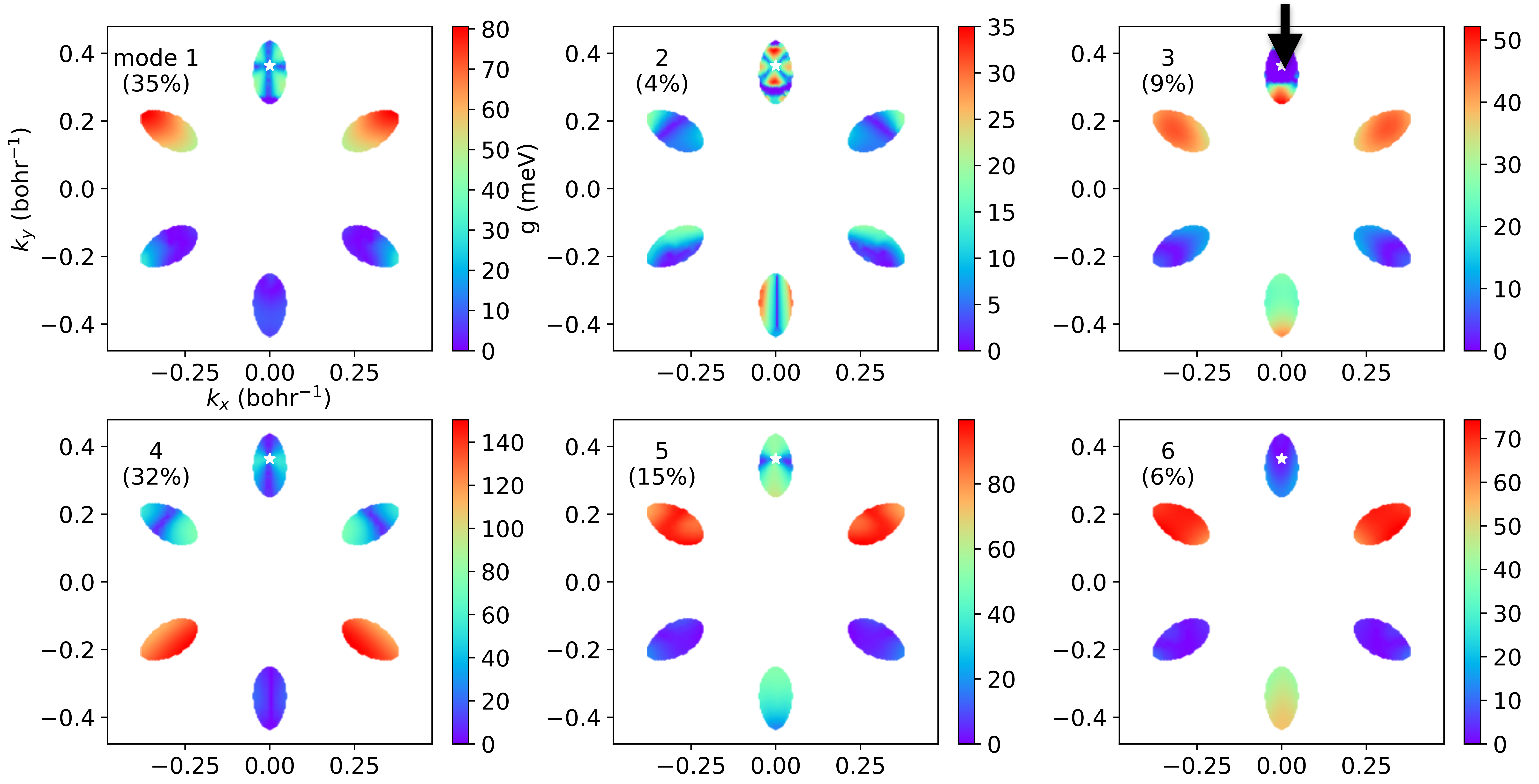
Works for any material dominated by screenable el-ph coupling

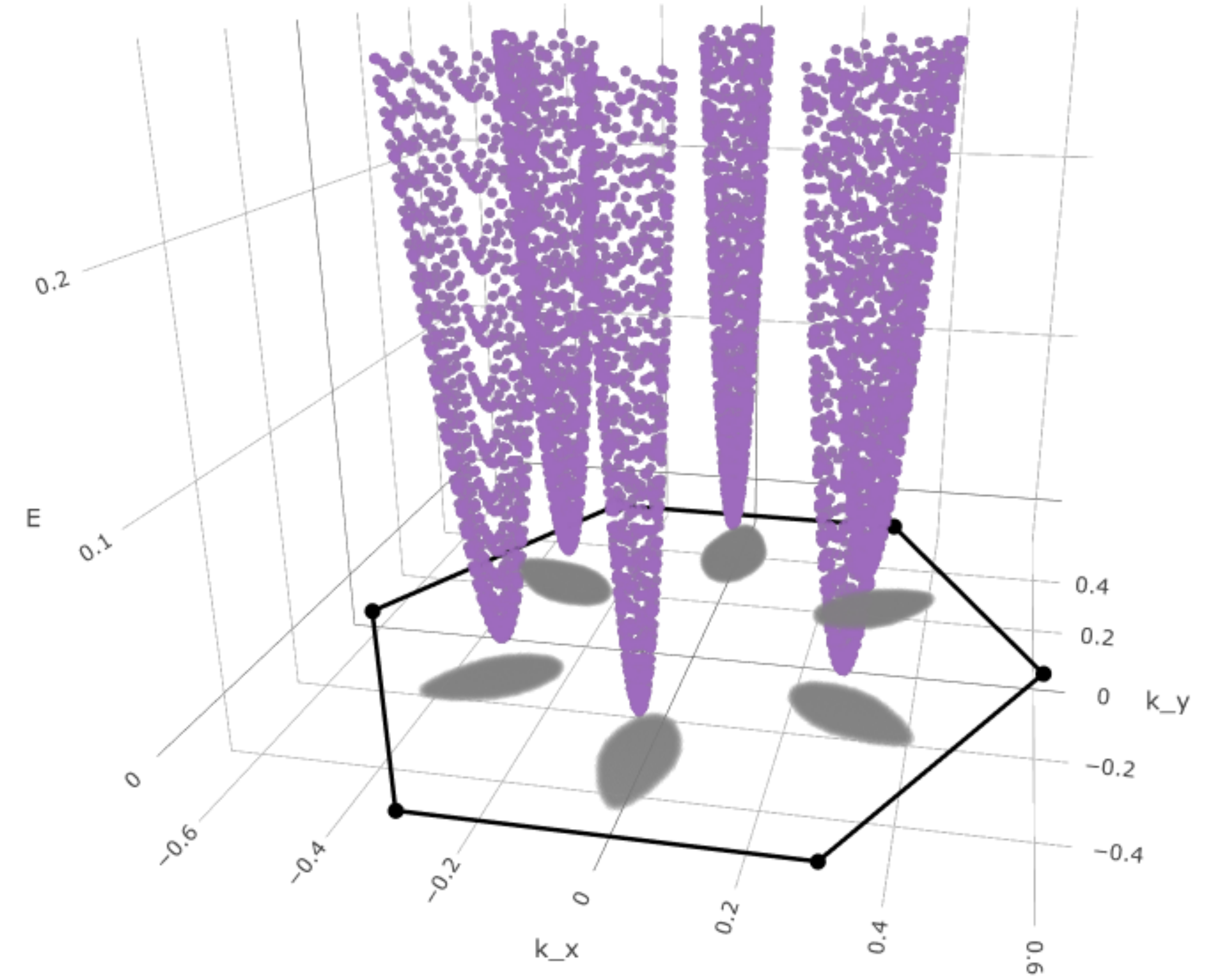
[T. Sohier, M. Gibertini, M. Verstraete, PRM 5, 024004 (2021)]

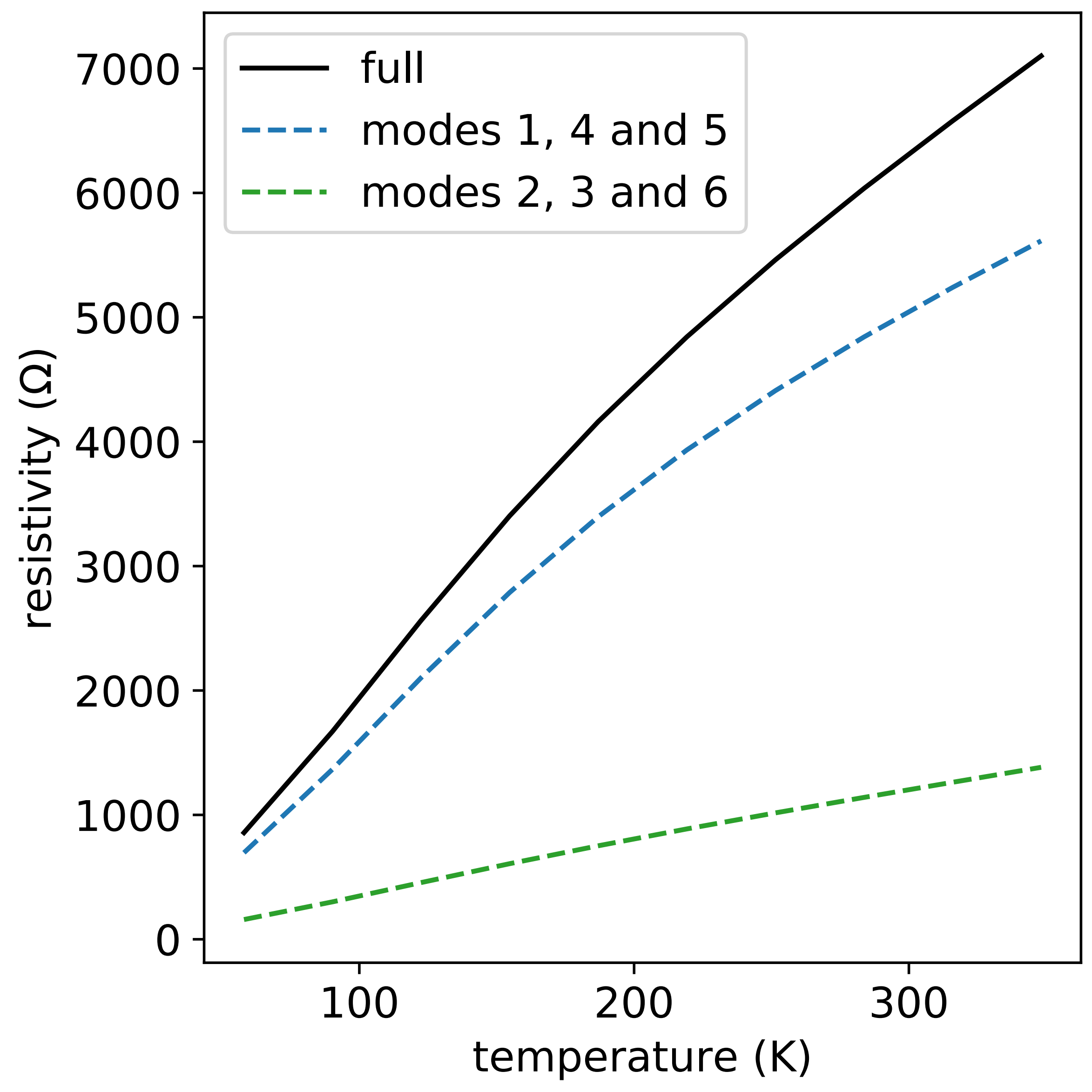
Valley-engineering

Arsenene: the role of intervalley scattering

Initial state







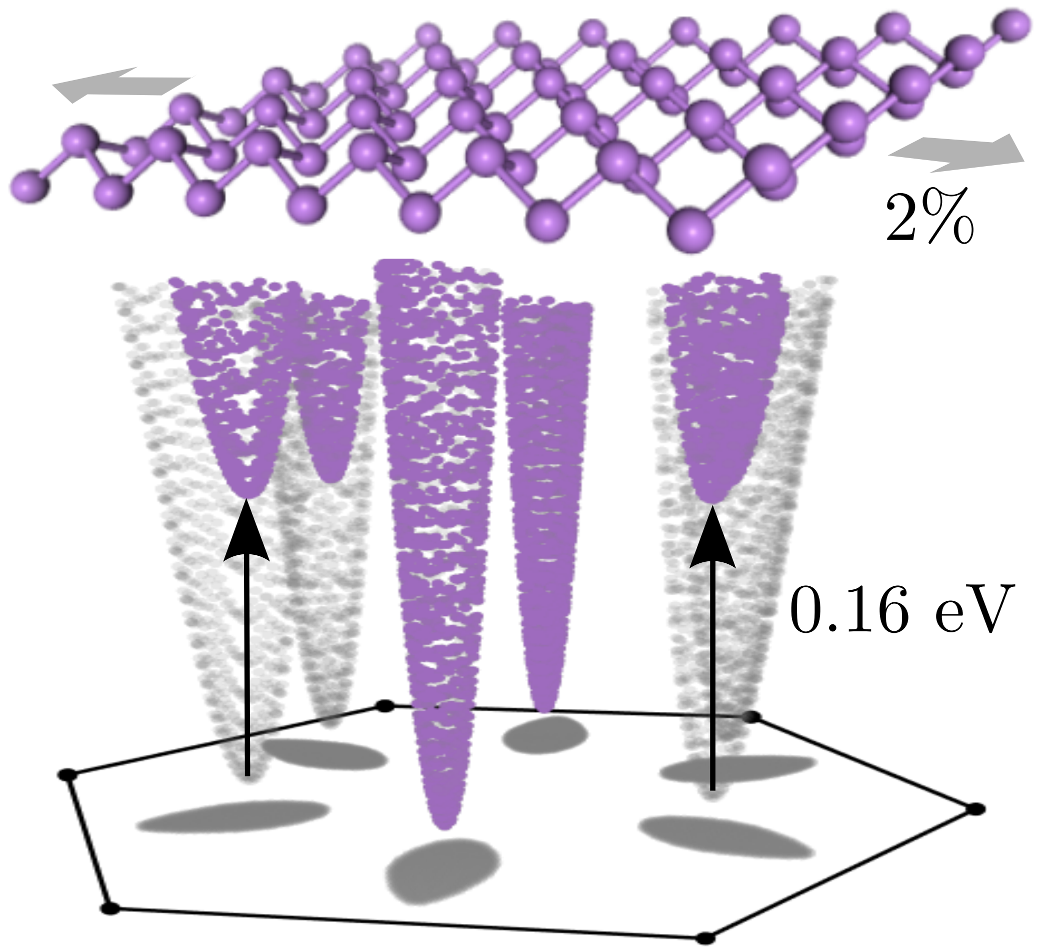
Transport dominated by intervalley scattering

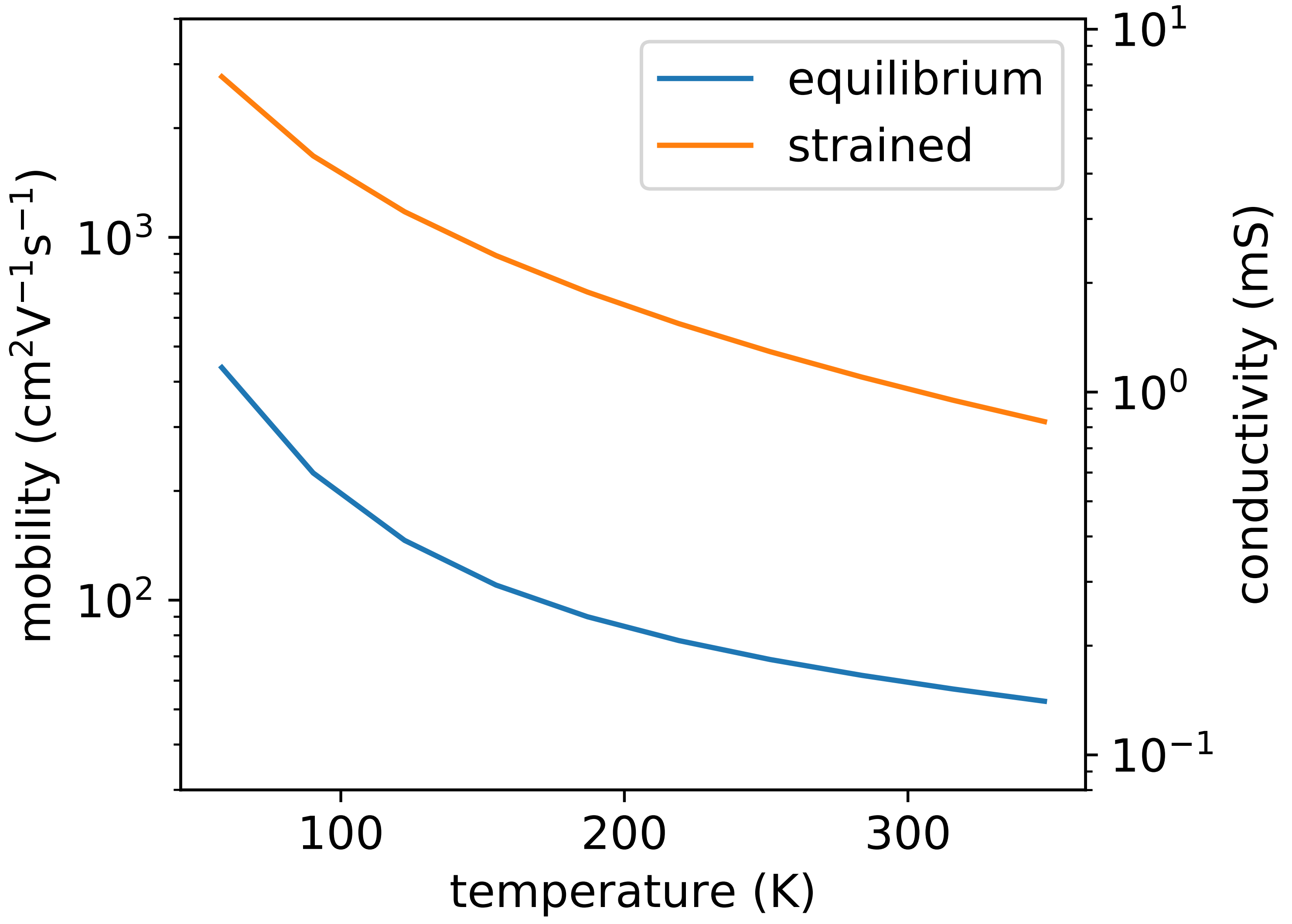
[T. Sohier, M. Gibertini, D. Campi, G. Pizzi, N. Marzari, Nano Letters 19, 3723 (2019)]

Valley-engineering

Mobility enhancement

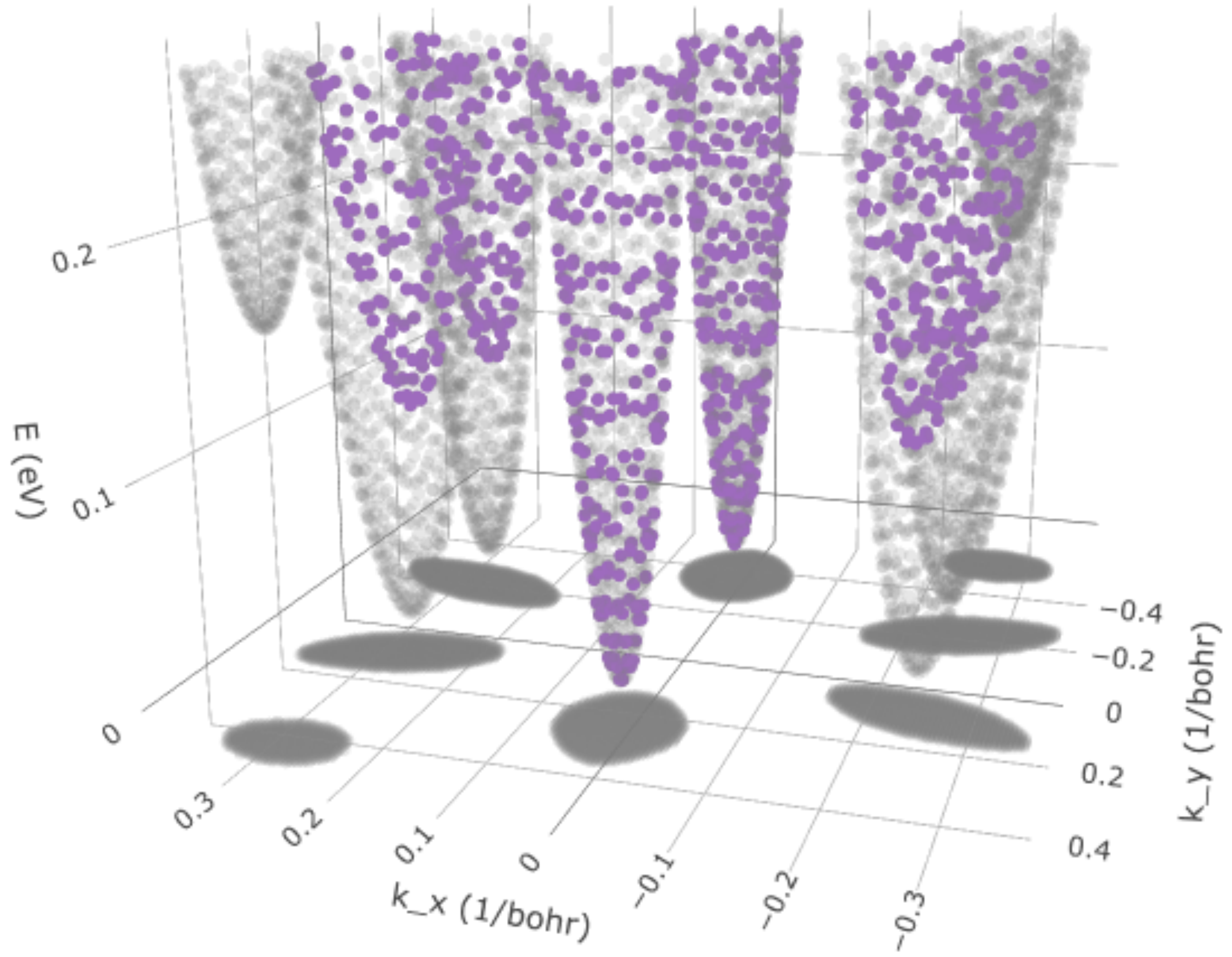
- Strain shifts the valleys out of reach, improving mobility by a factor 6.



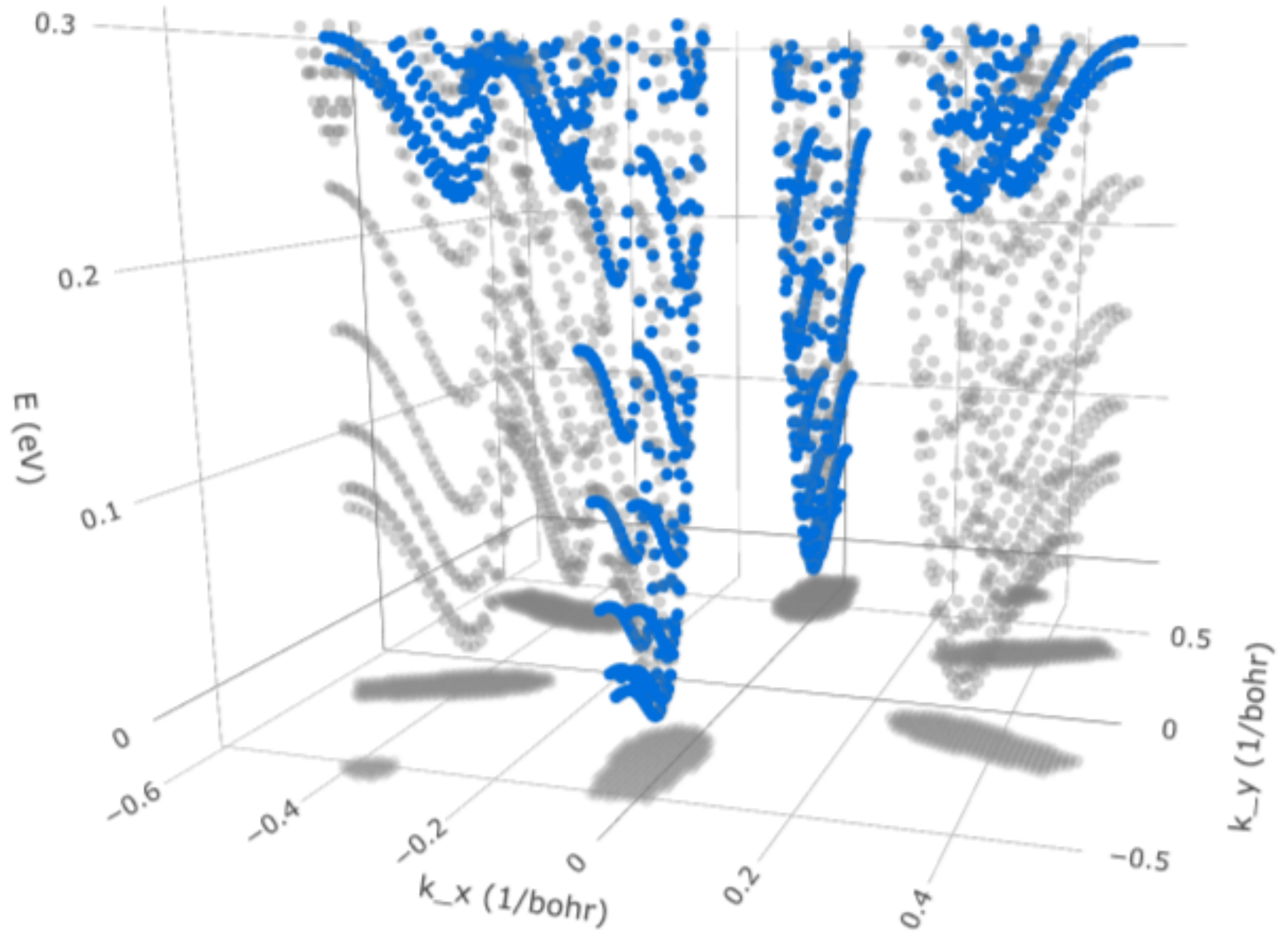


- Works for group V buckled monolayers, and others…

Antimonene



Blue Phosphorene



[T. Sohier, M. Gibertini, D. Campi, G. Pizzi, N. Marzari, Nano Letters 19, 3723 (2019)]

Perspectives for ML

- Descriptors:
  - Electronic structure features: number of valleys, Fermi velocity, DOS, etc…
  - Electron-phonon interactions: Intra- or intervalley, screenable or not, …
- Databases:
  - Need more data!

@ EPFL:
- N. Marzari
- M. Gibertini
- D. Campi
- G. Pizzi

@ ULiège
- M. Verstraete