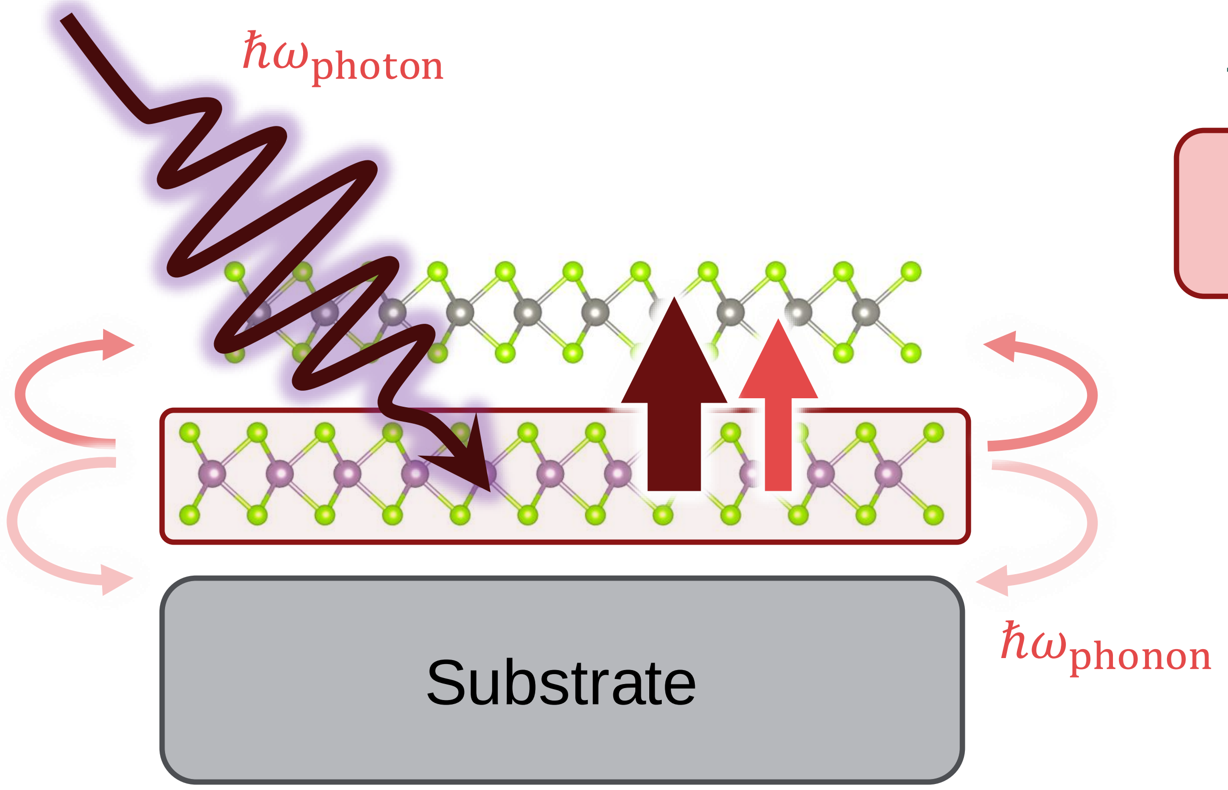
# Anharmonic Enhancement of Interfacial Phonon-Phonon Coupling in Twisted TMD Bilayers

JOHNATHAN D. GEORGARAS & FELIPE H. DA JORNADA

Electrons, Phonons, Electron-Phonon Scattering, and Phononics II

Room: 205D 3/4/2024, 3:00 PM – 6:00 PM

# Heat Management in Layered Transition Metal Dichalcogenide (TMD) Devices




## Heat Transfer Processes

*Timescale*

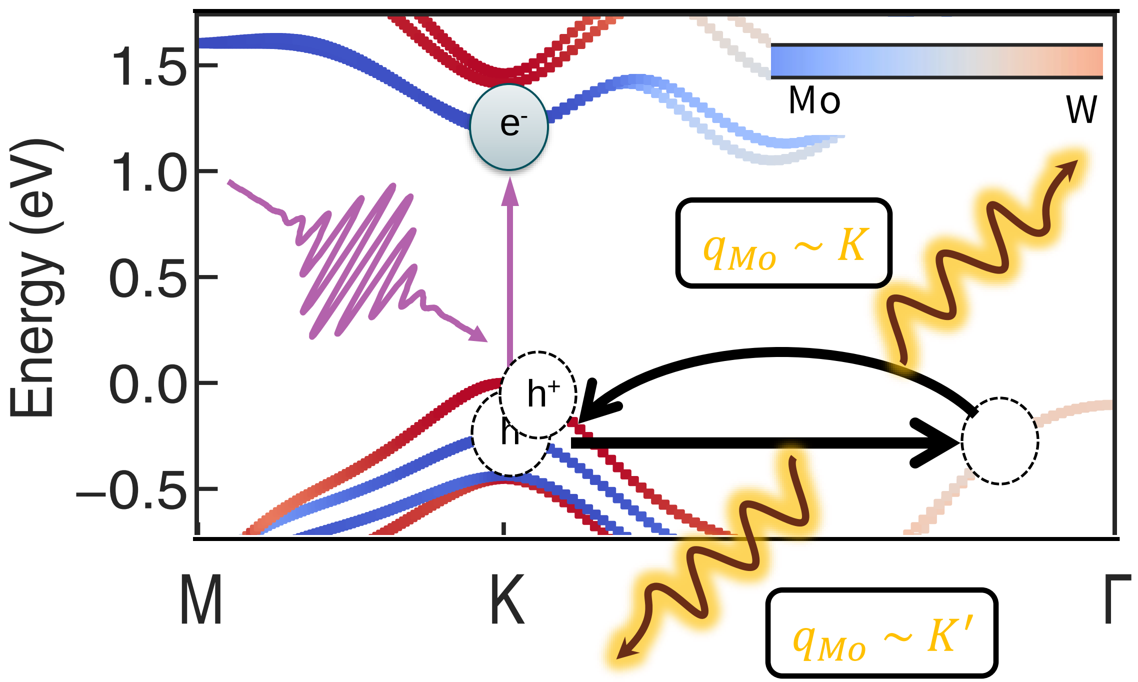
| | Timescale |
|---|---|
| Thermalization with substrate | ~1 ns |
| Interfacial thermalized conductance [1] | ~200 ps |
| "Hidden" interlayer phonon - phonon coupling [2] | ~20 ps |
| *Layer engineering ?* [3] | ? ps |

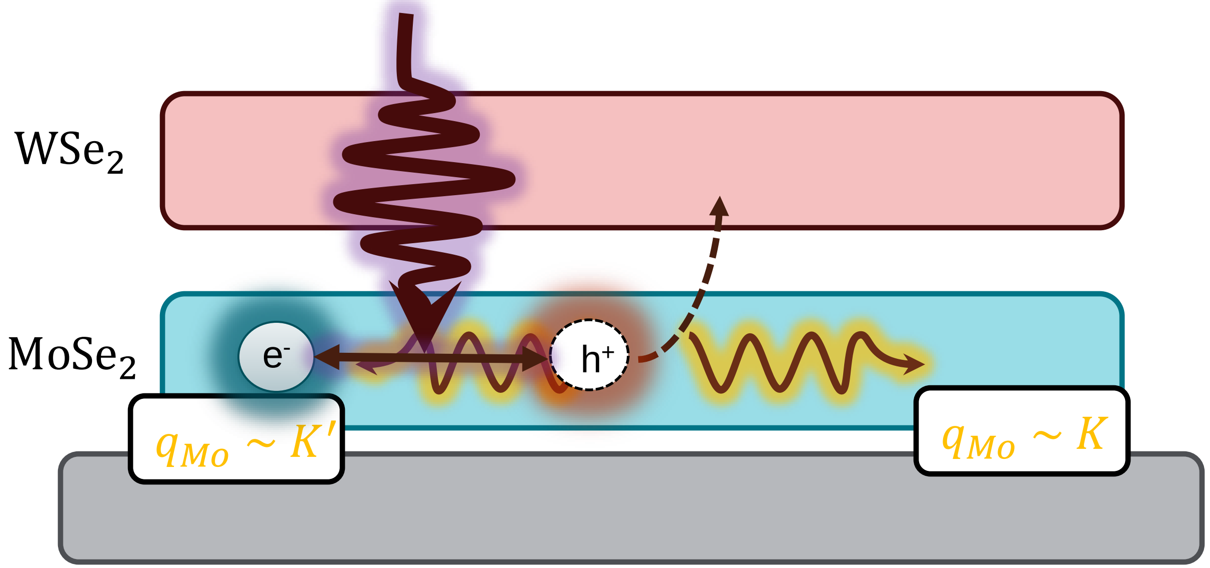

[1] Sood, A., Haber, J. B., Carlström, J., Peterson, E. A., Barre, E., **Georgaras, J. D.**, Reid, A. H., Shen, X., Zajac, M. E., Regan, E. C., Yang, J., Taniguchi, T., Watanabe, K., Wang, F., Wang, X., Neaton, J. B., Heinz, T. F., Lindenberg, A. M., da Jornada, F. H., & Raja, A. "Bidirectional Phonon Emission…" Nature Nanotechnology, **18,** 29. (2023).

[2] Johnson, A.*, **Georgaras, J. D**.*, Shen, X., Sood, A., Zeng, H., Saunders, P. , Kim, H., Yao, H., Heinz, T. F., Lindenberg, A., da Jornada, F. H., Luo, D. and Liu, F. Science Advances **10** (2024).

[3] **Georgaras, J. D**. & da Jornada, F. H. In Preparation (2024)

# "Hidden" Phonons Highways:
## Distribution of Remaining Phonons after Quasi-particle Relaxation




Electron-phonon scattering produces a ***non-thermal, highly polarized distribution*** of near-K wavevector phonons [1]



[1] Johnson, A.*, Georgaras, J. D.*, Shen, X., Sood, A., Zeng, H., Saunders, P. , Kim, H., Yao, H., Heinz, T. F., Lindenberg, A., da Jornada, F. H., Luo, D. and Liu, F. "Hidden phonon highways…", Science Advances **10** (2024).

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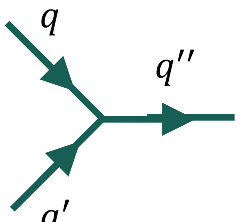
# “Hidden” Phonons Highways: Phonon-Phonon Interlayer Scattering Lifetimes from Perturbation Theory

*Scattering Rate $\Gamma(\omega)$ from Imaginary Part of Self-Energy*

$$\Gamma_q(\omega_q) = \frac{\pi}{2N}\sum_{q',q''} \frac{\hbar|\Phi_3(-q,q',q'')|^2}{8\omega_q\omega_{q'}\omega_{q''}} \boxed{\Delta(-q+q'+q'')}$$

$$\times \left[ \boxed{(n_{q'}+n_{q''}+1)\delta(\omega_q+\omega_{q'}-\omega_{q''})} + \boxed{2(n_{q'}-n_{q''})\delta(\omega_q-\omega_{q'}-\omega_{q''})} \right]$$

*Fusion*-like

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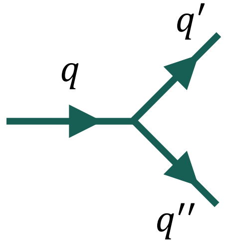


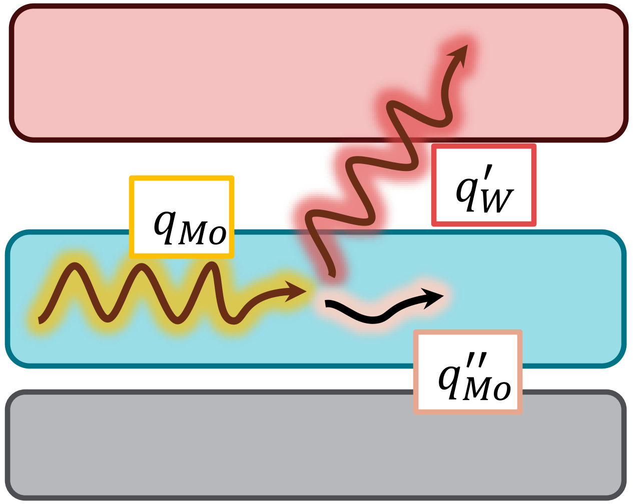

Explicitly calculate *phonon-phonon scattering* matrix elements $\Phi_3$

- Requires 3rd order force constant
- Expensive: $O(N^3)$ calculations, one for each triple atom set

Interlayer scattering: non-trivial to distinguish layer quality of phonon

- **Novel approach:** rotate from bilayer basis to monolayer basis $\Phi_3^{\mathrm{BL}} \rightarrow \Phi_3^{\mathrm{ML}}$

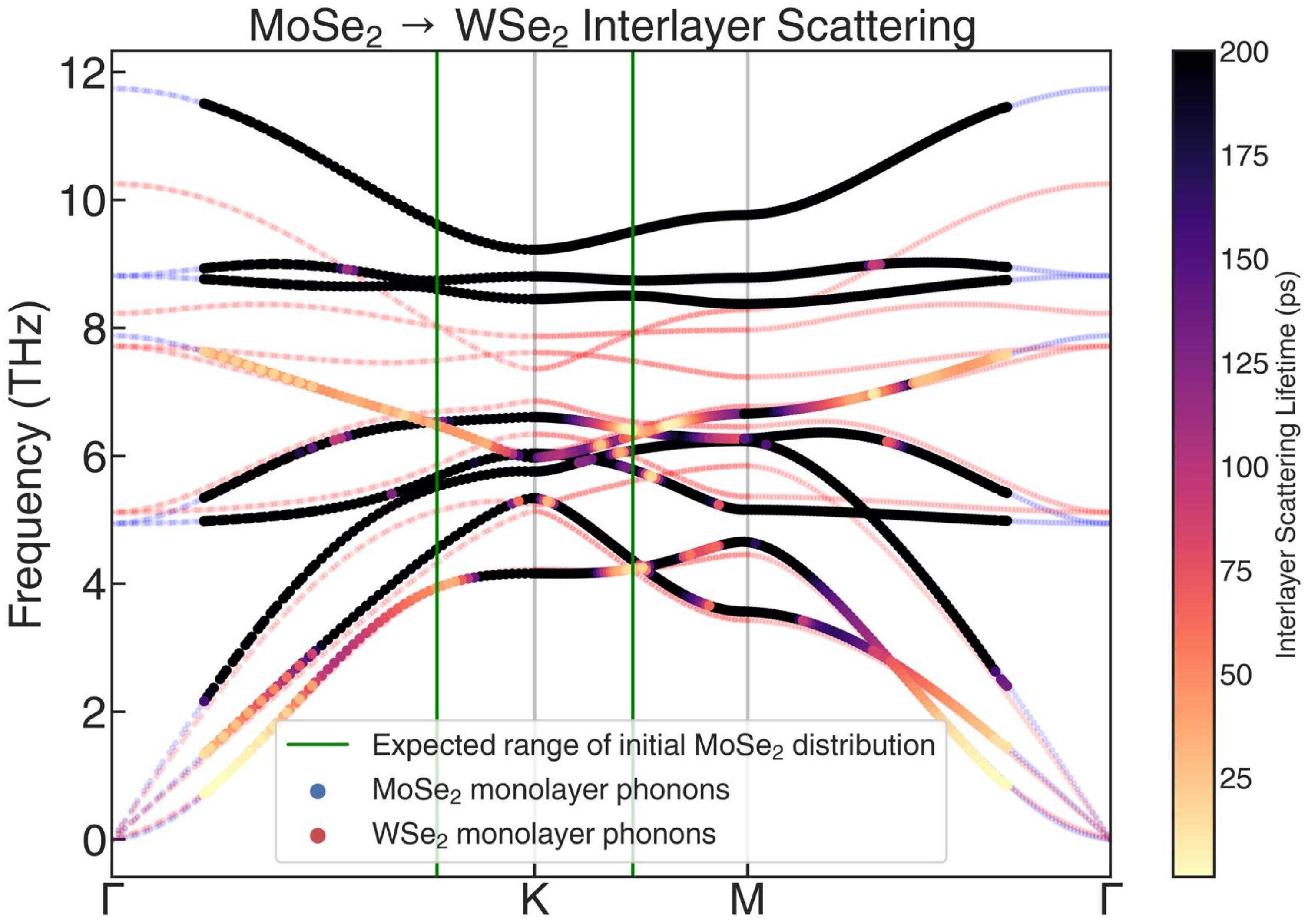
*Example Scattering Process*



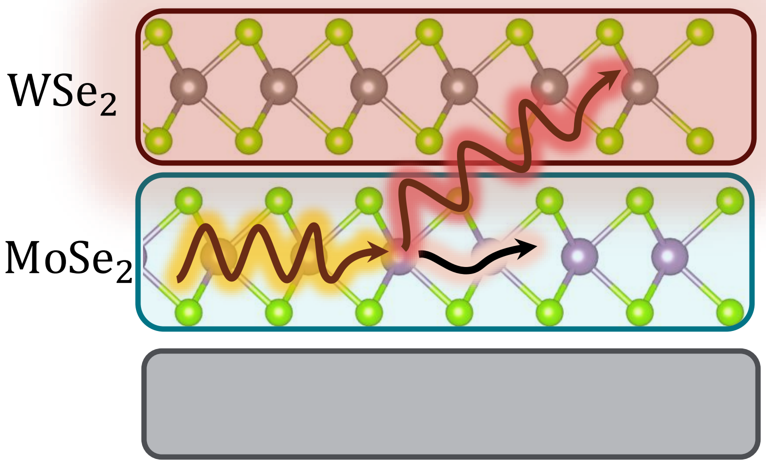

Initial phonon $q_{\mathrm{Mo}} \sim \mathrm{K}$ originating in Mo layer scattering to the W layer


[1] Johnson, A.*, Georgaras, J. D.*, Shen, X., Sood, A., Zeng, H., Saunders, P. , Kim, H., Yao, H., Heinz, T. F., Lindenberg, A., da Jornada, F. H., Luo, D. and Liu, F. “Hidden phonon highways…”, Science Advances **10** (2024).

# Phonon Interlayer Scattering Lifetimes in TMD Heterostructures




*Scattering from initial* $\mathrm{MoSe_2}$ *phonon* $q_{Mo} \sim K$*: primarily in* $\mathrm{WSe_2}$ *layer*




Interlayer non-thermal transfer $\sim\mathbf{20\ ps}$
**ORDER OF MAGNITUDE FASTER** than the thermalized distribution $\sim\mathbf{200\ ps}$



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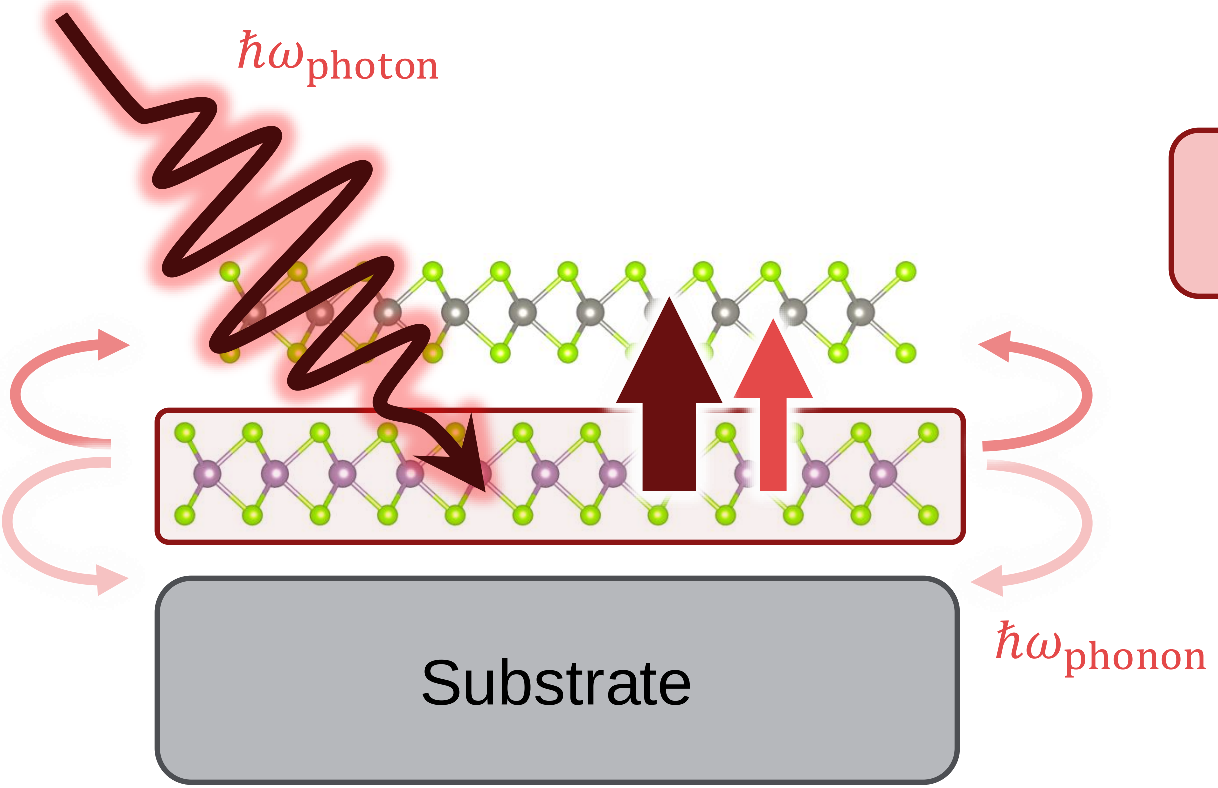


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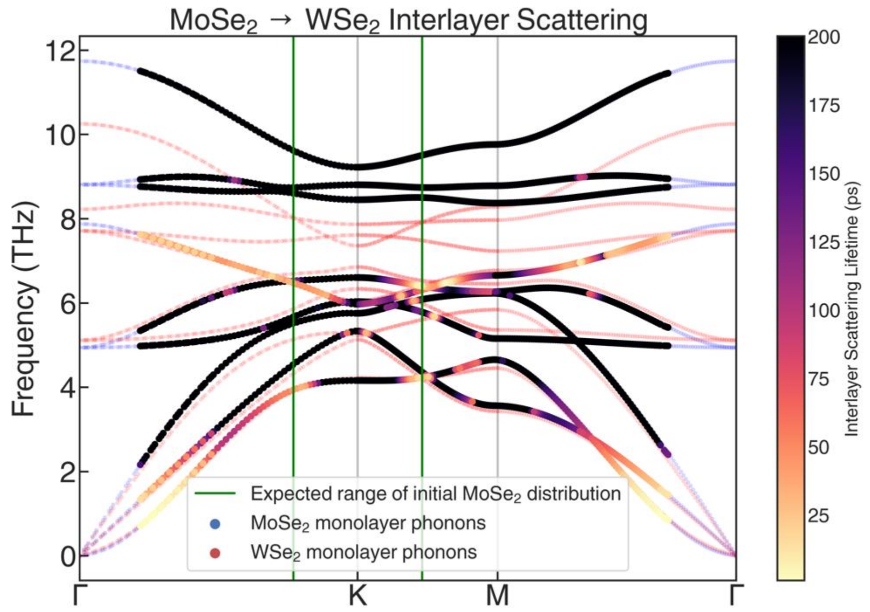
# Layer Selection for Interlayer Phonon – Phonon Coupling

*Useful Heuristic:*

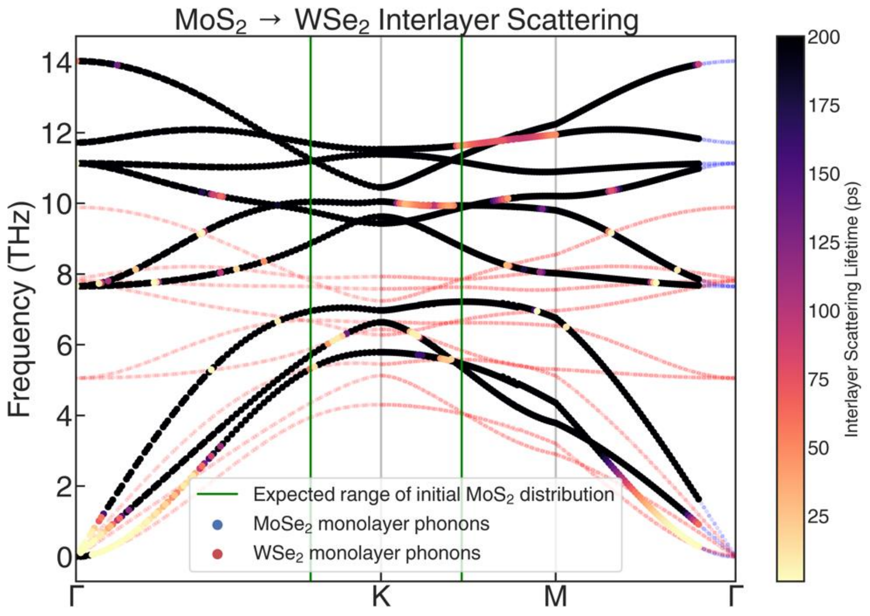
- **For a $K_{Mo} \rightarrow K_{W} + \Gamma_{Mo}$,** interlayer energy transfer is highest near band overlap due to a conservation of energy and momentum

**Beyond layer selection:**
can we further engineer and dynamically control band overlap to increase interlayer coupling?

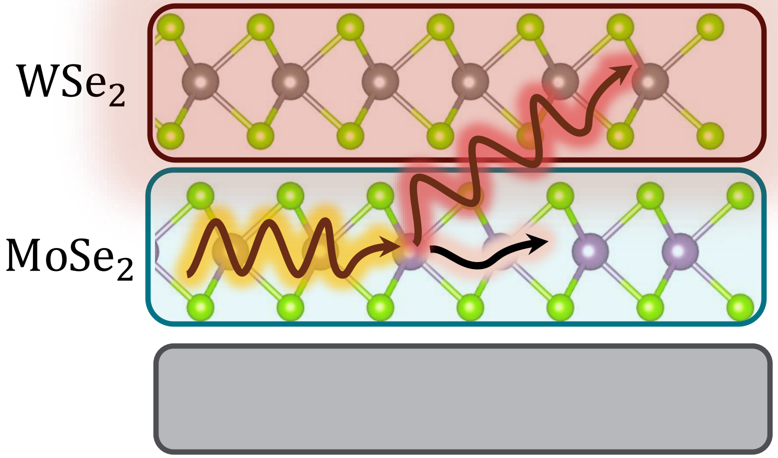
Homo-chalcogen




Hetero-chalcogen




*Scattering from initial phonon $q_{Mo} \sim K$: primarily in $\mathrm{WSe_2}$ layer*

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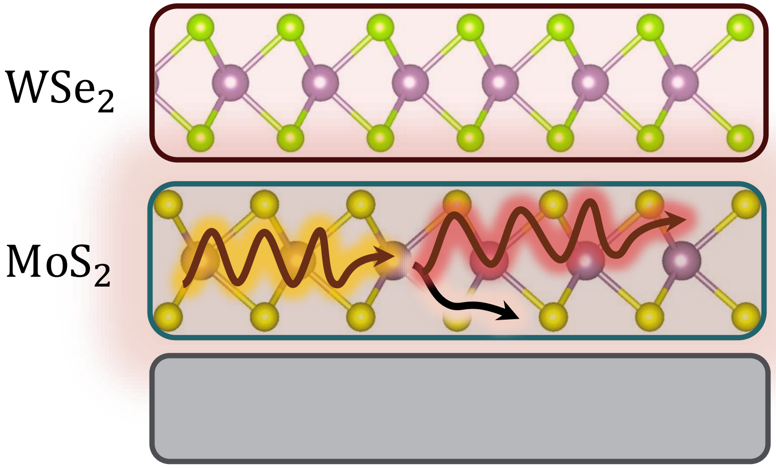
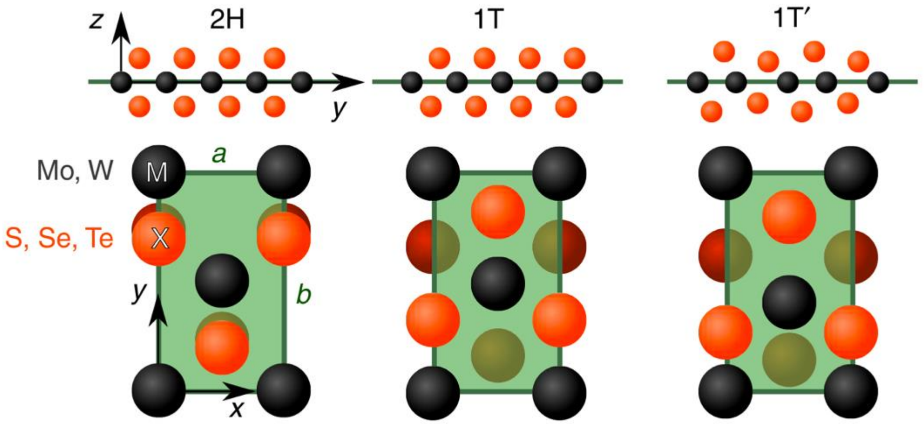
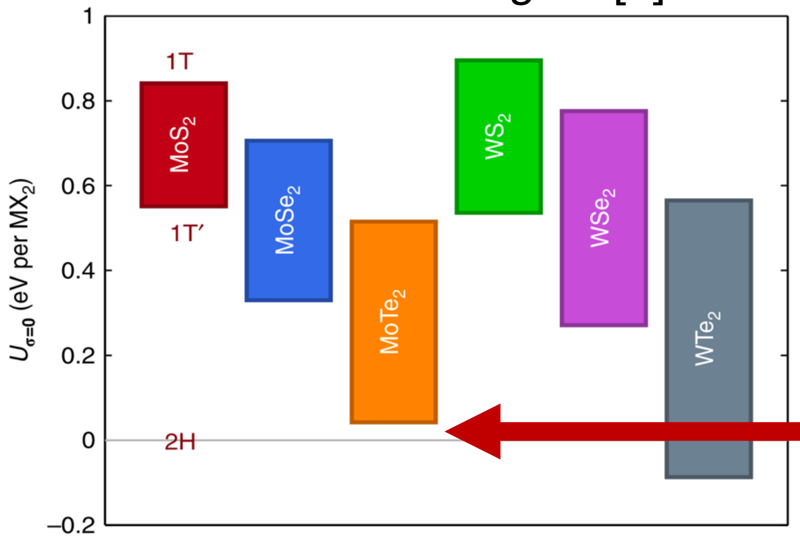


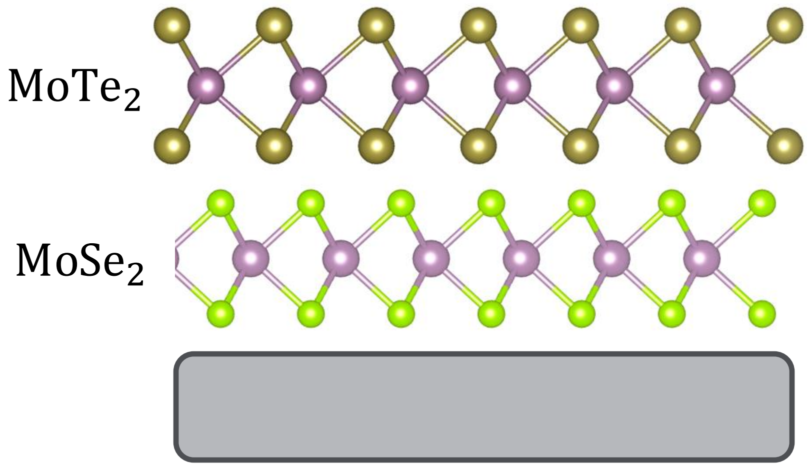
# Layer Engineering:
# Phase Transition in Transition Metal Dichalcogenides

*Three metastable phases of TMDs* [1]




*Relative Energies* [1]




Can we leverage increased anharmonicity from a phase change to **enhance interlayer phonon coupling?**



[1] Duerloo et al, Nature Comm. **7** (2014)

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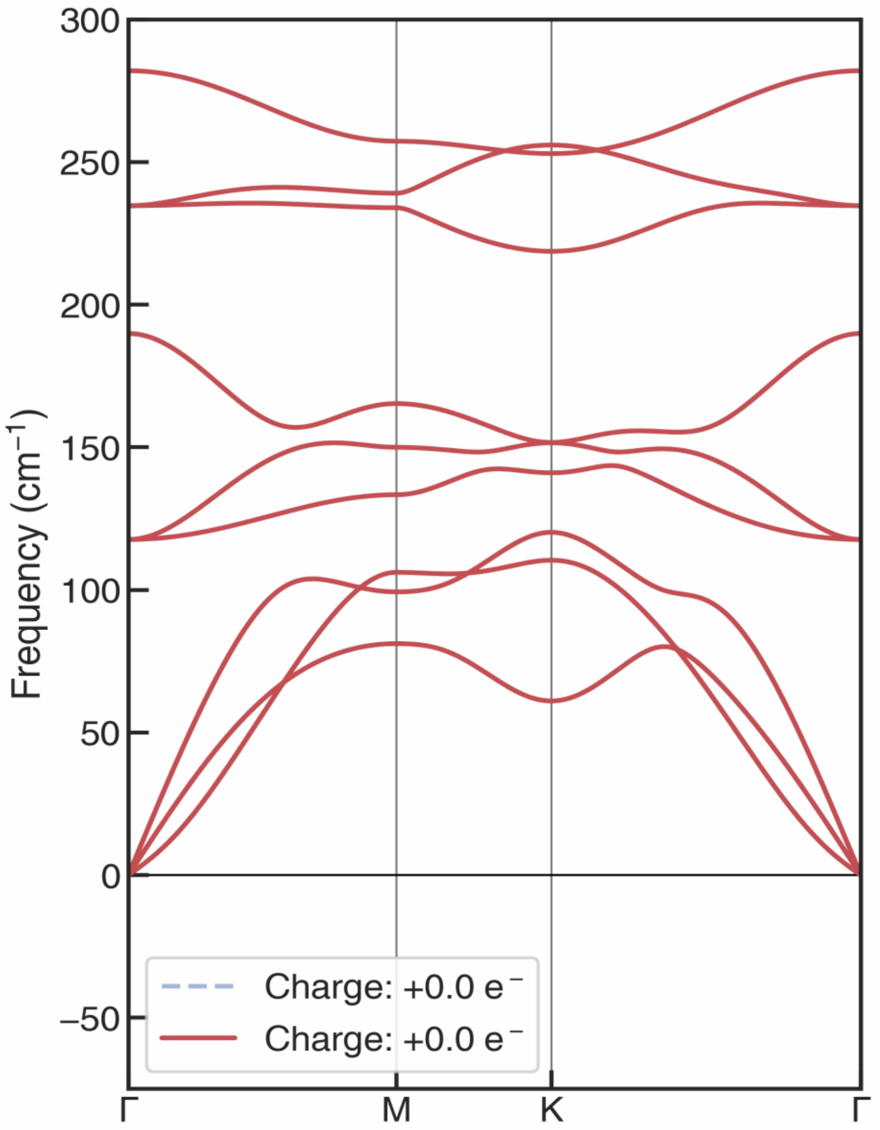
# Layer Engineering:
# Phase Transition in Monolayer $MoTe_2$ by Charge Doping

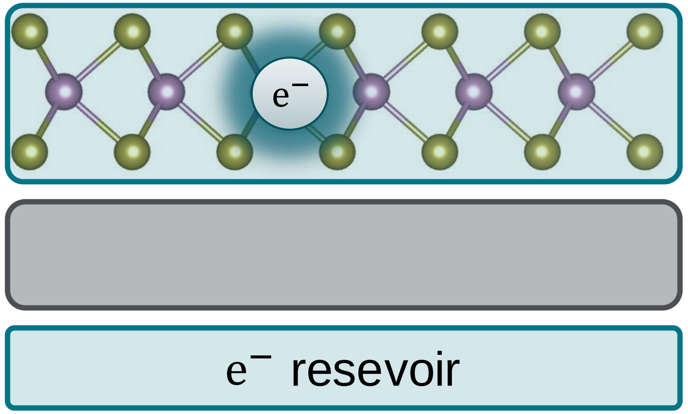
*Softening Phonon Modes in Monolayer* $MoTe_2$

- Method to induce 2H → 1T' phase change in $MoTe_2$:
  - **Electrostatic gating [1]**
  - Ionic liquid gating [2]
  - Photo-induced phase transition [3]

- **DFT:** *Ab initio* relaxation and forces for charged monolayer $MoTe_2$ to determine phonons by *finite displacement method*.




$MoTe_2$




Charge doping allows for dynamic control of phonon band structure and interatomic anharmonicity


[1] Li, Reed et al, Nature Comm. **7** (2016)
[2] Zakhidov et al, ACS Nano **14** (2020)
[3] Guan et al, PRL **128** (2022)

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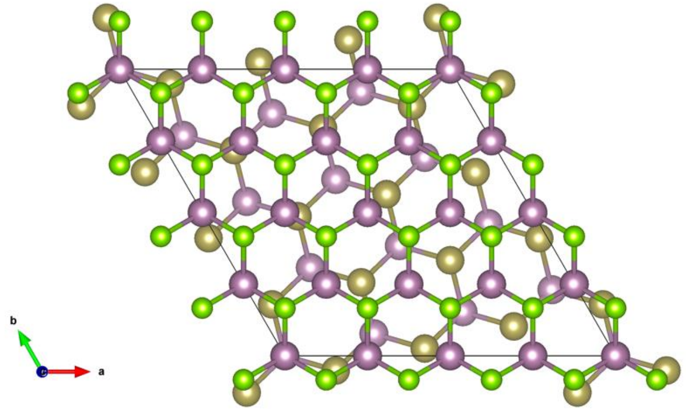
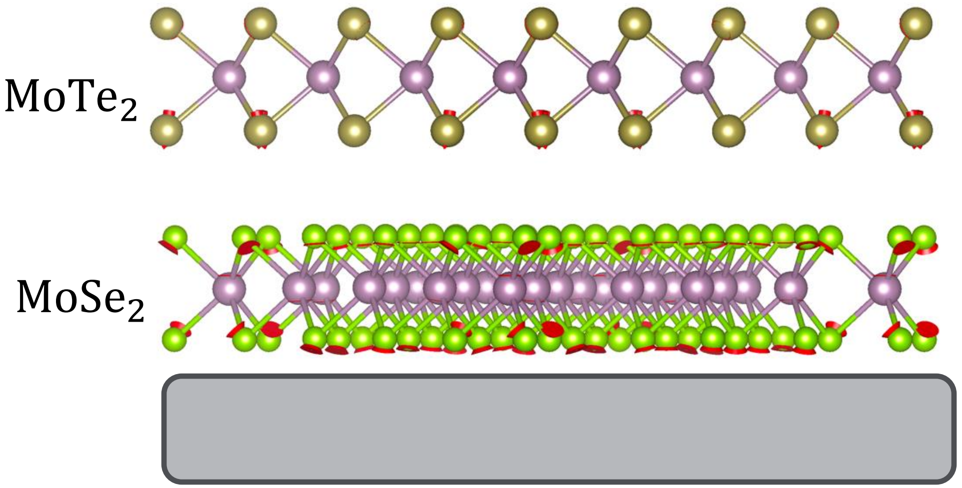
# Layer Engineering:
# Increasing Anharmonicity by Charge Doping in Twisted TMDs

*Twisted Bilayer* $\mathrm{MoTe_2/MoSe_2}$



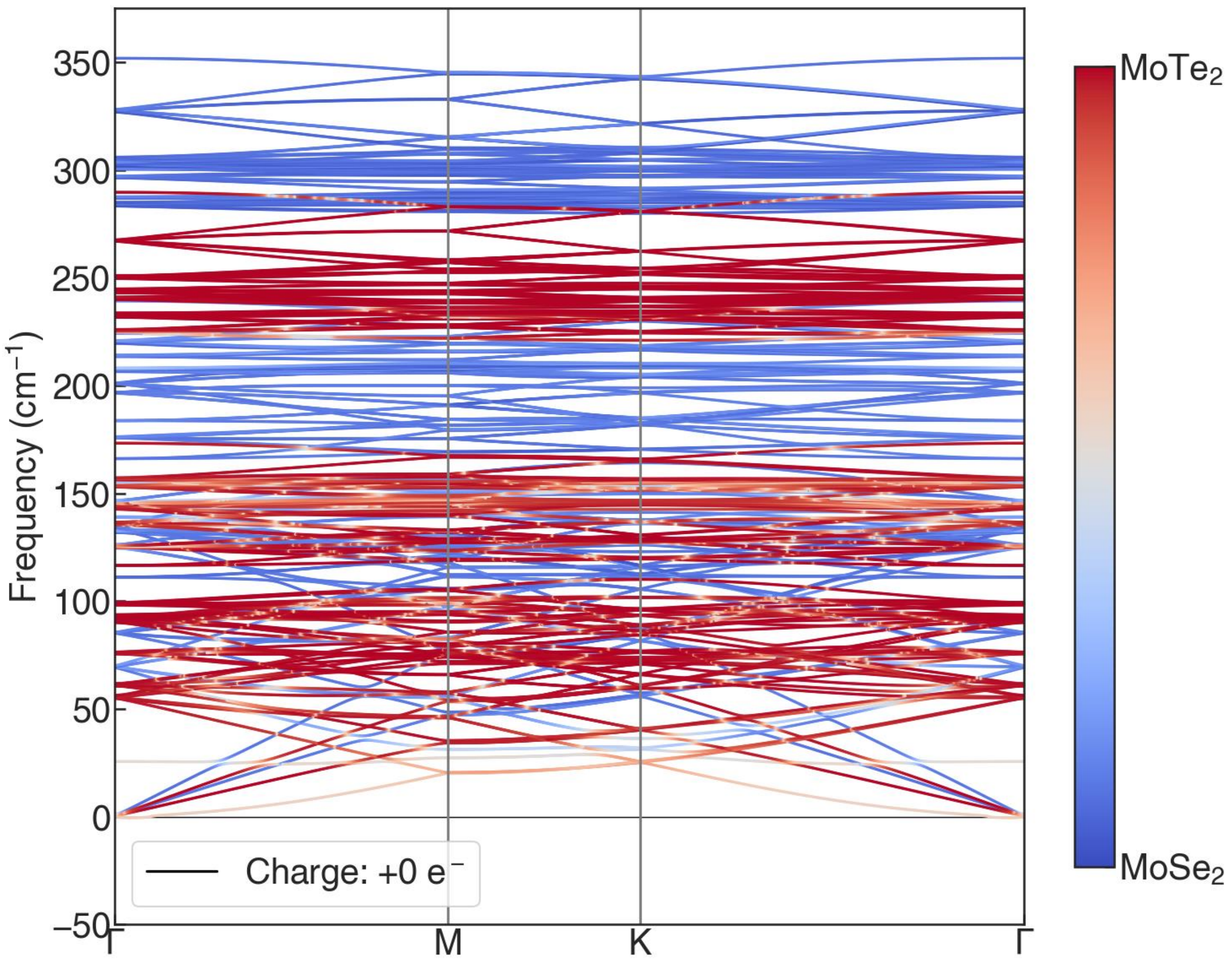



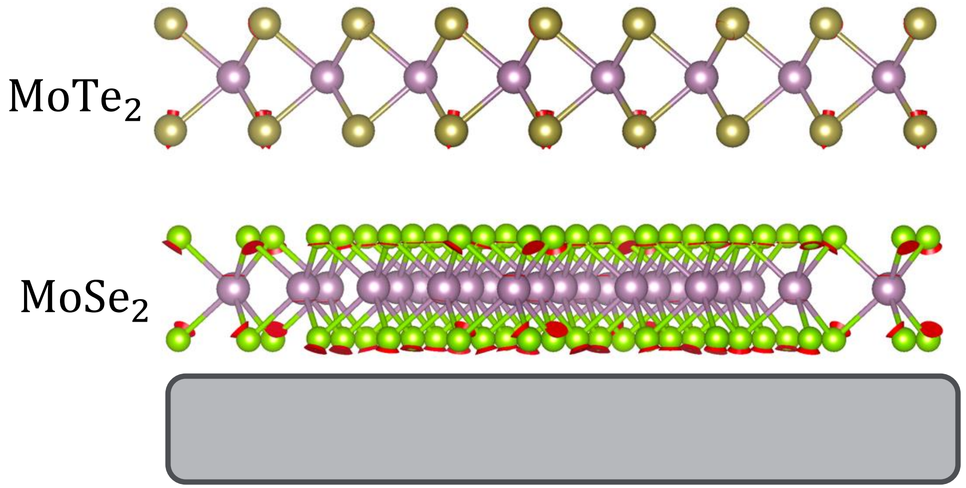



[1] Georgaras, J. D. & da Jornada, F. H. In Preparation (2024)

# Layer Engineering: Increasing Anharmonicity by Charge Doping in Twisted TMDs

*Softening Phonon Modes in* $MoTe_2/MoSe_2$

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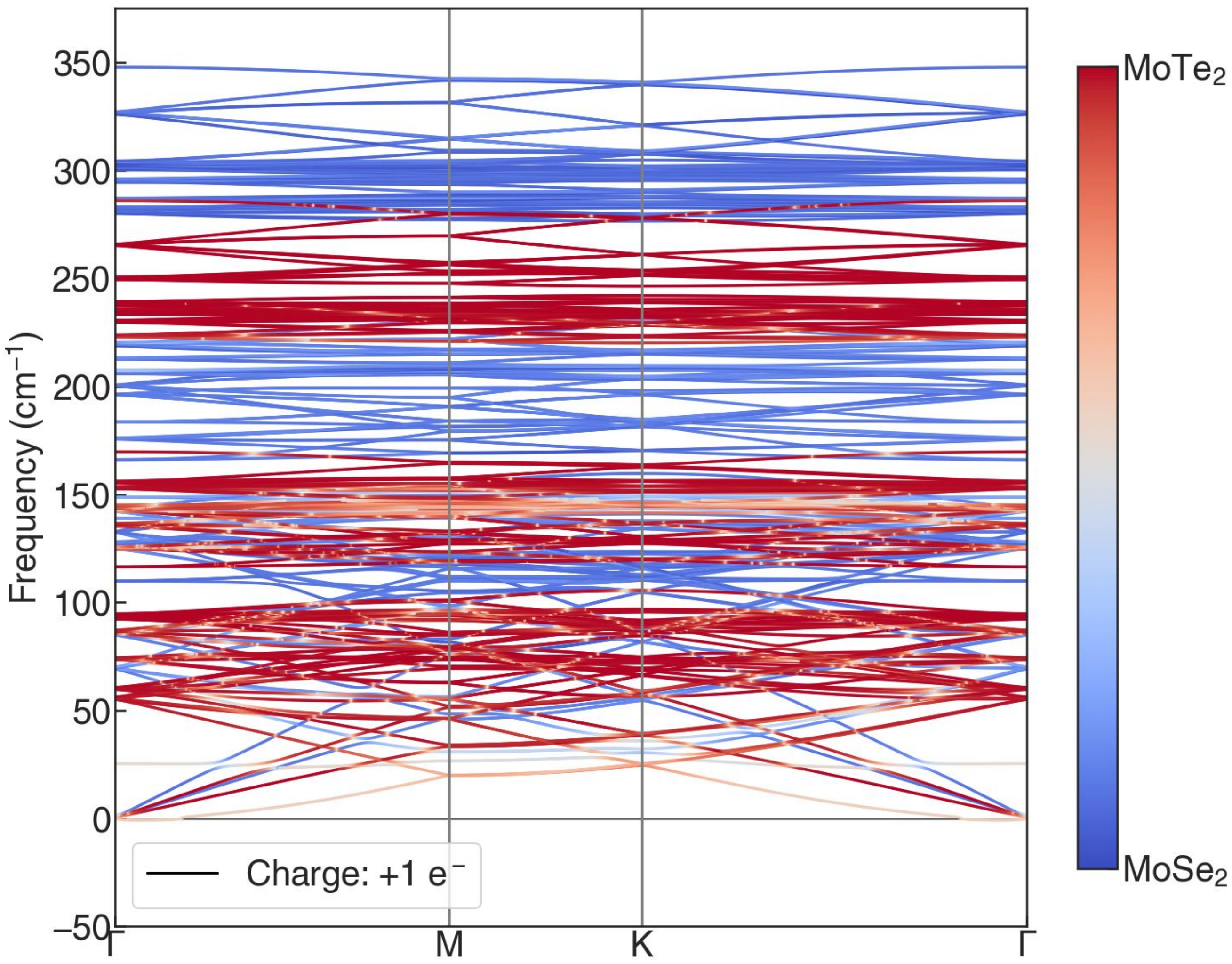


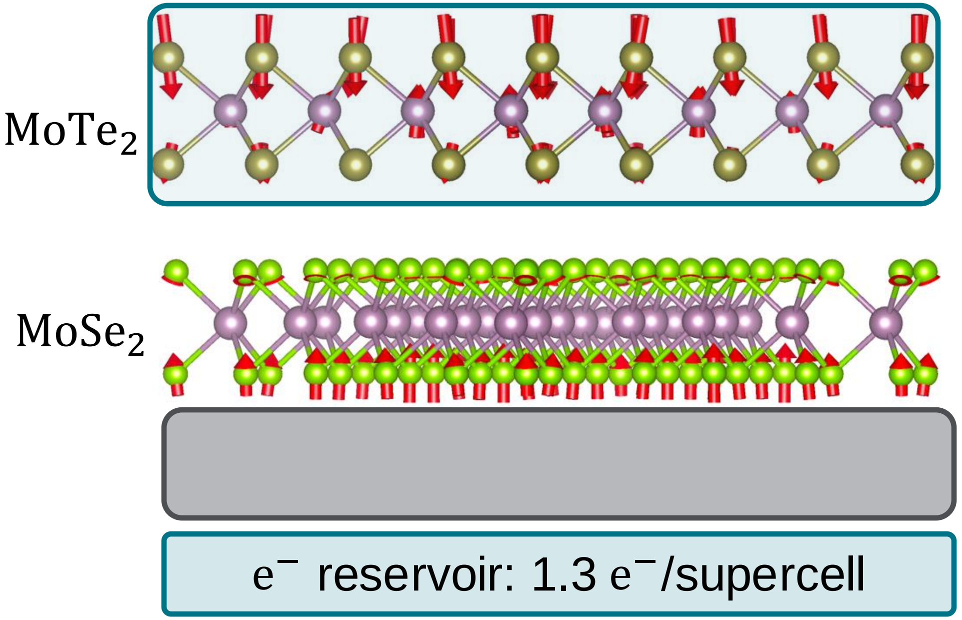


- Interatomic bonds weaken, increasing bond anharmonicity and allowing more favorable stacking



[1] Georgaras, J. D. & da Jornada, F. H. In Preparation (2024)

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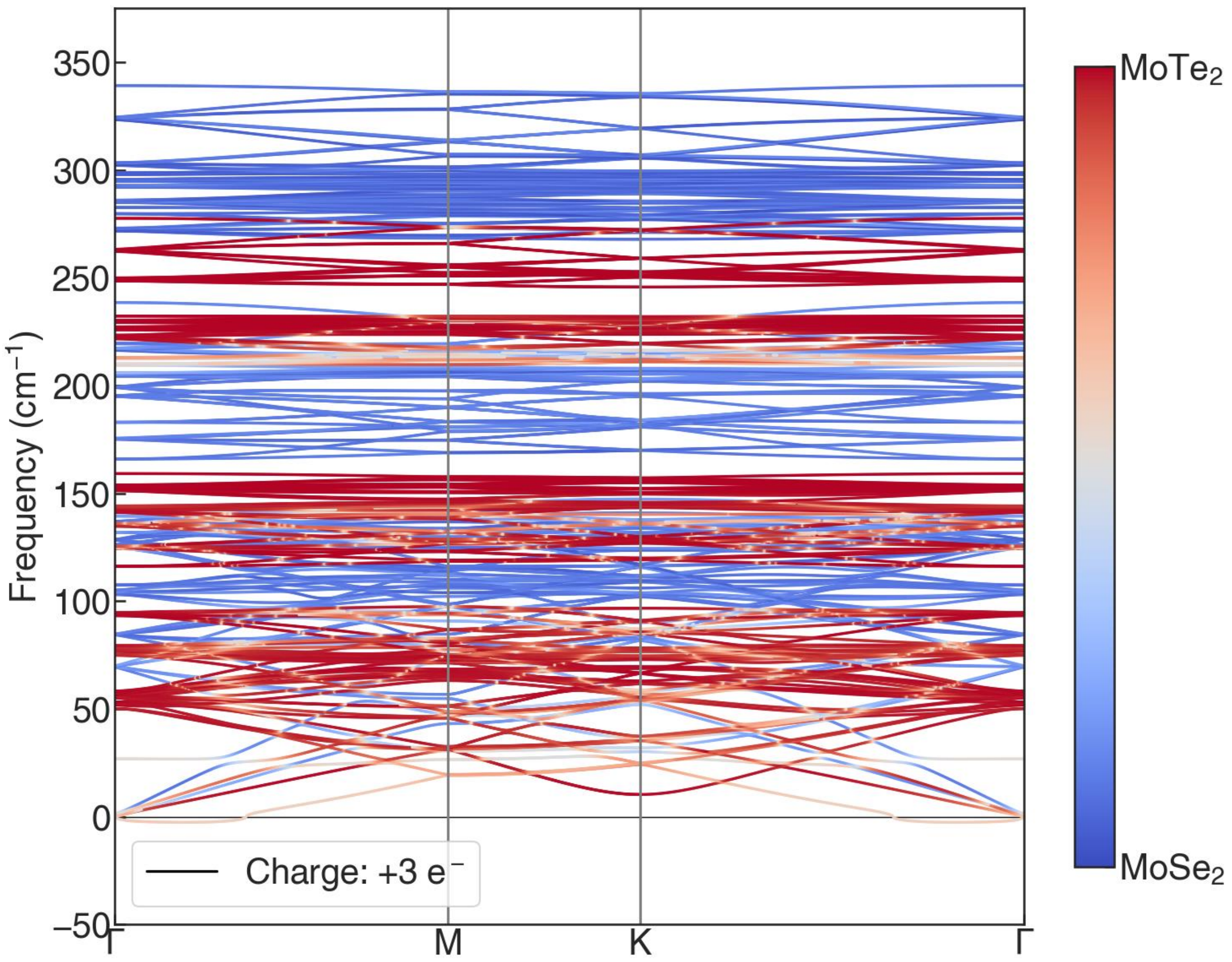


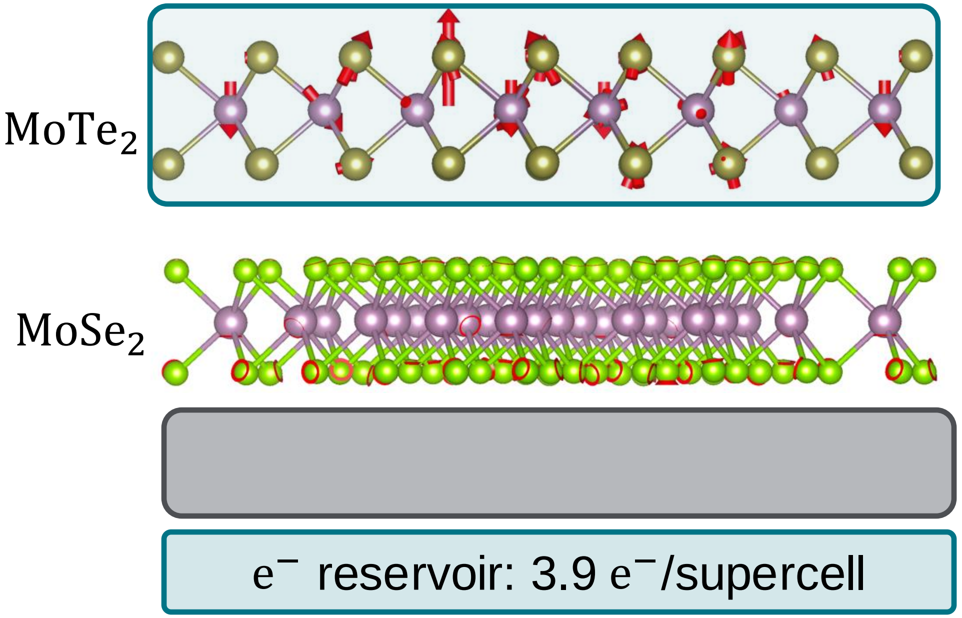


- Interatomic bonds weaken, increasing bond anharmonicity and allowing more favorable stacking
- Phonons in $MoTe_2$ layer soften as layer approaches phase transition



[1] Georgaras, J. D. & da Jornada, F. H. In Preparation (2024)

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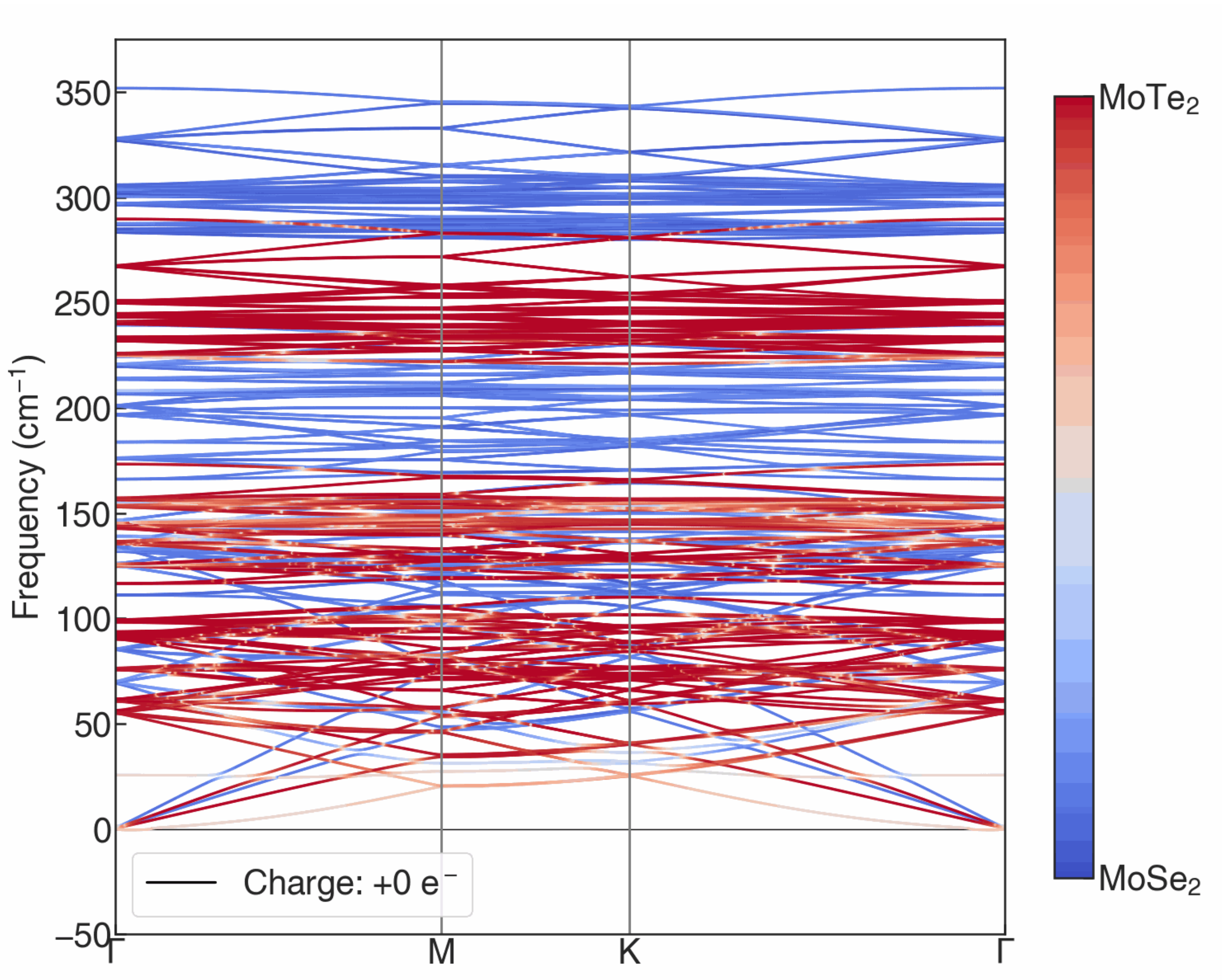


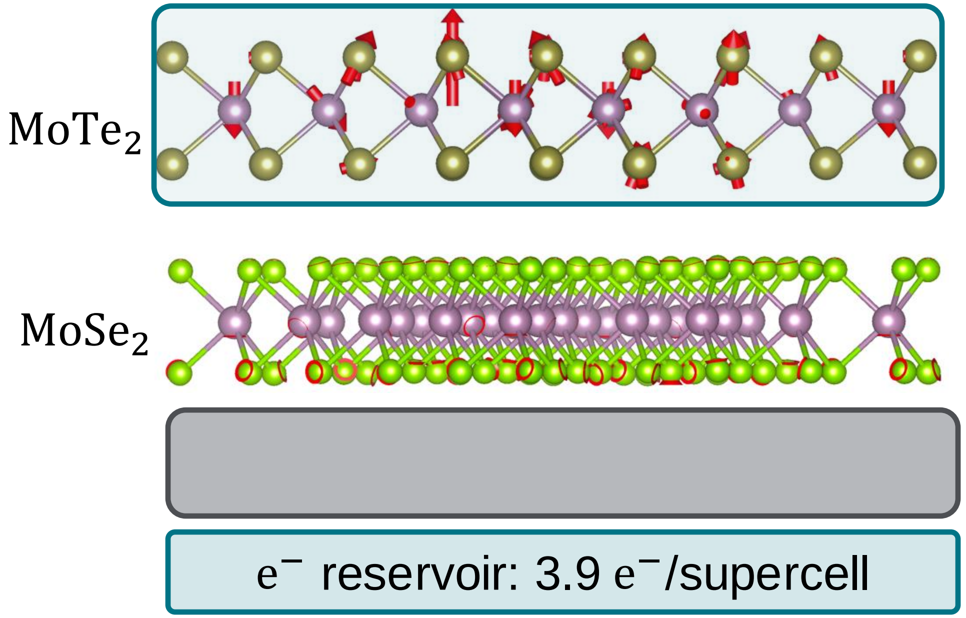


- A bonds weaken, increasing bond anharmonicity and allowing more favorable stacking
- Phonons in $MoTe_2$ layer soften as layer approaches phase transition
- **Greater overlap seen in lowest optical and acoustic branches as phonons frequency decreases**



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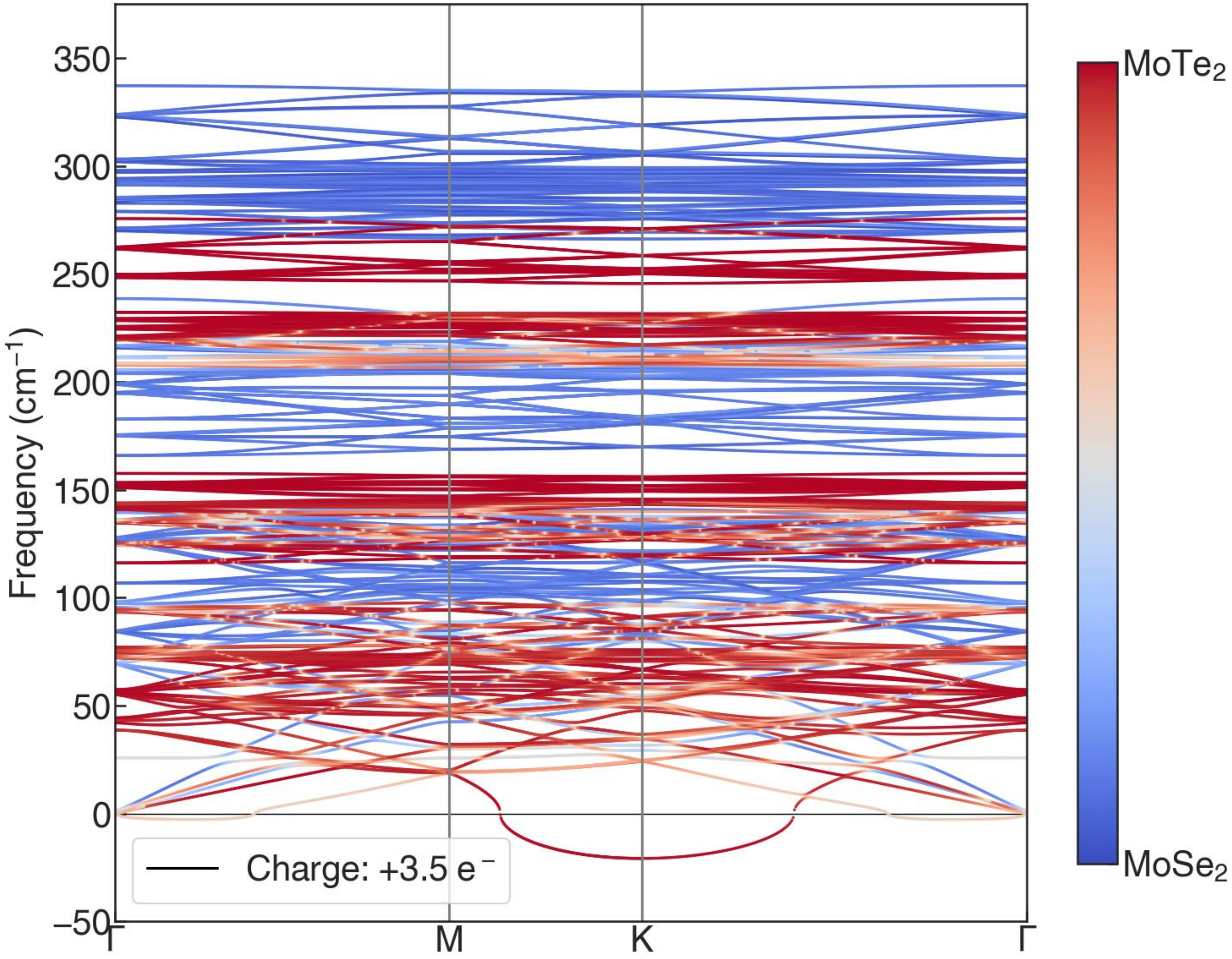



## *Looking Forward*

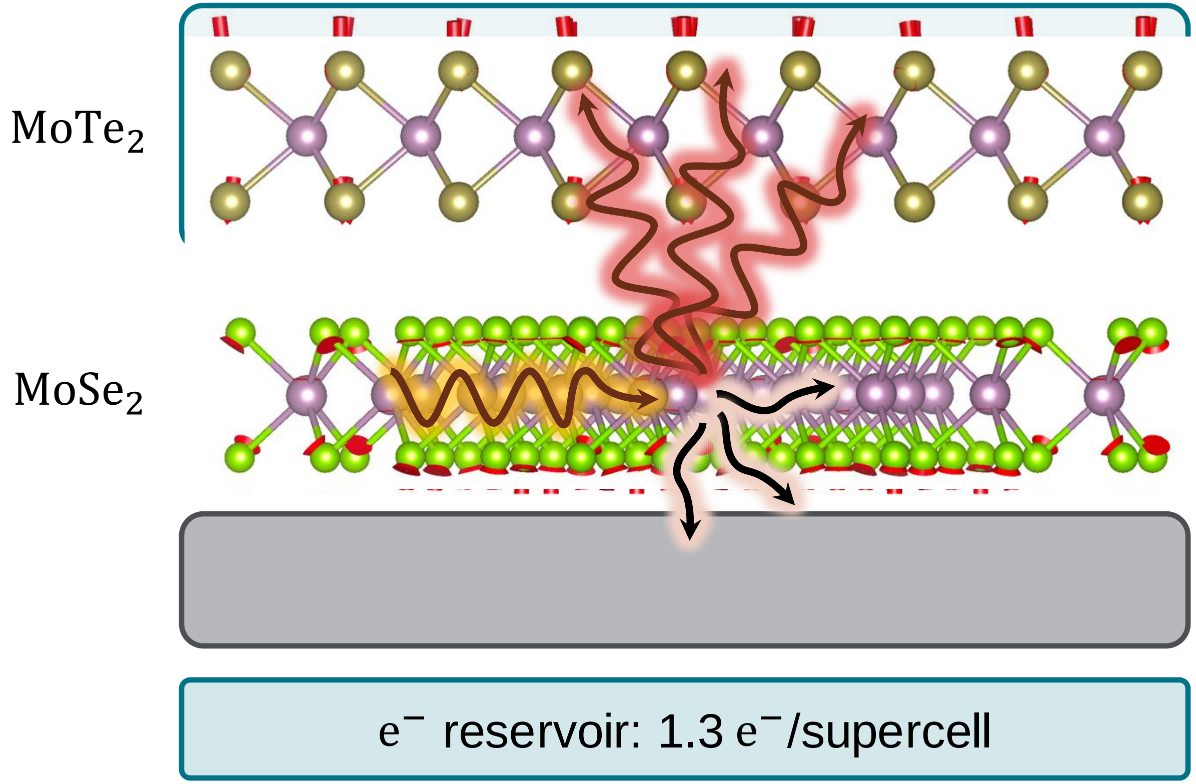
- Explicit calculation of **interlayer phonon scattering lifetimes** at different charge-doping.
- **Figure-of-merit** for band overlap and "inter-phase anharmonicity"
- Understanding local **moiré reconstruction** upon charge-doping



[1] Georgaras, J. D. & da Jornada, F. H. In Preparation (2024)

# Summary: Charge-Doping Tuned Interlayer Phonon-Phonon Coupling

1. Non-thermal distributions of phonons scatter heat in TMD heterostructures **an order of magnitude faster** than thermalized conduction [1]
2. Devised method to accurately calculate interlayer phonon-phonon scattering via basis rotation. [1]
3. Band overlap heuristic for enhancing interlayer 3-phonon processes involving layer-hybridized $q \sim \Gamma$ phonons. [2]
4. **Electrostatic charge-doping of $\mathbf{MoTe_2}$** shows dynamic control of resonant overlap to efficiently extract heat from adjacent layer. [2]





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[2] Georgaras, J. D. & da Jornada, F. H. In Preparation (2024)

# Acknowledgements and Thanks


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U.S. DEPARTMENT OF ENERGY | Office of Science

NERSC

FRONTERA
TACC | NSF | TEXAS

Natural Sciences and Engineering Research Council of Canada
Conseil de recherches en sciences naturelles et en génie du Canada

Canada

**Fang Liu** (Stanford)

Amalya Johnson

**Tony Heinz** (Stanford)

**Felipe Jornada** (Stanford)

**Aditya Sood** (Stanford)

**Aaron Lindenberg** (Stanford)

**Duan Luo** (SLAC)