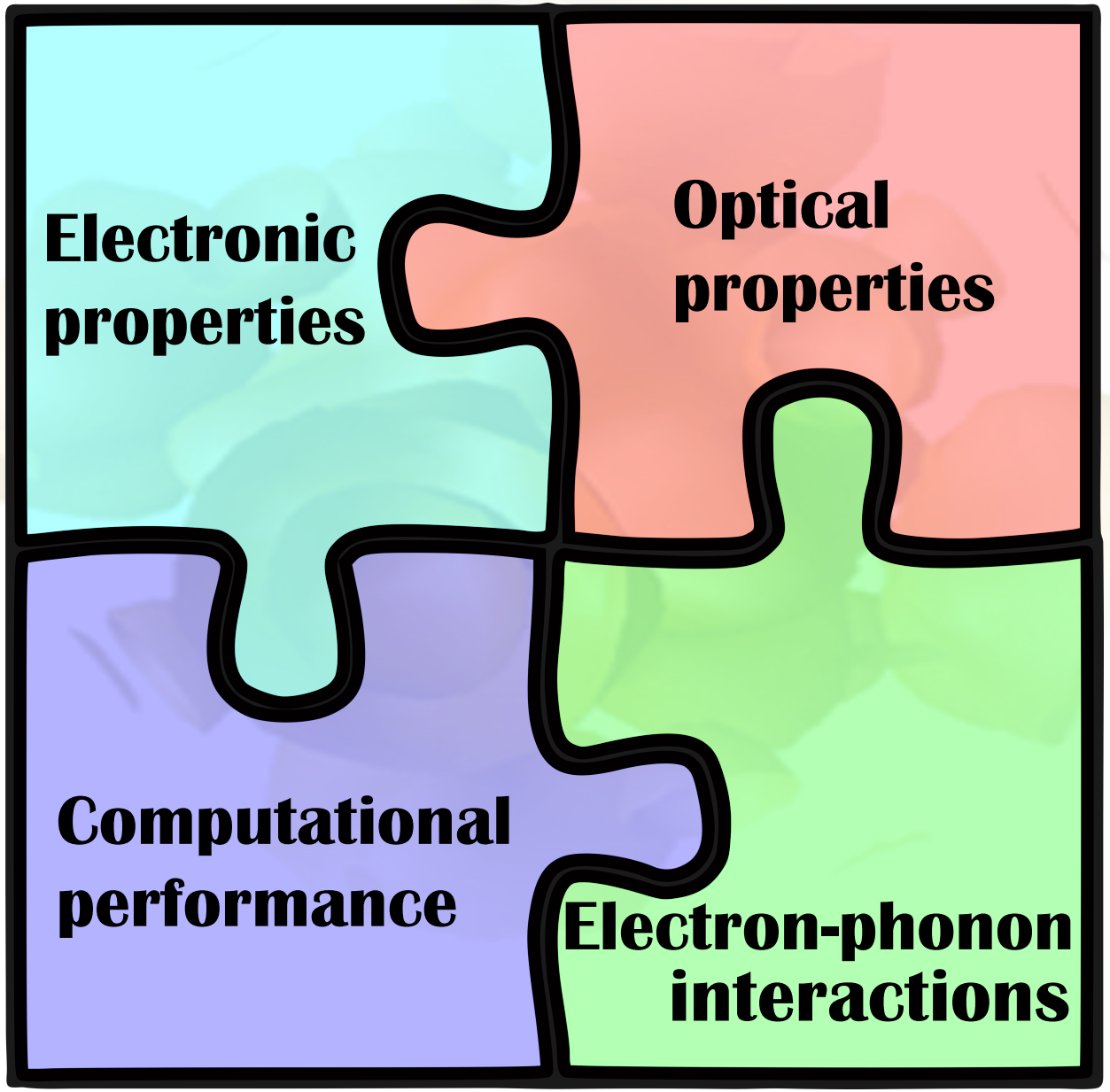
Hybrid excitations and electron-phonon interactions in pyrene@MoS_2

Ignacio Gonzalez Oliva, HU Berlin

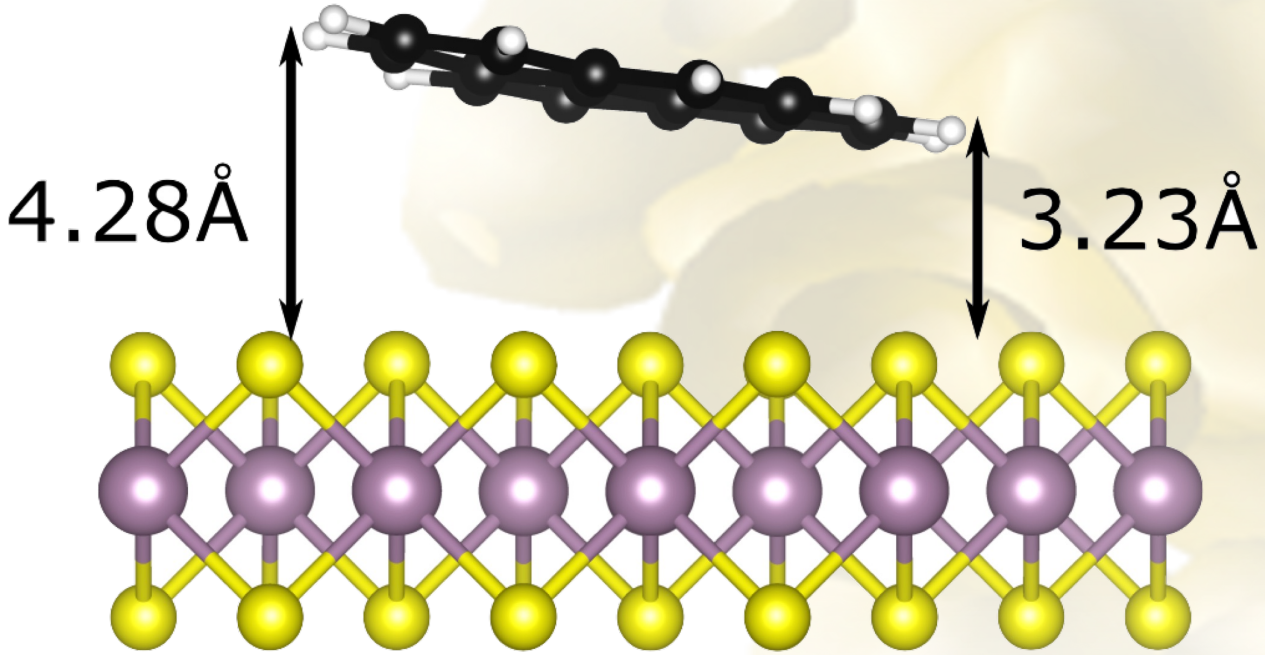
Content

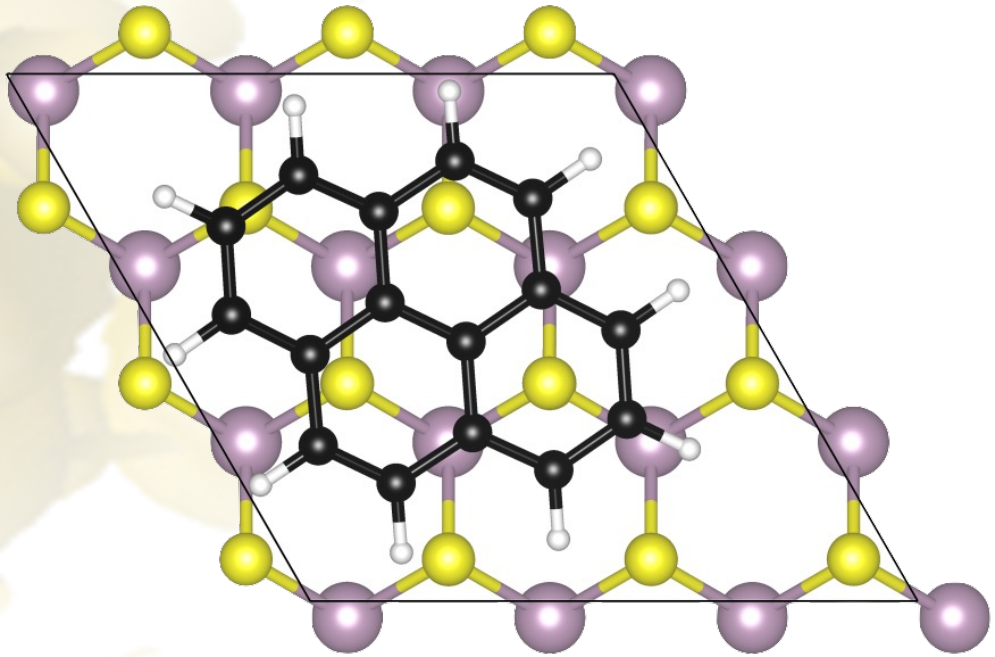

Electronic properties
Optical properties
Computational performance
Electron-phonon interactions


exciting

Electronic properties of pyrene@MoS_2

I. Gonzalez Oliva *et al.*, Phys. Rev. Materials. **6**, 054004 (2022).





Electronic properties of pyrene@MoS_2

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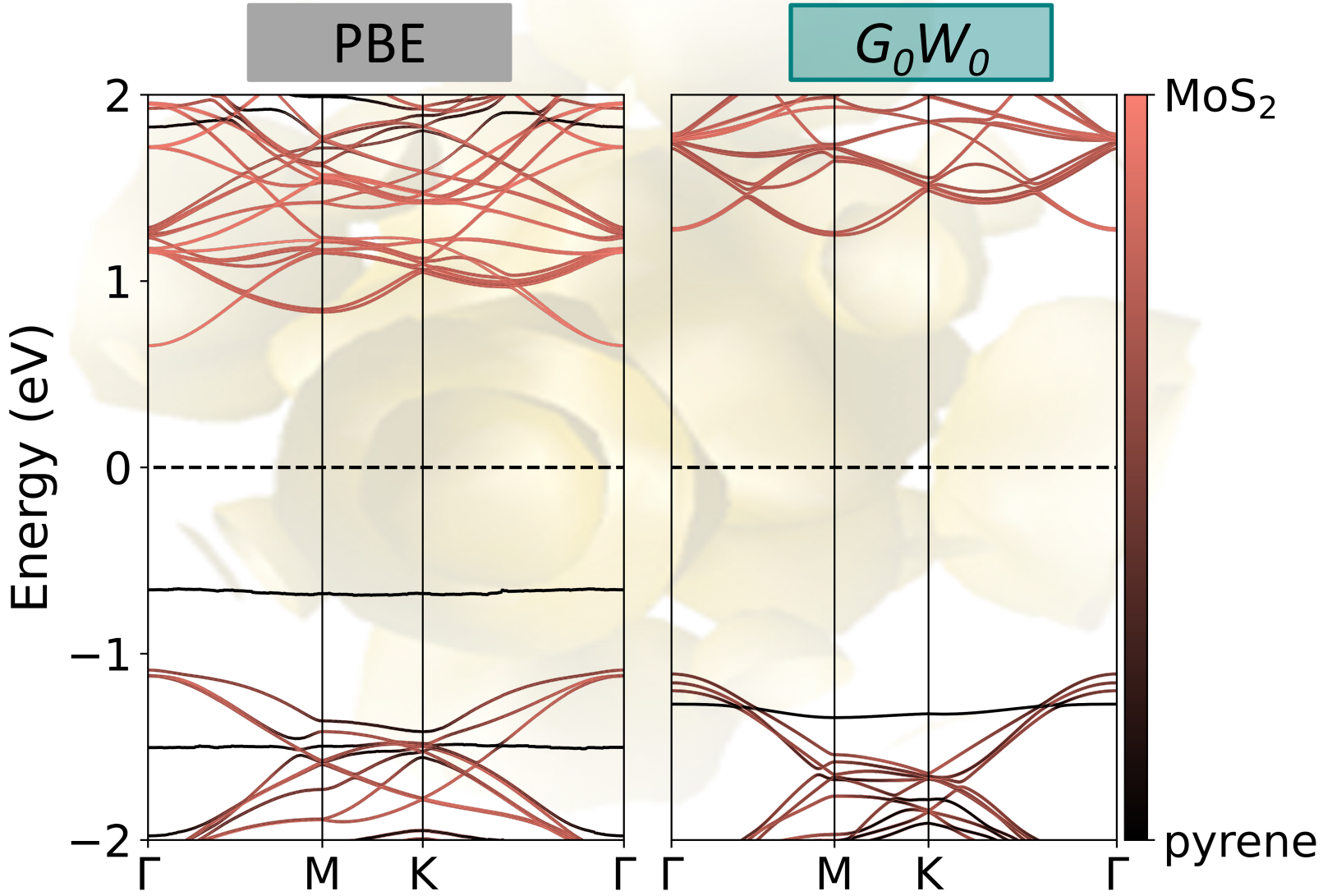
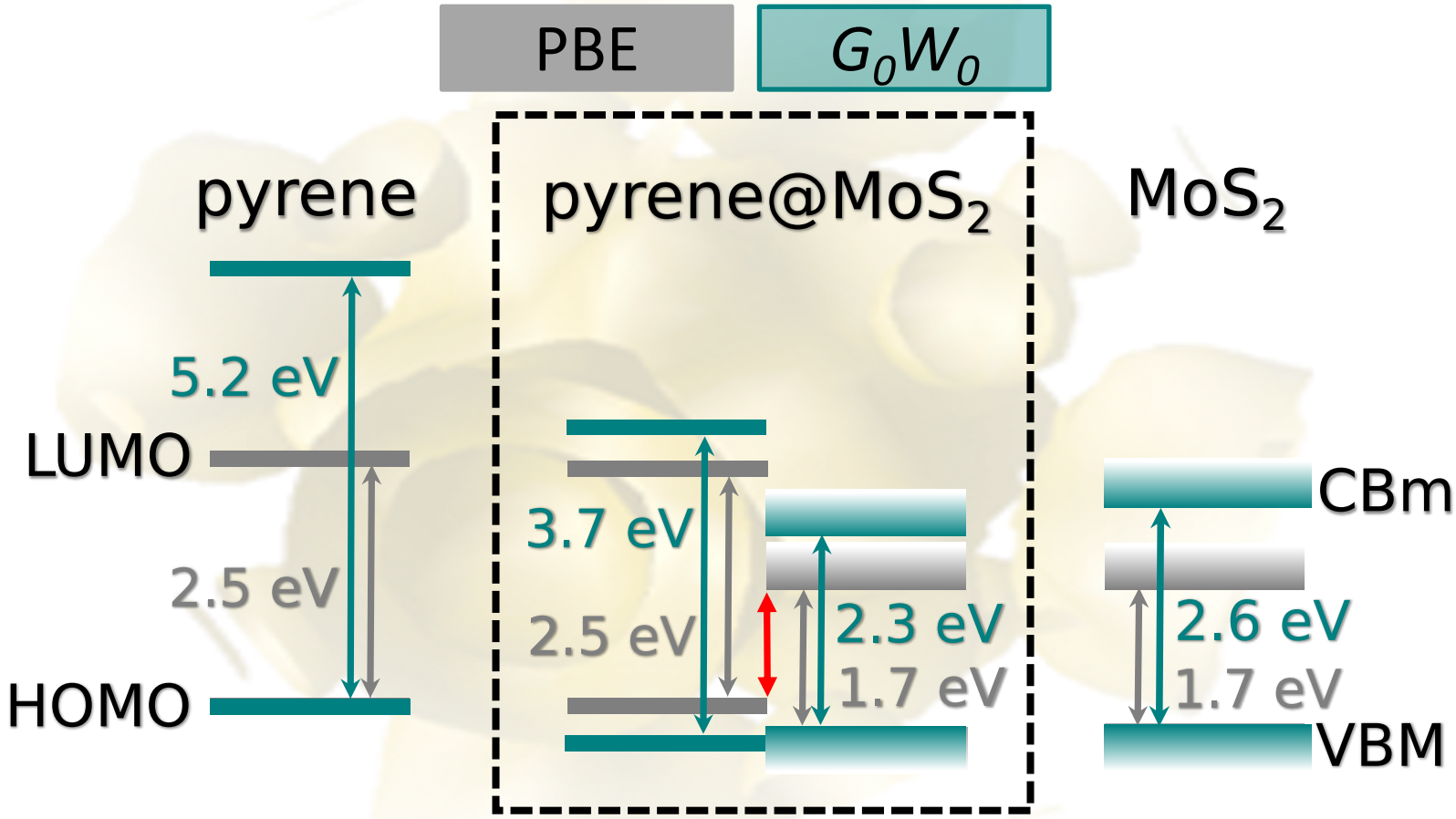


Level alignment scheme

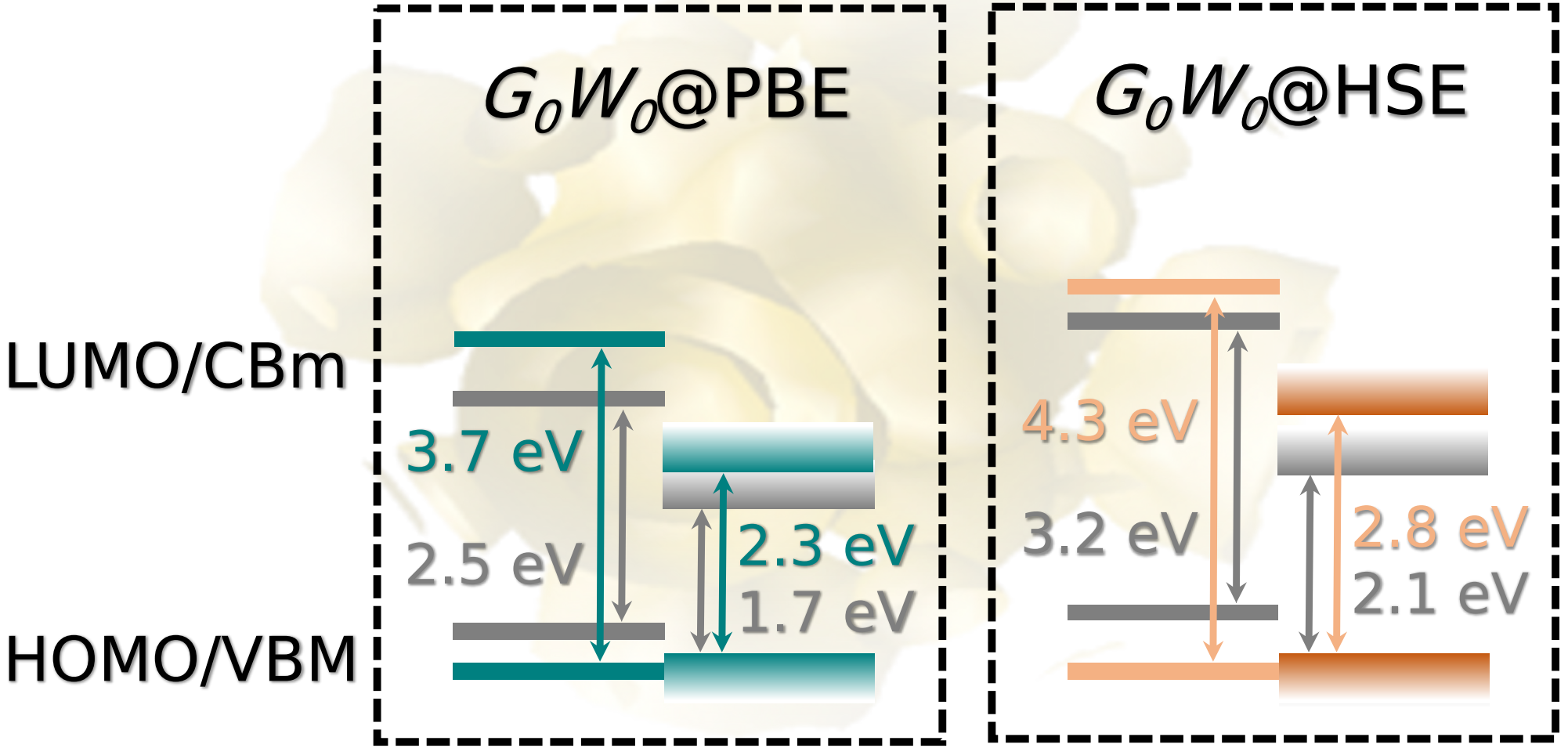
I. Gonzalez Oliva *et al.*, Phys. Rev. Materials. **6**, 054004 (2022).



Correction	Value (eV)
Self-energy	2.5
Polarization-induced	-1.5

Level alignment scheme (HSE vs PBE)

I. Gonzalez Oliva *et al.*, phys. stat. sol. (a) (2023); in print.



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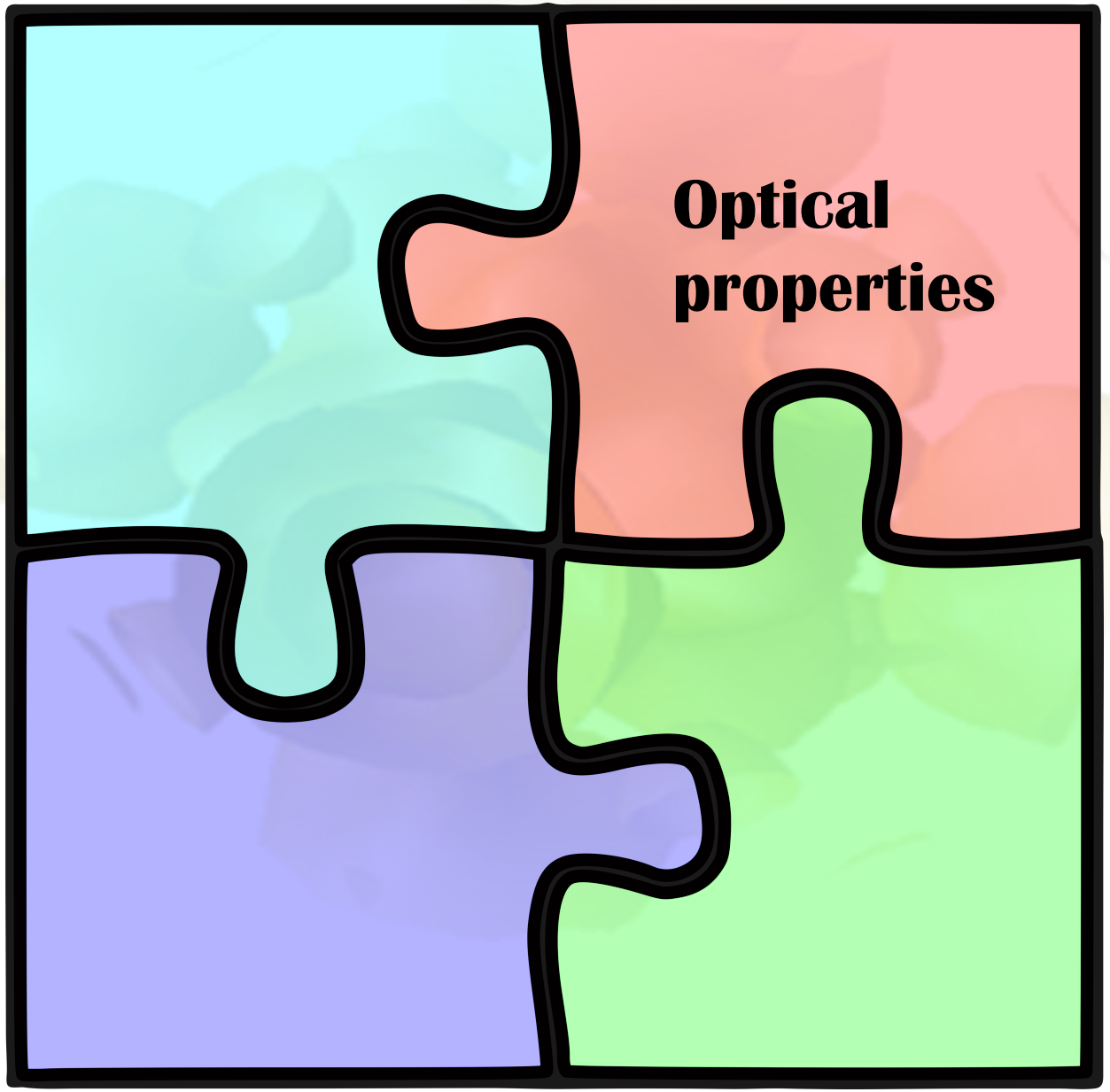
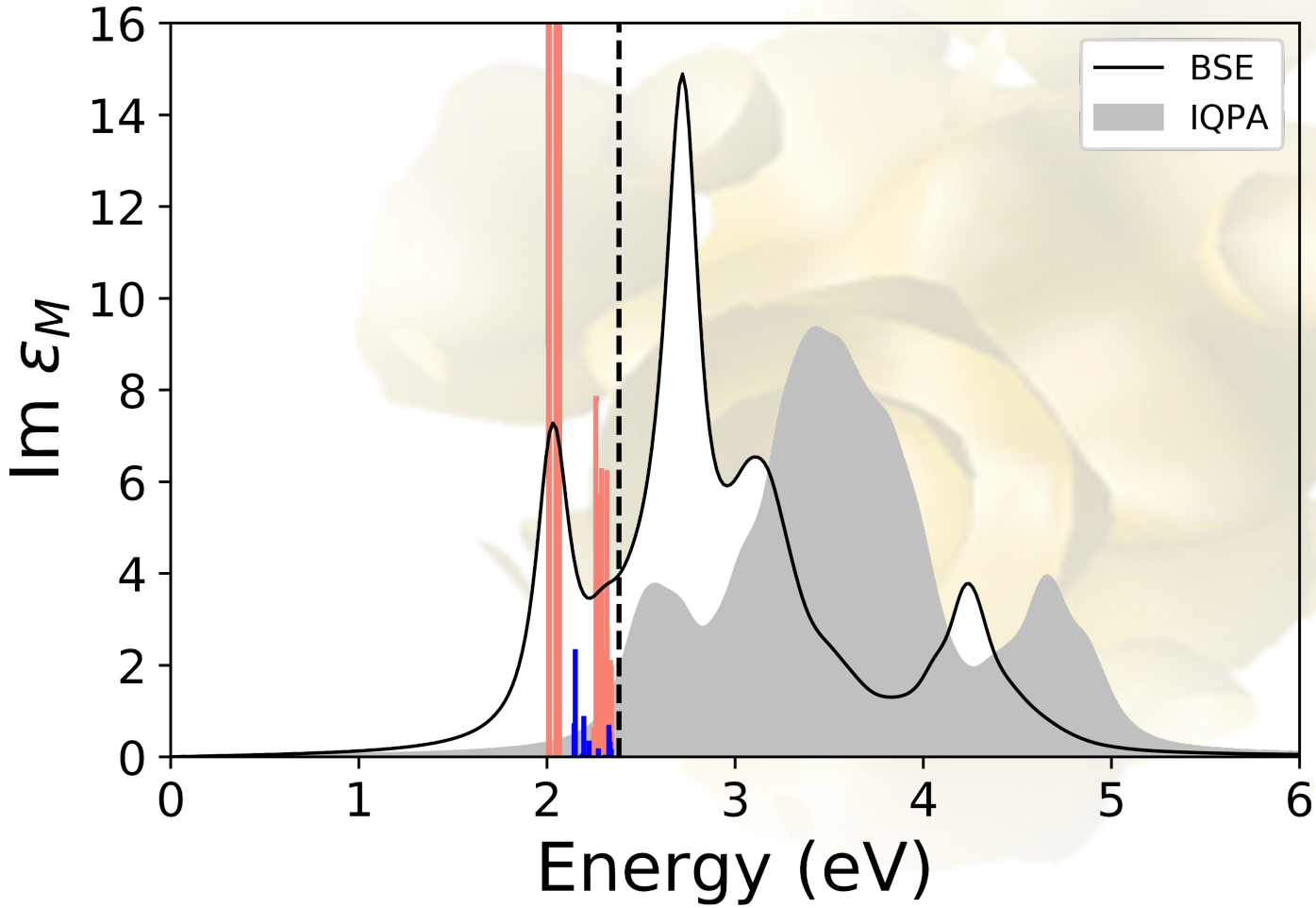


Absorption spectrum

I. Gonzalez Oliva *et al.*, Phys. Rev. Materials. **6**, 054004 (2022).

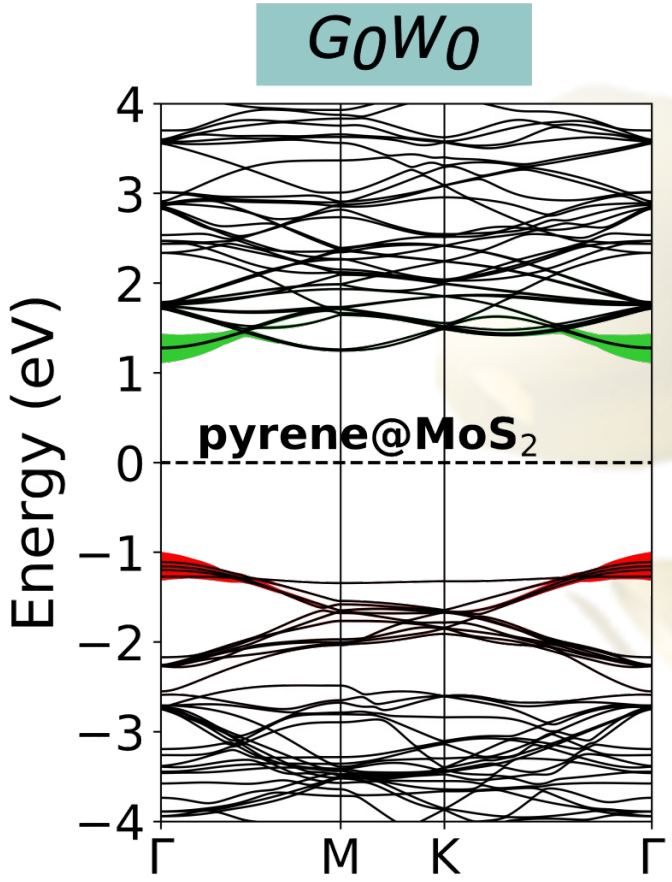


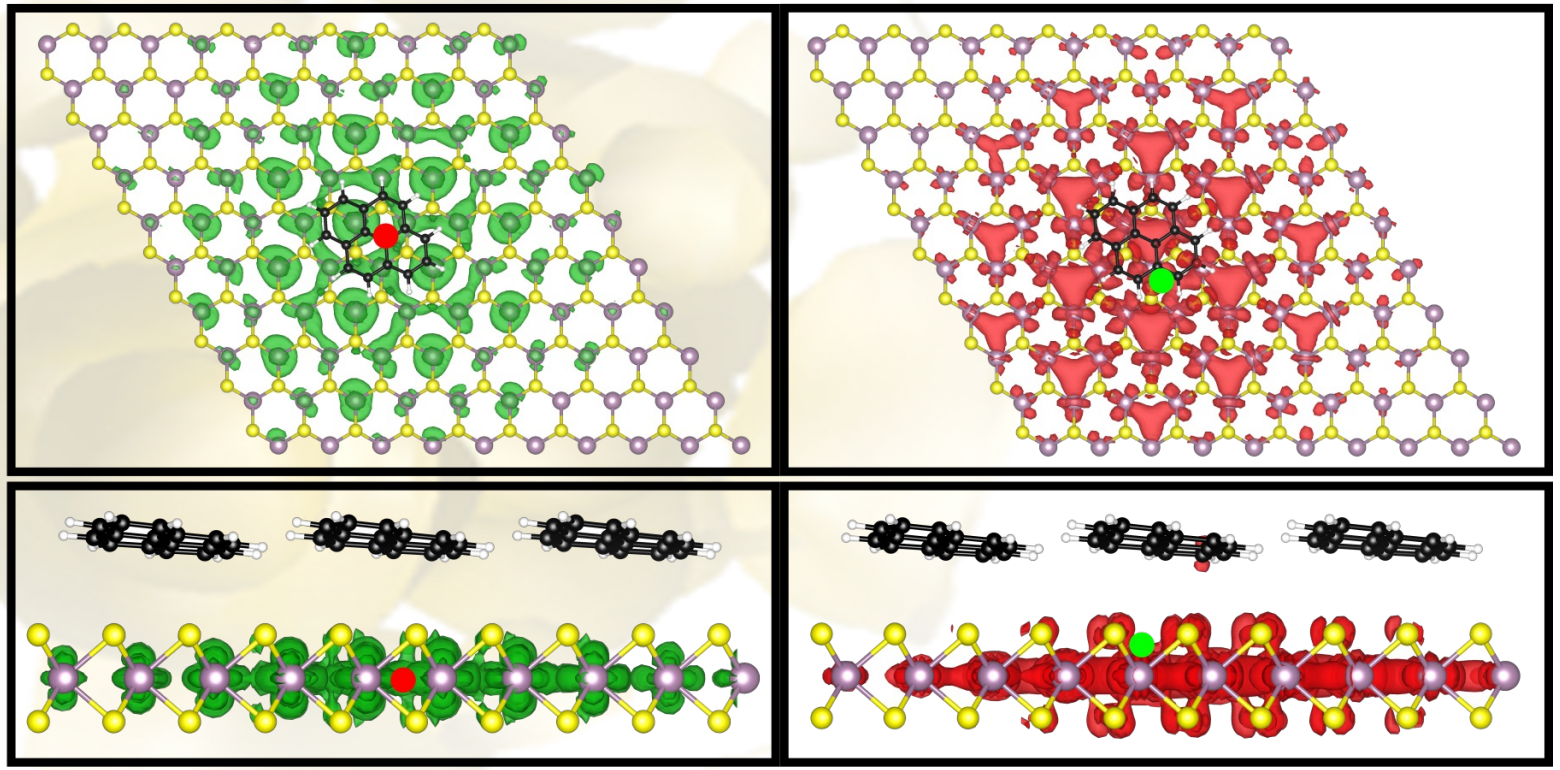
Type of excitons

1. MoS_2 (red)
2. Charge-transfer (blue)

MoS_2 exciton

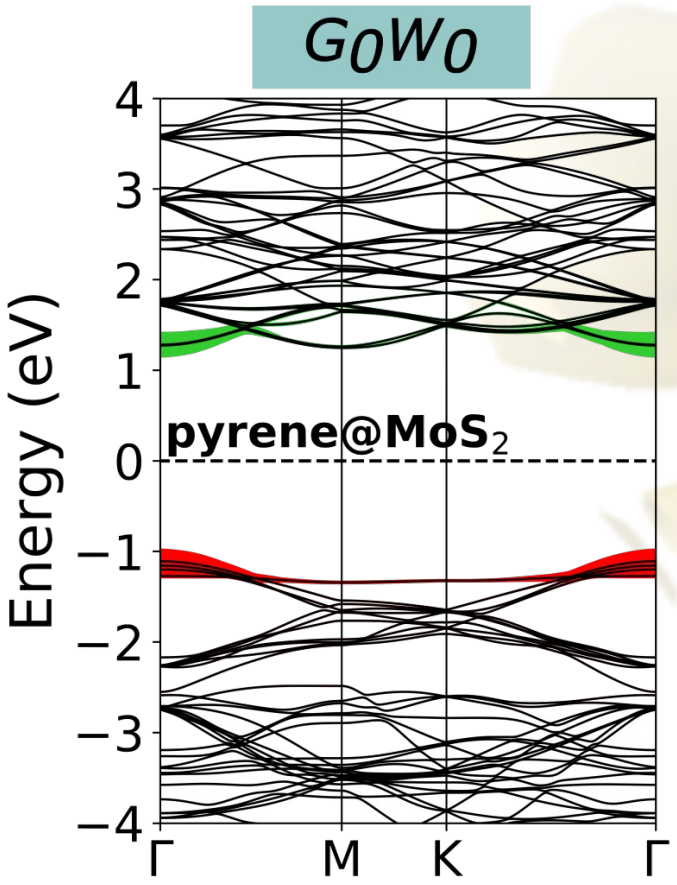
I. Gonzalez Oliva *et al.*, Phys. Rev. Materials. **6**, 054004 (2022).

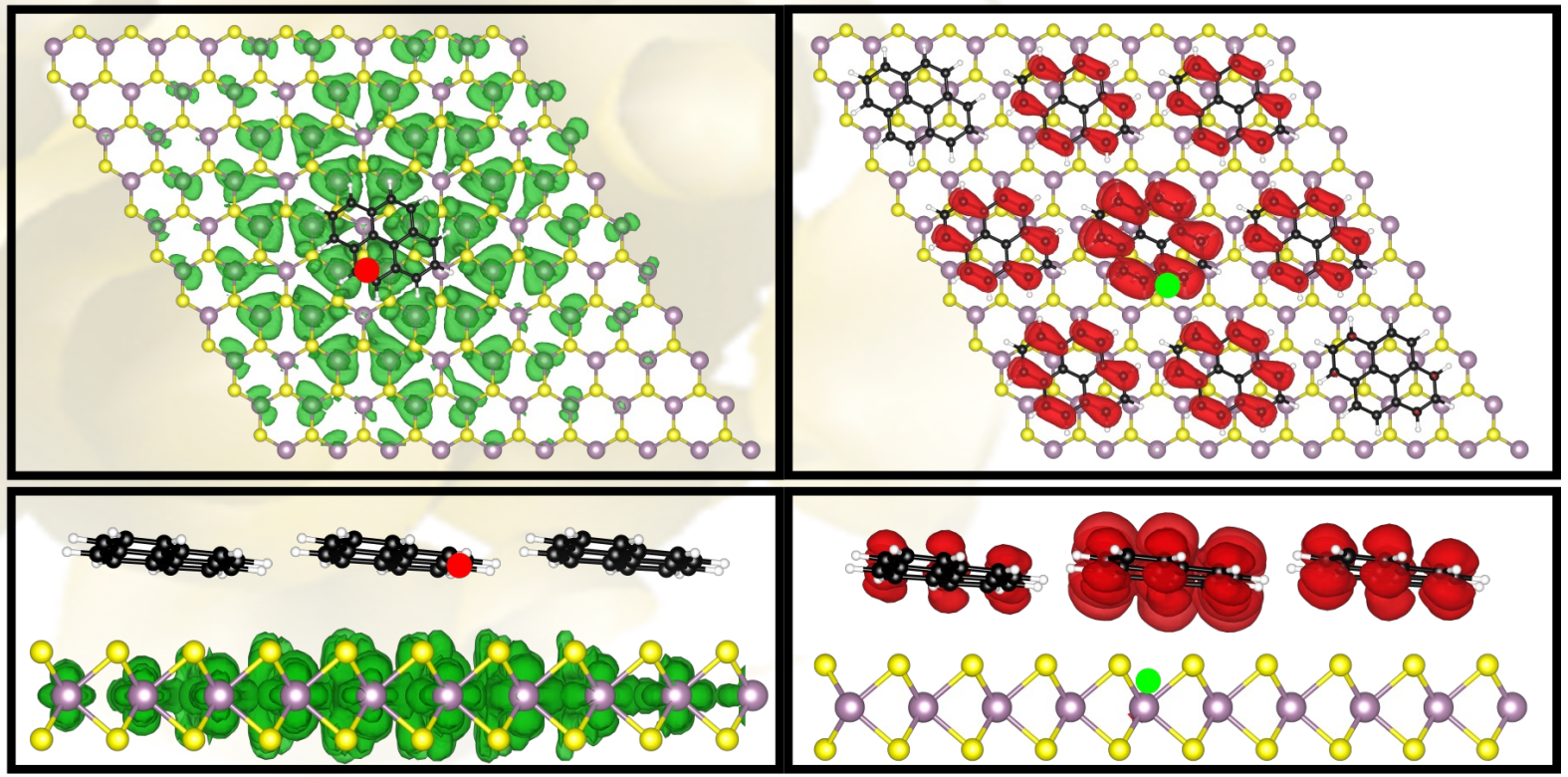




Charge-transfer exciton

I. Gonzalez Oliva *et al.*, Phys. Rev. Materials. **6**, 054004 (2022).





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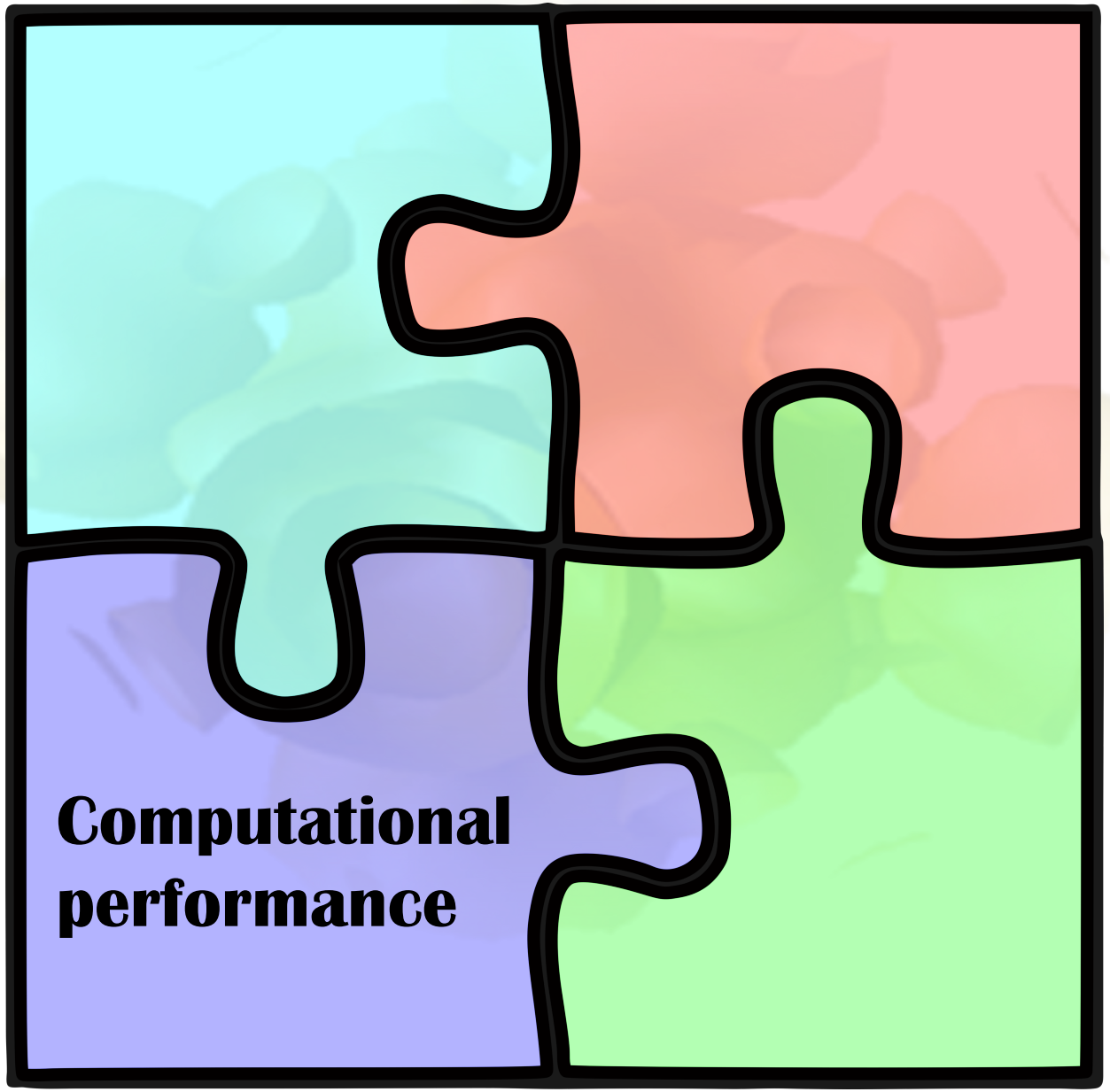
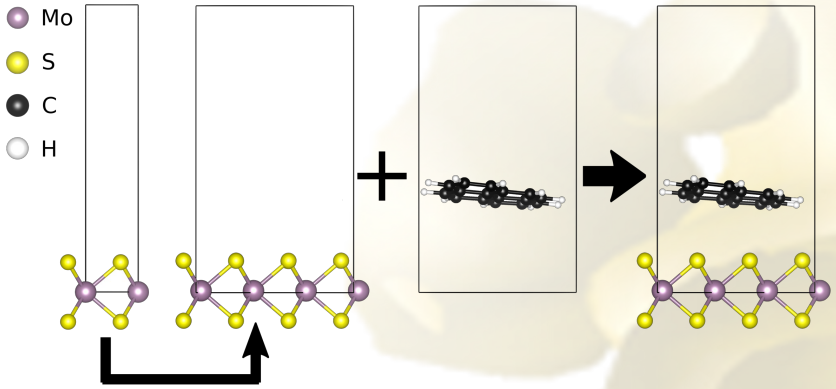
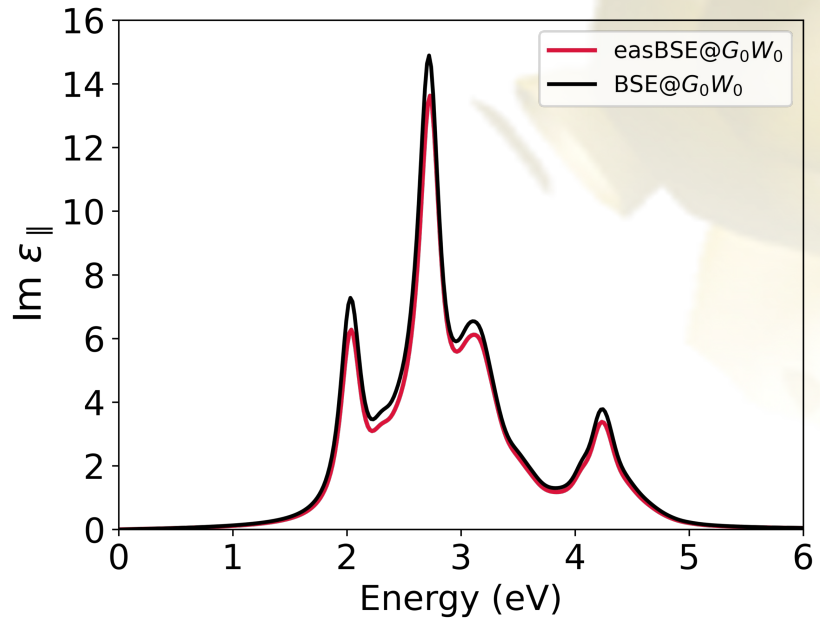


Speeding up BSE calculations

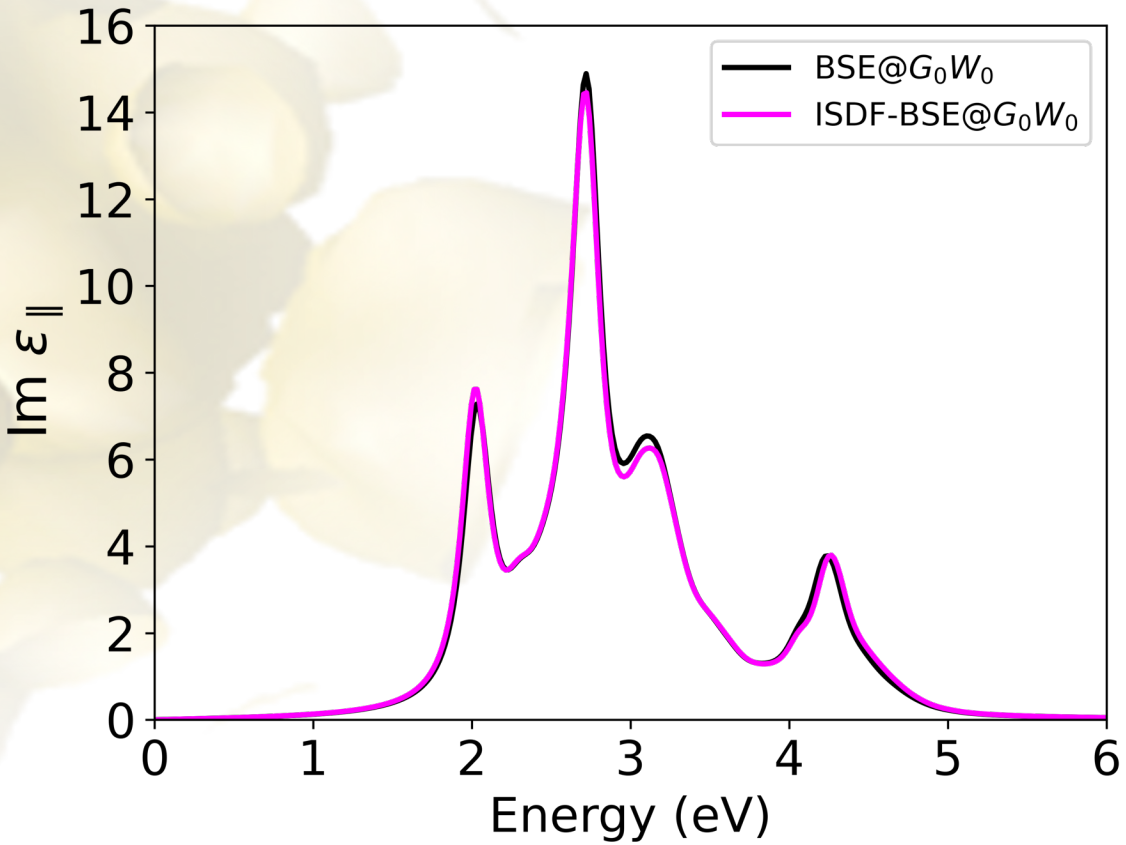
I. Gonzalez Oliva *et al.*, phys. stat. sol. (a) (2023); in print.

Expasion-addition screening





Interpolative separable density fitting (ISDF)



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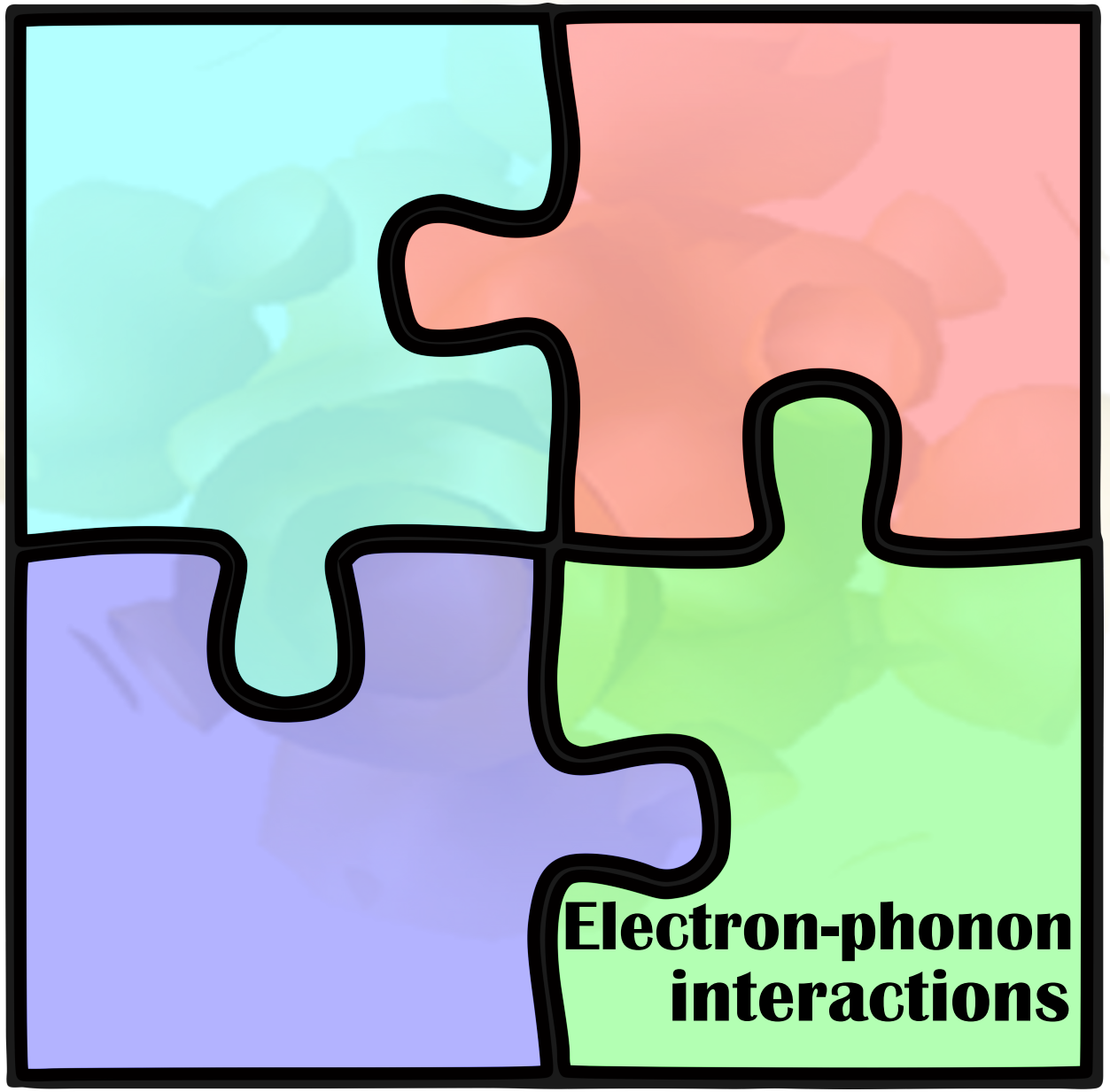
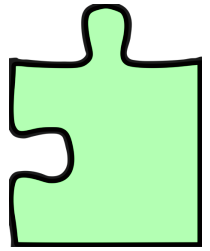


Electron-phonon interaction: self-energy



Disclaimer: The following is not yet release in `exciting` and the results are under analysis

However, they look very promising ☺

$$\Sigma_n^{e-ph}(\mathbf{k},\omega) \approx \sum_{\nu m} \int \frac{d\mathbf{q}}{V_{BZ}} |g_{mn\nu}(\mathbf{k},\mathbf{q})|^2 [\dots]$$

$$[\dots] = \left[\frac{1 - f_{m\mathbf{k}+\mathbf{q}} + n_{\nu\mathbf{q}}}{\omega - \epsilon_{m\mathbf{k}+\mathbf{q}} - \omega_{\nu\mathbf{q}} - i\eta} + \frac{f_{m\mathbf{k}+\mathbf{q}} + n_{\nu\mathbf{q}}}{\omega - \epsilon_{m\mathbf{k}+\mathbf{q}} + \omega_{\nu\mathbf{q}} - i\eta} \right]$$

Electrons → DFT(+GW)

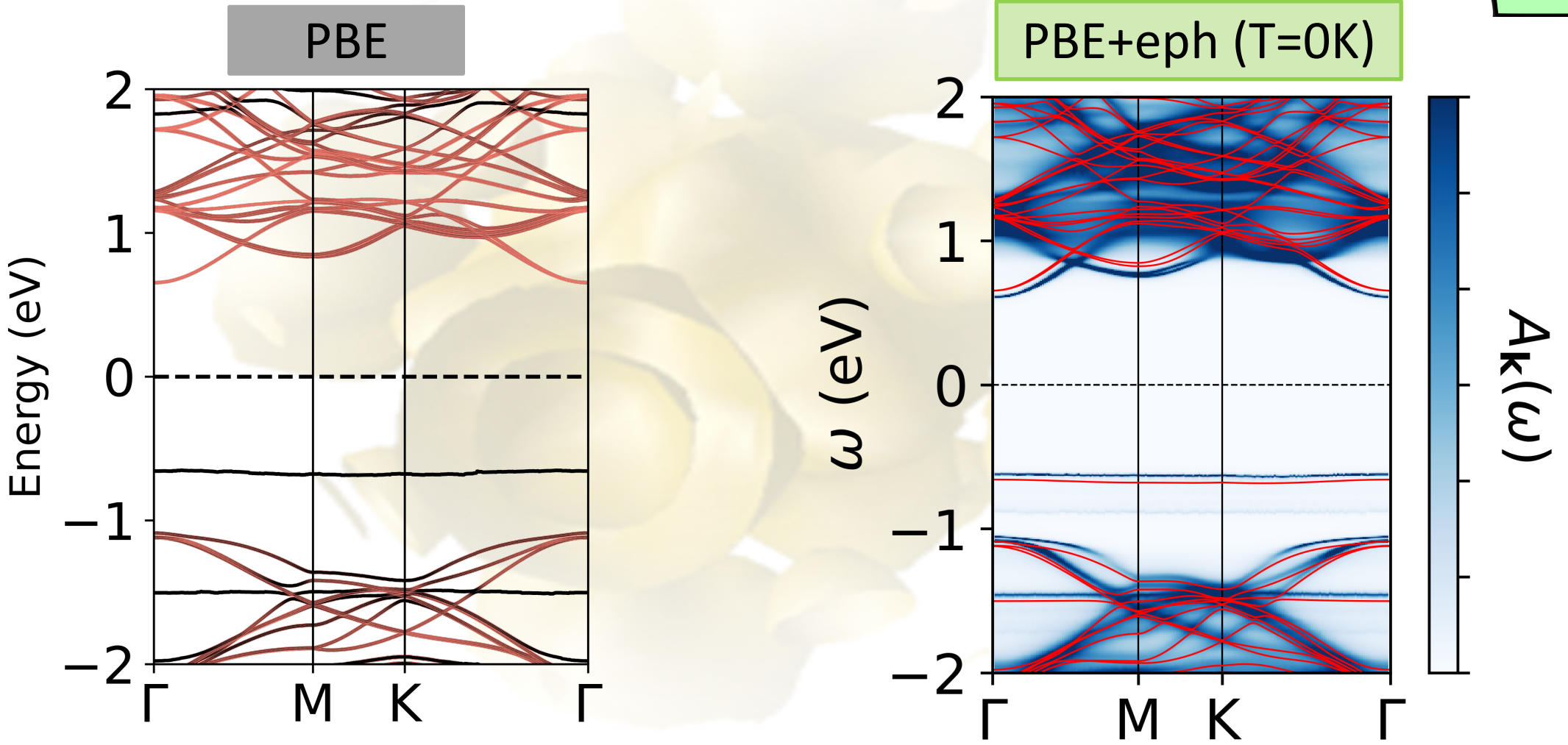
Phonons → DFT (DFPT)

Coupling → DFT (DFPT)

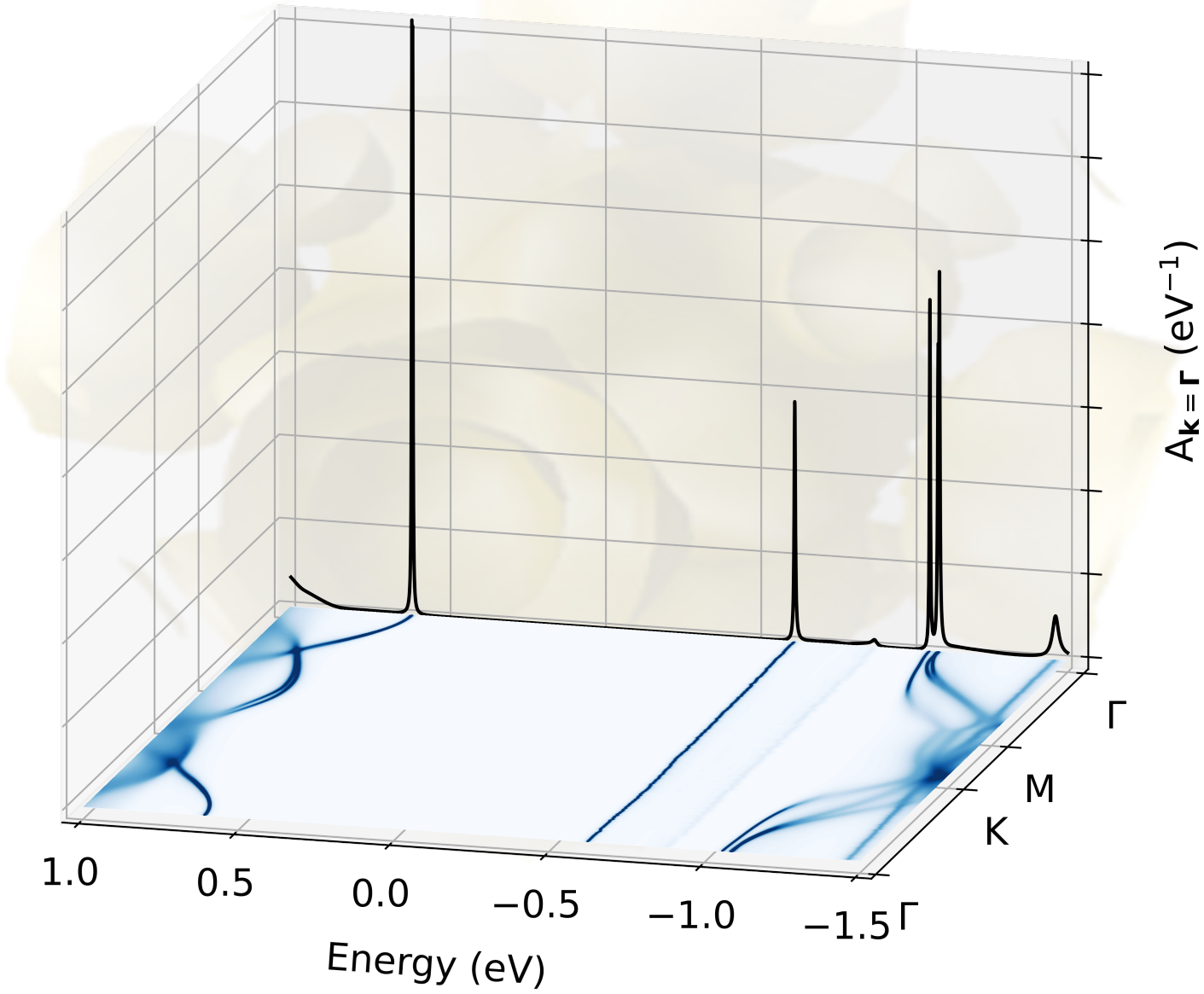
Many-body momentum-resolved DOS (direct comparison with ARPES)

$$A(\omega,\mathbf{k}) = -\frac{1}{\pi} \sum_n \frac{\mathrm{Im}\Sigma_n(\mathbf{k},\omega)}{(\omega - \epsilon_{n\mathbf{k}} - \mathrm{Re}\Sigma_n(\mathbf{k},\omega))^2 + \mathrm{Im}\Sigma_n(\mathbf{k},\omega)^2}$$

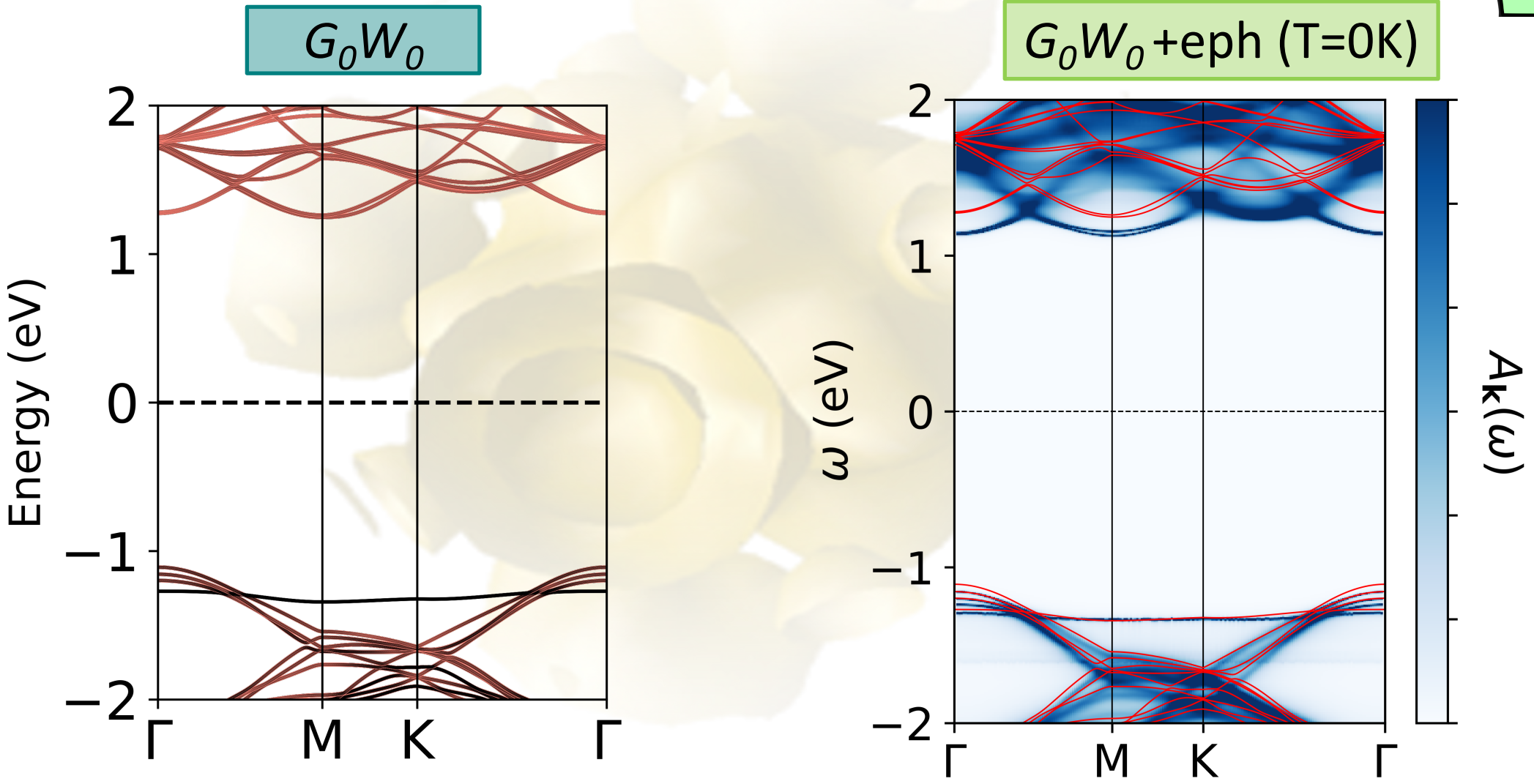
Electron-phonon spectral function



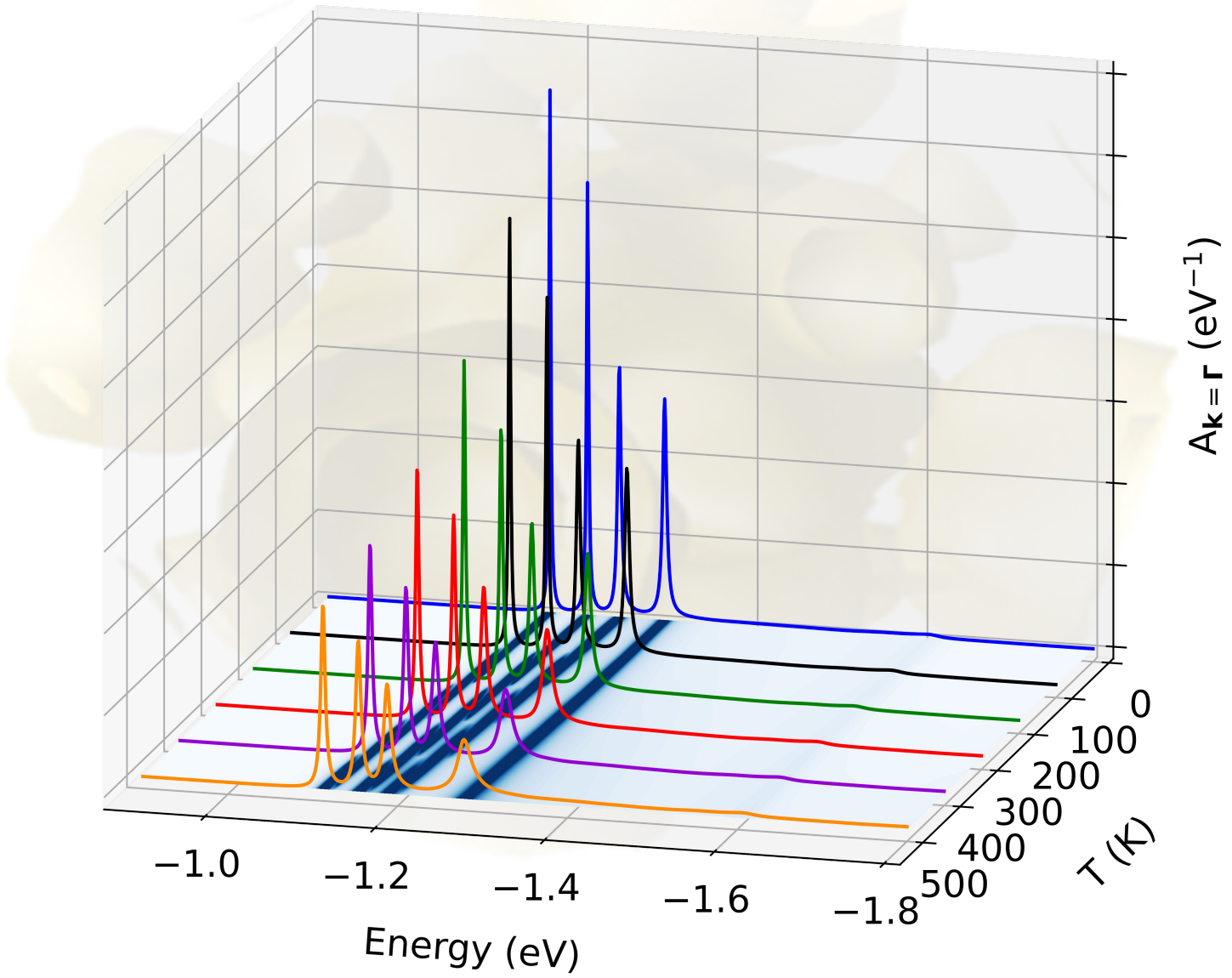
Electron-phonon spectral function at $k = \Gamma$



Electron-phonon spectral function + G_0W_0



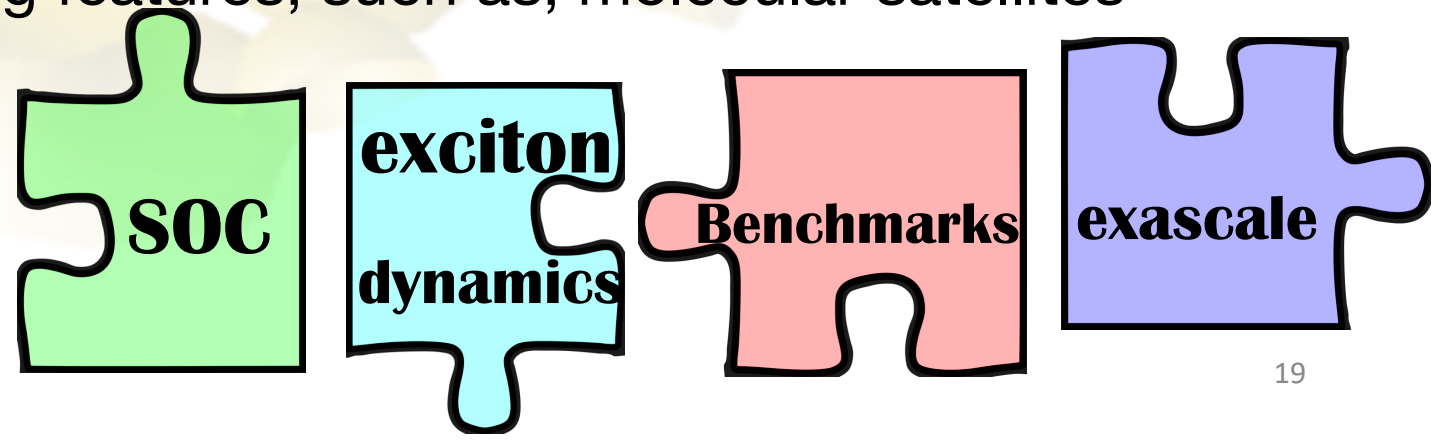
Temperature effects



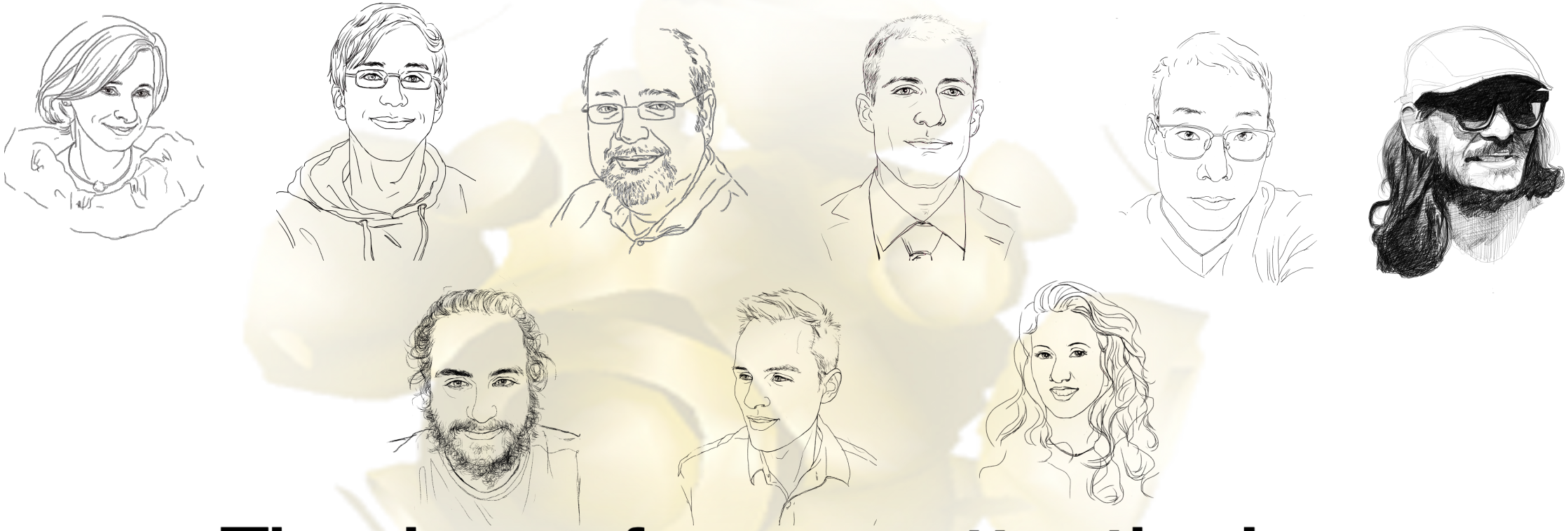
Summary and outlook

- G_0W_0 calculations reveal changes in the type of level alignment
- HSE improves the HOMO-LUMO gap and MoS_2 gap but does not change the level alignment
- Two types of excitons are described: MoS_2 like and charge-transfer
- New implementations speed up BSE calculations
- Initial results show interesting features, such as, molecular satellites

Puzzle is not complete!



Acknowledgements



Thank you for your attention!

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