Ab-initio study of the temperature effects on the optical properties of transition metal dichalcogenides

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Motivation

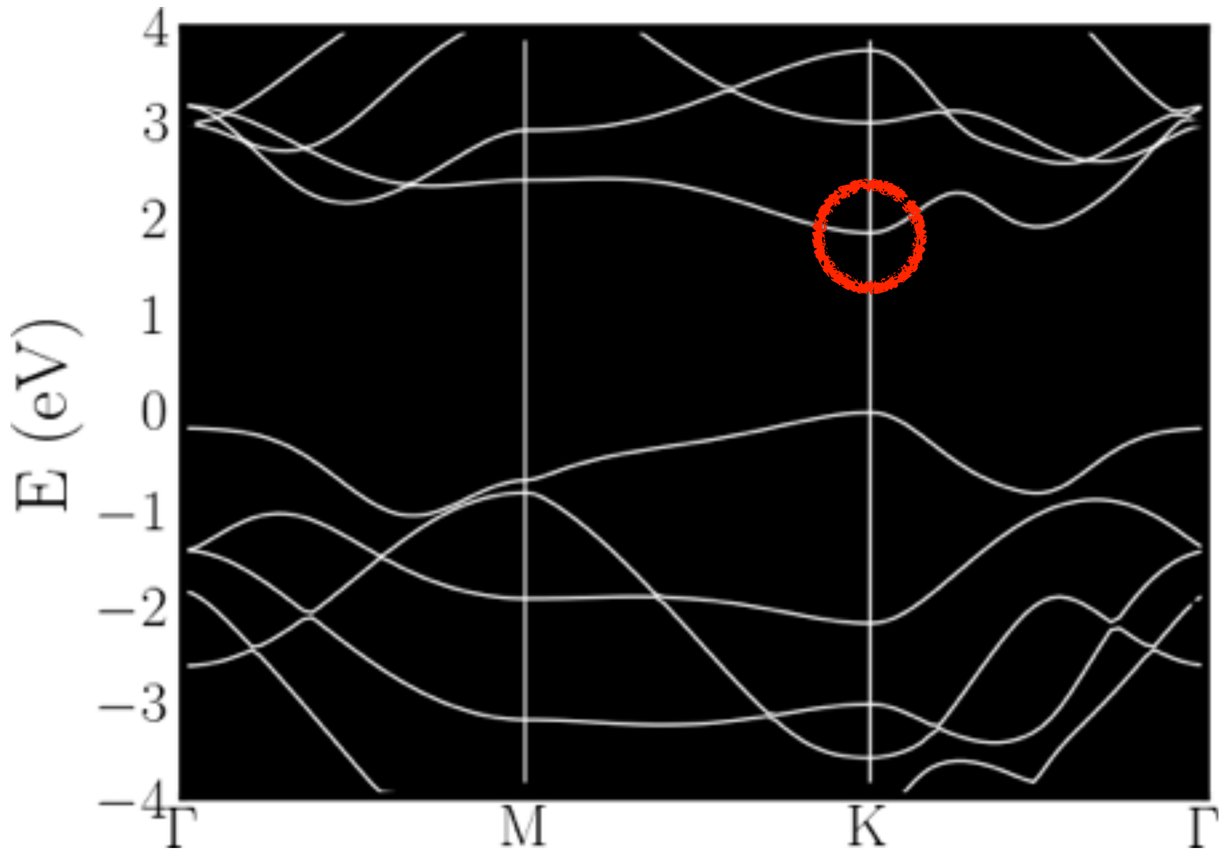
Realistic description of electronic structure, electron-lattice interaction

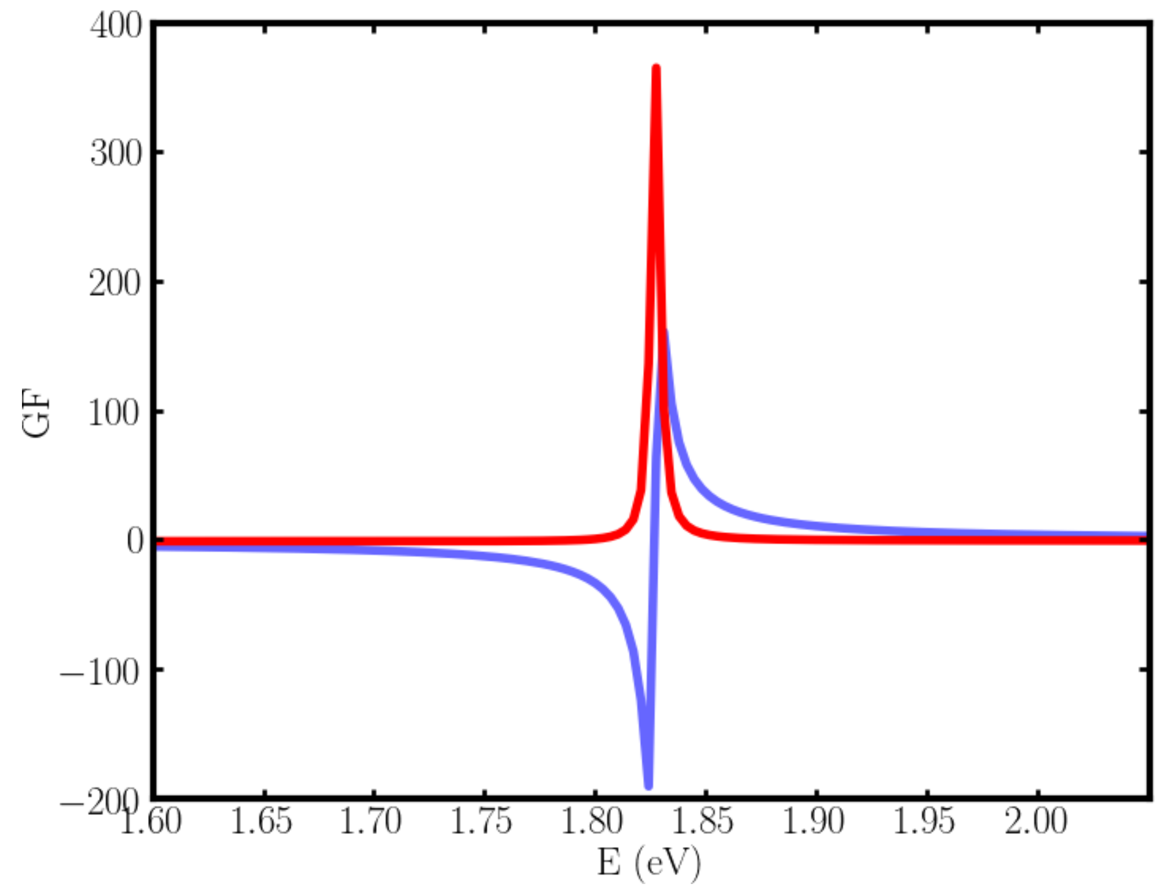
Temperature effects: bandgap renormalization, finite lifetime for electrons and excitons

First step for a carrier relaxation study. Application in valley depolarization, ultra-fast spectroscopy

A few formulas…

$$\bar{H} = \bar{H}_{el} + \bar{H}_{ion} + \bar{H}_{el-ion}$$

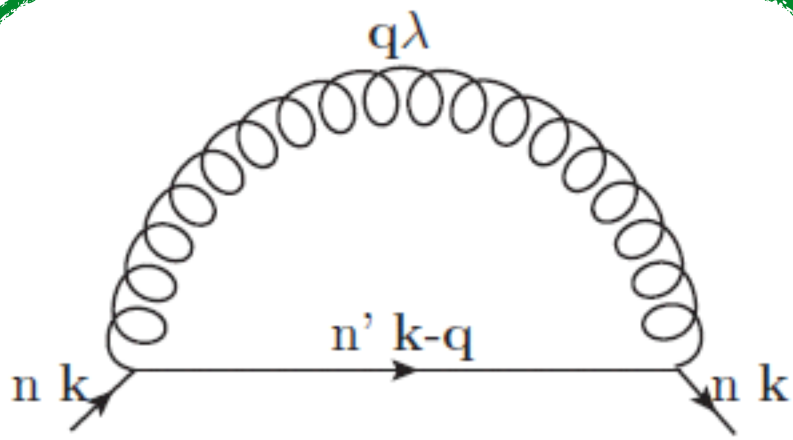




Without lattice-interaction: Electronic states have infinite lifetimes

Reality: Lattice-interaction gives them a finite lifetime

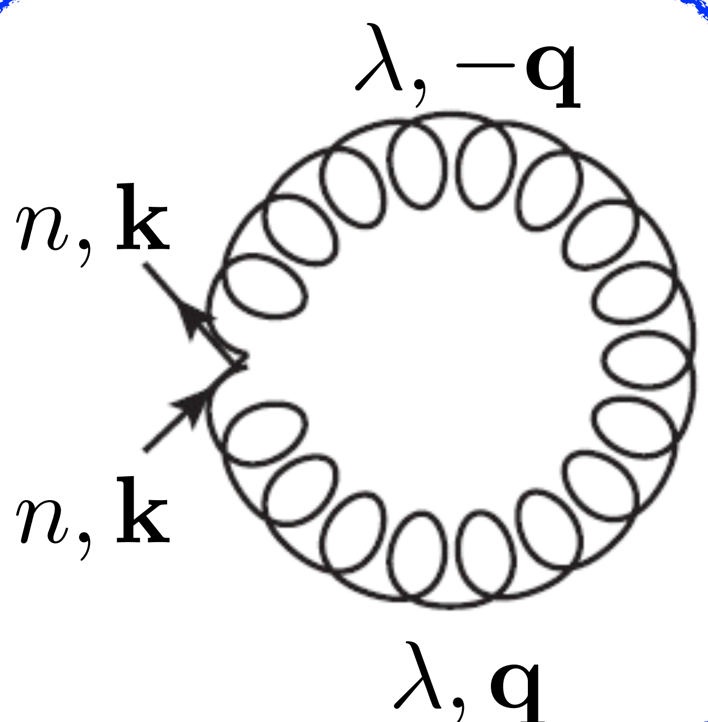
A few formulas…



Scattering of an electron by a phonon

Temperature enters via population of phonon states

$$\Sigma^{Fan}_{n\mathbf{k}}(i\omega_i, T) = \sum_{\mathbf{q}\lambda\mathbf{n}'} \frac{|g^{\mathbf{q}\lambda}_{nn'\mathbf{k}}|^2}{N_q} \left[\frac{N_{\mathbf{q}\lambda}(T) + 1 - f_{n'\mathbf{k}-\mathbf{q}}}{i\omega - \varepsilon_{n'\mathbf{k}-\mathbf{q}} - \omega\mathbf{q}\lambda - i0^+} + \frac{N_{\mathbf{q}\lambda}(T) + f_{n'\mathbf{k}-\mathbf{q}}}{i\omega - \varepsilon_{n'\mathbf{k}-\mathbf{q}} + \omega\mathbf{q}\lambda - i0^+} \right]$$



$$\Sigma^{DW}_{n\mathbf{k}}(T) = \sum_{\mathbf{q}\lambda} \Lambda^{\mathbf{q}\lambda,-\mathbf{q}\lambda}_{nn\mathbf{k}} (2N_{\mathbf{q}\lambda}(T) + 1)$$

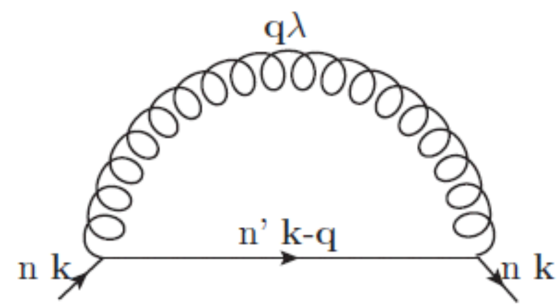
Two-phonon scattering process

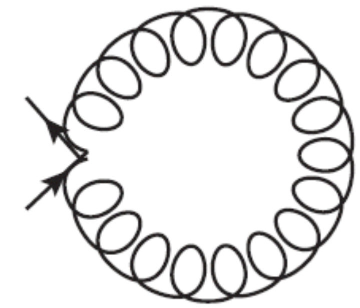
Quasiparticle approximation

$$E_{n\mathbf{k}} = \varepsilon_{n\mathbf{k}} + Z_{n\mathbf{k}} \left[\Sigma^{Fan}_{n\mathbf{k}}(\varepsilon_{n\mathbf{k}}, T) + \Sigma^{DW}_{n\mathbf{k}}(T) \right]$$

Complex energy

Bare energy





$$A_{n\mathbf{k}}(\omega, T) = \frac{Z_{n\mathbf{k}}(T)|\Gamma_{n\mathbf{k}}(T)|}{\pi \left[(\omega - \Re[E_{n\mathbf{k}}](T))^2 + \Gamma^2_{n\mathbf{k}}(T) \right]}$$

$g^{\mathbf{q}\lambda}_{nn'\mathbf{k}}$

QUANTUM ESPRESSO

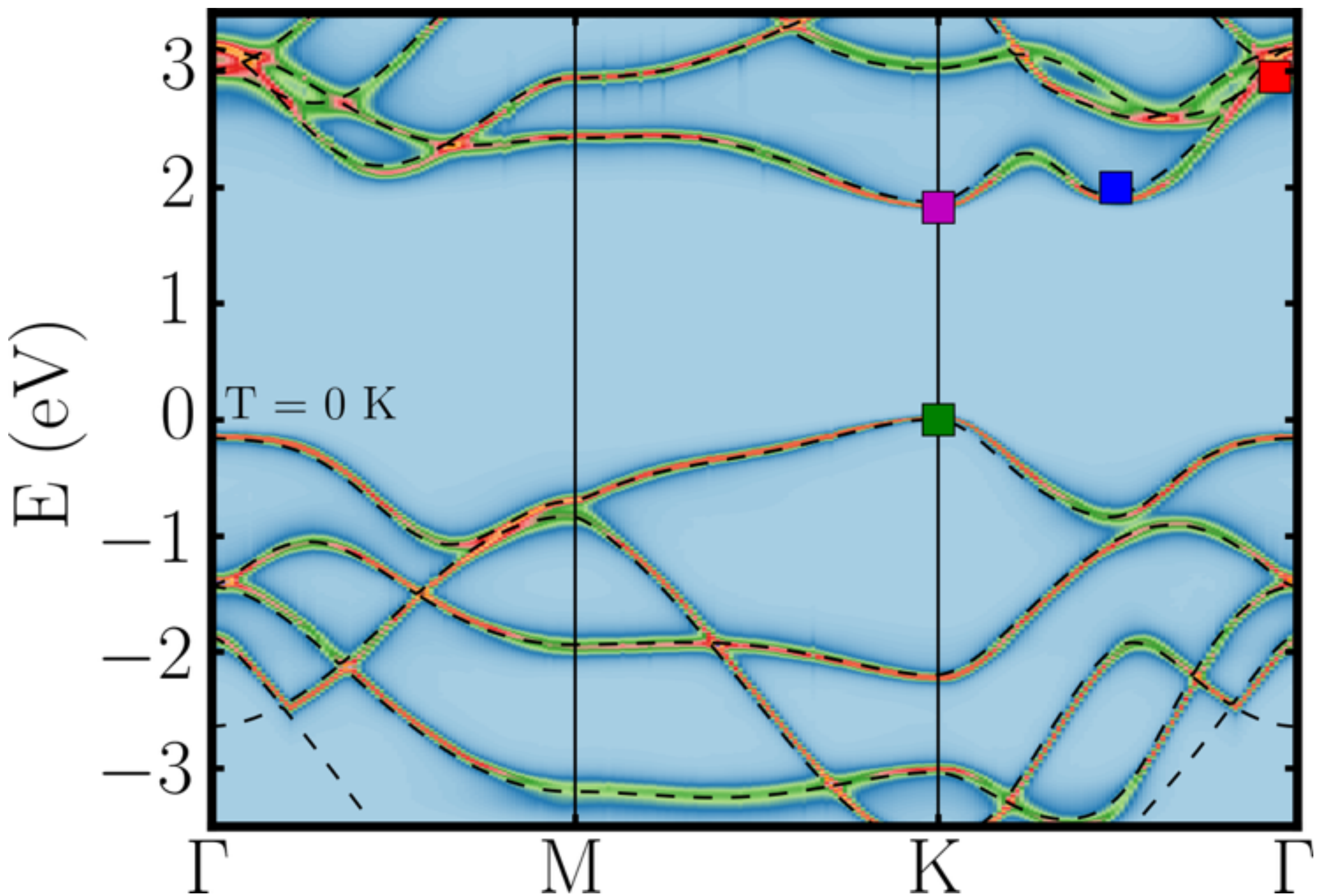
DFPT

$\Sigma_{n\mathbf{k}}(T)$

Yambo ©

MBPT

Single-layer MoS



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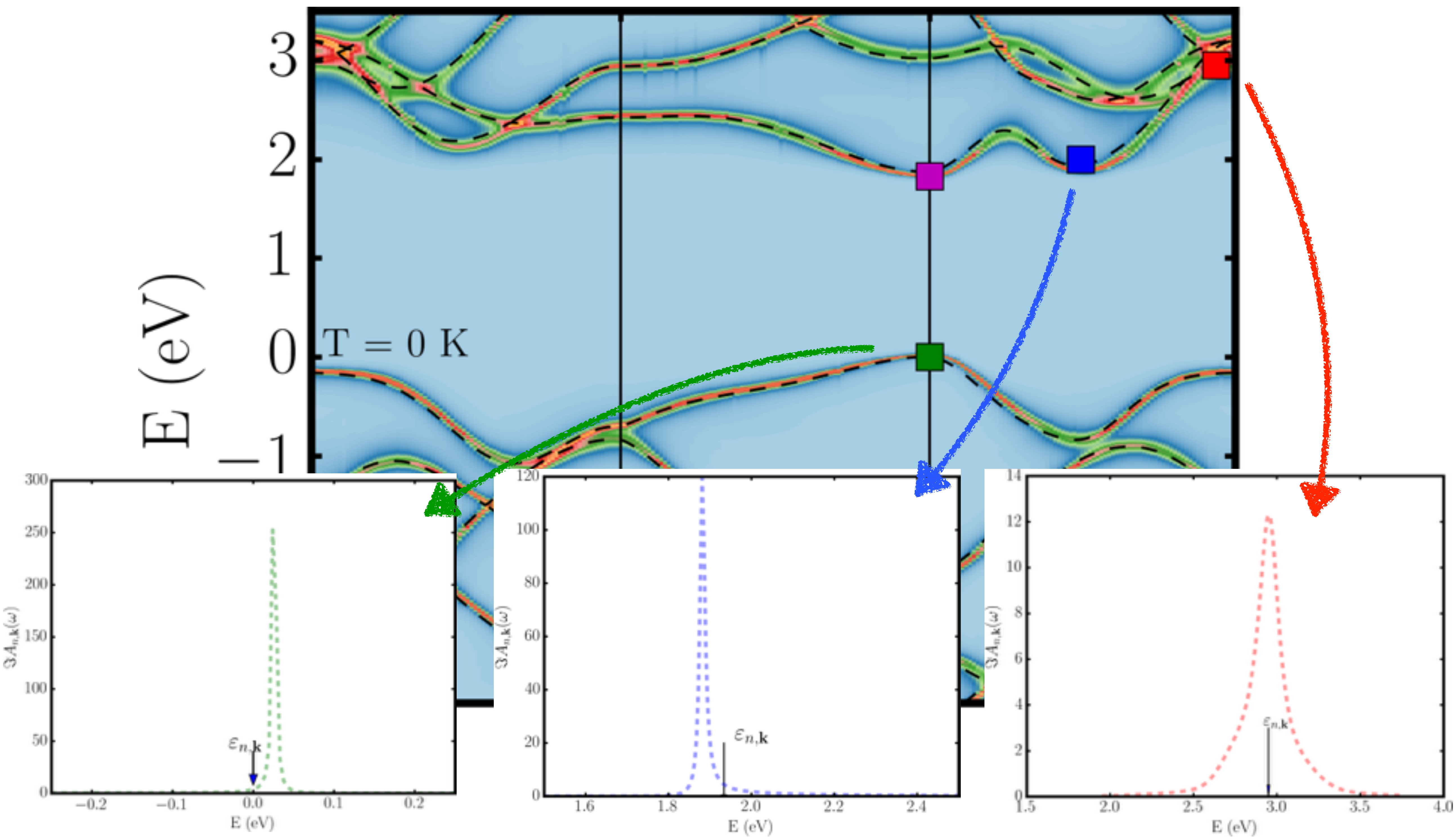

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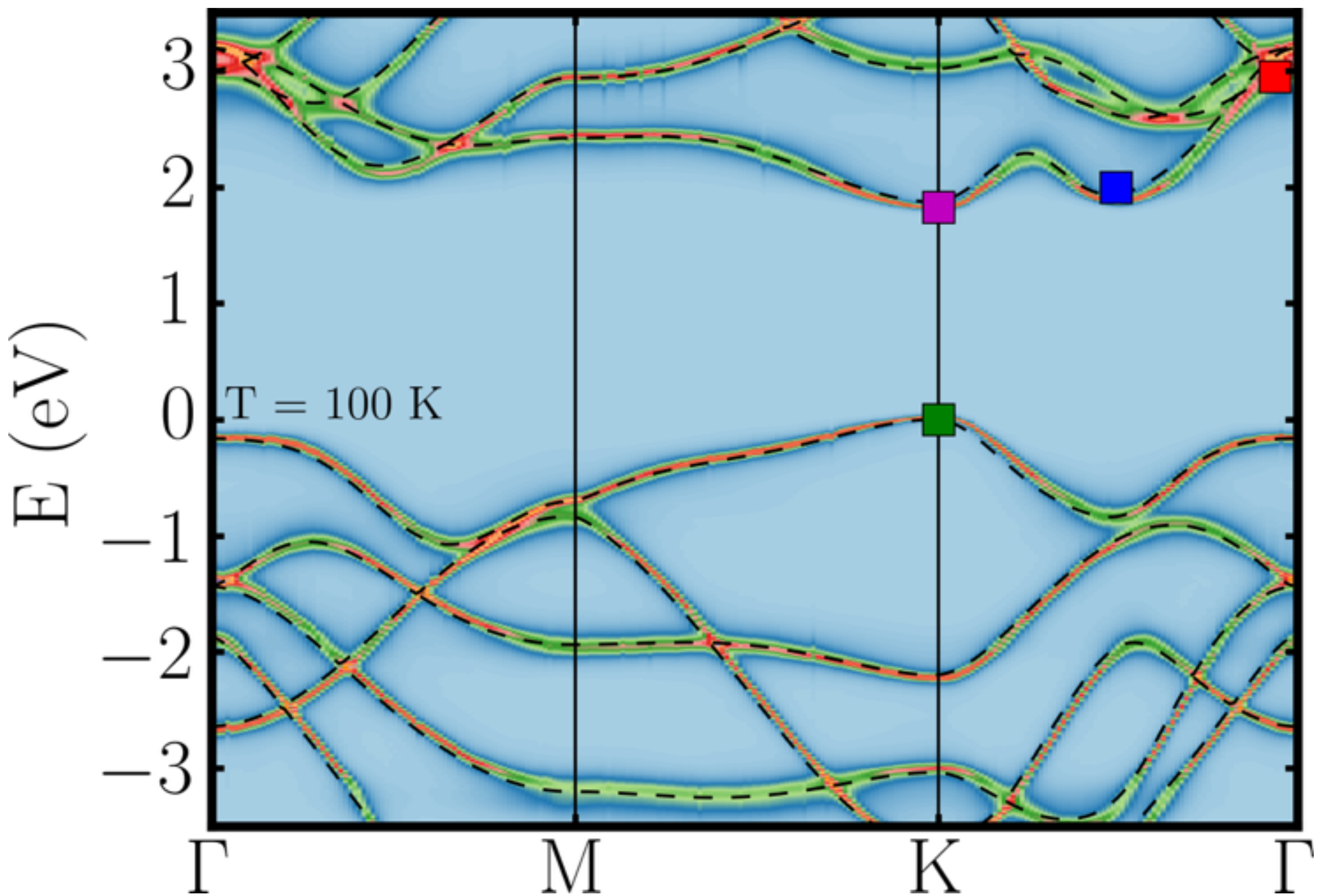

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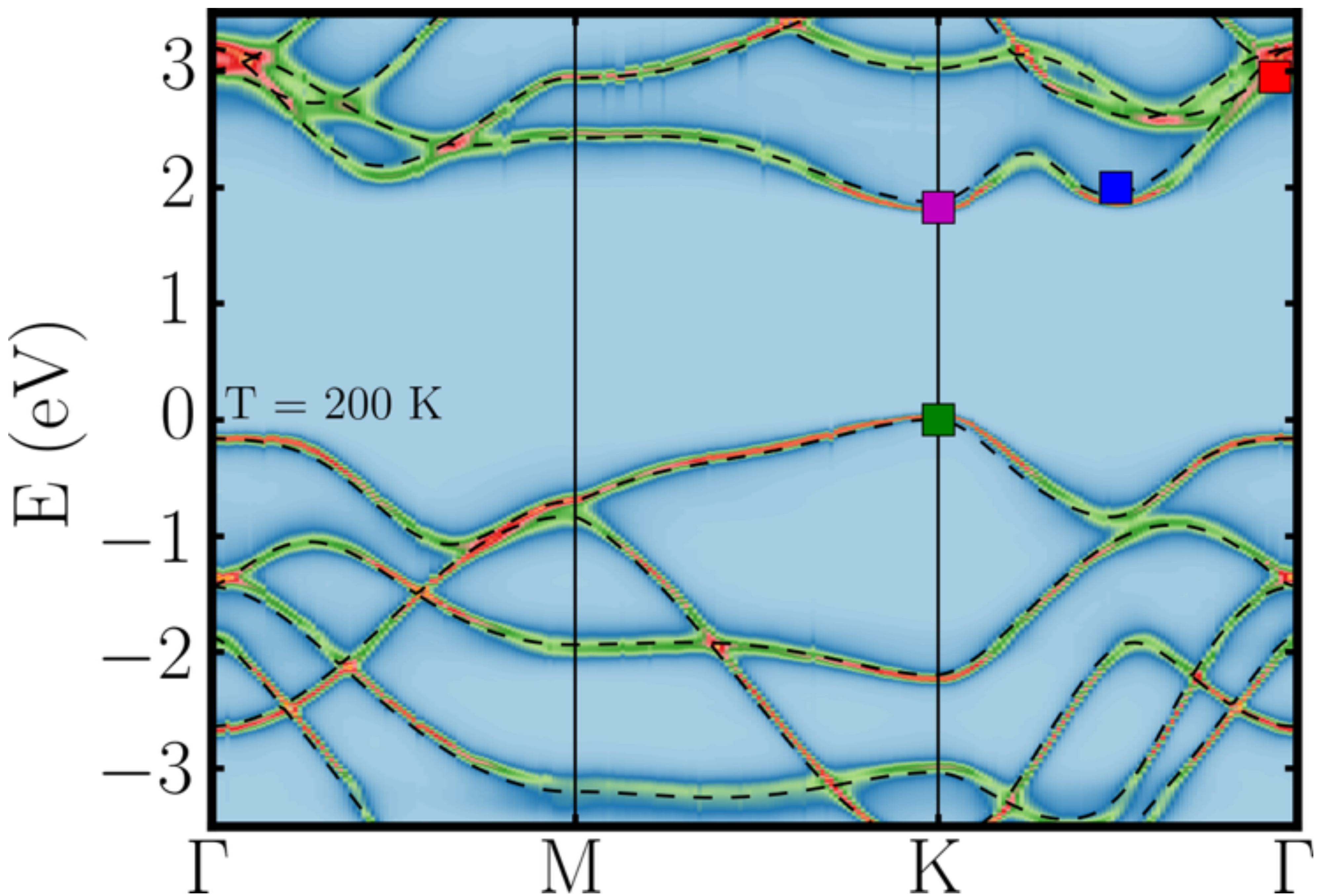

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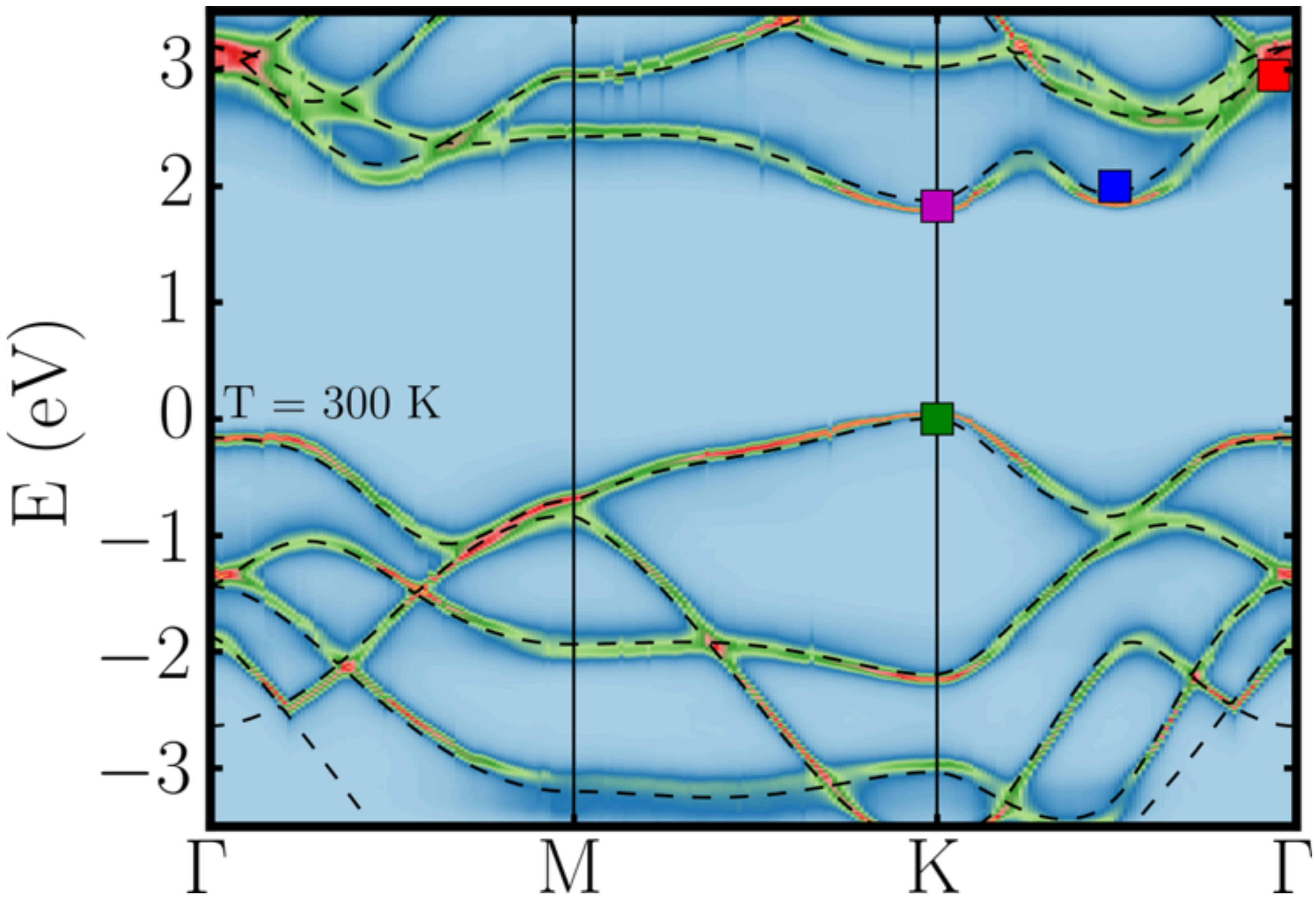

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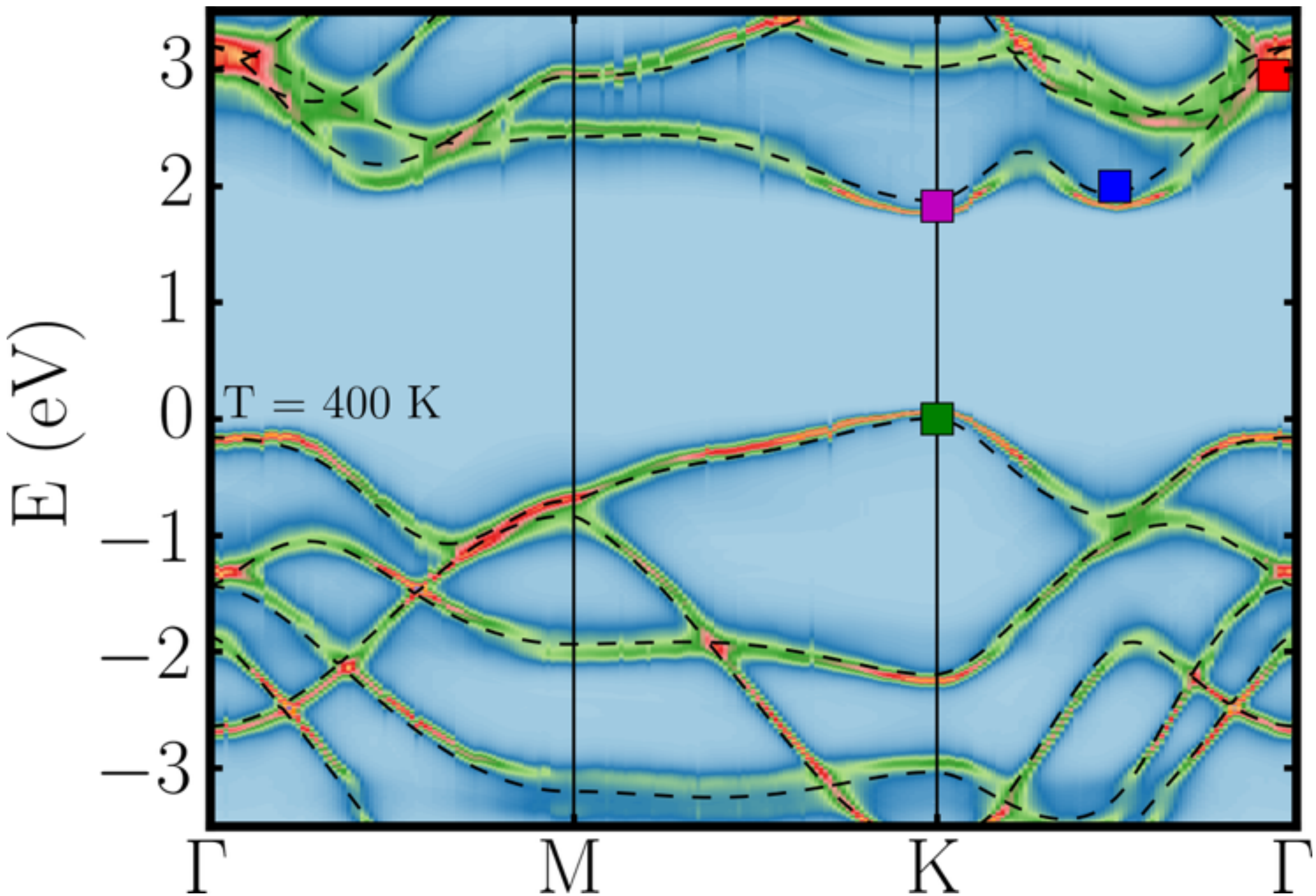

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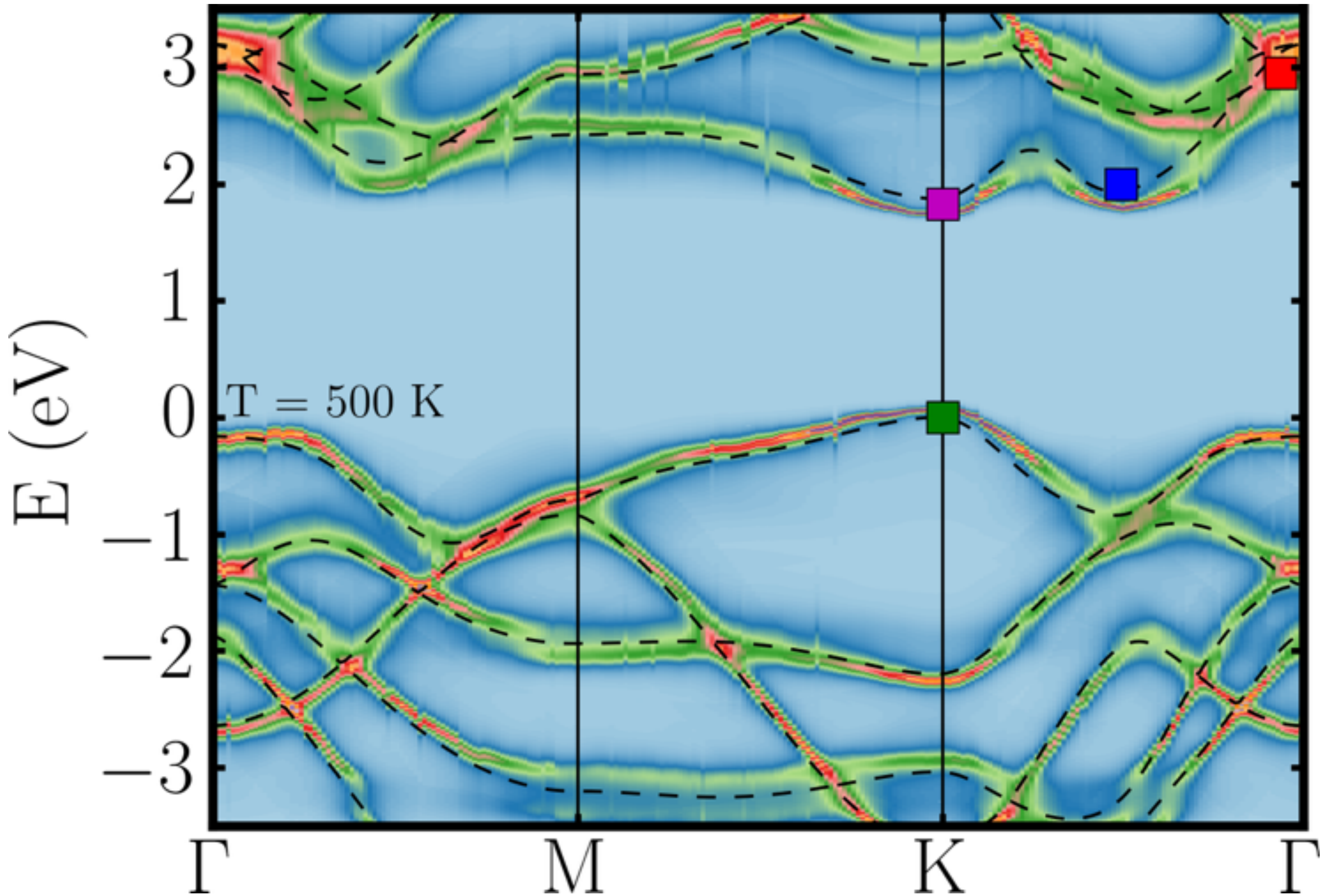

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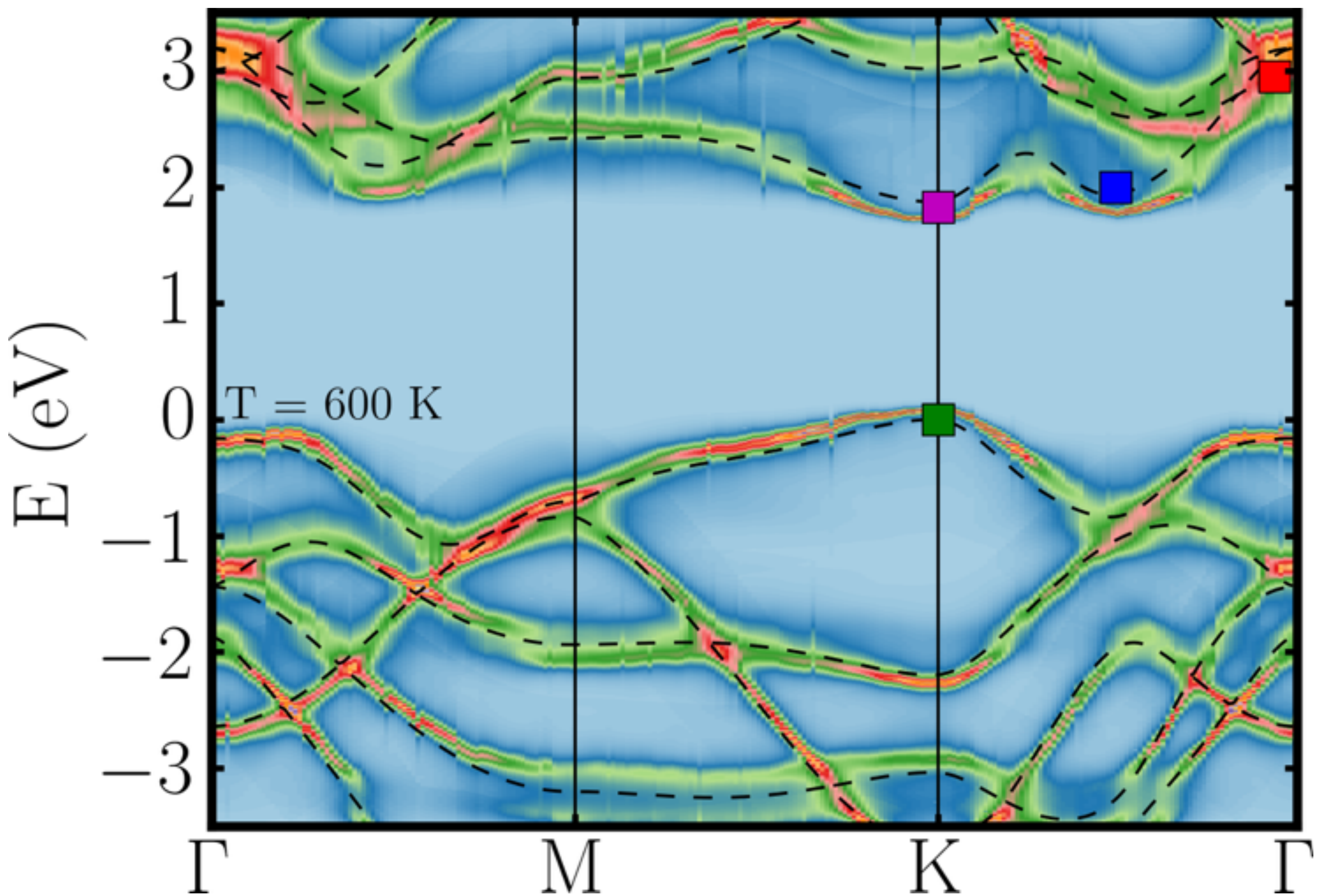

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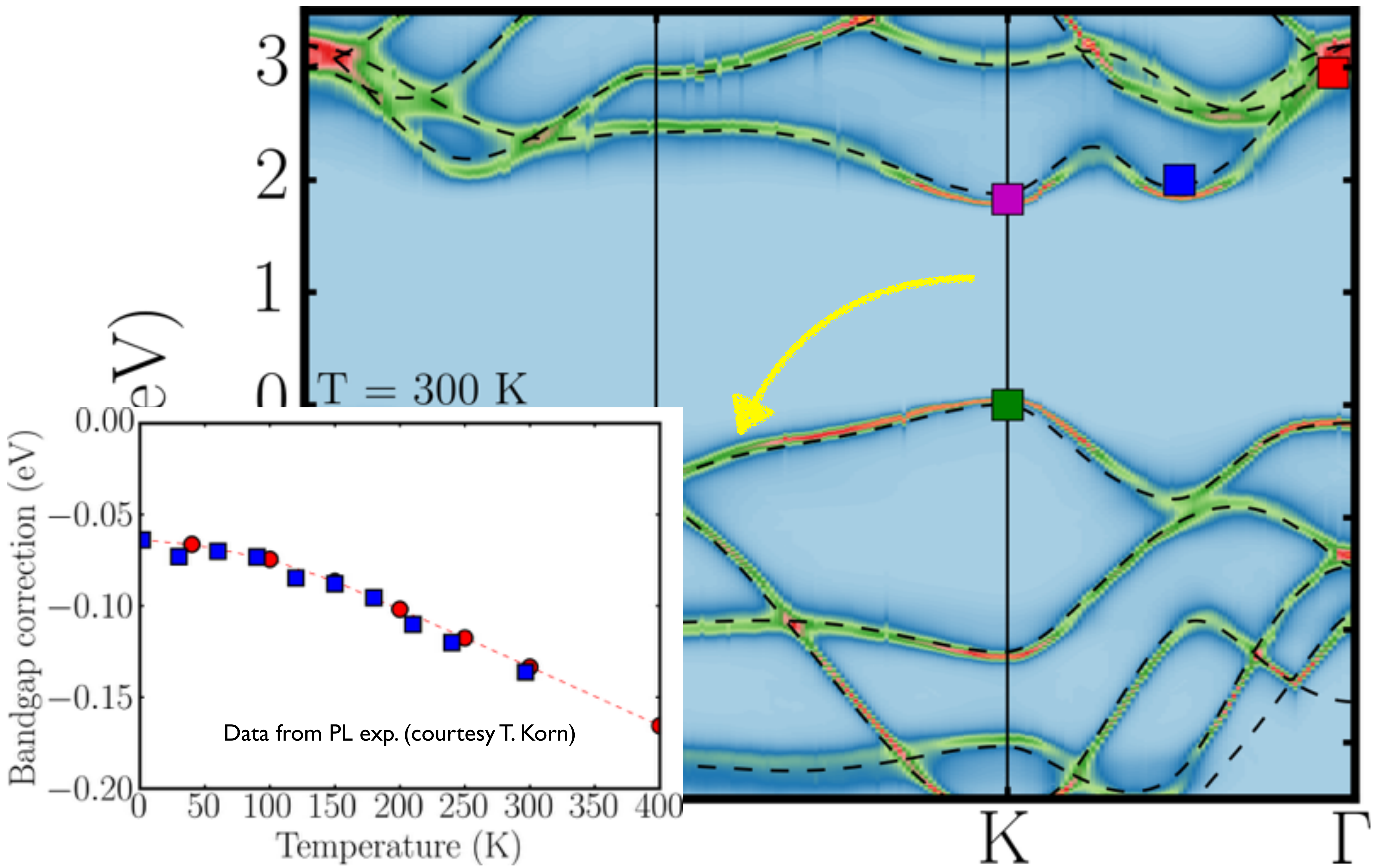
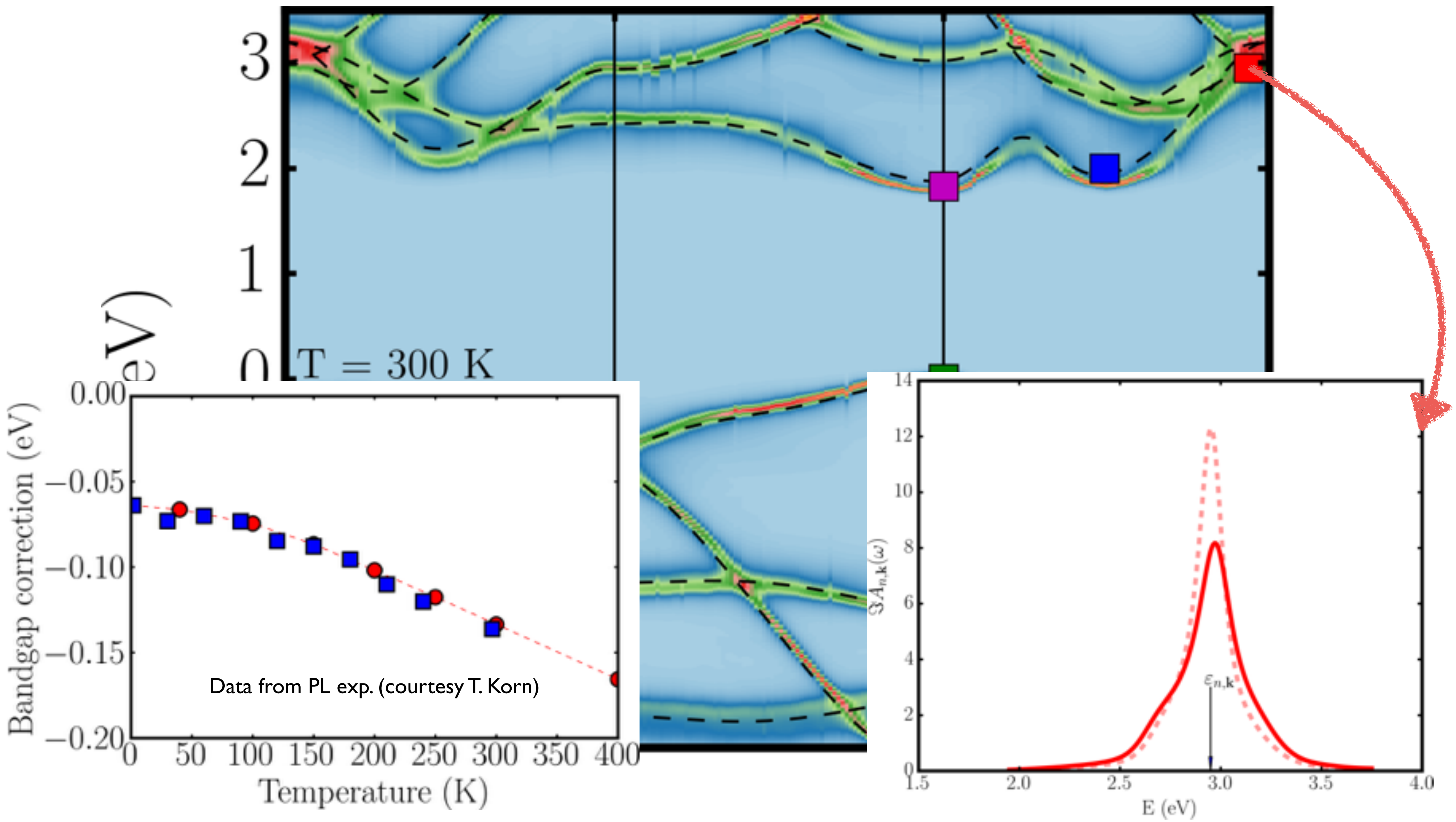


Bandgap renormalization

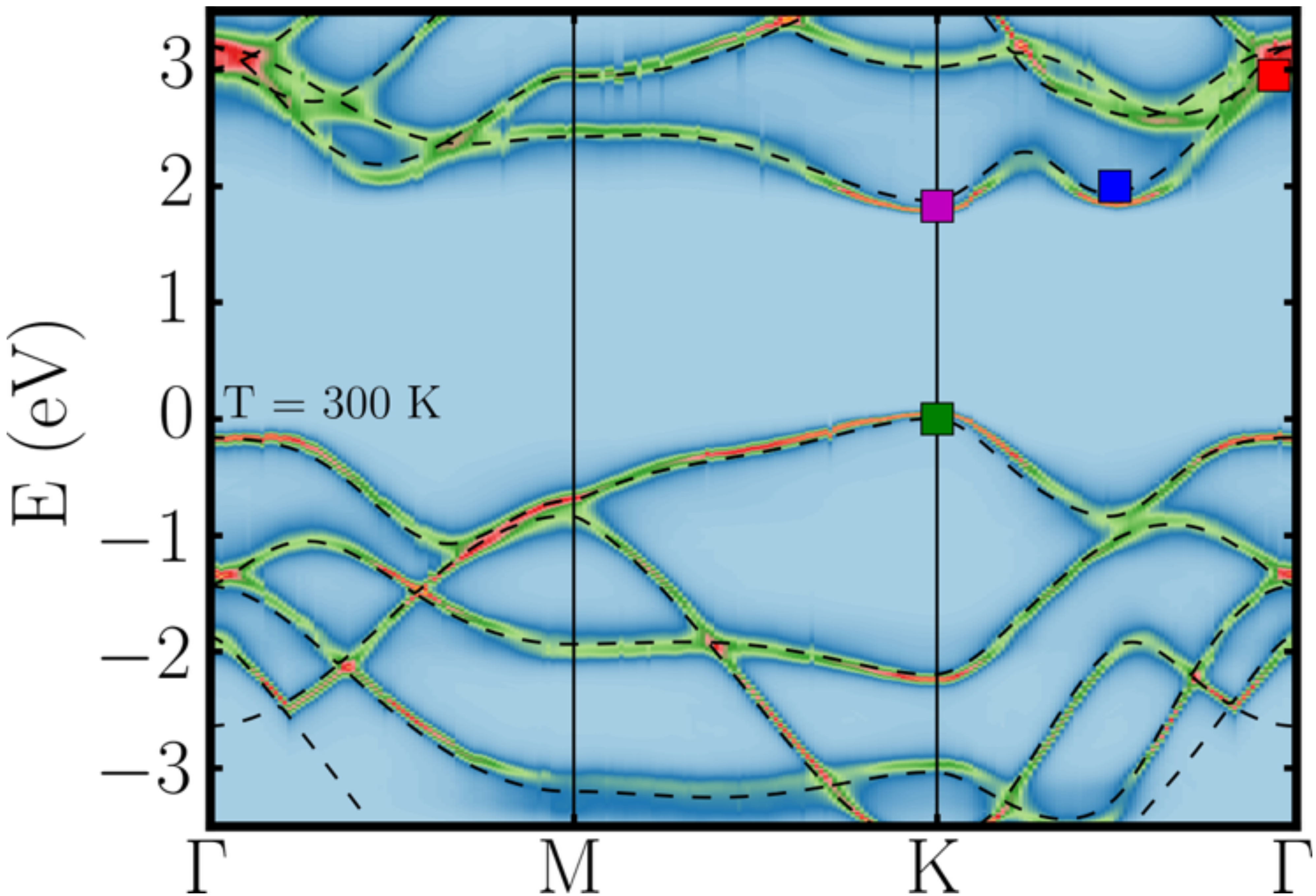
Single-layer MoS



Bandgap renormalization

Breakdown QP approx.

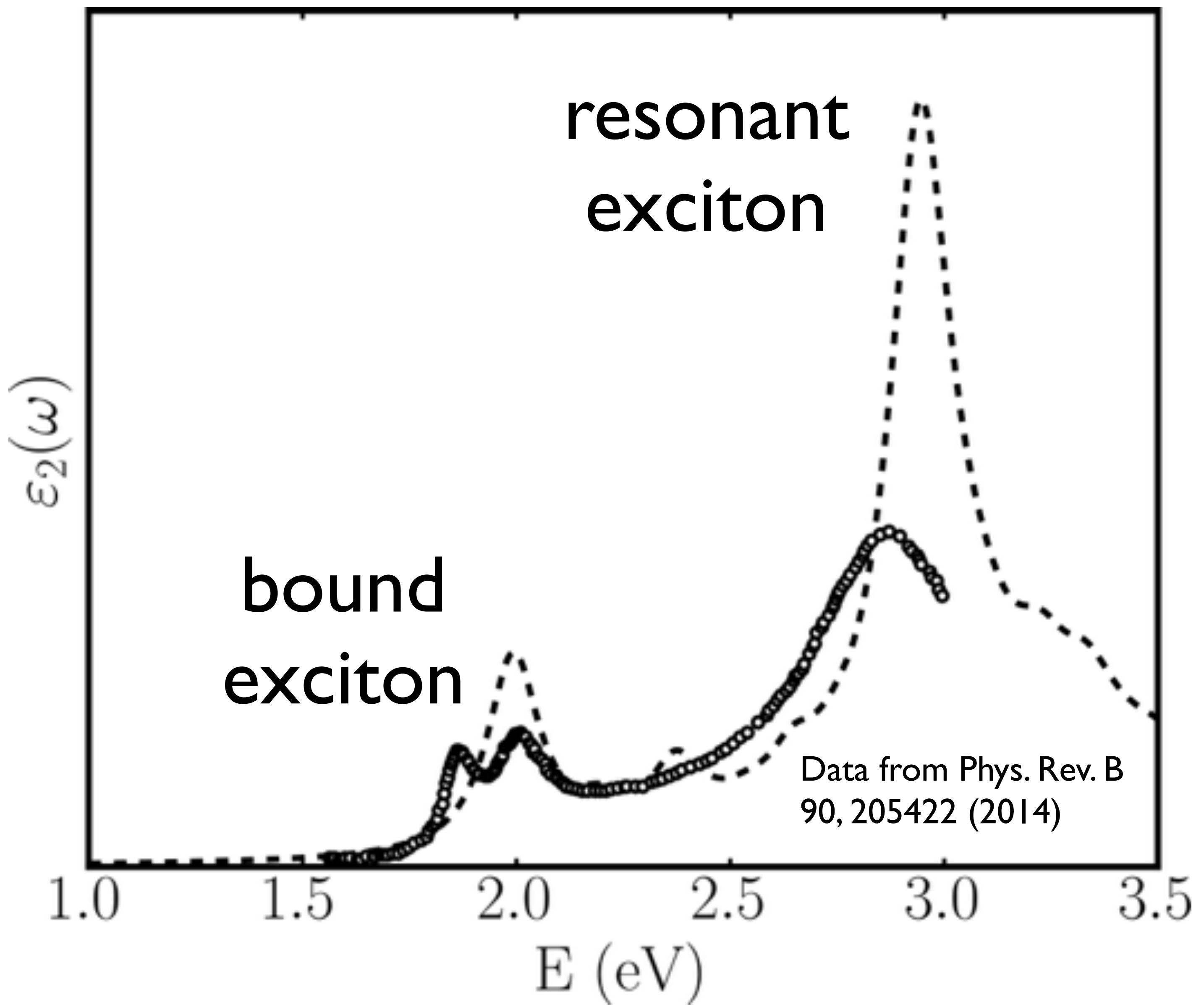
Single-layer MoS

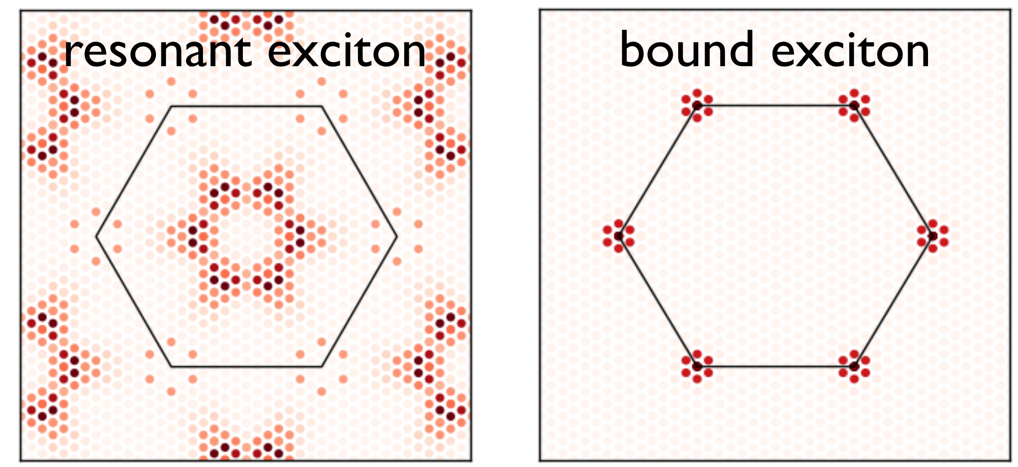


What about optical properties (excitons) ?

Excitons at finite temperature

$$H^{FA}_{ee',hh'} = (E_e - E_h)\delta_{eh,e'h'} + (f_e - f_h)\Xi_{ee',hh'}$$





Experiments show lines with different broadenings

Spectra usually obtained at room temperature

Homogeneous broadening for all the peaks

A. Molina-Sánchez et al., Phys. Rev. B 88, 045412 (2013)

Excitons at finite temperature

$$H_{ee',hh'}(T) = H^{FA}_{ee',hh'} + \left[\Delta E_e(T) - \Delta E_h(T)\right] \delta_{eh,e'h'}$$

Bethe-Salpeter Hamiltonian is not hermitian
Excitons acquire a finite lifetime

A. Marini, Physical Rev. Lett. 101, 106405 (2008)

Excitons at finite temperature

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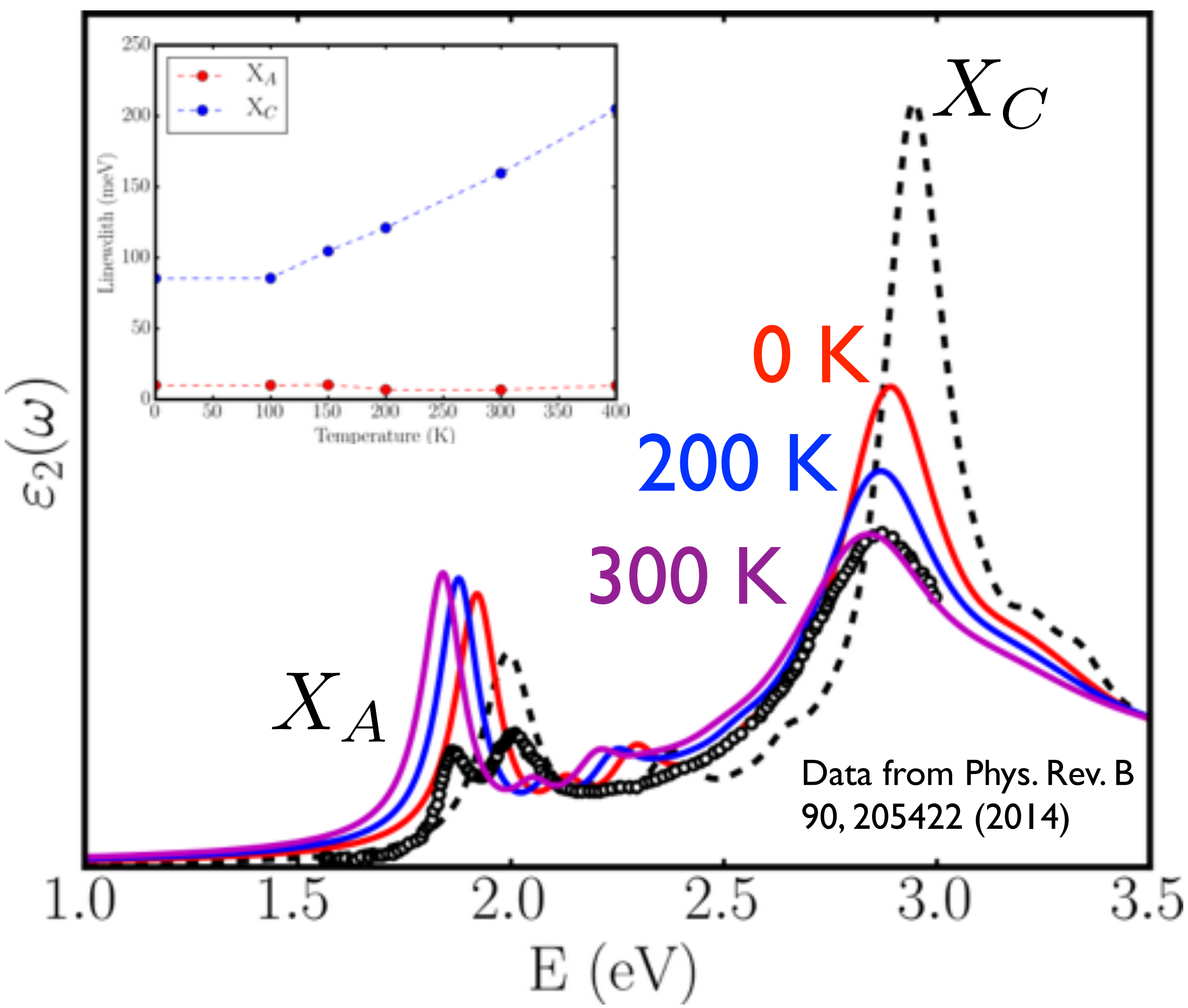
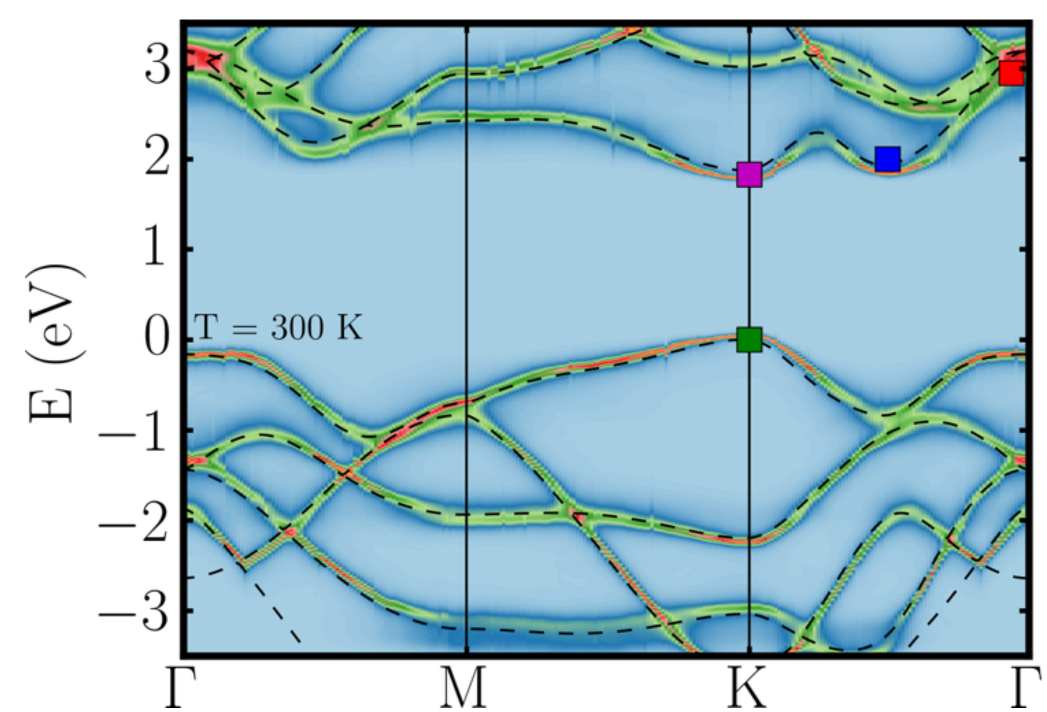




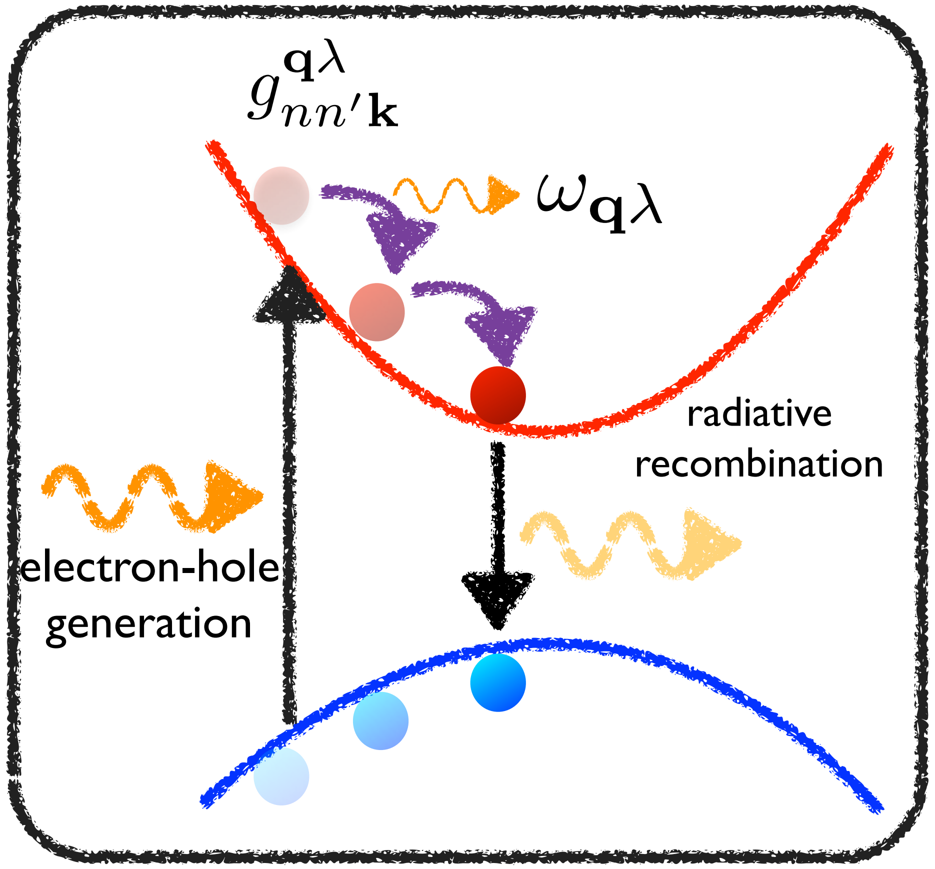
Conclusions and ongoing work

Temperature effects are important for an accurate bandgap value and for a realistic description of the bands

Breakdown of quasiparticle approximation

Lifetime of quasiparticles (excitons)

Carrier relaxation for ultra-fast optics



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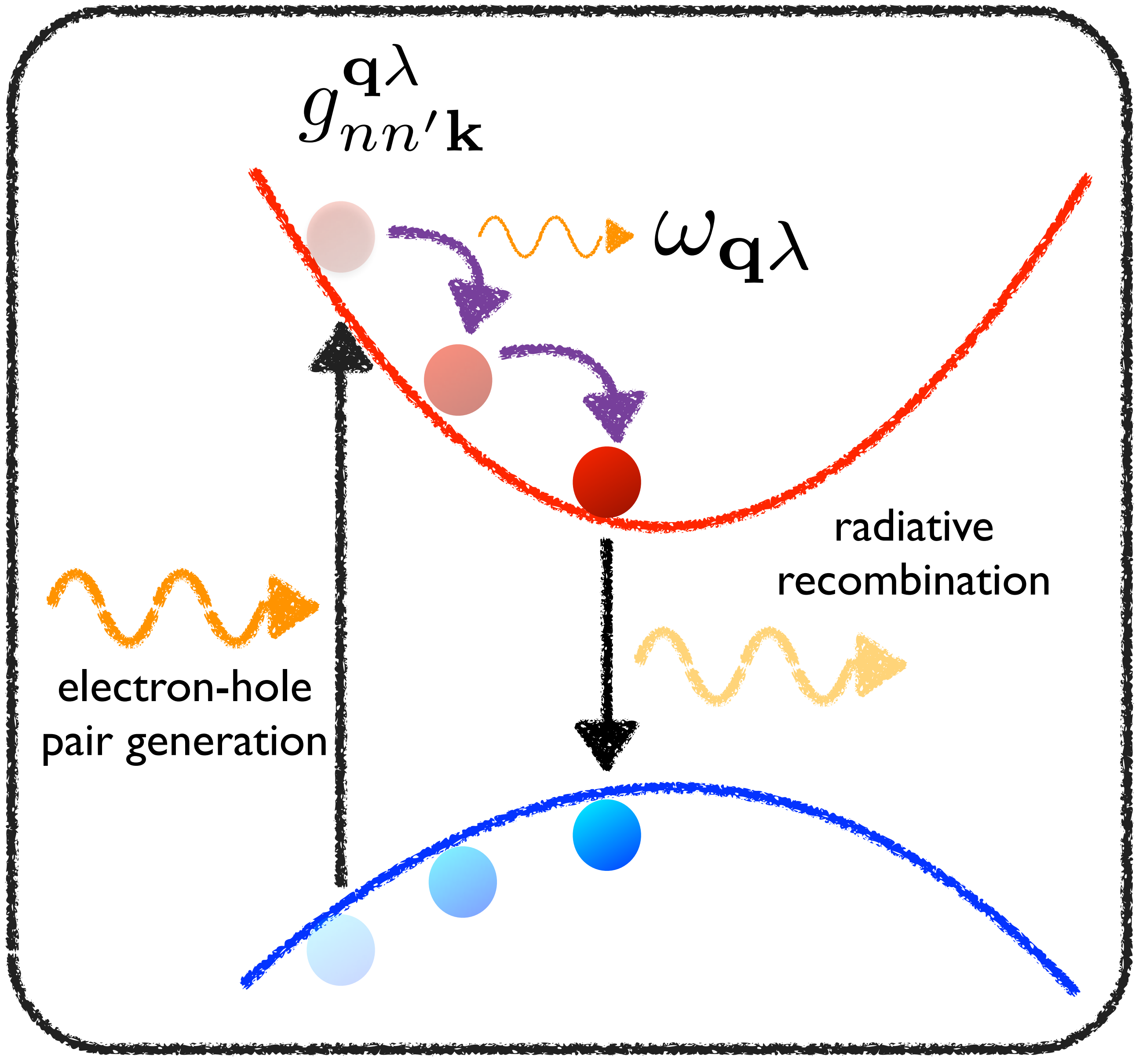
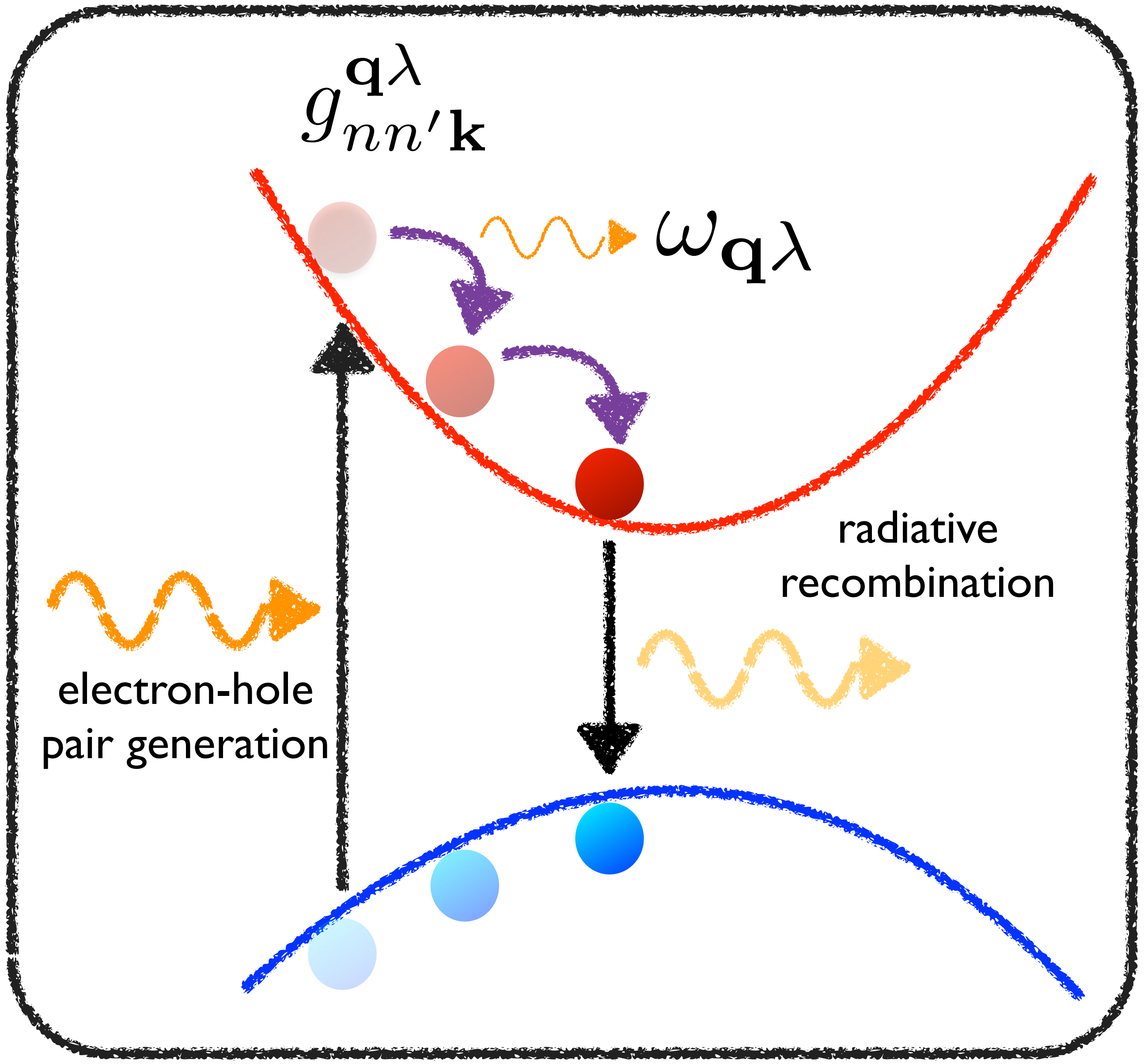
CNR Rome, Italy

Carrier relaxation for ultra-fast optics





$g_{nn'\mathbf{k}}^{\mathbf{q}\lambda}$
$\omega_{\mathbf{q}\lambda}$
radiative recombination
electron-hole pair generation