Challenges and prospects of first principles simulations of materials *out of equilibrium*

Vojtech Vlcek

Workshop for AI-Powered Materials Discovery — June 24, 2025

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Motivation 1: *driven* materials properties

- **Transient/Driven states**: Floquet-Bloch, IMT/MIT, magnetic ordering, exciton-magnon coupling …

Mahmood et al., Nature Physics 12, 306 (2016)
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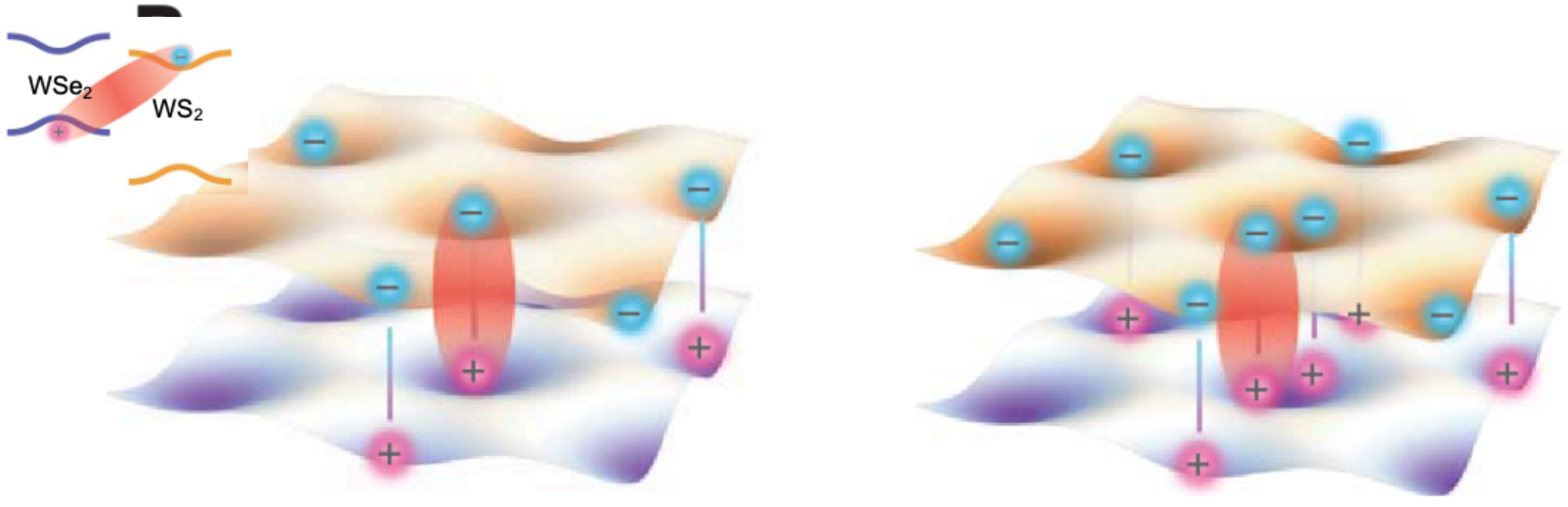
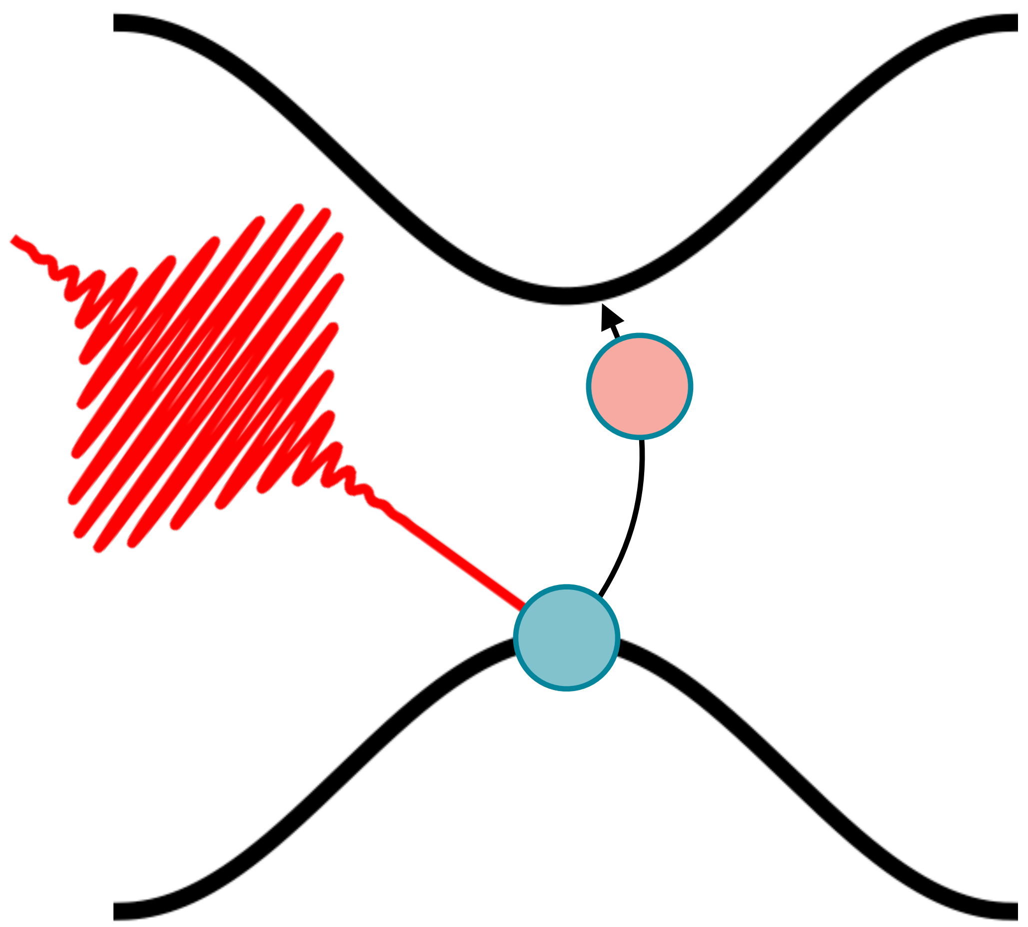


Xiong, et al. Science 380, 860 (2023)

- Diverse **applications of transient properties**: on-chip photonic computation, condensate-based optoelectronics, exciton-polariton mediated high-T superconductivity, & QIS…
  - FET (100GHz) ⇒Optical: THz ⇒PHz



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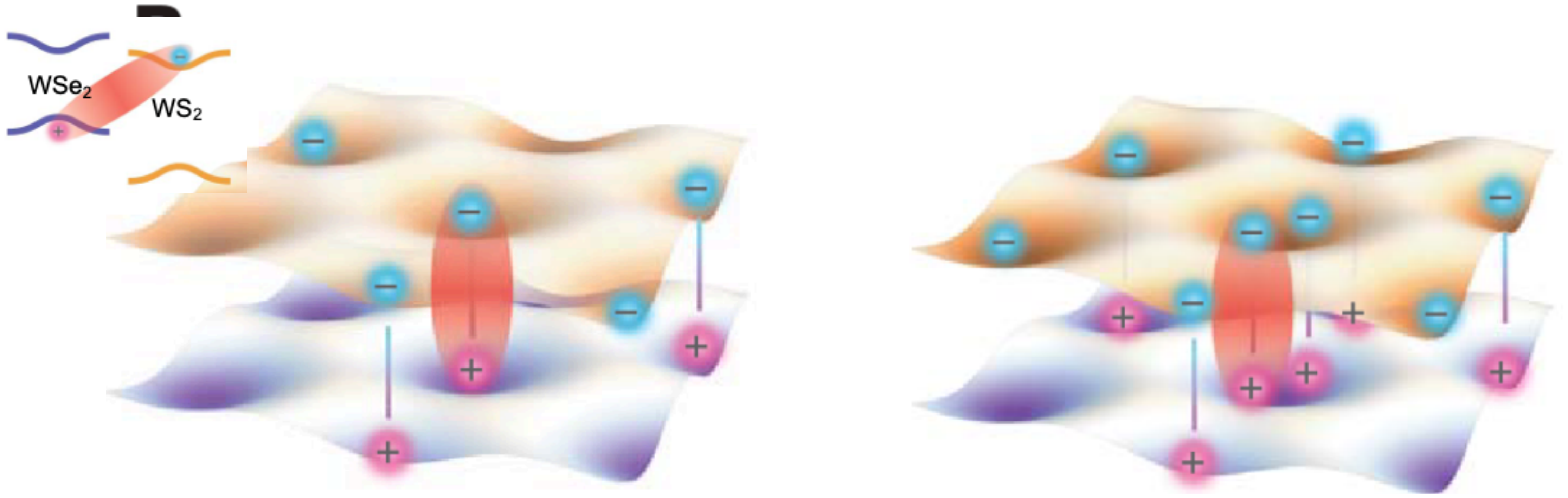

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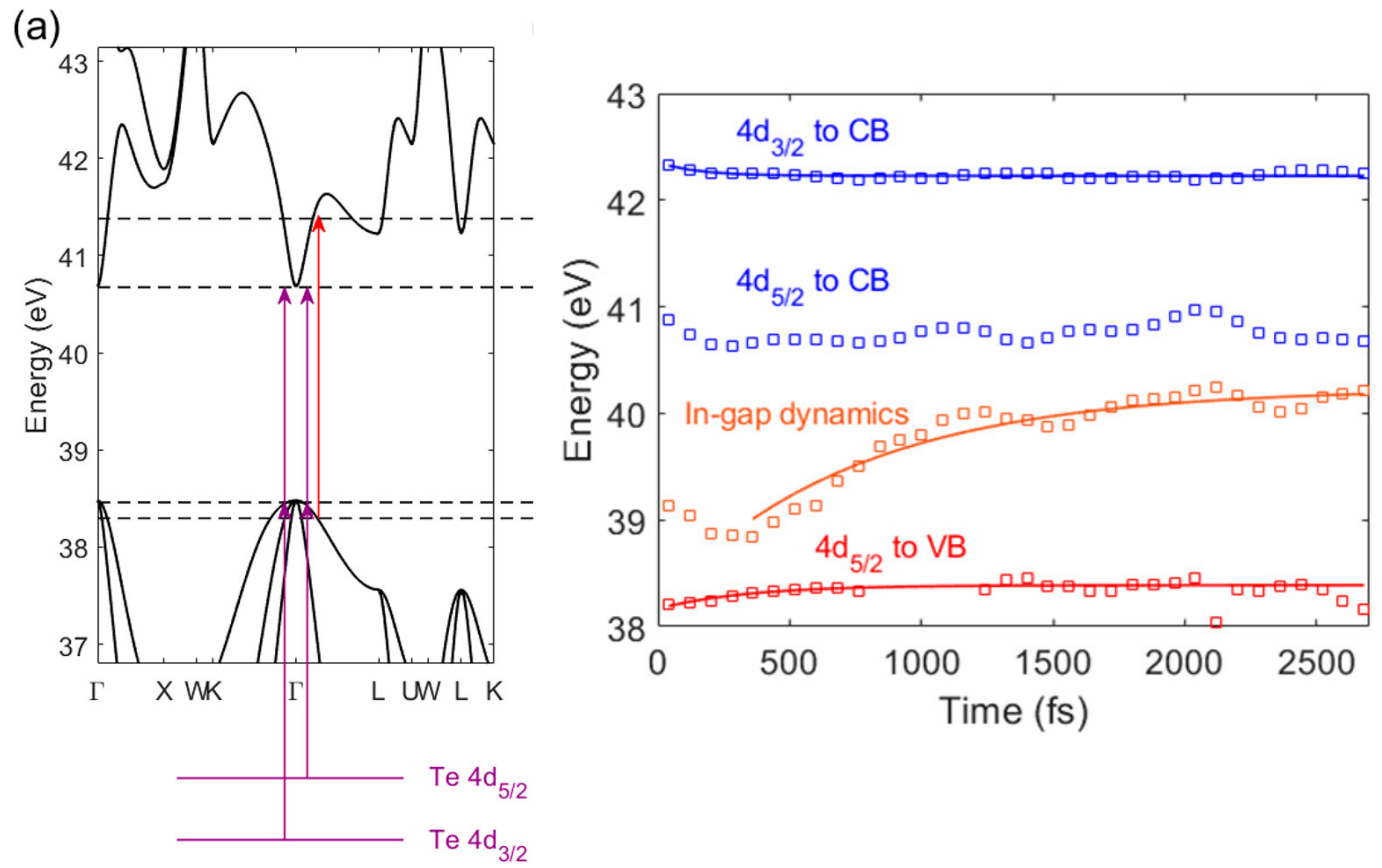

H. Liu et al. J. Phys. Chem. Lett., 14, 2106 (2023)

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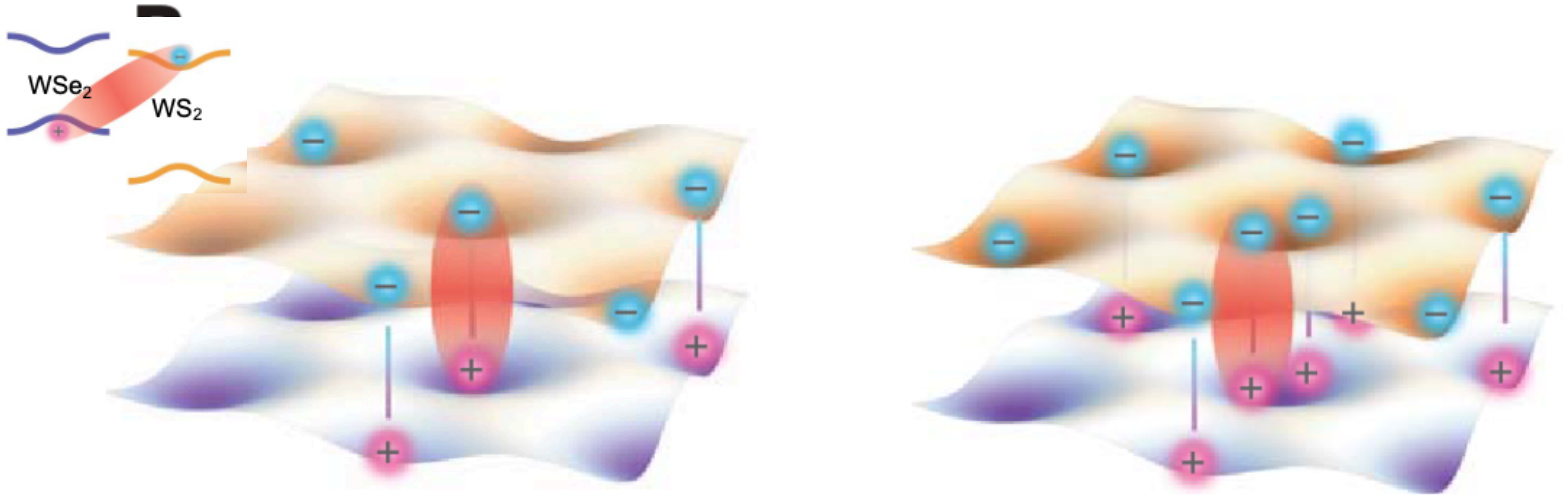

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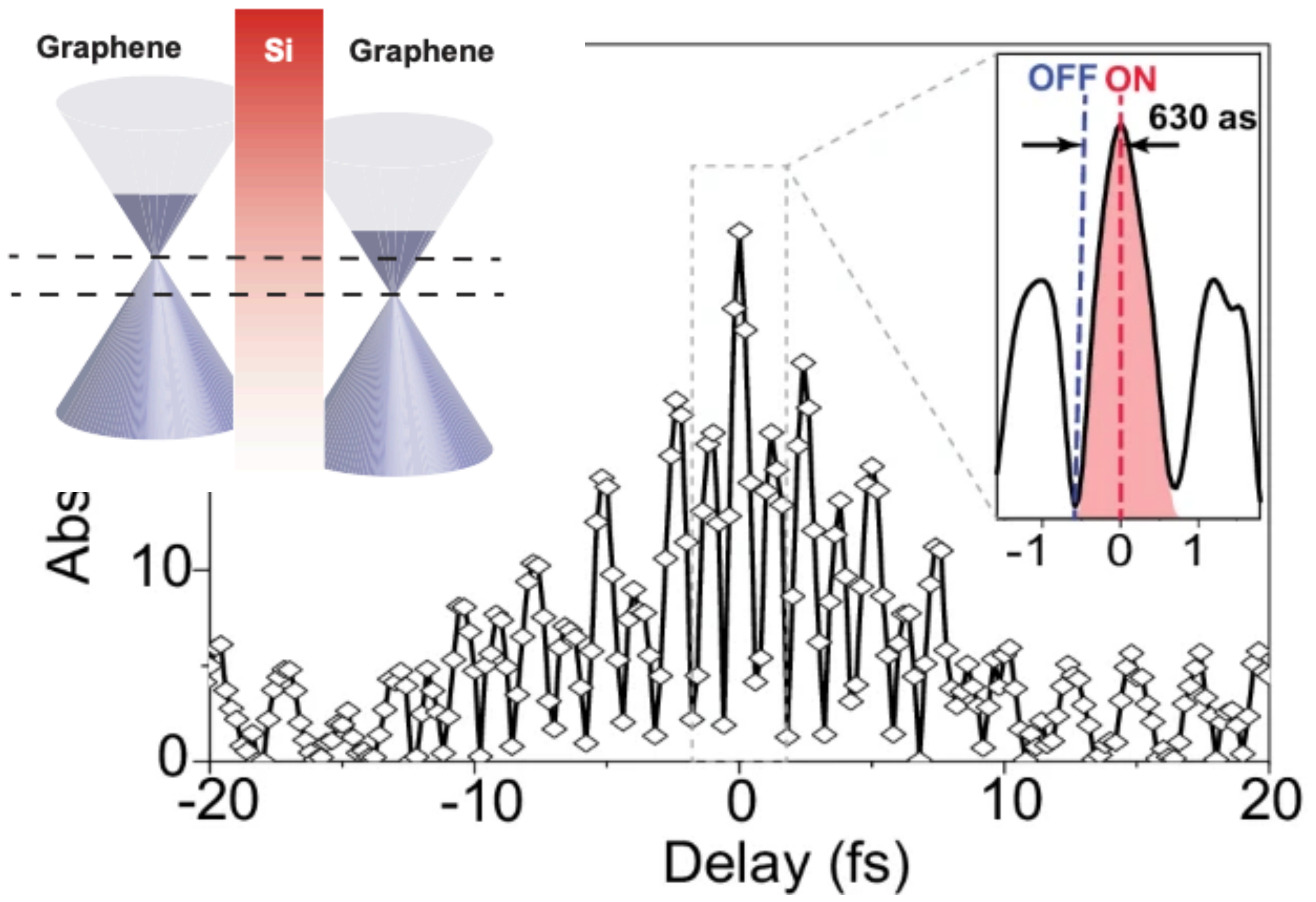

Sennary et al., Nature Comm,16, 4335 (2025)

Motivation 2: witnessing electronic structure dynamics

G Inzani and M Lucchini, J. Phys. Photonics 7 022001 (2025)

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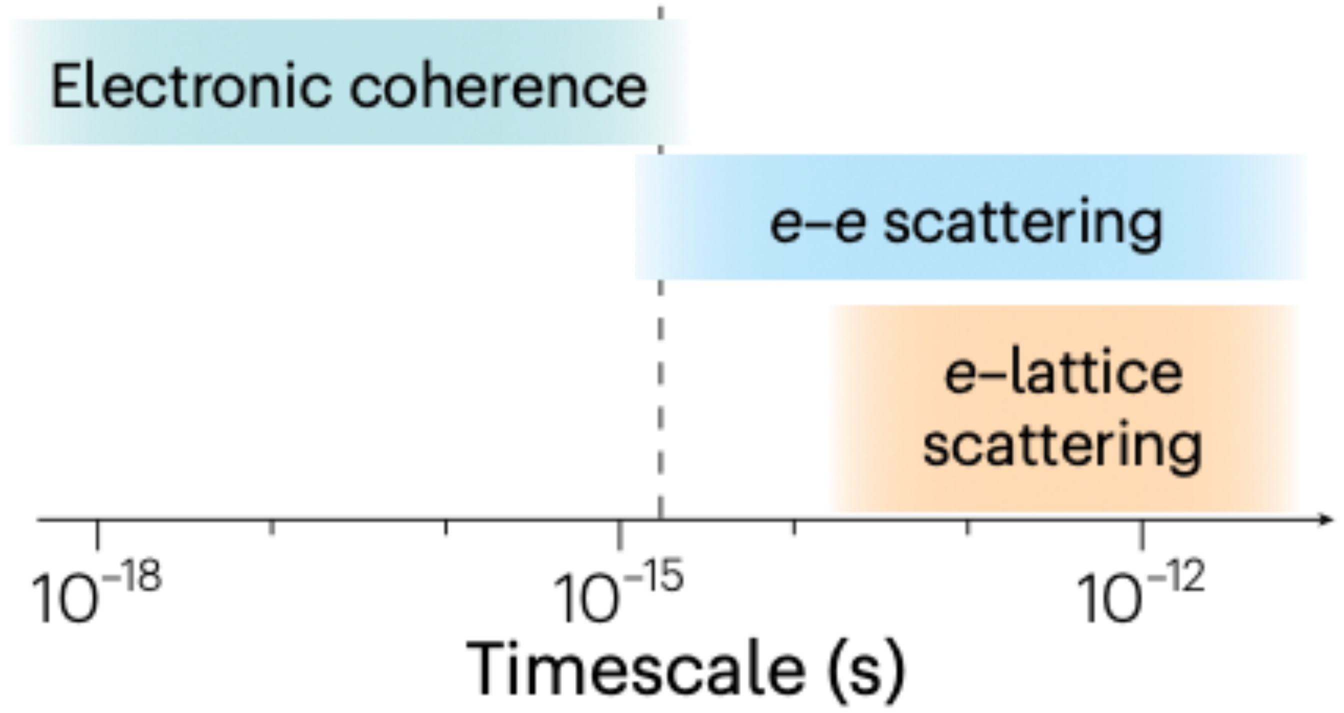

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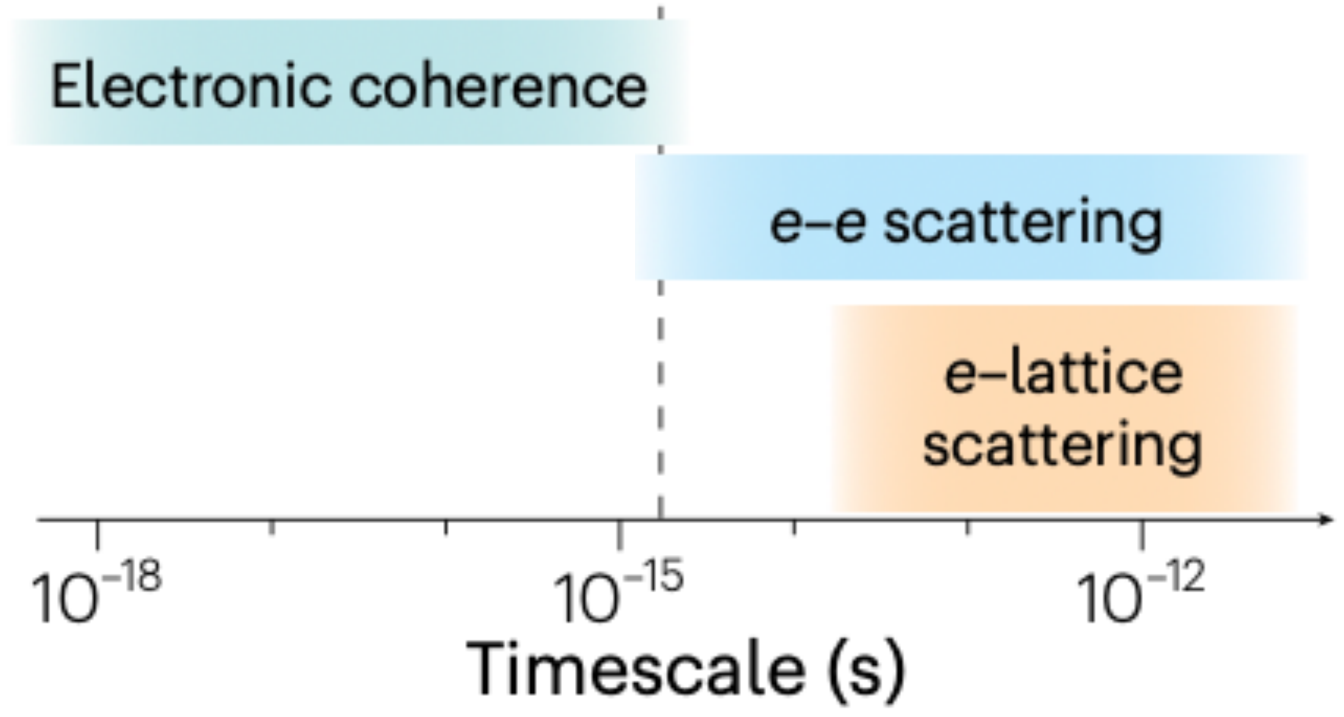

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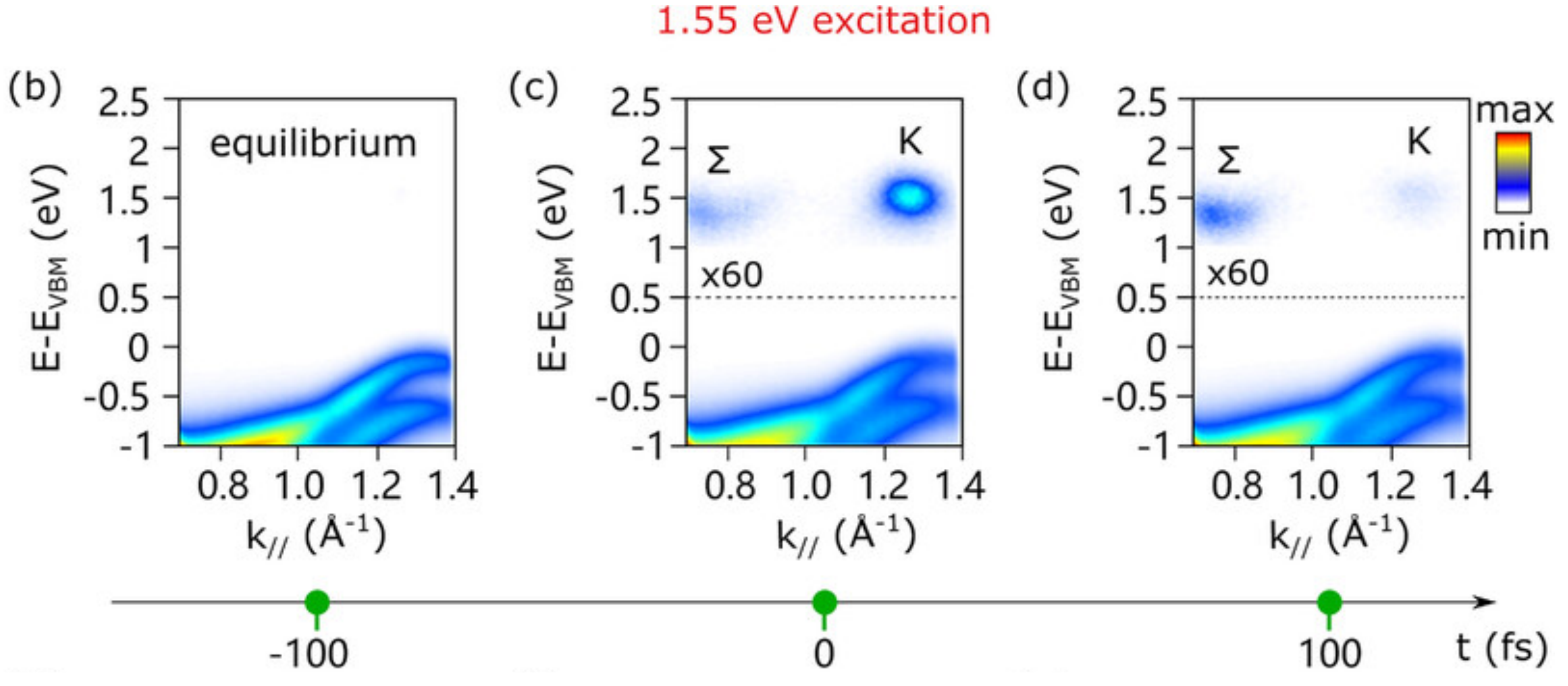

Dong , Puppin et al. Natural sciences (2021)

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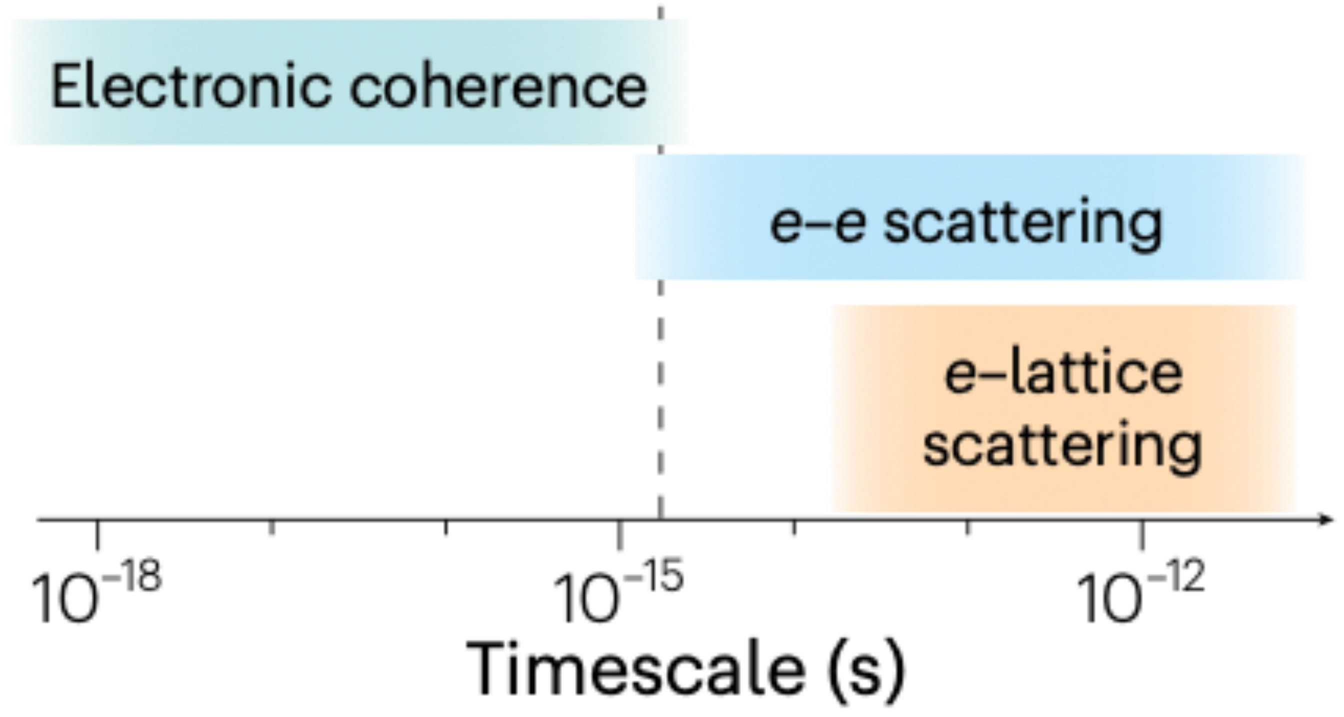

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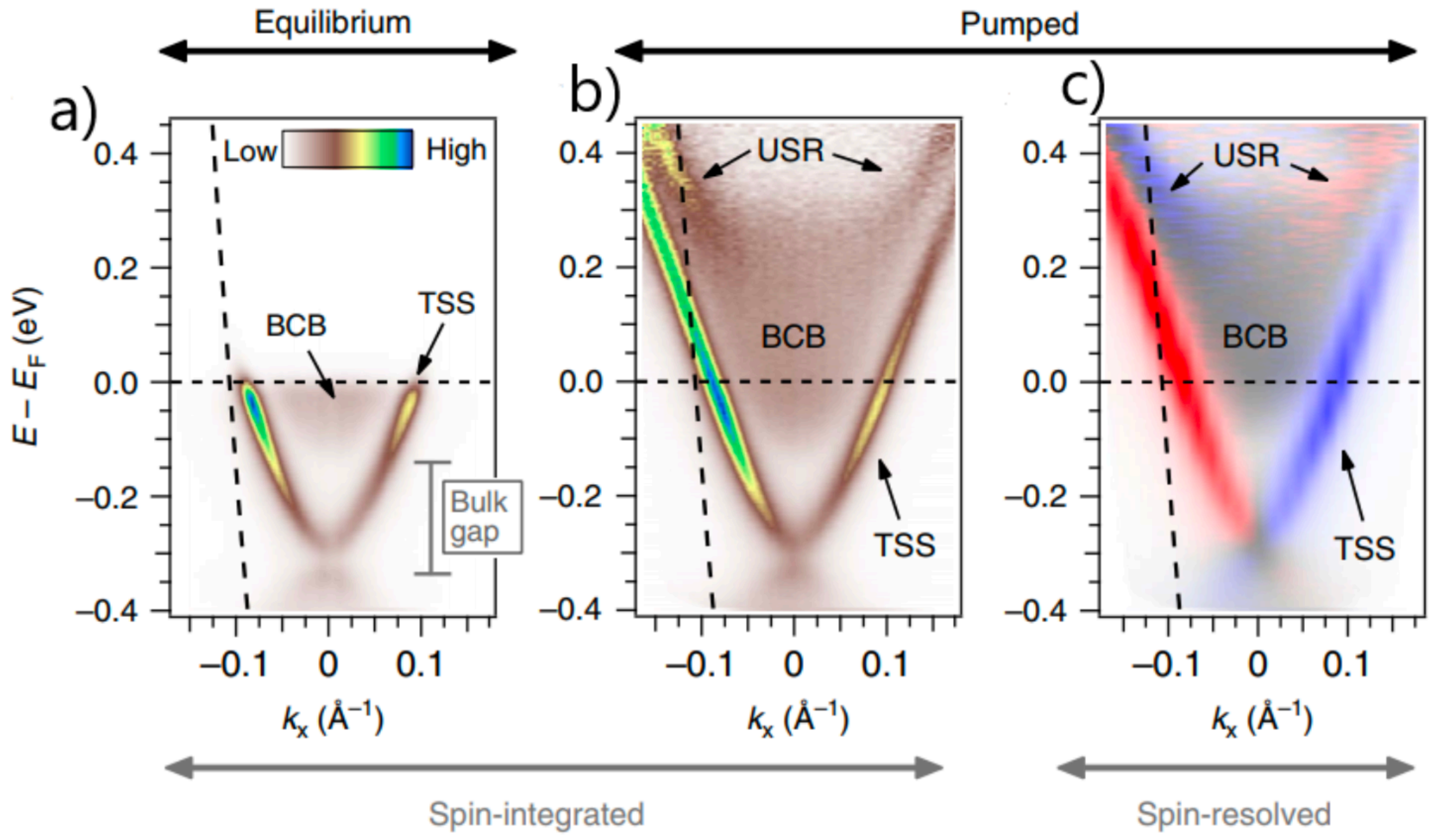

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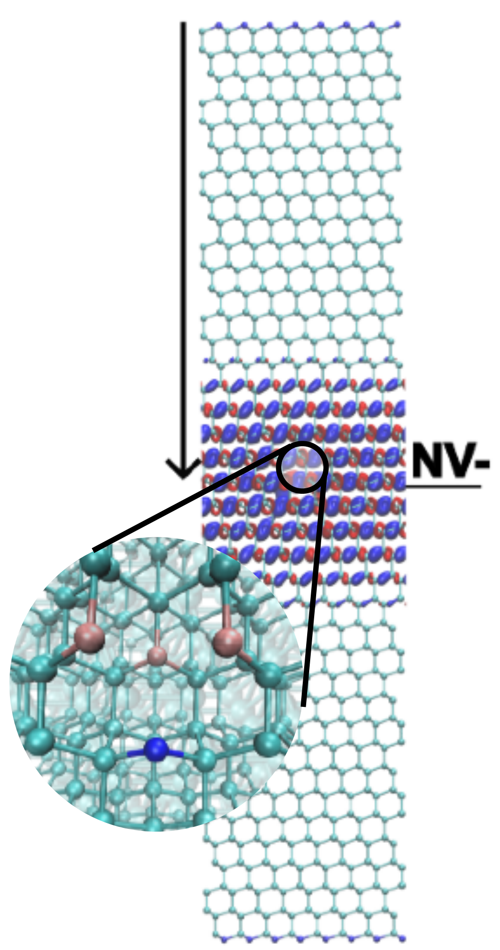
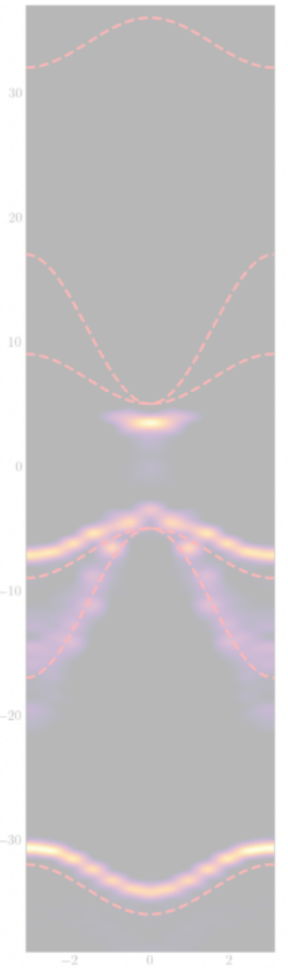
First Principles Materials Simulations - challenges (& prospects)

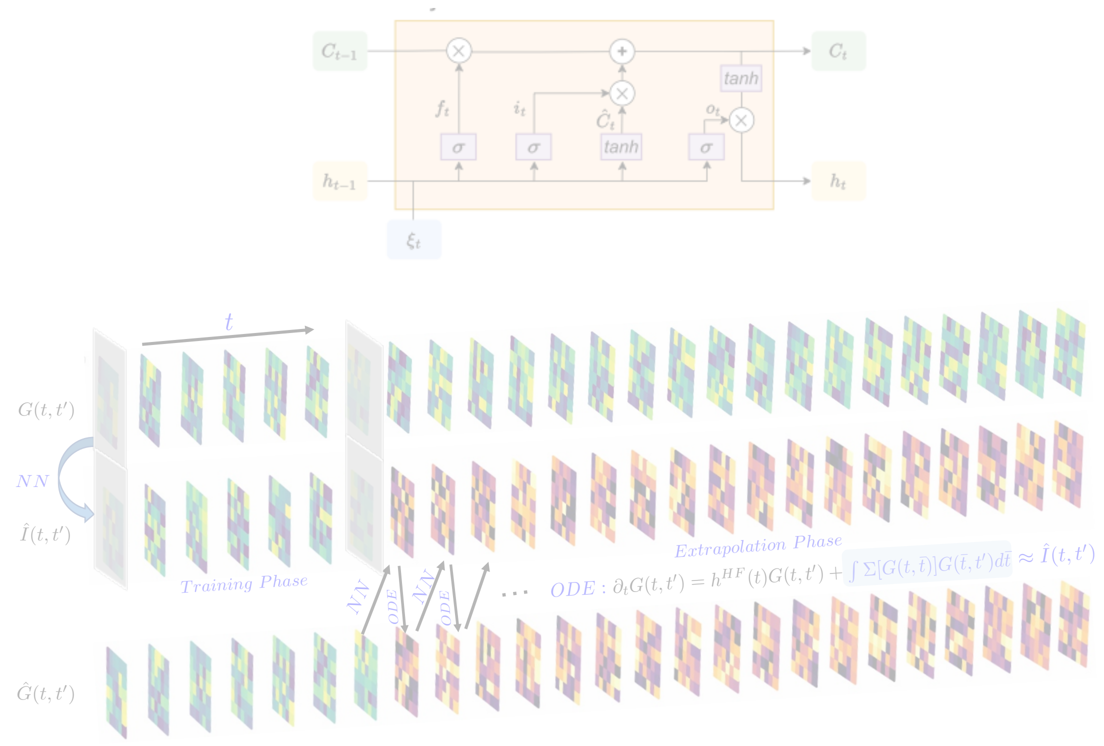
Inspirational tale: equilibrium



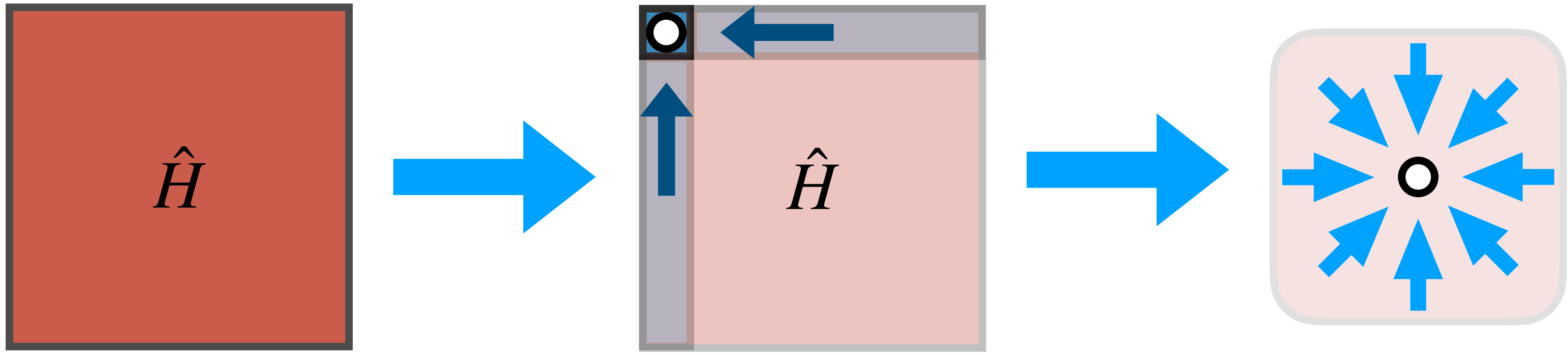
Extending current frameworks to non-equilibrium



Opportunities for *new* approaches



Theory Framework - *complexity reduction*



- direct evolution of observables, i.e., correlation functions
  - One body space-time correlator

    ⇔ (S)TARPES
  - Two-body correlators and observables

    ⇔ particle-hole, i.e., optical (contracted)

    ⇔ particle-particle in 2E ARPES

"Complexity conservation"

large dimensionality

⇩

downfolded energy/time dependence

Efficient Implementation of Equilibrium Simulations:

- Scaling Up Many-Body Calculations:

— ***intrinsic vs apparent complexity (Kolmogorov)***

  - Exploiting compression techniques:

adapted from: John, arXiv:2102.06968 (2021)

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    — response functions and basis
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- ⇒ *Excited state calculations for systems with thousands of electrons*

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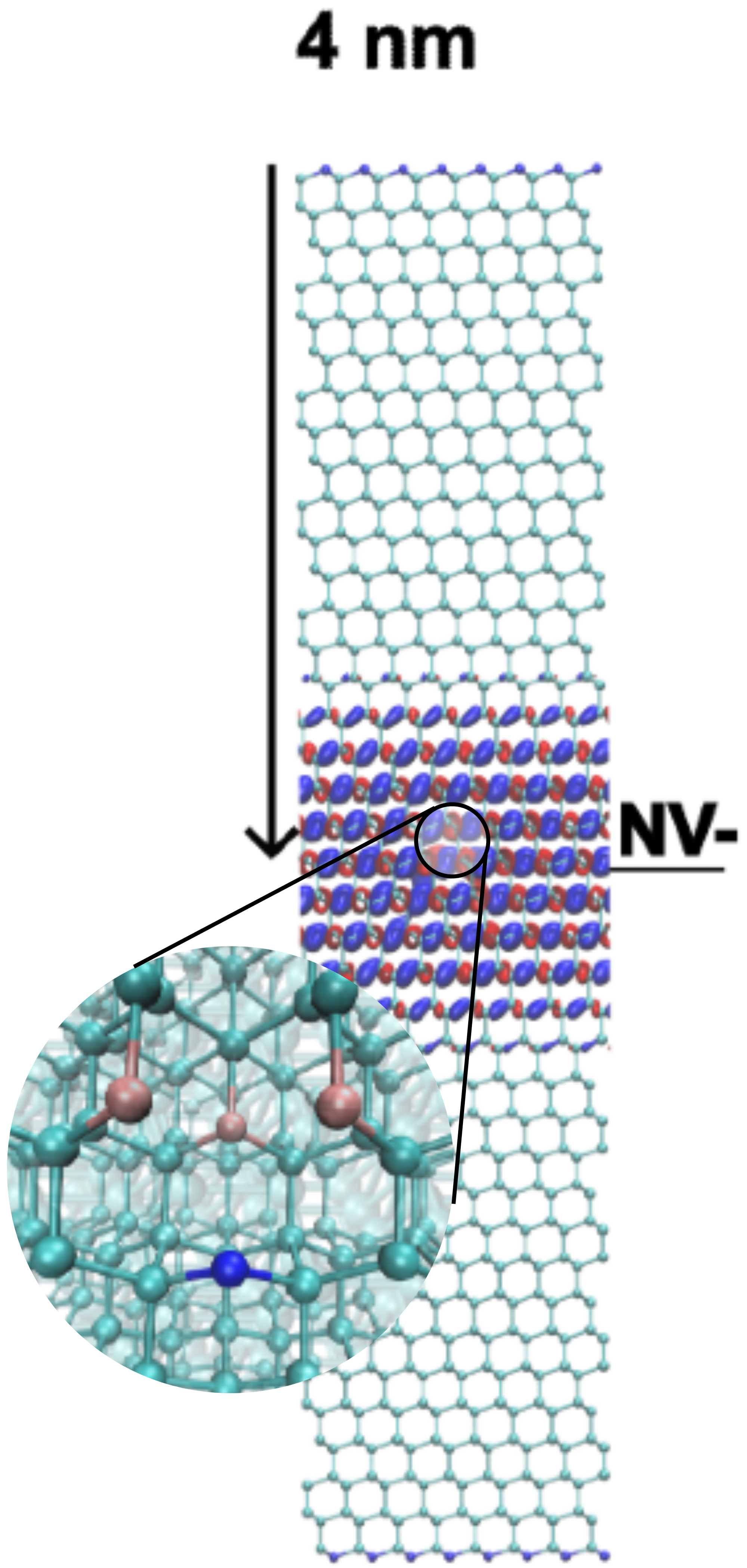

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Apelian, Canestraight, Liu, VV, Nano Letters 24 (38), 11882 (2024)
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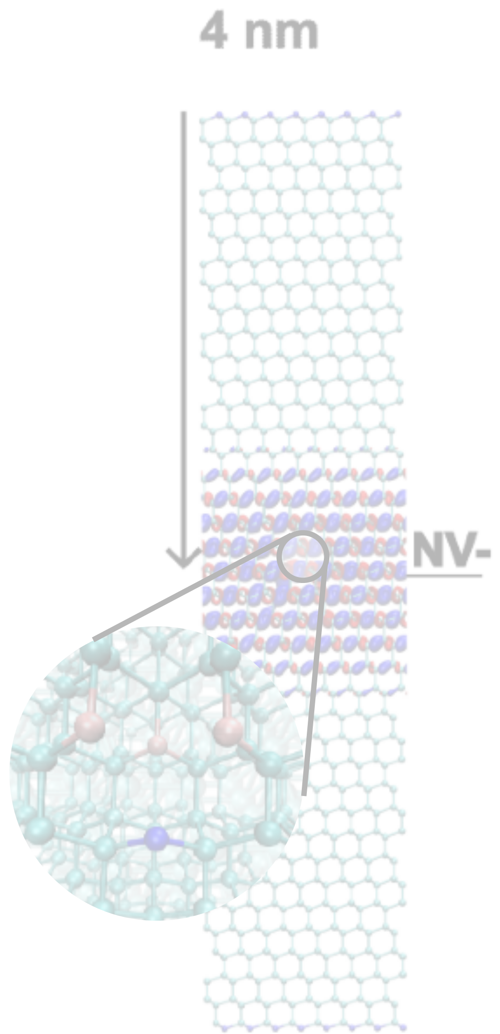

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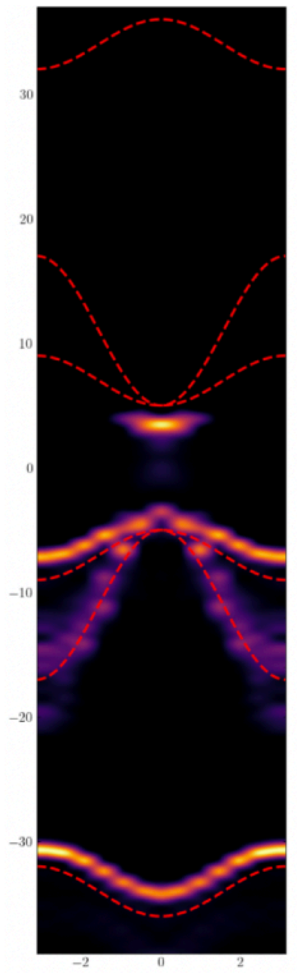

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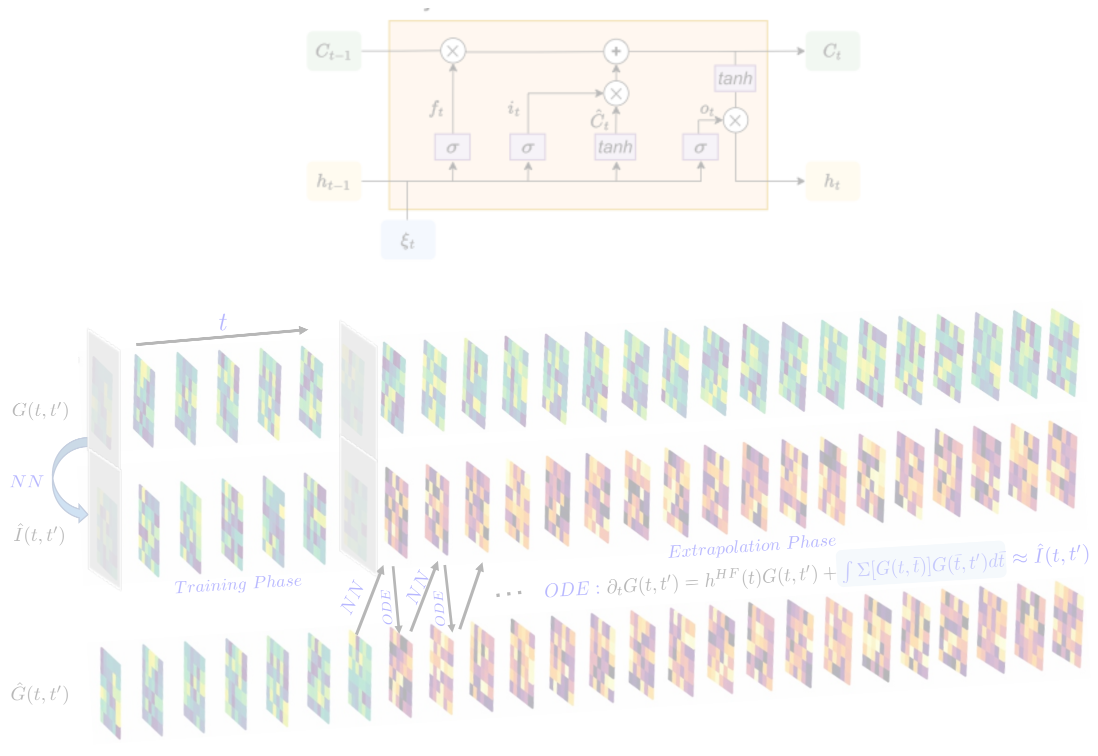
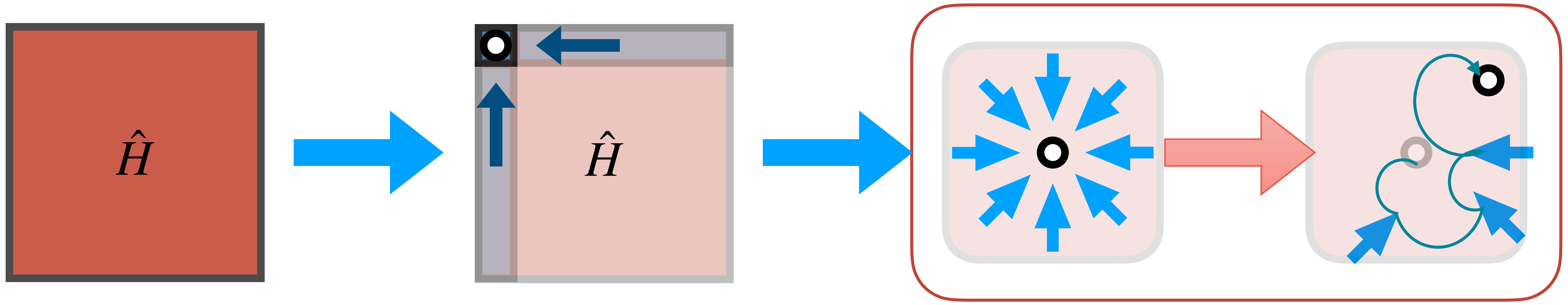


Non-equilibrium theory extension and its challenges



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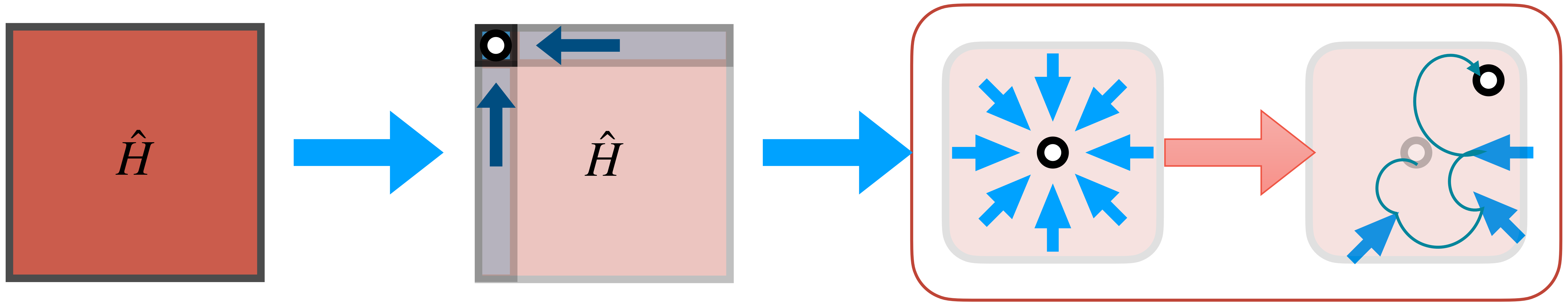

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- Information about *time* and energy $\Rightarrow$ *A tale of* ***two times***

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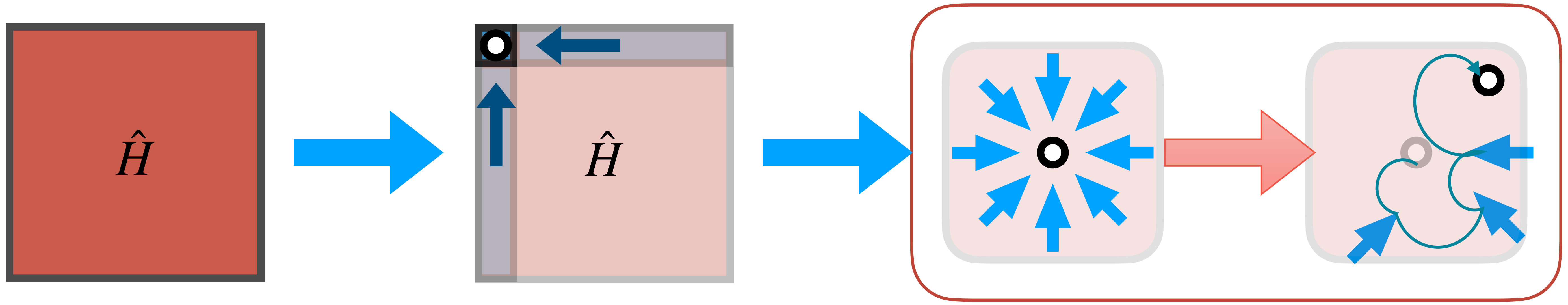

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  - Property of Interest (Observables & Correlators)
  - Intrinsic Interaction Strength
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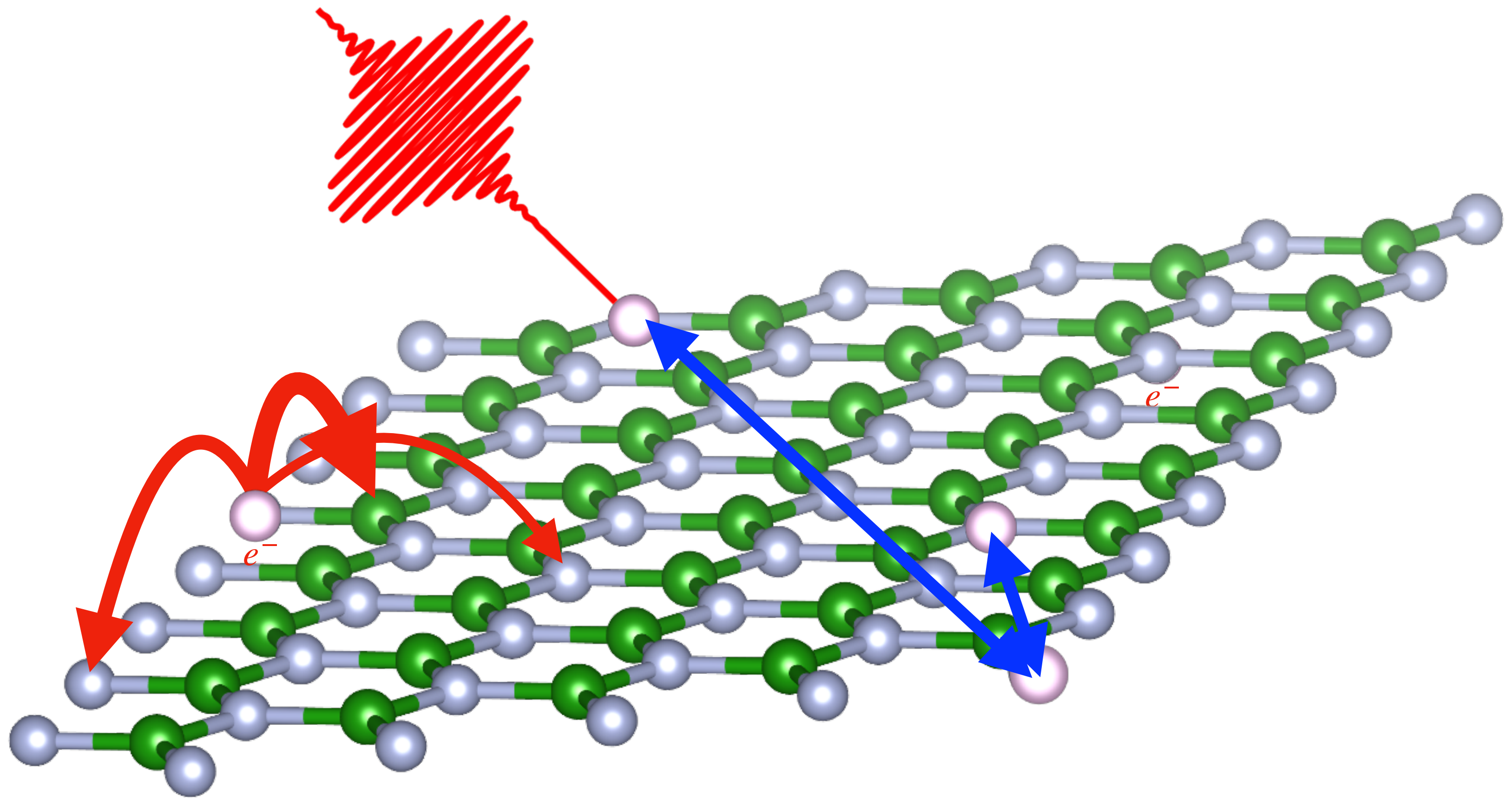

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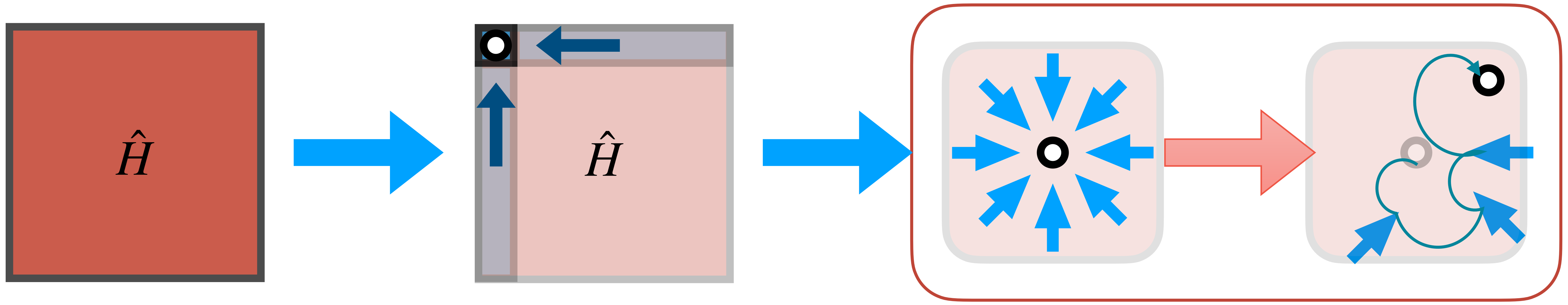

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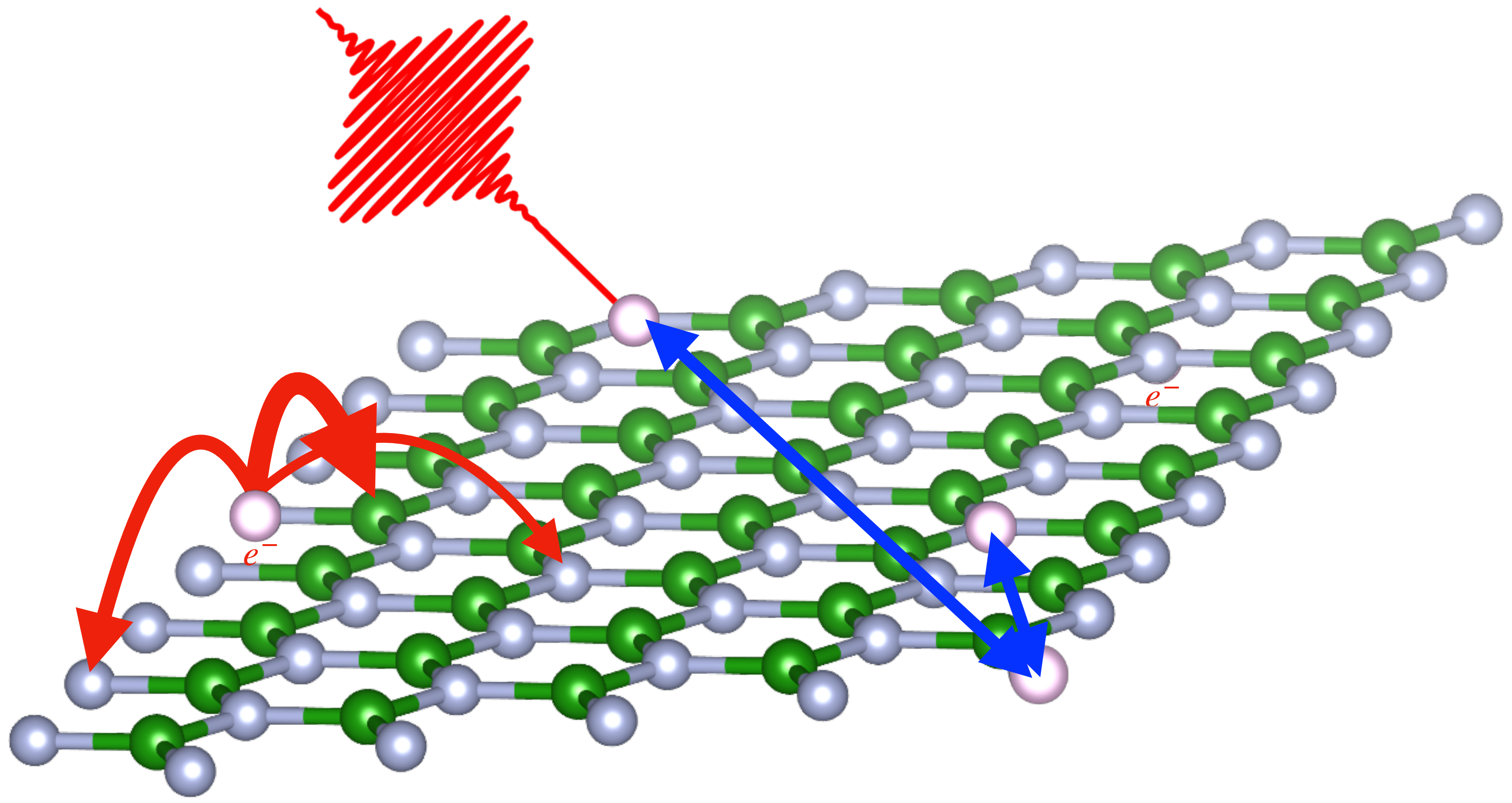

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Time-resolved photoemission spectra — “*two time*” correlations

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  - Spectra directly depend on **two times**

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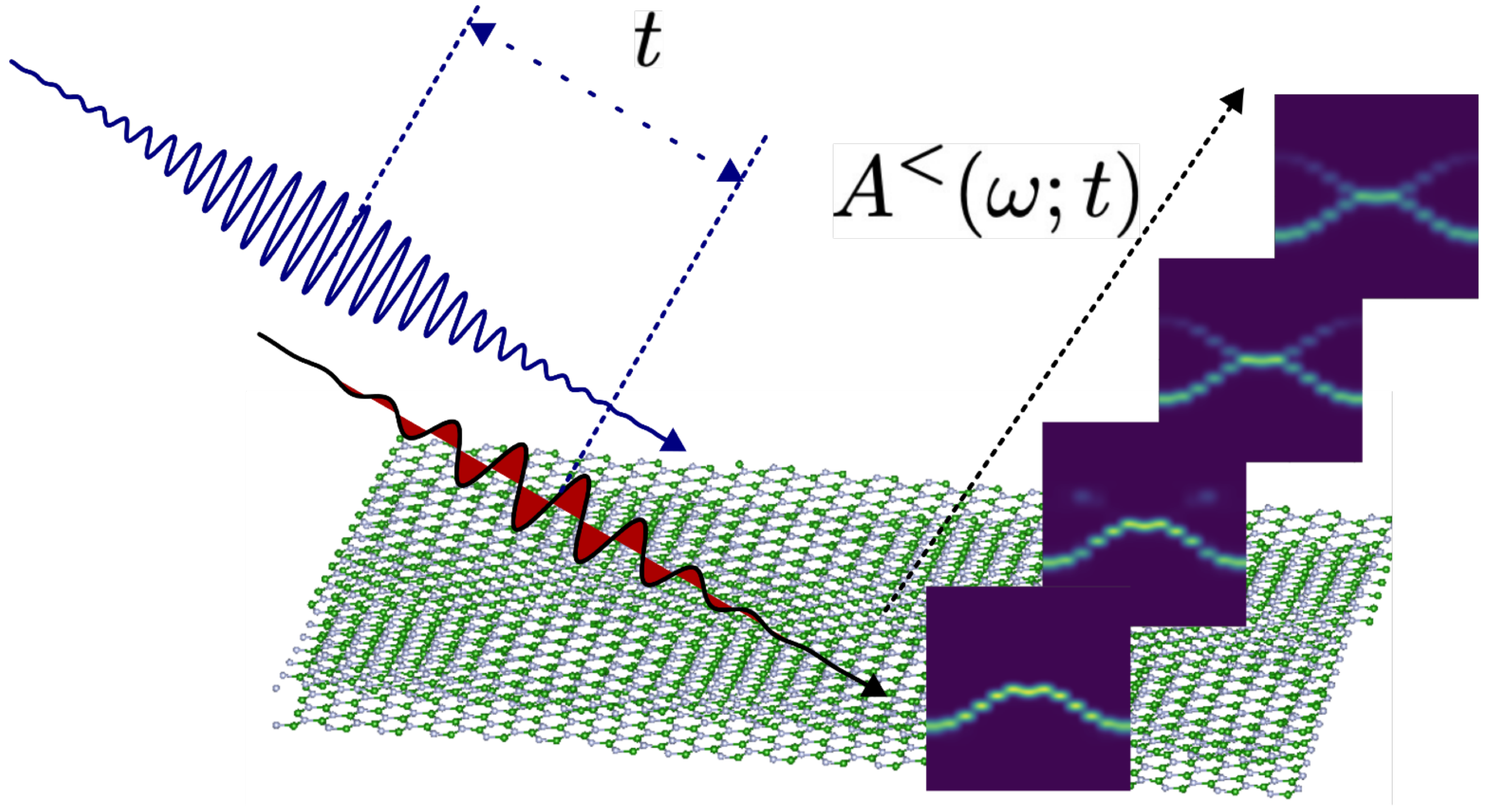

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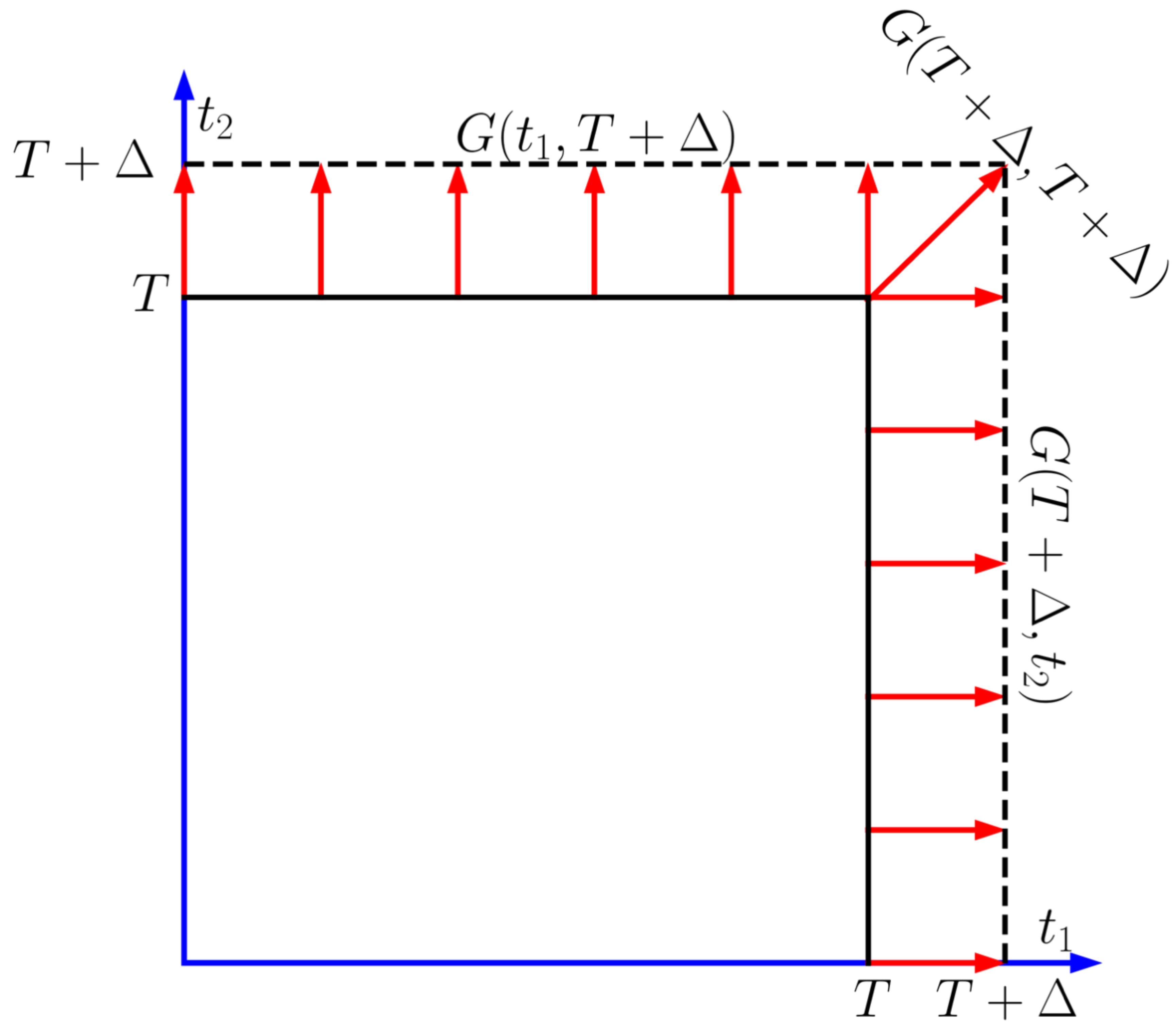

Lipavsky, Spicka, Velicky Phys. Rev. B 34, 6933 (1986)

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Joost, Schlunzen, Bonitz Phys. Rev. B 101, 245101 (2020)

Perfetto, Pavlyukh, Stefanucci Phys. Rev. Lett. 128, 016801 (2022)

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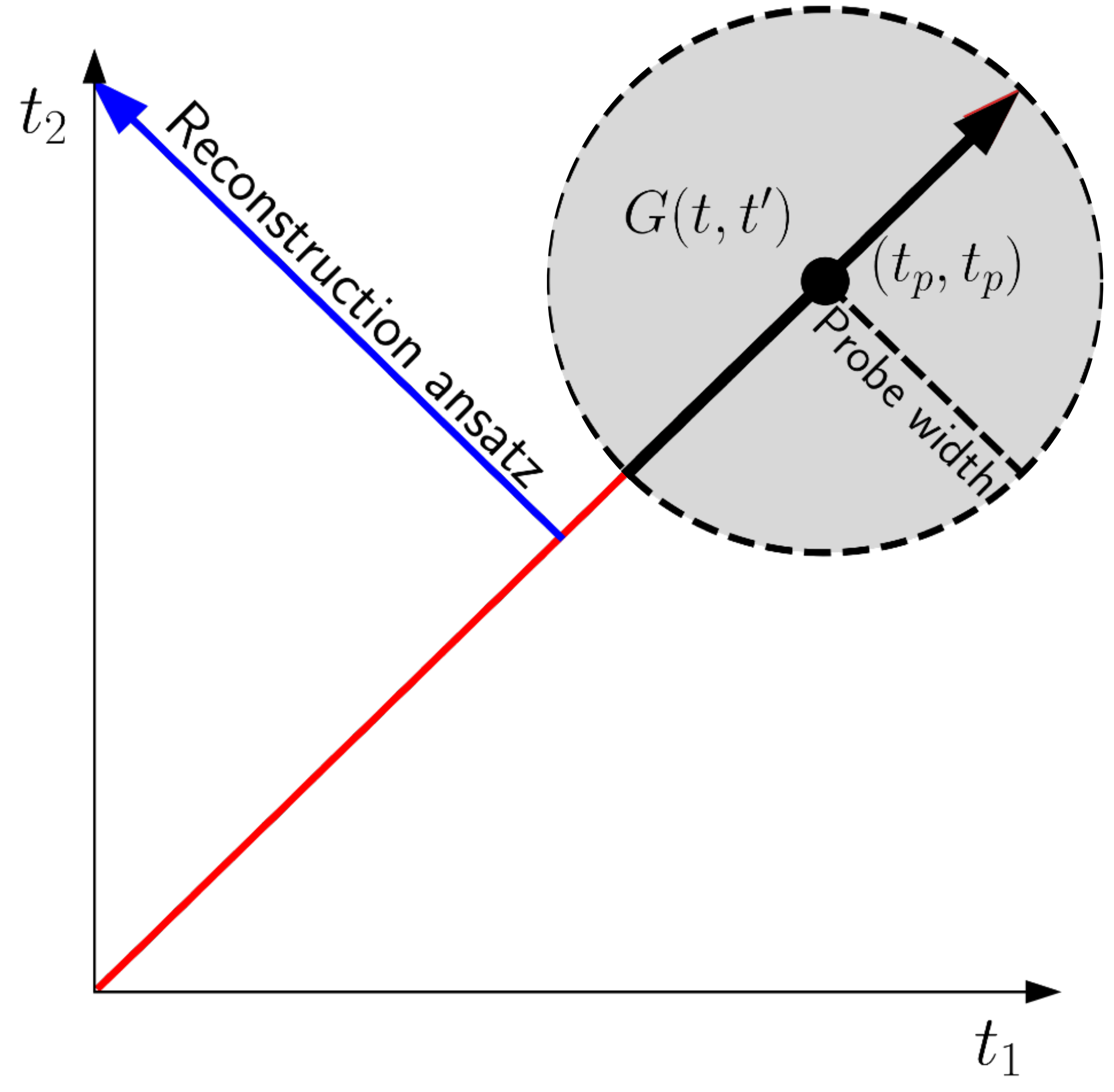

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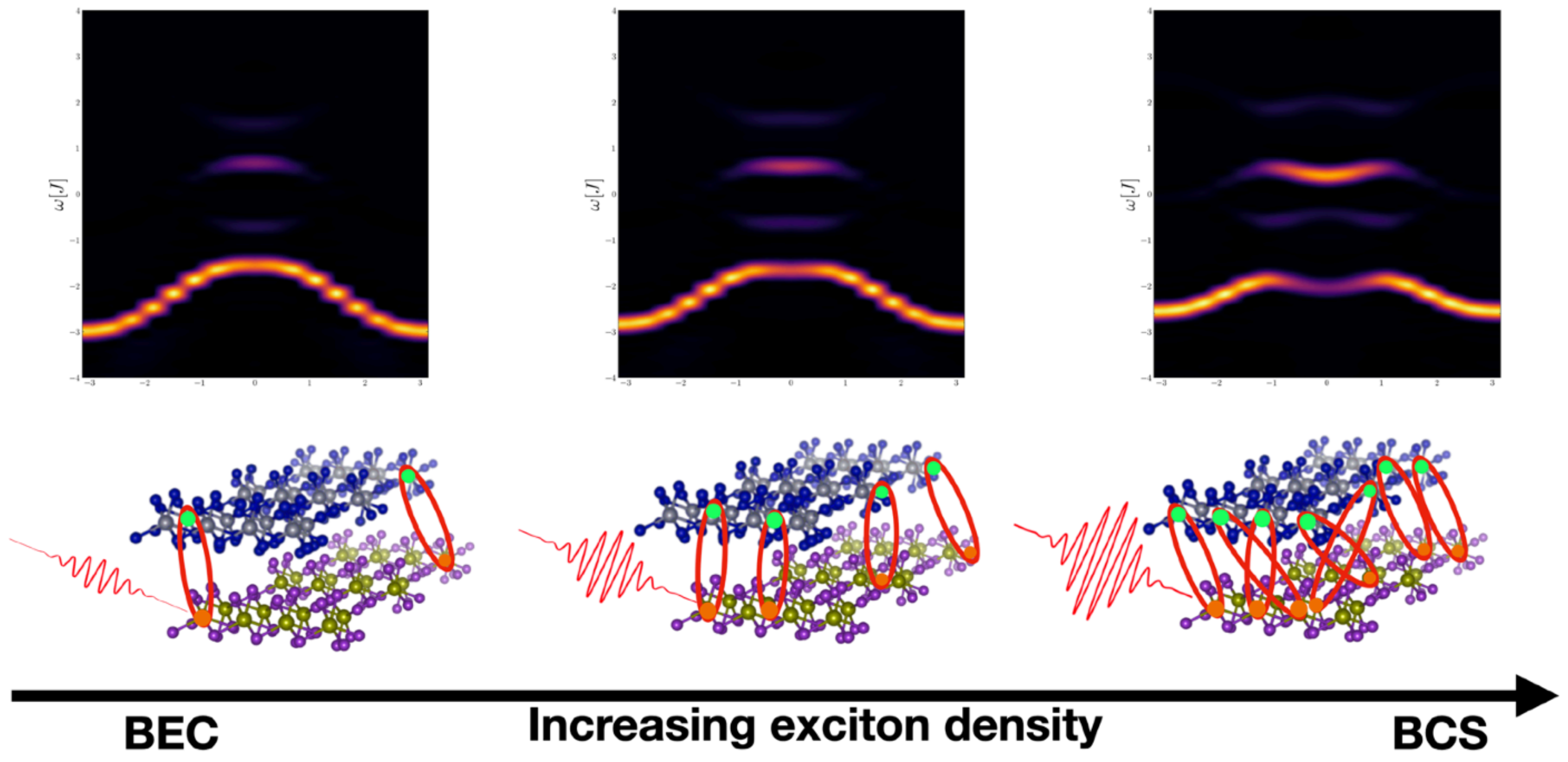

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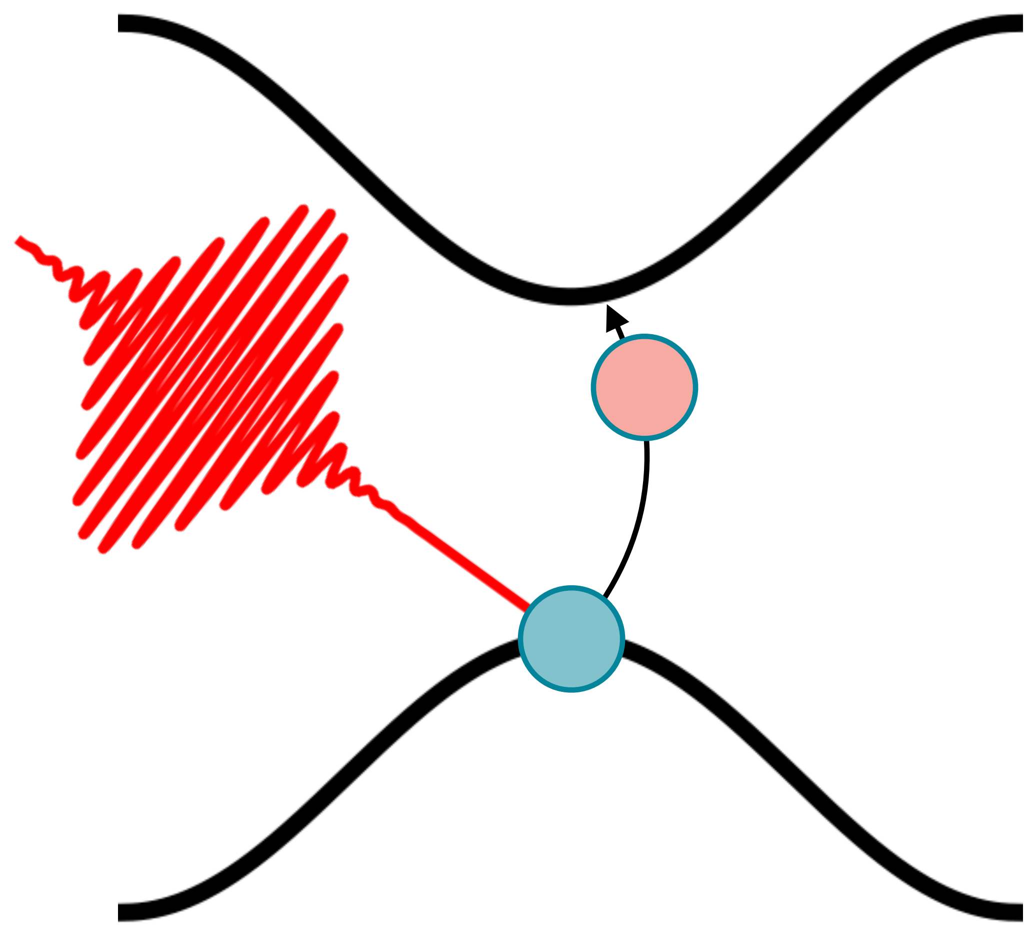

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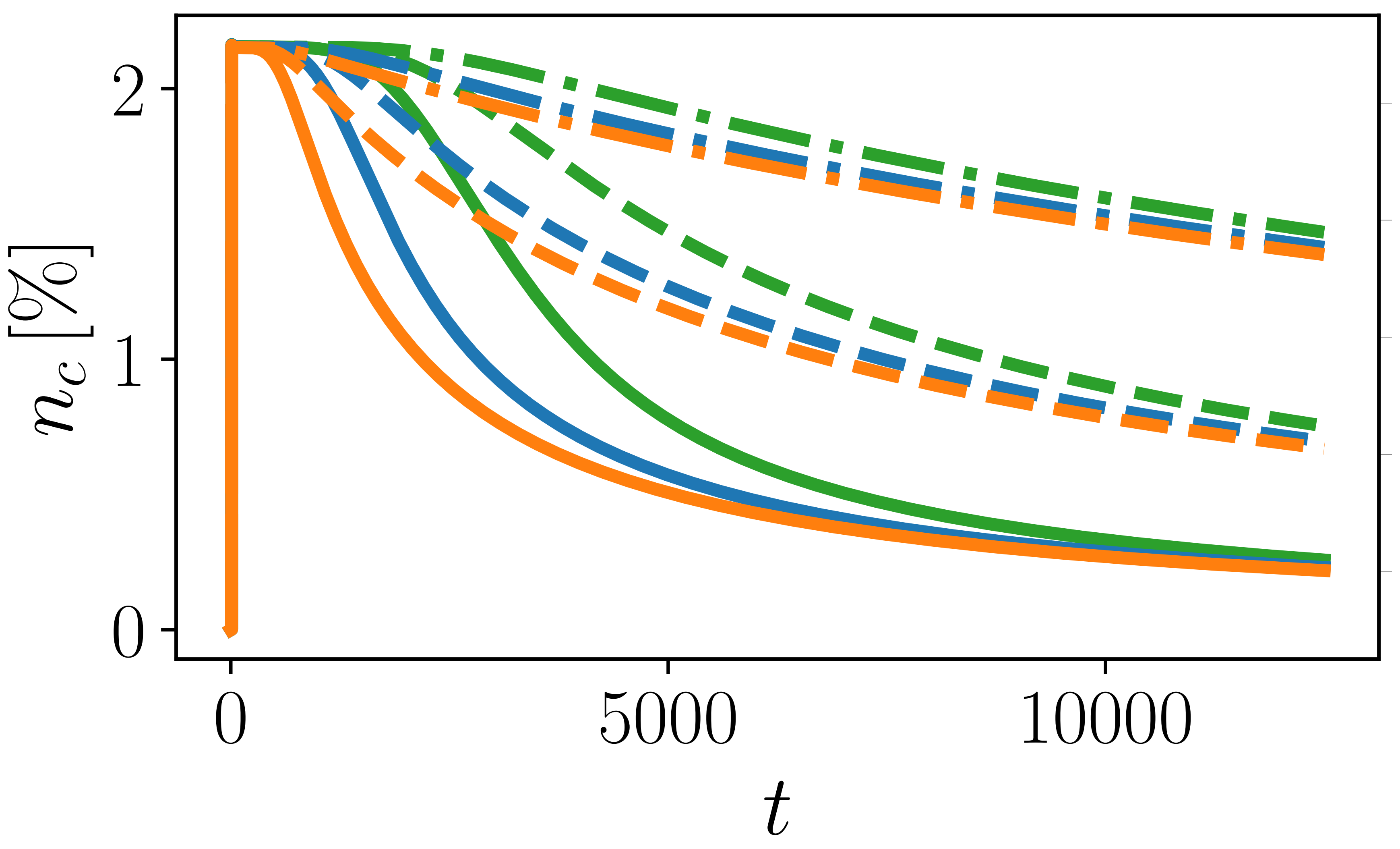

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Blommel, Perfetto, Stefanucci, **VV** arXiv:2505.01541 (2025)

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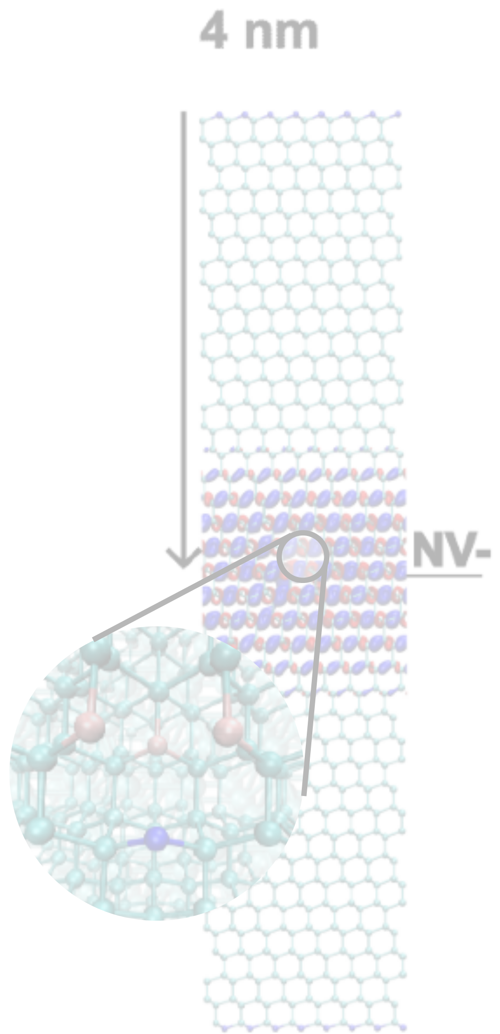

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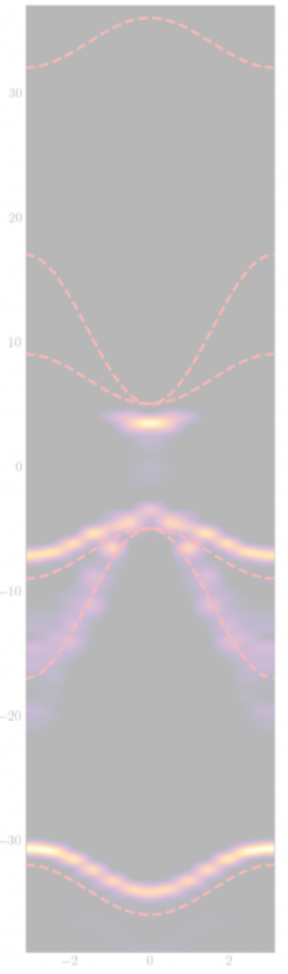

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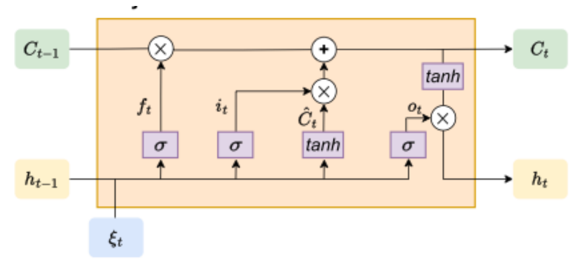
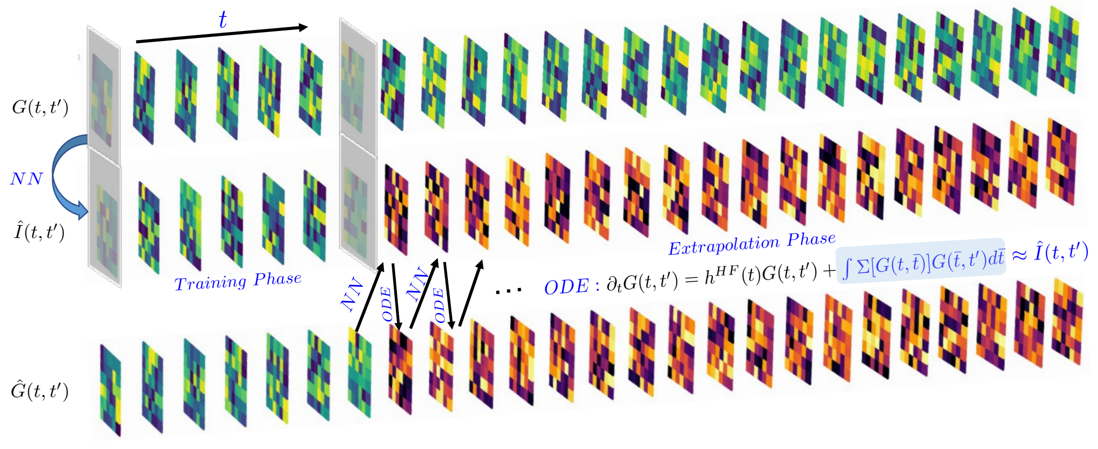
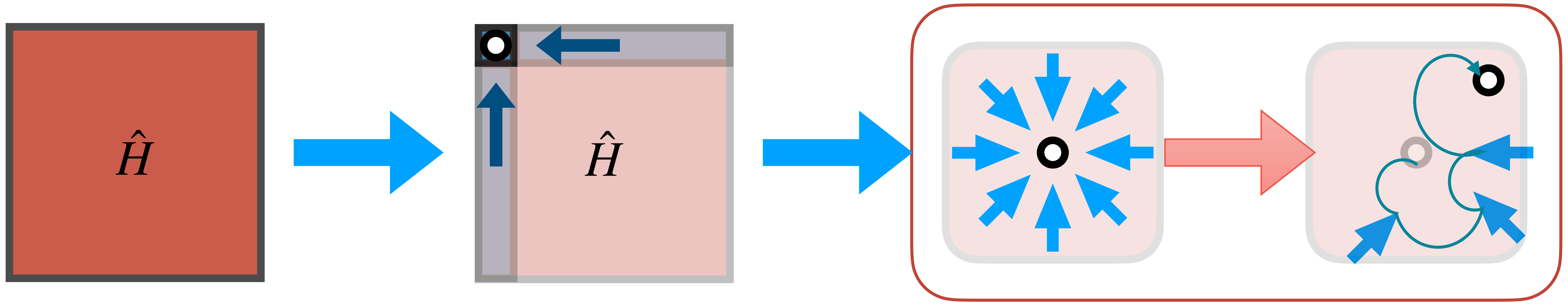




Numerical and Data-driven methods



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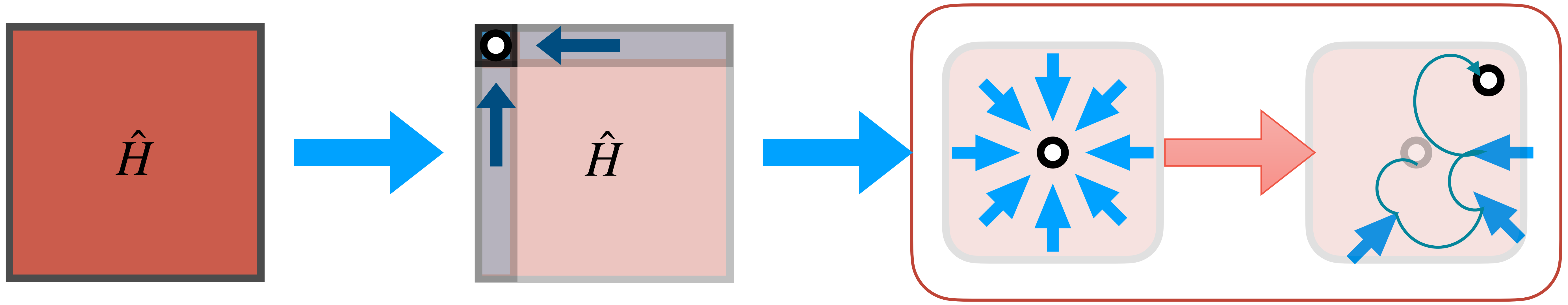

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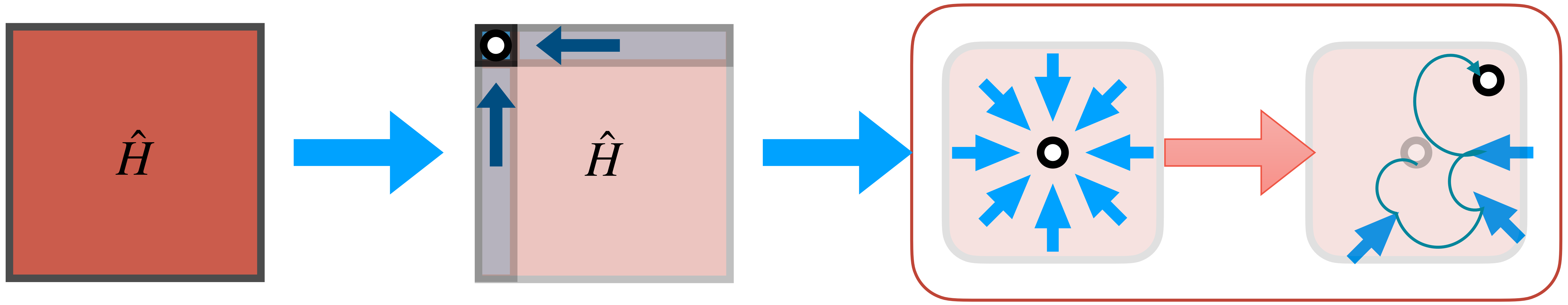

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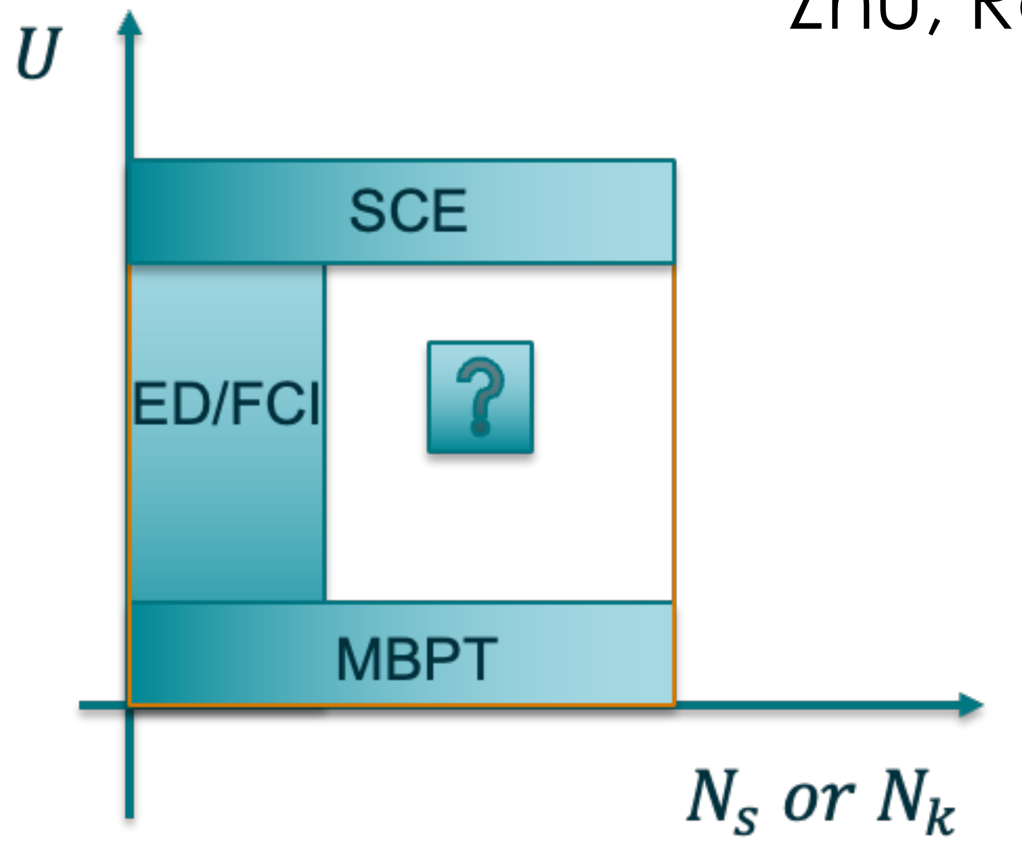
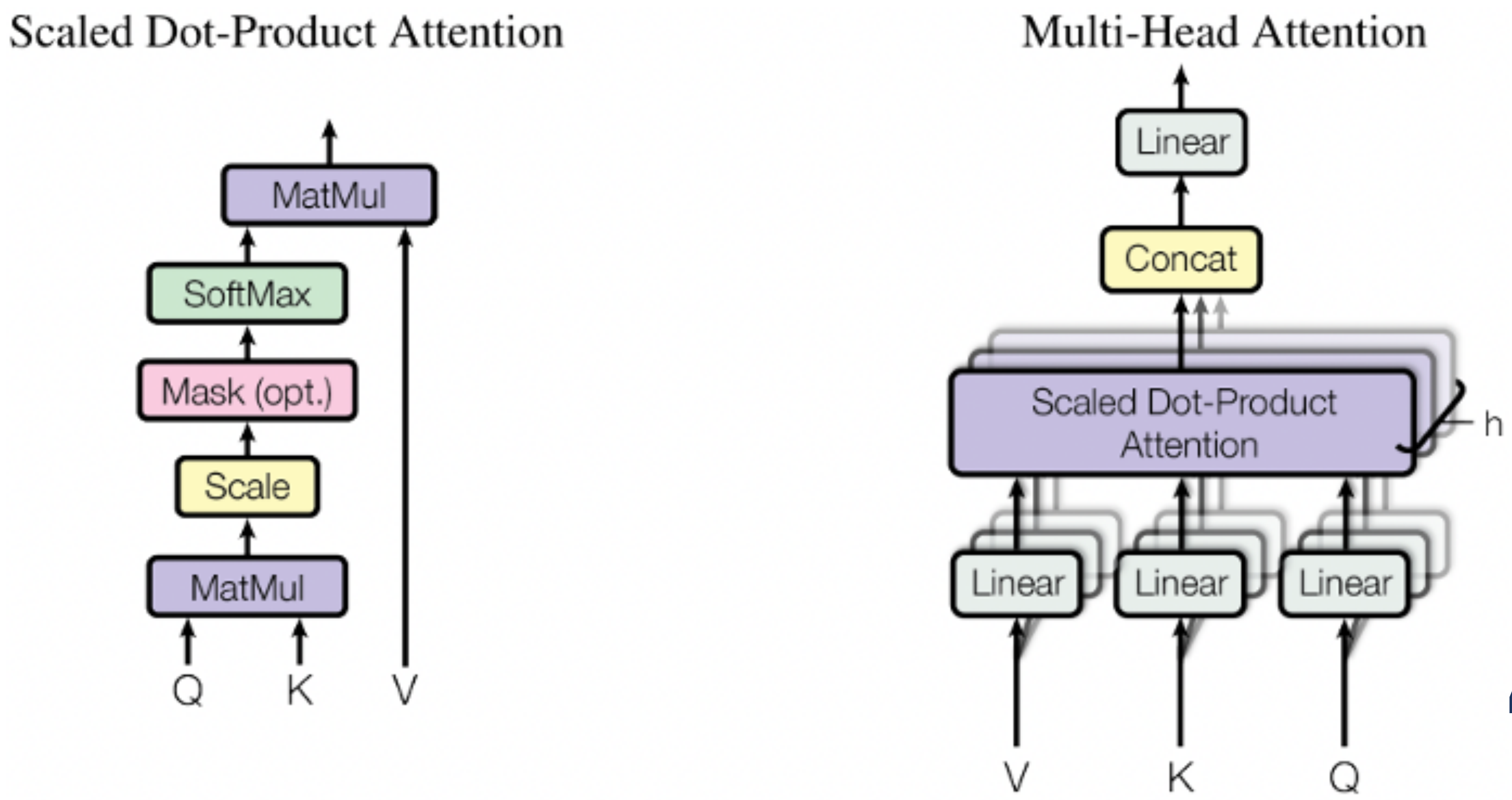
Zhu, Rosenberg, Huang, Bassi, Yang, Zhang, arXiv:2504.14483 (2025)





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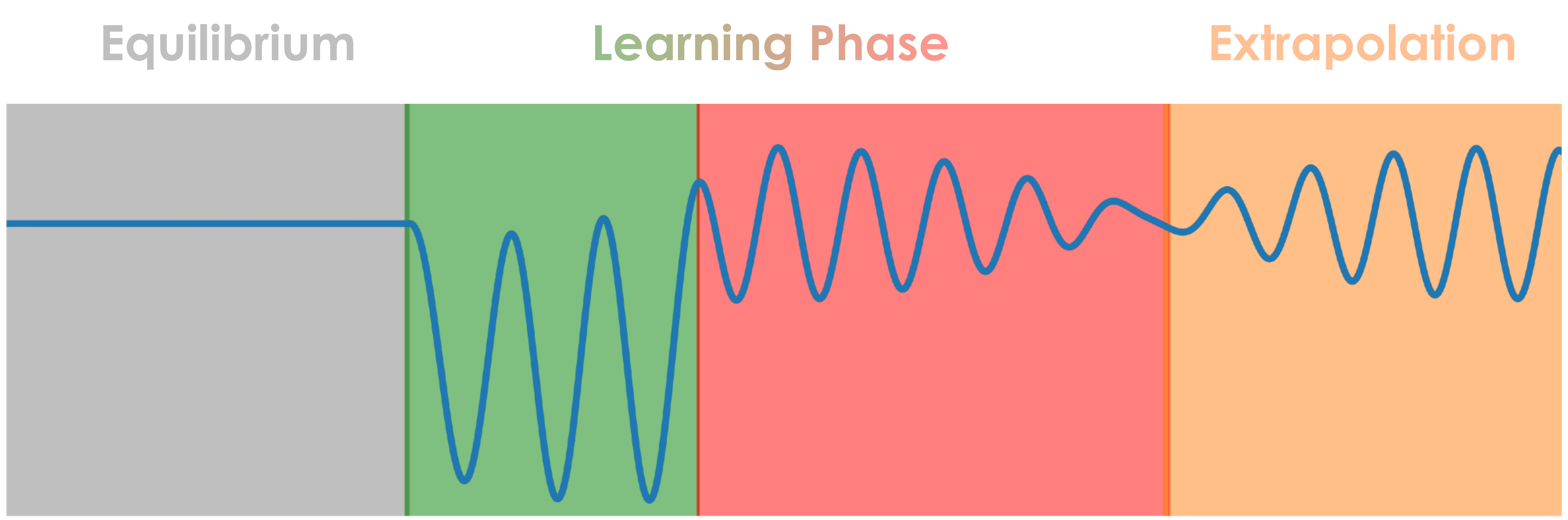
Numerical extrapolations — mapping on generalized linear dynamics



Reeves, Yin, Zhu, Ibrahim, Yang, **VV** Phys Rev B 7, 075107 (2023)

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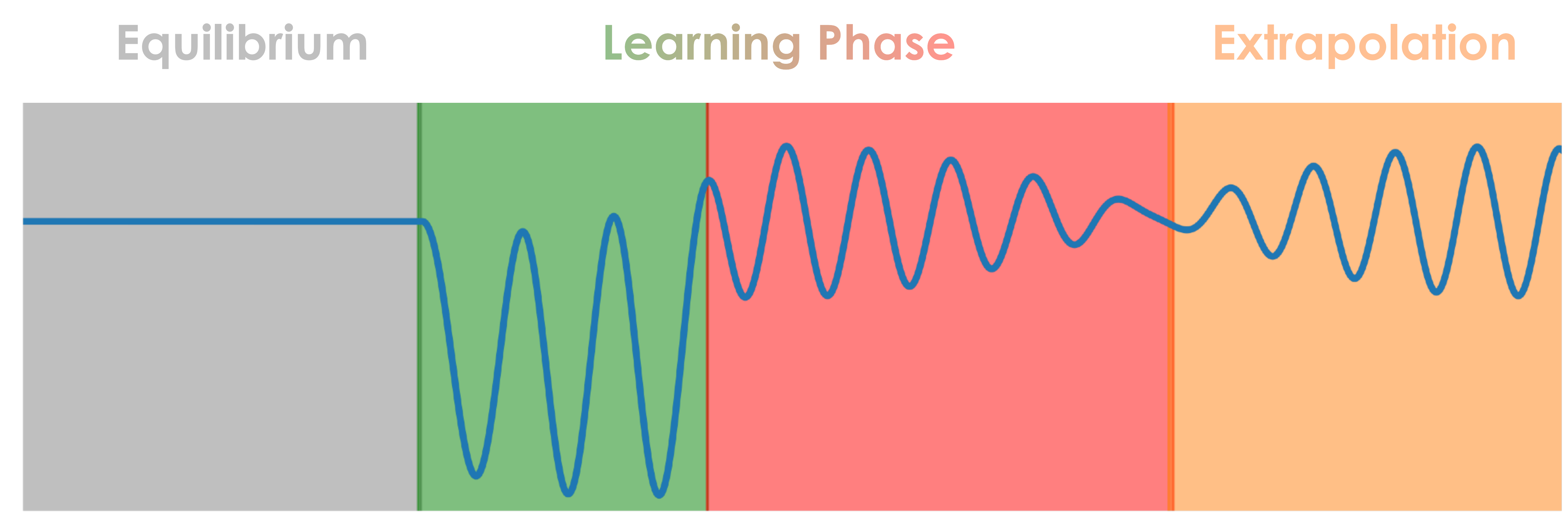

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Koopman PNAS 17, 315–8 (1931)

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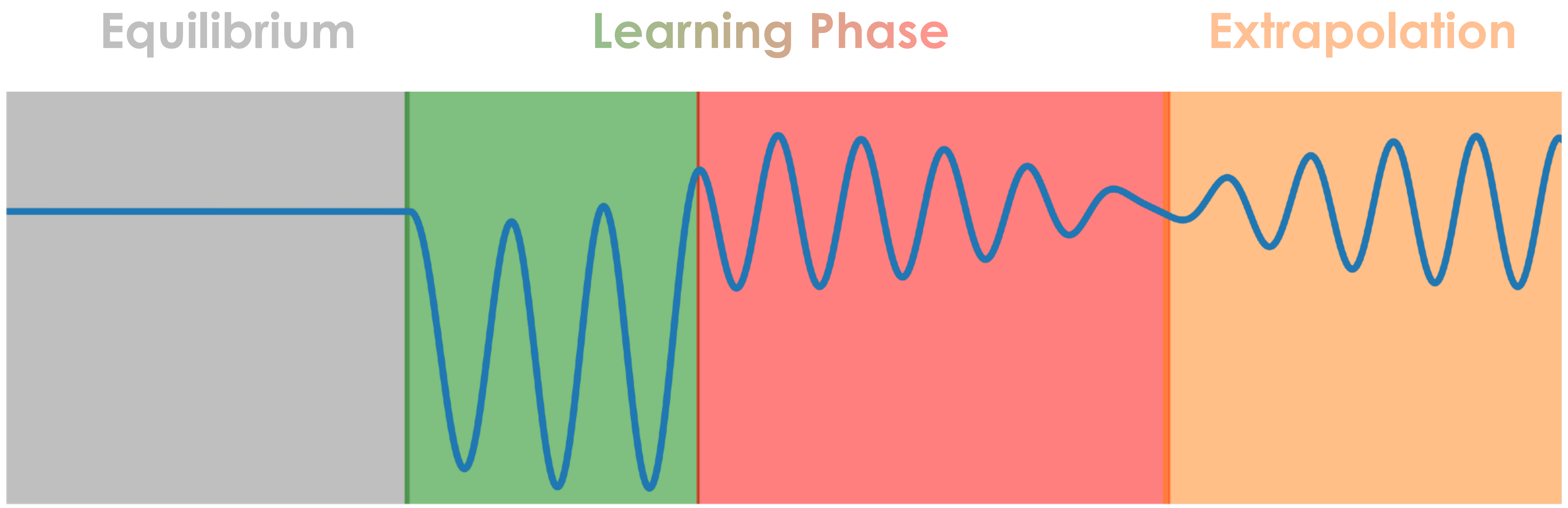

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$$\frac{d\hat{G}(t)}{dt} = \hat{f}(\hat{G}(t), t) \underset{\text{DMD assumption}}{\Rightarrow} \hat{G}(t+\Delta t) \approx \hat{A}\hat{G}(t)$$

$$\hat{A} \; X_1 \approx X_2$$

$$X_1^T = [G(t_0), \ldots G(t_m)] \qquad X_2^T = [G(t_1), \ldots G(t_{m+1})]$$

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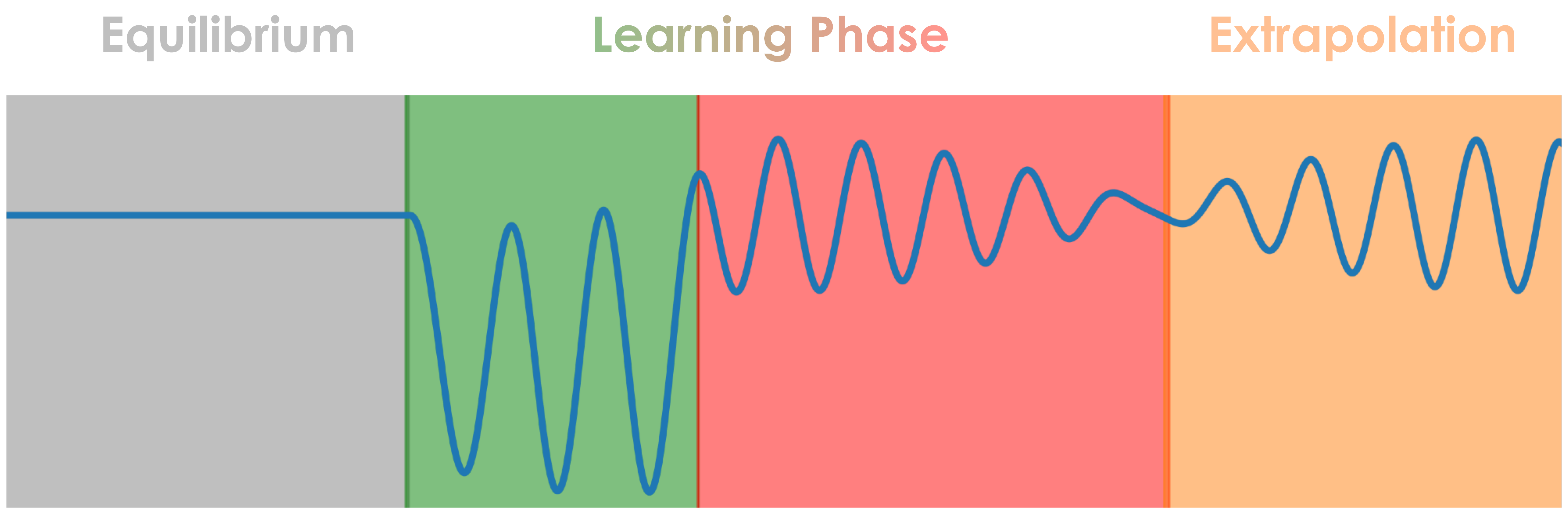

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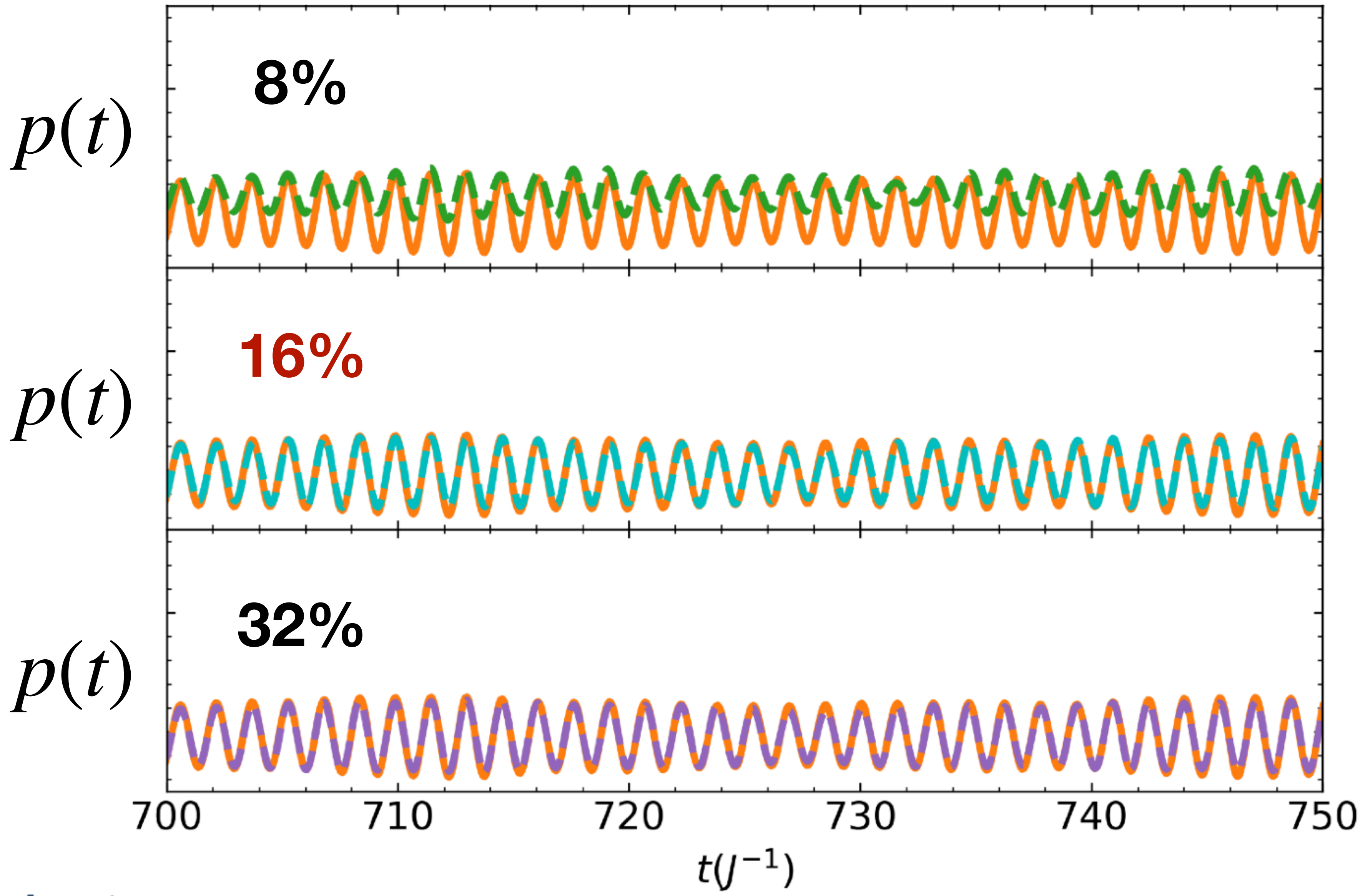

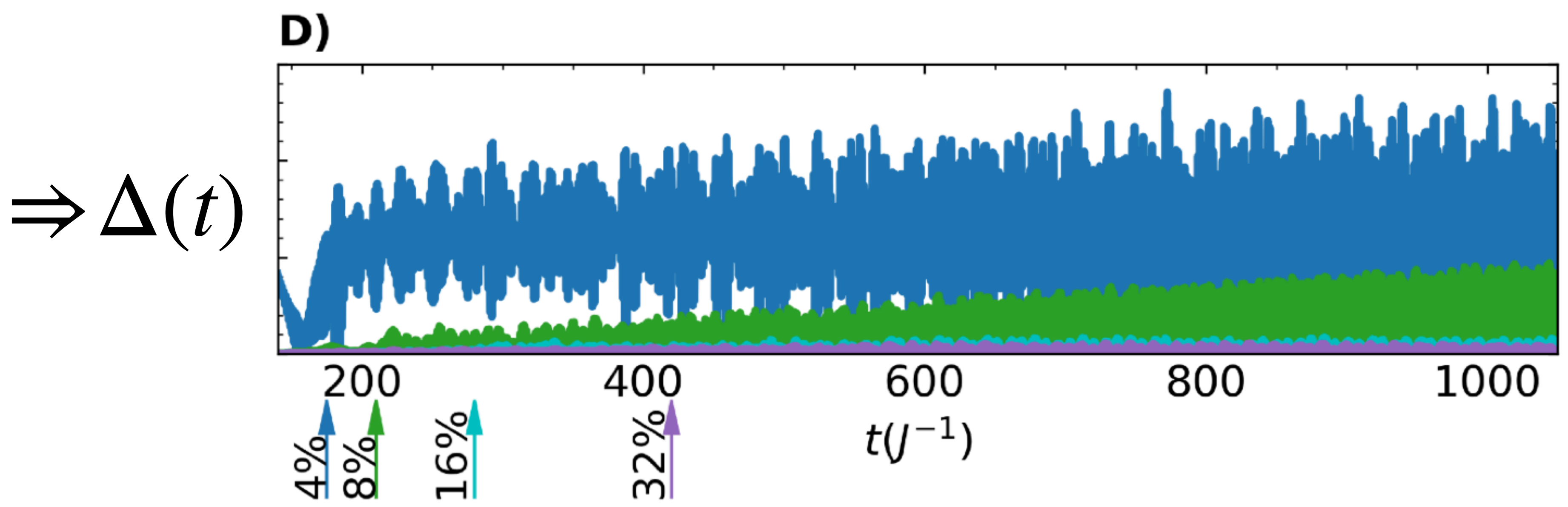

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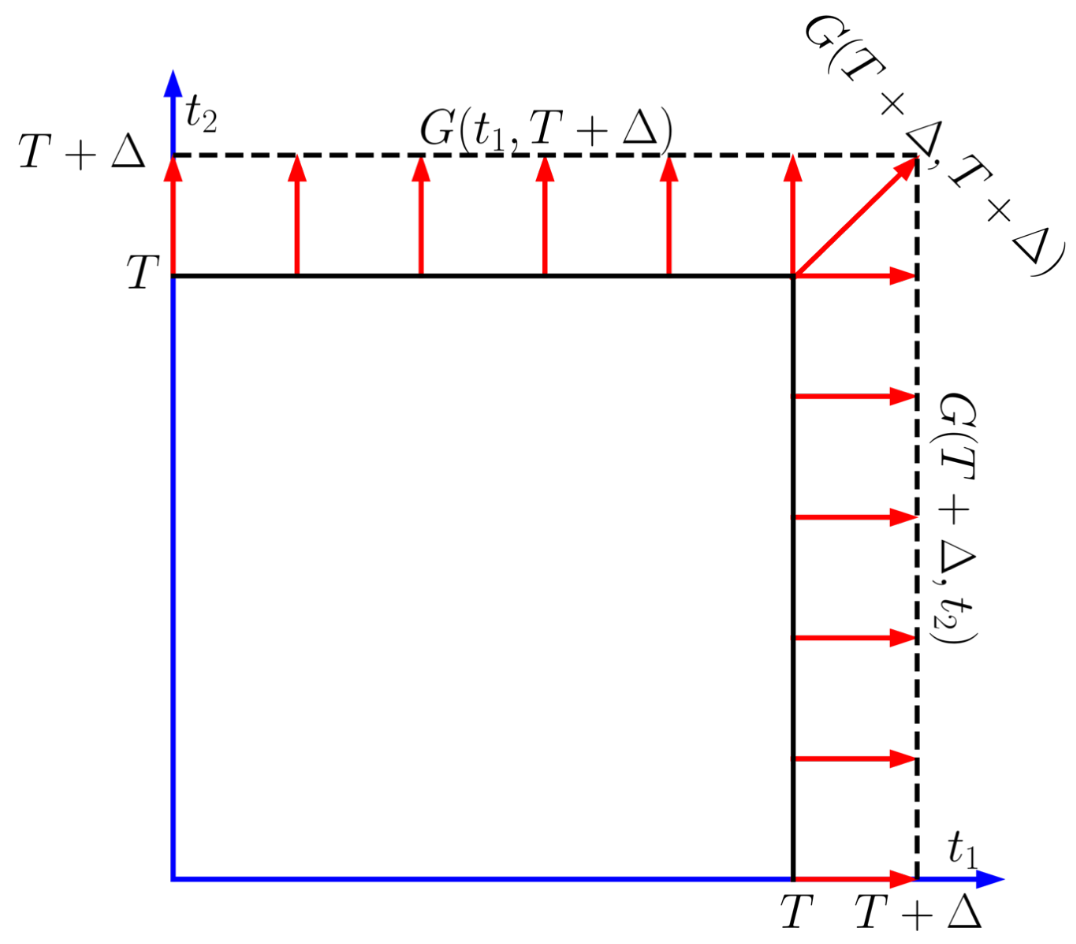
From Equations to Operators: Rethinking Dynamics

- Similar to the equilibrium self-energy ⇔ dynamics as operator learning:

$$\left[i\partial_t - h^{(0)}(t)\right] G(t,t') = \delta(t,t') + I(t,t') \quad \Rightarrow \quad i\partial_t G(t,t') = A[G(t,t')] + \tilde{I}[G(t,t')]$$

integro-differential equations

functional operator



$$\boxed{G(t,t')} \Rightarrow \boxed{I(t,t')} \Rightarrow \boxed{\tilde{I}(t,t')}$$

Bassi, et al. Machine Learning with Applications 15, 100524 (2024)

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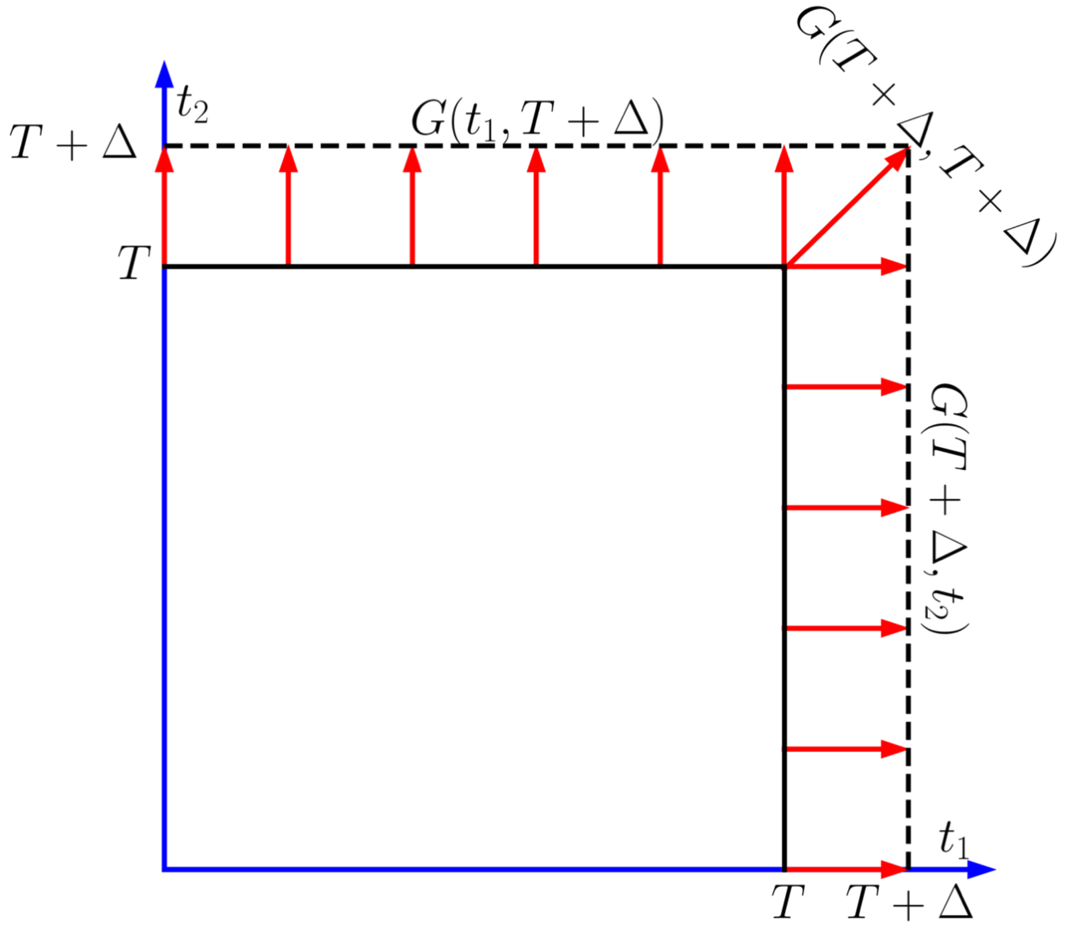

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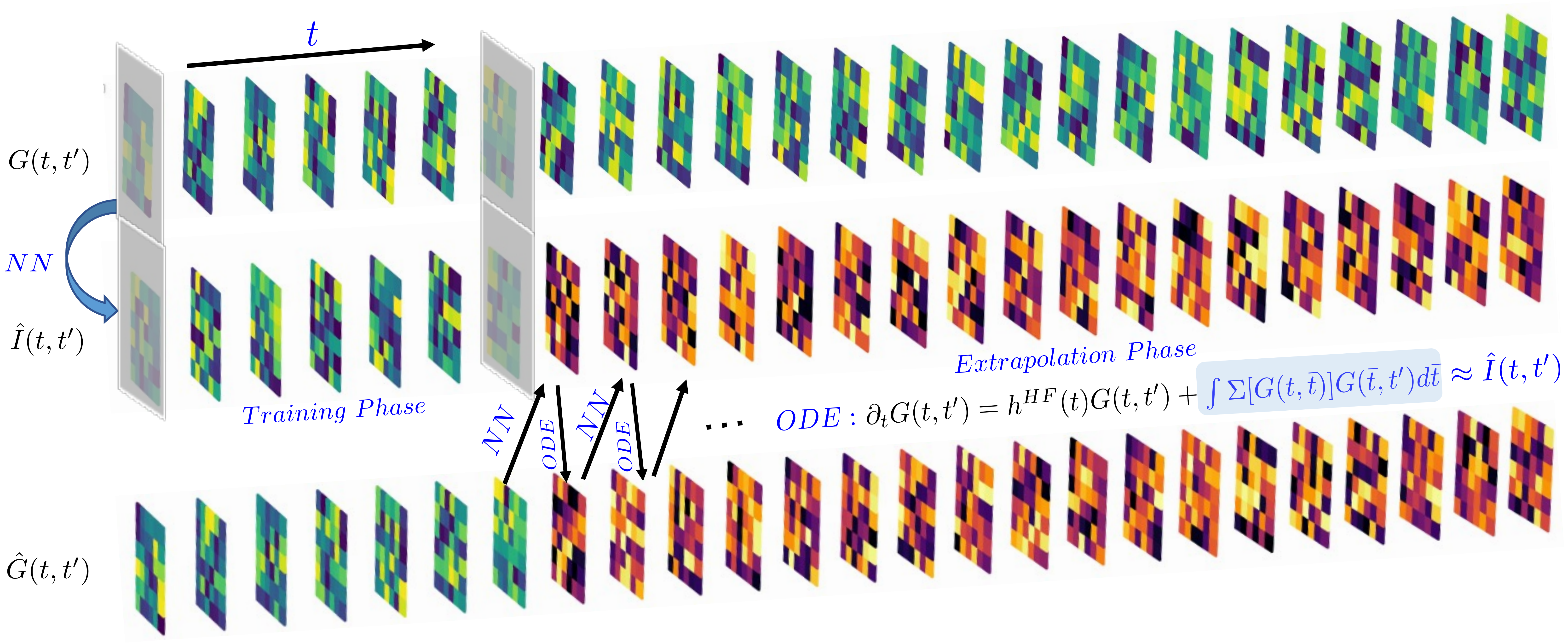

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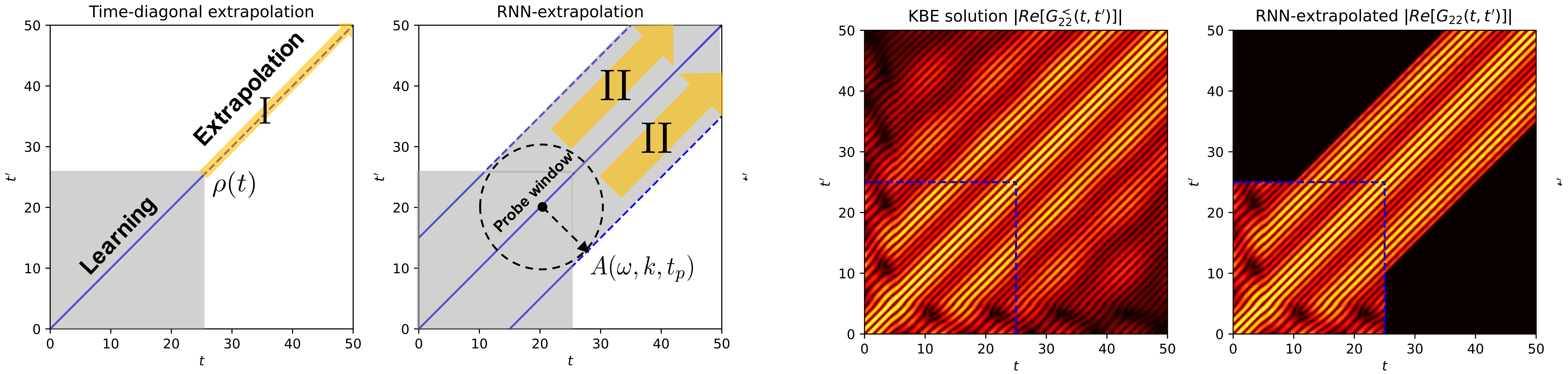

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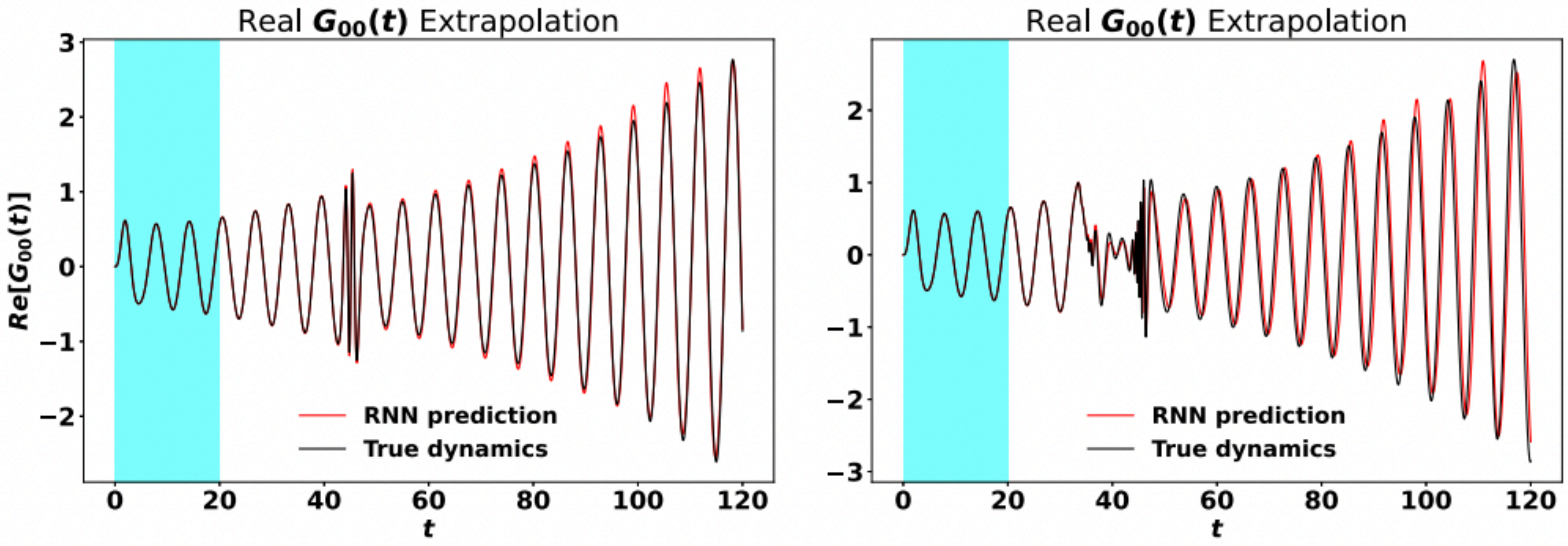

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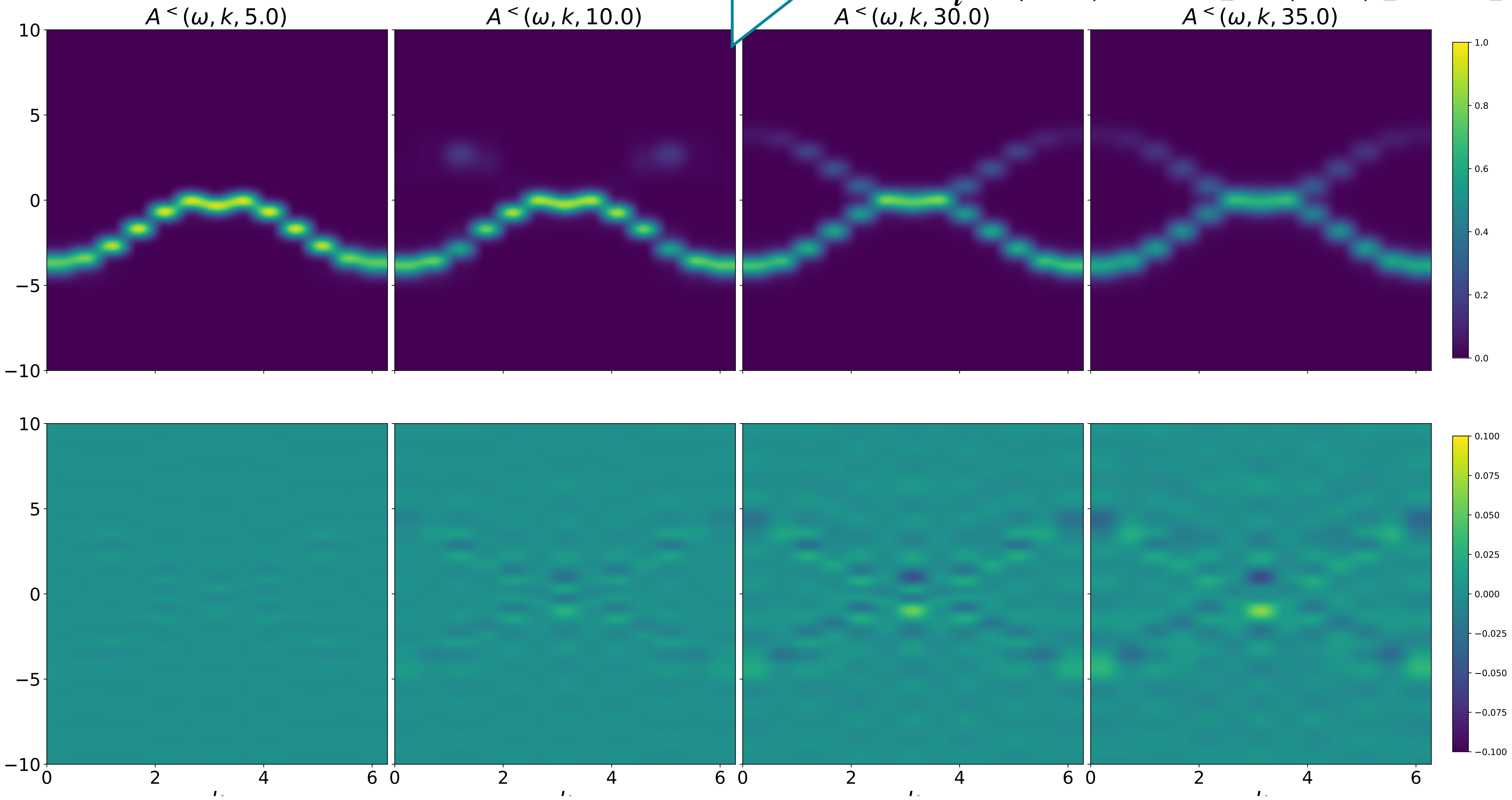

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Many Questions and Even More Opportunities:

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- Final thought: **Hardware acceleration** ⇒ speeds up target *algebraic operations* and *learning*
  - GPUs, TPUs, FPGAs?
  - Stochastic and Neuromorphic computing?

Morningstar et al., PRX Quantum 3, 020331 (2022)
Czischek et al., SciPost Phys. 12, 039 (2022)

Acknowledgments

+ G **Stefanucci**, E **Perfetto**, Y **Zhu**, C **Yang**, K **Ibrahim**, …

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