



Ultracompact entangled photon sources based on 2D semiconductors

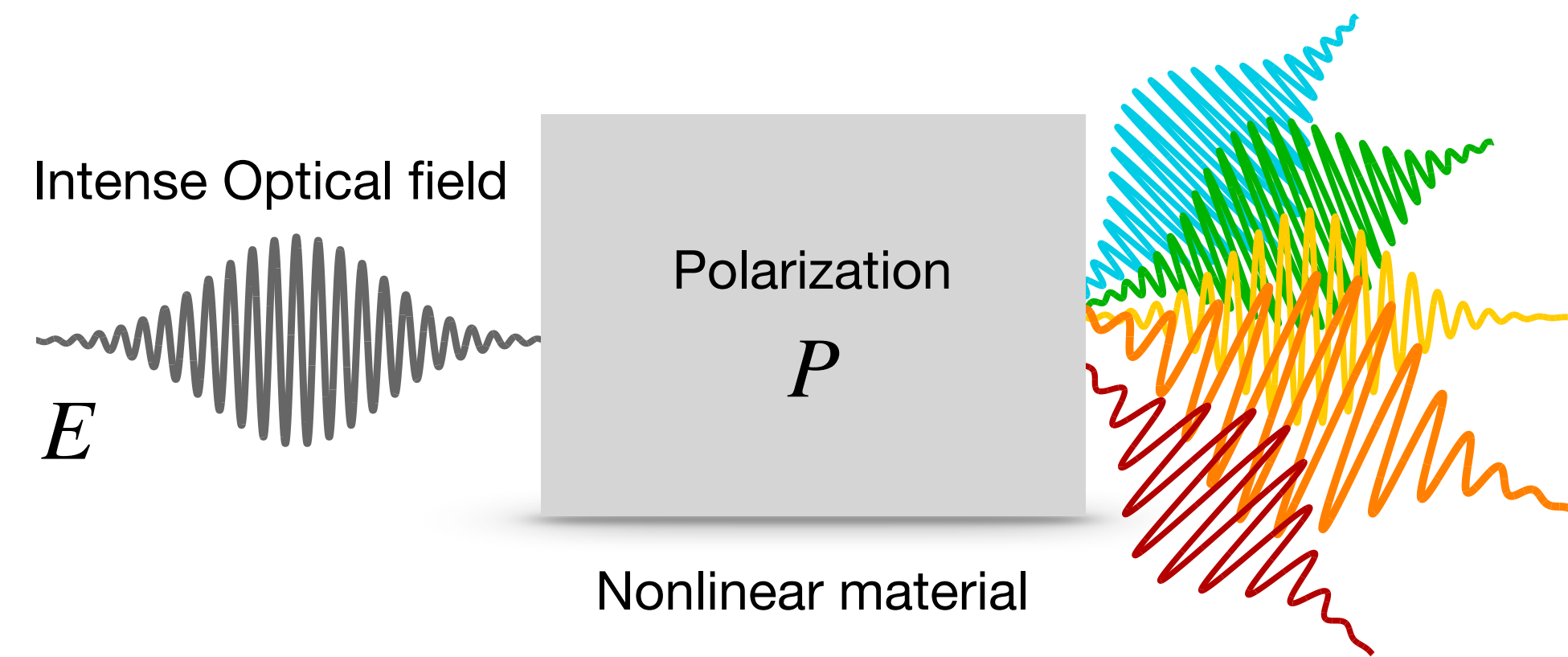
Chiara Trovatello – chiara.trovatello@polimi.it

Assistant Professor – Politecnico di Milano, Milano, Italy

Marie Skłodowska-Curie Global Fellow – Columbia University, New York, NY, USA

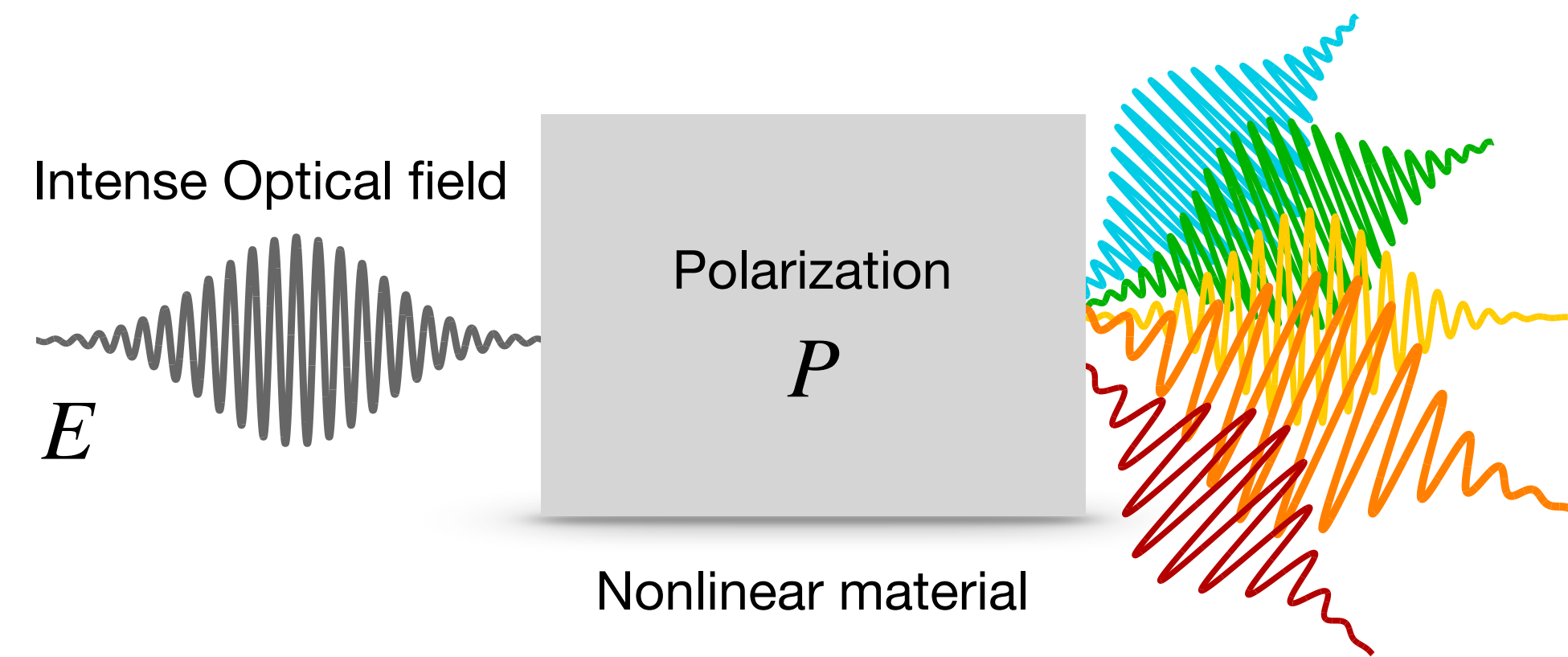


Nonlinear optics: mathematical operations with photons



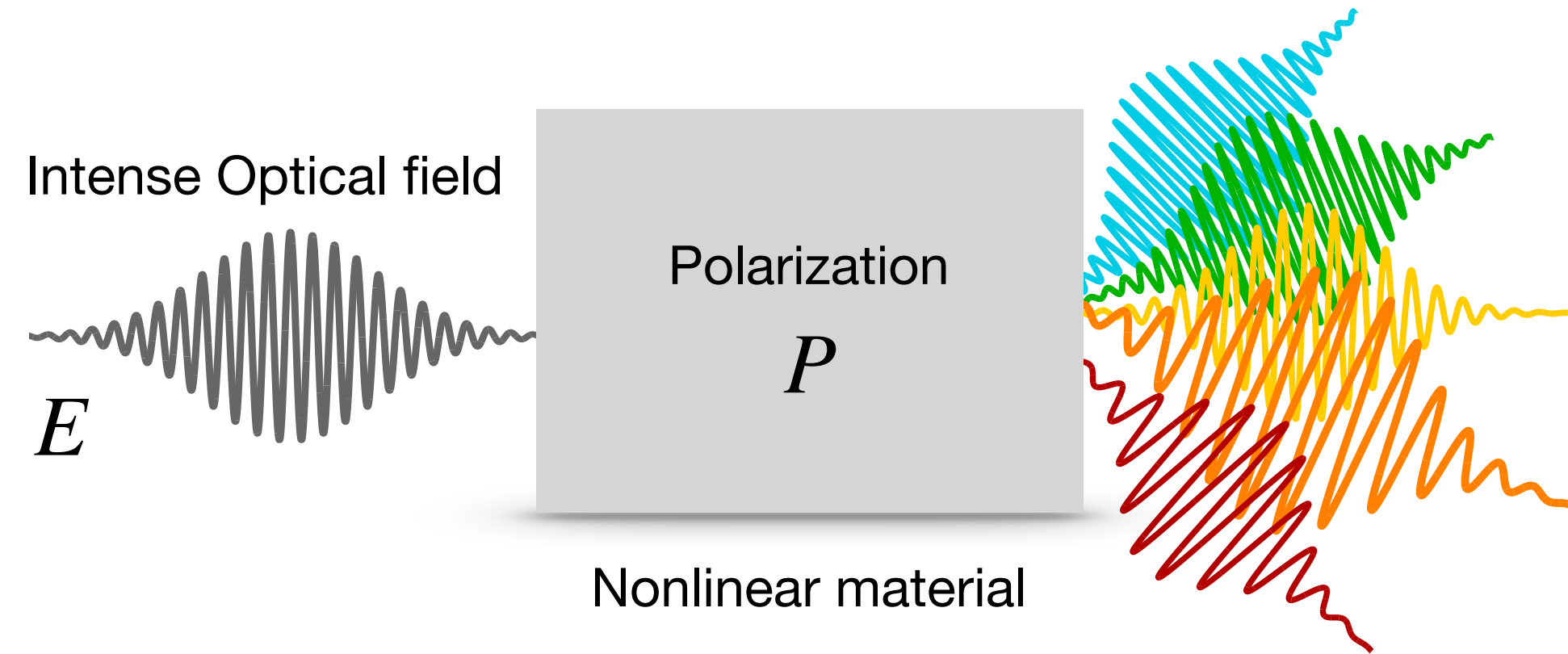
Polarization $P = \epsilon_0(\chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \dots)$

Nonlinear optics: mathematical operations with photons



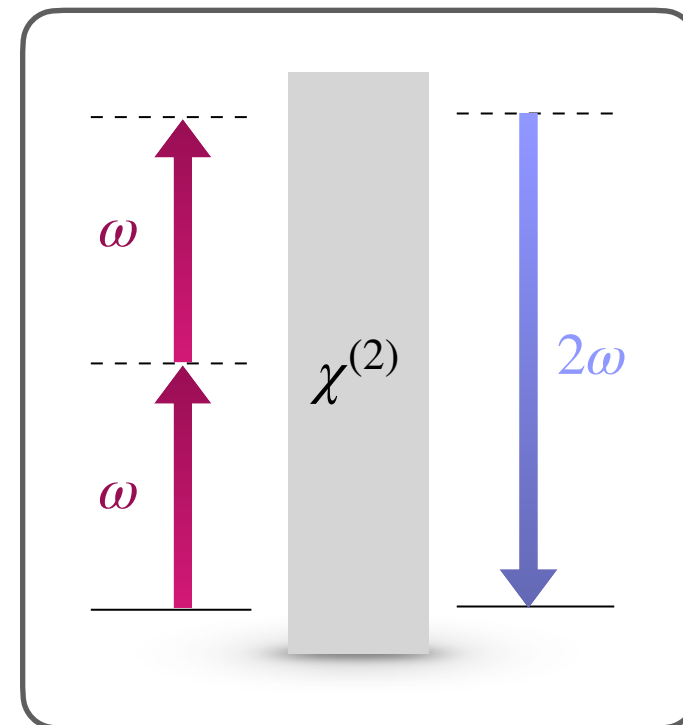
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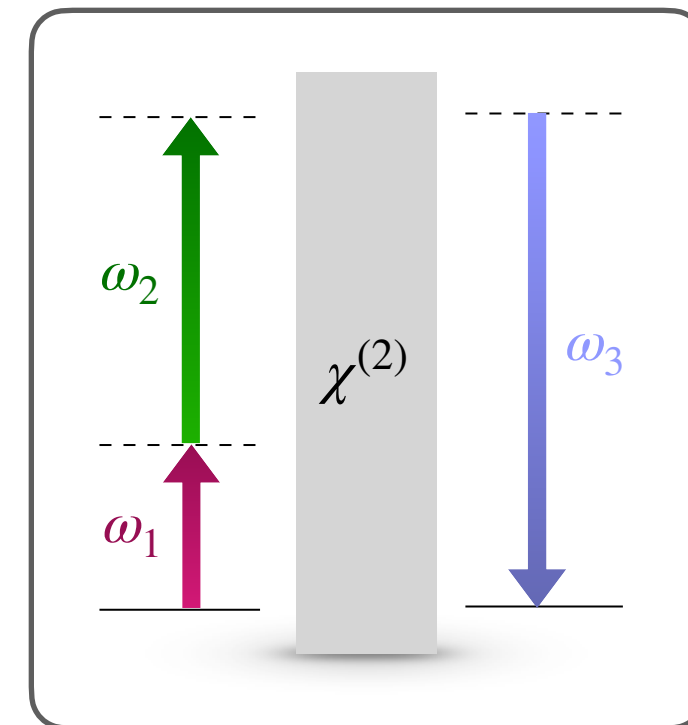
$$\text{Polarization } P = \epsilon_0(\chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \dots)$$

$$\omega + \omega = 2\omega$$



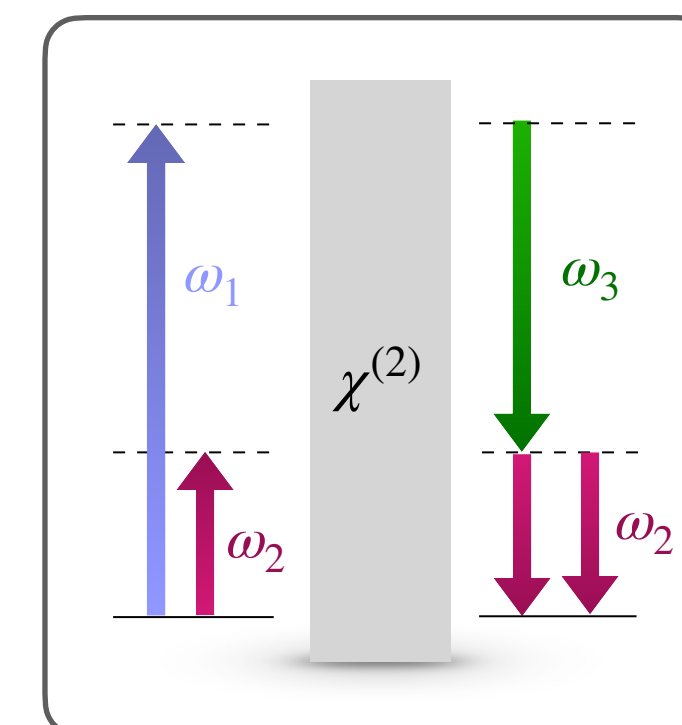
Second Harmonic Generation (SHG)

$$\omega_1 + \omega_2 = \omega_3$$



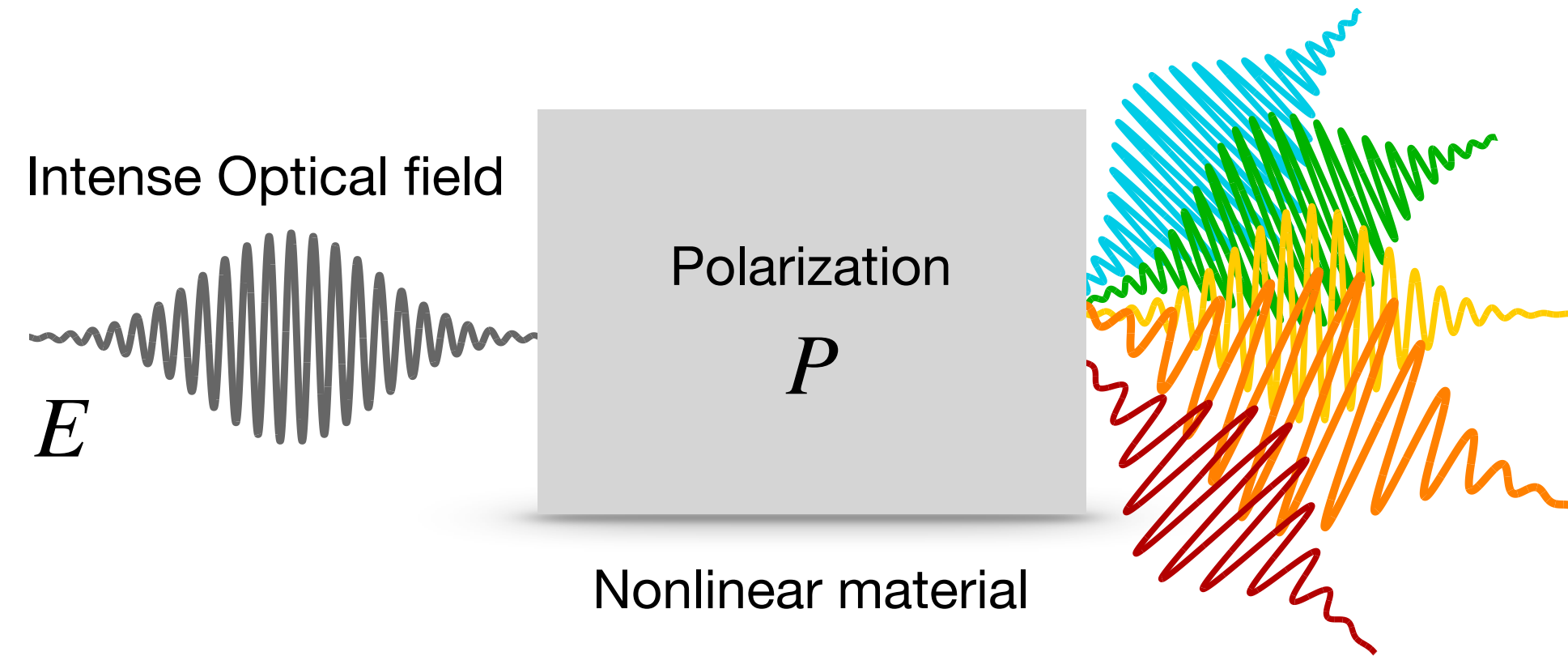
Sum Frequency Generation (SFG)

$$\omega_1 - \omega_2 = \omega_3$$



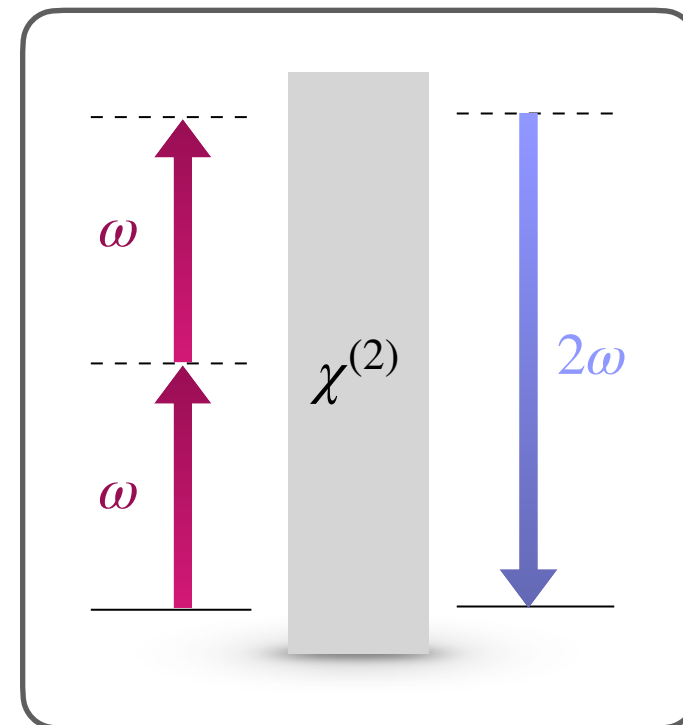
Difference Frequency Generation (DFG)
Optical Parametric Amplification (OPA)

Nonlinear optics: mathematical operations with photons



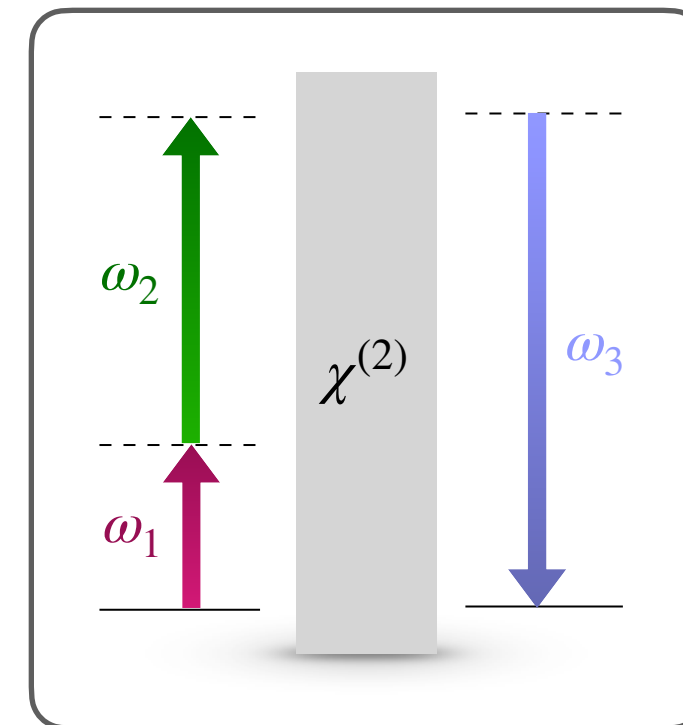
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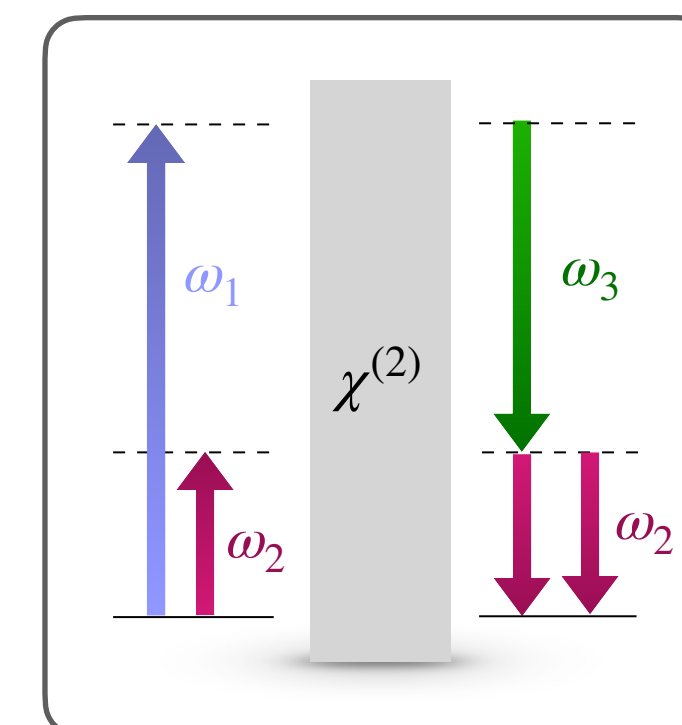
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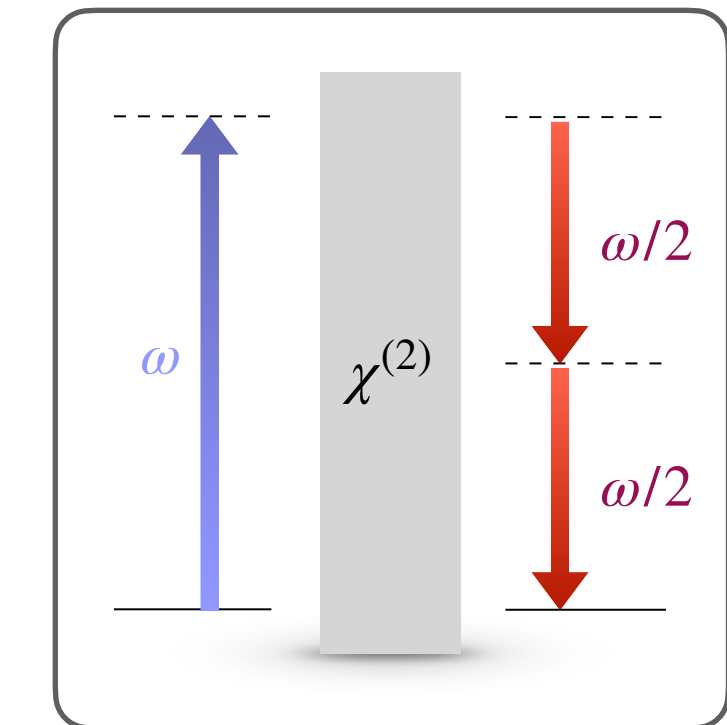
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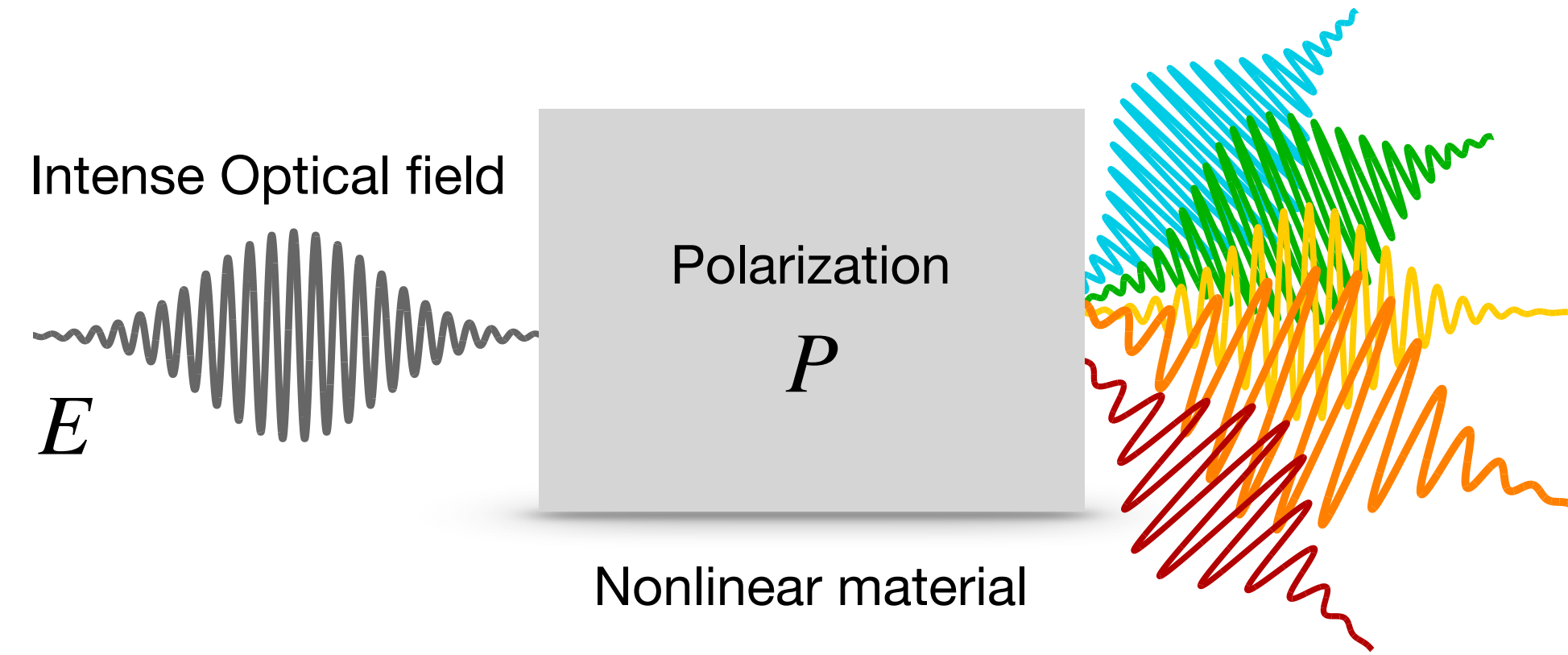
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$$\omega \rightarrow \omega/2 + \omega/2$$



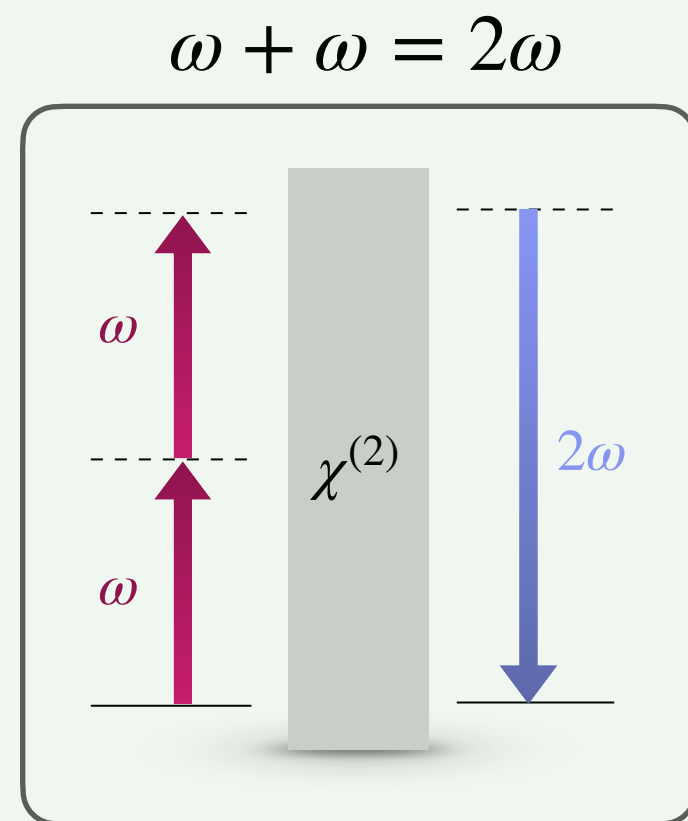
Spontaneous Parametric Down-conversion (SPDC)

Nonlinear optics: mathematical operations with photons

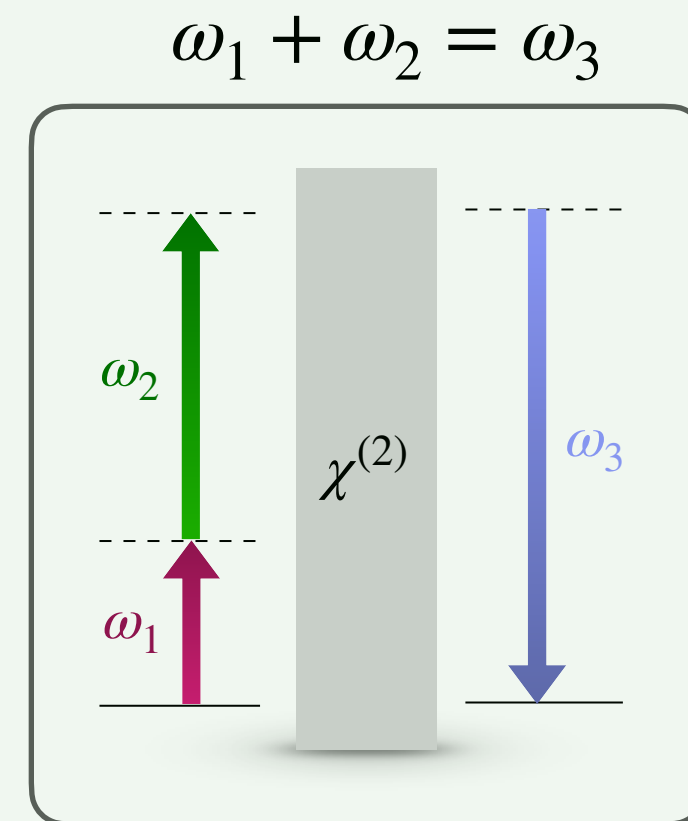


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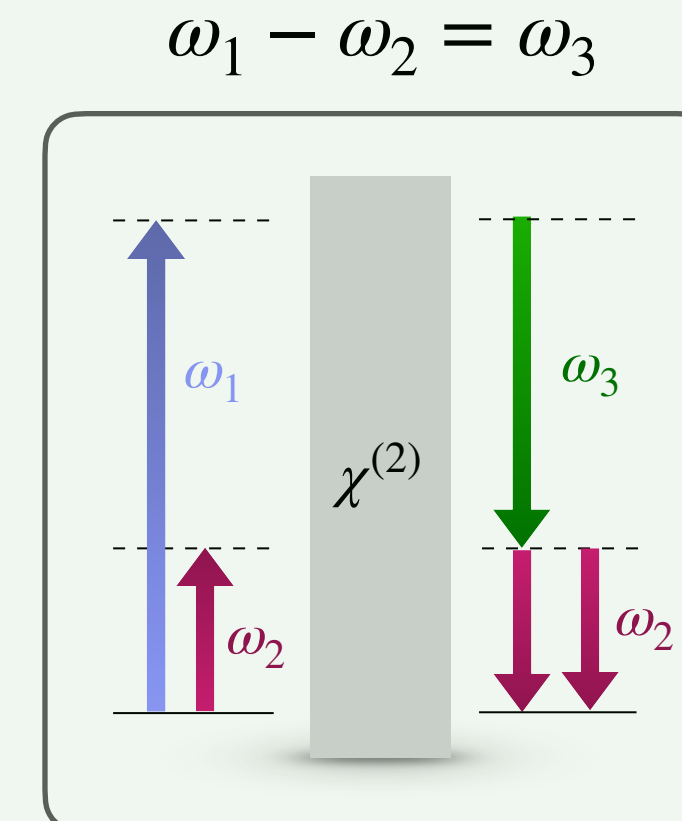
Classical light



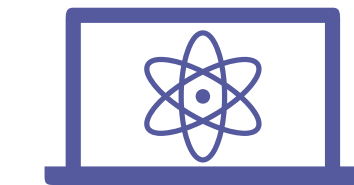
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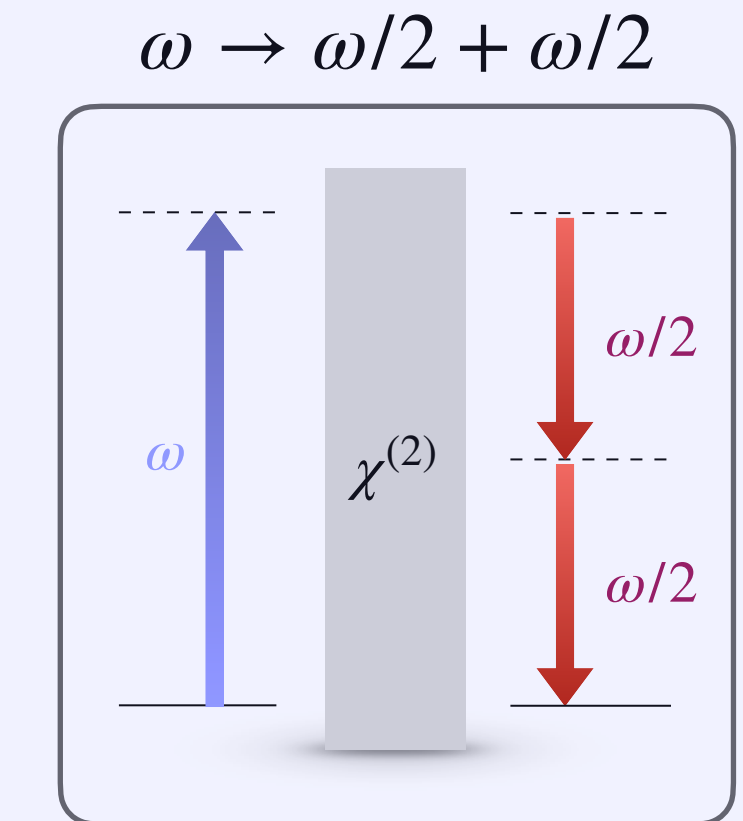
Sum Frequency Generation (SFG)



Difference Frequency Generation (DFG)
Optical Parametric Amplification (OPA)



Quantum light



Spontaneous Parametric Down-conversion (SPDC)

Efficiency of nonlinear crystals

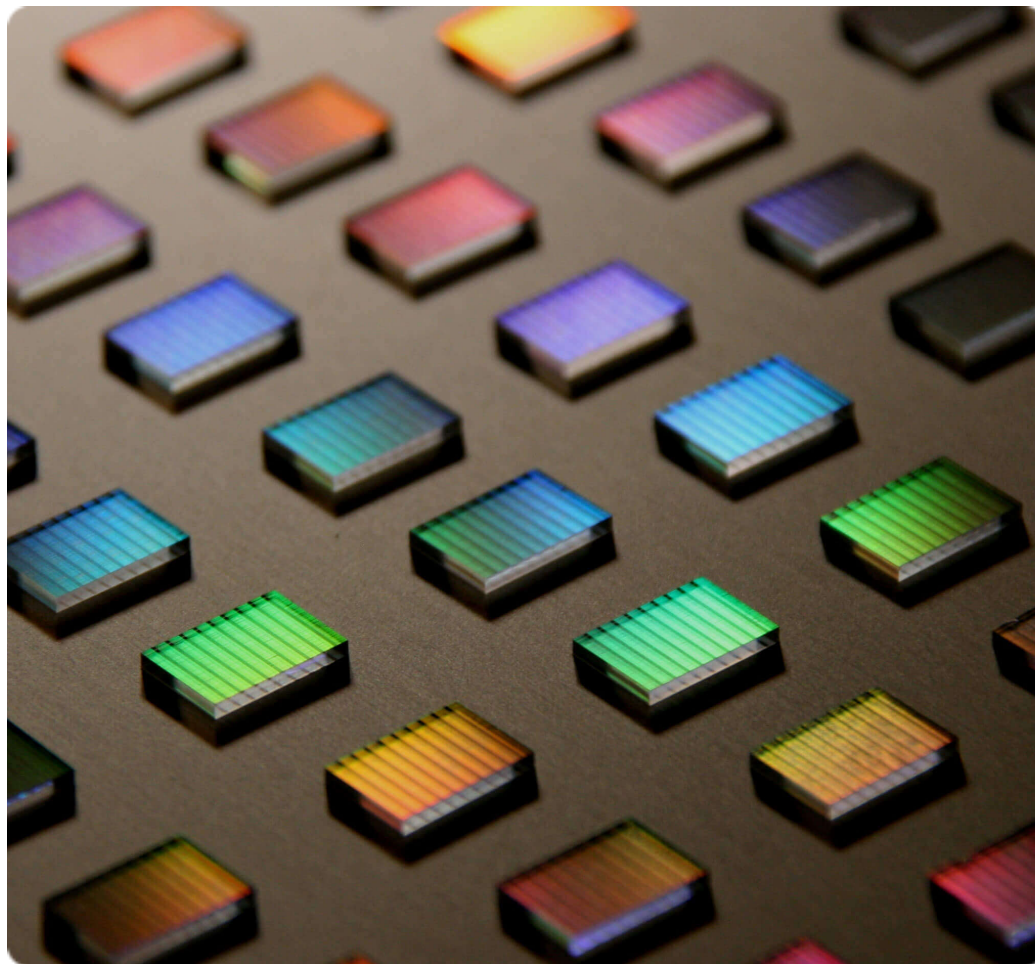
Bulk nonlinear crystals

(e.g. BBO)



$$|\chi^{(2)}| \sim 10^{-12} \text{ m/V}$$

(e.g. PPLN)



$$|\chi^{(2)}| \sim 10^{-11} \text{ m/V}$$

C. Trovatello et al., “Tunable Optical Nonlinearities in Layered Materials” ACS Photonics, 11(8), 2860–2873 (2024)

O. Dogadov et al., “Parametric Nonlinear Optics with Layered Materials and Related Heterostructures, Laser & Photonics Reviews, 2100726 (2022)

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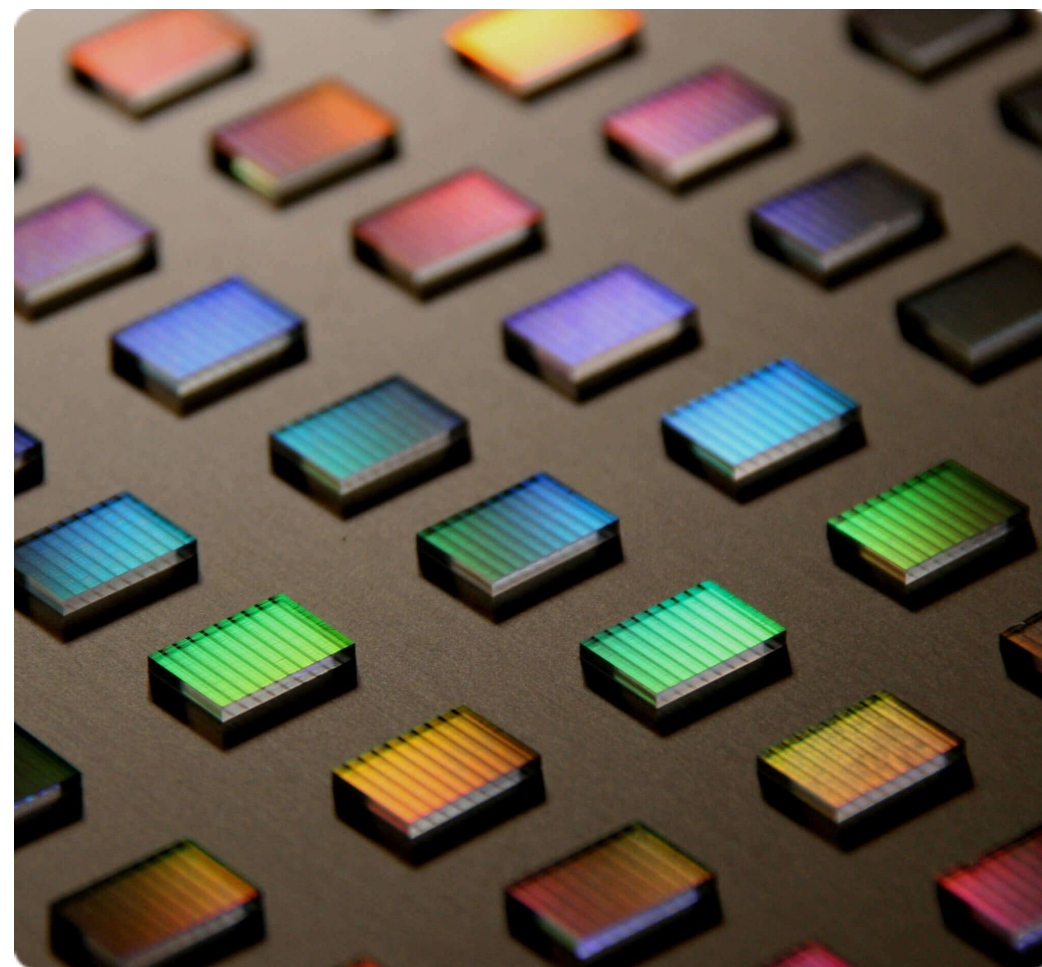
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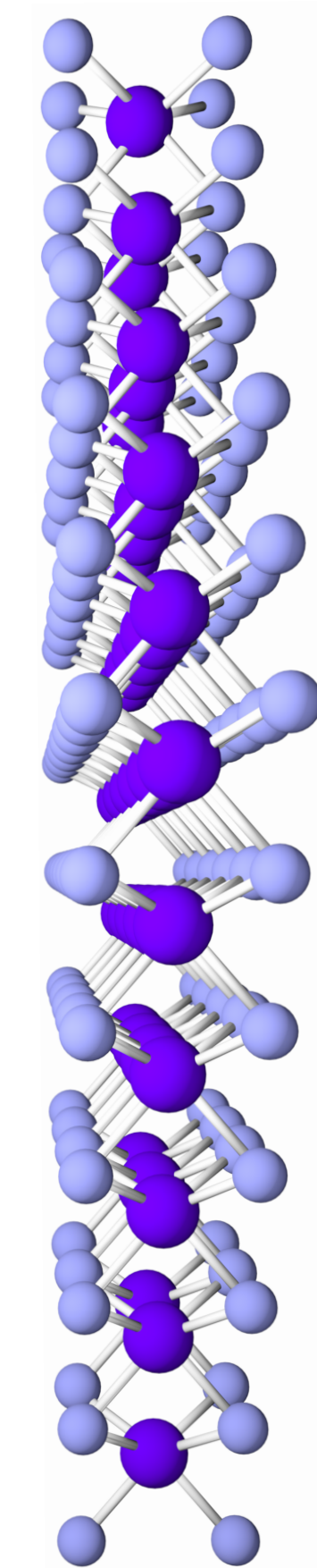
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2D nonlinear crystals

(e.g. TMDs)



MoS₂, MoSe₂
WS₂, WSe₂

$$|\chi^{(2)}| \sim 10^{-10} - 10^{-9} \text{ m/V}$$

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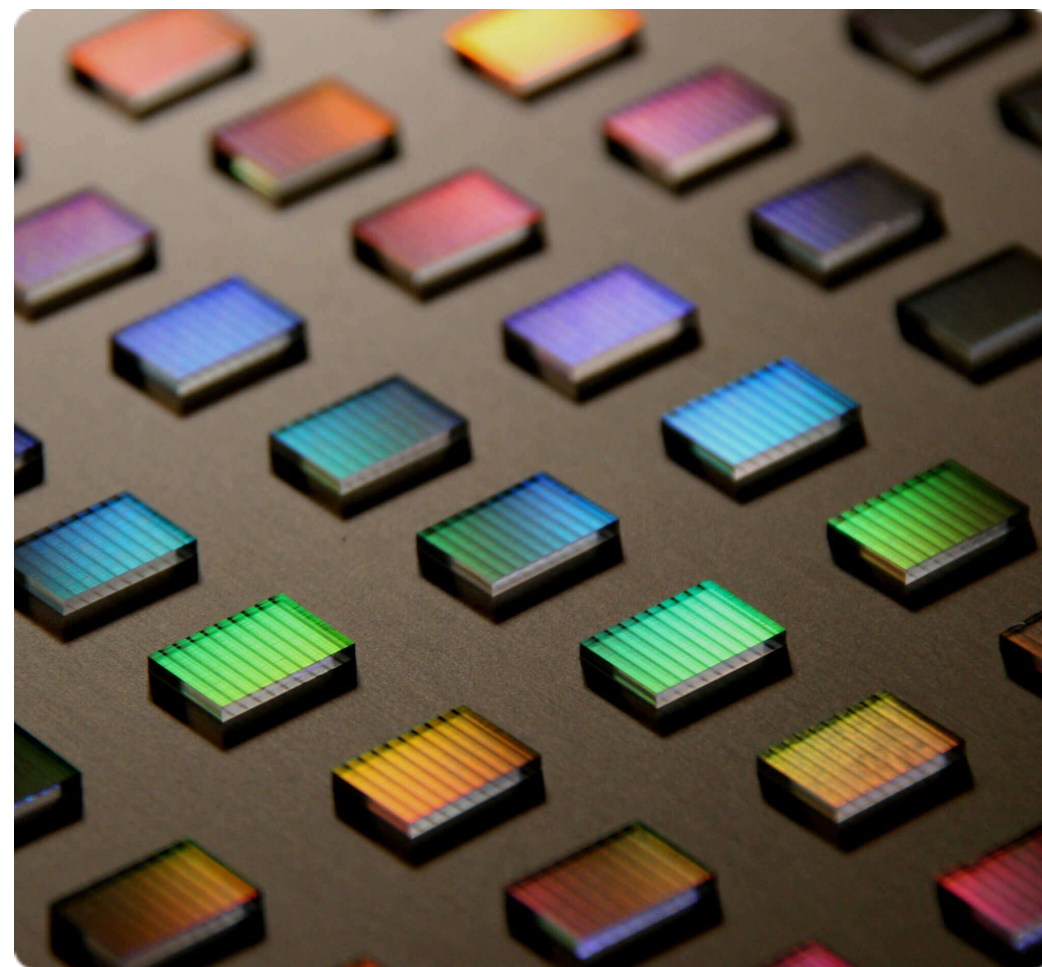
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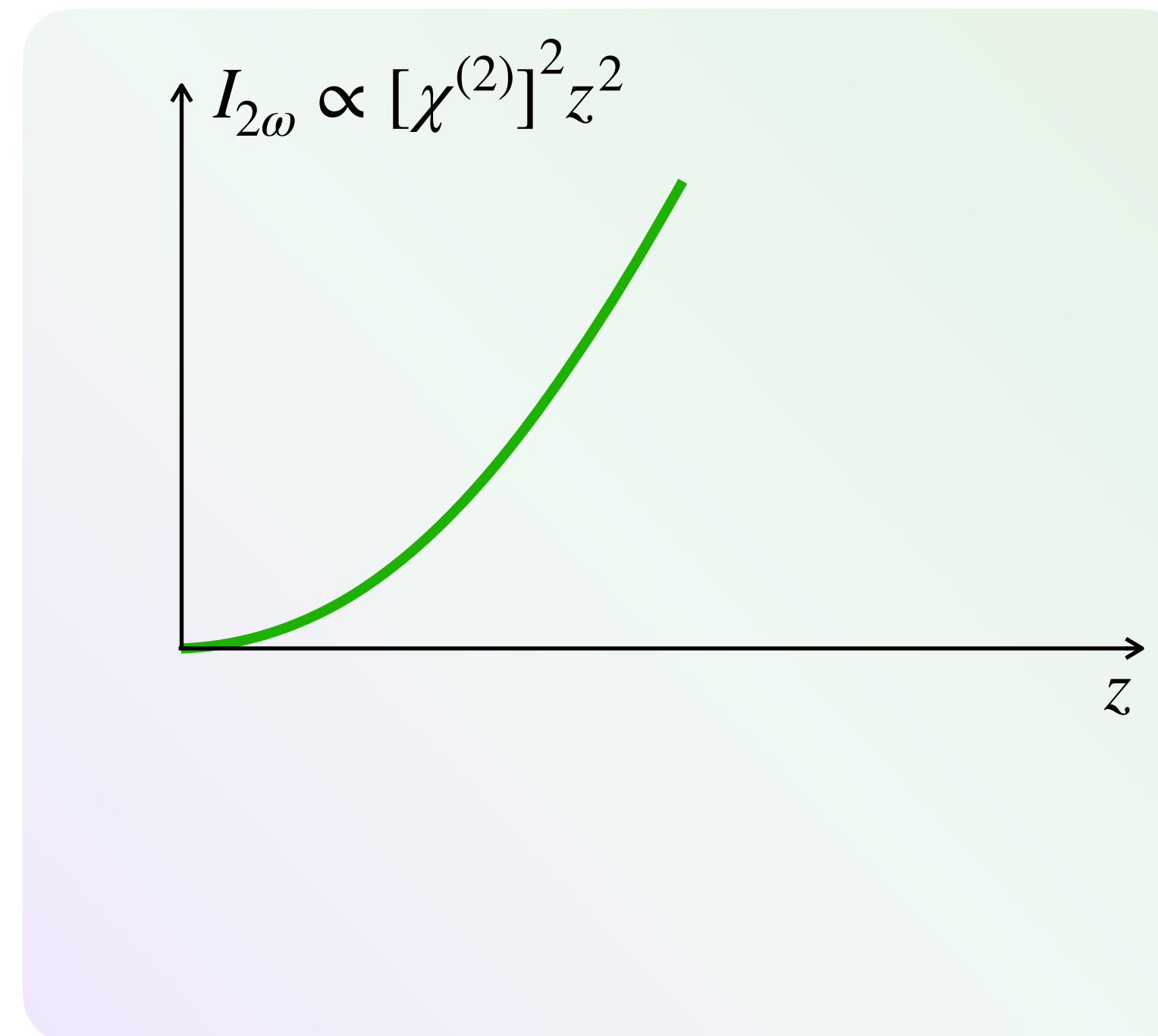
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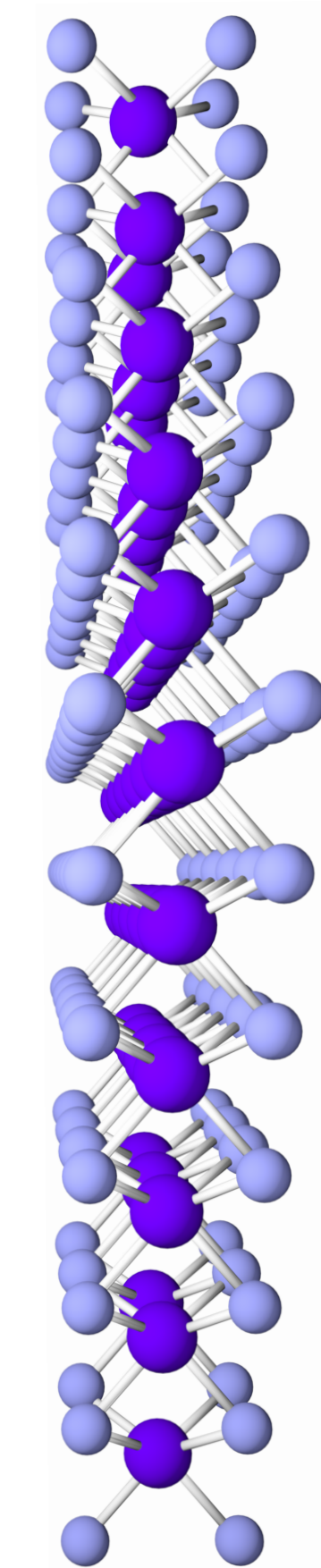
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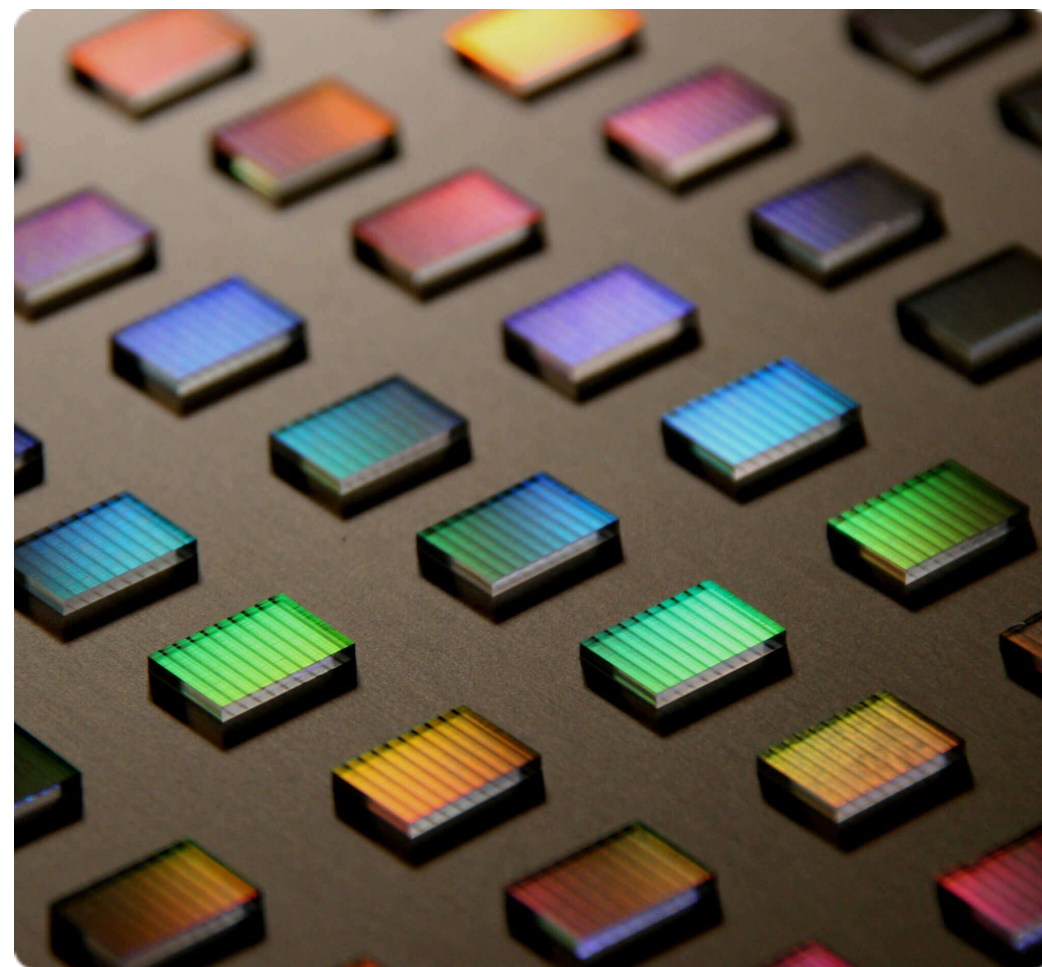
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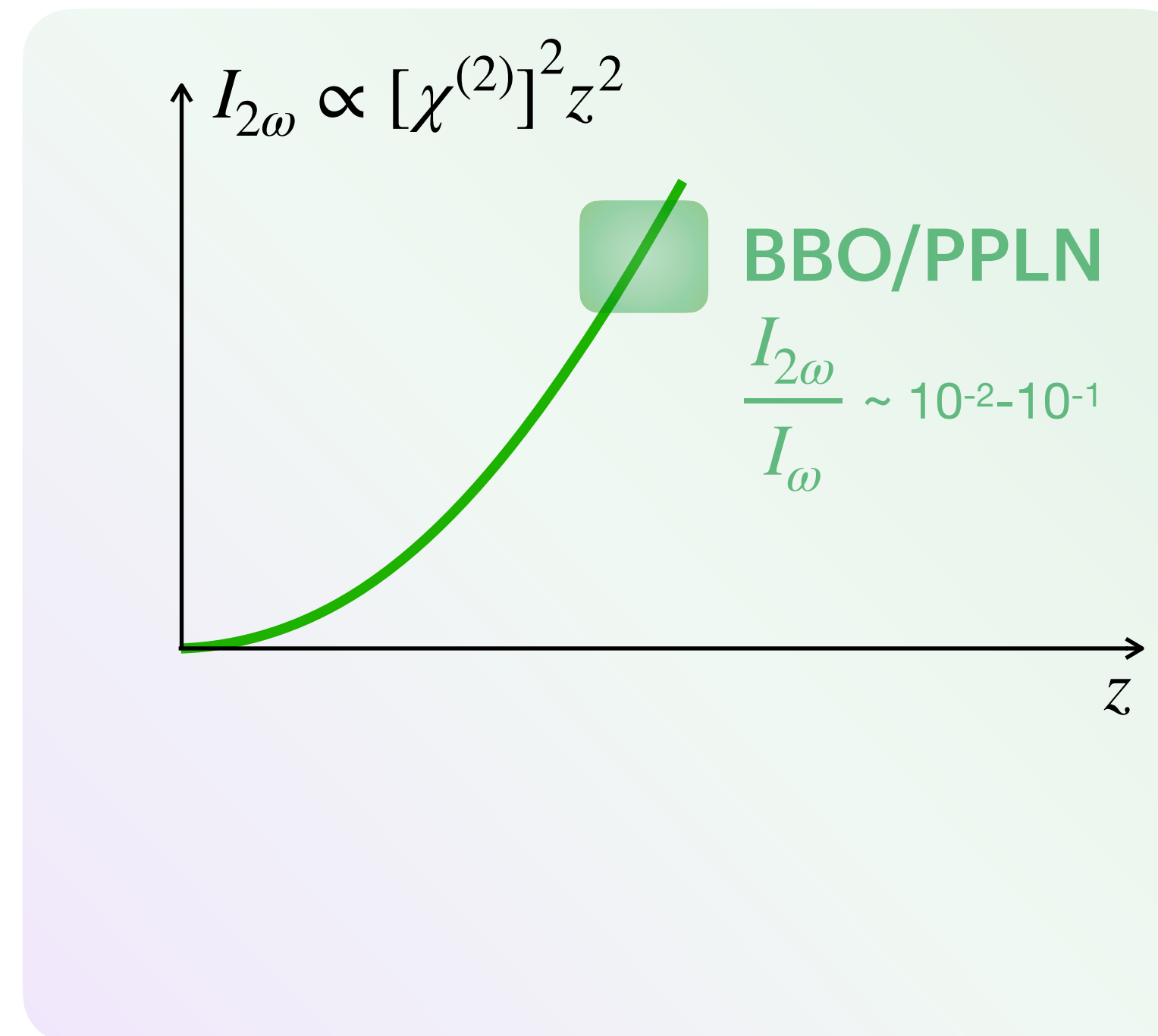
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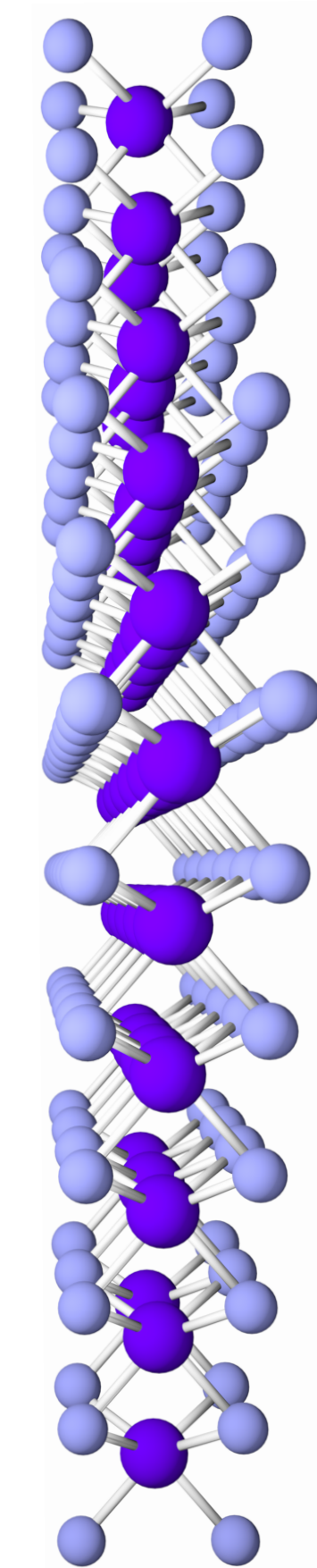
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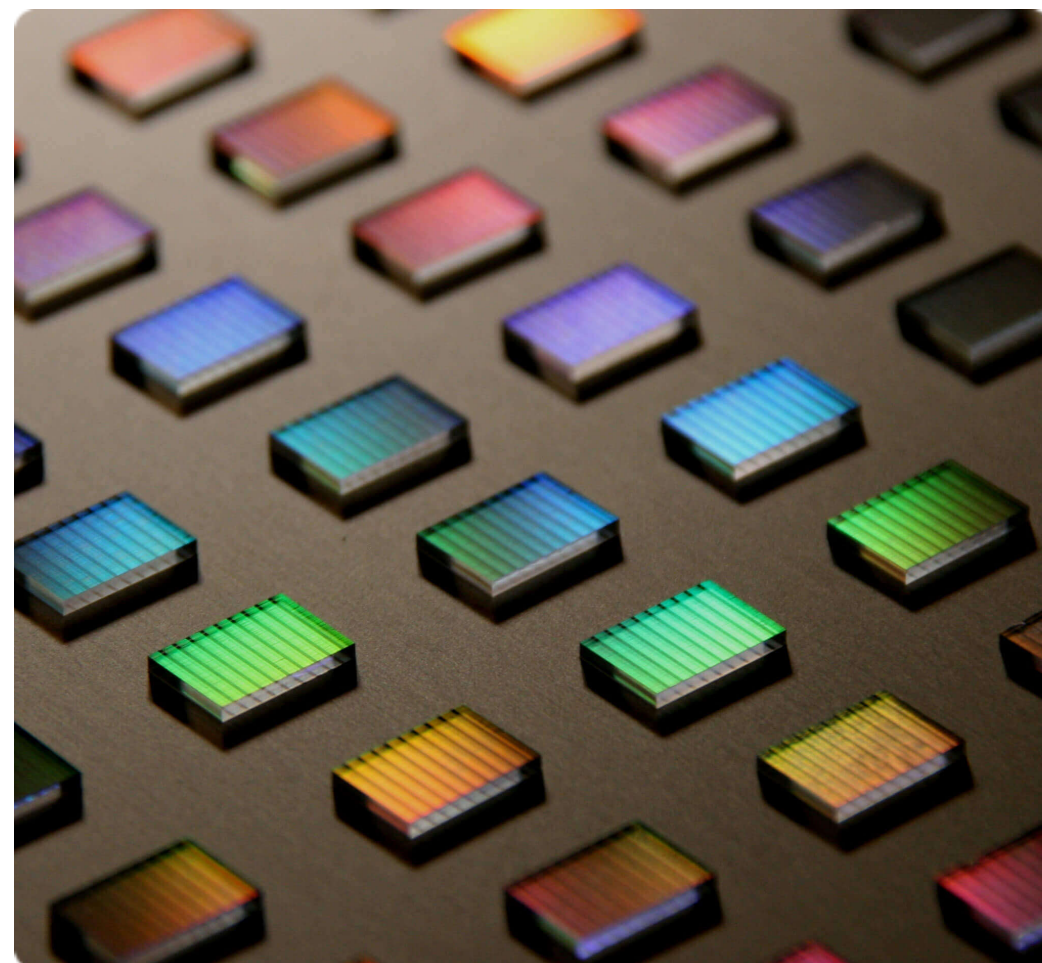
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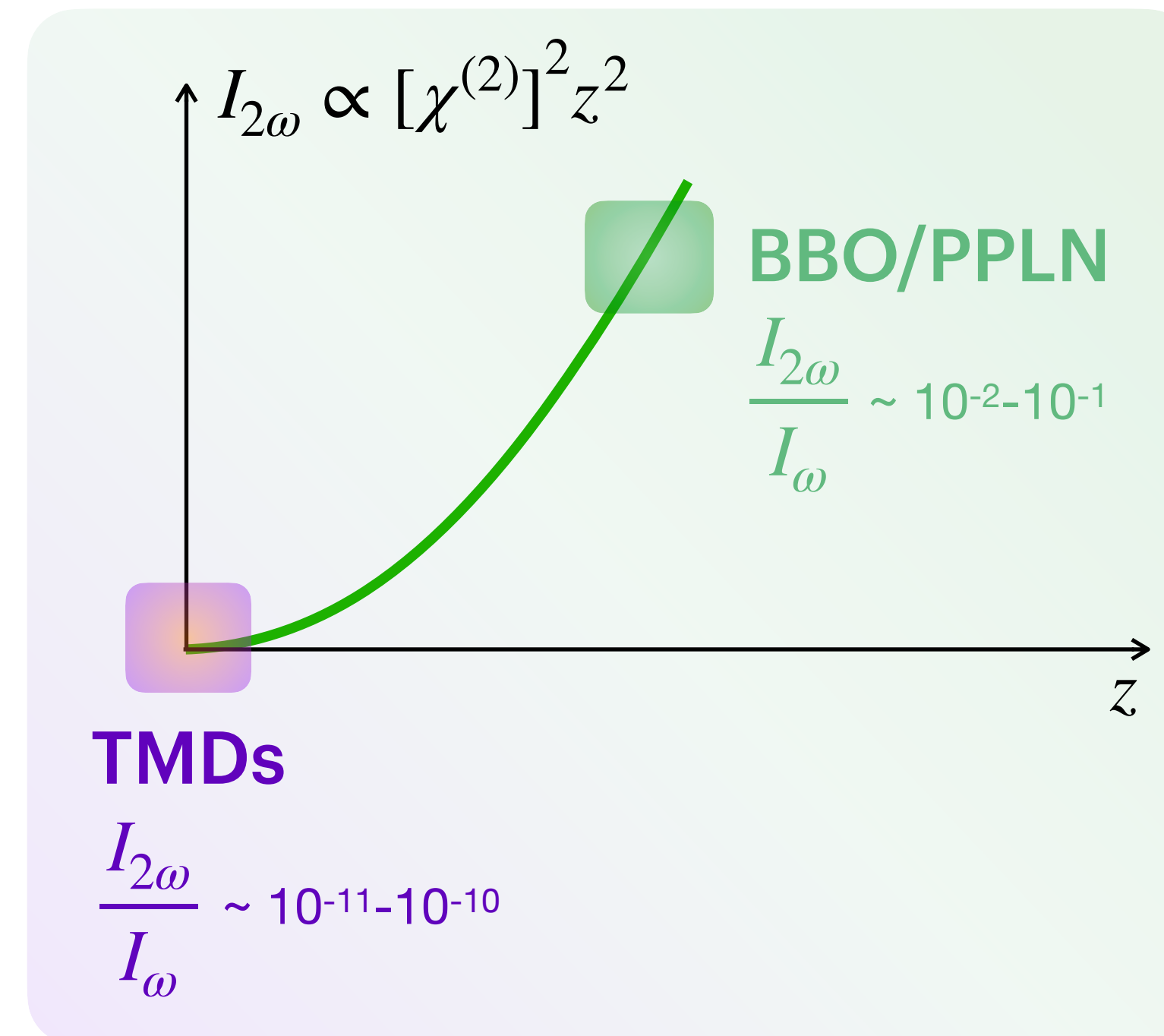
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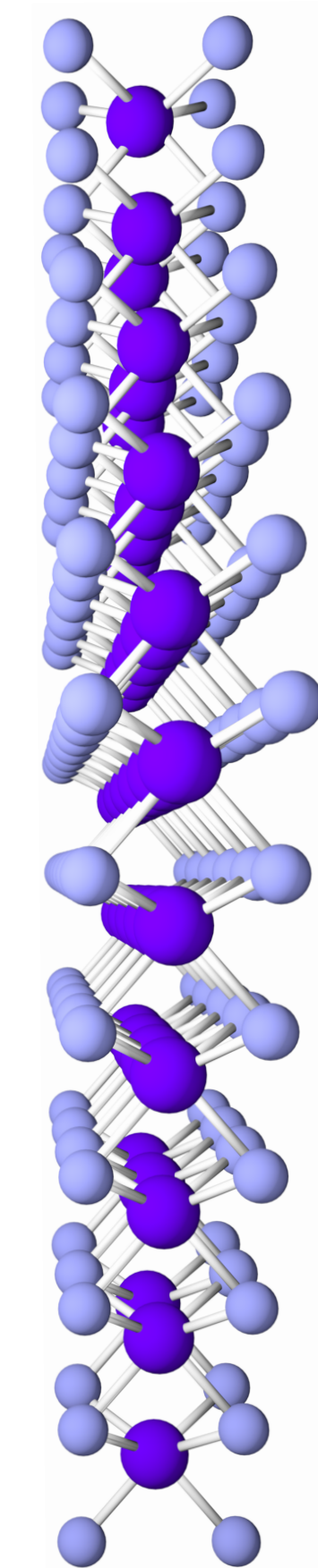
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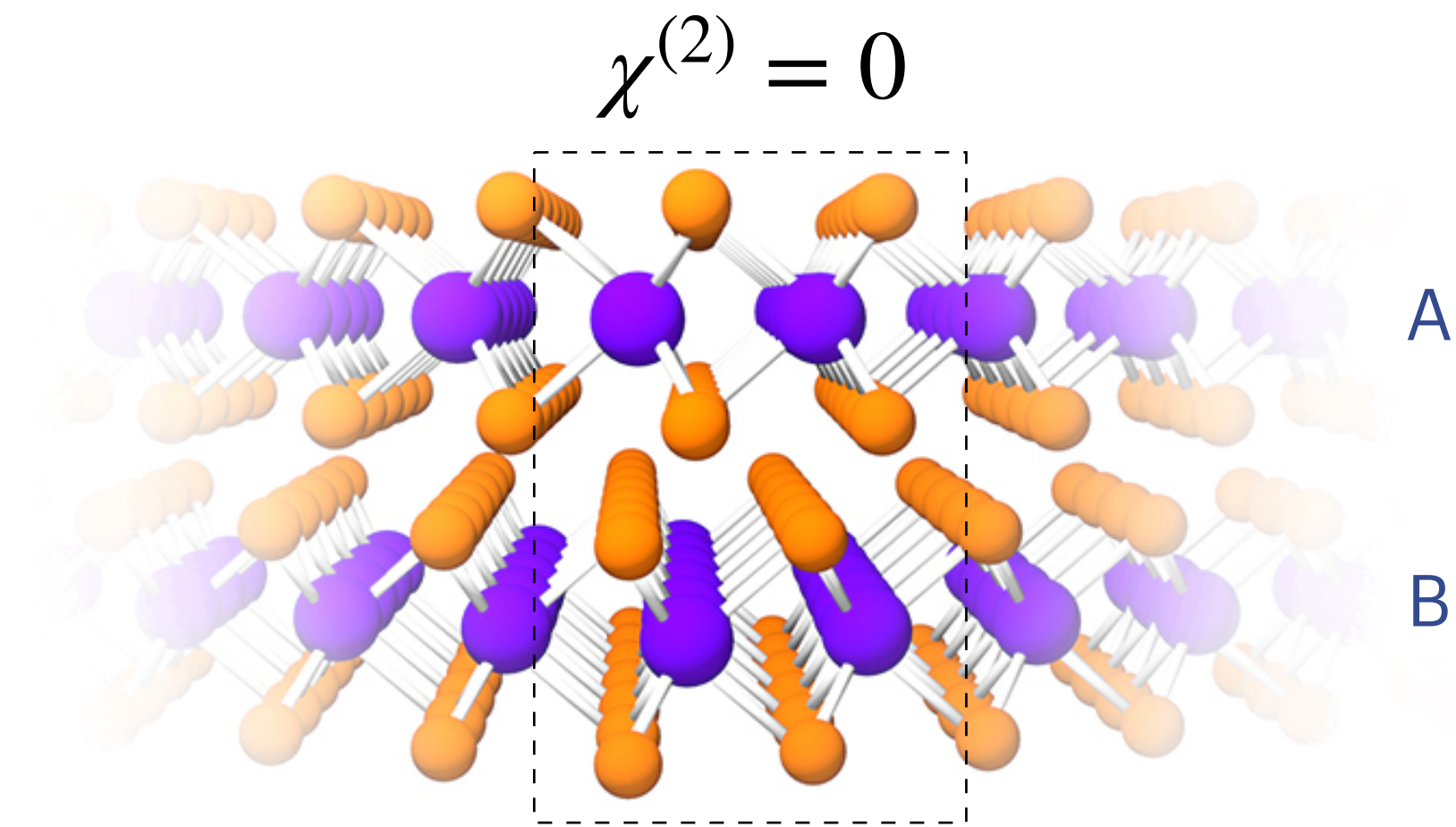
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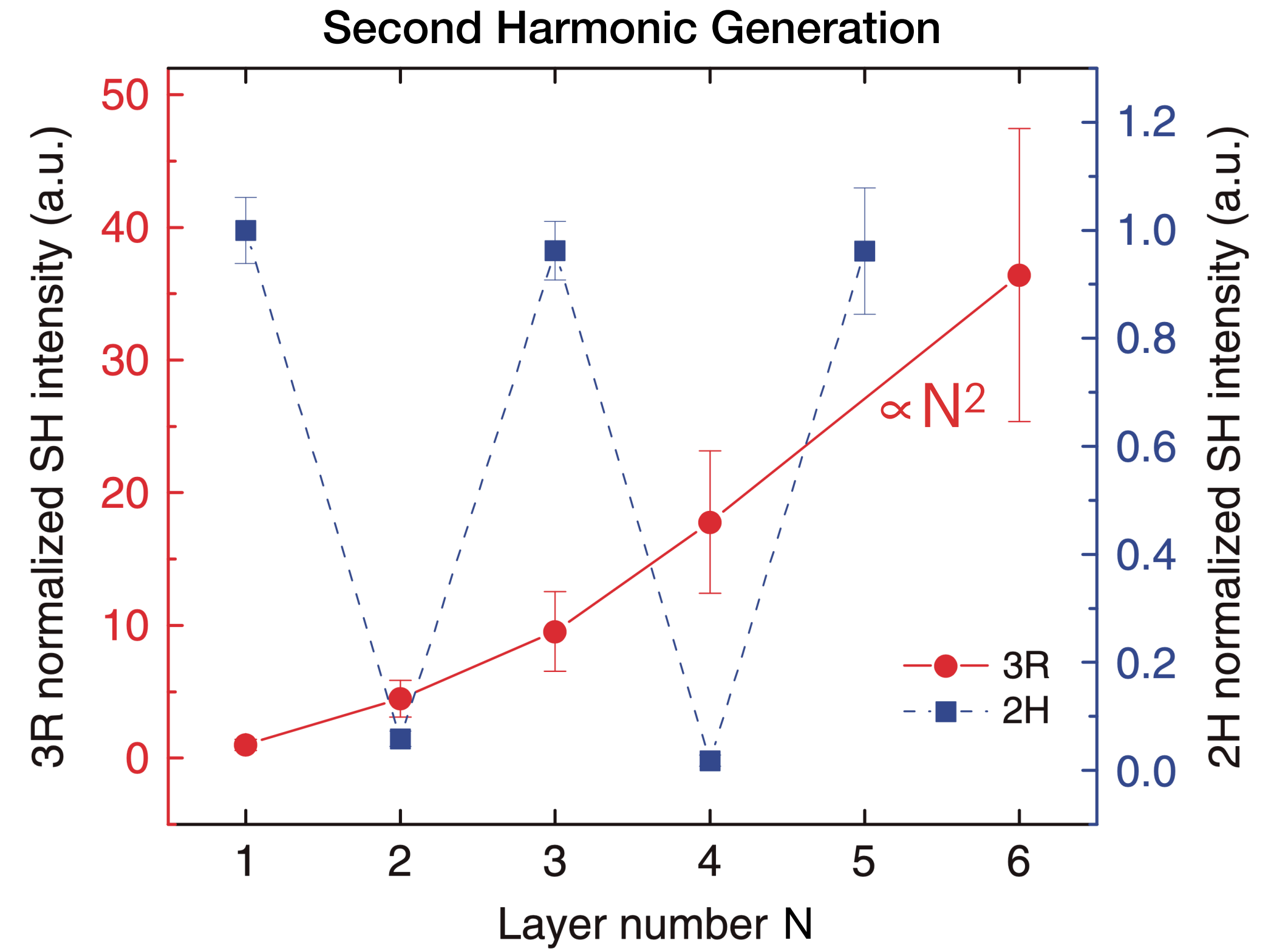
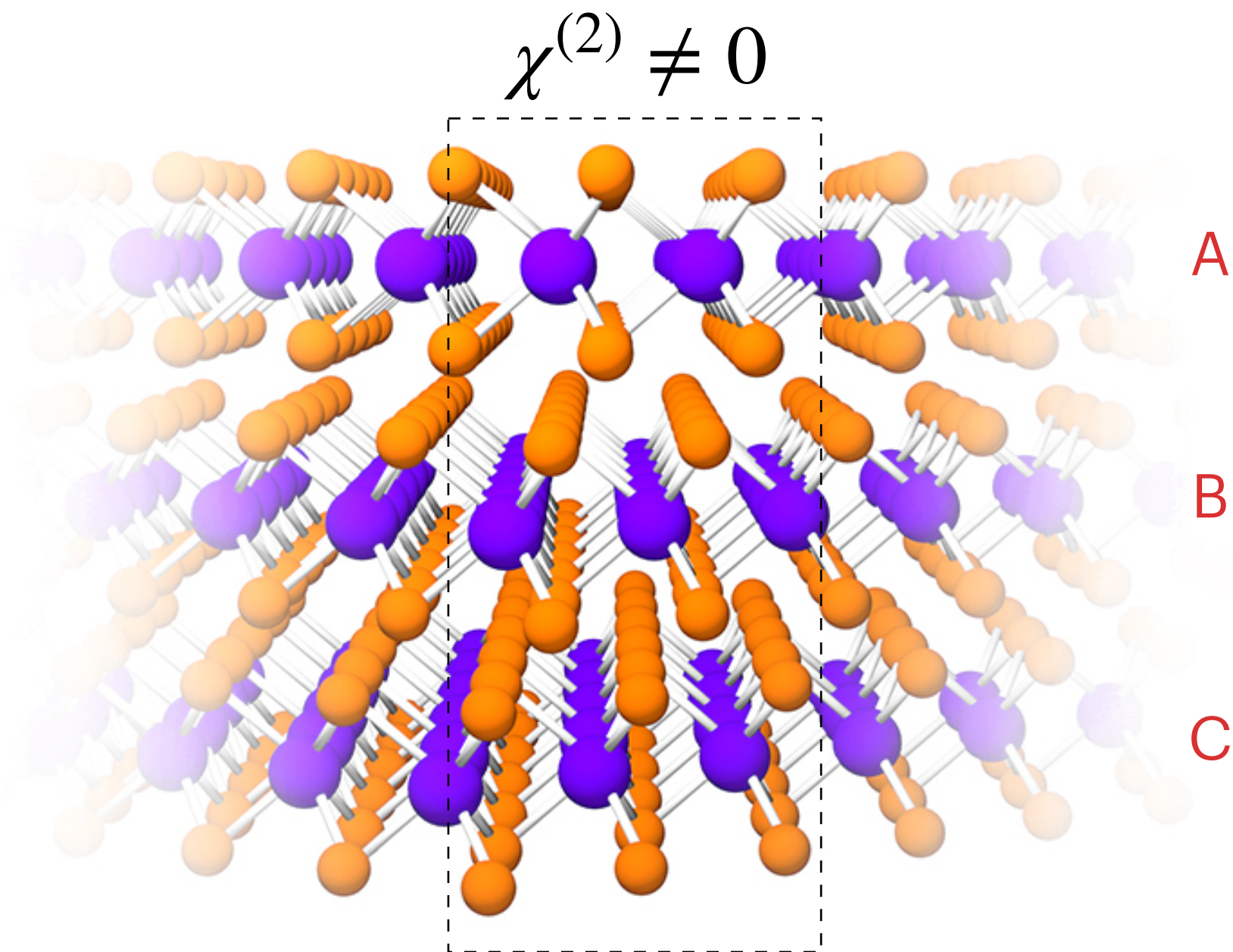
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TMD crystal structure-dependent SHG

2H

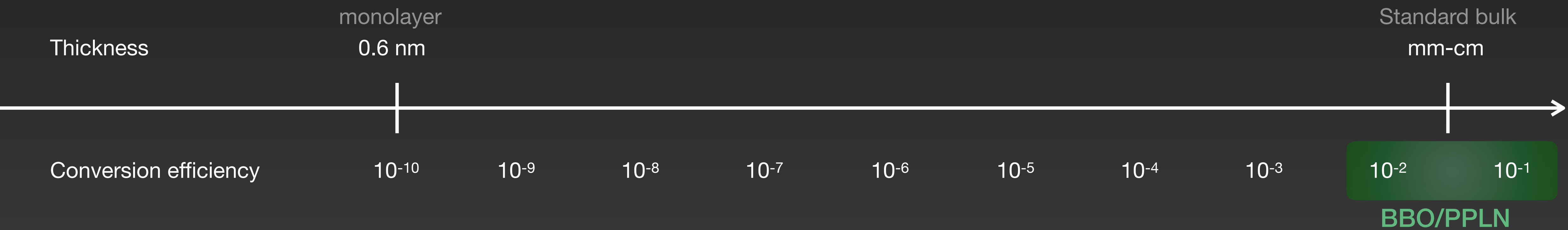


3R

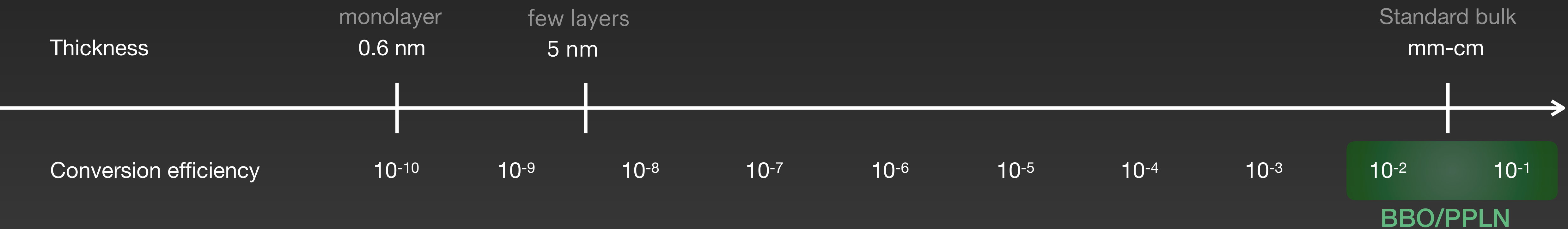


M. Zhao et al. *Light Sci. App.*, **5**, e16131 (2016)

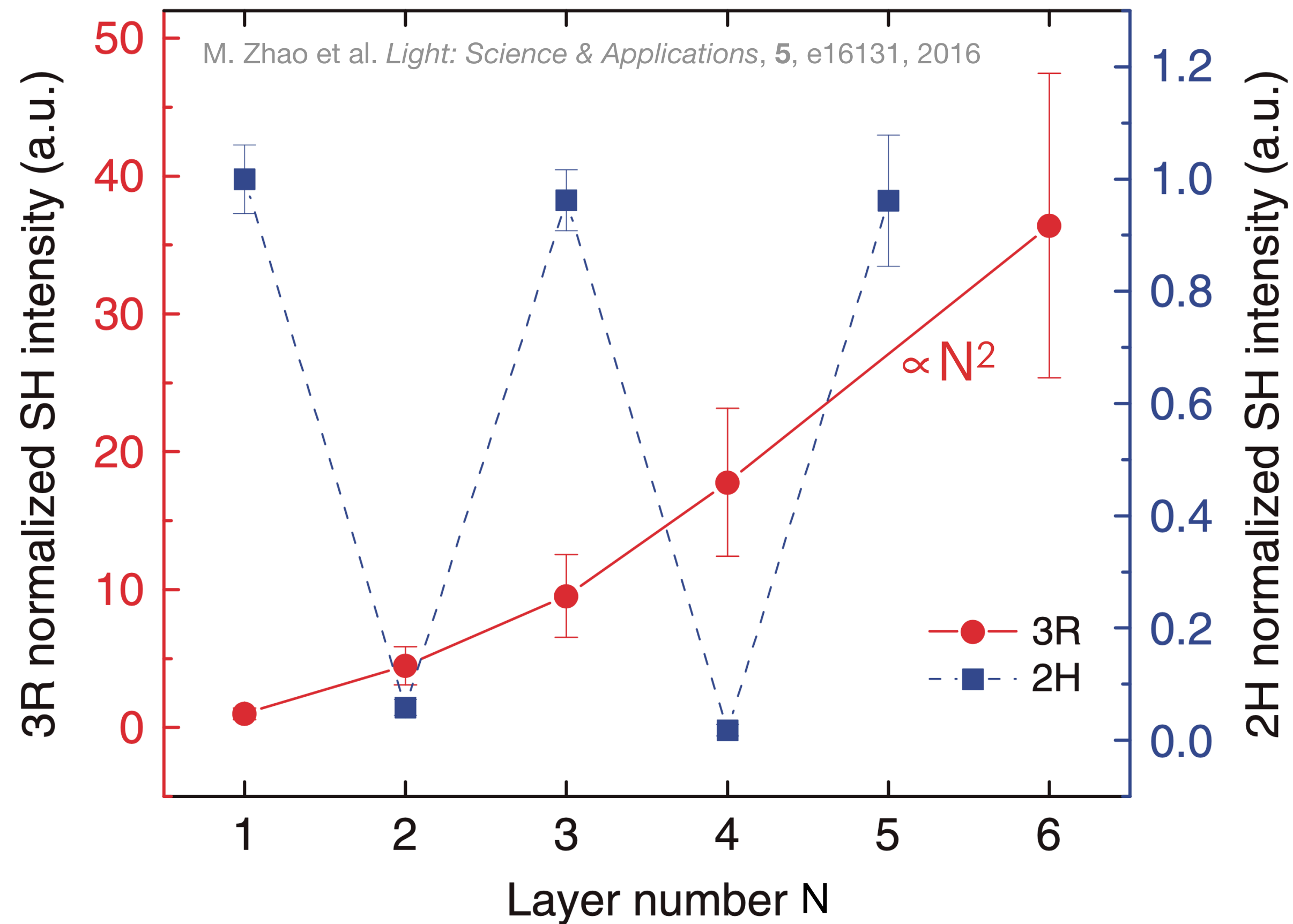
The “conversion efficiency – thickness” arrow



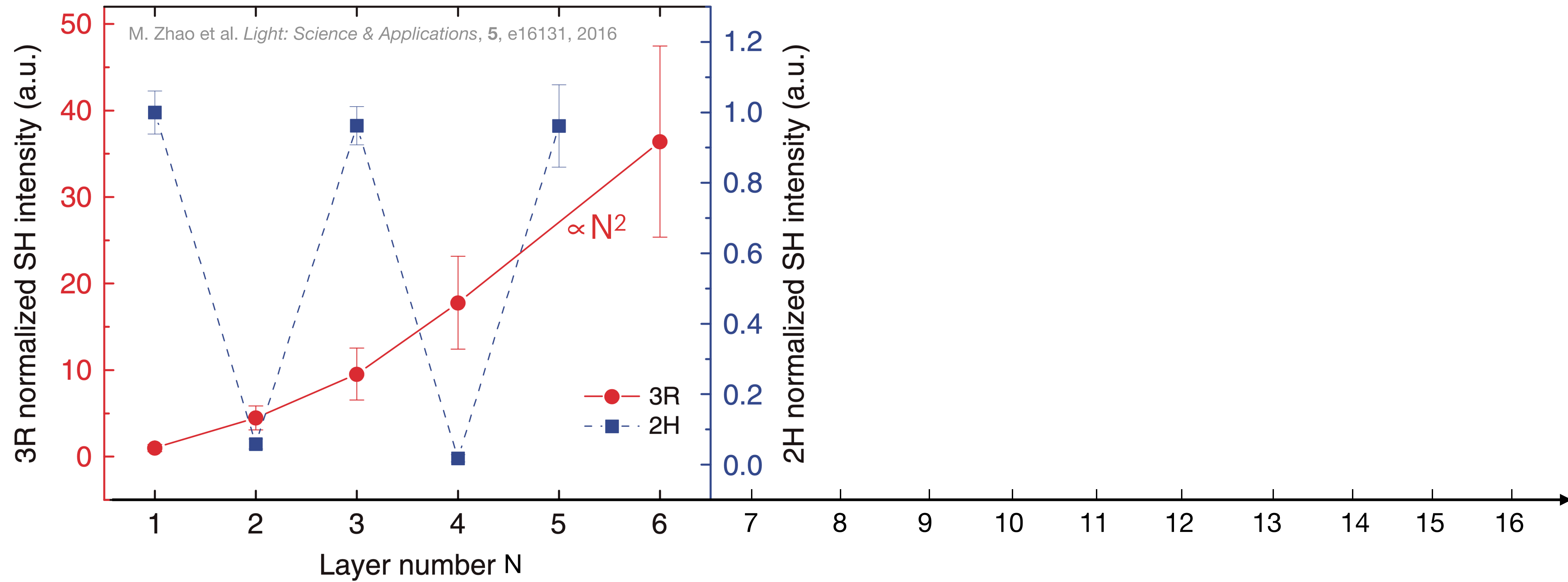
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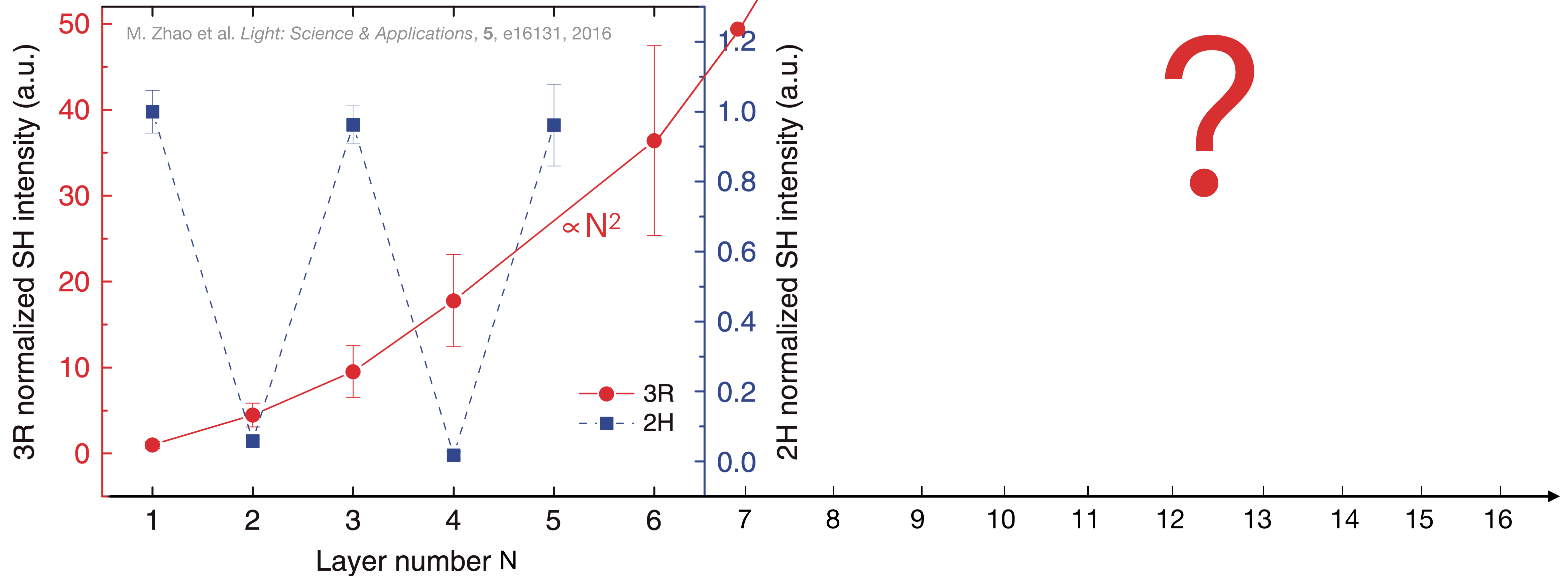
Thickness dependent SHG in 3R-MoS₂



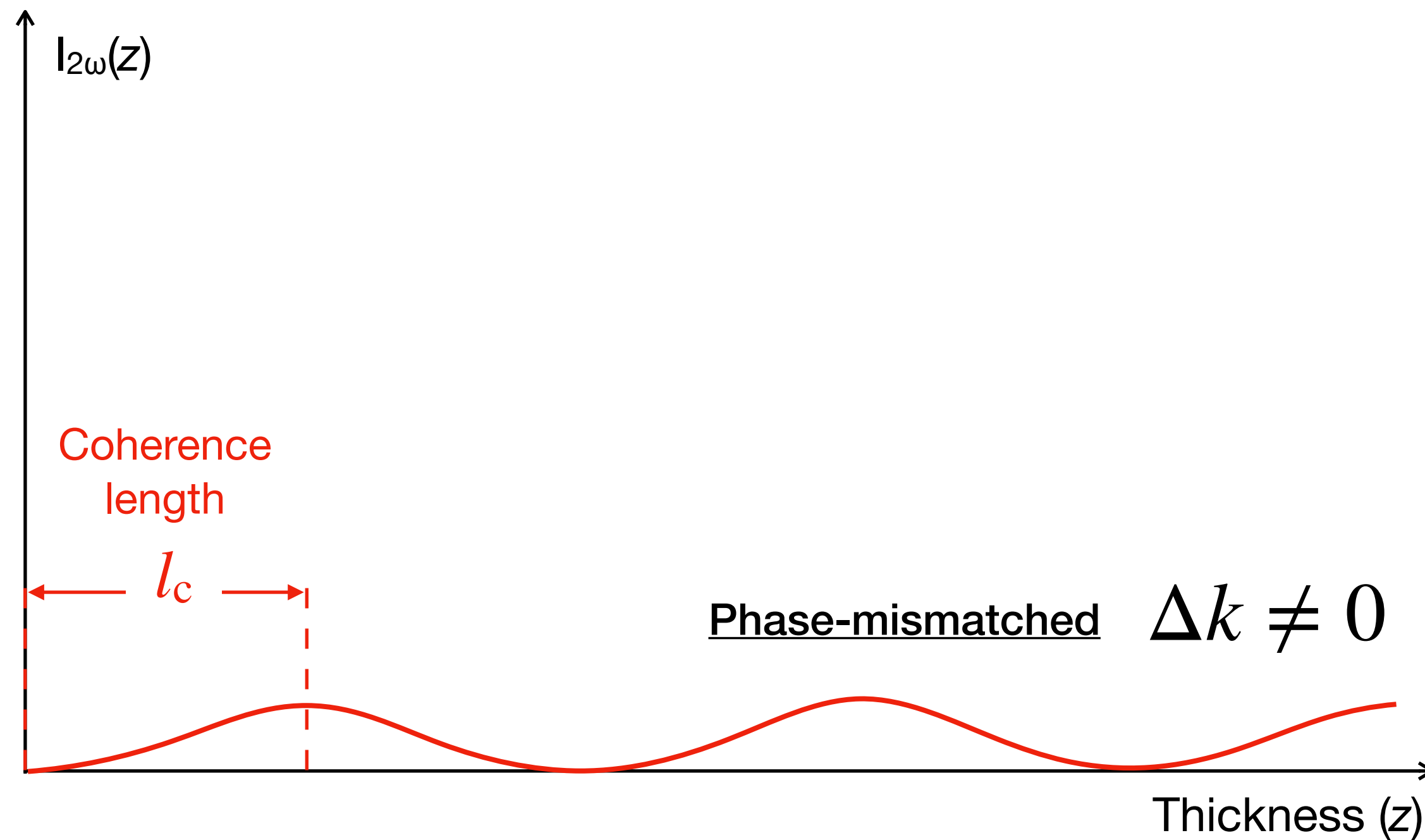
Thickness dependent SHG in 3R-MoS₂



Thickness dependent SHG in 3R-MoS₂



Phase mismatch revealed by the coherence length in 3R-MoS₂



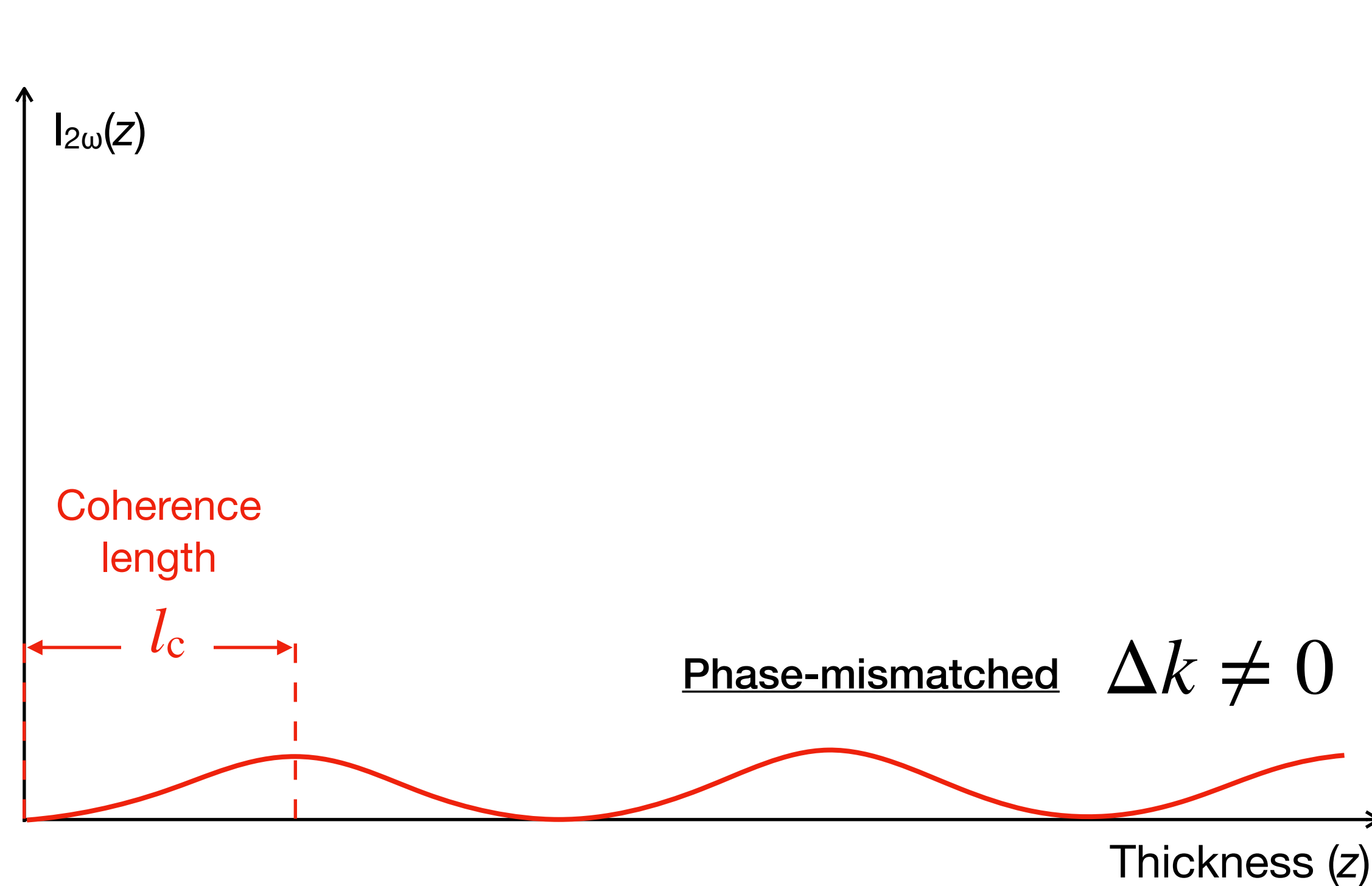
$$I_{2\omega} = \Gamma^2 I_{\omega}^2 \left[\frac{2}{\Delta k} \sin\left(\frac{\Delta k z}{2}\right) \right]^2$$

$$\Gamma^2 = \frac{2\omega^2 [\chi^{(2)}]^2}{4\epsilon_0 c^3 n_{\omega}^3}$$

Phase mismatch

$$\begin{aligned} \Delta k &= k_{2\omega} - 2k_{\omega} \\ &= \frac{2\omega}{c} (n_{2\omega} - n_{\omega}) \end{aligned}$$

Phase mismatch revealed by the coherence length in 3R-MoS₂

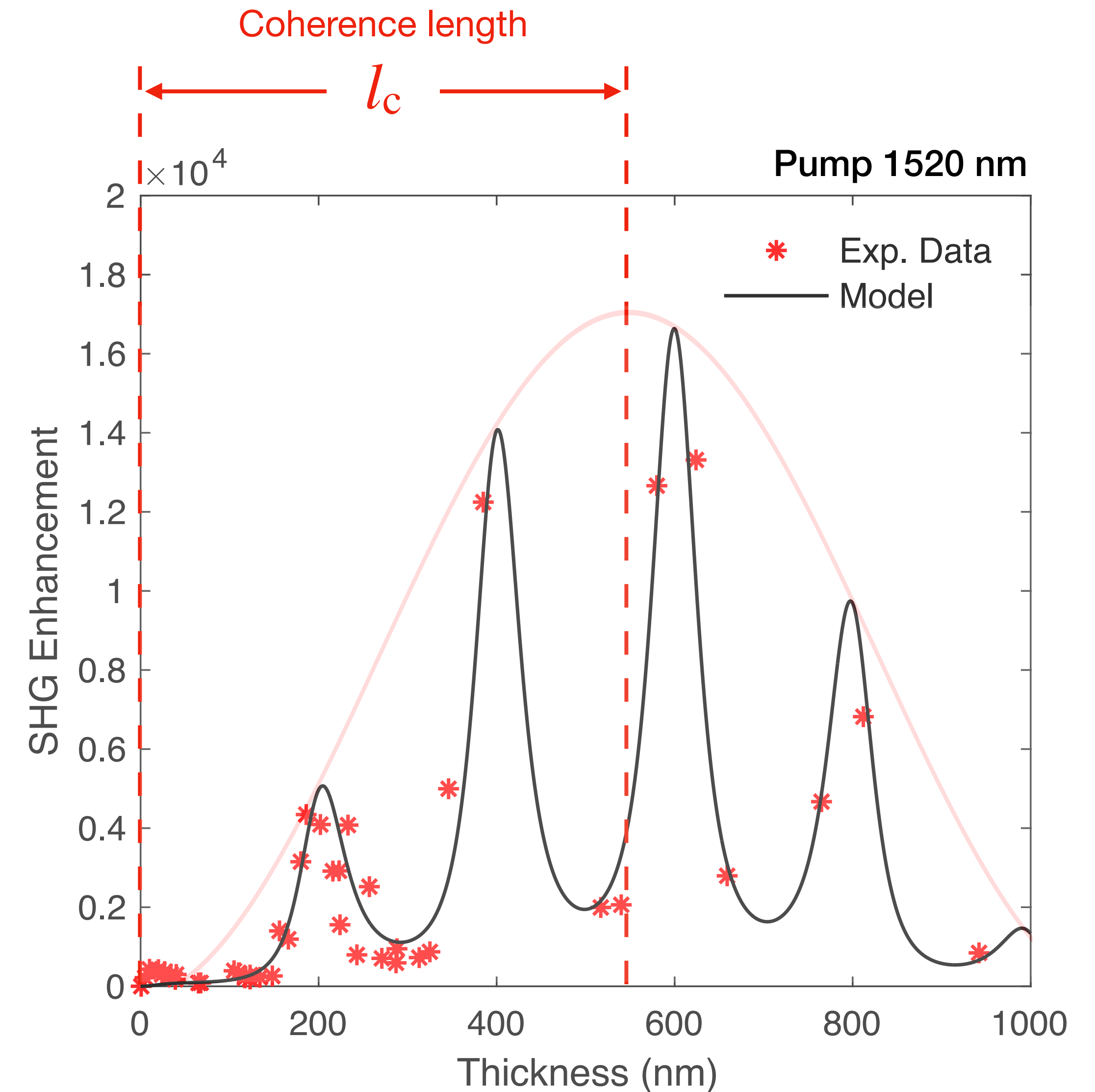


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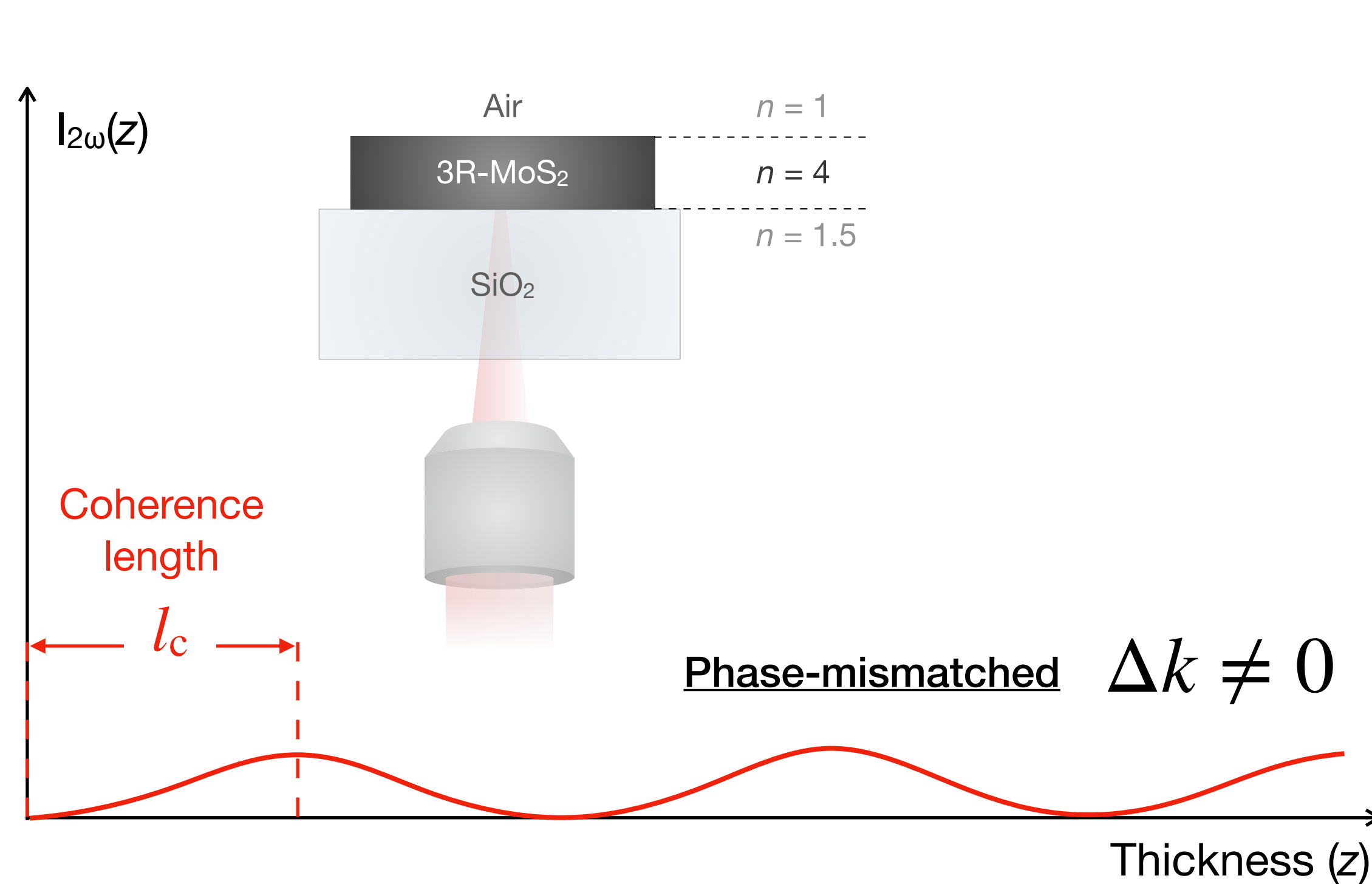
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Xu, X., Trovatiello, C., Mooshammer, F. et al. *Nature Photonics*, **16**, 698–706 (2022)

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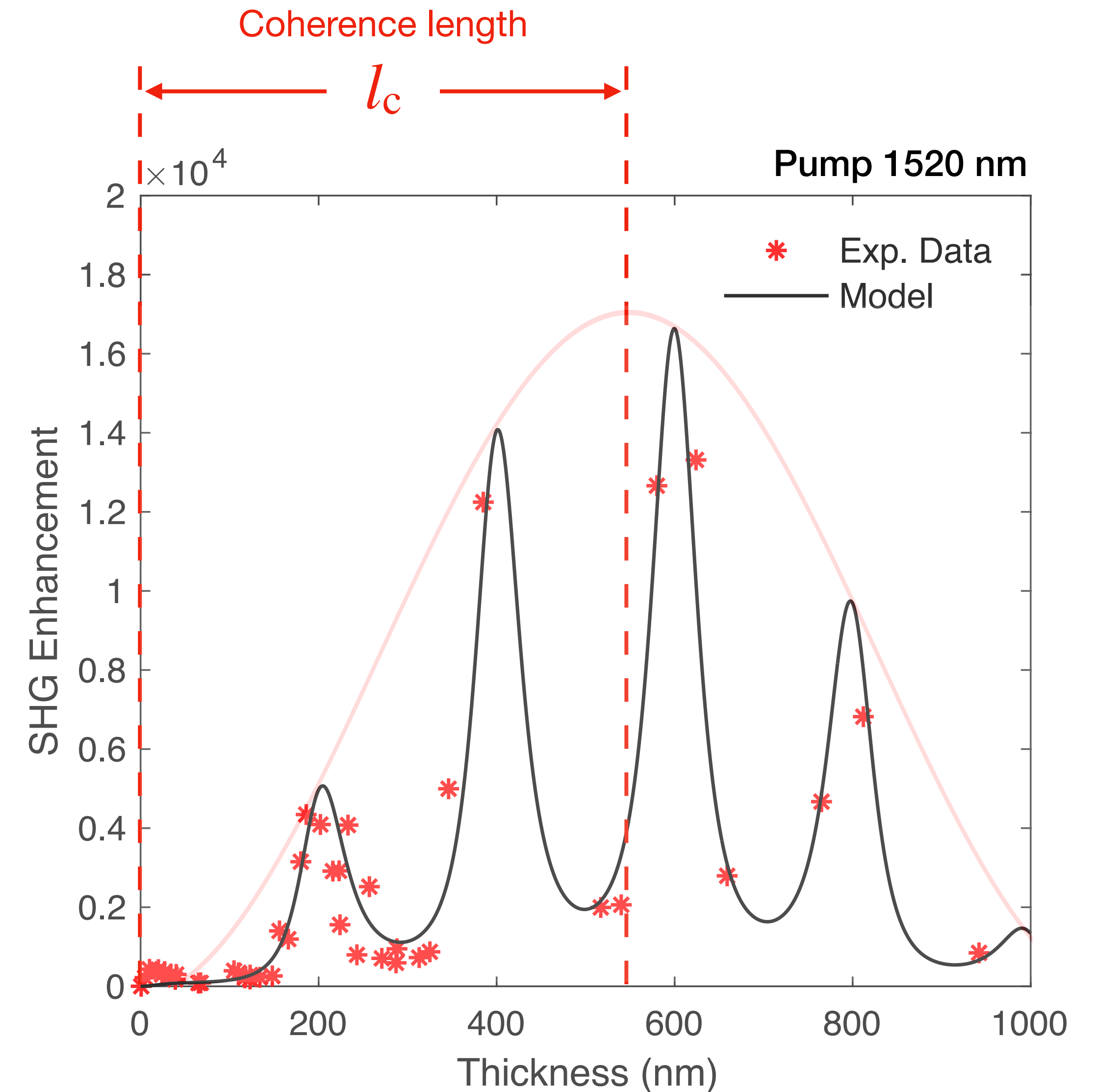


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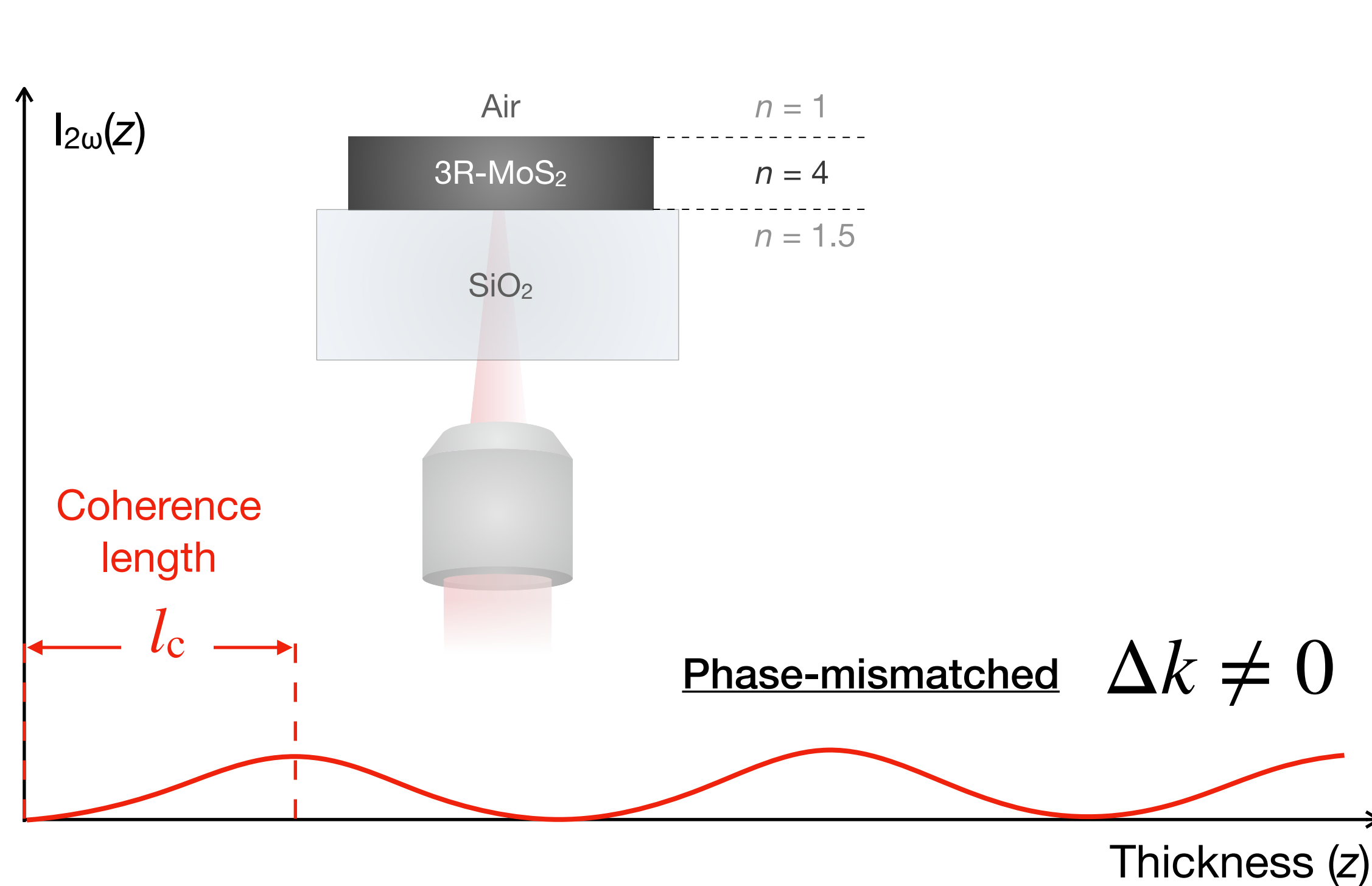
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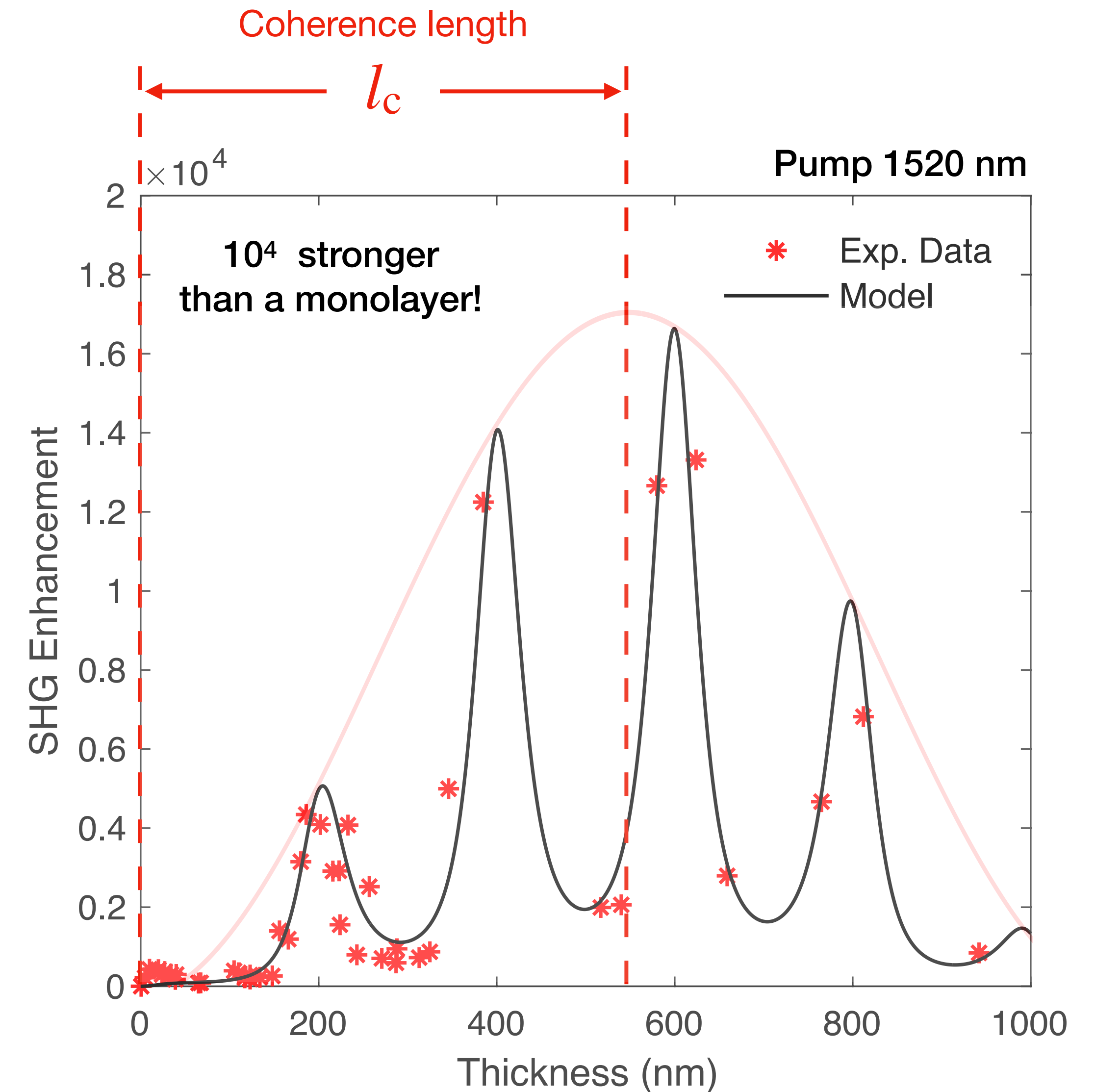


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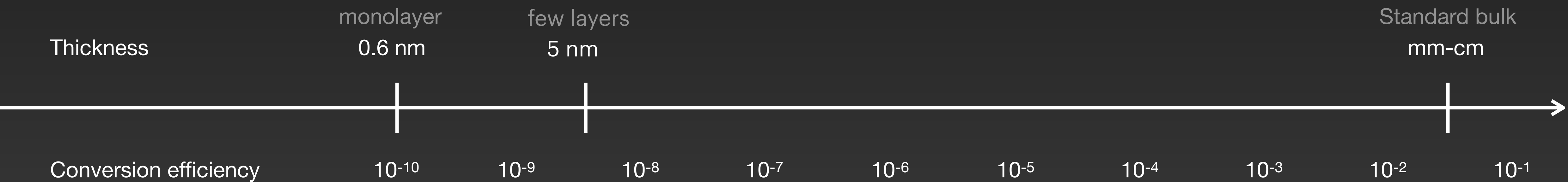
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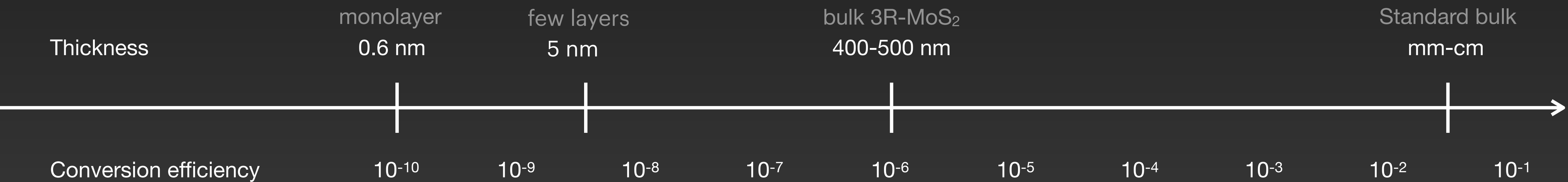


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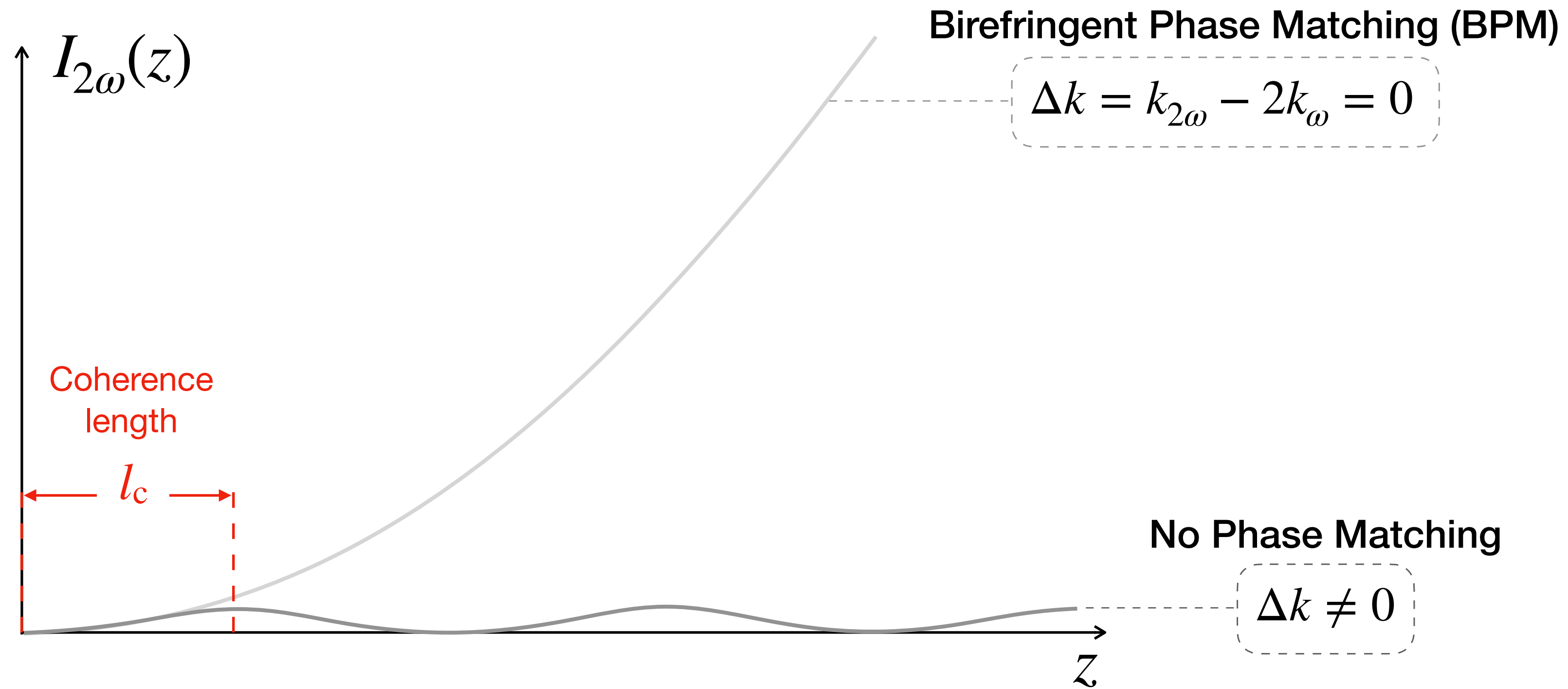
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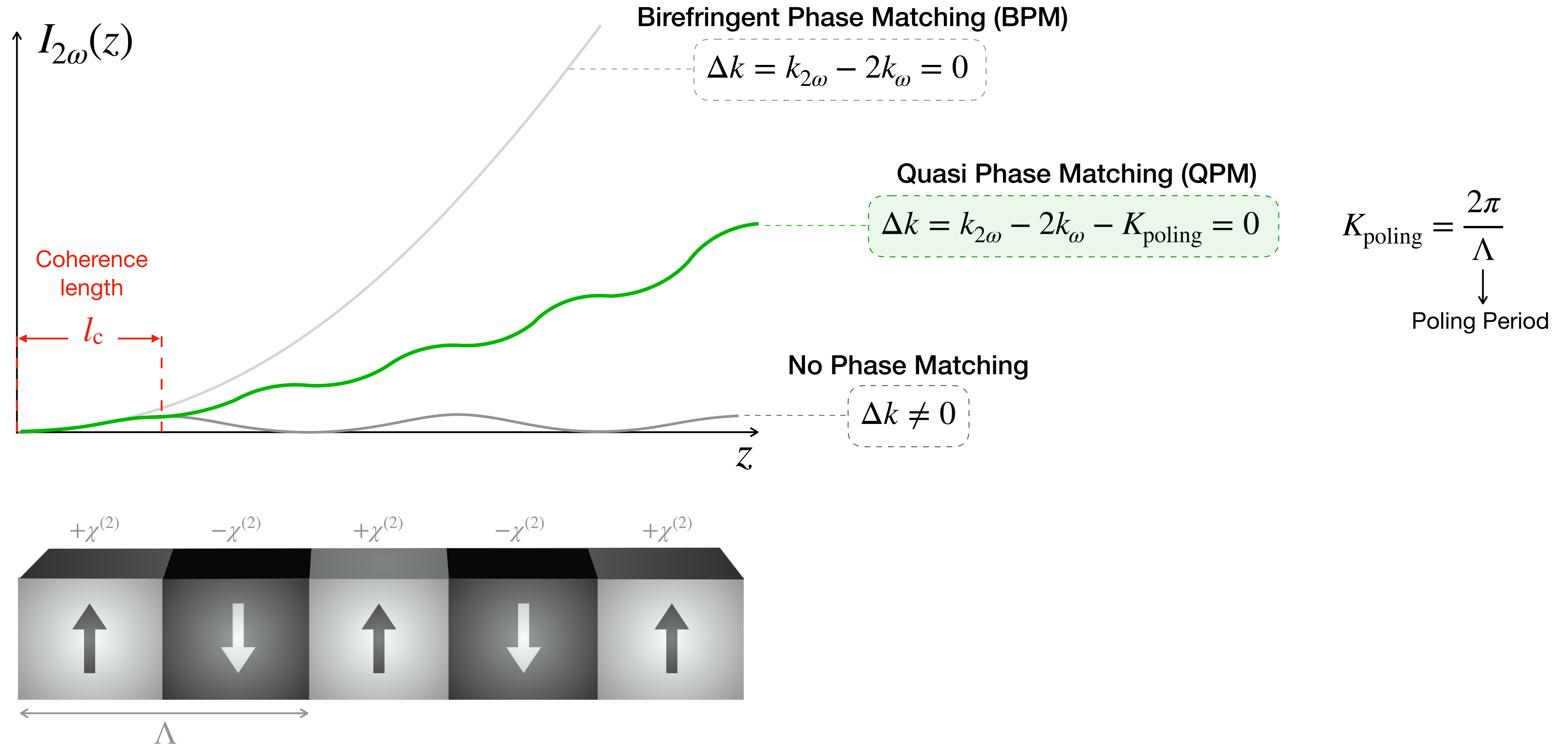


Phase matching



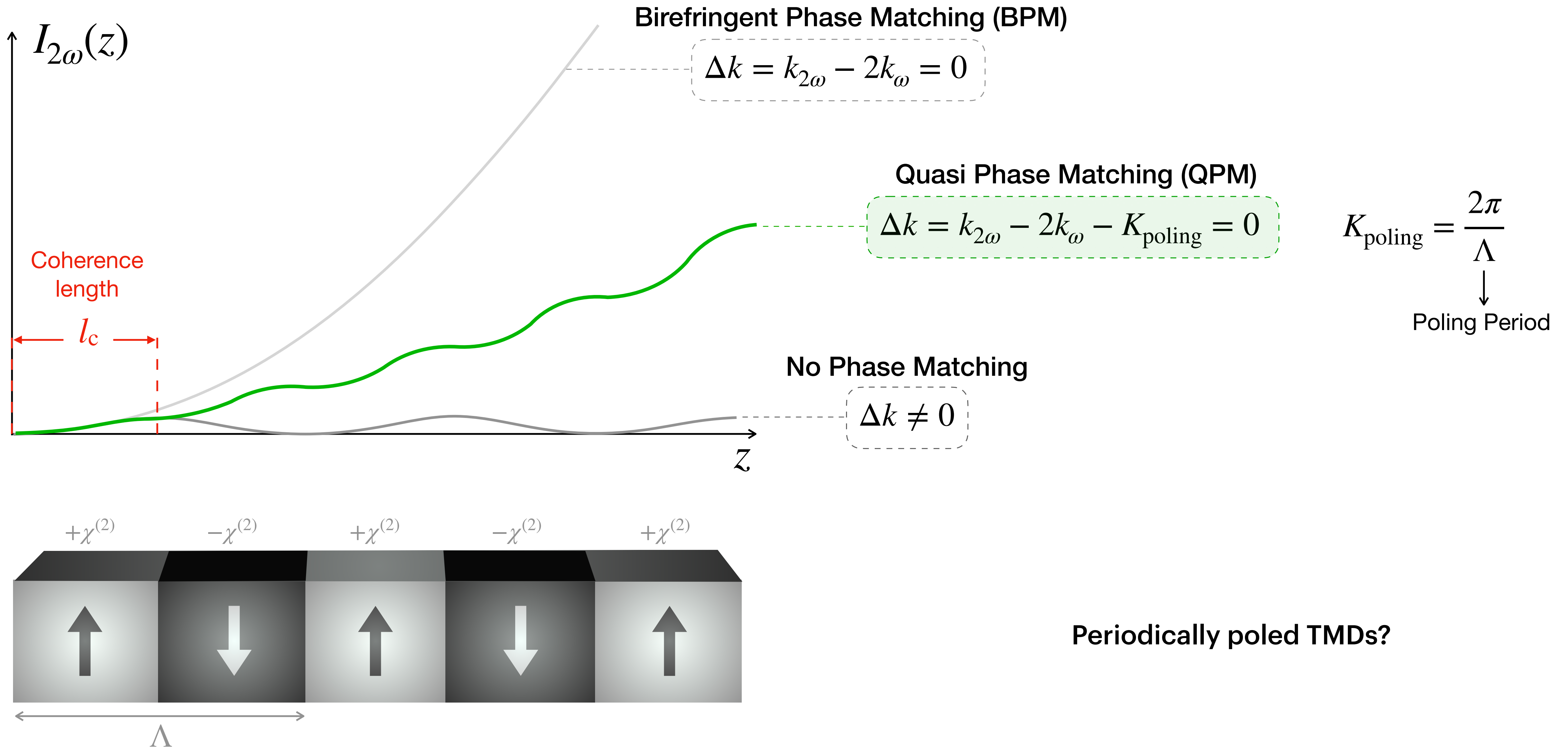
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Phase matching



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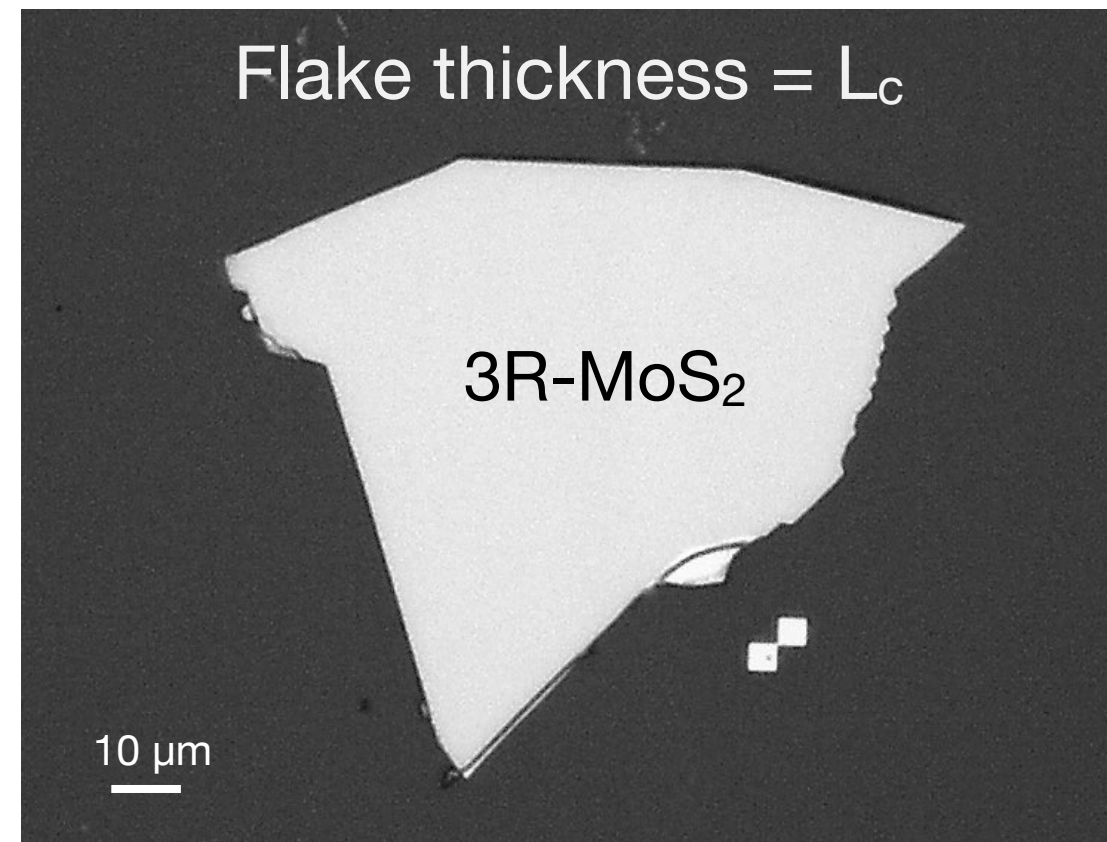
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Introducing periodically poled transition metal dichalcogenides (PPTMDs)

Cut and ~~paste~~ twist&stack



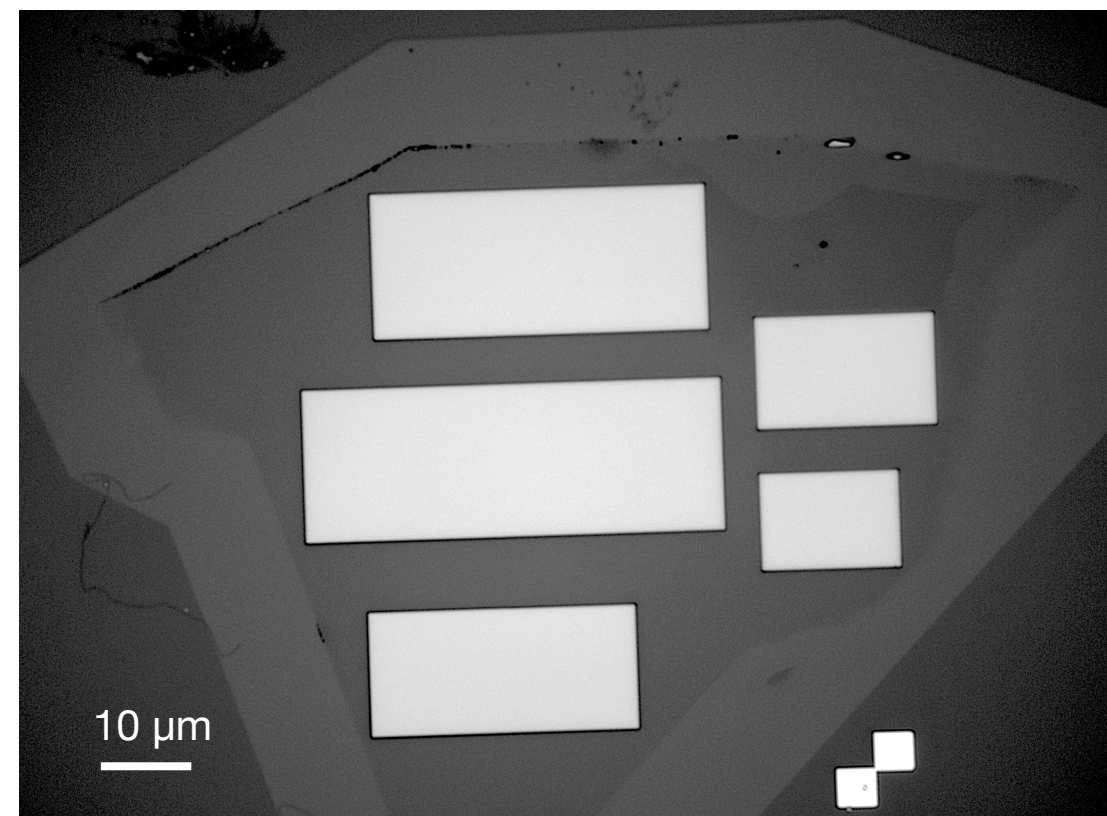
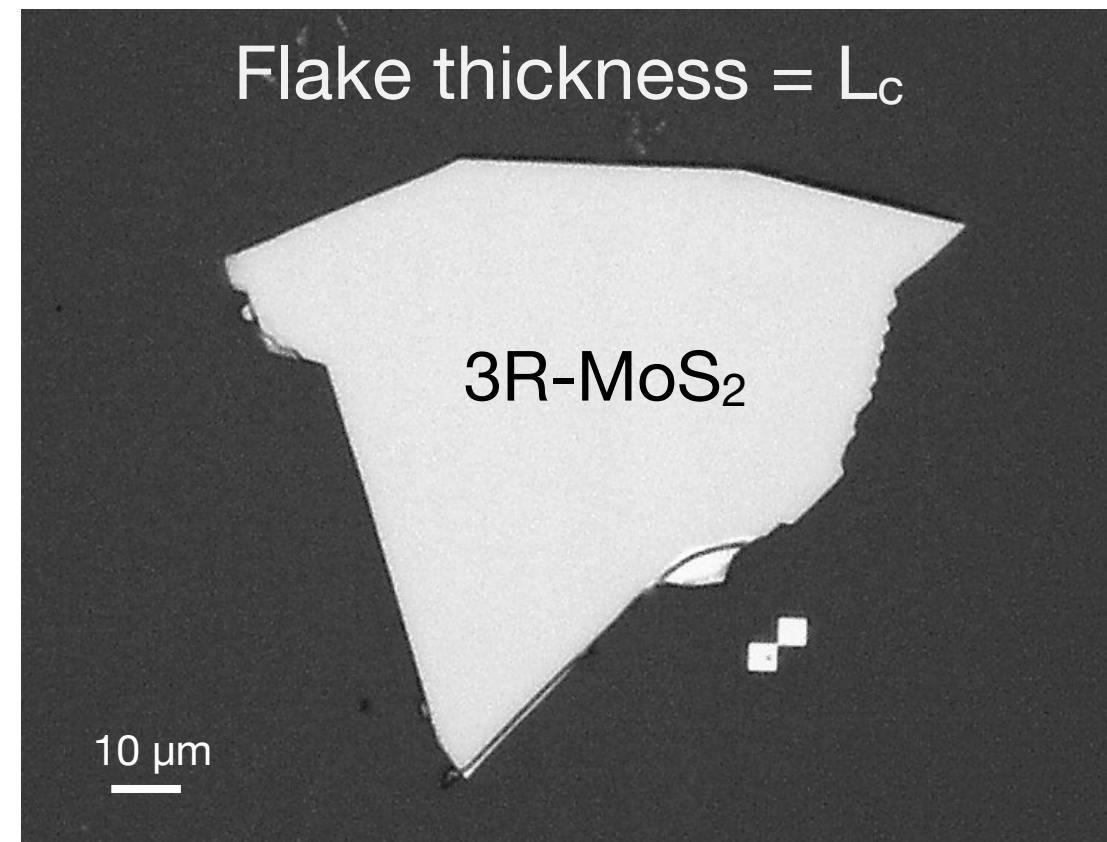
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U.S. Provisional Patent, Application No. 63/607, 309

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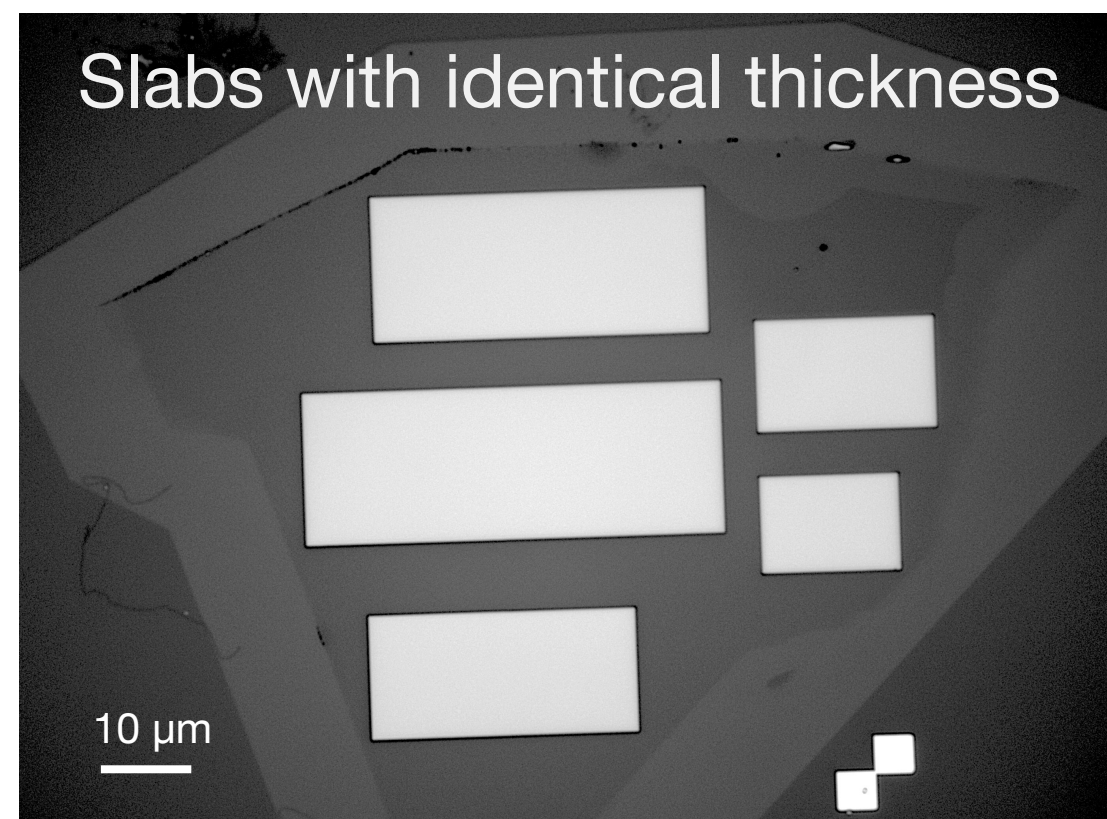
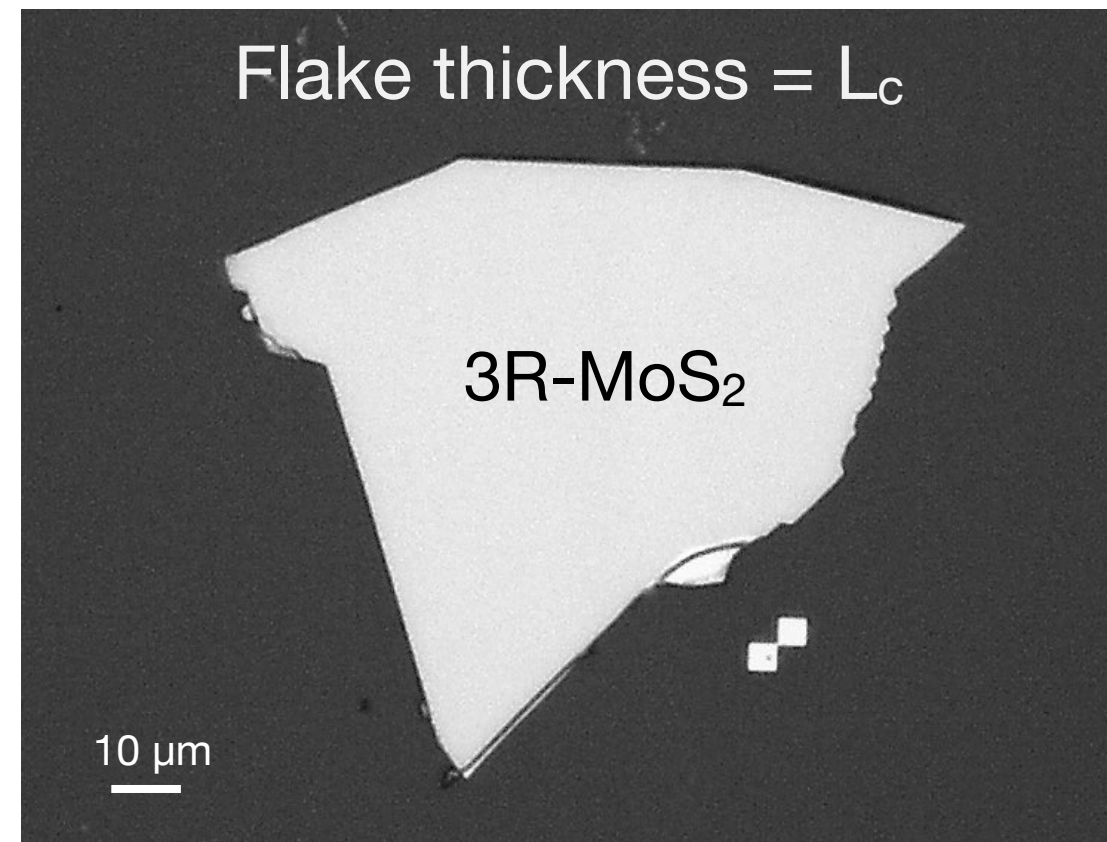
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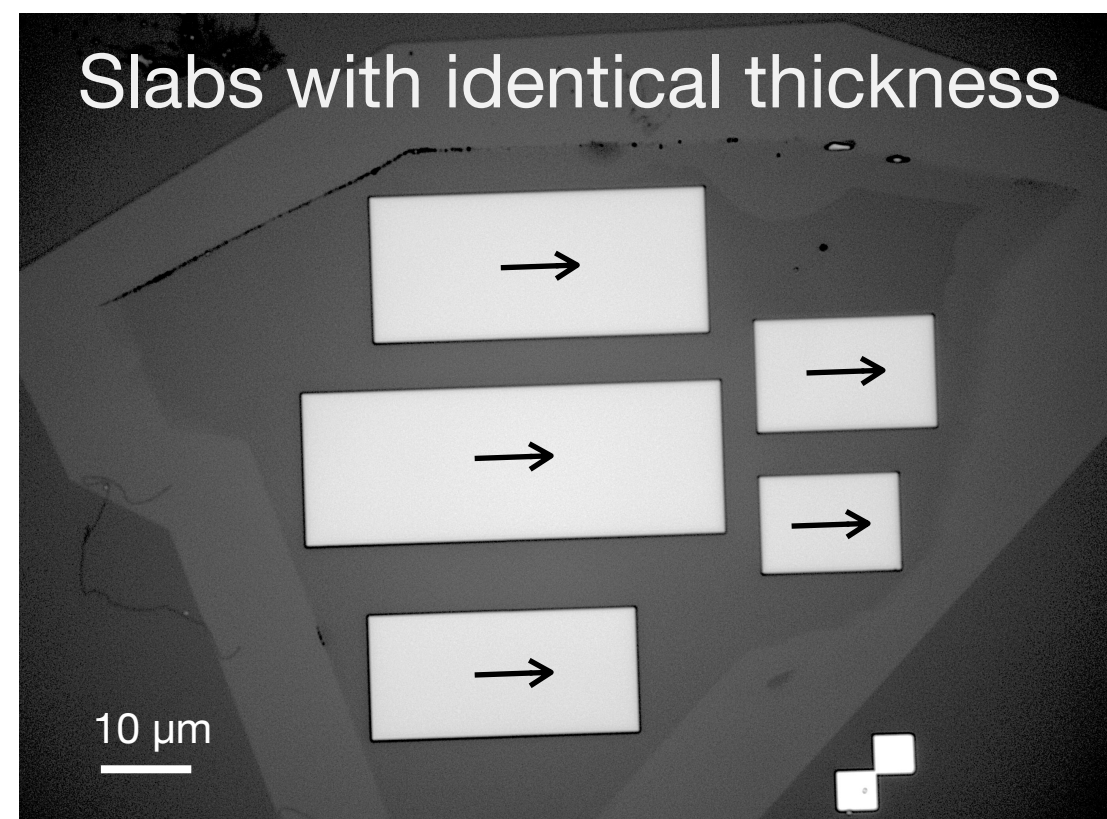
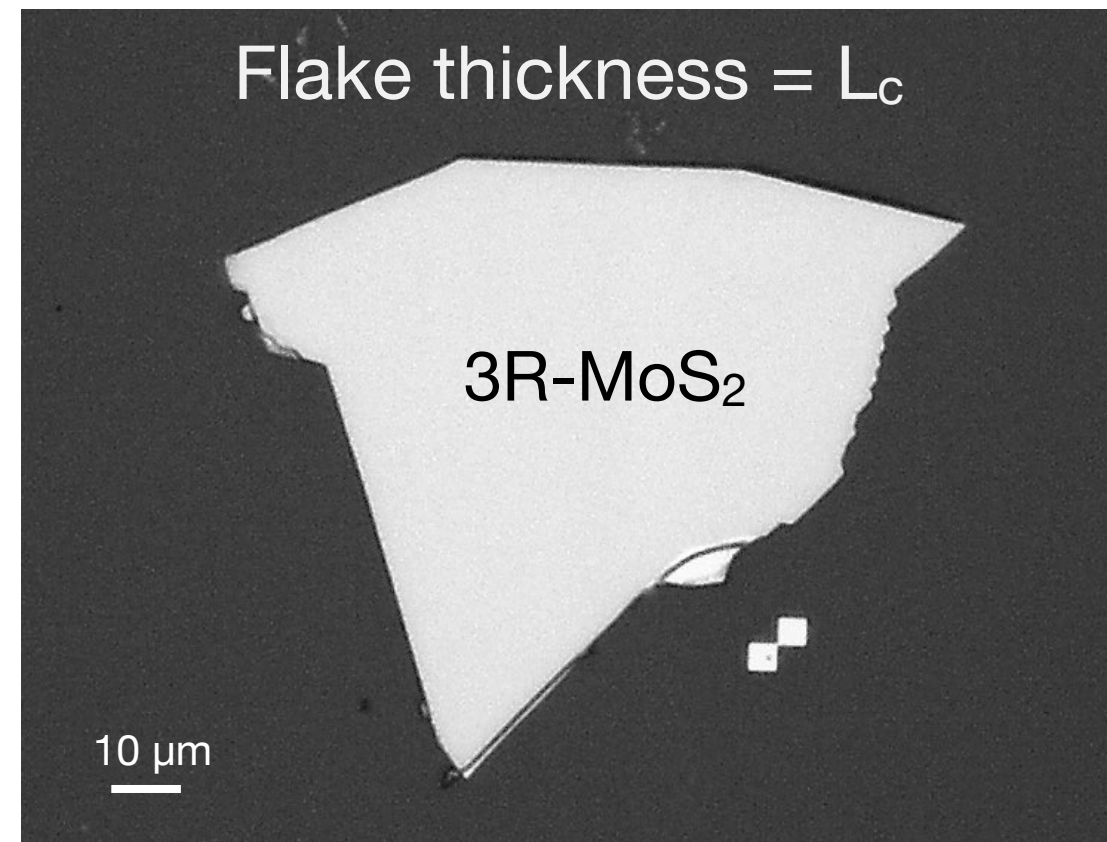
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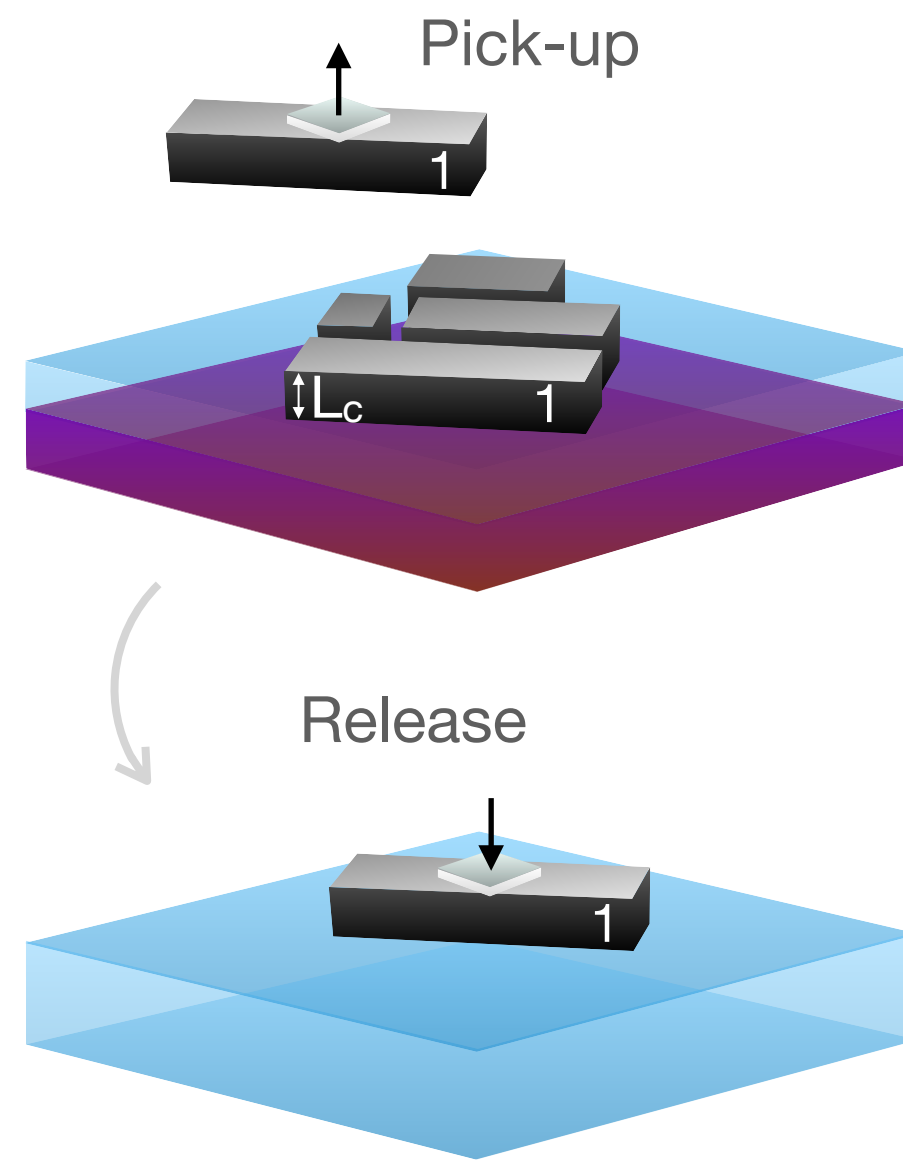
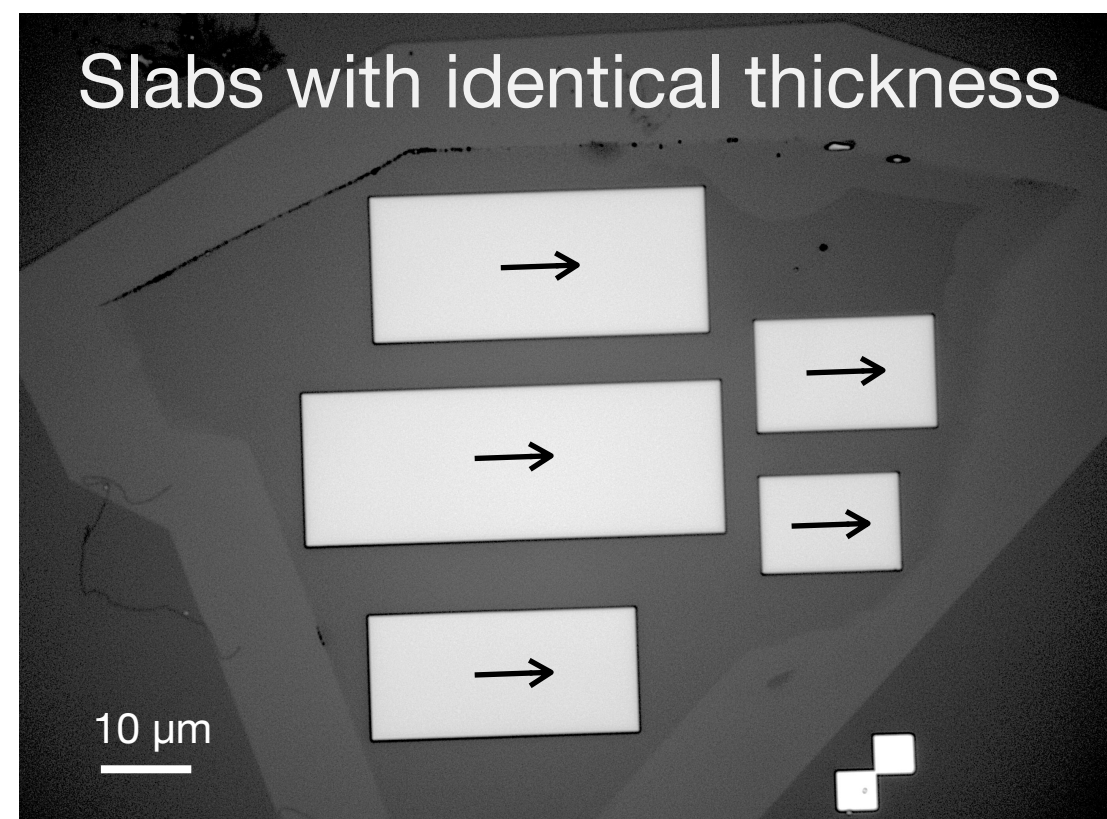
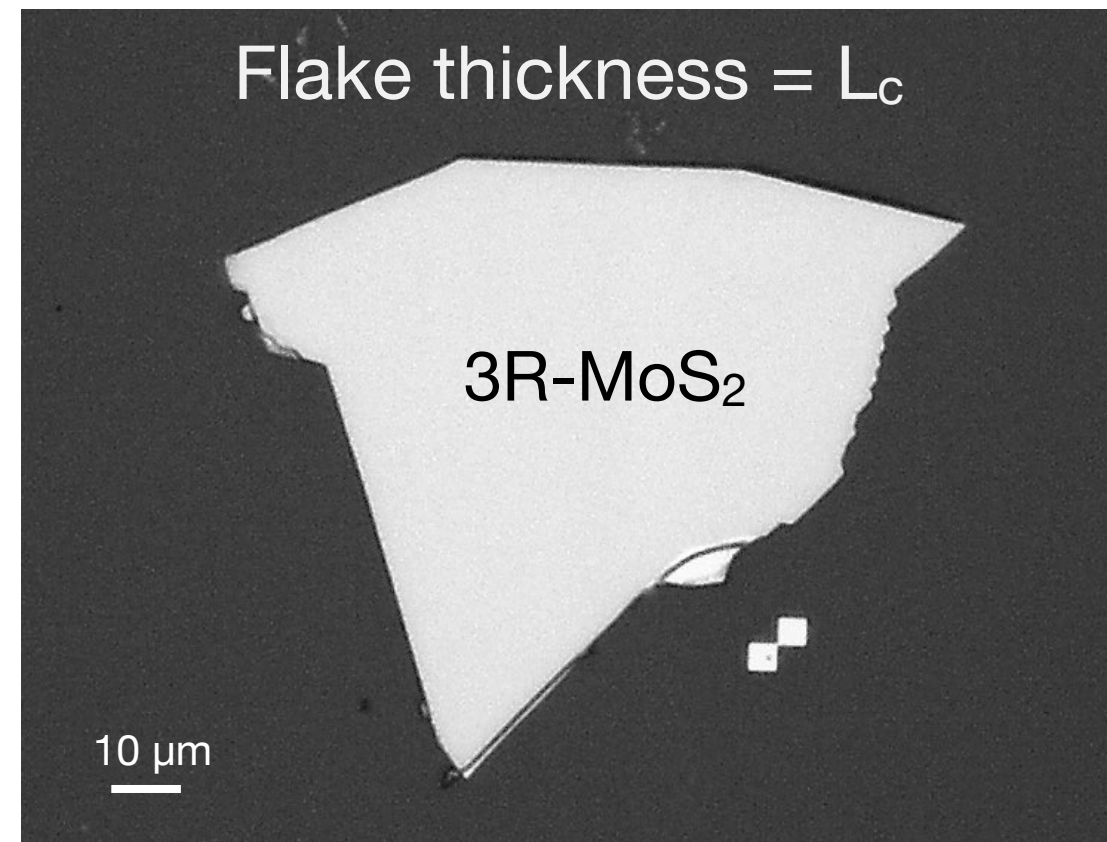
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Introducing periodically poled transition metal dichalcogenides (PPTMDs)

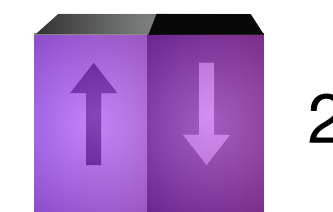
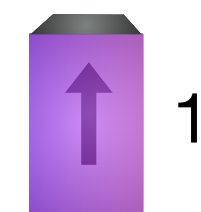
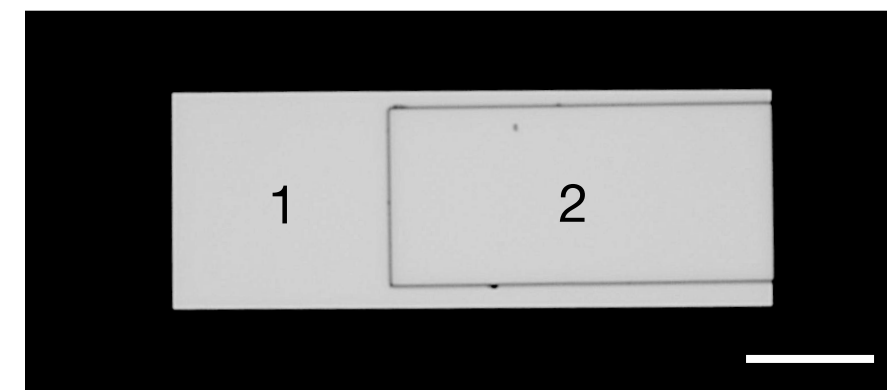
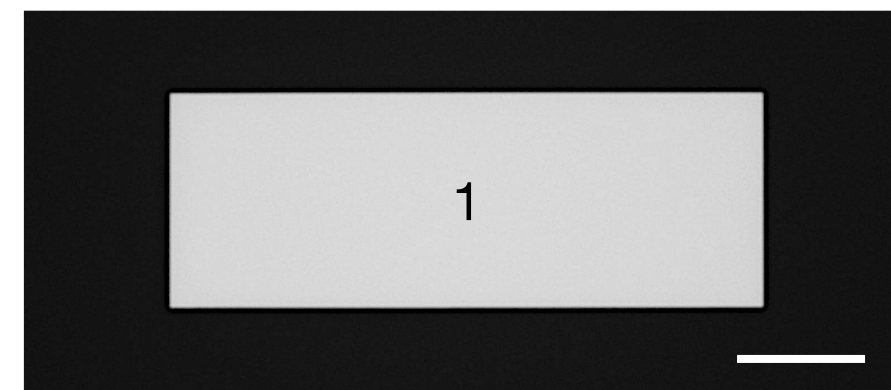
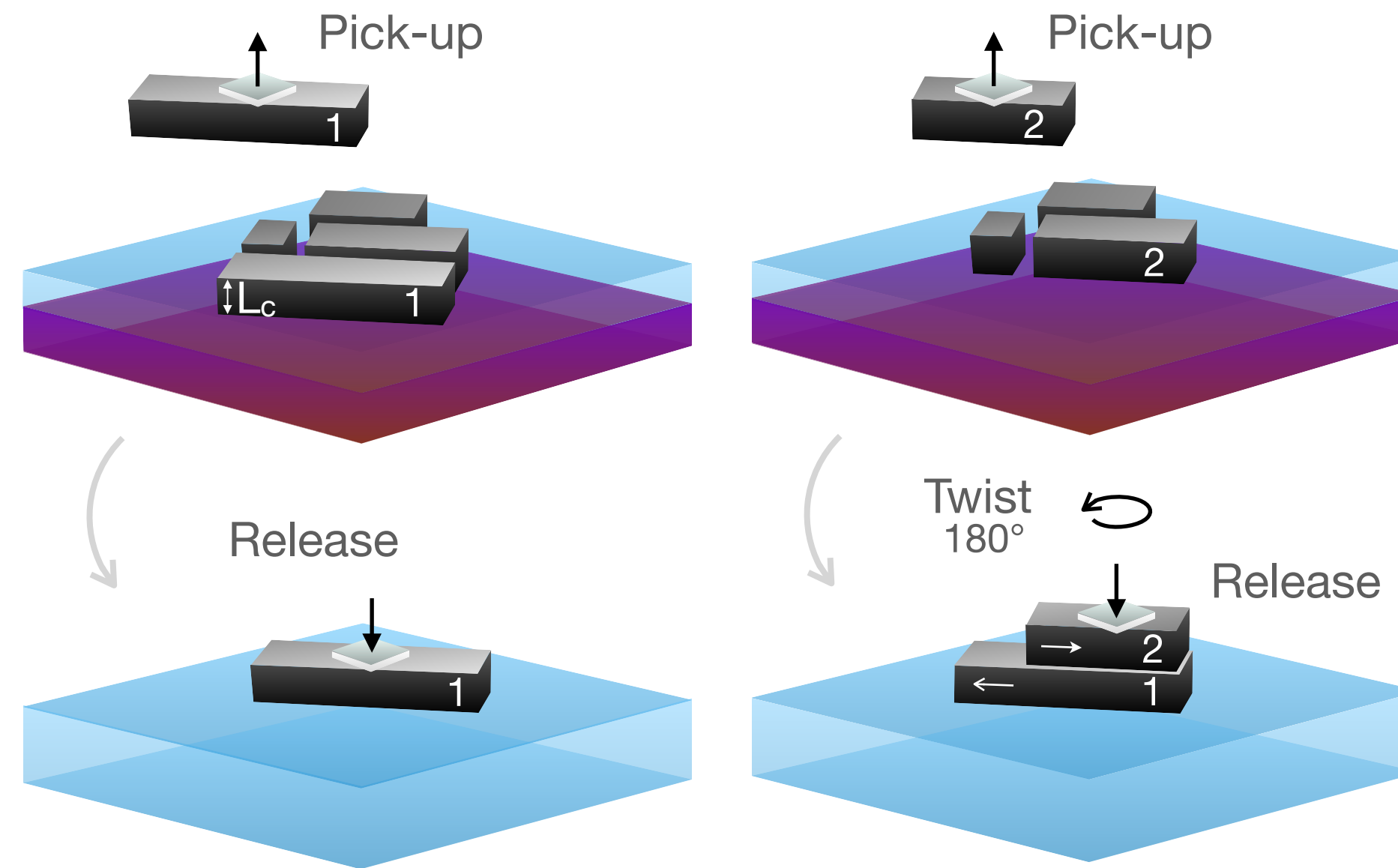
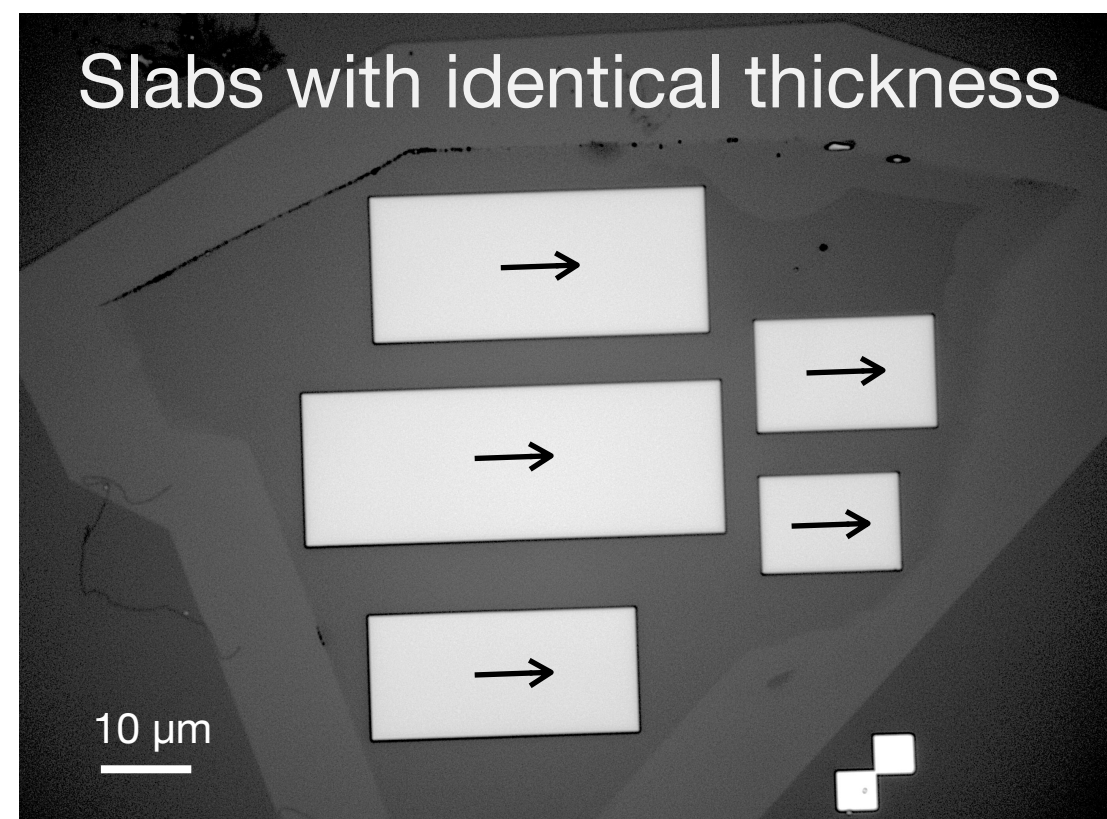
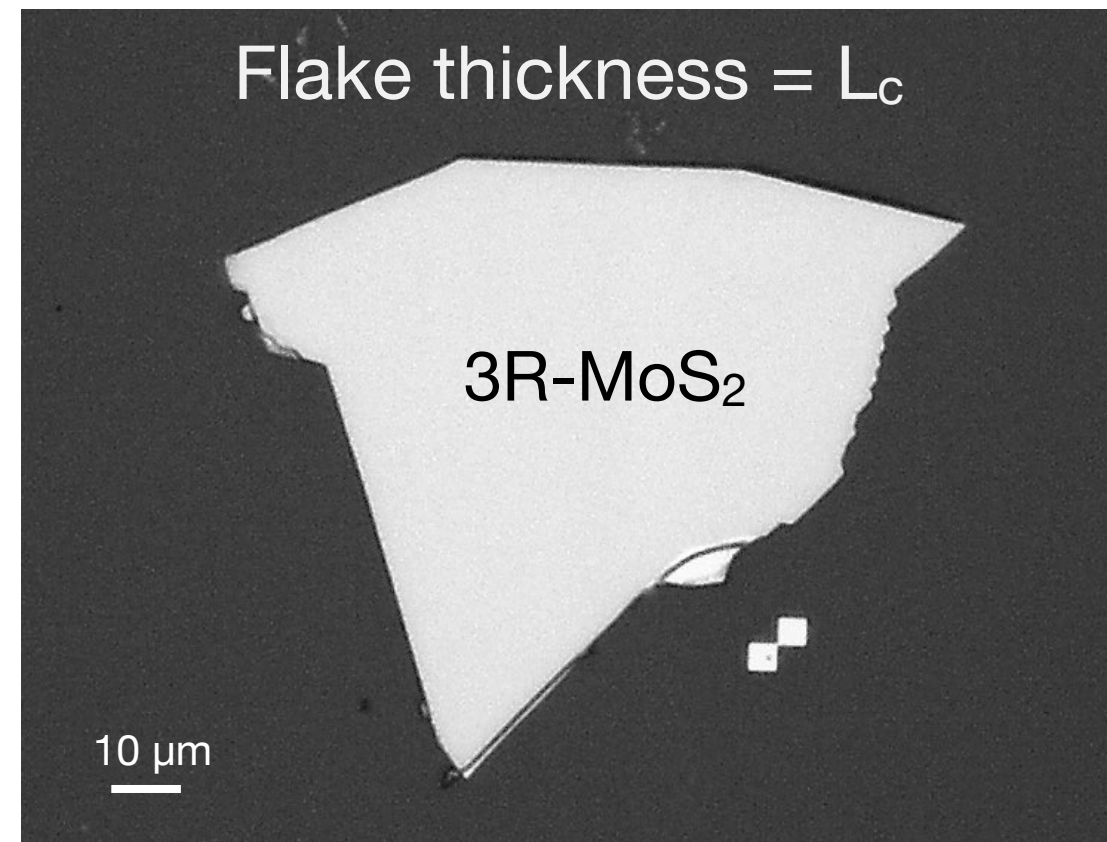
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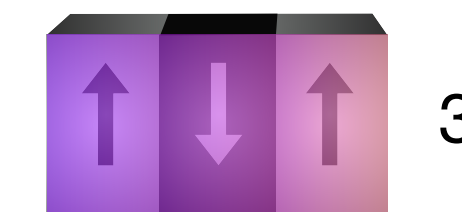
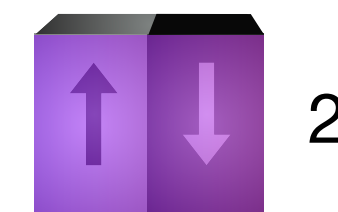
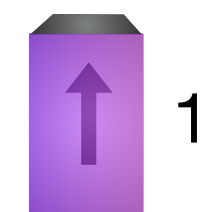
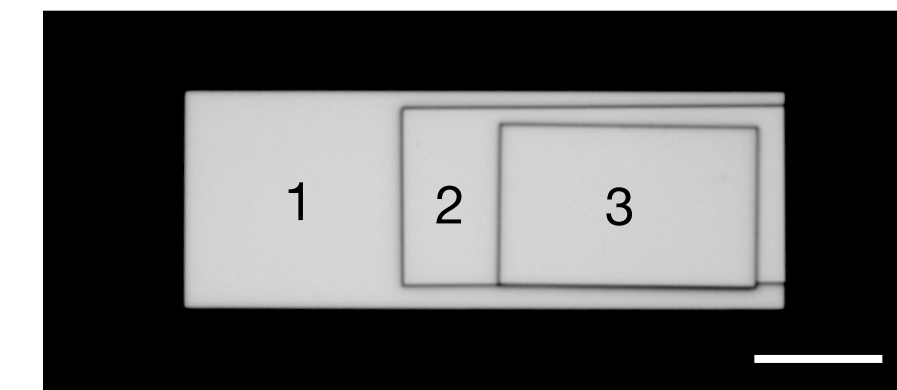
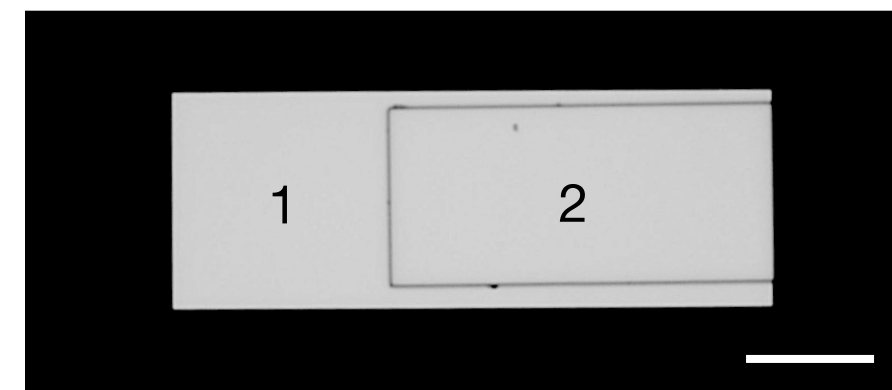
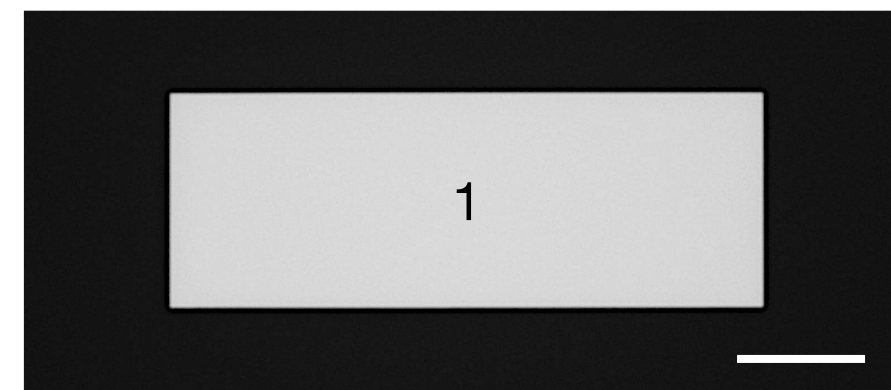
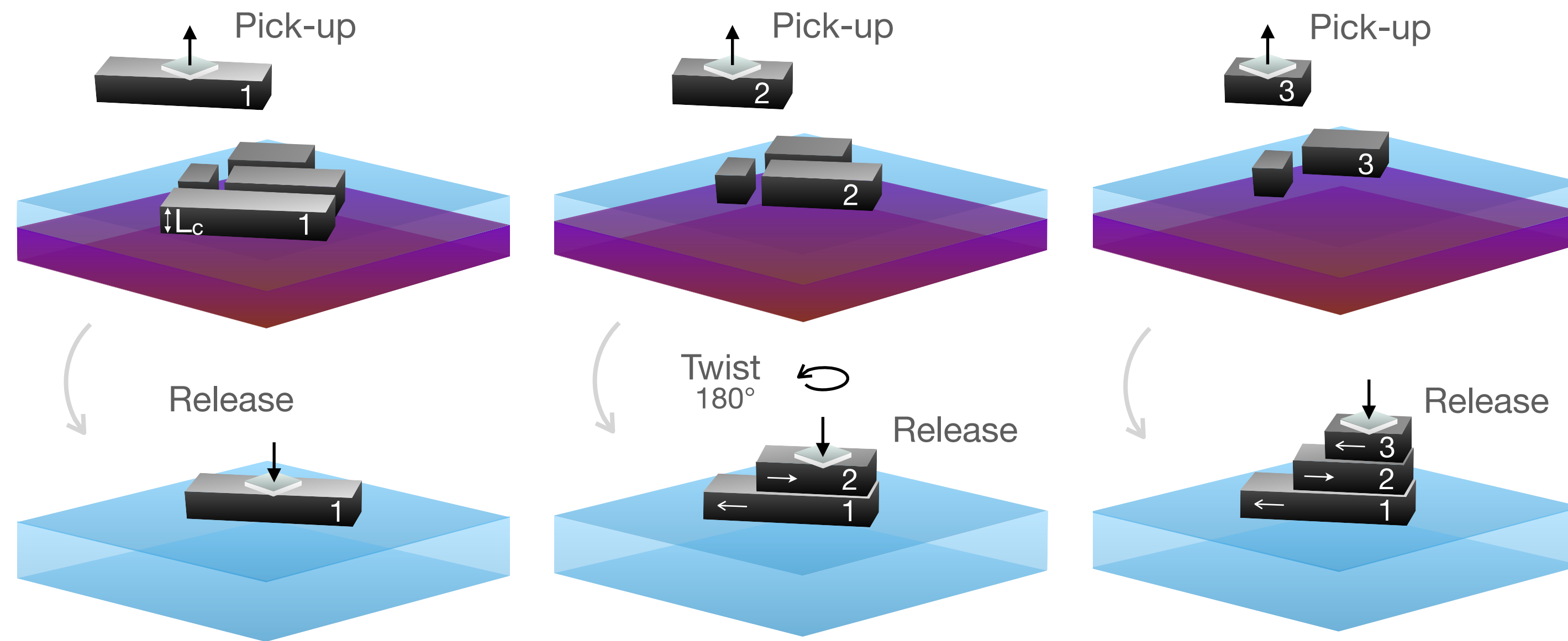
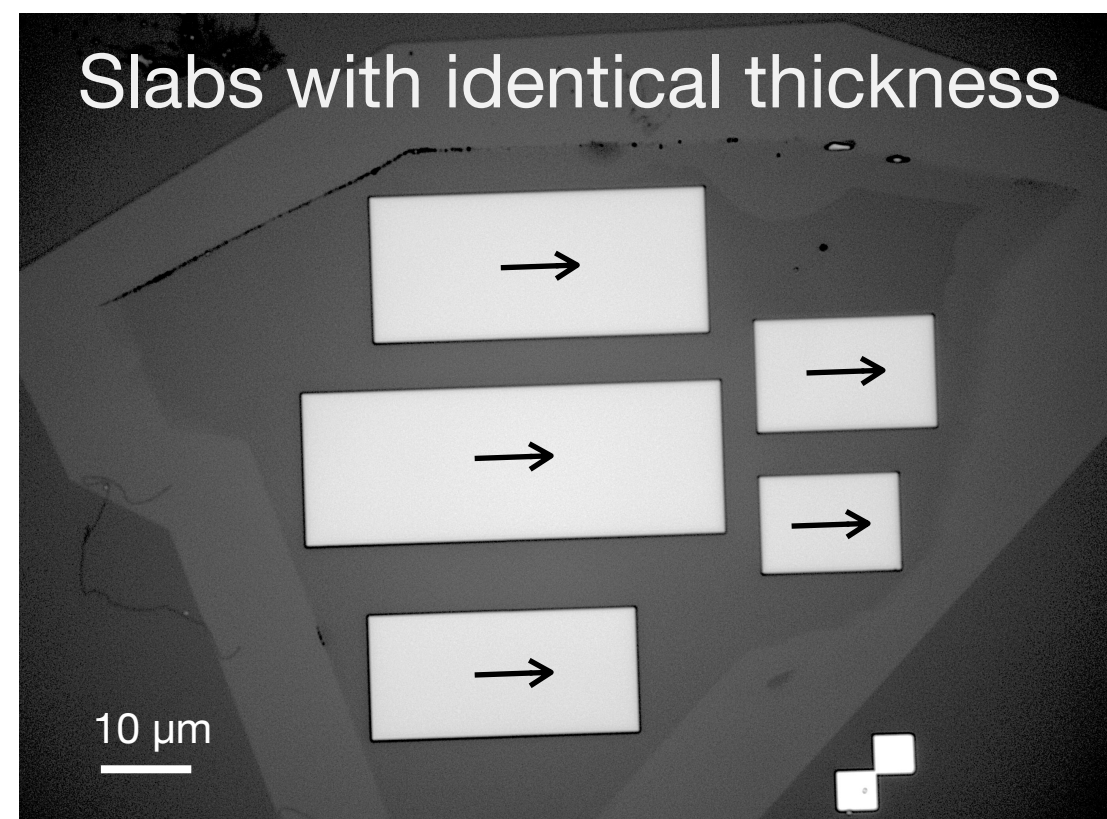
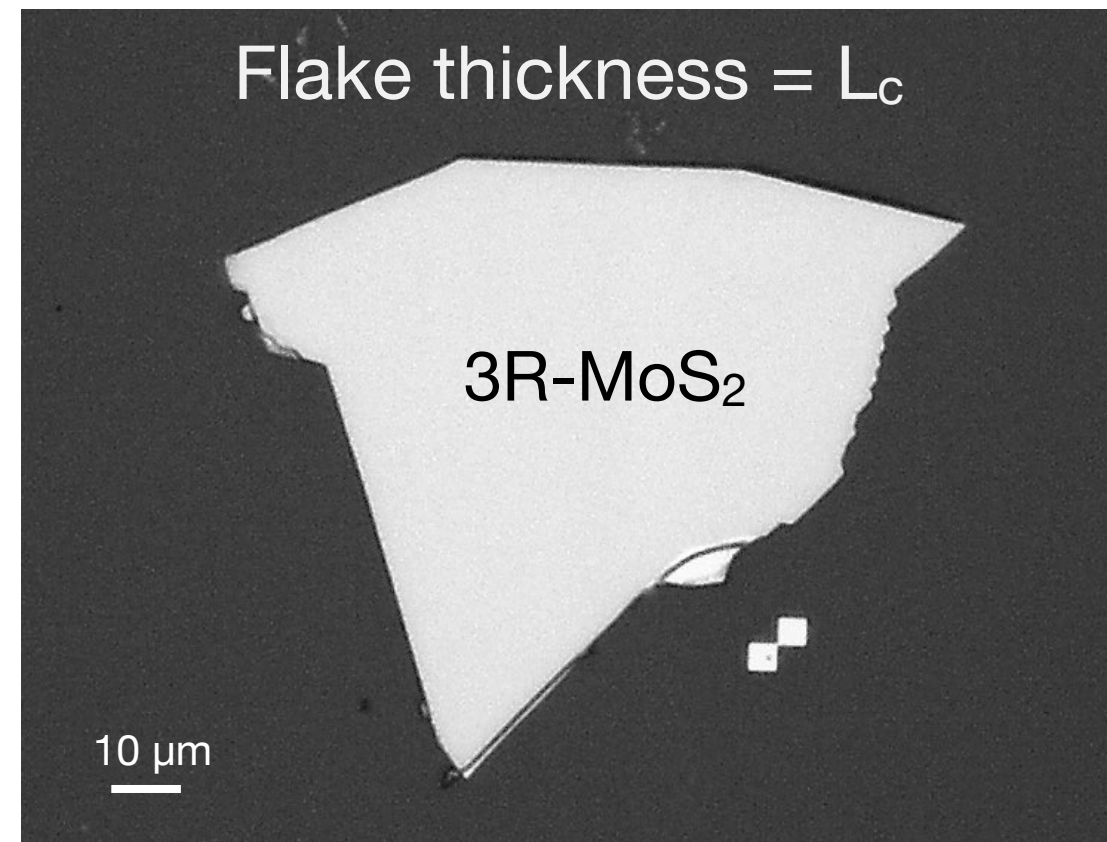
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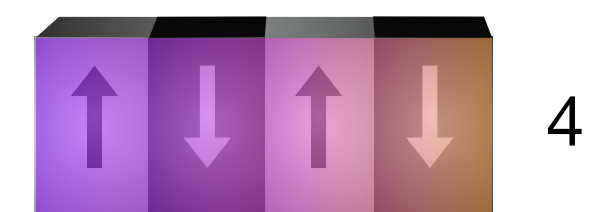
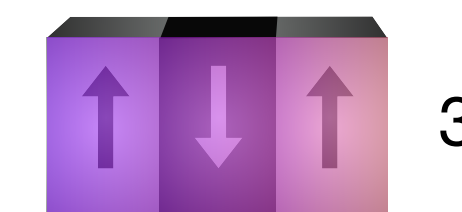
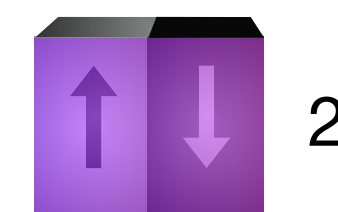
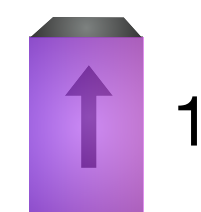
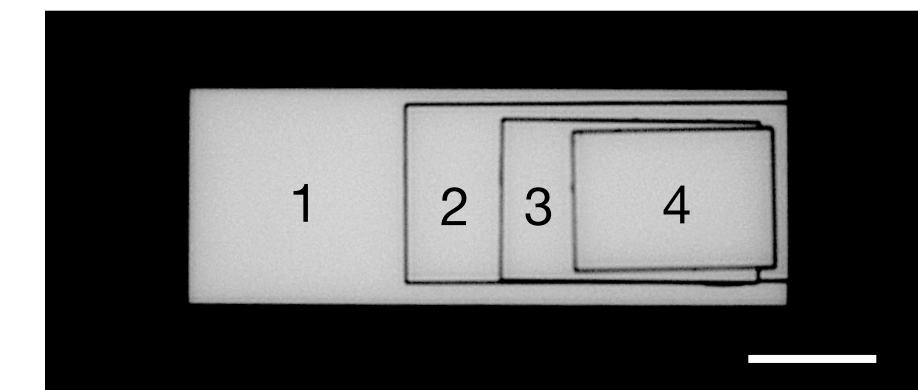
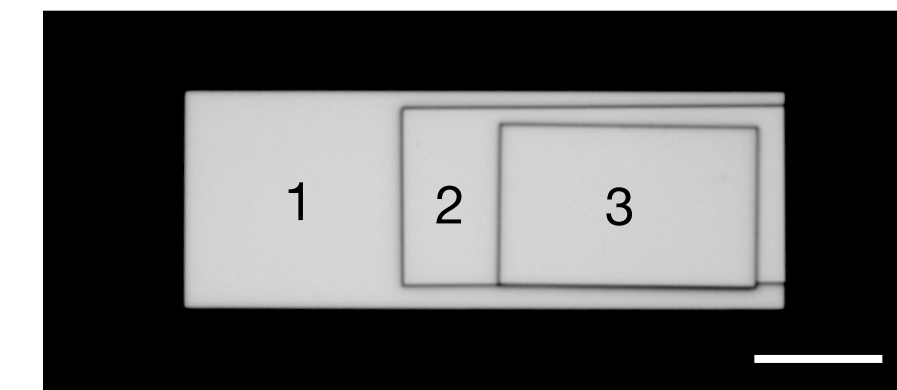
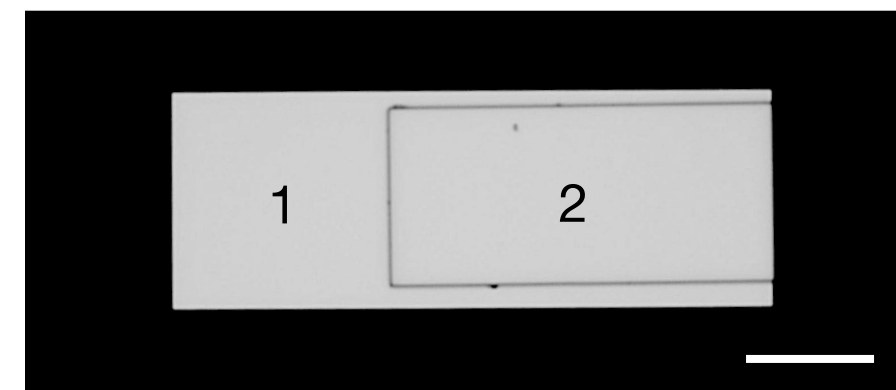
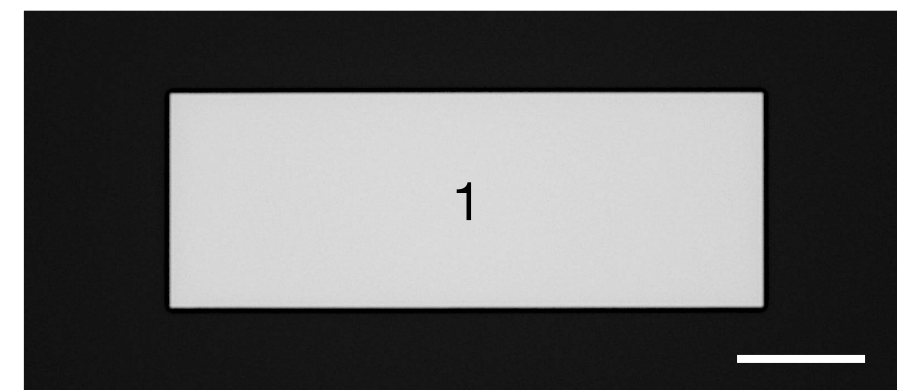
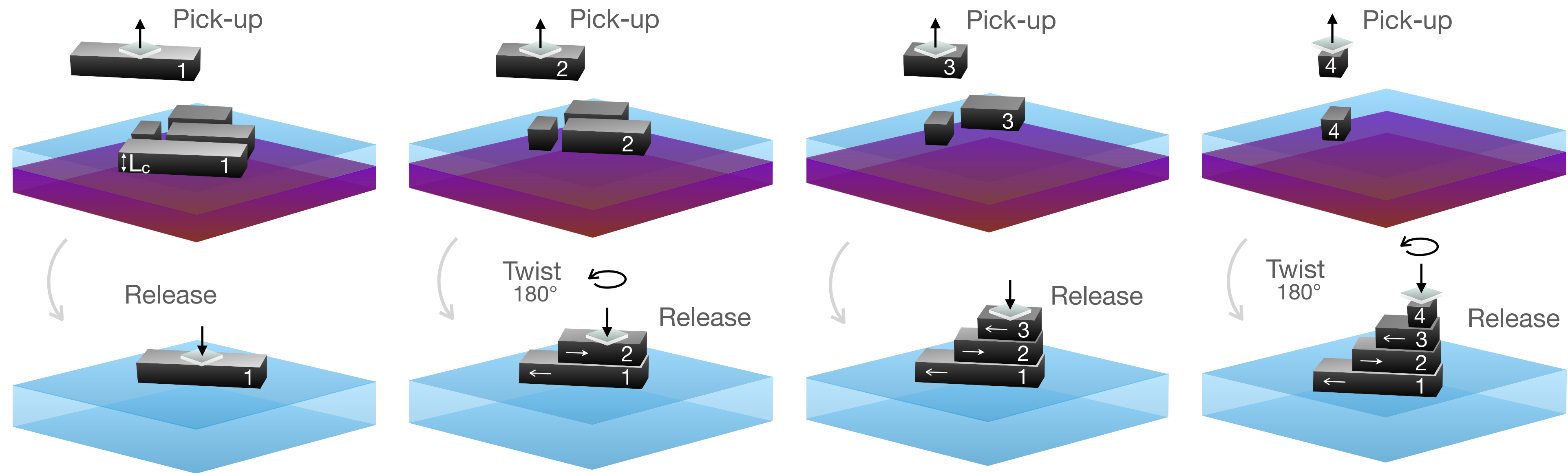
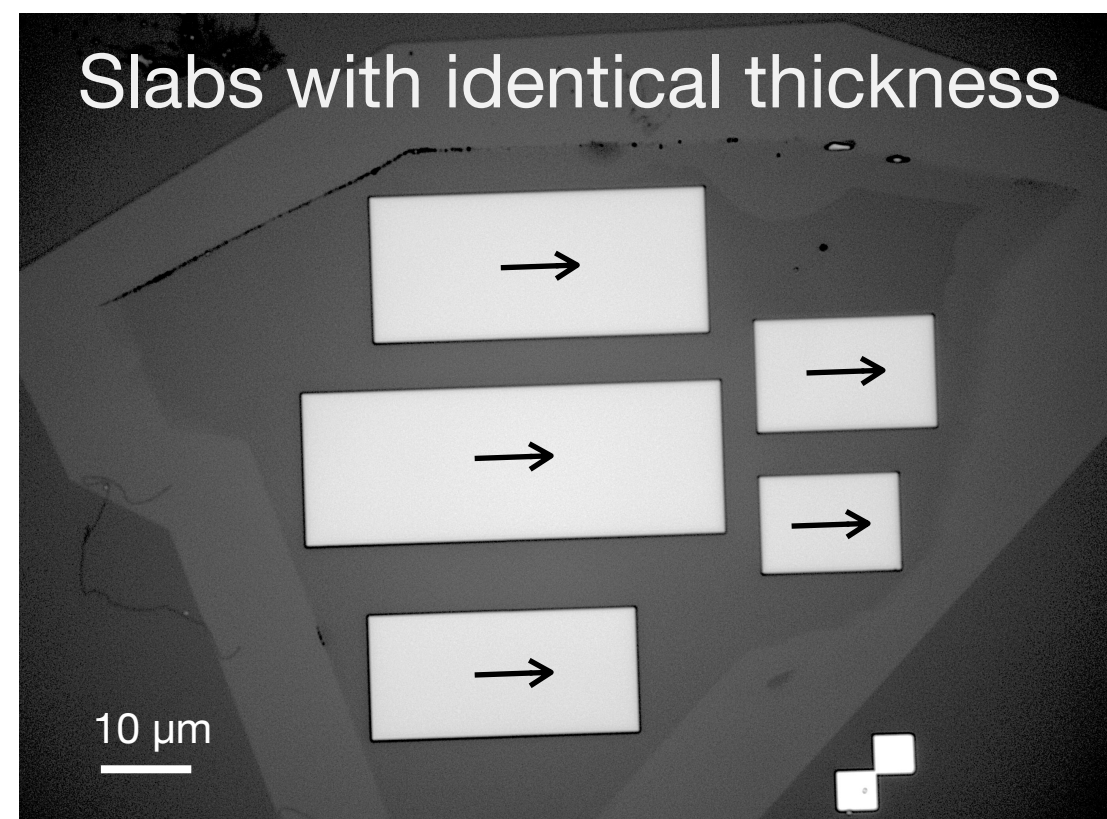
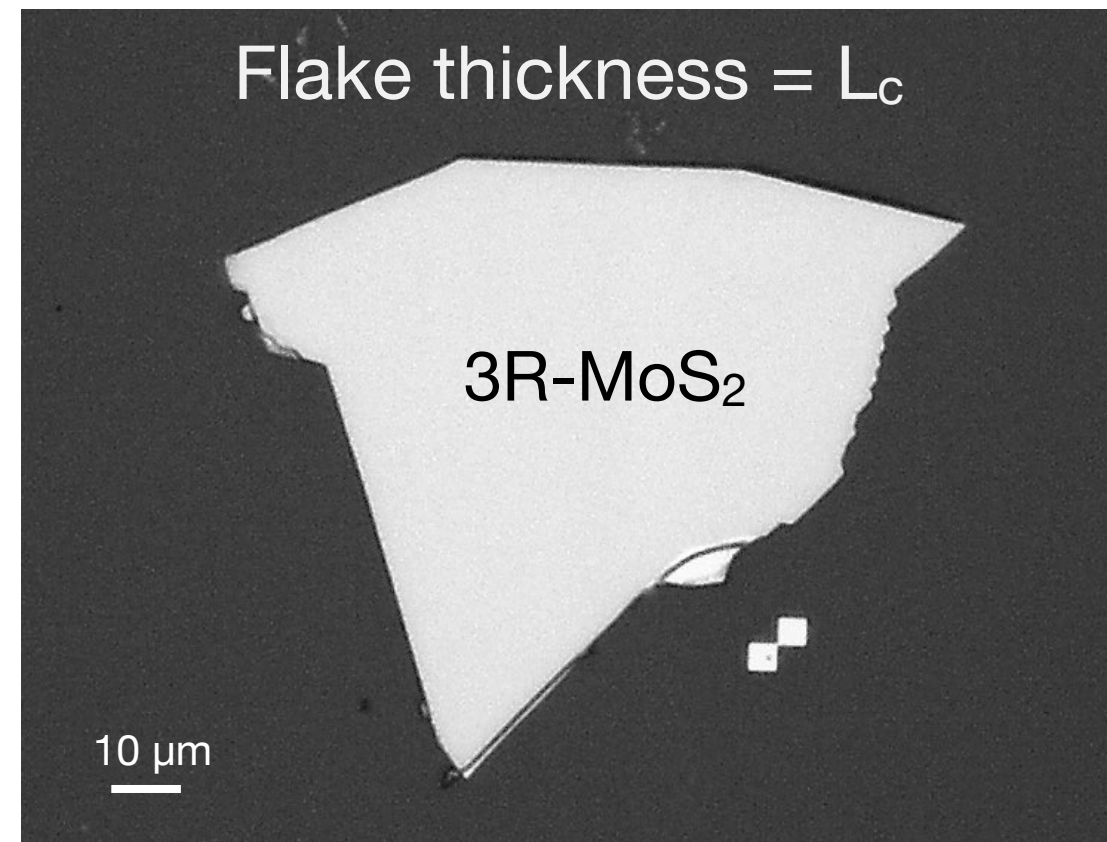
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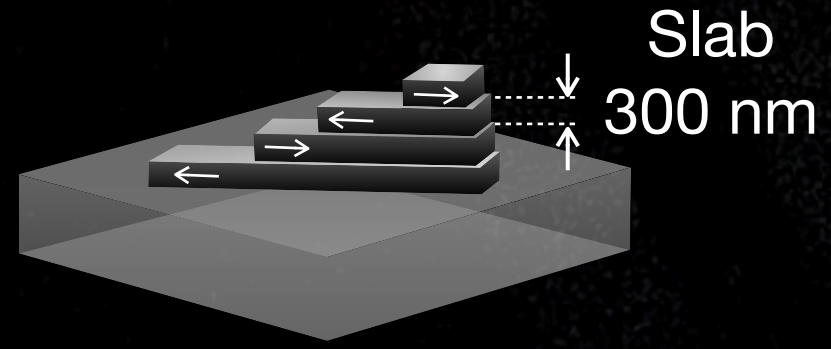
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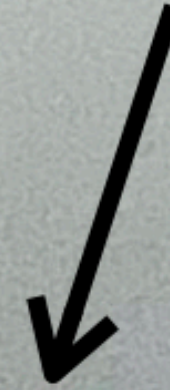


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Macroscopic efficiencies with Microscopic crystals

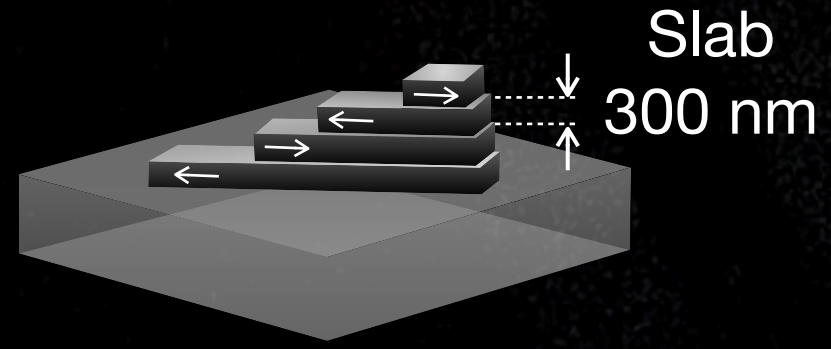


SHG from a 1.2 μm
thick PPTMD

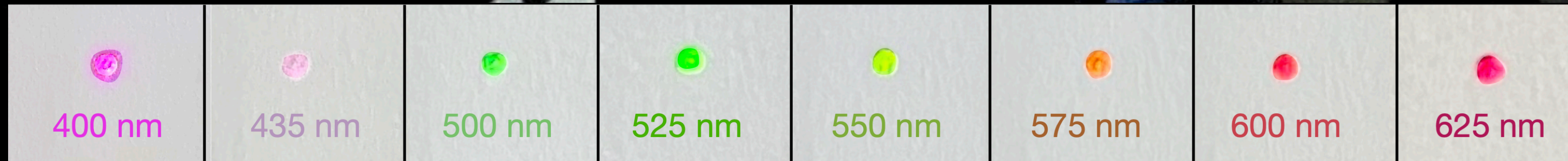


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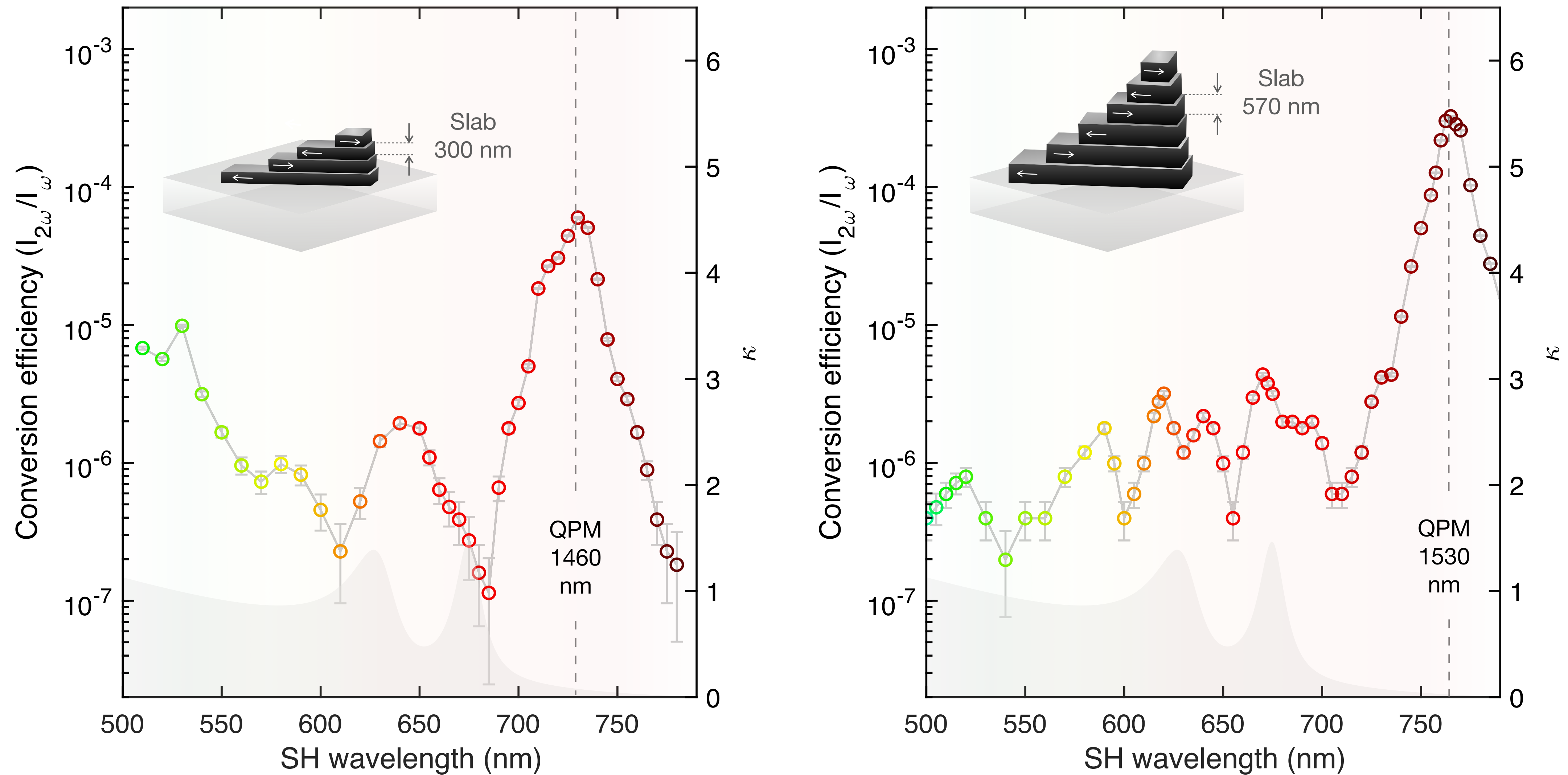
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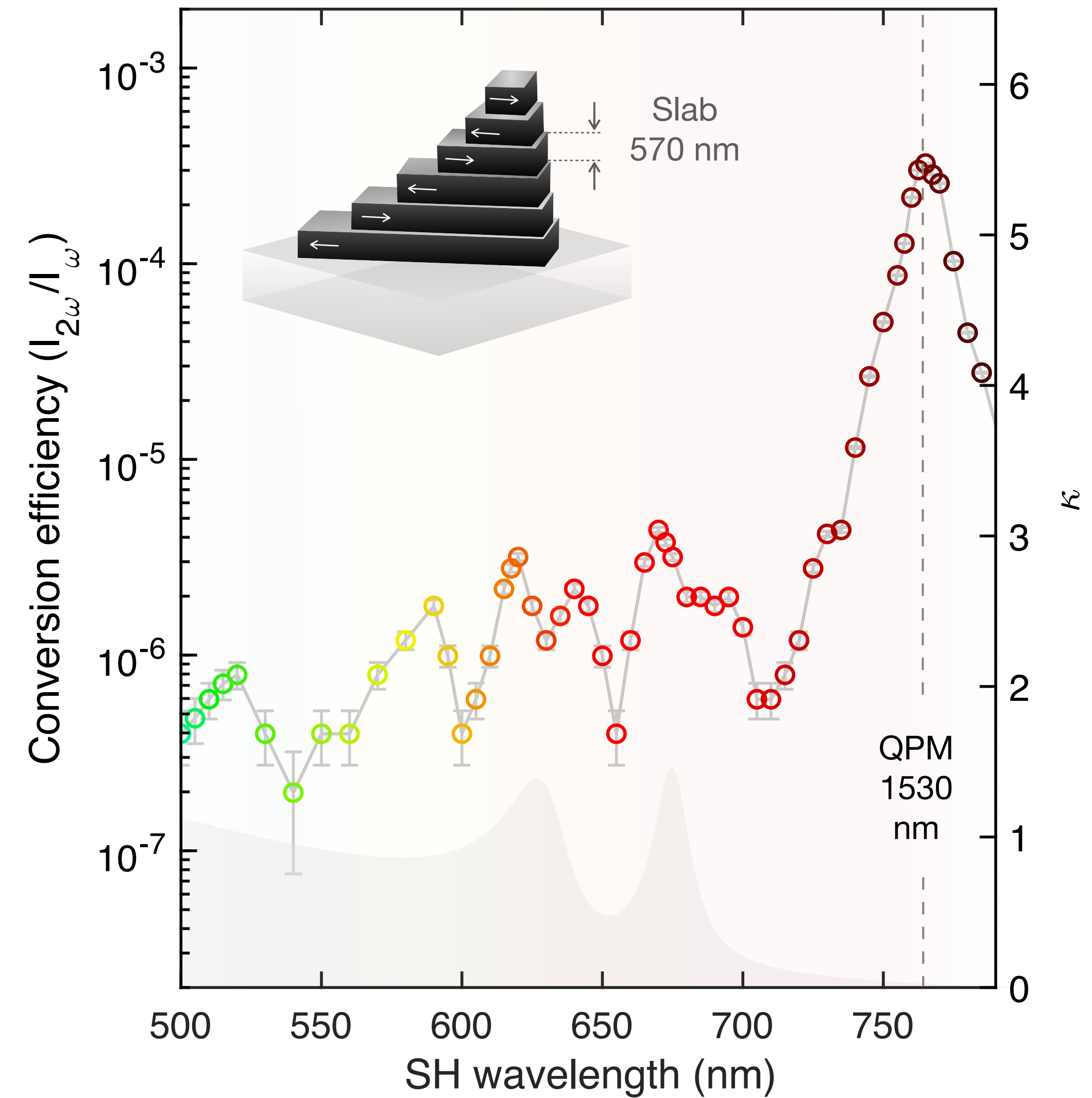
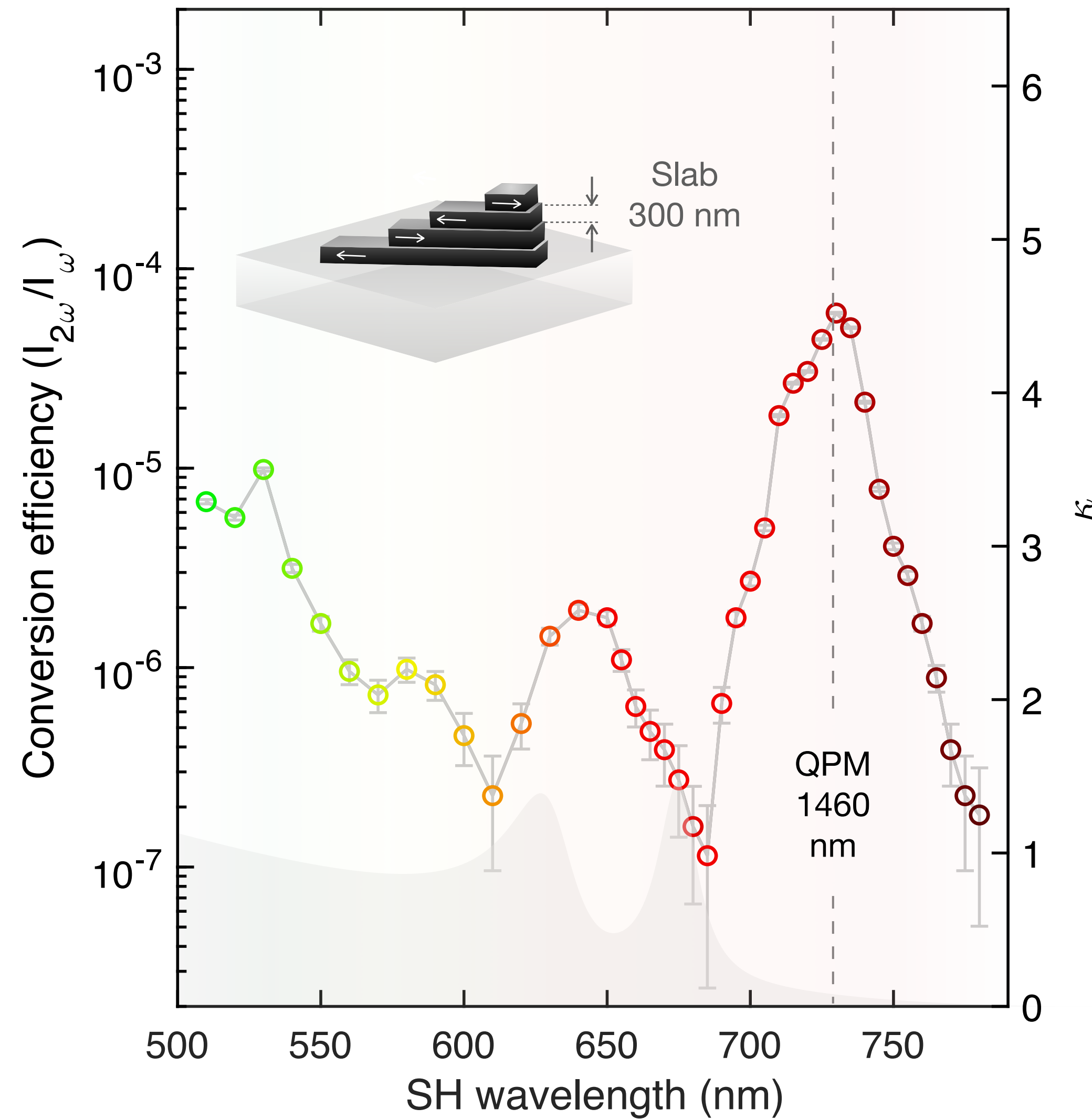
Thickness-tunable quasi-phase-matching bandwidth



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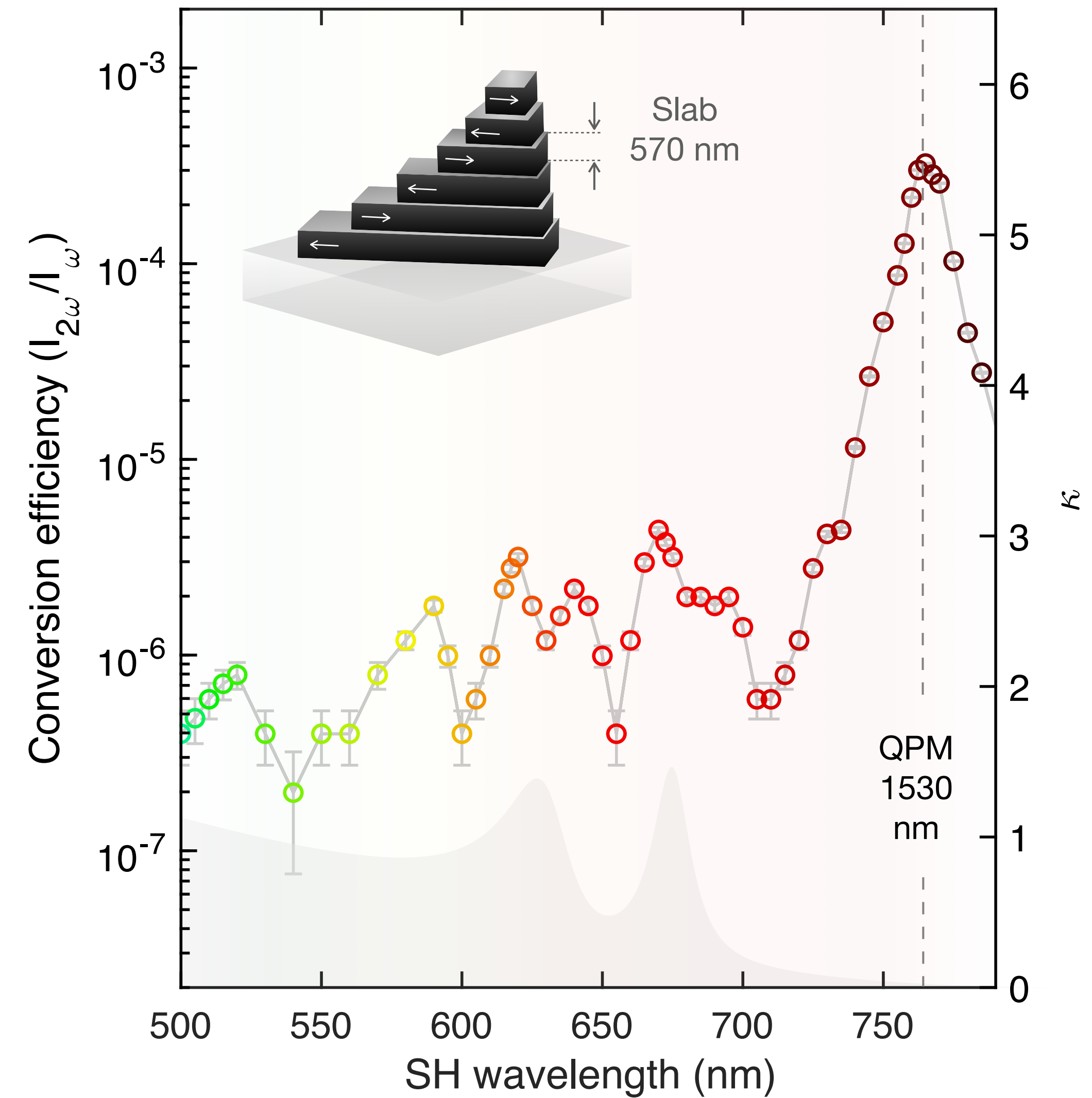
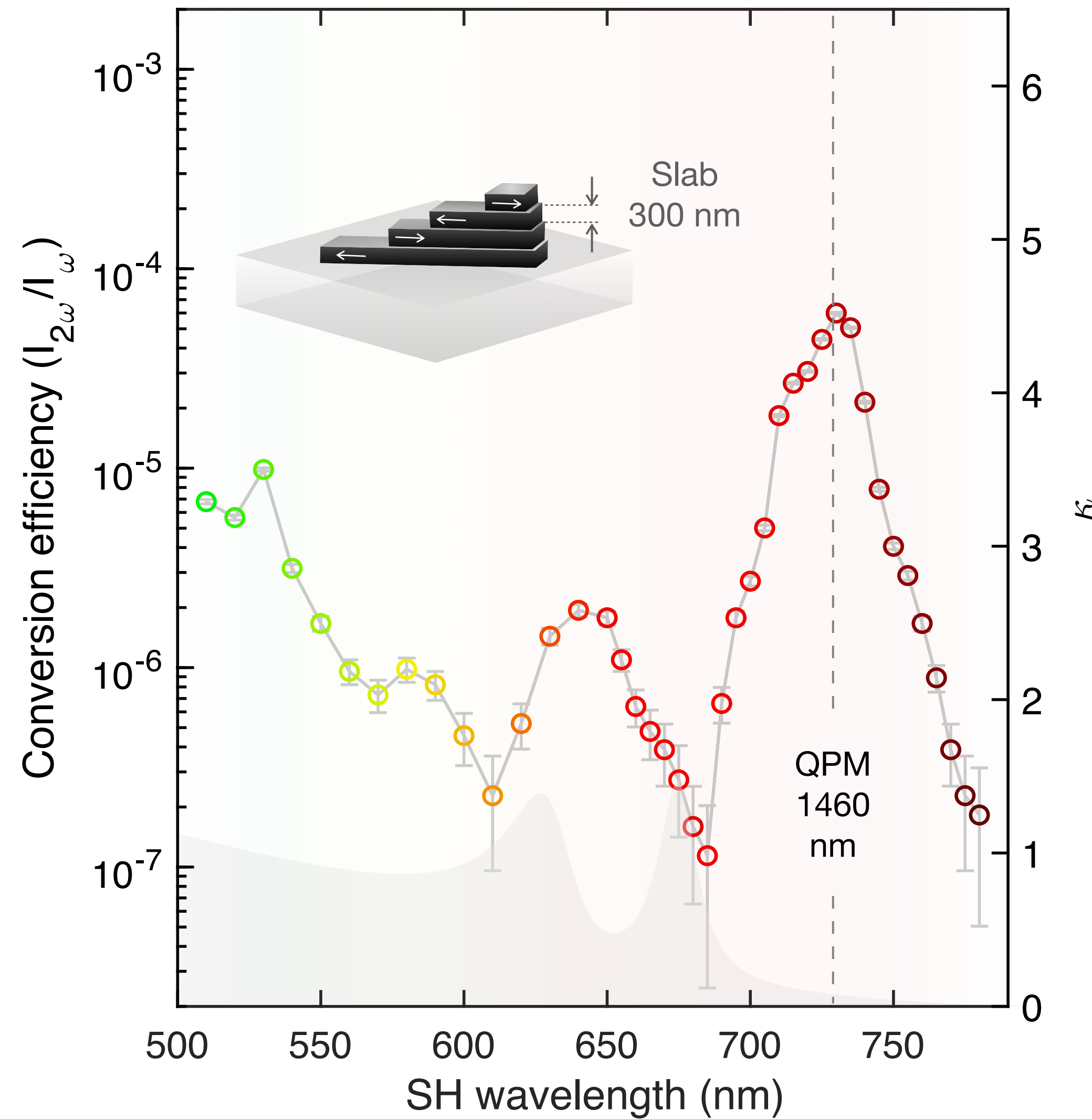
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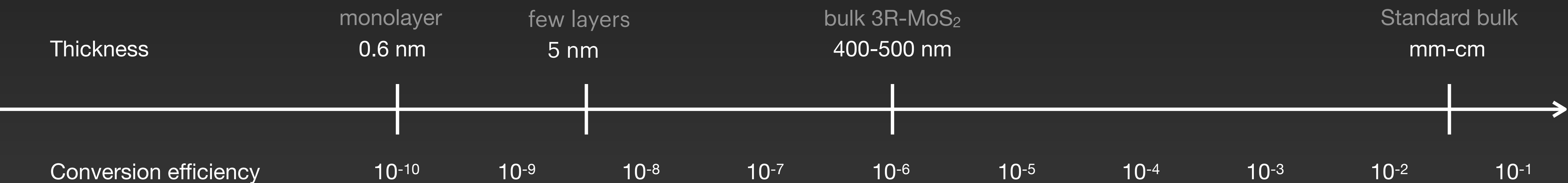
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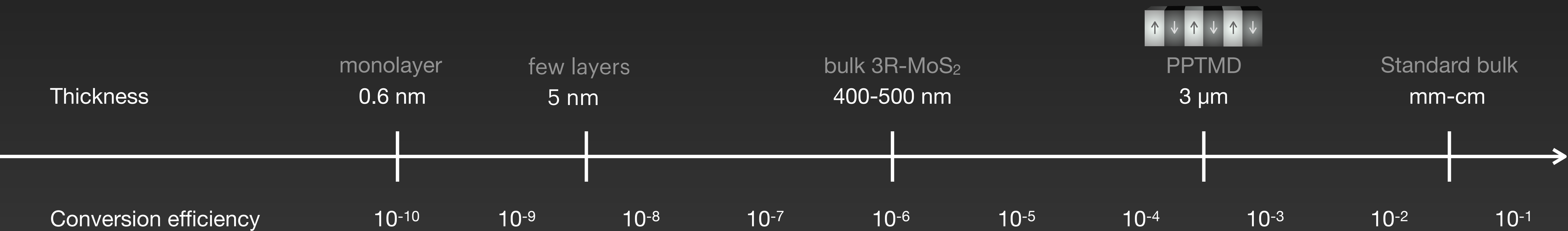
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The “conversion efficiency – thickness” arrow
QUASI PHASE MATCHED LAYERED SEMICONDUCTORS



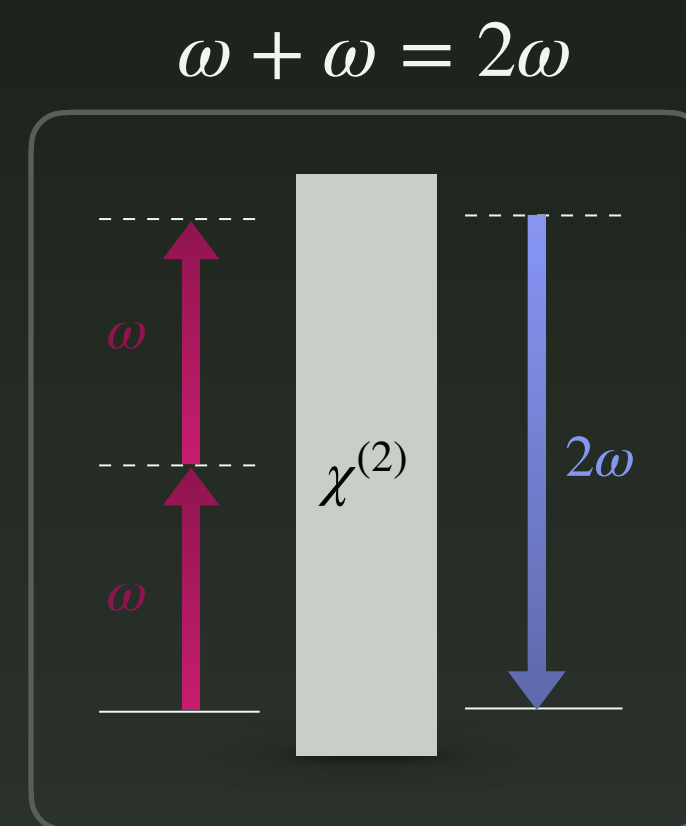
The “conversion efficiency – thickness” arrow

QUASI PHASE MATCHED LAYERED SEMICONDUCTORS



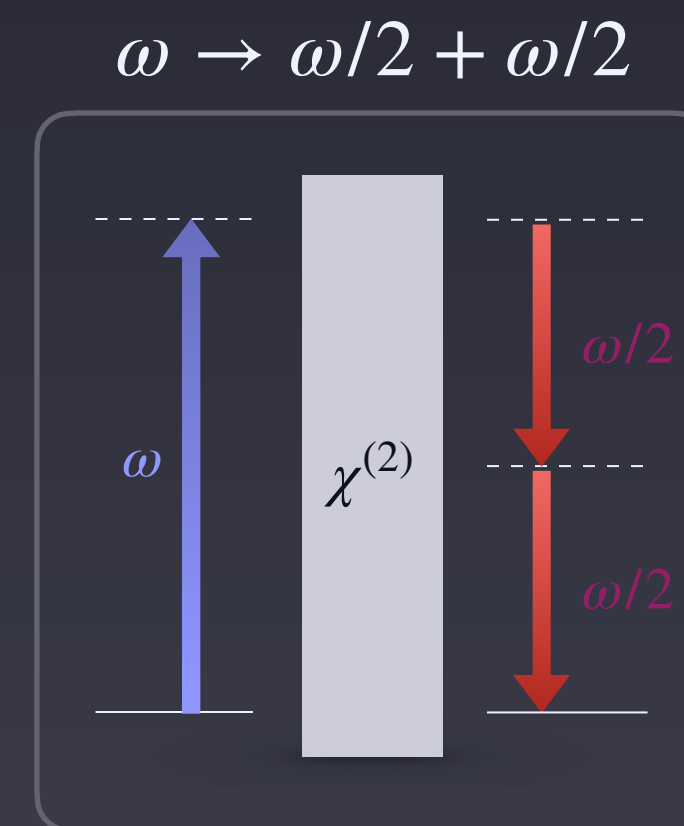
Quasi-phase-matched
Spontaneous Parametric Down-Conversion
from PPTMDs

Classical light



Second Harmonic Generation
(SHG)

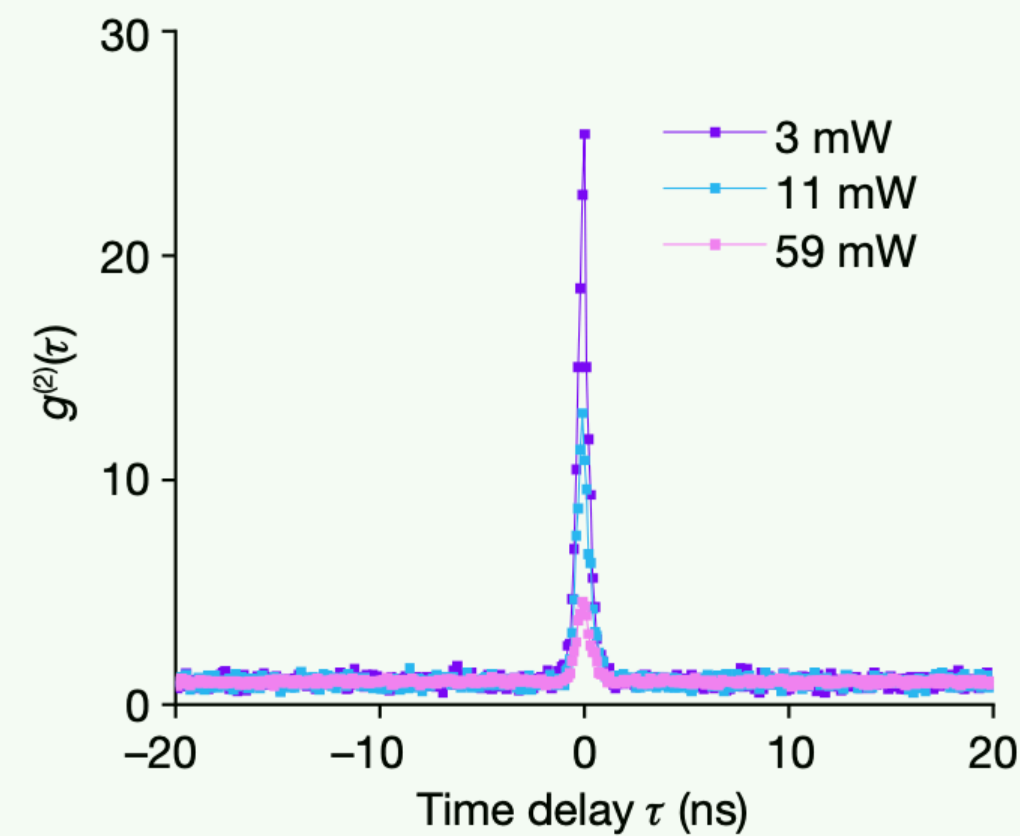
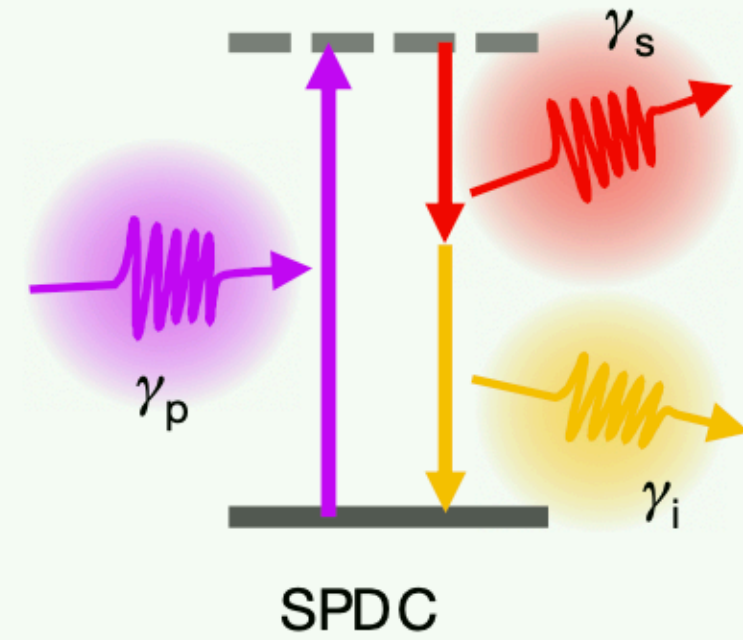
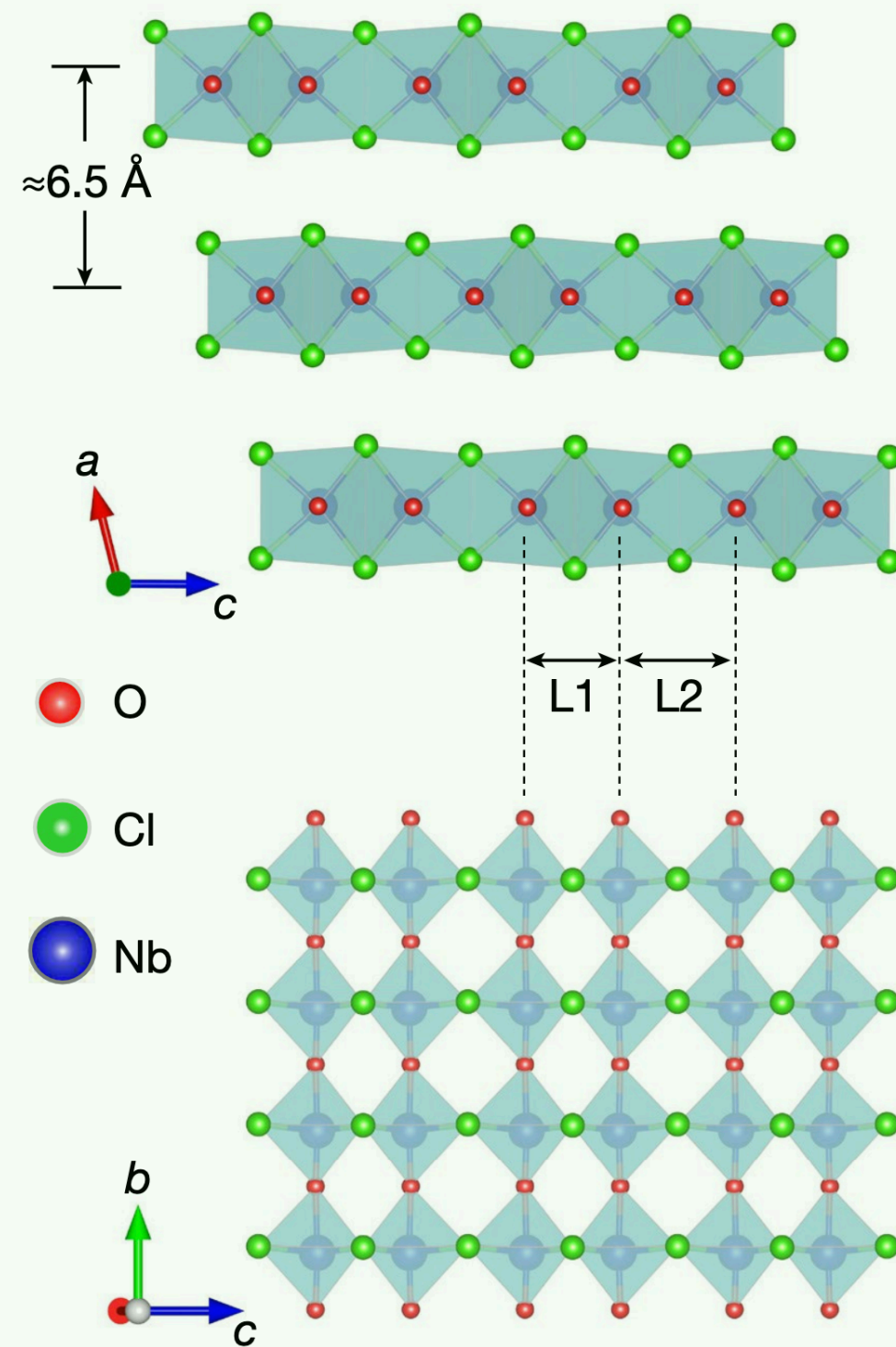
Quantum light



Spontaneous Parametric
Down-conversion (SPDC)

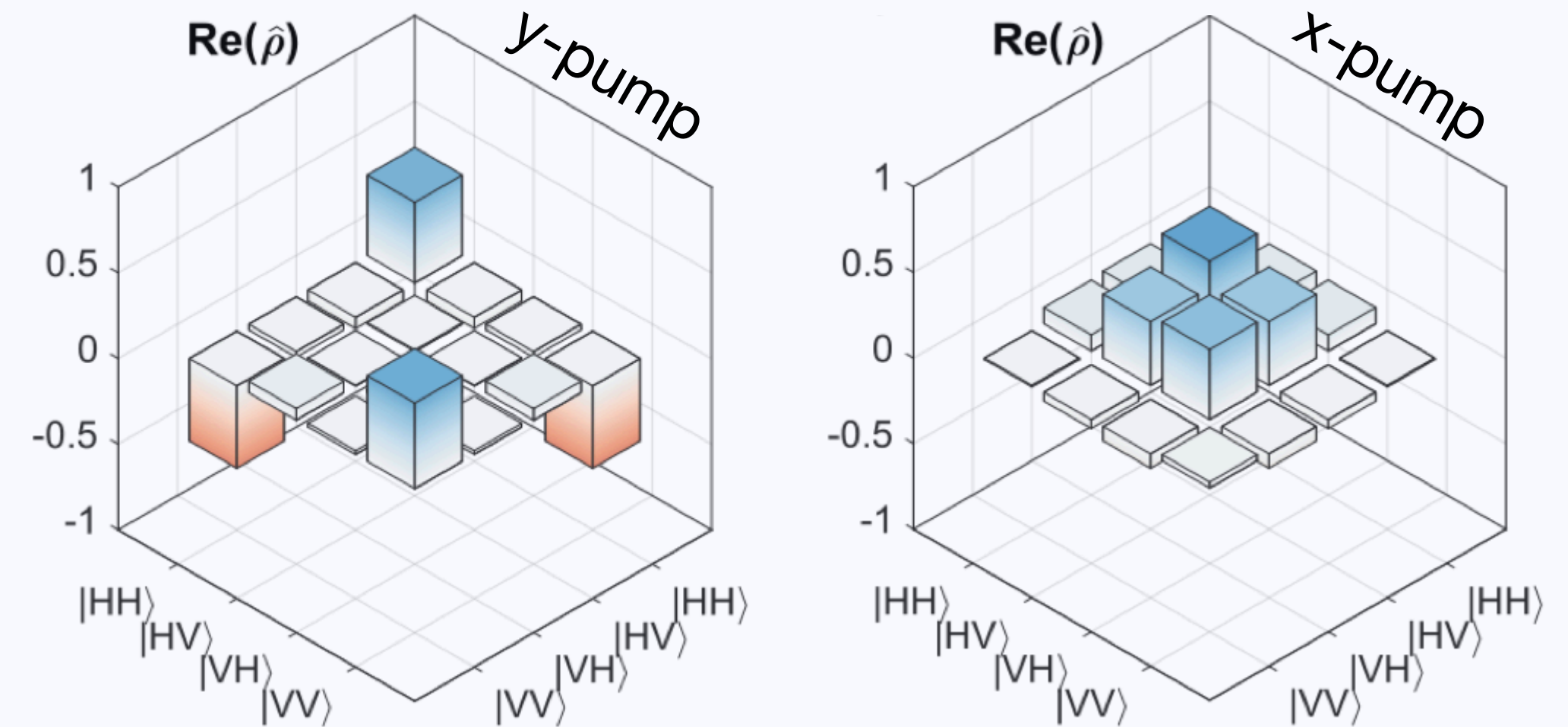
Spontaneous parametric down-conversion (SPDC) in van der Waals crystals

NbOCl₂



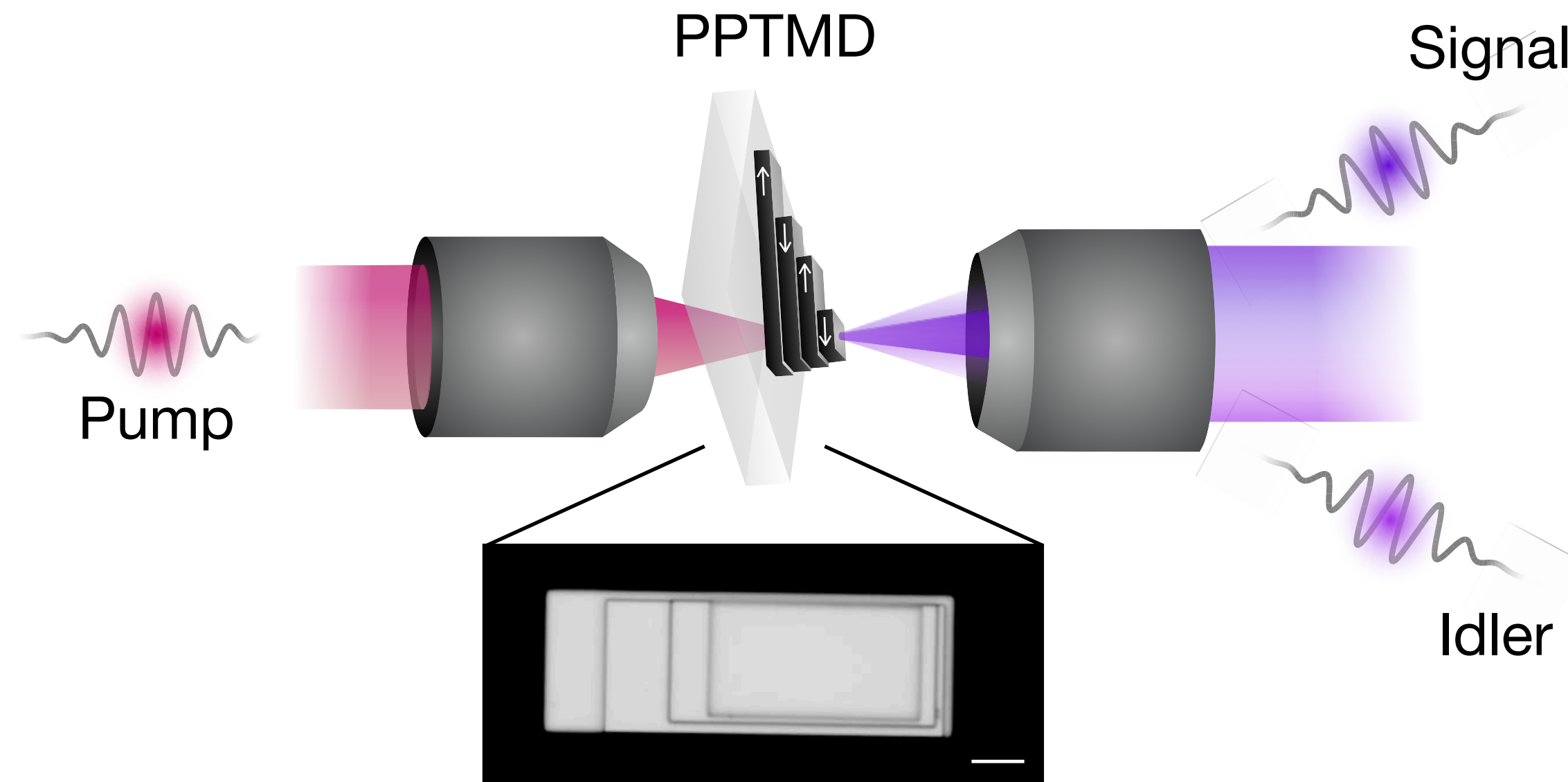
Guo, Q., Qi, XZ., Zhang, L. *et al.* "Ultrathin quantum light source with van der Waals NbOCl₂ crystal" *Nature* **613**, 53–59 (2023)

3R-MoS₂



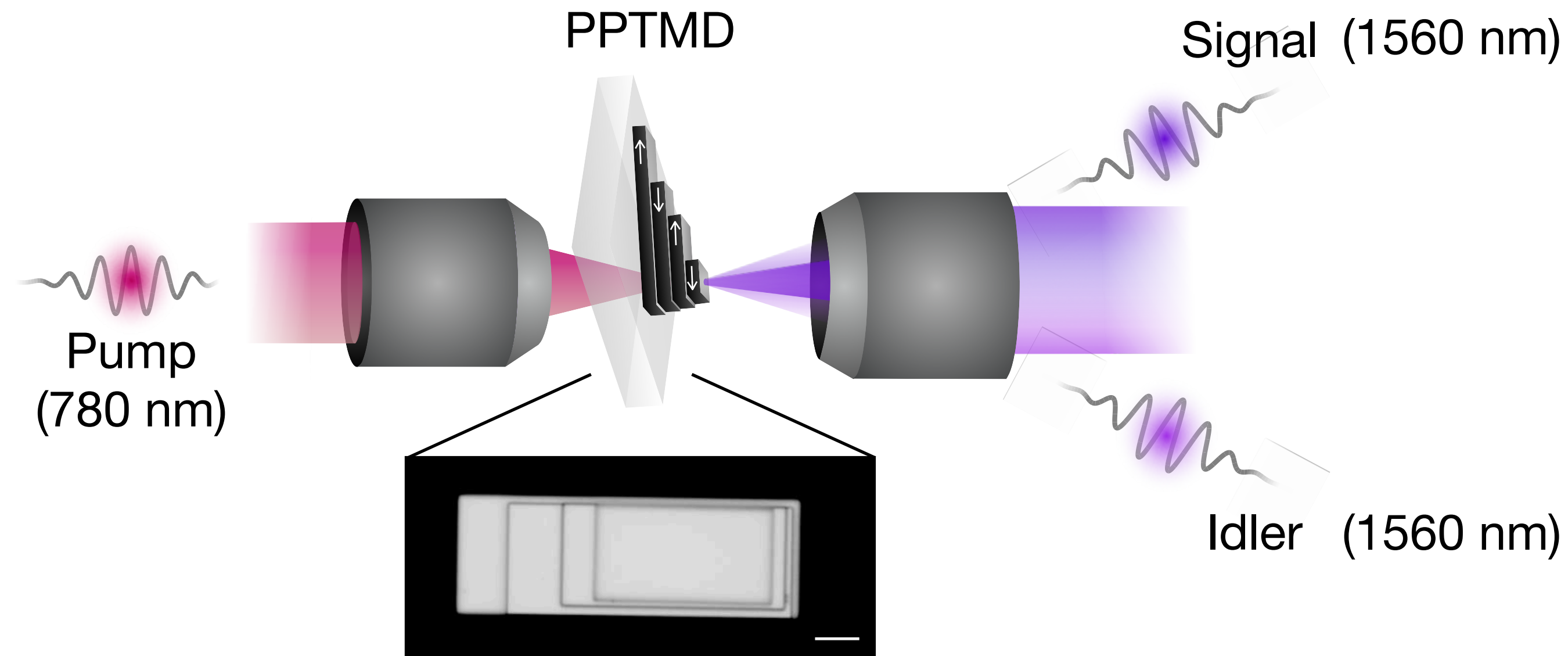
Maximilian A. Weissflog *et al.* "A Tunable Transition Metal Dichalcogenide Entangled Photon-Pair Source" [arXiv:2311.16036](https://arxiv.org/abs/2311.16036) (2023)

Quasi-phase-matched SPDC from PPTMDs



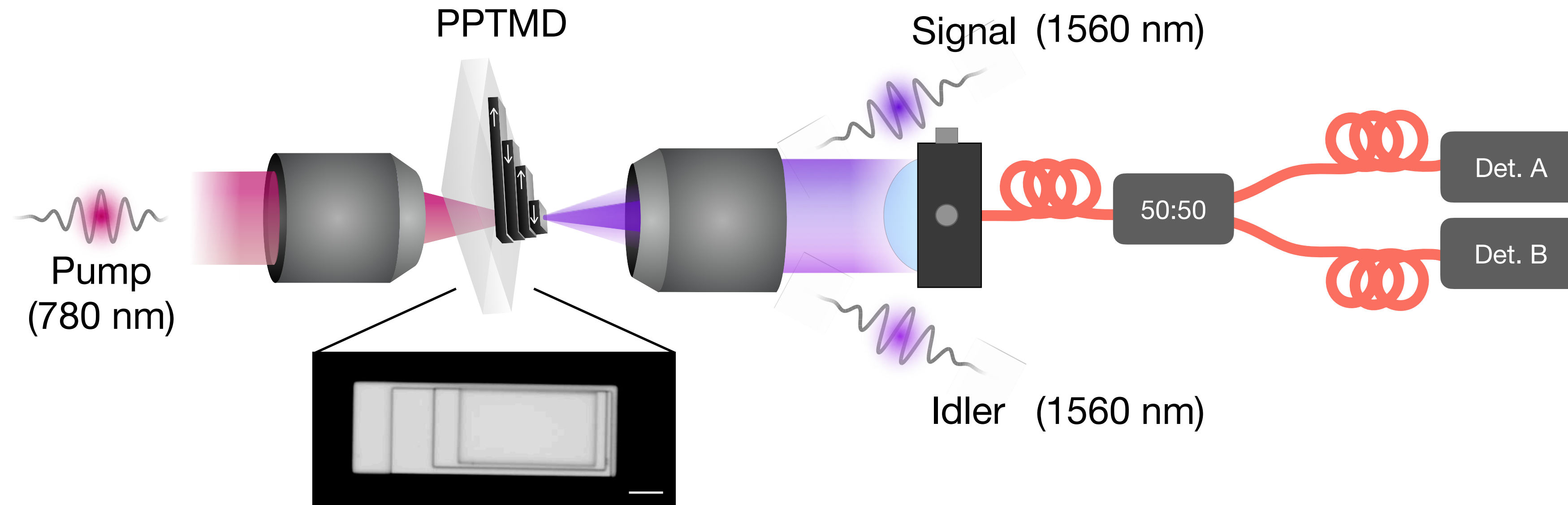
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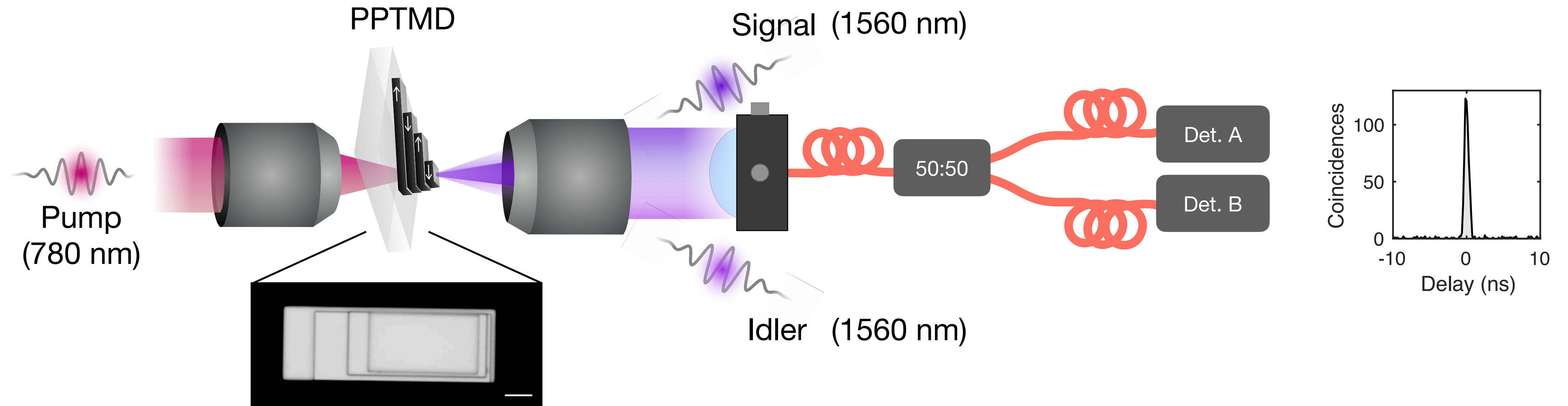
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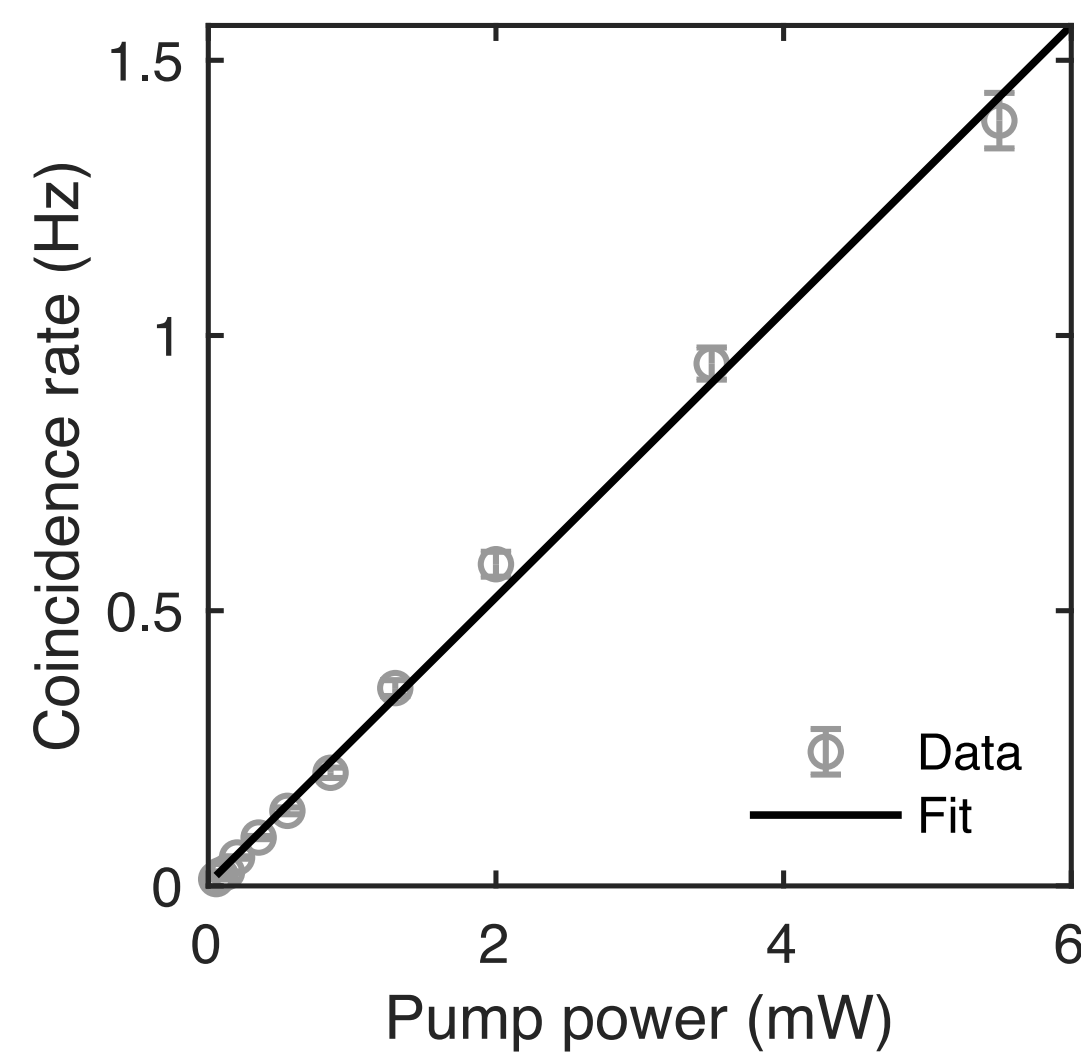
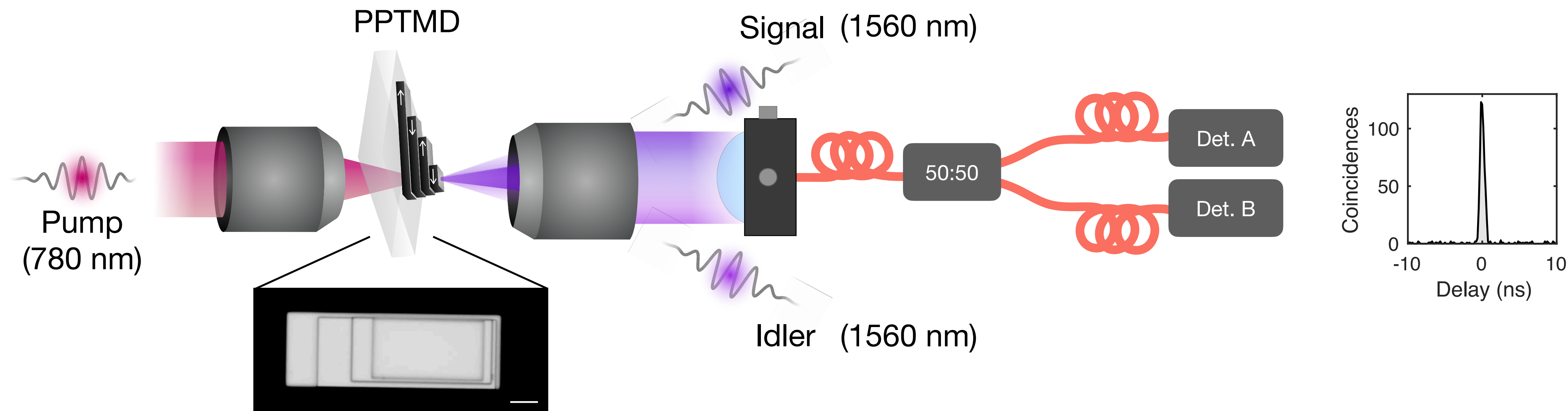
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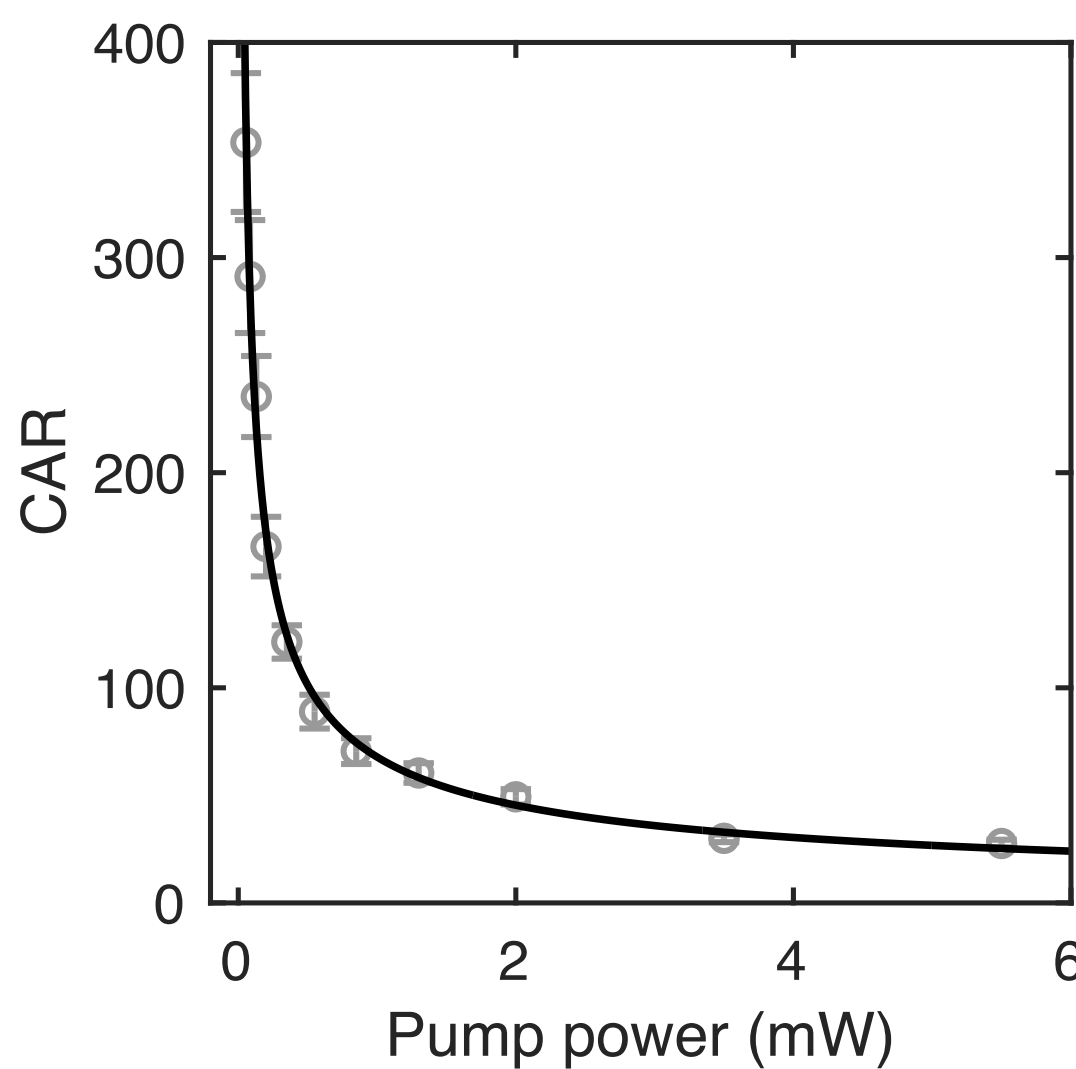
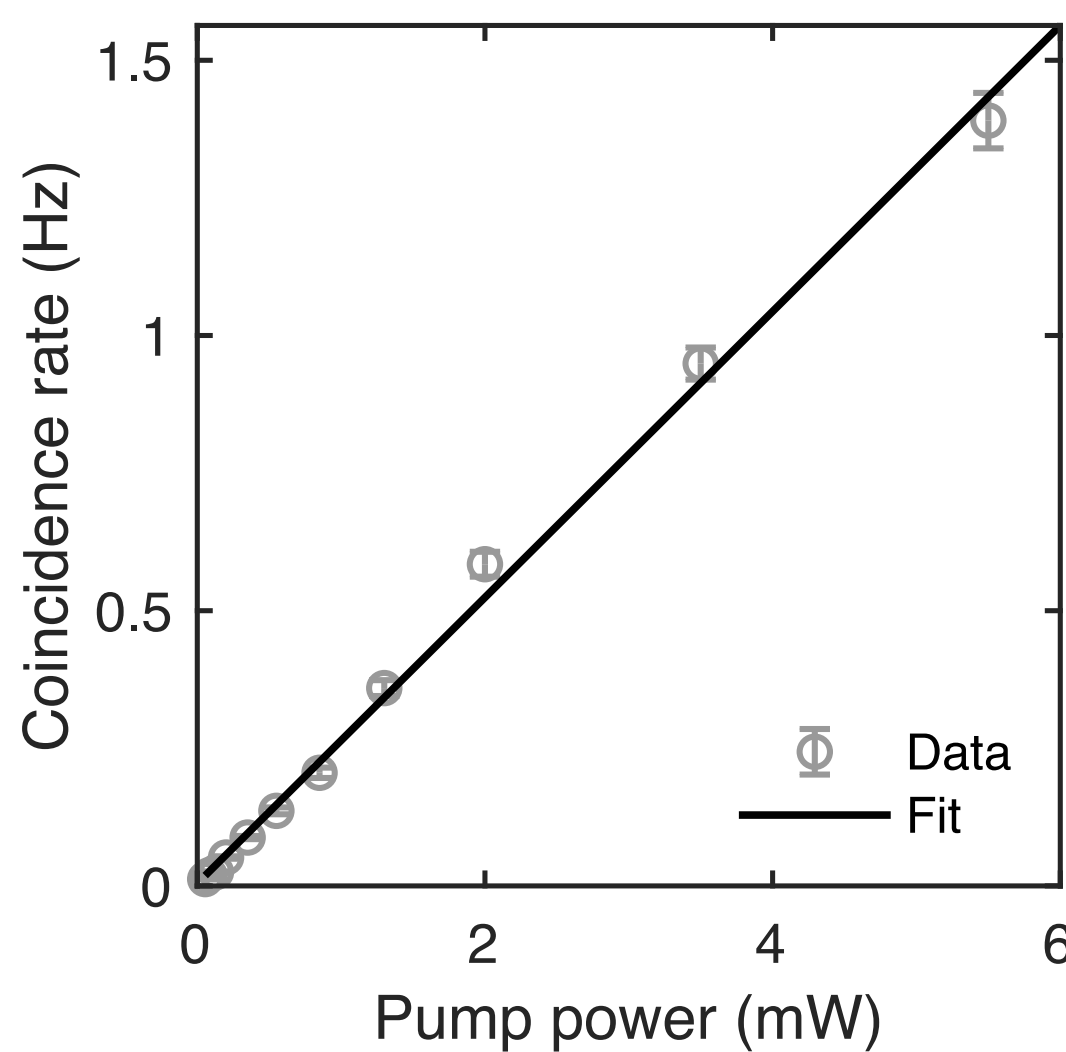
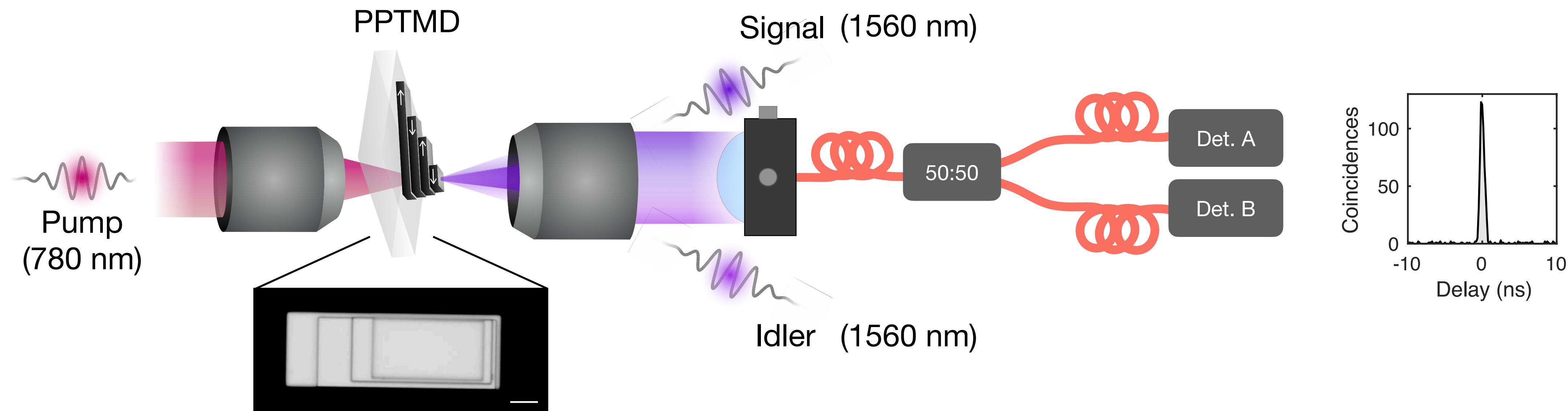
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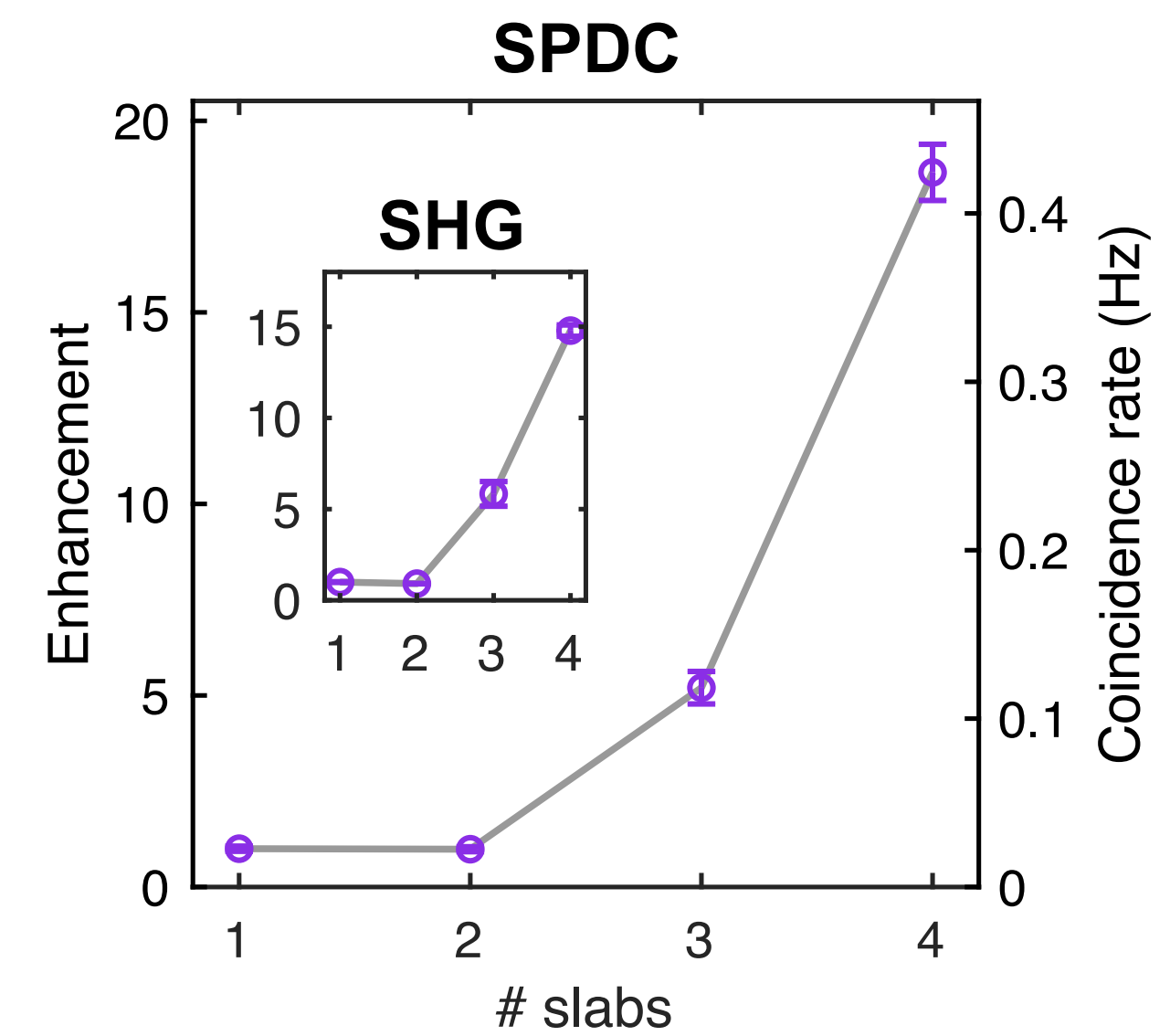
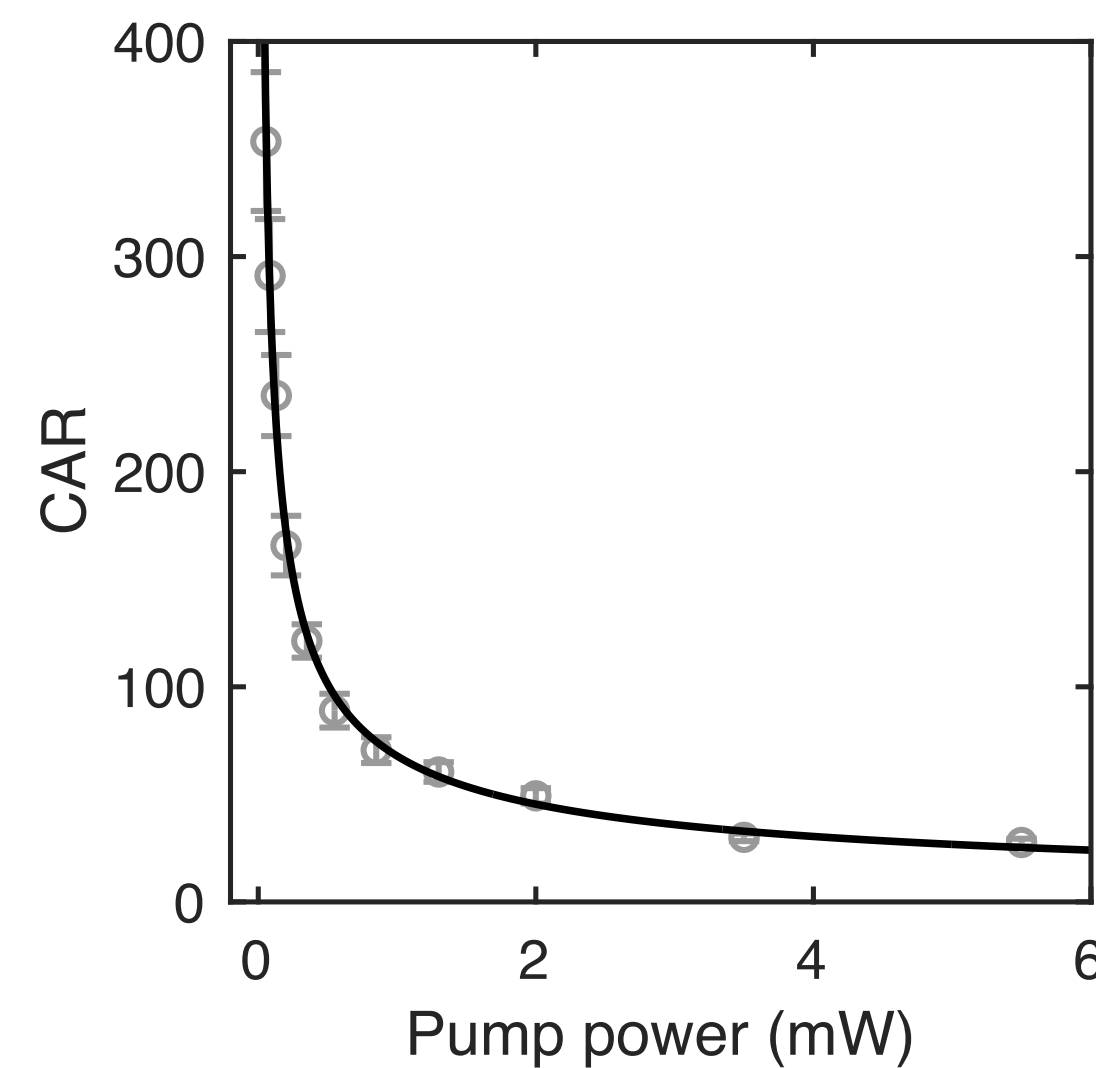
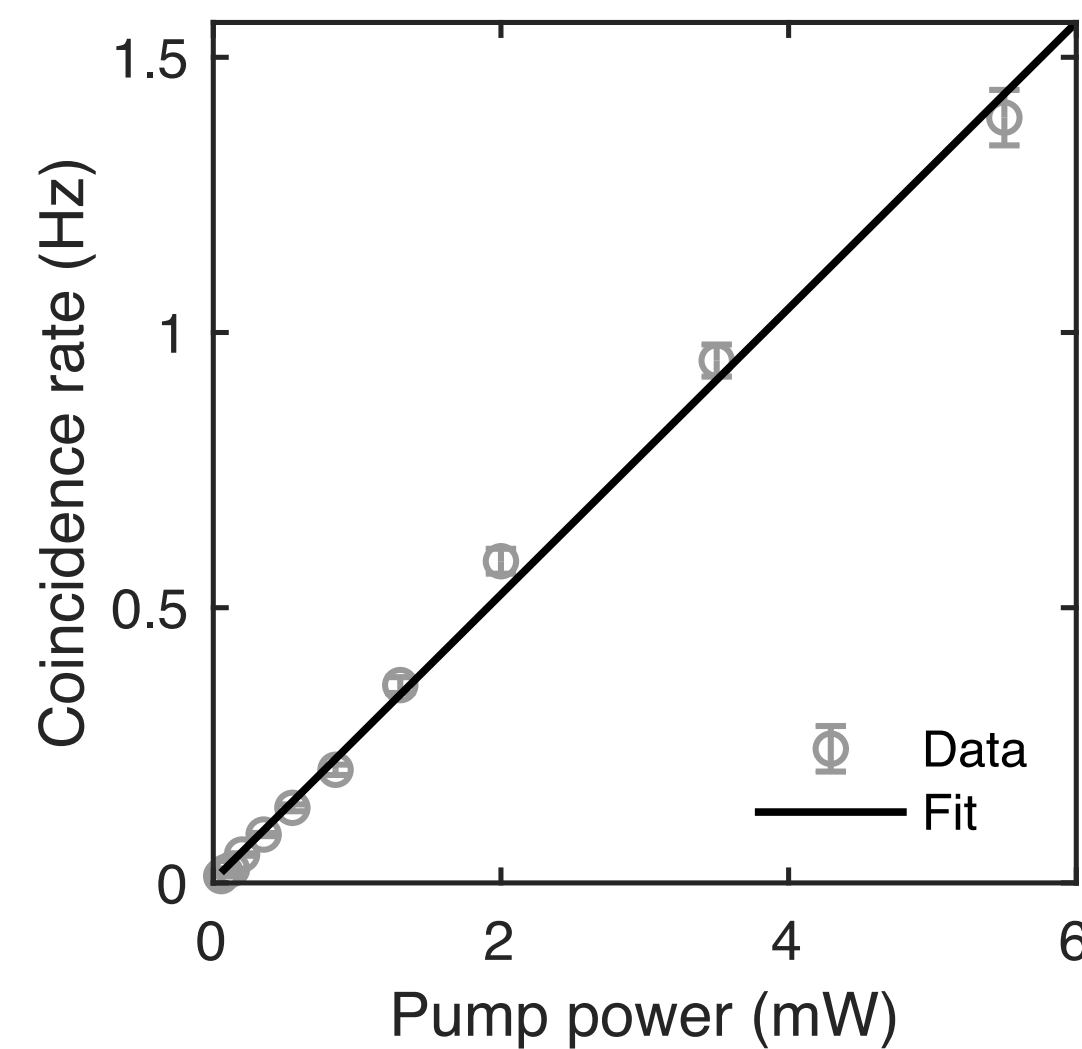
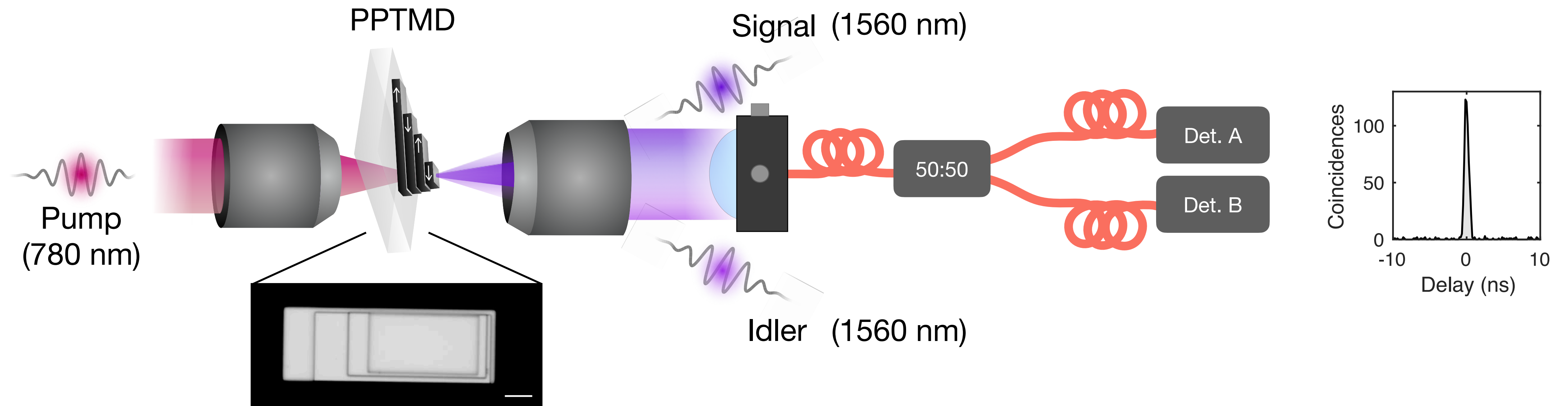
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Roadmap

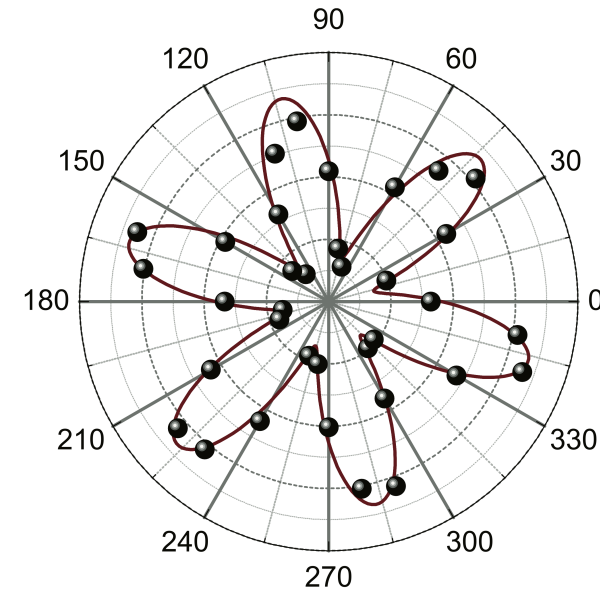
NONLINEAR OPTICS WITH TMDs

SHG from TMDs

2013

L. M. Malard et al., Phys. Rev. B
87, 201401(R), 2013

Y. Li. et al., Nano Lett. **13**,
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2024

See also works from De Paula, Heinz, Sun,
Eilenberger, Yu, Staude, Pertsch,
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Roadmap

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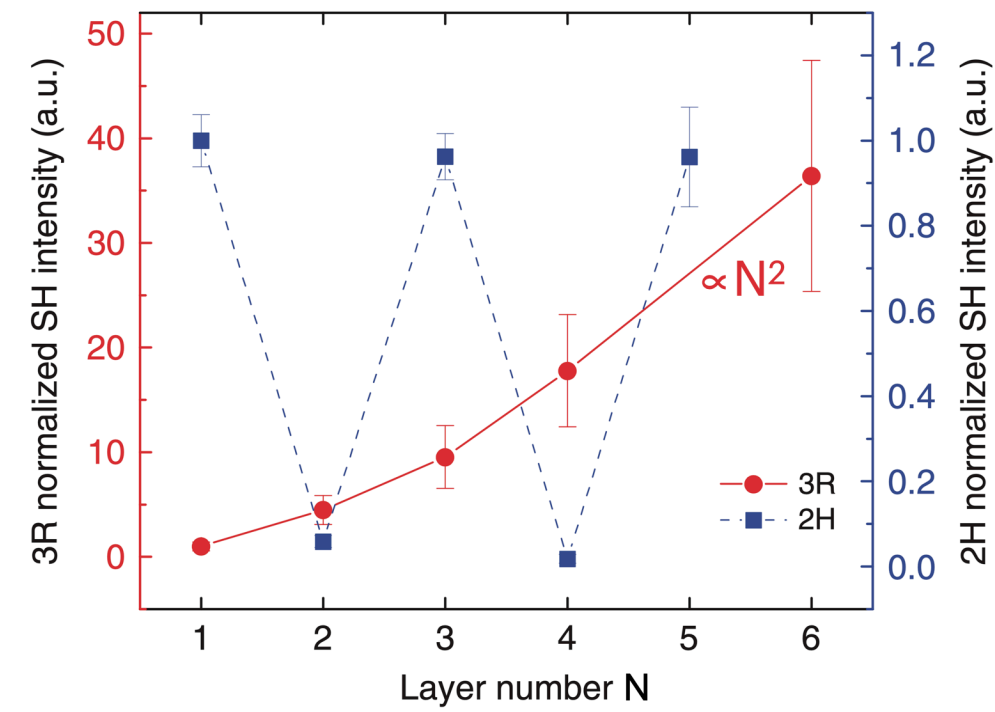
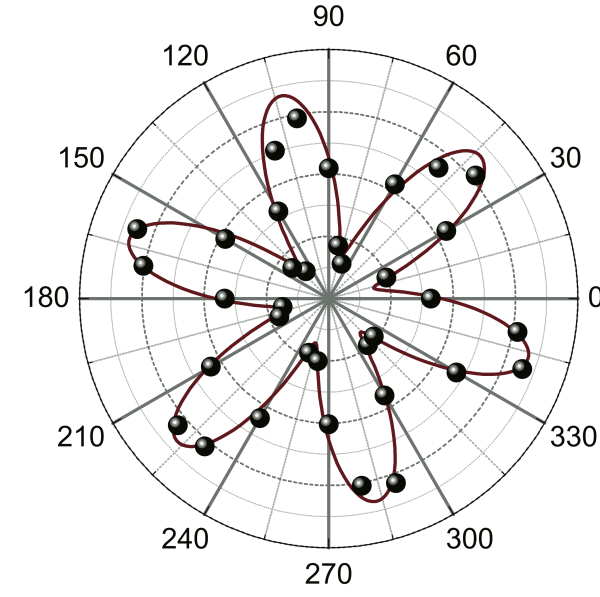
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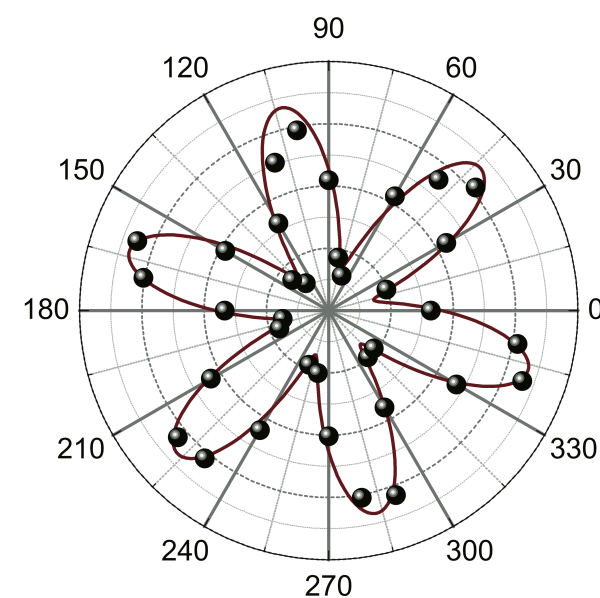
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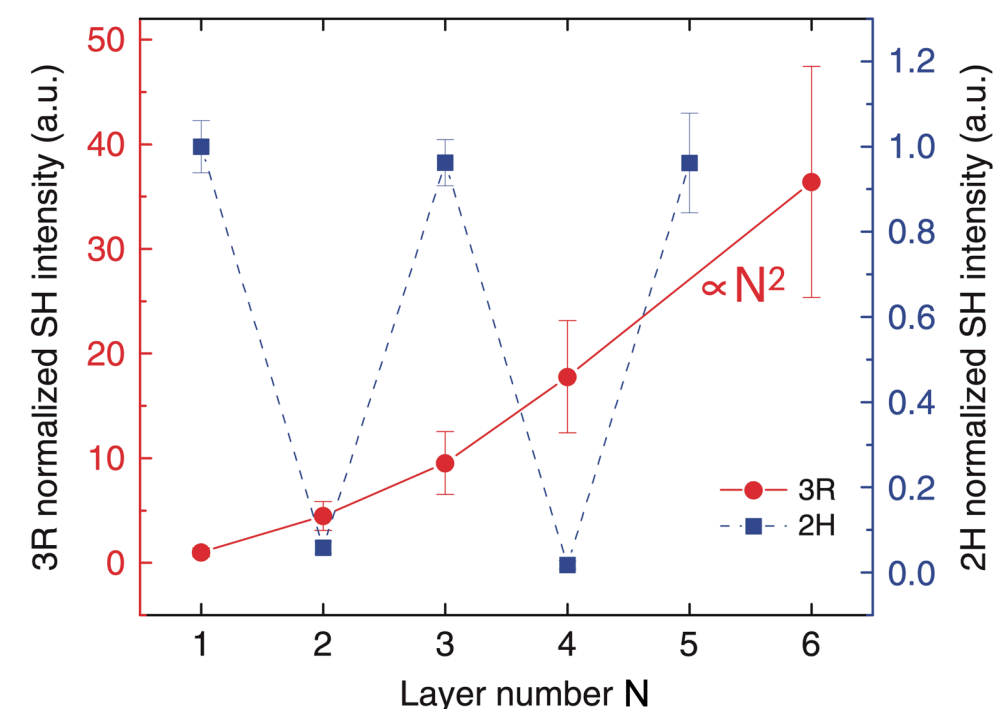
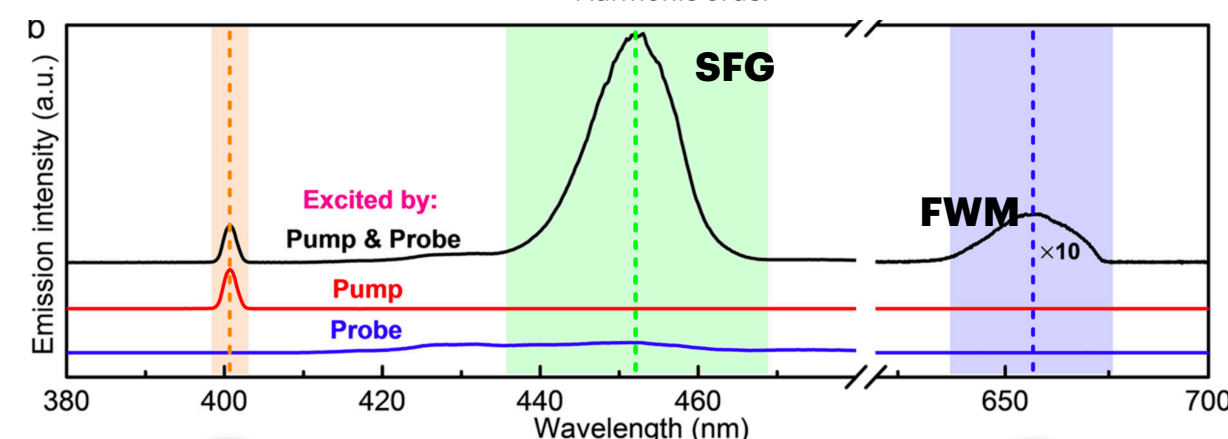
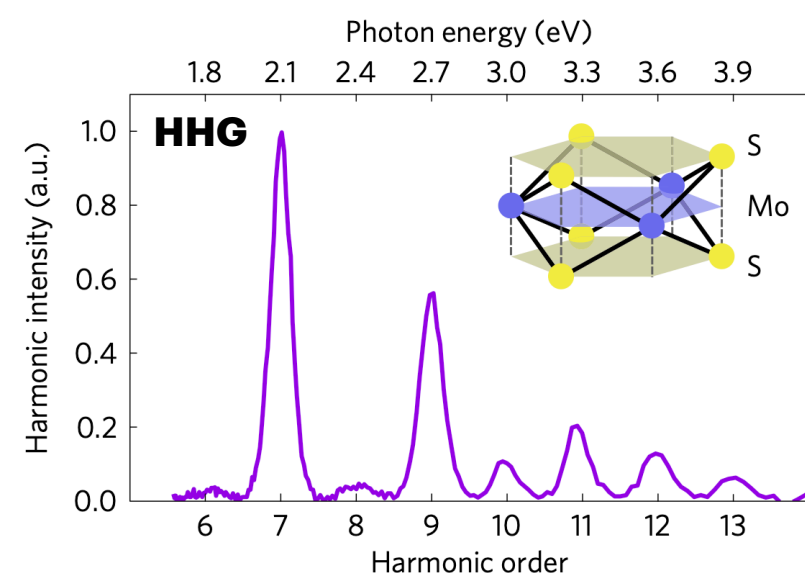
M. Zhao et al., *Light Sci. & App.*, **5**,
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D. Li et al. ACS Nano, **10**, 3,
3766-3775, 2016

H. Liu et al., *Nature Physics*, **13**,
6-10, 2017

SFG, THG, FWM, HHG



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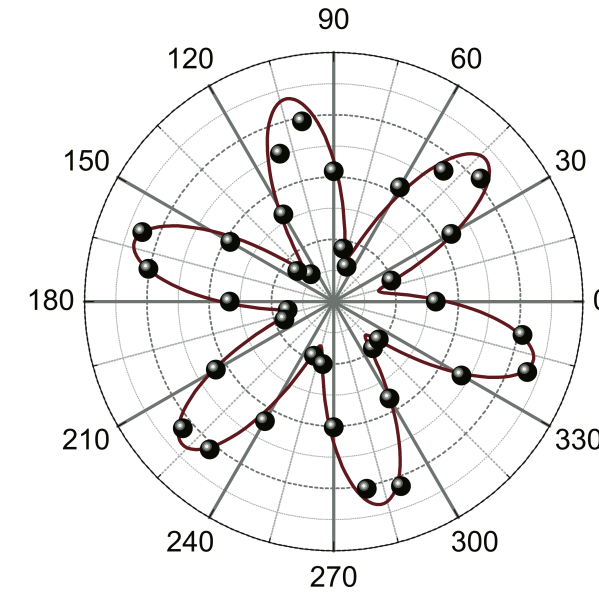
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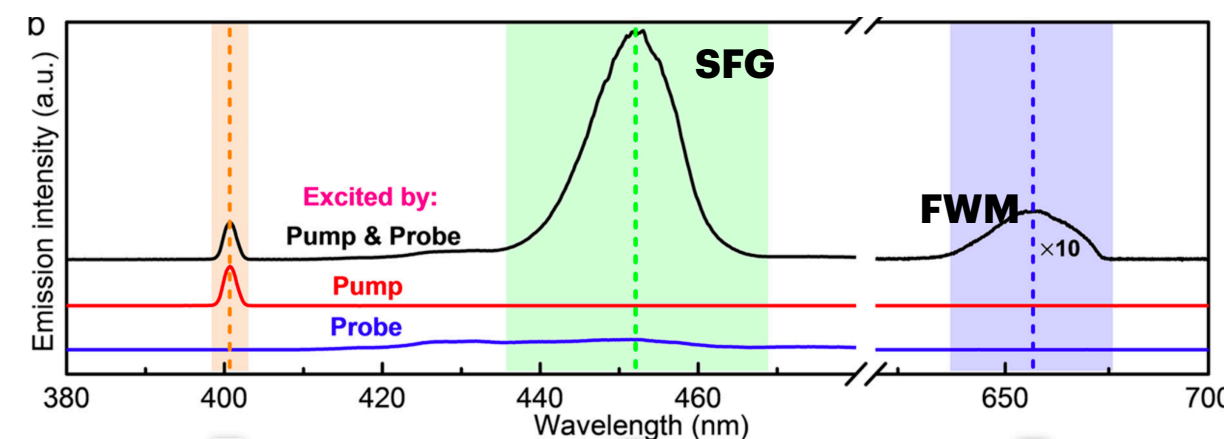
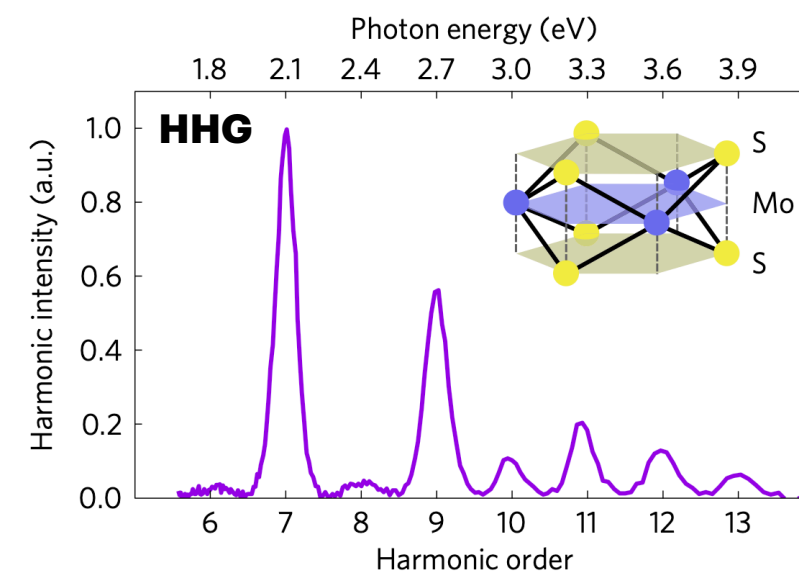
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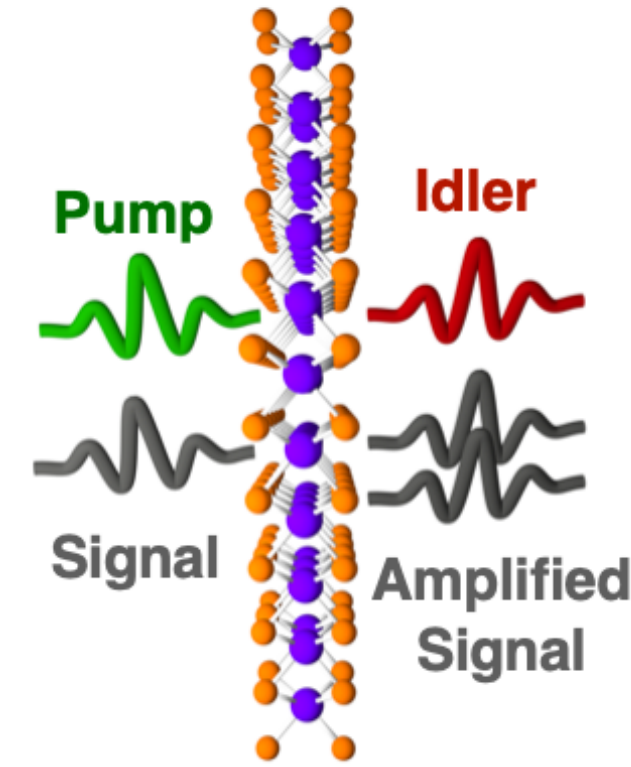
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SFG, THG, FWM, HHG



OPA in monolayer TMDs

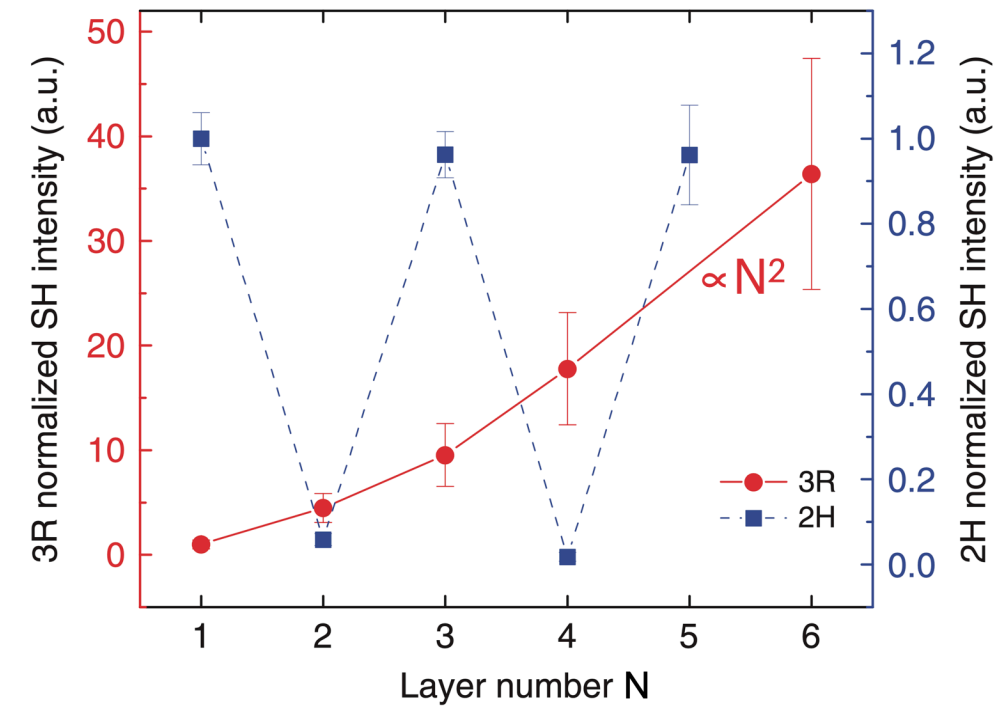


C. Trovatello et al., Nature Photonics, **15**, 6-10, 2021

2024

SHG in 2H and 3R-TMD

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Roadmap NONLINEAR OPTICS WITH TMDs

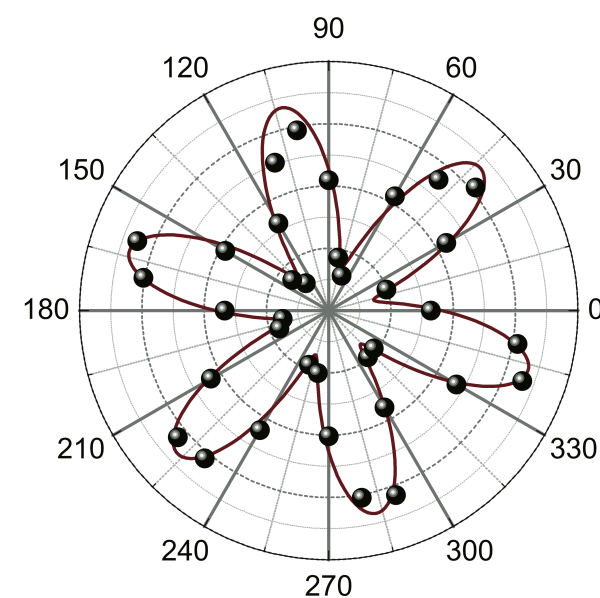
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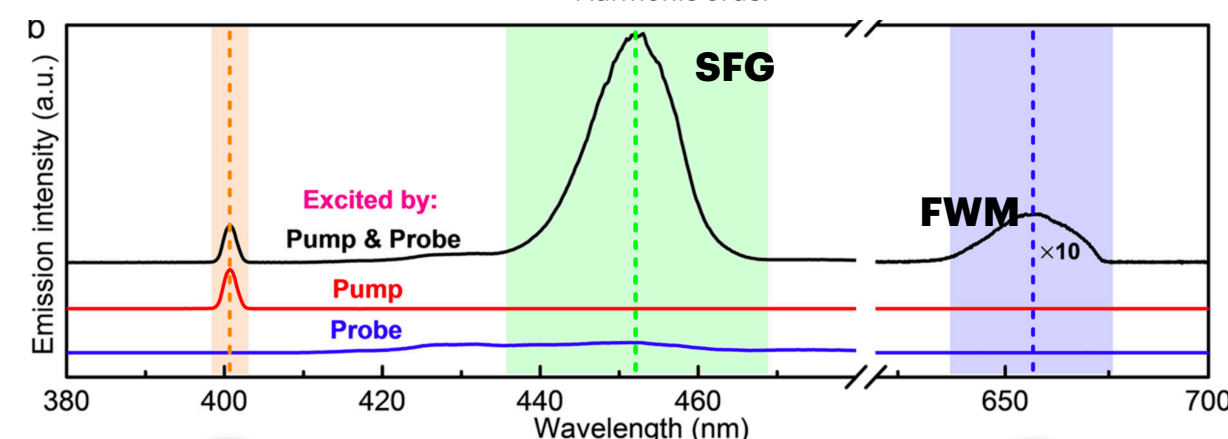
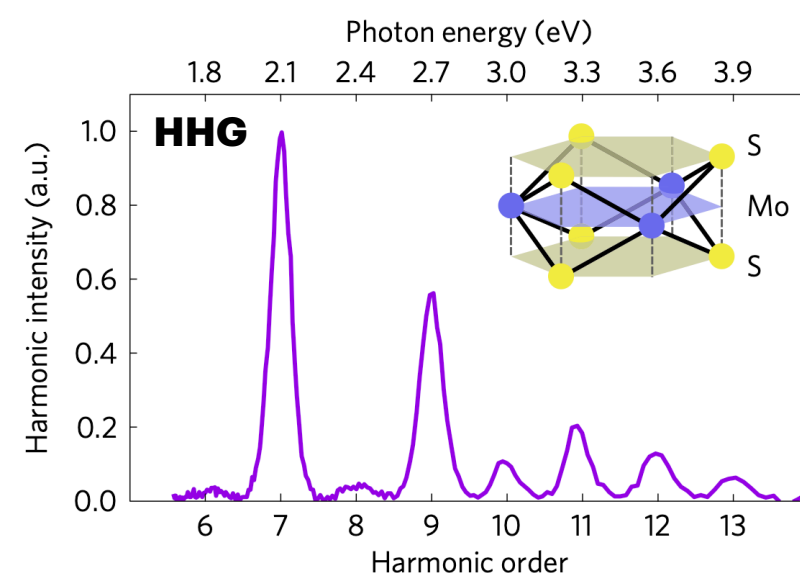
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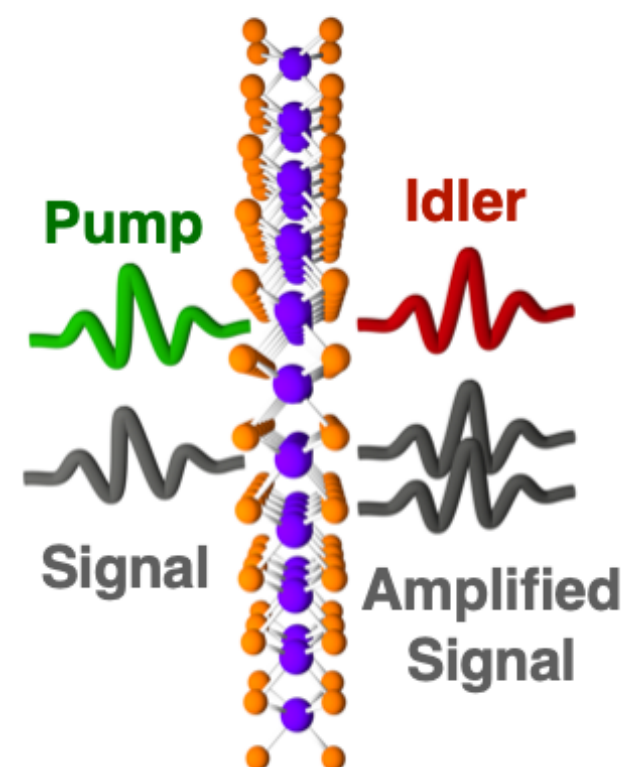
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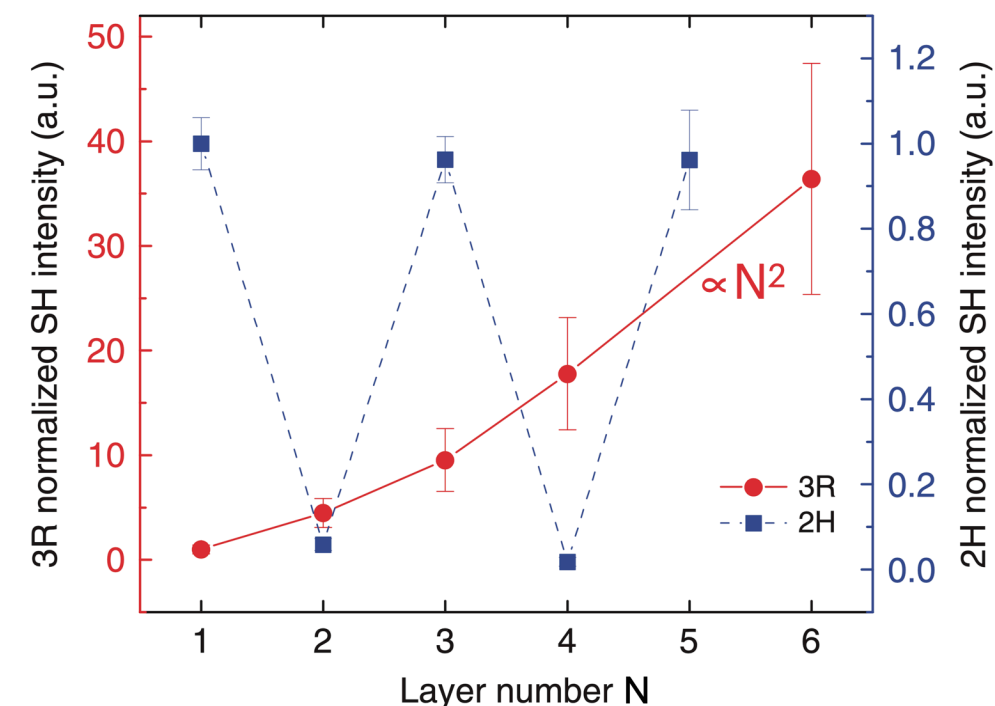


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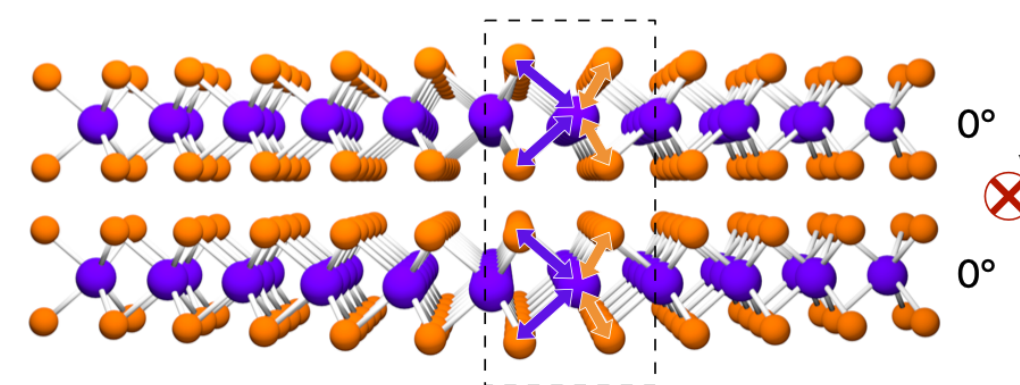
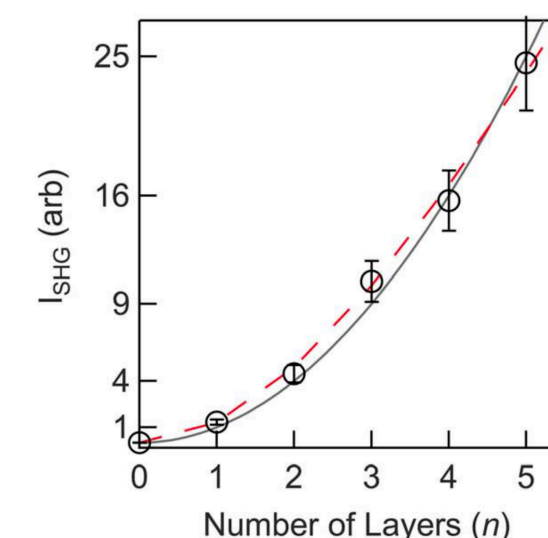
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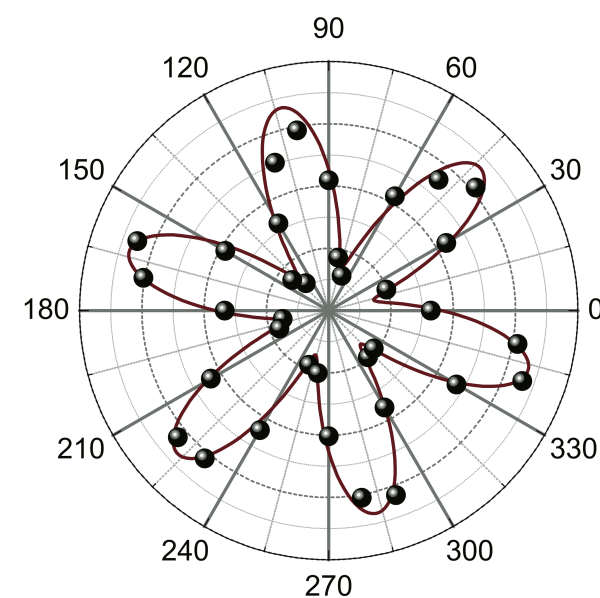
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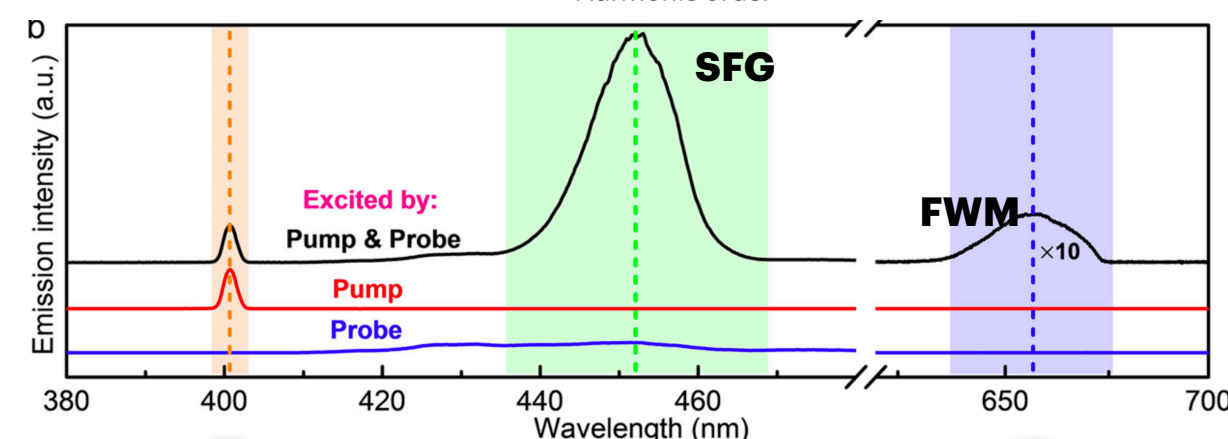
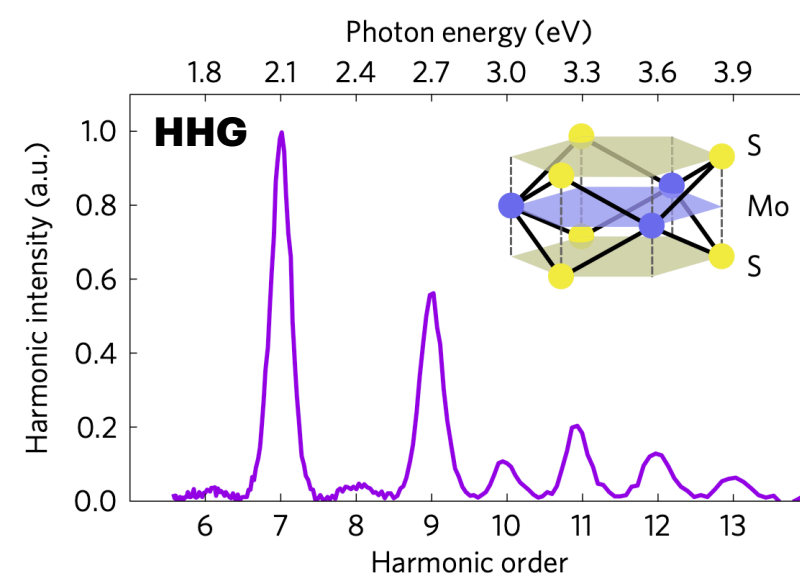
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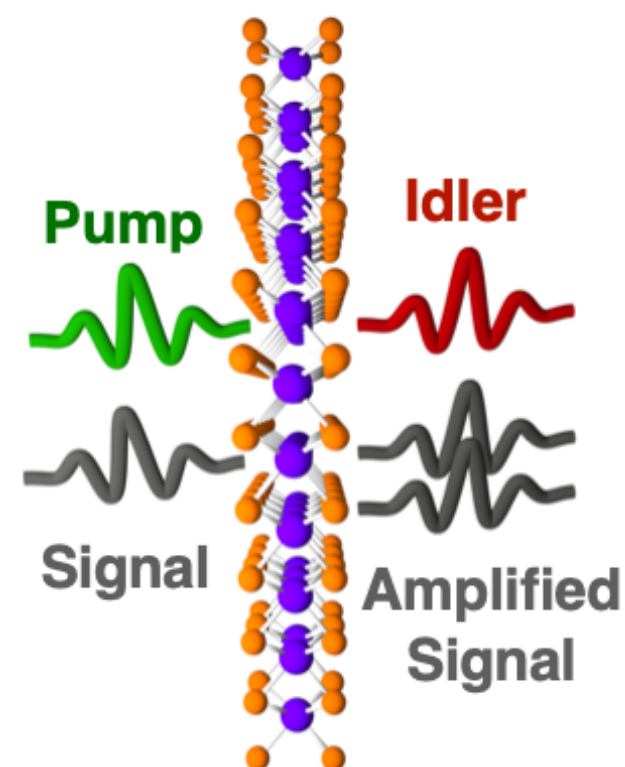
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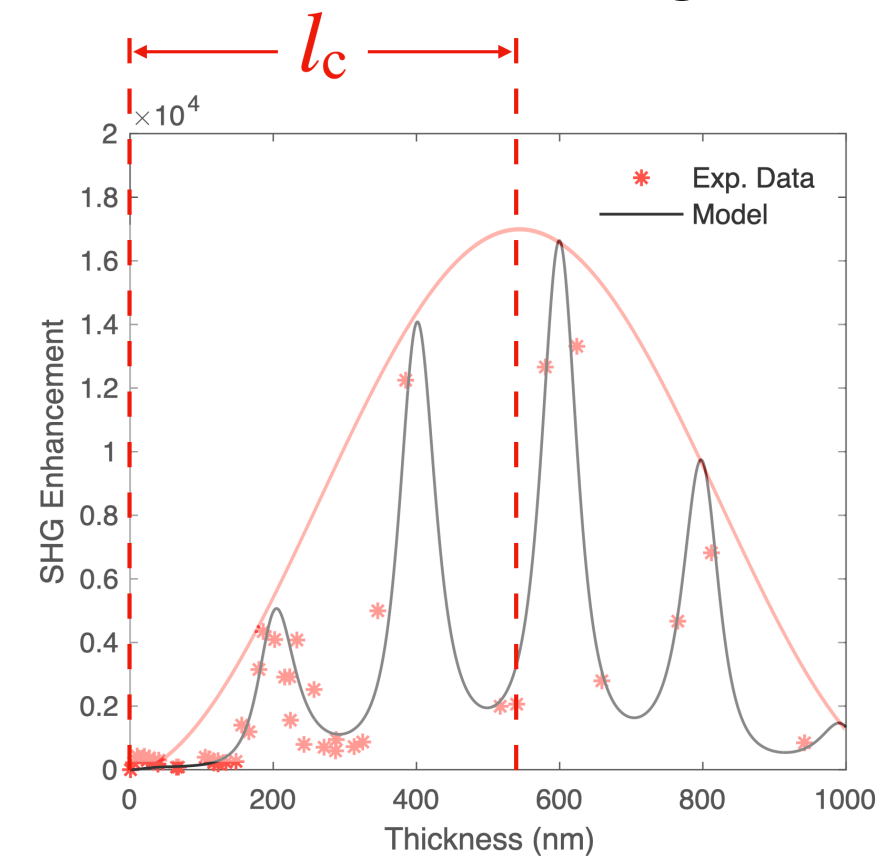


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C. Trovatello et al., Nature Photonics, **15**, 6-10, 2021

Coherence length

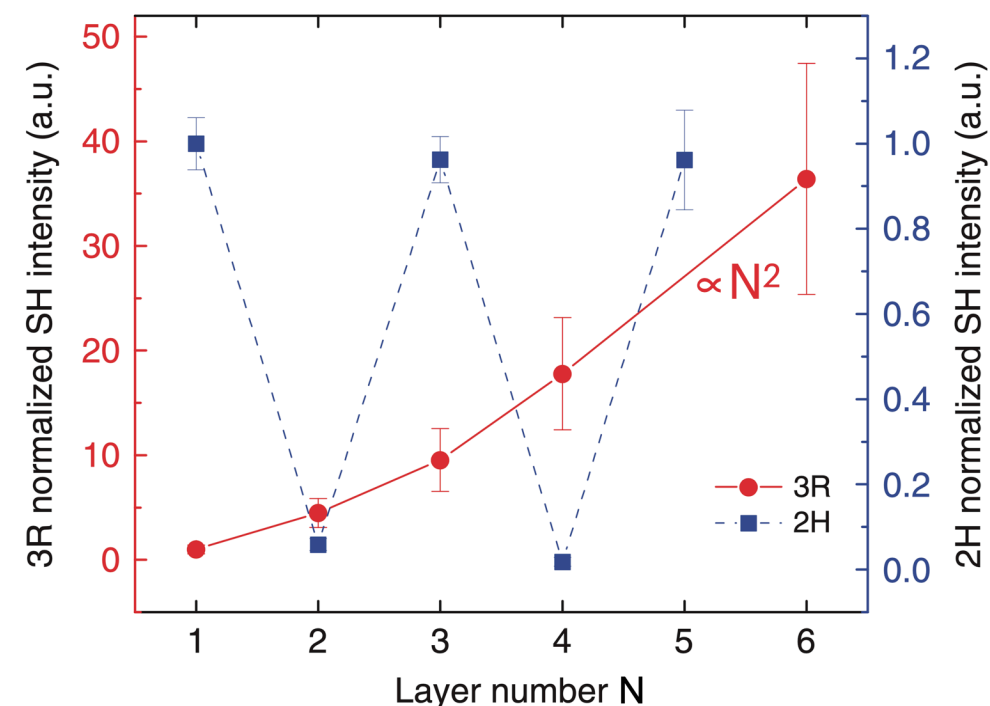


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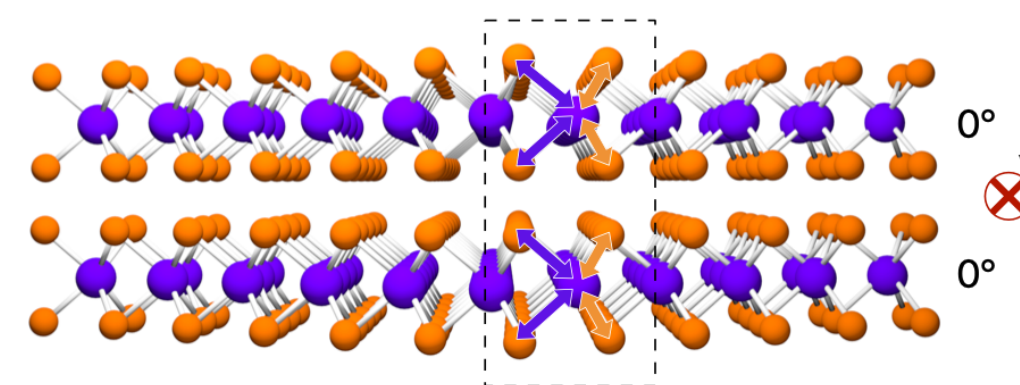
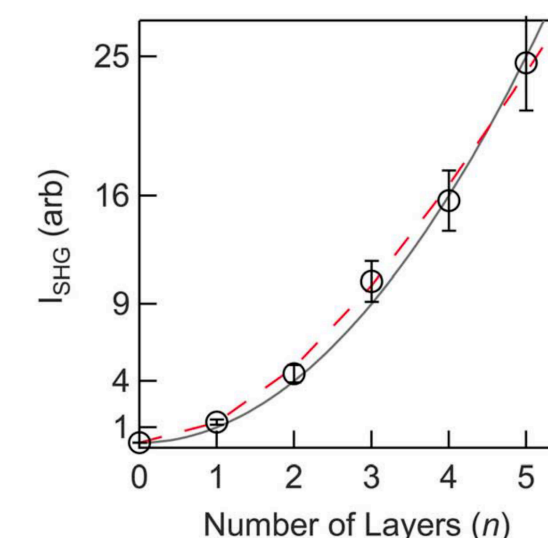
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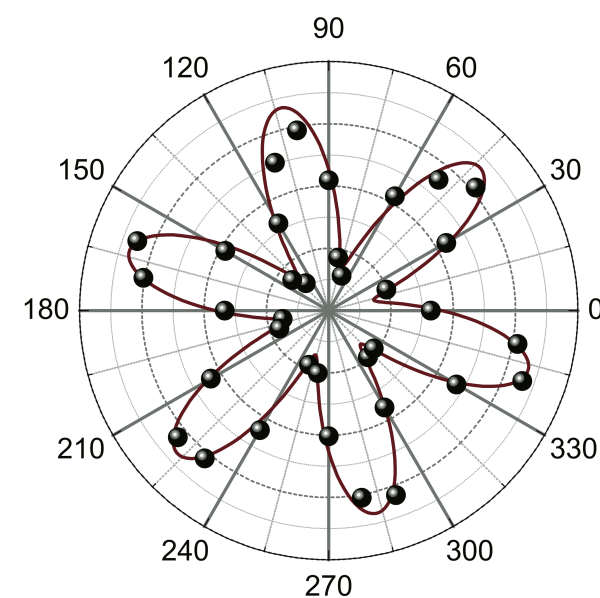
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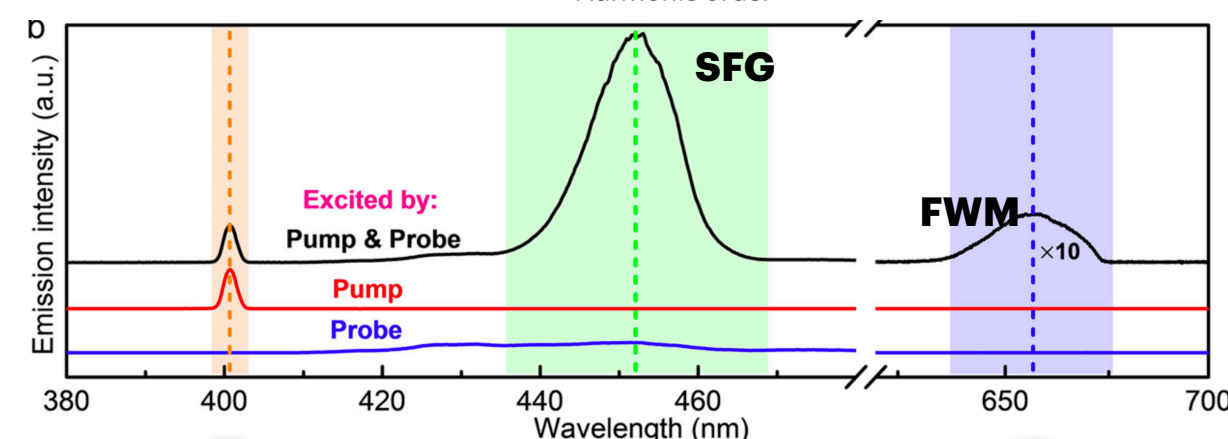
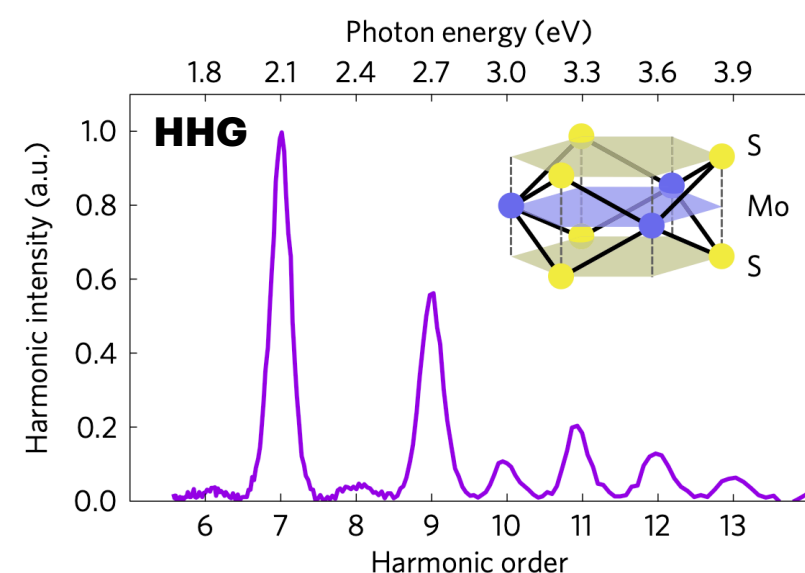
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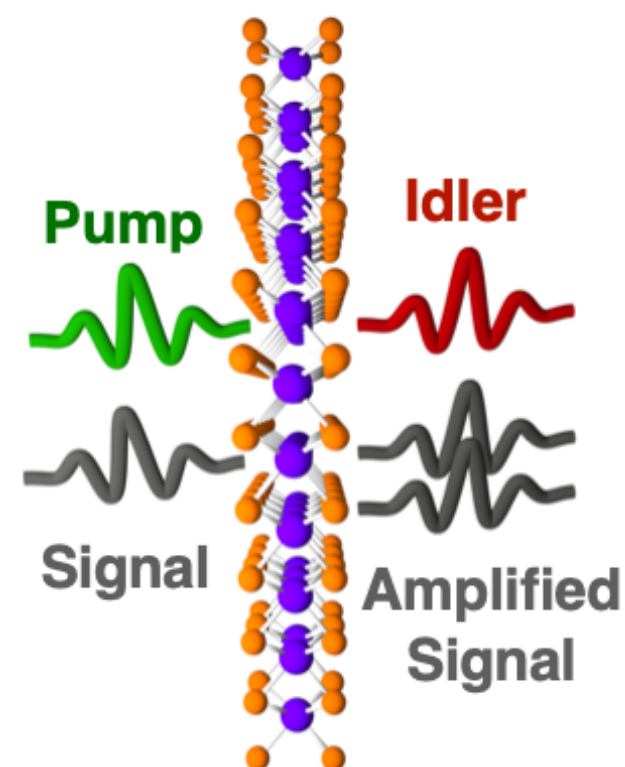
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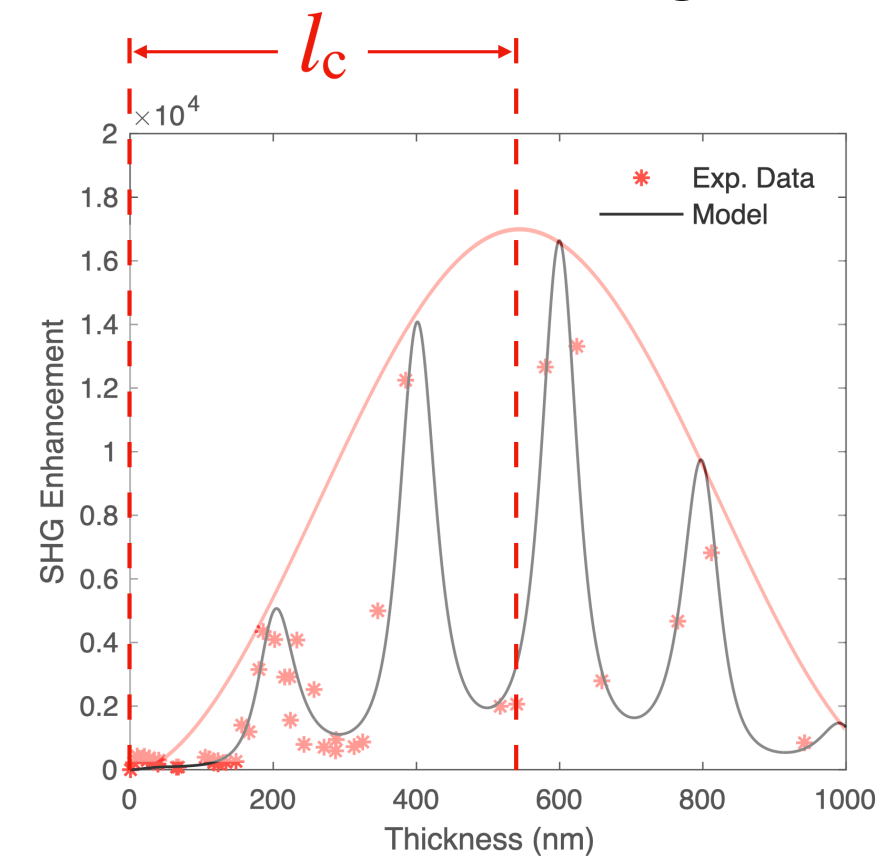


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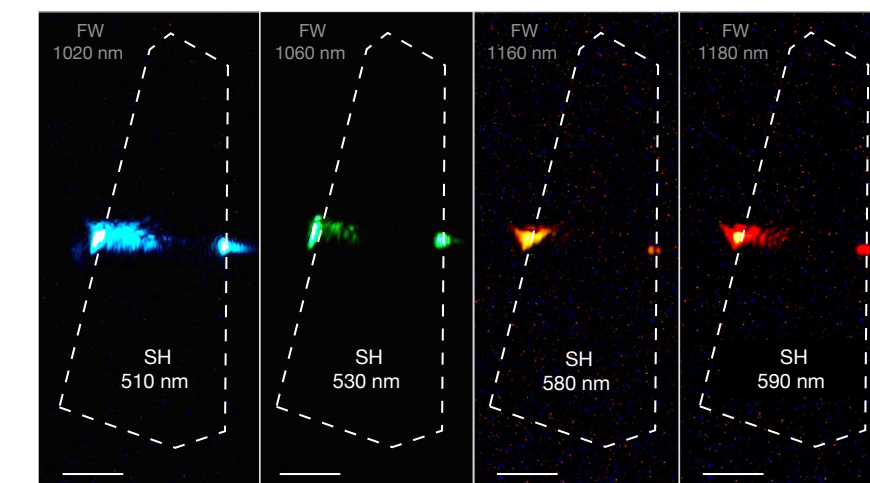
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TMD waveguides

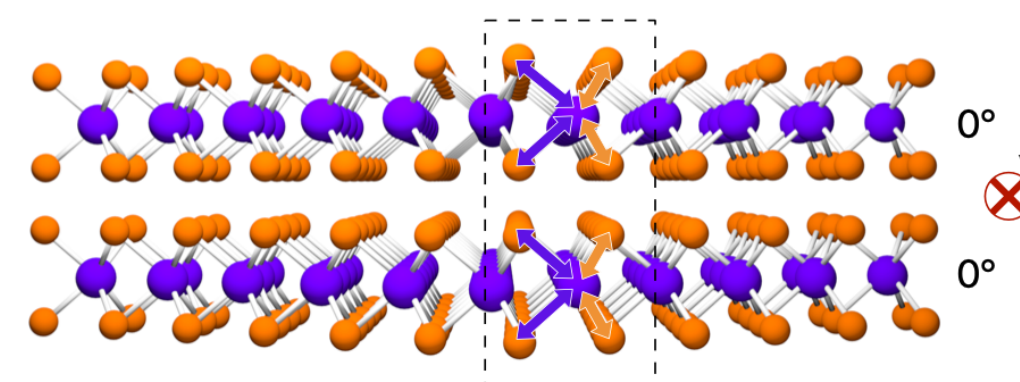
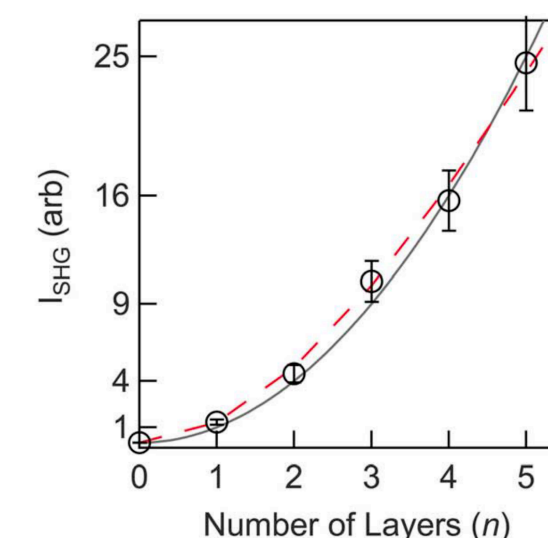


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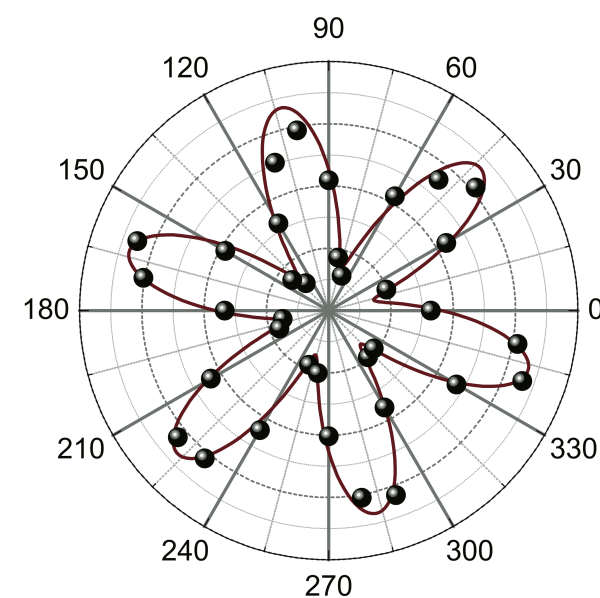
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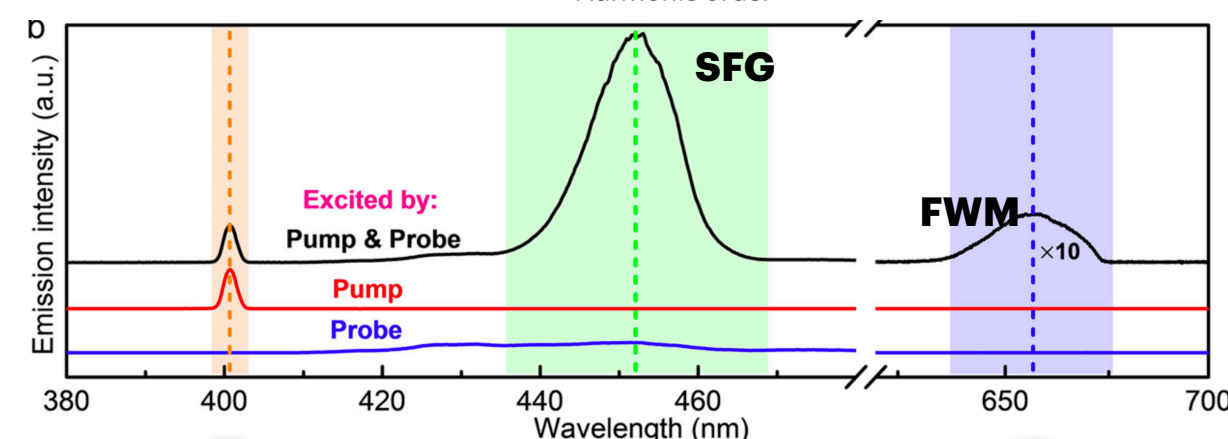
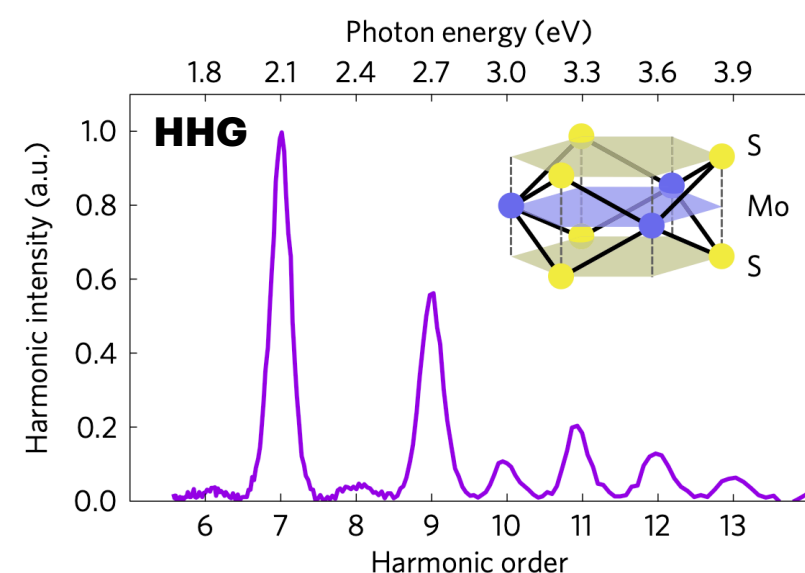
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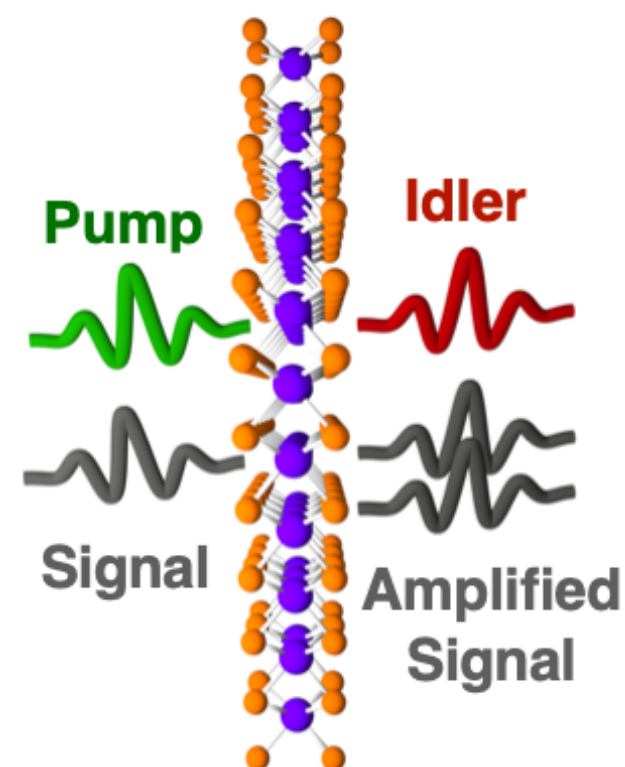
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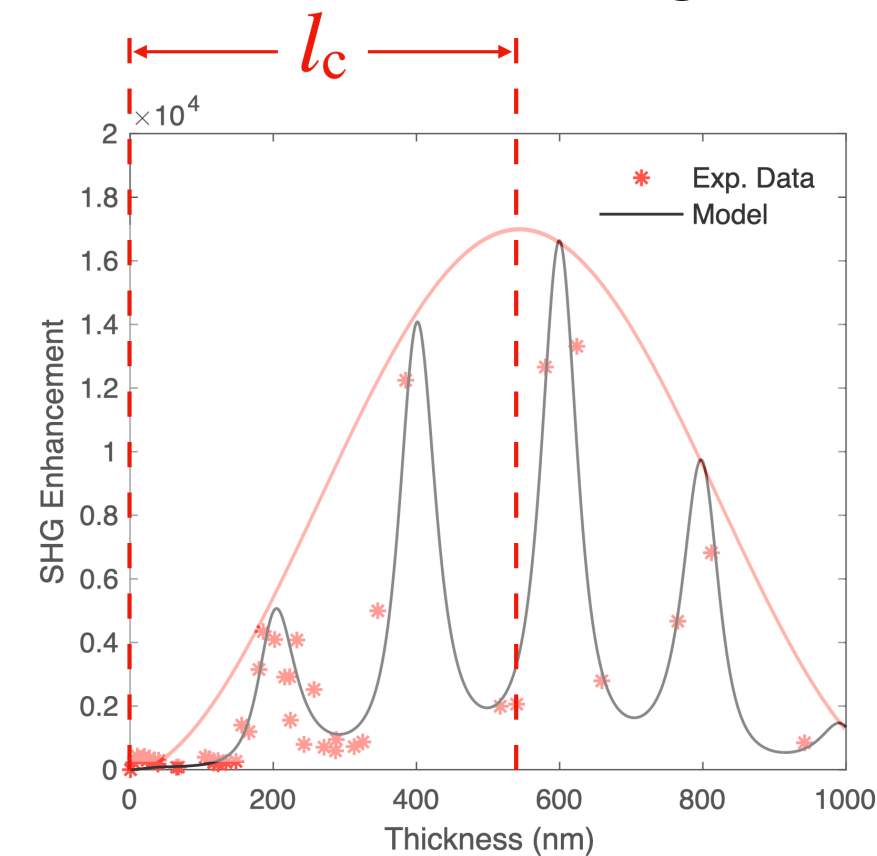


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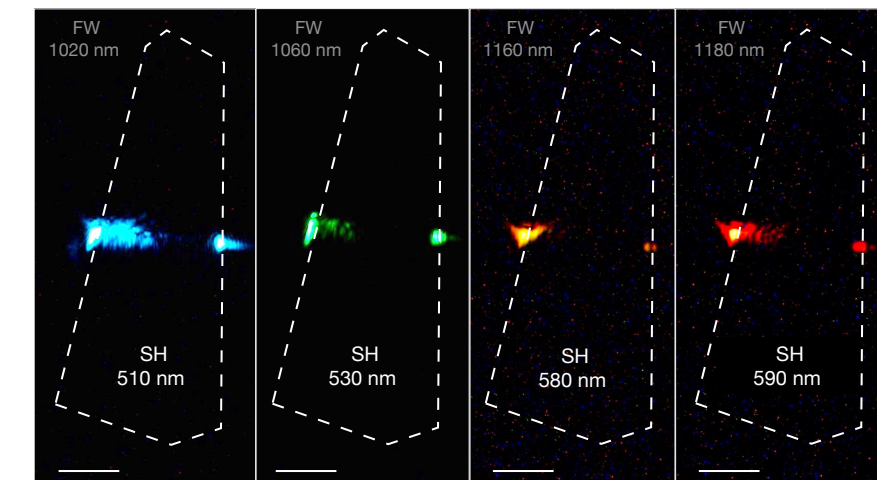
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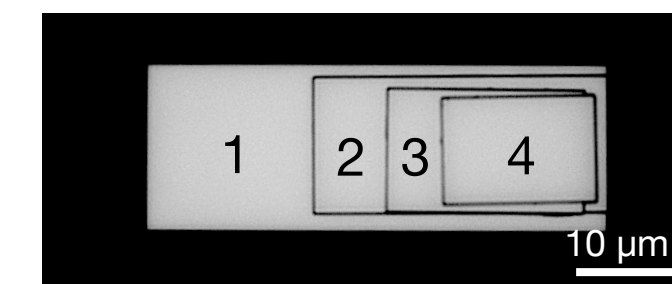
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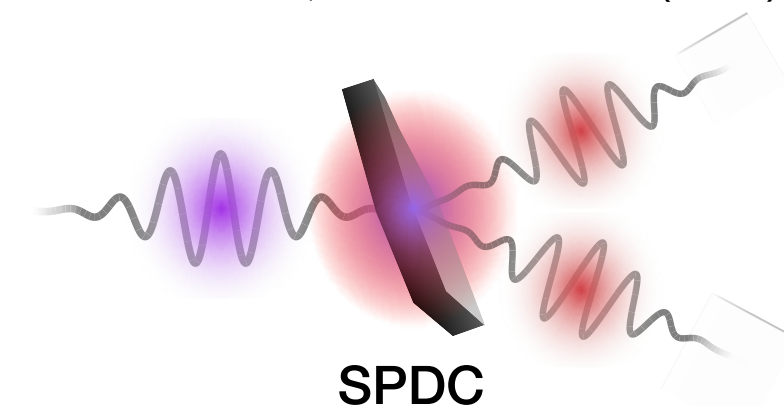
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Periodically Poled TMDs



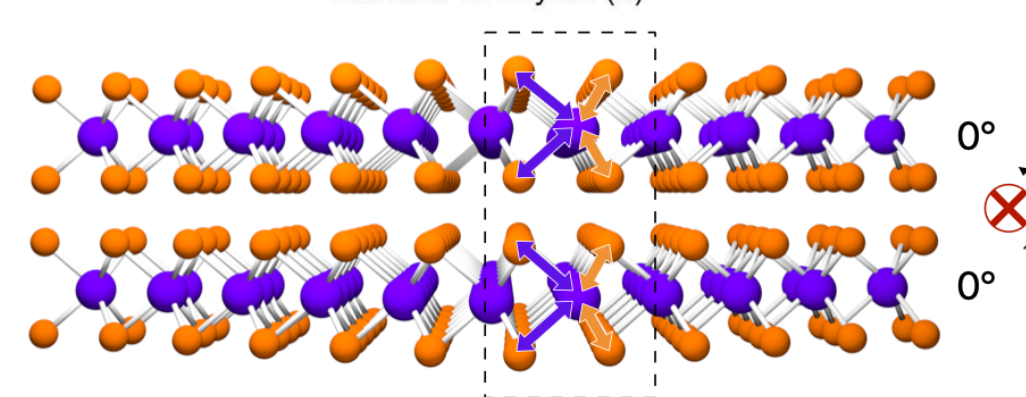
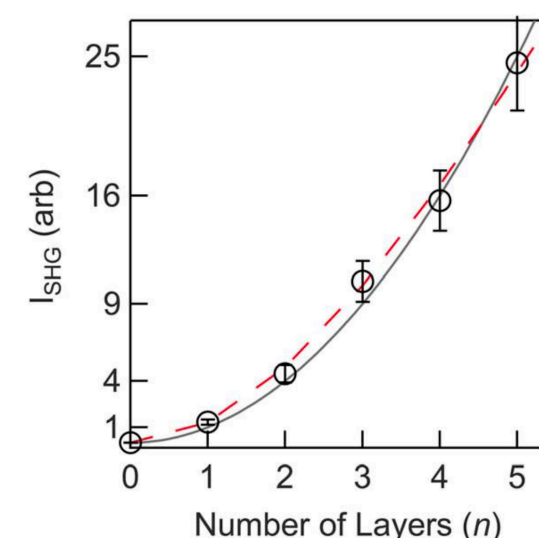
C. Trovatiello et al., Nature Photonics (2025)



On-chip integrable,
programmable,
microscopic
entangled photon source

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N² enhancement in artificial AA stacks



Nonlinear optics

Quantum optics

Integrated optics



Low-dimensional materials

Acknowledgements



P. James Schuck

Xinyi Xu

Zhi Hao (Benson) Peng

Changhwan Lee

Kaiyuan Yao

Emanuil Yanev

Xiaoyang Zhu

Fang Liu

James Hone

Nicola Curreli

Dmitri Basov

Fabian Mooshammer

Cory Dean

Birui Yang

Michal Lipson

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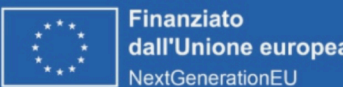
Andrew Ye



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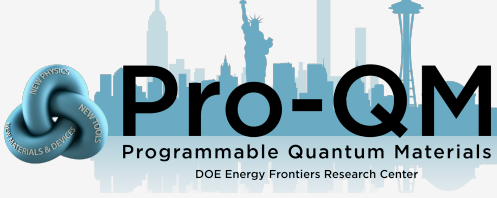
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DI RIPRESA E RESILIENZA



NQSTI
National Quantum Science
and Technology Institute



PIONEER
MARIE SKŁODOWSKA-CURIE ACTION



Pro-QM
Programmable Quantum Materials
DOE Energy Frontiers Research Center

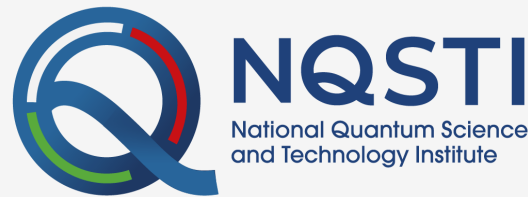
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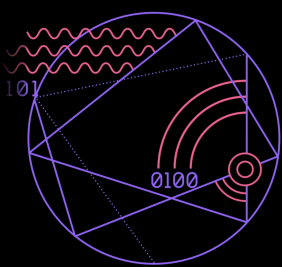
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Use photonics. Find a solution. Change the world.

2024**AMBASSADORS**



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Ultracompact entangled photon sources based on 2D semiconductors

Chiara Trovatello – chiara.trovatello@polimi.it

Assistant Professor – Politecnico di Milano, Milano, Italy

Marie Skłodowska-Curie Global Fellow – Columbia University, New York, NY, USA



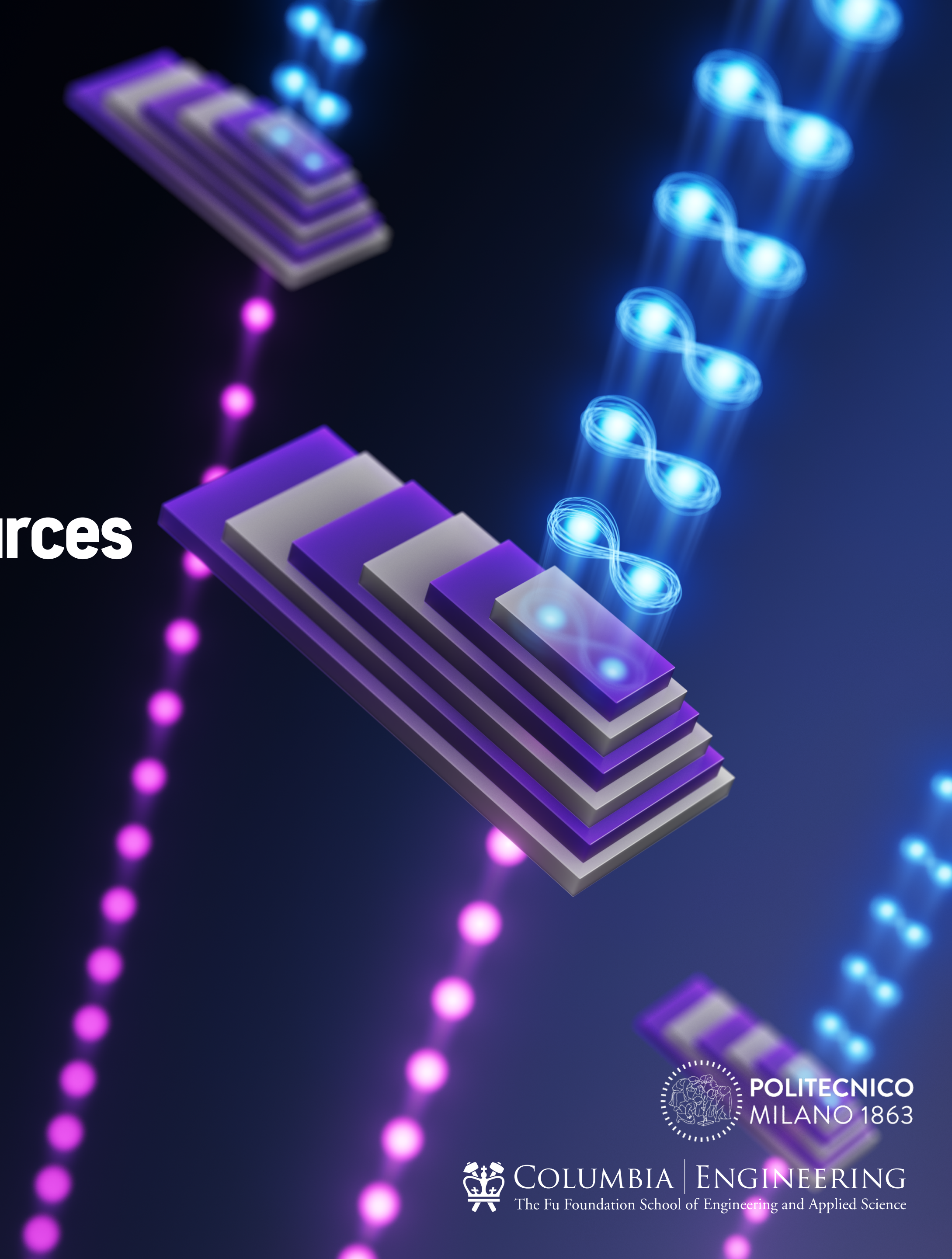


Extra: Ultracompact entangled photon sources based on 2D semiconductors

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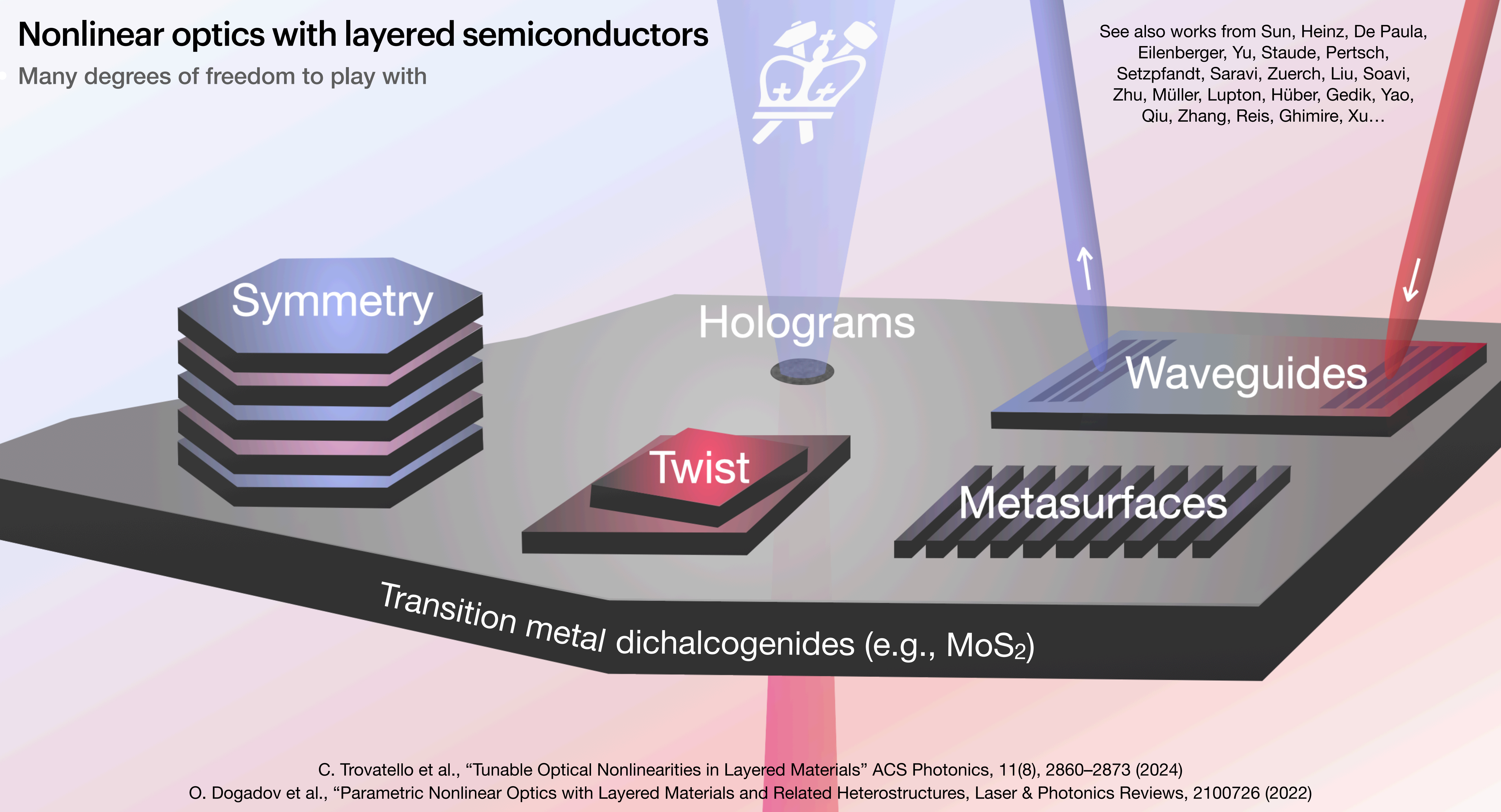
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Nonlinear optics with layered semiconductors

- Many degrees of freedom to play with

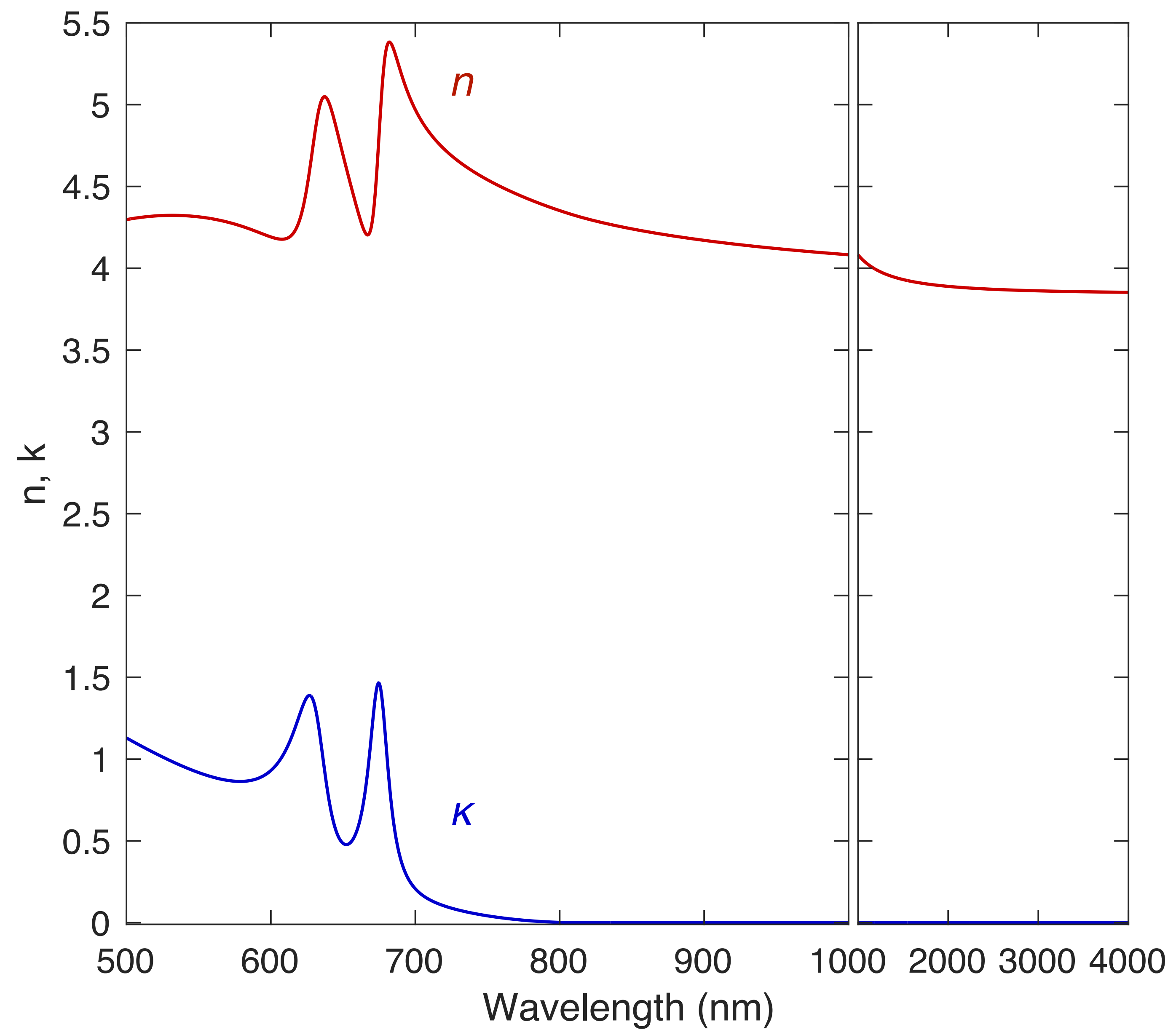
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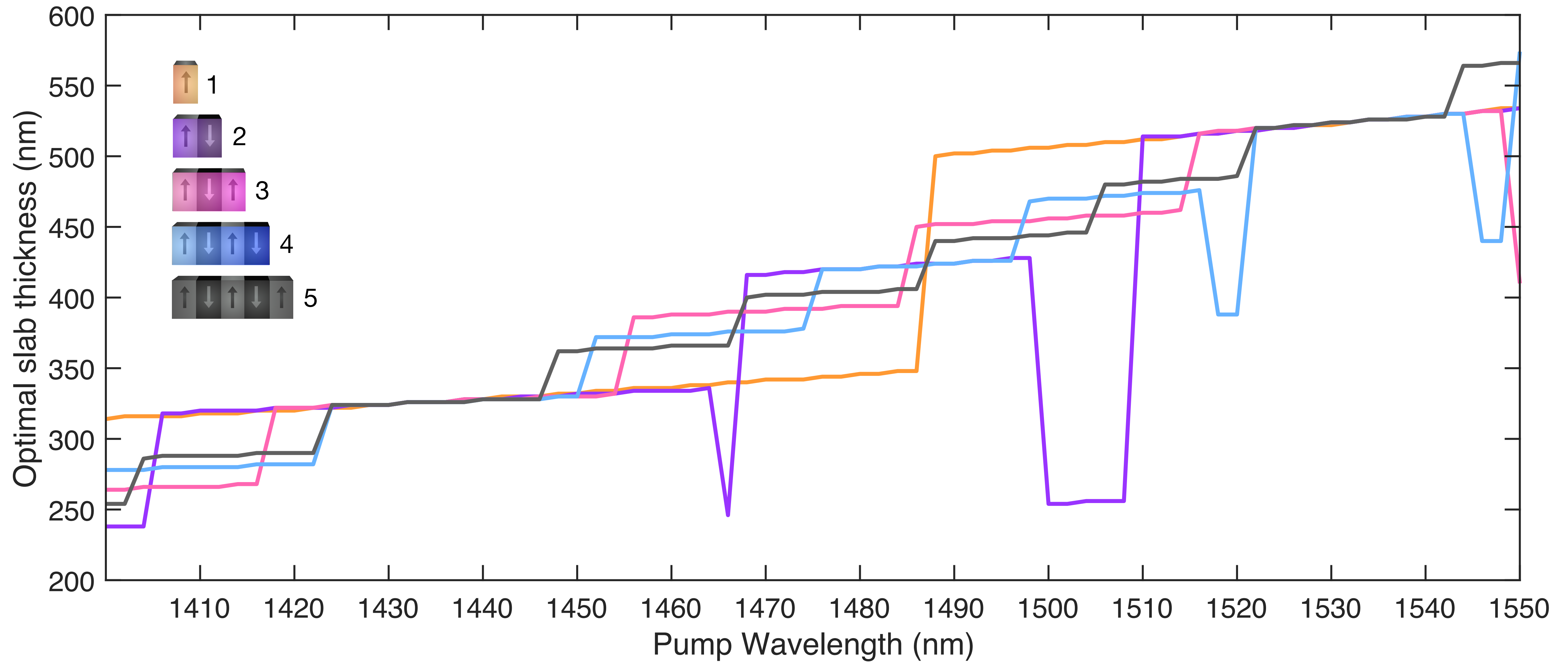
C. Trovatello et al., “Tunable Optical Nonlinearities in Layered Materials” ACS Photonics, 11(8), 2860–2873 (2024)

O. Dogadov et al., “Parametric Nonlinear Optics with Layered Materials and Related Heterostructures, Laser & Photonics Reviews, 2100726 (2022)

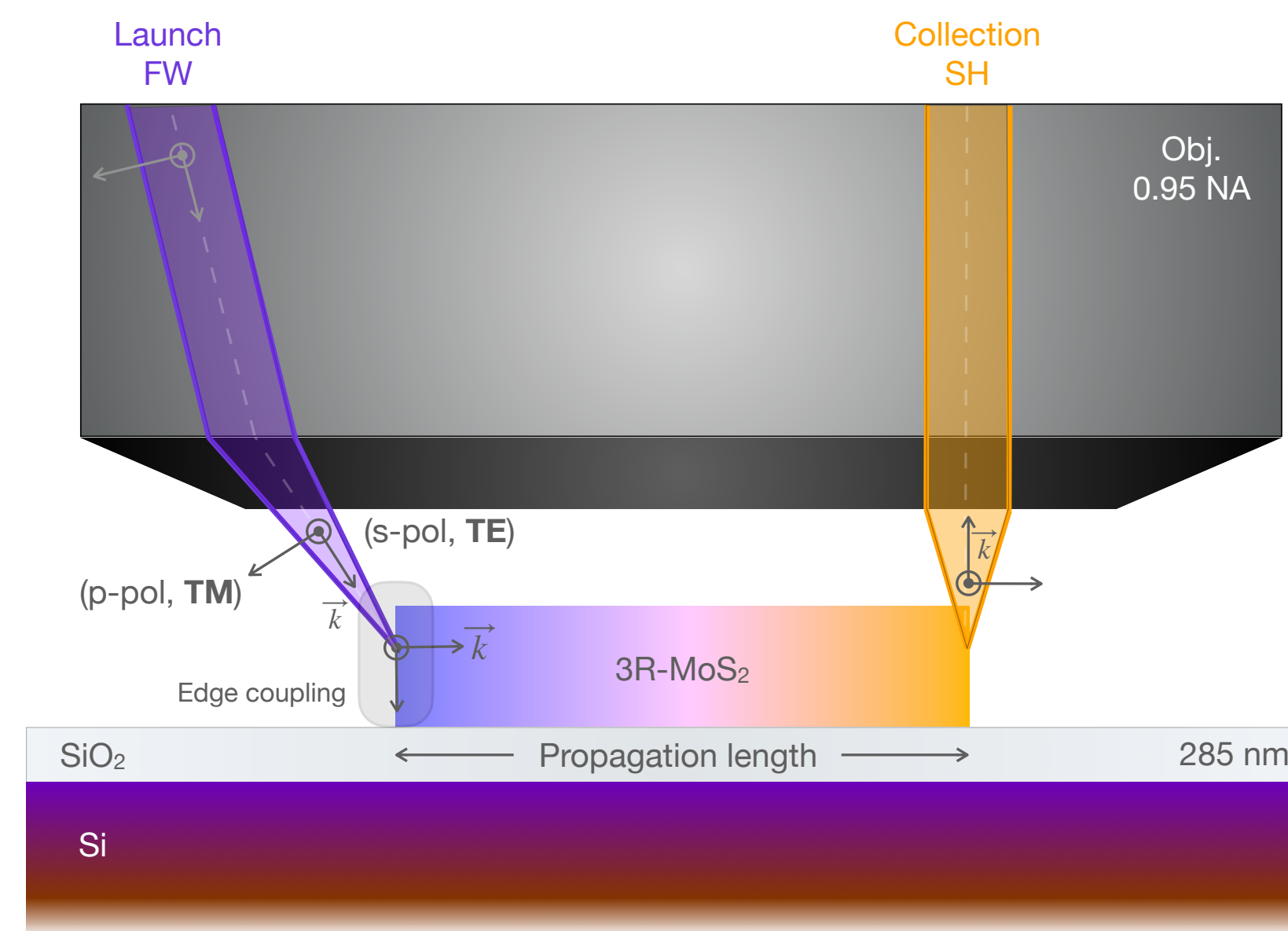
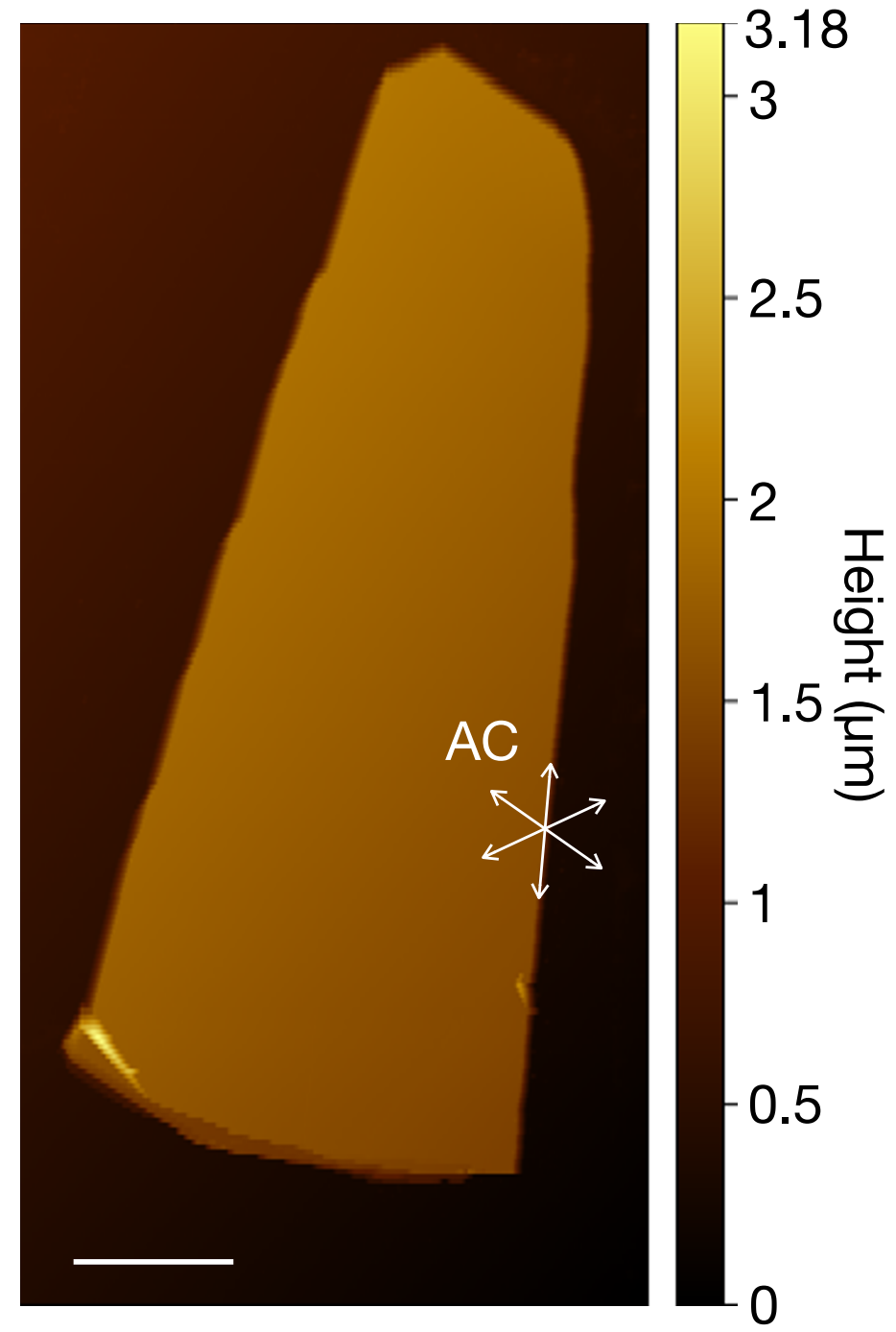
3R-MoS₂ refractive index



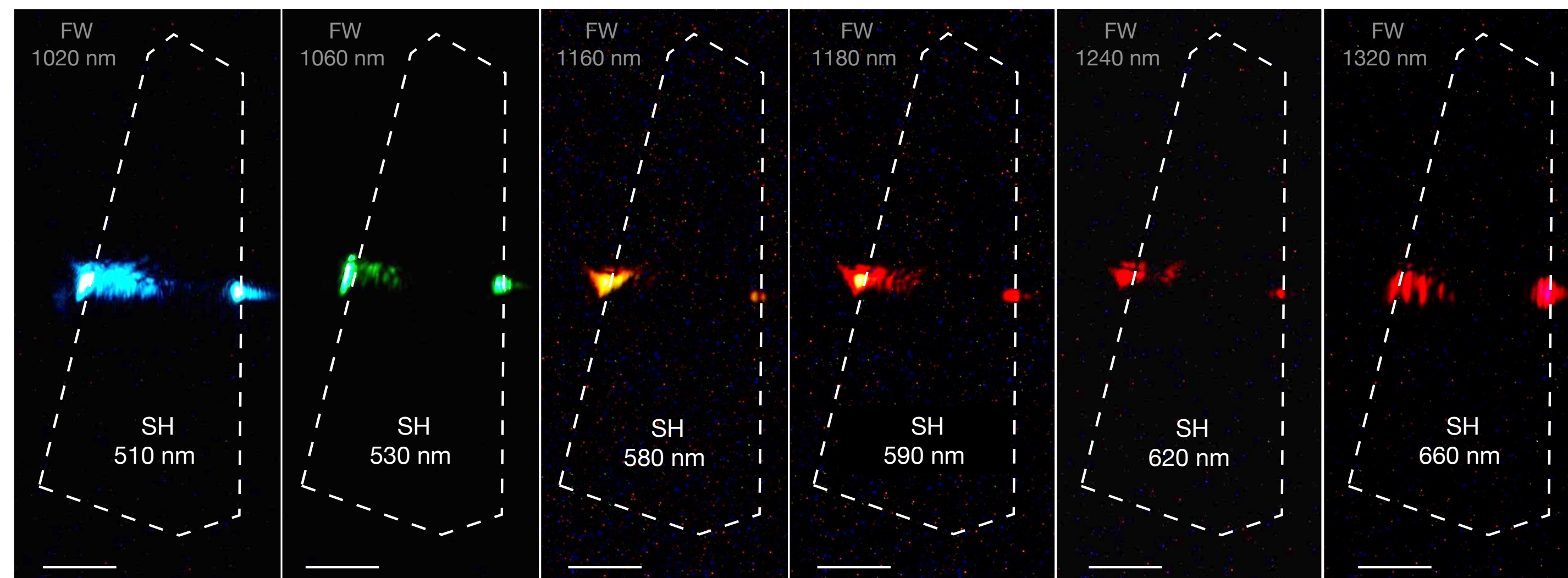
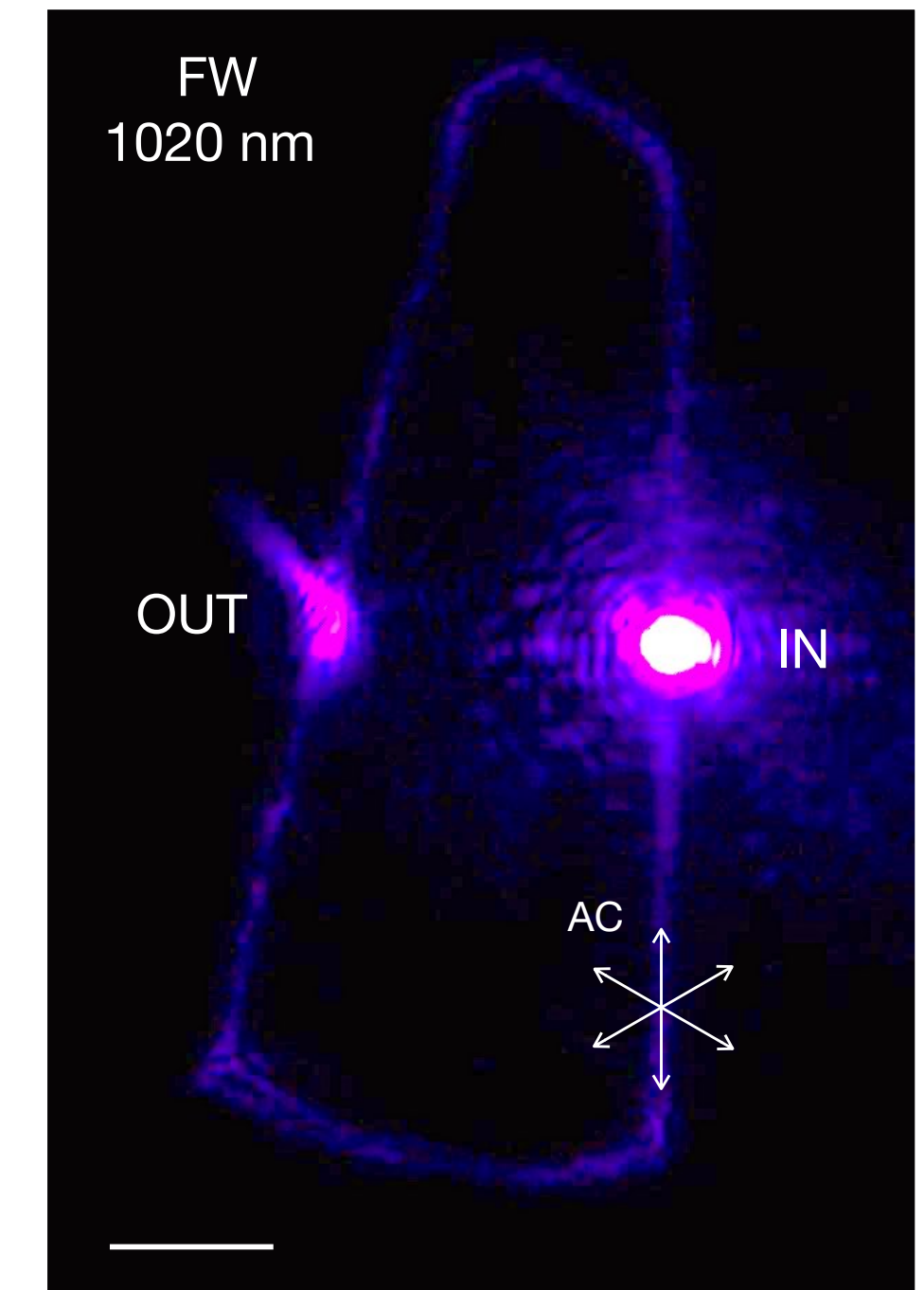
Theory model



A new geometry for TMDs

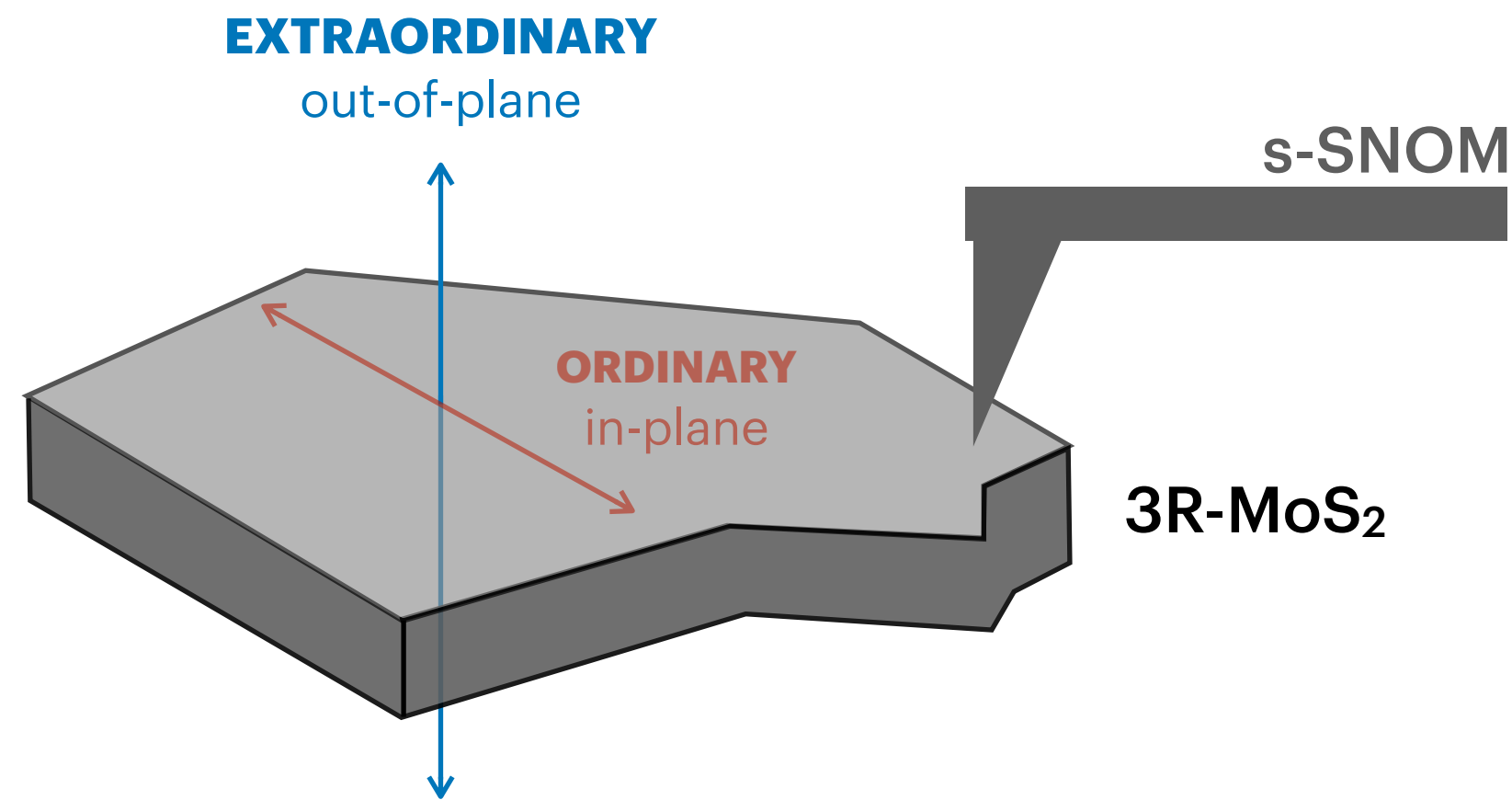


Xu, X., Trovatello, C., Mooshammer, F. *et al.* Towards compact phase-matched and waveguided nonlinear optics in atomically layered semiconductors. *Nature Photonics*, **16**, 698–706 (2022)

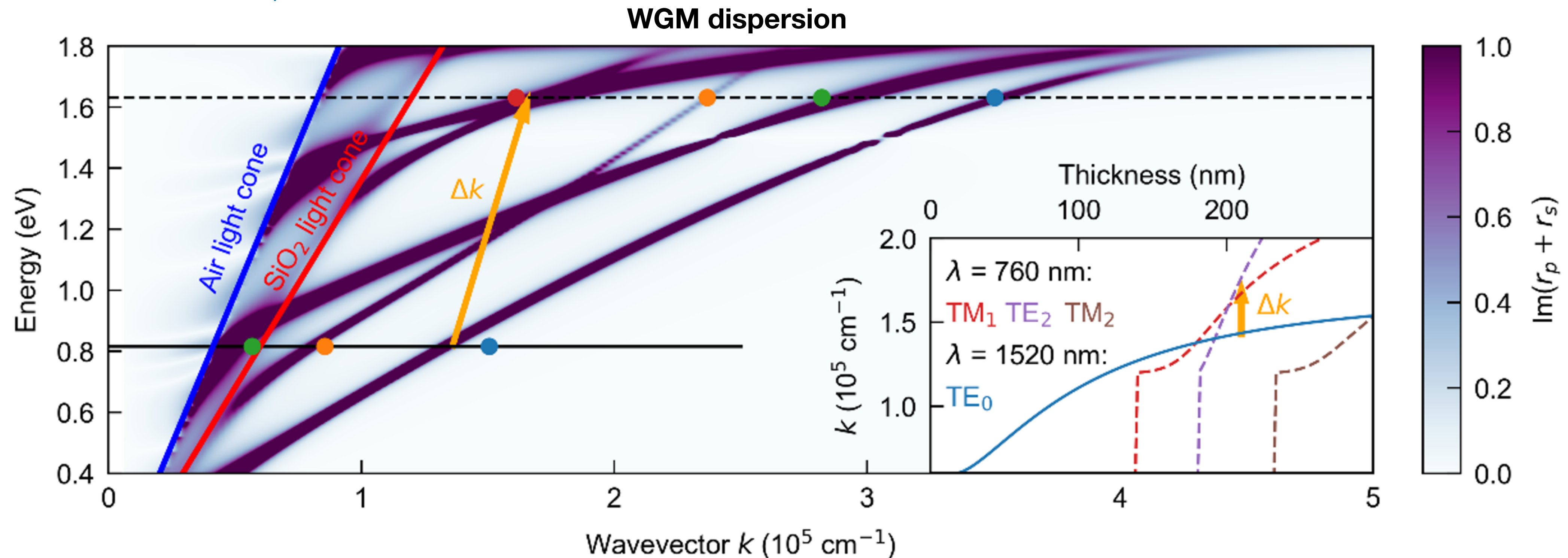


The TMD is a waveguide!

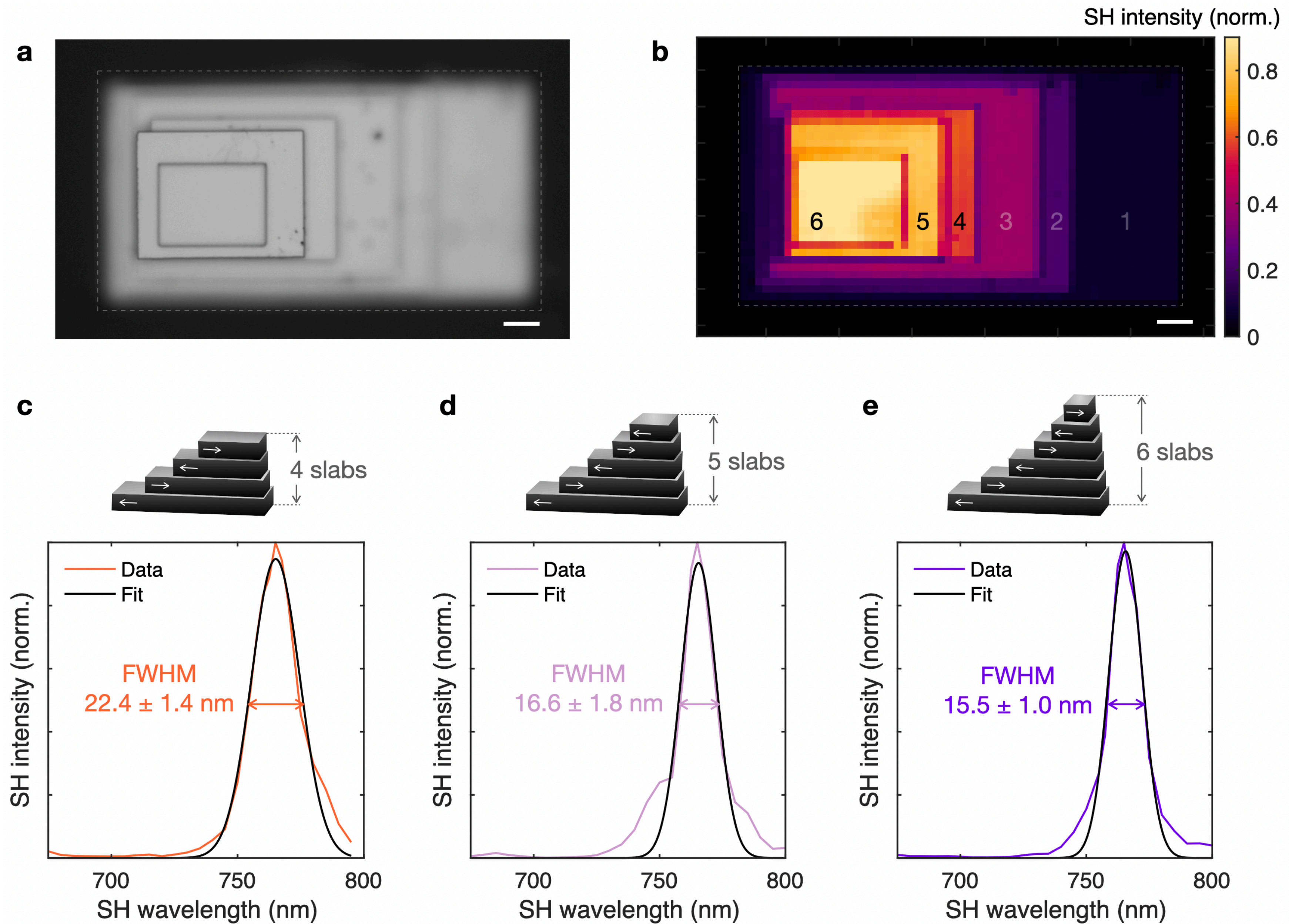
Towards phase-matched SHG in a waveguide geometry



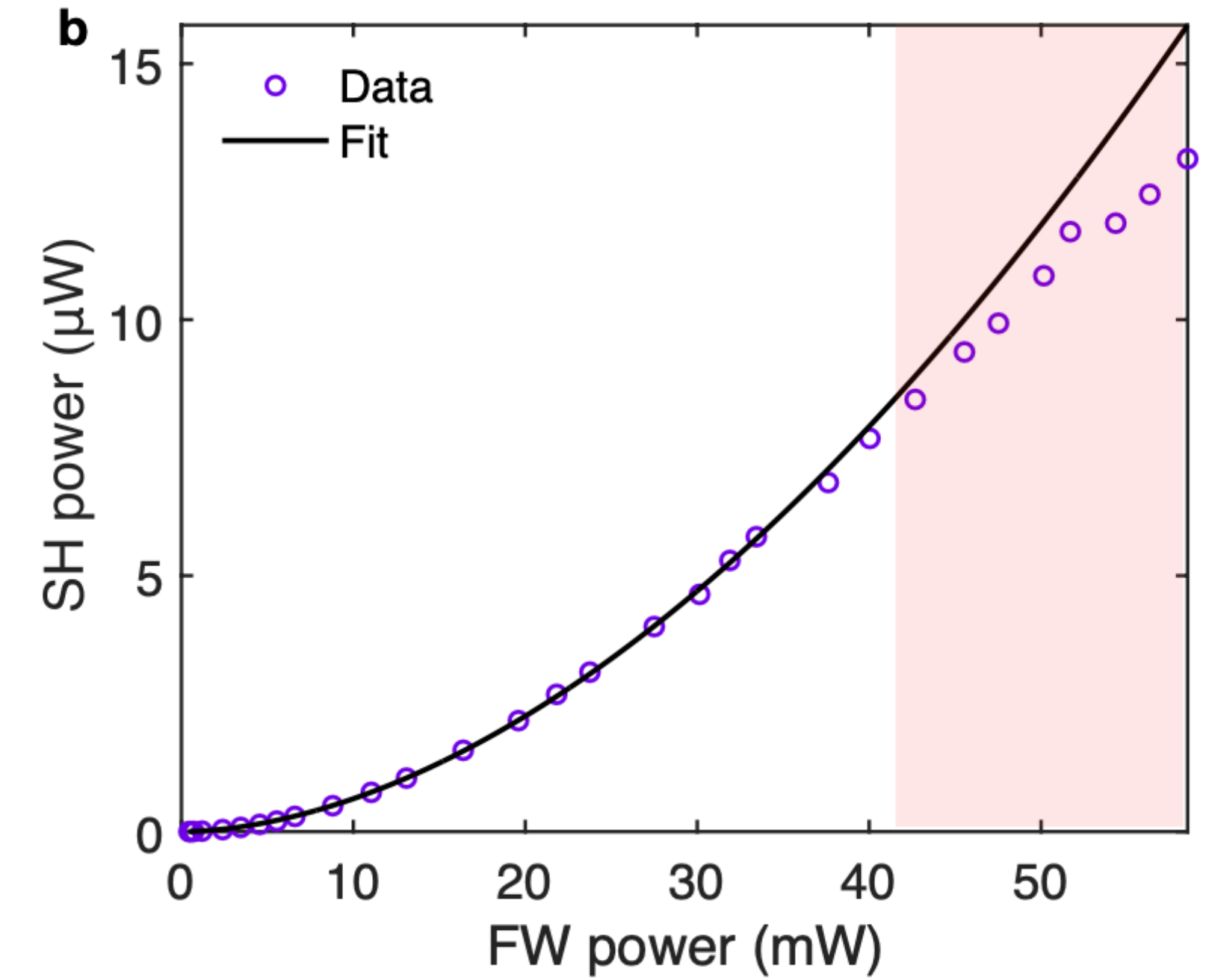
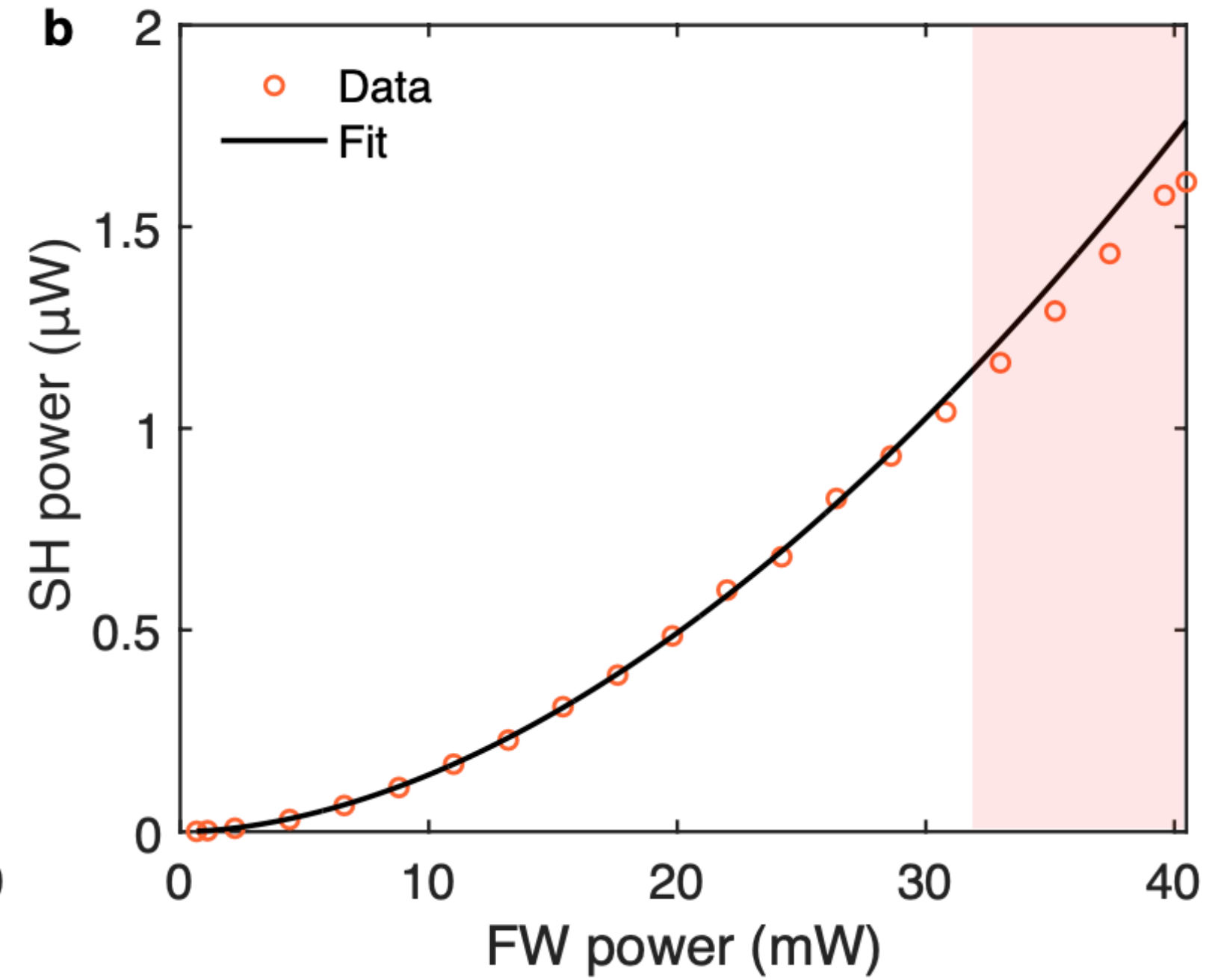
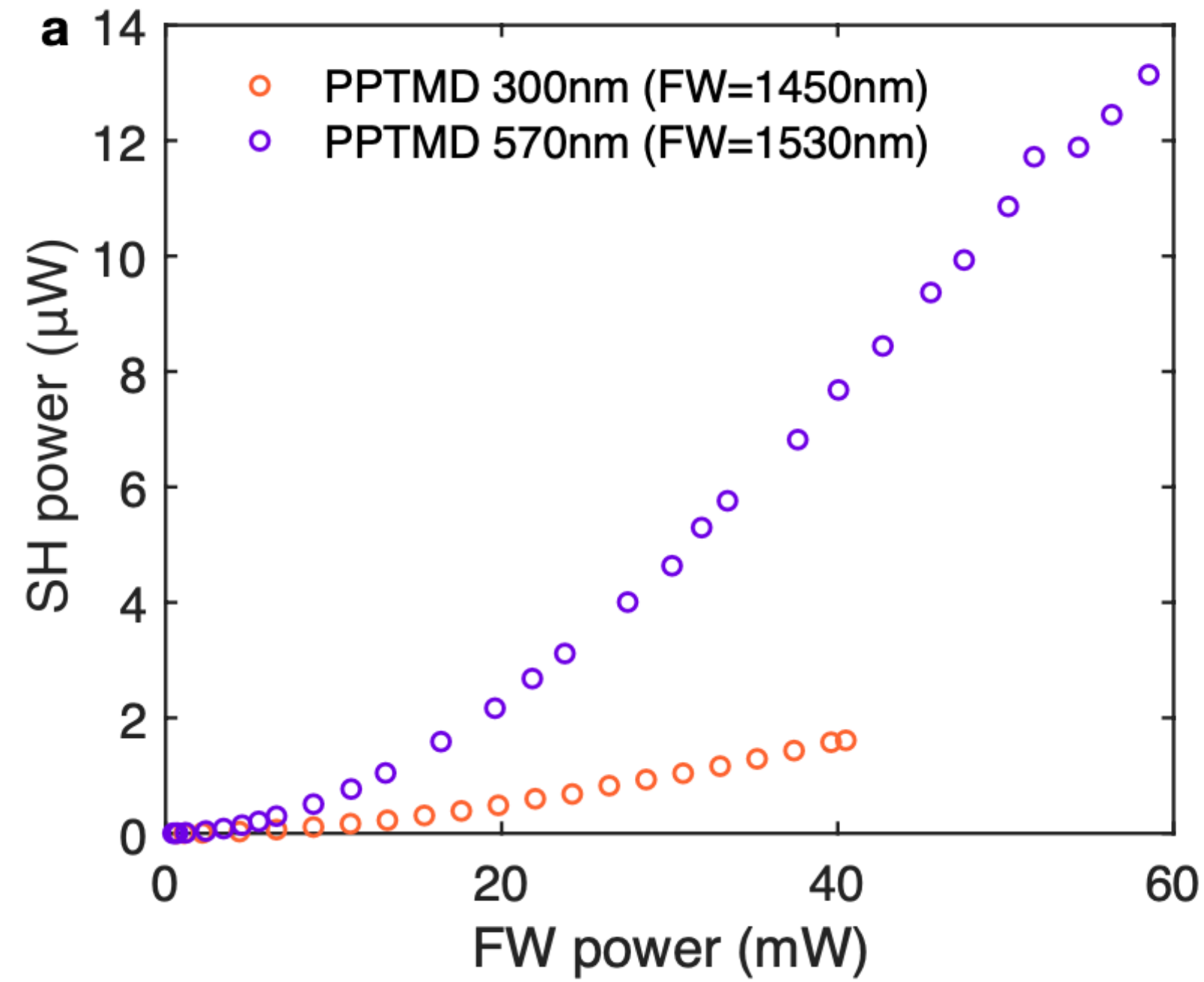
By tailoring the **thickness** of the 3R-MoS₂ slab, the TE₀ modes at ω and selected higher-order modes at 2ω can be **phase-matched** in a **waveguide geometry**



Bandwidth



SHG Power Dependence



Transition metal dichalcogenides (TMDs)

Weak out-of-plane van der Waals forces
Strong in-plane covalent bonds

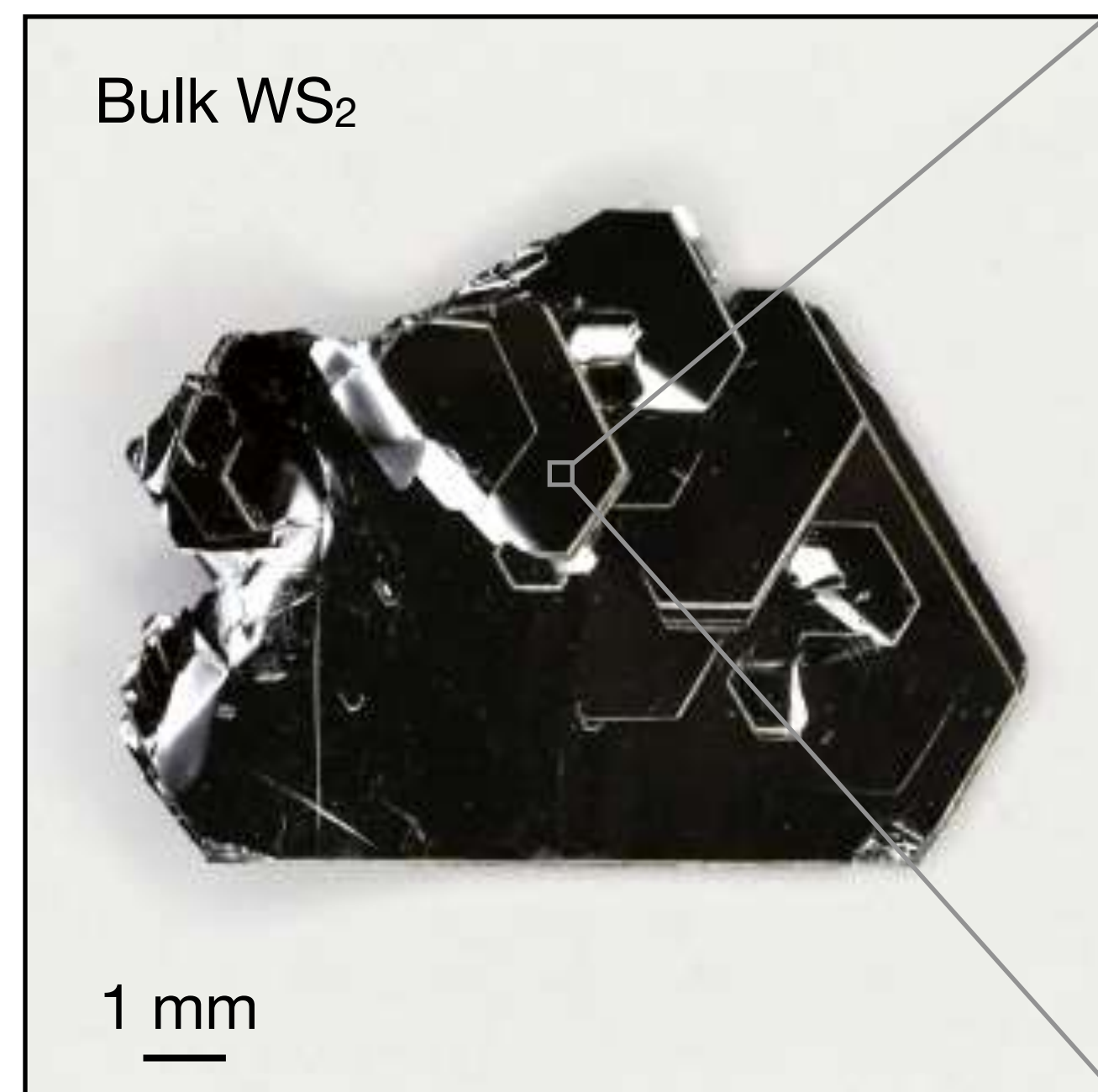
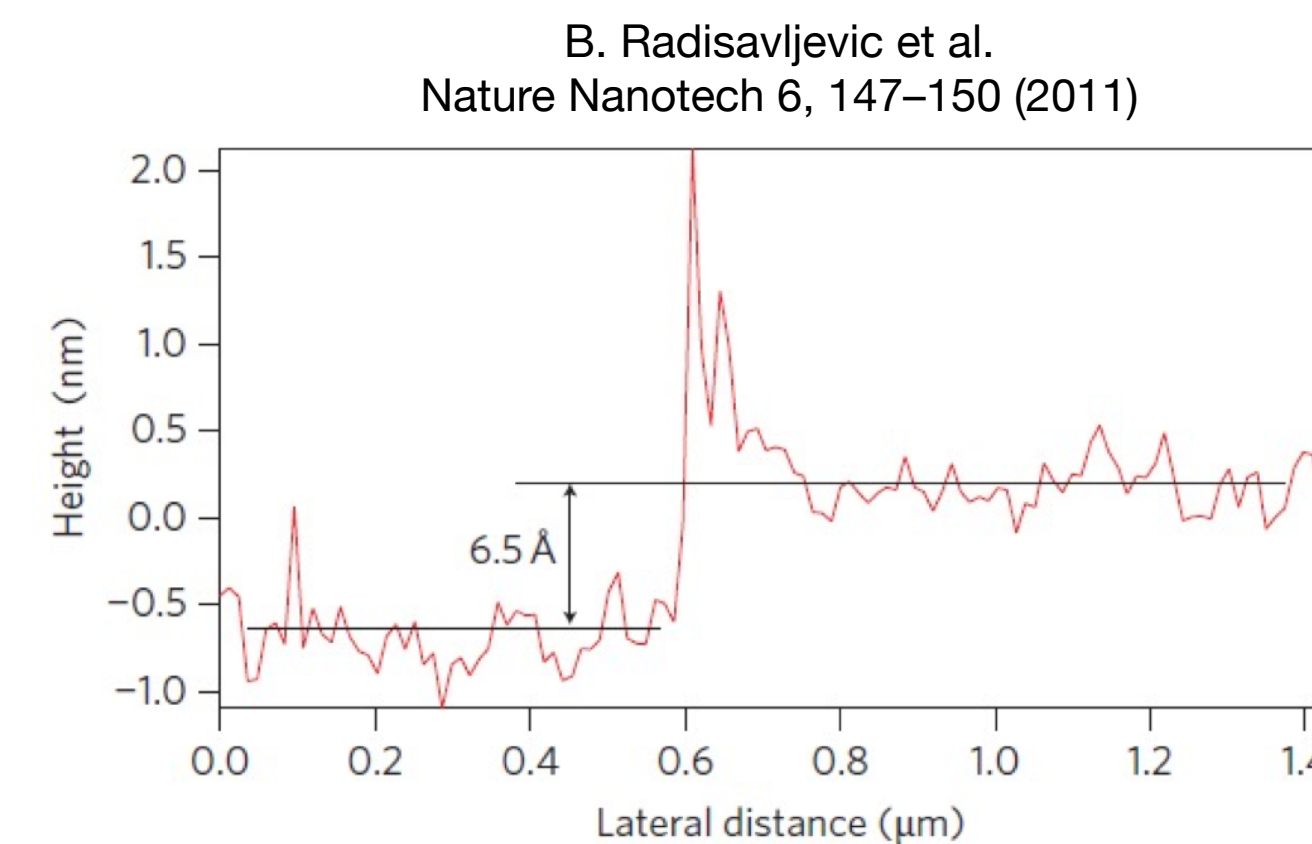
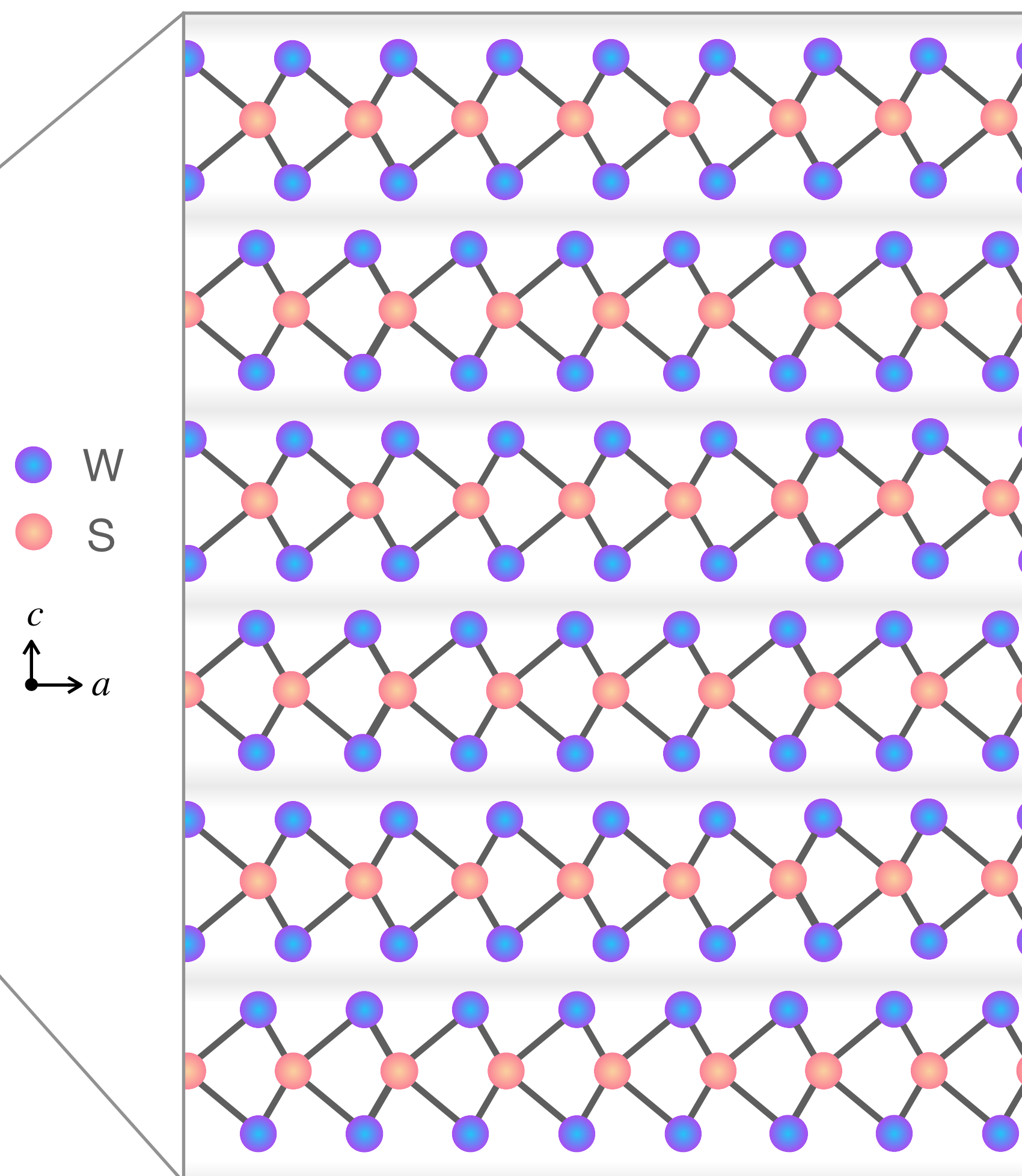


Photo credits: HQ Graphene



Bulk



Monolayer

