Quantum Light Sources

using
nanocavities and molecular vibrations

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Introduction: single photons and photon pairs

Single photon sources based on quantum emitters

Ex.: Quandela QD source

- **Improve some parameters**
  (wavelength, tunability, scalability, photon rate, etc.)

- **A bridge between material science, chemistry and quantum optics**

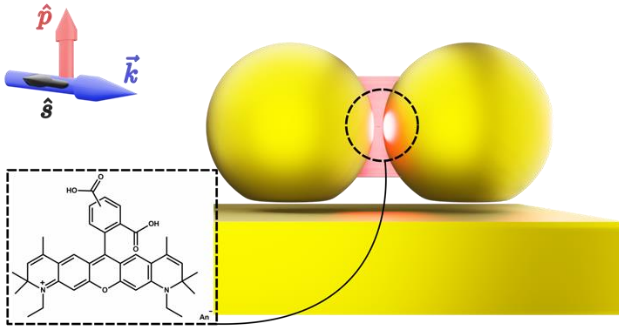
Photon pair sources based on nonlinear crystals

- Parametric down-conversion $\chi^{(2)}$
- Four-wave mixing $\chi^{(3)}$

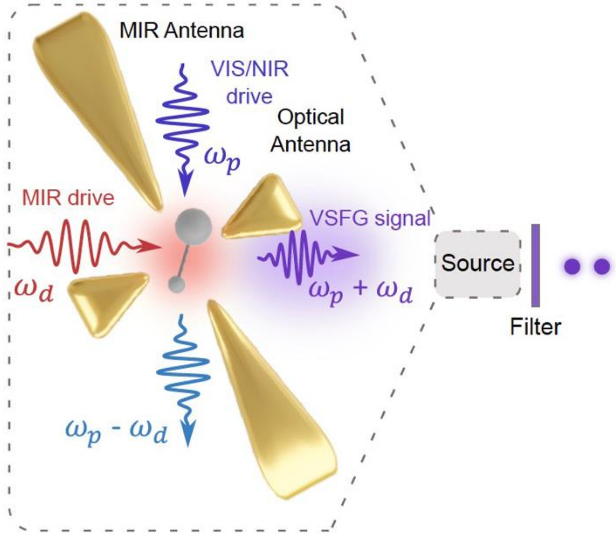
EPFL

I. A fluorophore in a nanocavity:

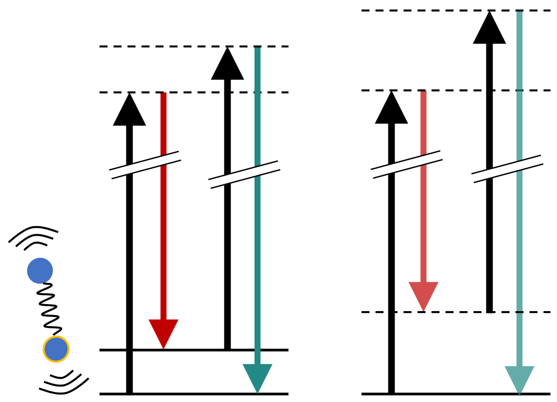
Giant Purcell factors and Lamb shifts



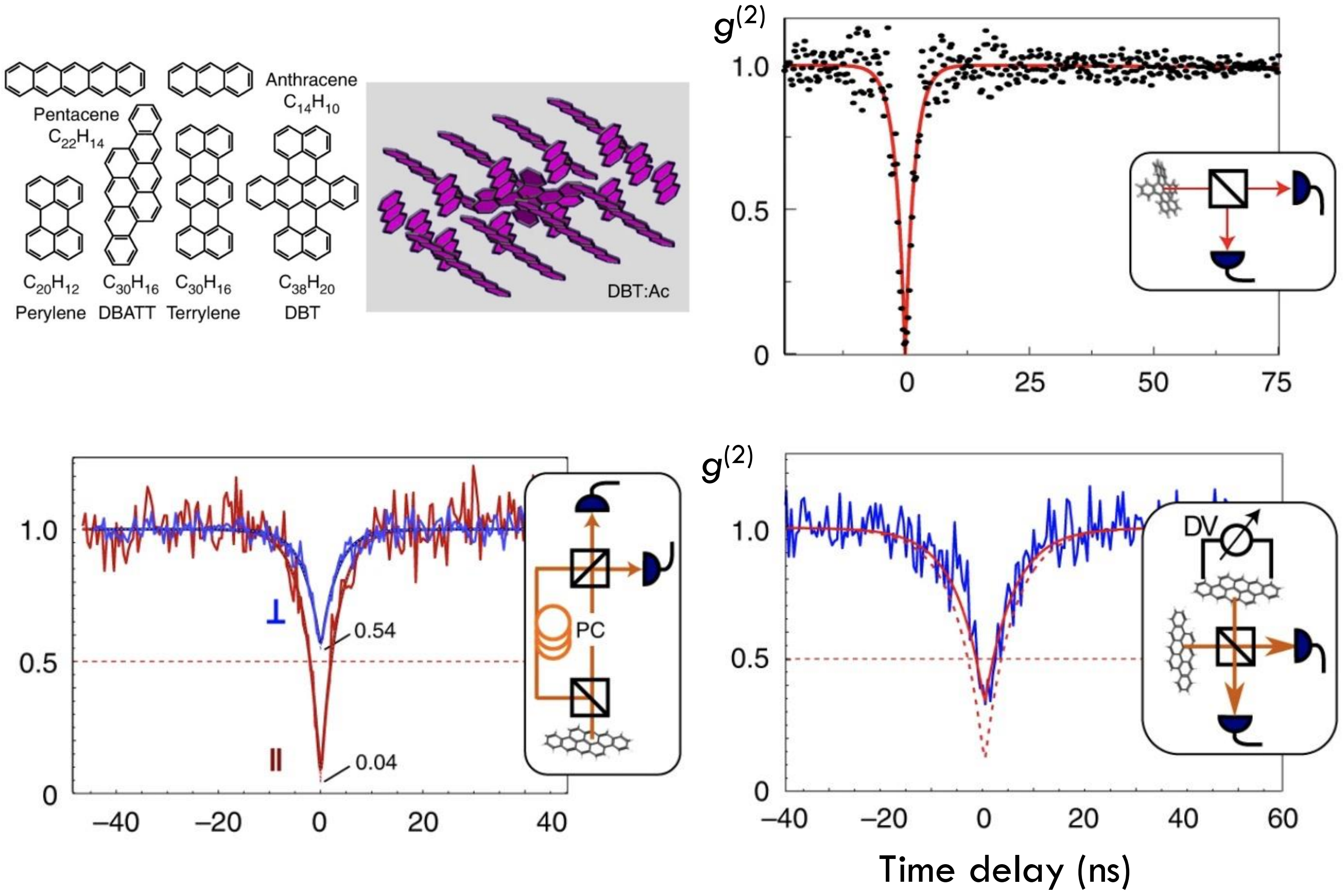
II. Single-molecule vibrational blockade



III. Polarisation entanglement from interfering four-wave mixing pathways



Single-molecule quantum emitters

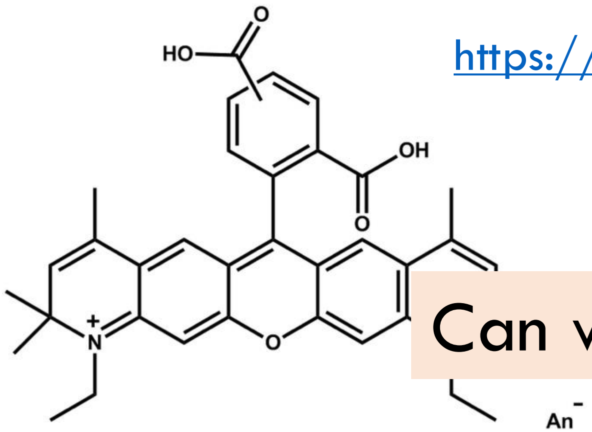


Hanbury-Brown & Twiss

Hong, Ou & Mandel

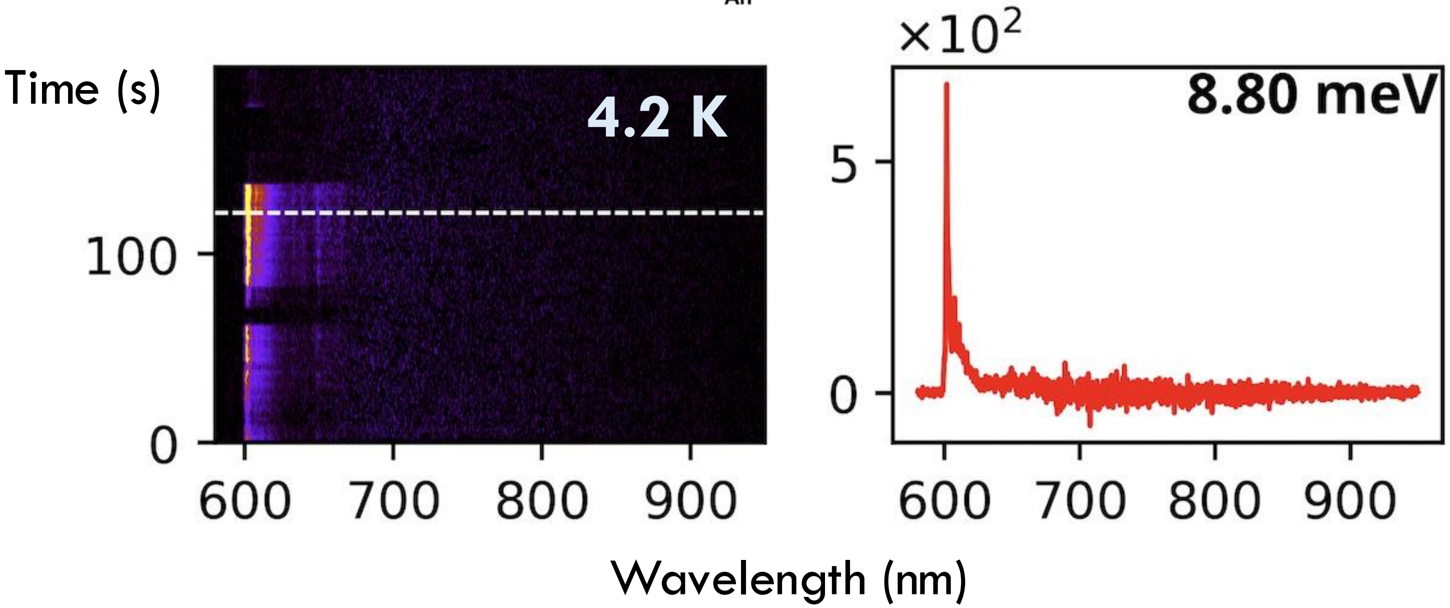
Toninelli et al. *Single organic molecules for photonic quantum technologies*. ***Nature Materials* 20,** 1615–1628 (2021)

A commercial fluorophore at 4 Kelvin



https://www.atto-tec.com/ATTO-590.html

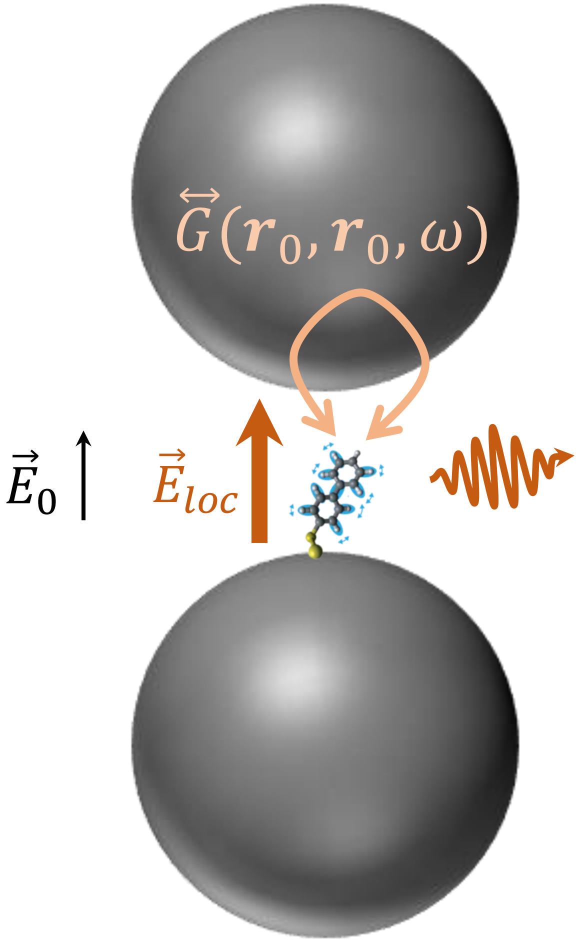
Can we reduce the excited-state lifetime by 5 x 10^4?



Far from lifetime-limited linewidth:

3 ns ⇔ 0.22 μeV

Light-Emitter interaction in a nanocavity



1. Enhancement of incident field ➢ **increased excitation rate**

2. Modification of the electromagnetic environment characterised by the dyadic Green's function $\overleftrightarrow{G}(\boldsymbol{r}_0, \boldsymbol{r}_0, \omega)$
   - ➢ **Purcell effect** $\propto \mathrm{Im}\{\overleftrightarrow{G}(\boldsymbol{r}_0, \boldsymbol{r}_0, \omega)\}$ (total LDOS)
   - ➢ **Lamb shift** $\propto \mathrm{Re}\{\overleftrightarrow{G}(\boldsymbol{r}_0, \boldsymbol{r}_0, \omega)\}$

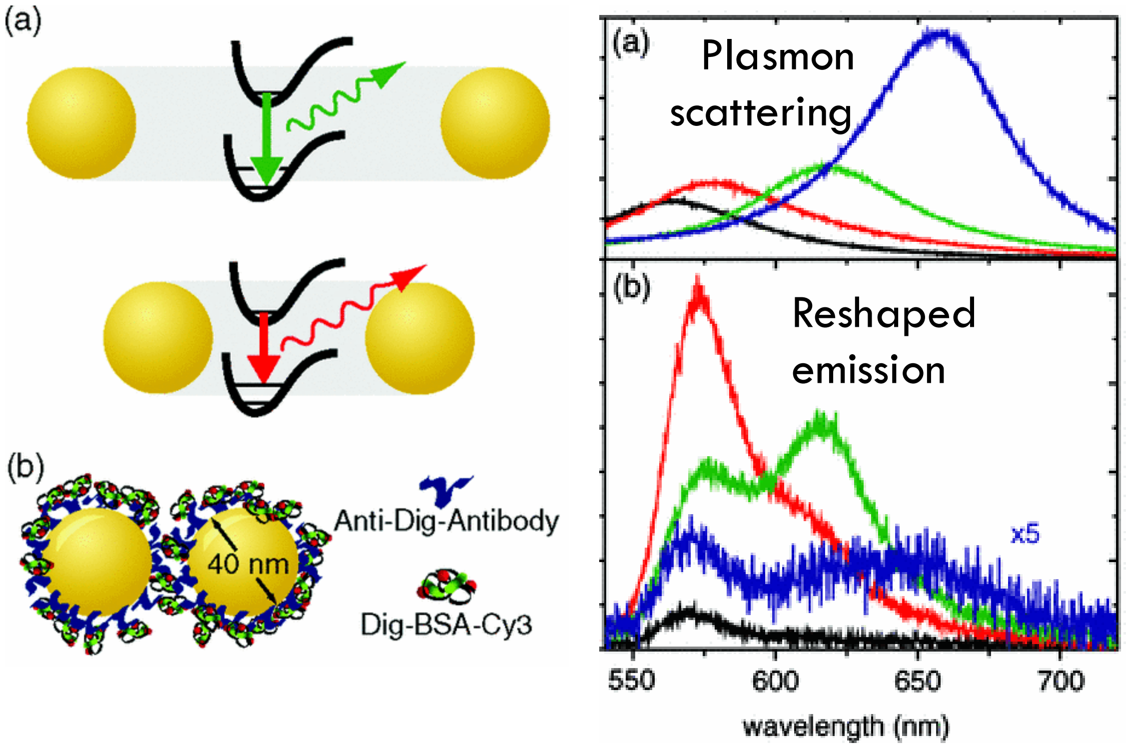
3. Reshaping (filtering) of far-field emission spectrum by

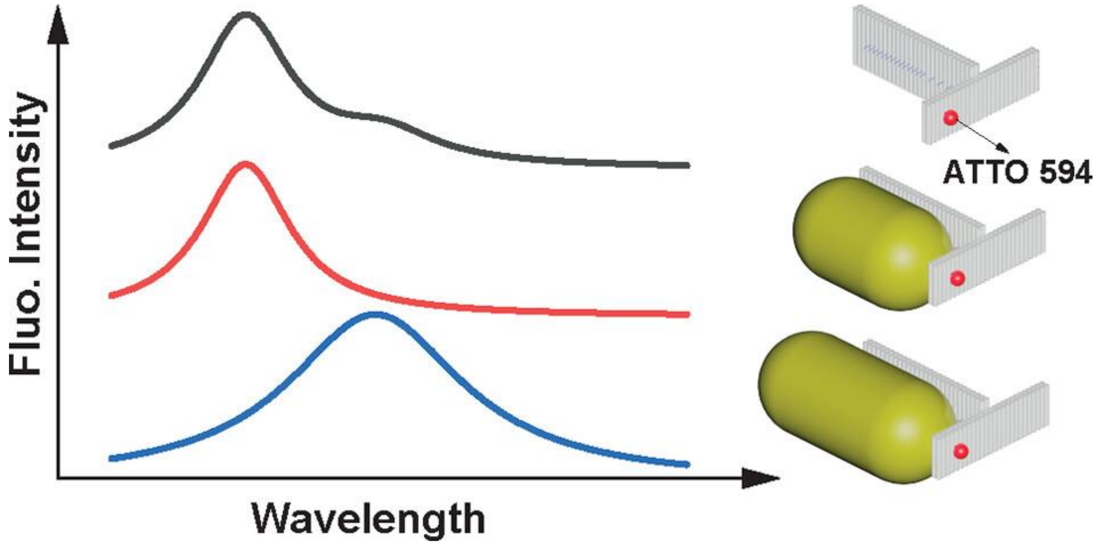
   $\gamma_{\mathrm{cav}}^{\mathrm{rad}}(\omega)$ (radiative LDOS)

📖 P. Yao *Phys. Rev. B* **80**, 195106 (2009)

Context: Reshaping fluorophore emission

DNA origami allows deterministic coupling of single fluorophores with plasmonic nanocavities

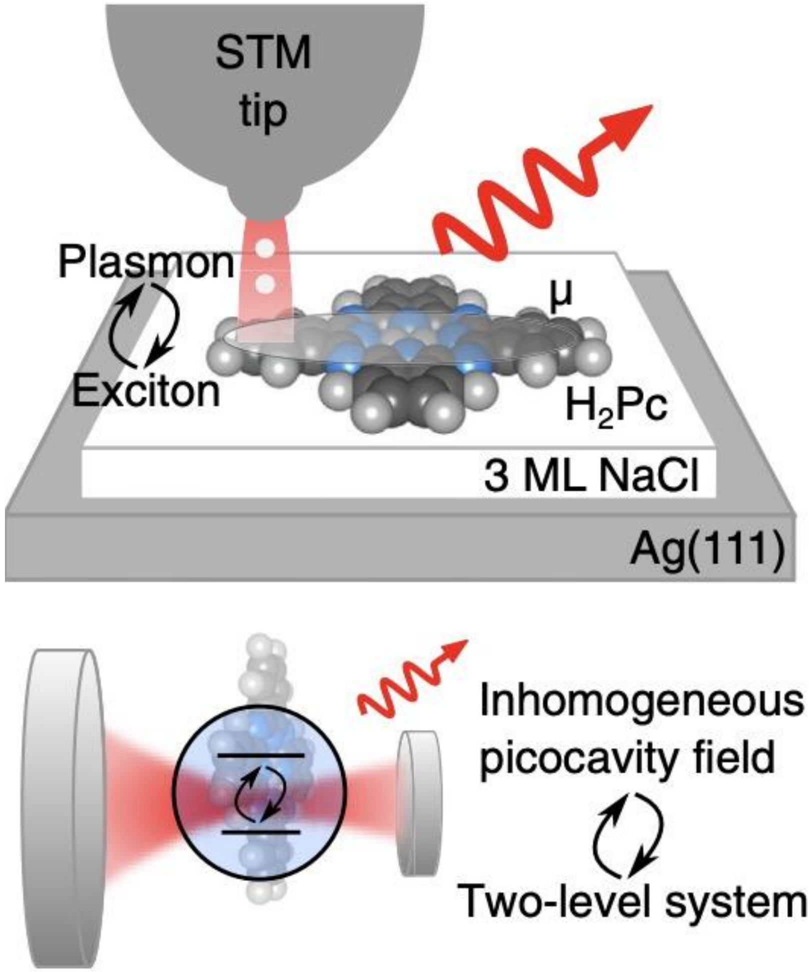


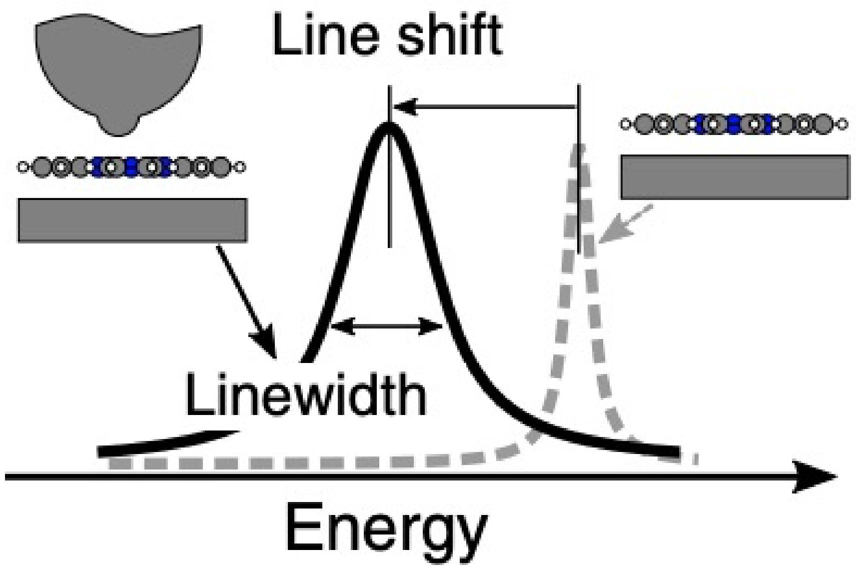


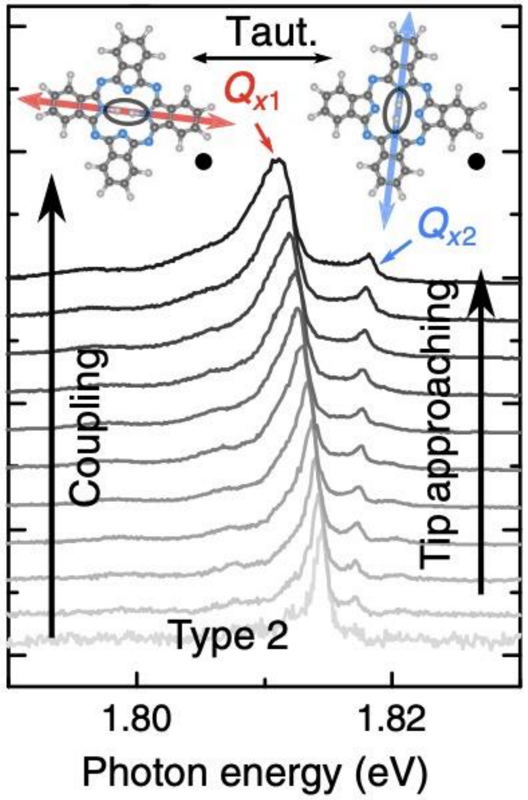
M. Ringler *Phys. Rev. Lett.* **100**, 203002 (2008)

M. Sanz-Paz *Nano Lett.* **23**, 6202–6208 (2023)

Context: Lamb and Purcell on a molecule



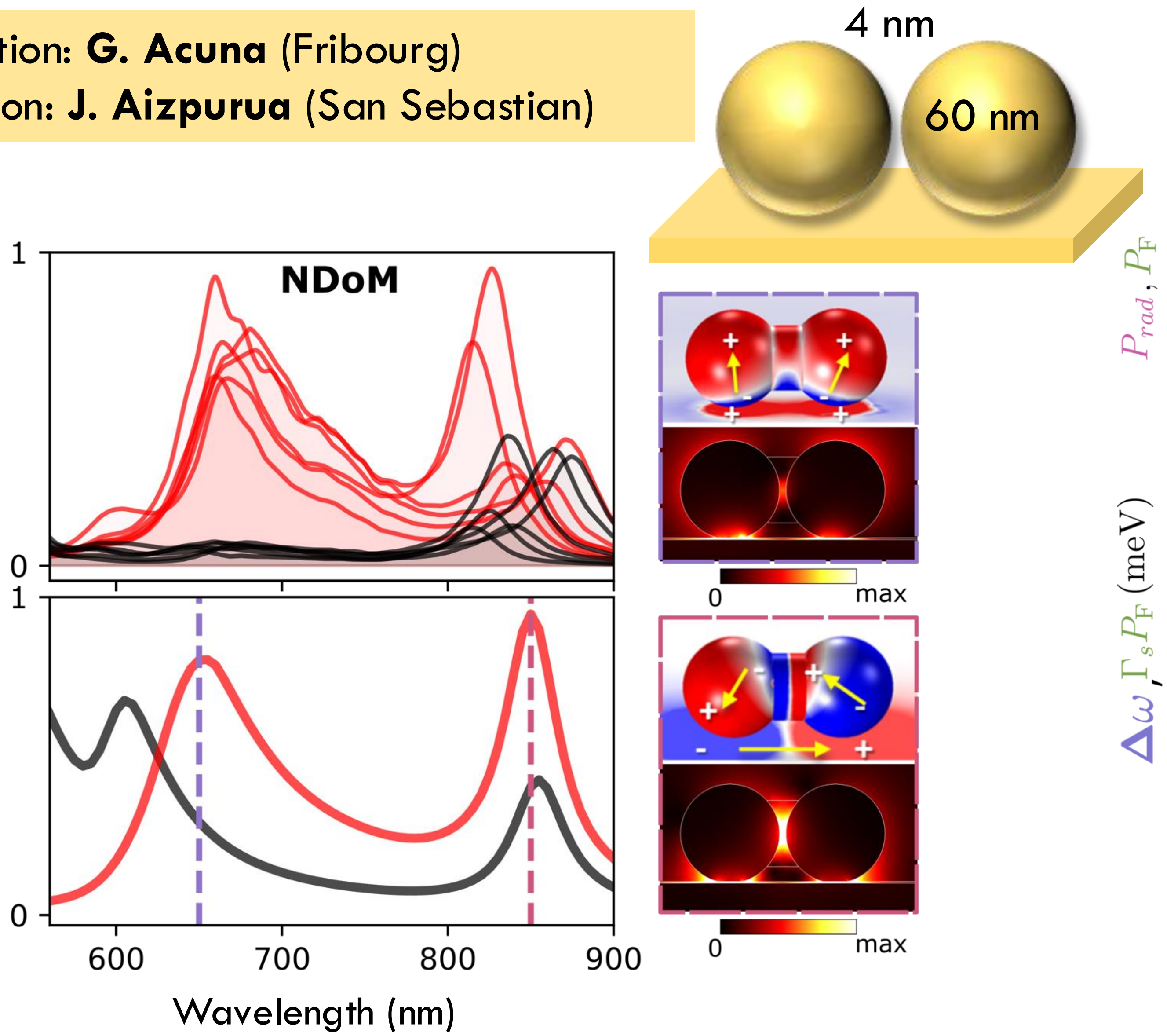


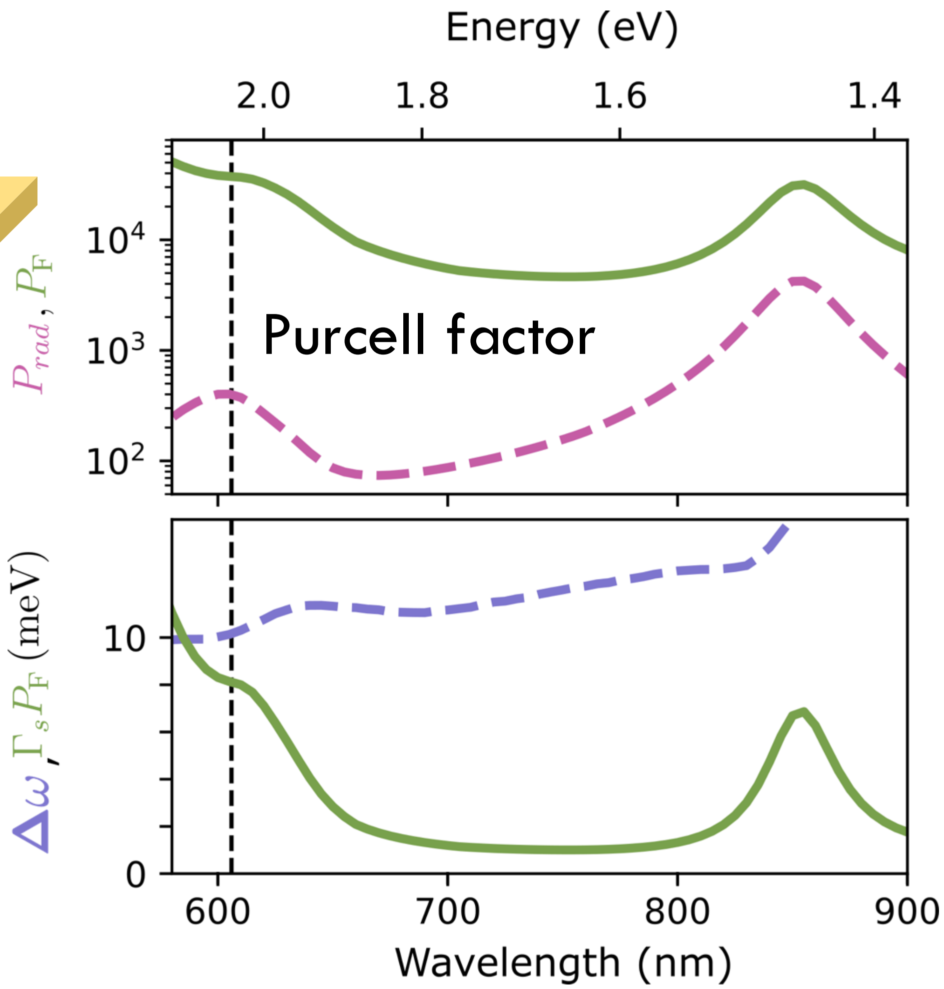


A. Rosławska *Phys. Rev. X* **12**, 011012 (2022)

Nanodimer on Mirror (NDoM)

Fabrication: **G. Acuna** (Fribourg)
Simulation: **J. Aizpurua** (San Sebastian)



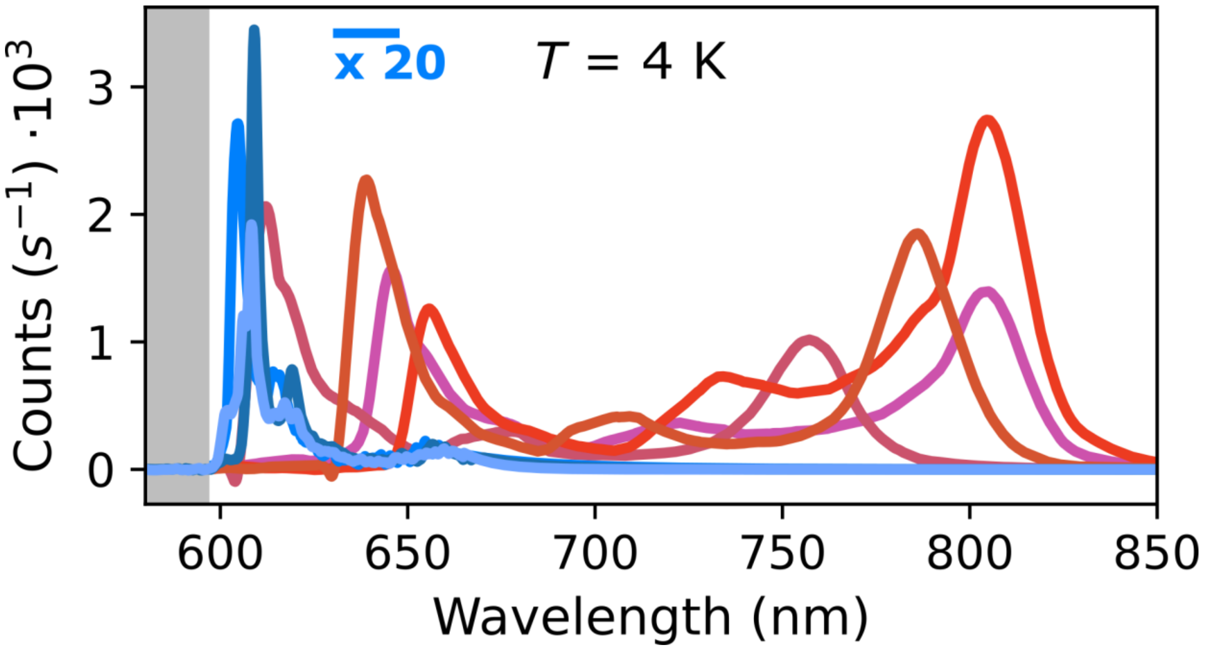


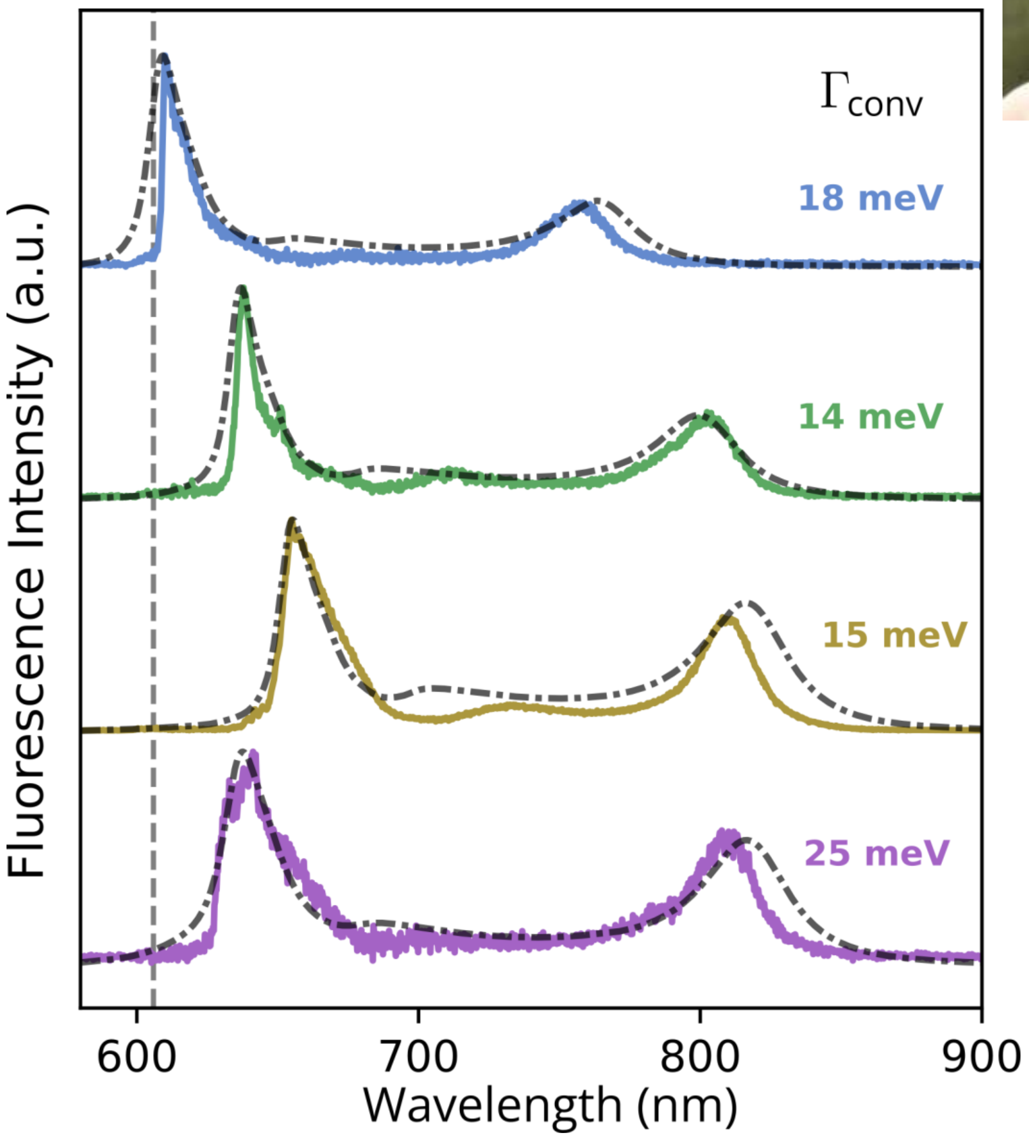
$\Gamma_s P_F$ → Purcell broadening
$\Delta\omega$ → Lamb Shift

S. Verlekar, M Sanz-Paz, M Zapata-Herrera *et al.* ***ACS Nano*** (2025)

Complete reshaping at $T = 4$ K

Sachin Verlekar

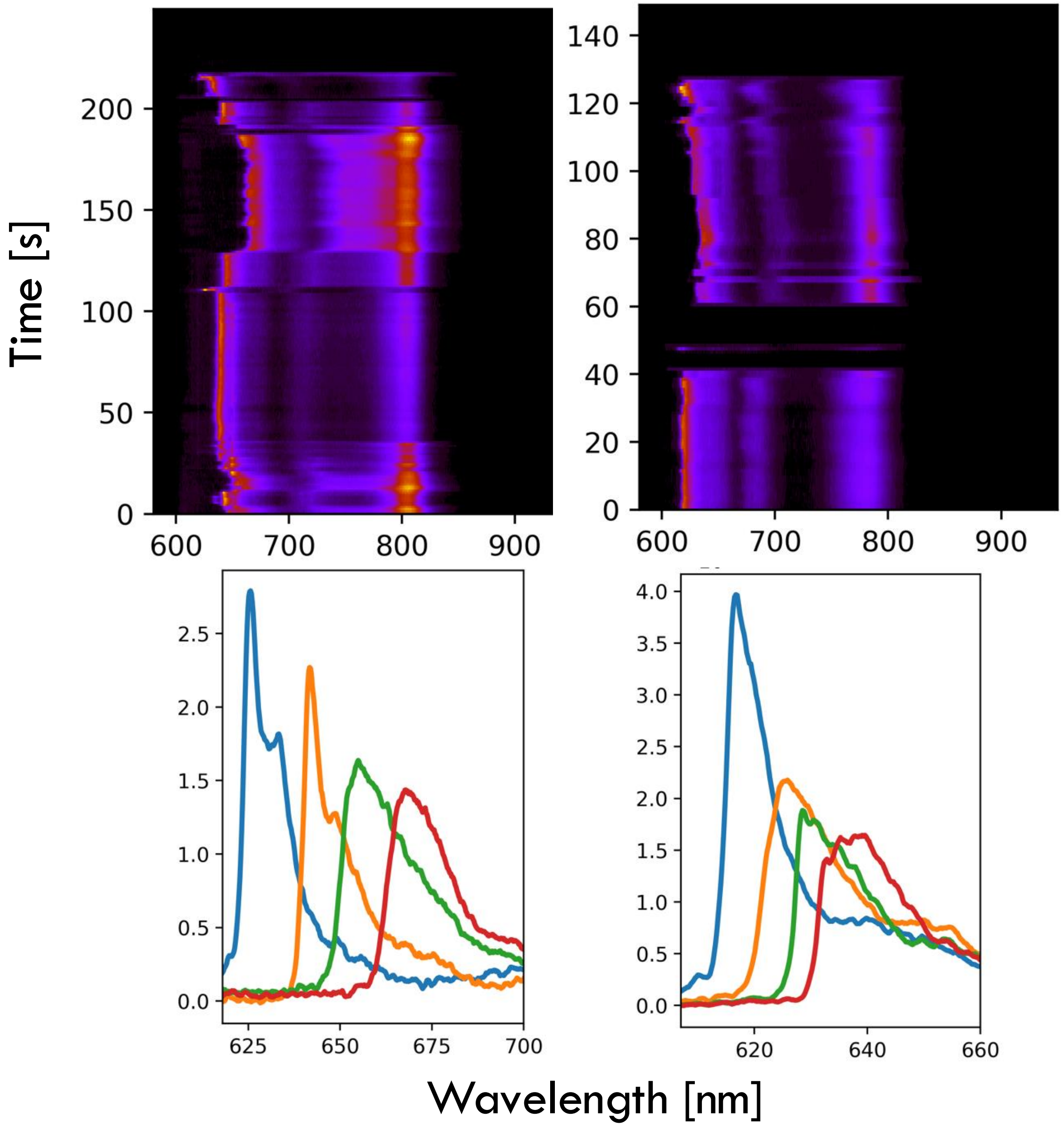




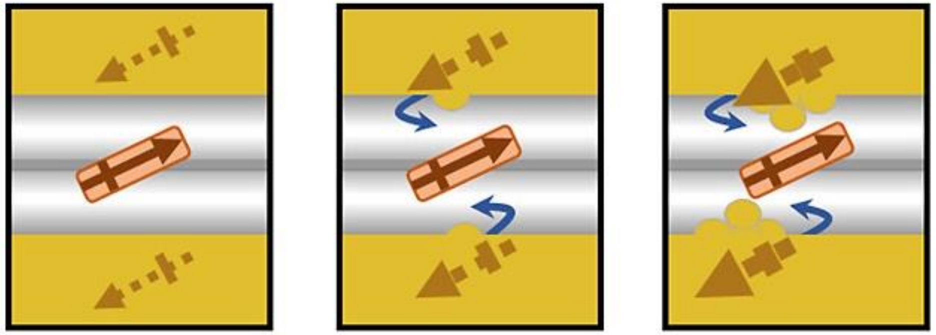
- Average broadening: 20 meV ($P_F \sim 10^5$)
- Corresponding lifetime: 30 fs

S. Verlekar, M Sanz-Paz, M Zapata-Herrera *et al.* ***ACS Nano*** (2025)

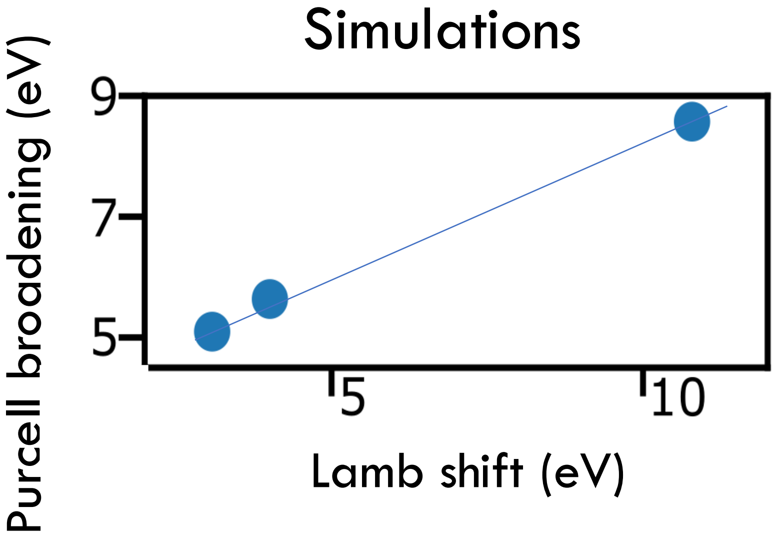
Fluctuating Purcell factor and Lamb shift



Possible mechanism: picocavities



S. Rochetti *Nano Lett.* **23**, 5959 (2023)



Conclusions

- We leverage the **giant Purcell effect and Lamb shift in nanocavities** to tailor the emission spectrum toward the infrared
- Results are compatible with a **lifetime-limited emission linewidth**
- Indistinguishable photons?
- Violations of Kasha's rule?

EPFL

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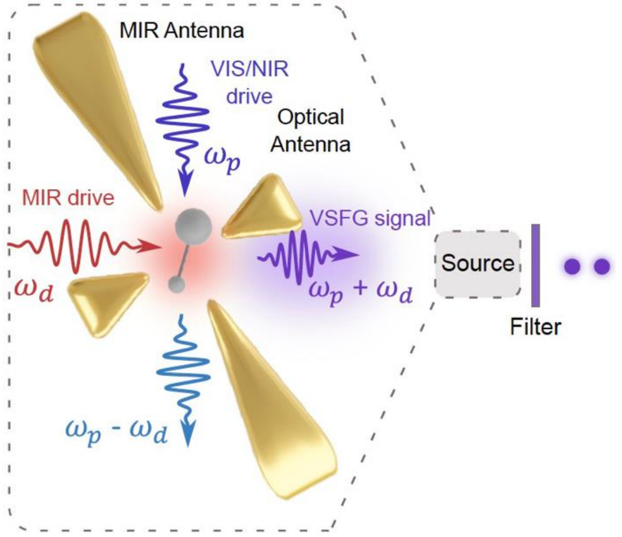
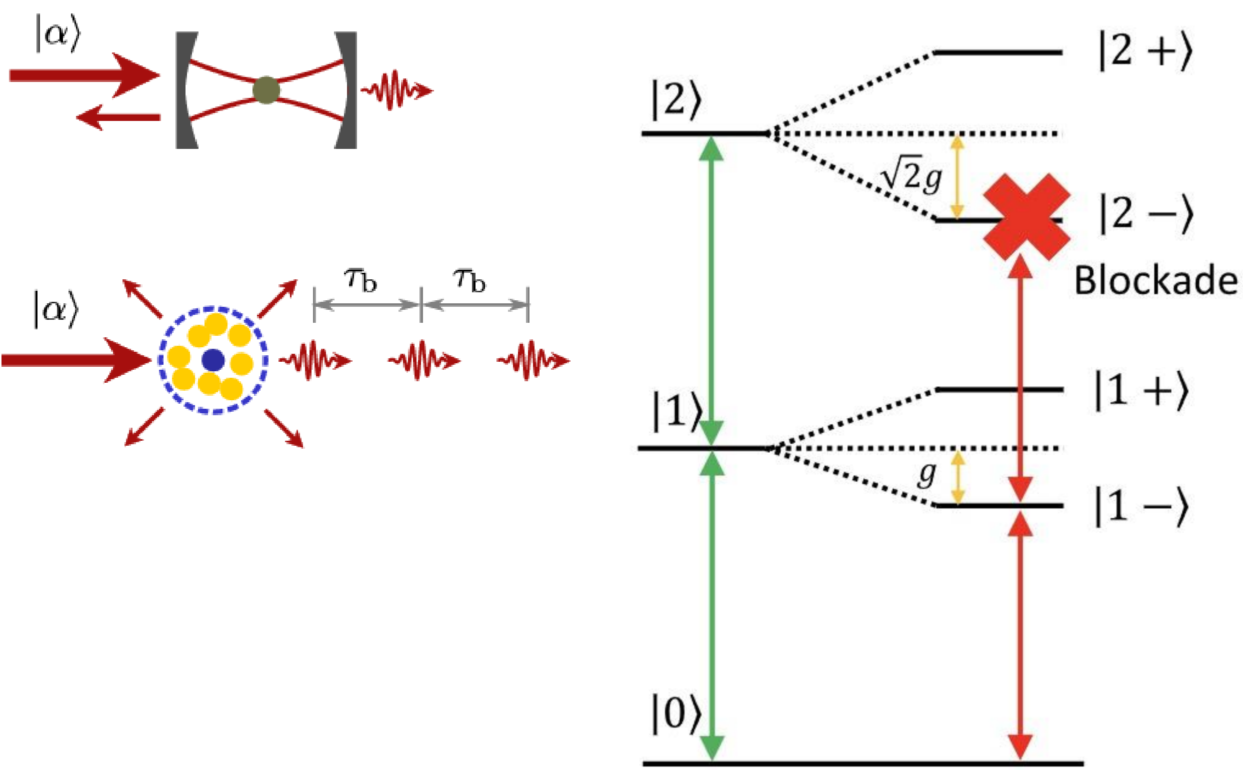


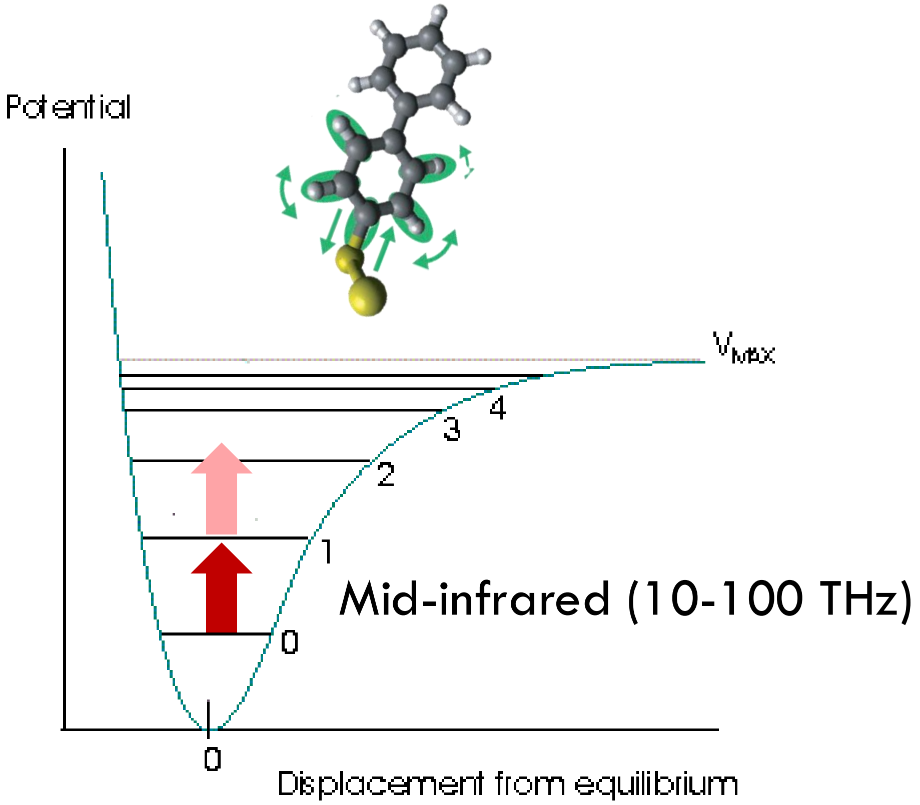
III. Polarisation entanglement from interfering four-wave mixing pathways

Photon blockade: concept

Conventional photon blockade



Molecular vibration: anharmonicity

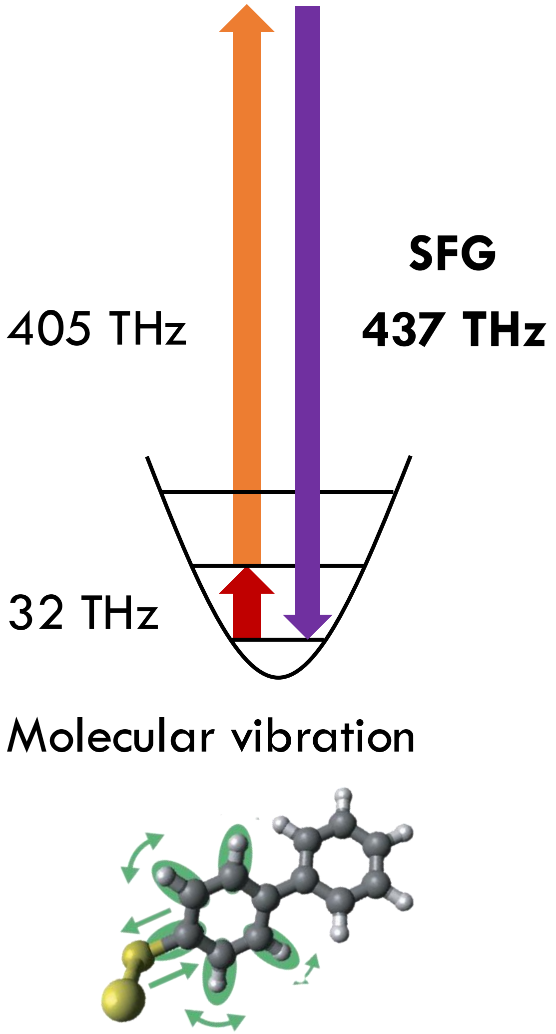


H. J. Snijders *Phys. Rev. Lett.* **121**, 043601 (2018)

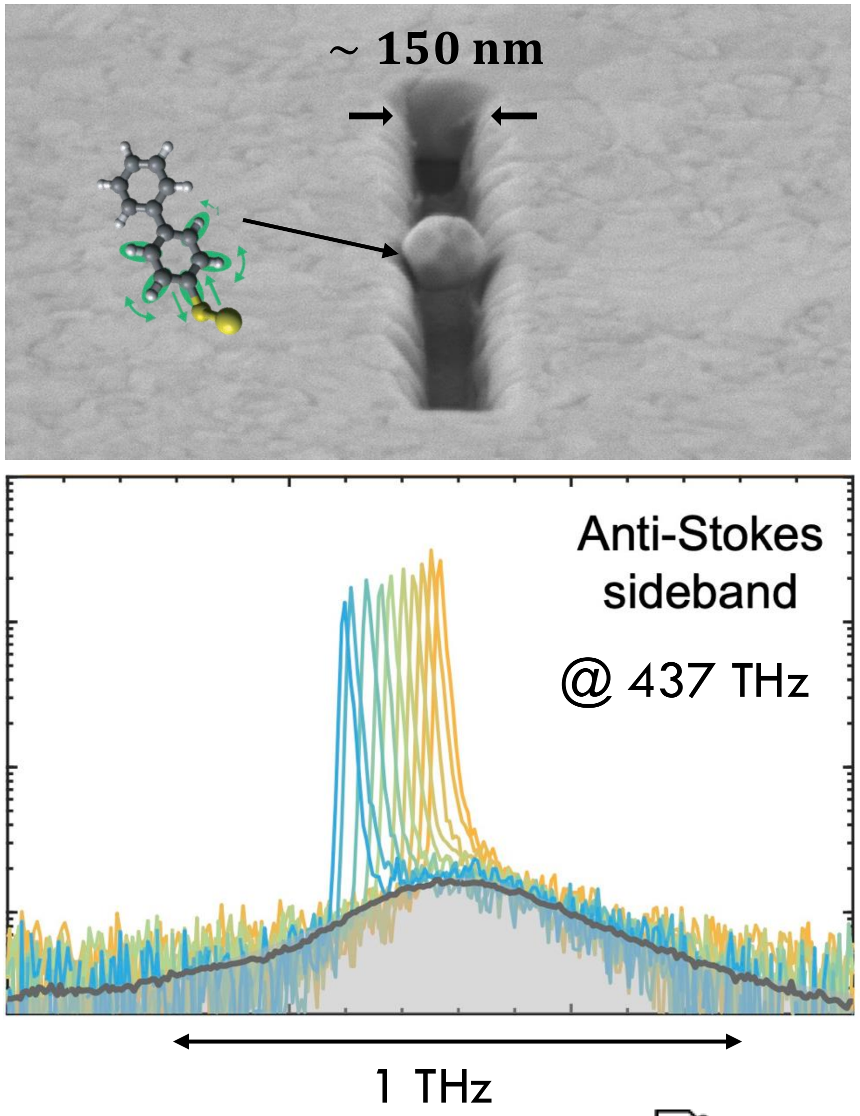
J.D. Pritchard *Annual Review of Cold Atoms and Molecules* 301-350 (2013)

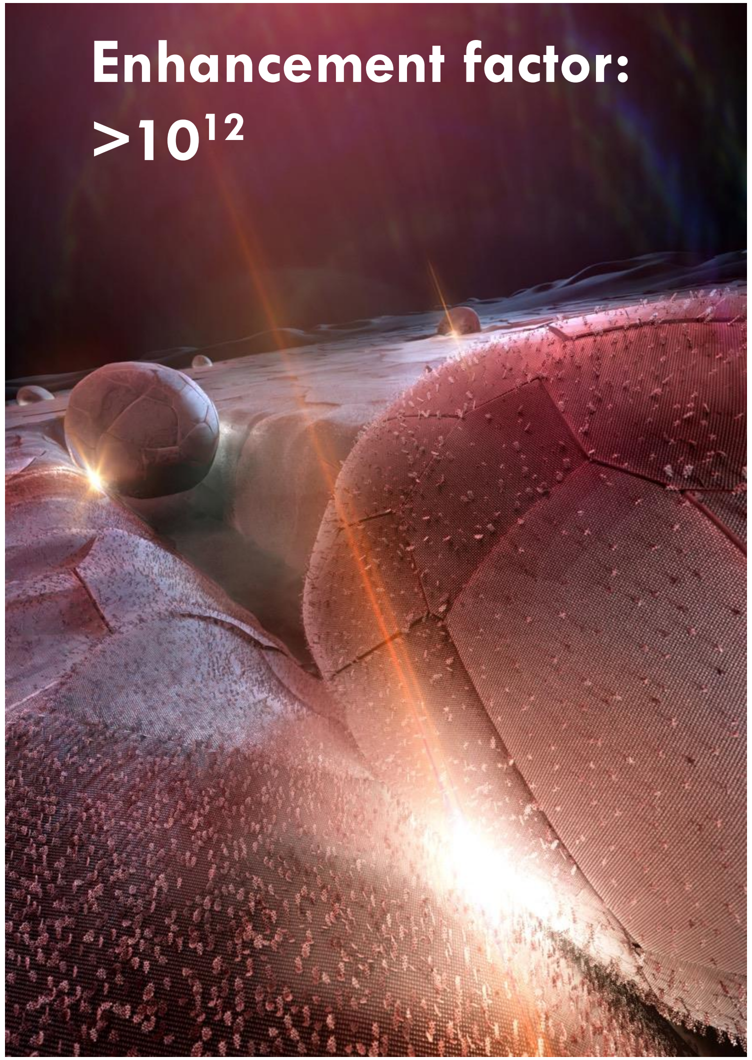
E. Zubizarreta Casalengua *Laser & Photonics Reviews* **14**, 1900279 (2020)

Vibrational sum-frequency generation (SFG)



"Nanoparticle-in-slit"



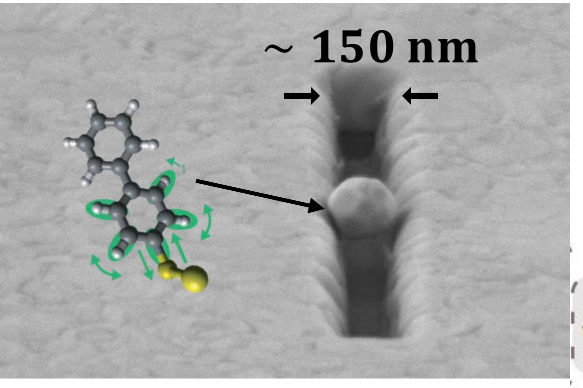


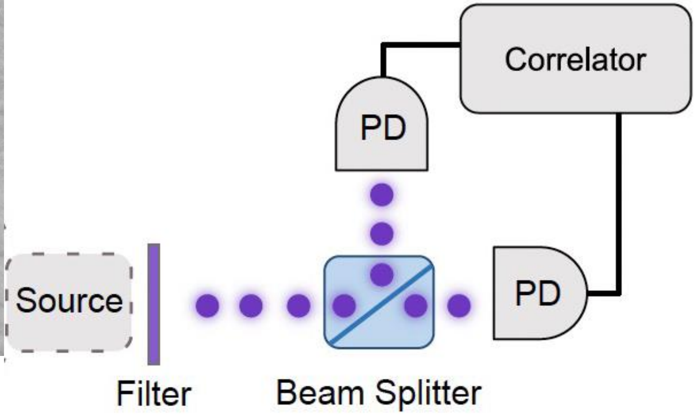
W. Chen et al. ***Science*** 374, 1264-1267 (2021)

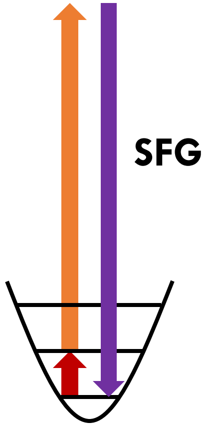
Single-molecule SFG: anharmonicity

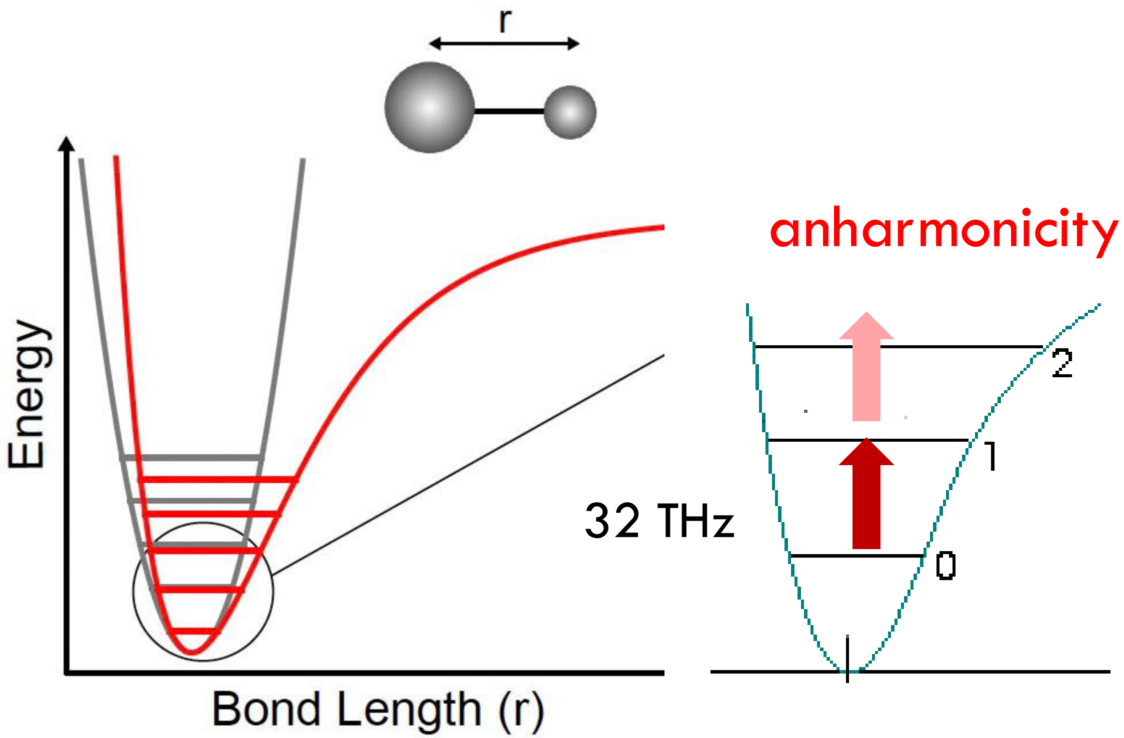
Fatemeh Moradi Kalarde

Collaboration : Johannes Feist & Carlos Sanchez Munos, Madrid





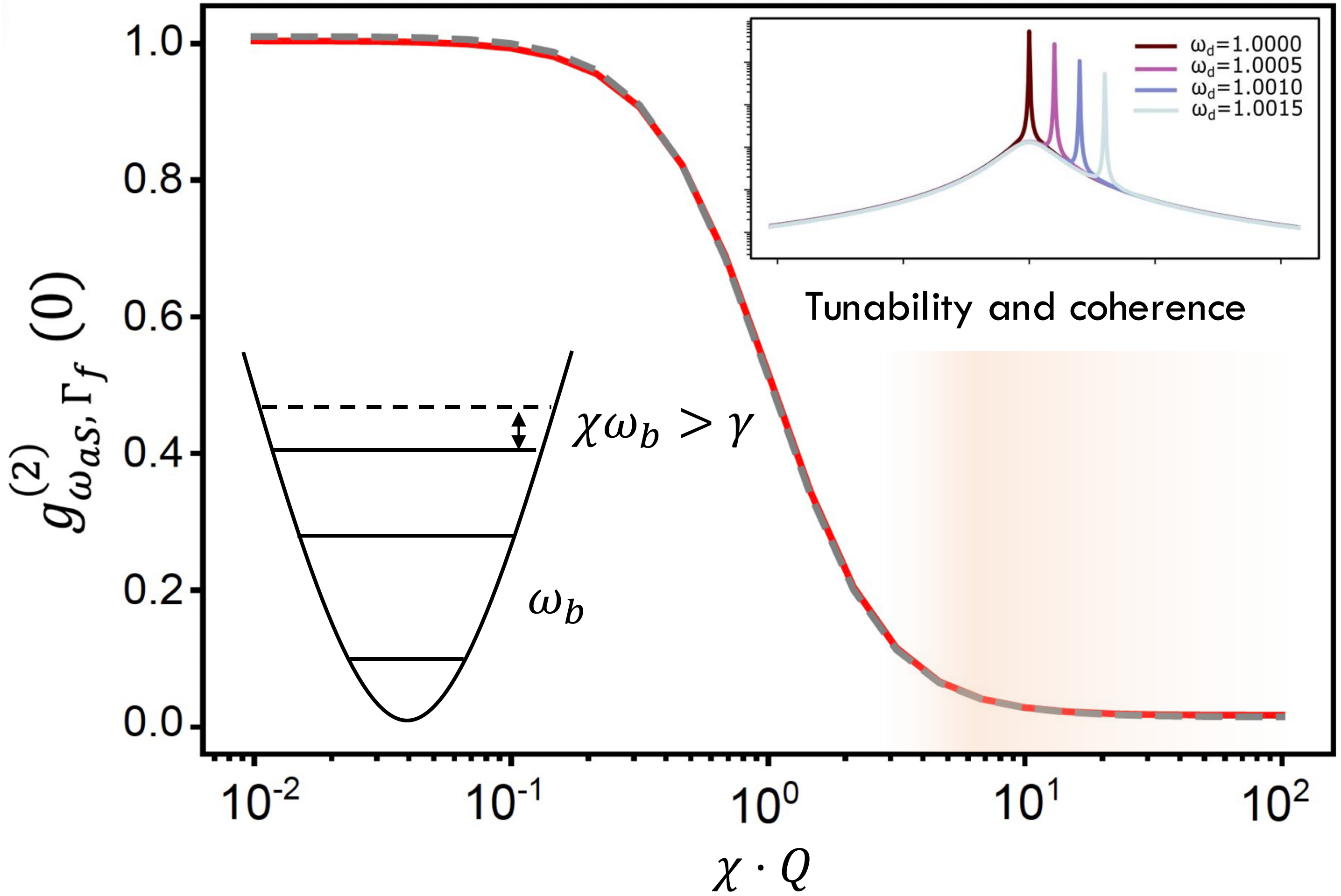




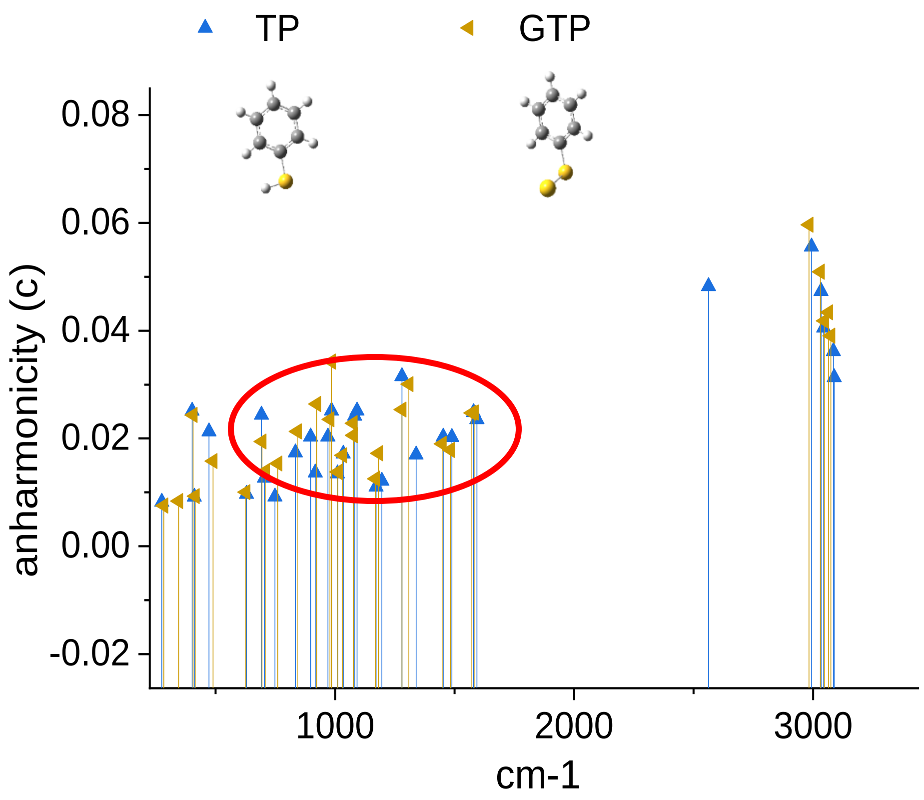
F. Moradi Kalarde *et al.* ***Nanophotonics***; 14: 59–73 (2025)

Single-molecule SFG: antibunching

Upconverted photon antibunching under MIR drive



Anharmonicity computed by DFT



Typical values for small organic molecules:

$$Q \sim 200;\ \ \chi \simeq 2 \times 10^{-2}\ \rightarrow \chi \cdot Q \sim 4$$

Christophe Galland, Swiss Quantum Days 2025

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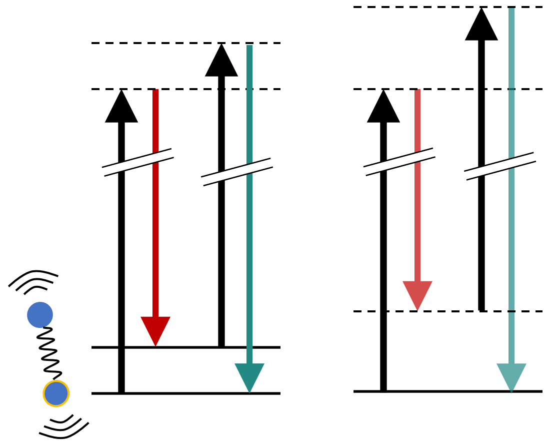
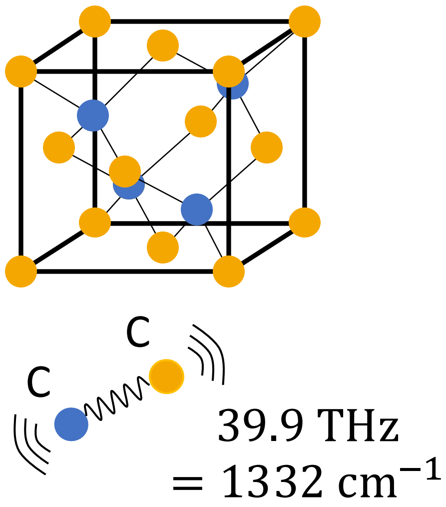
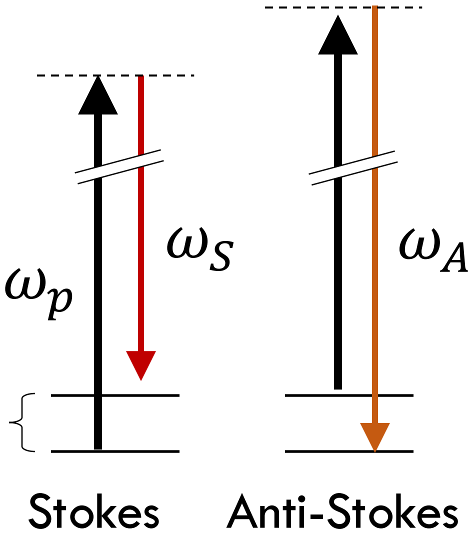


Raman scattering & Four-wave mixing in diamond

Spontaneous Raman scattering

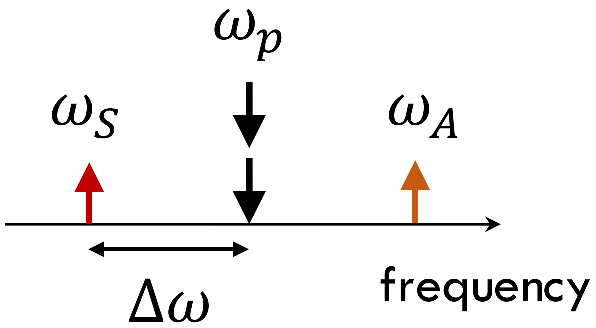
2 carbon atoms per unit cell

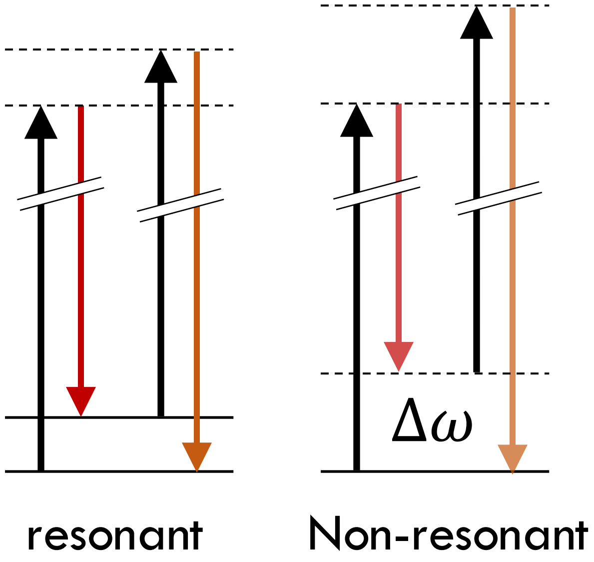




Four-wave mixing

$\chi^{(3)}$ processes
for two incoming photons at ω_p



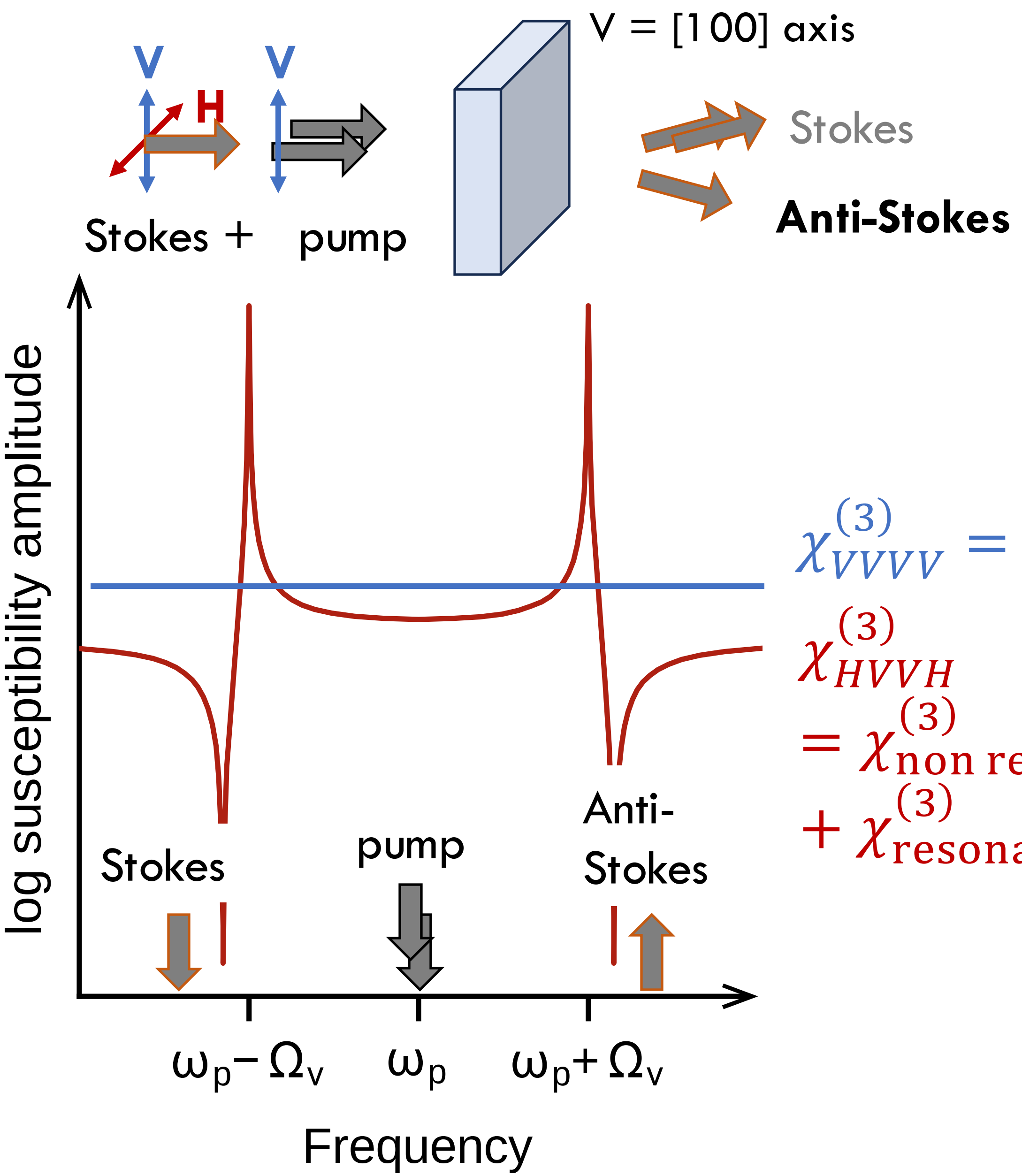


Interfering pathways:

$$\chi^{(3)}_{\text{non res.}} + \chi^{(3)}_{\text{resonant}}(\Delta\omega)$$

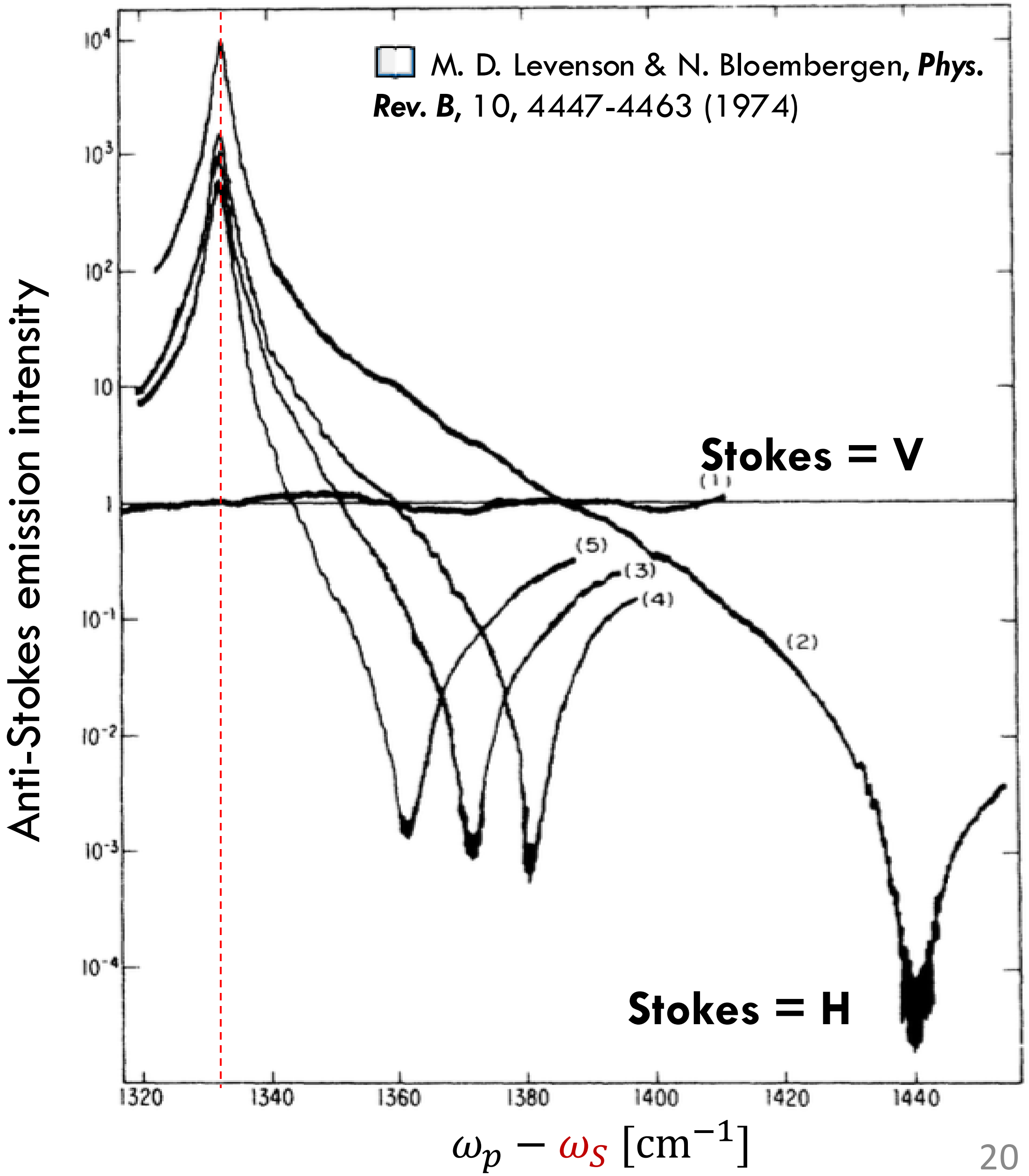
T. A. Freitag *Phys. Rev. A* **108**, L051501 (2023)

Coherent anti-Stokes Raman spectroscopy (CARS)

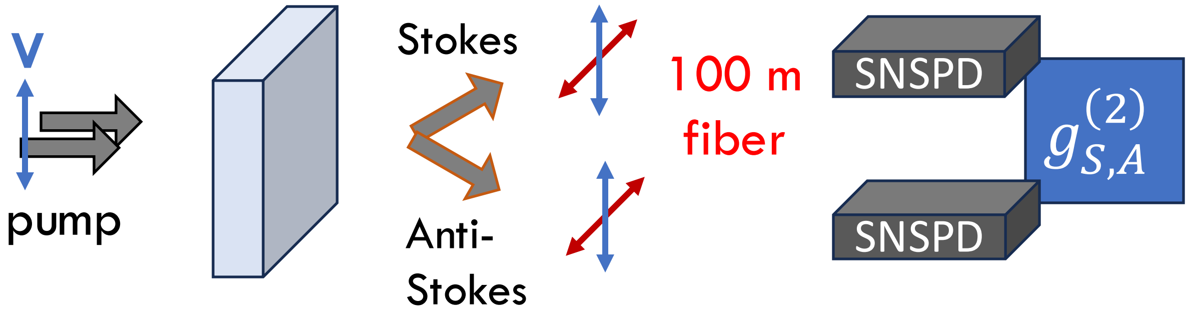


$\chi^{(3)}_{VVVV} = \chi^{(3)}_{\text{non res.}}$

$\chi^{(3)}_{HVVH} = \chi^{(3)}_{\text{non res.}} + \chi^{(3)}_{\text{resonant}}(\Delta\omega)$



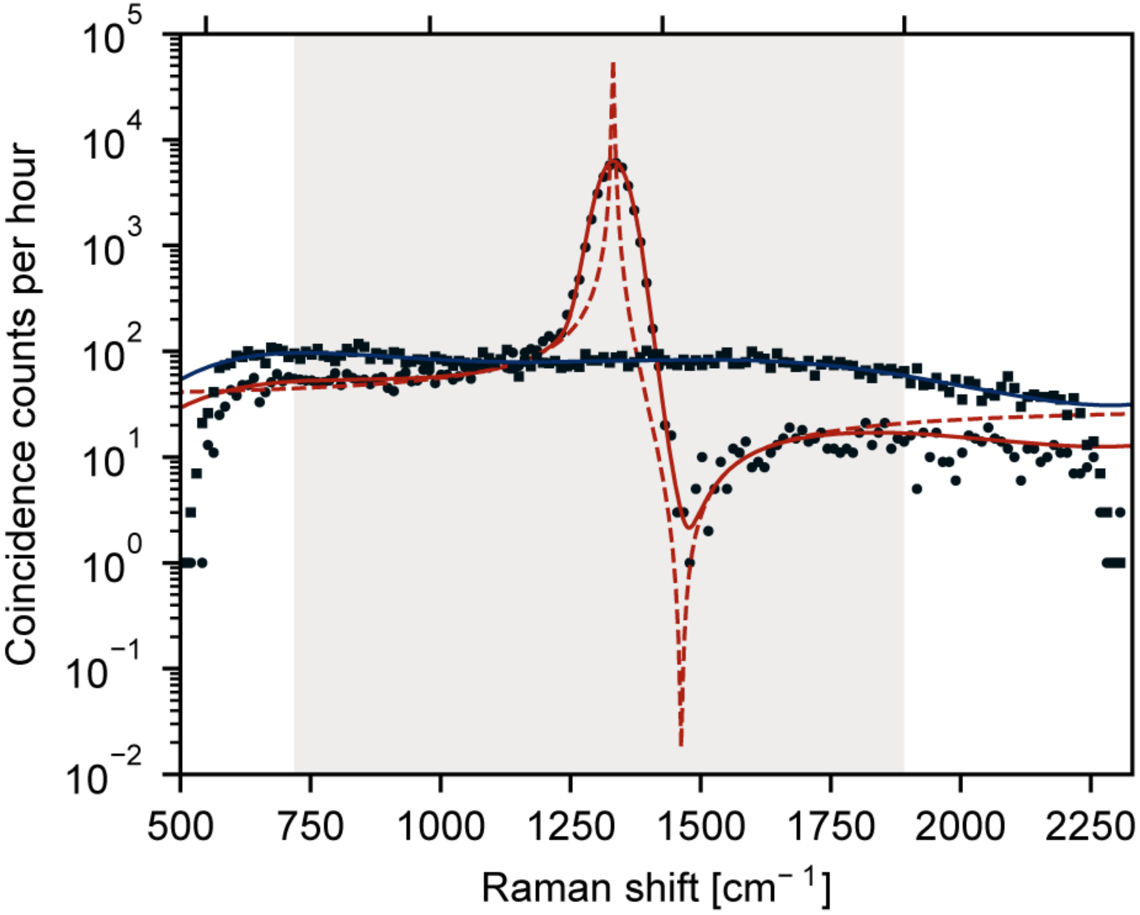
Measurement-induced coherent spectroscopy

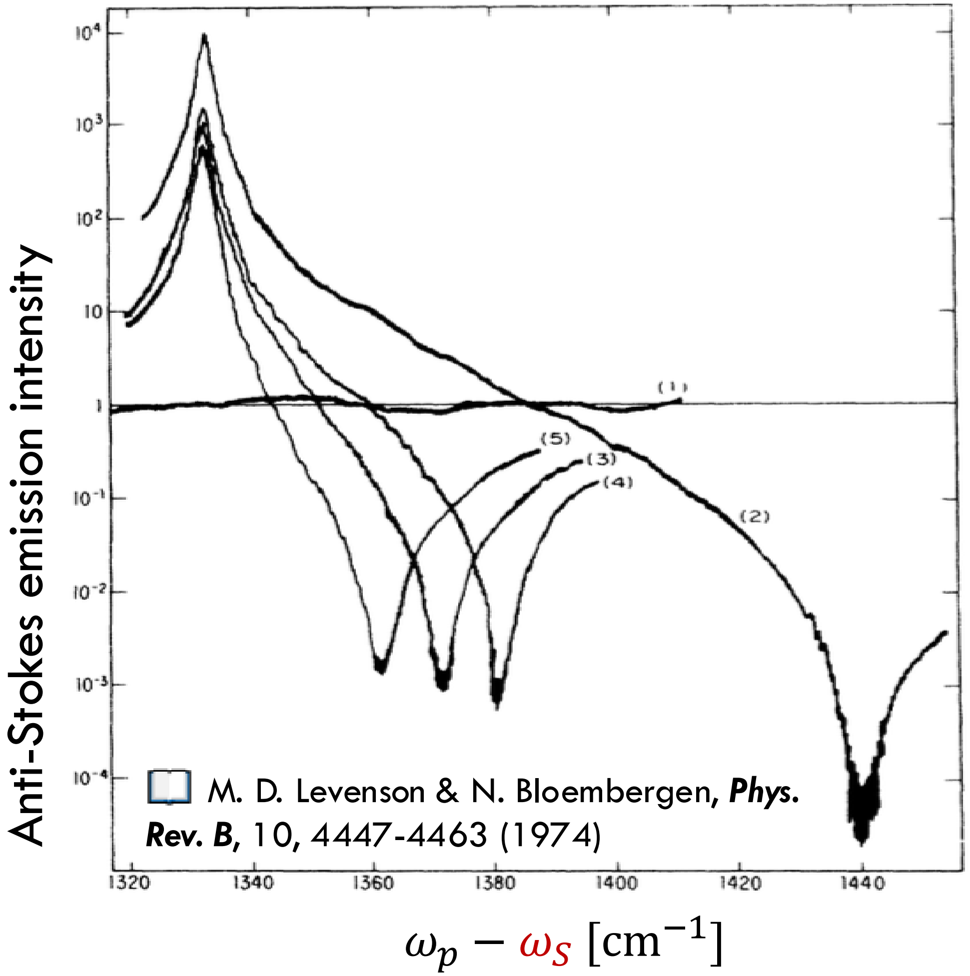


V. Vento *et al.* arXiv:2408.11477

See also V. Vento *et al.* ***Nature Comm.*** **14**, 2818 (2023)

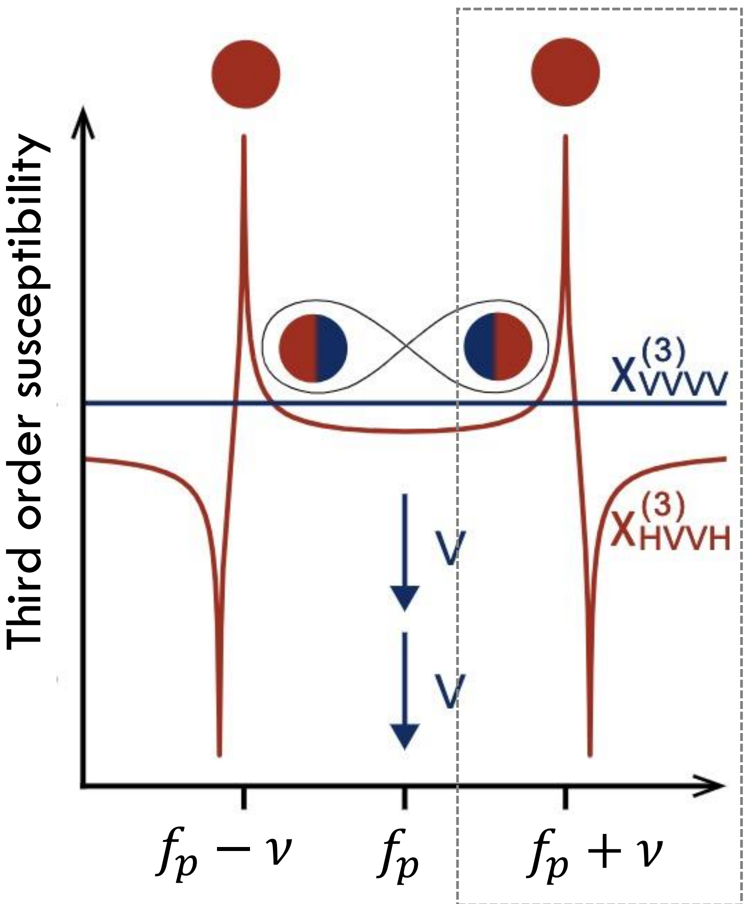
Valeria Vento





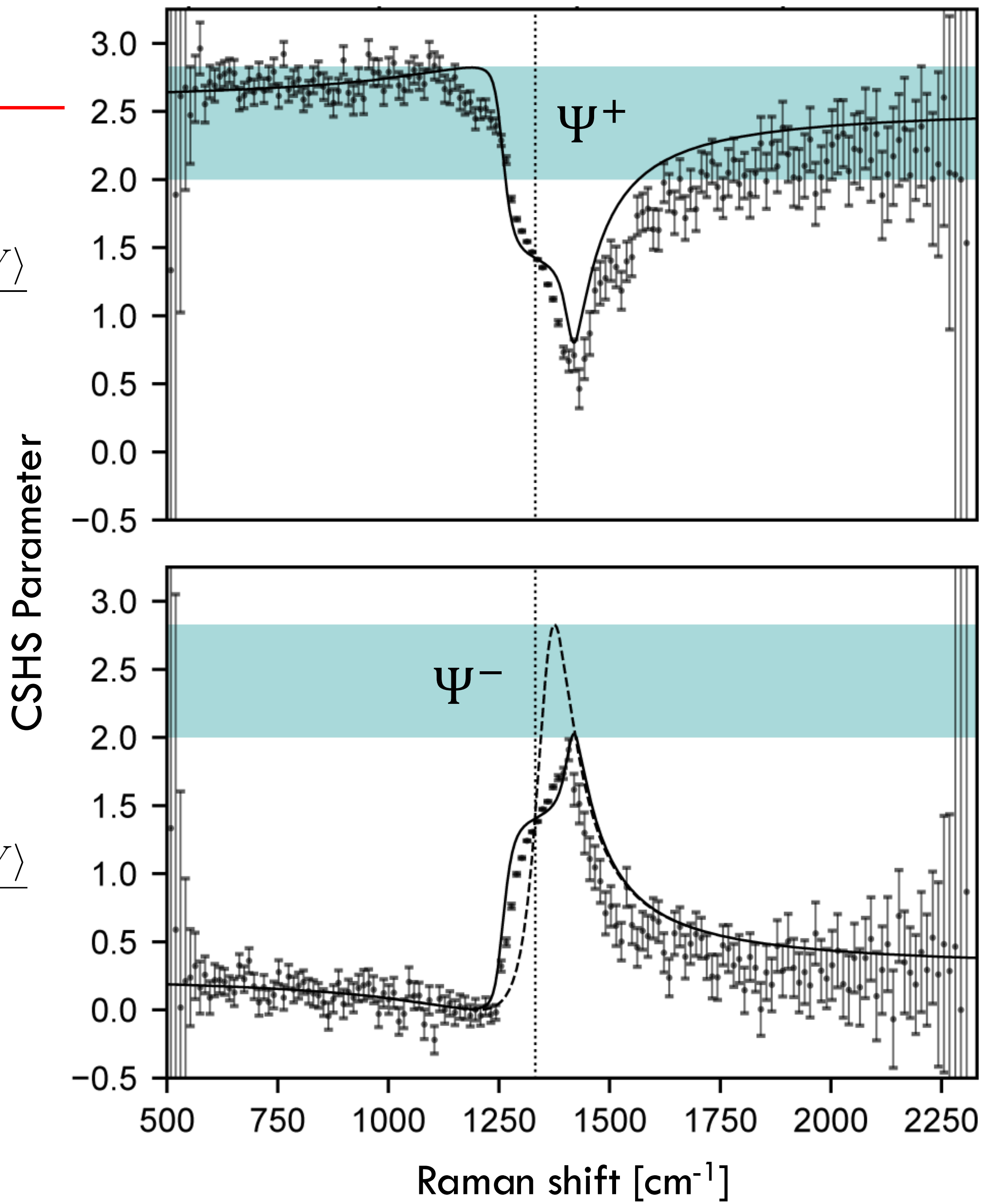
M. D. Levenson & N. Bloembergen, ***Phys. Rev. B***, 10, 4447-4463 (1974)

Polarisation Entanglement



$$|\Psi_+\rangle = \frac{|HH\rangle + |VV\rangle}{\sqrt{2}}$$

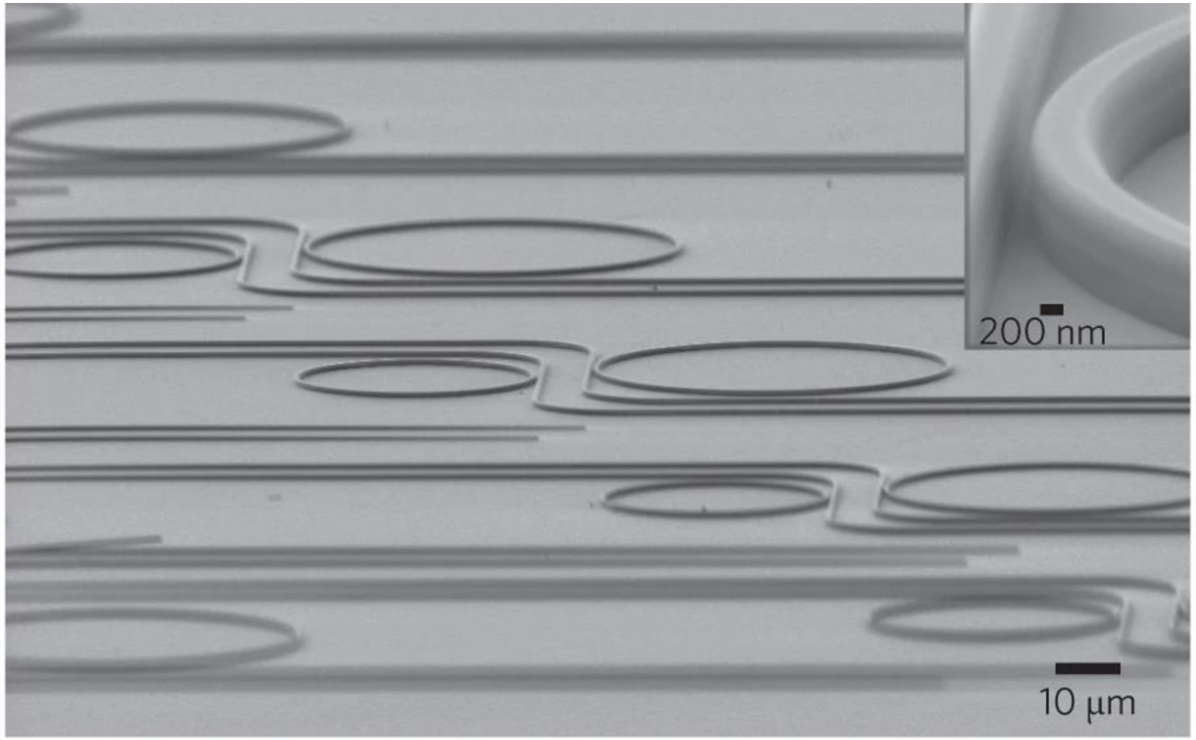
$$|\Psi_-\rangle = \frac{|HH\rangle - |VV\rangle}{\sqrt{2}}$$

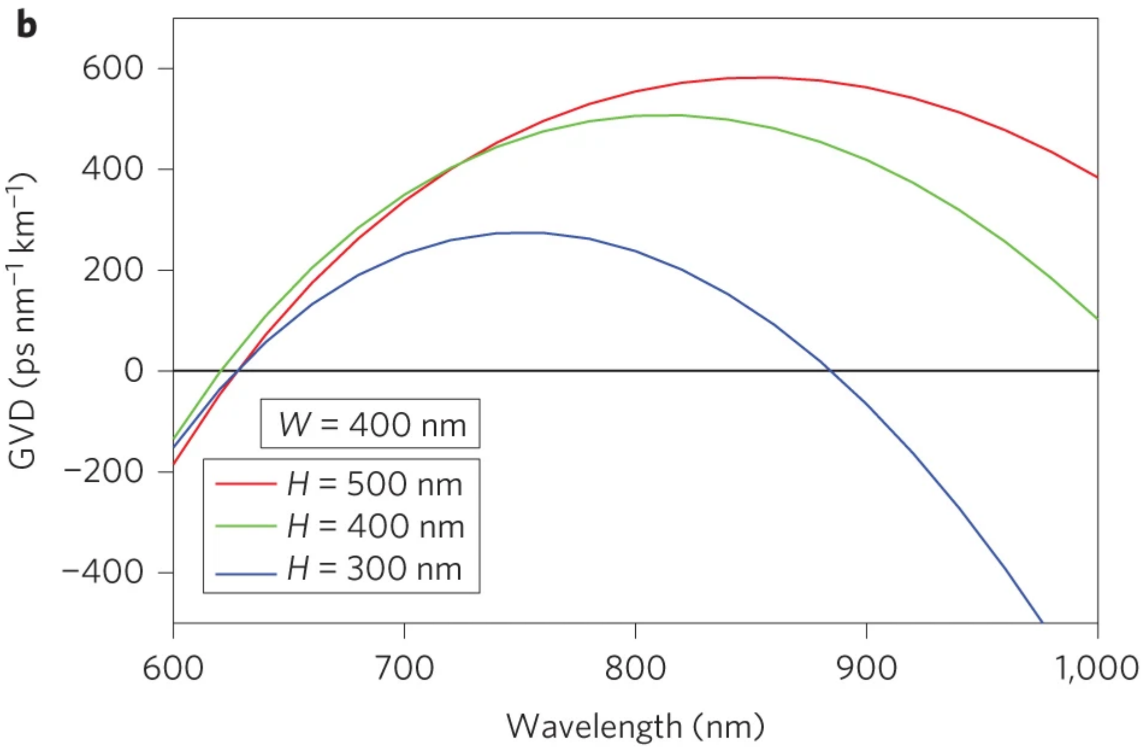


V. Vento *et al.* arXiv:2408.11477

Prospects for bright source of entangled photons

Diamond waveguide engineering to achieve broadband phase-matching of $\chi^{(3)}$ processes

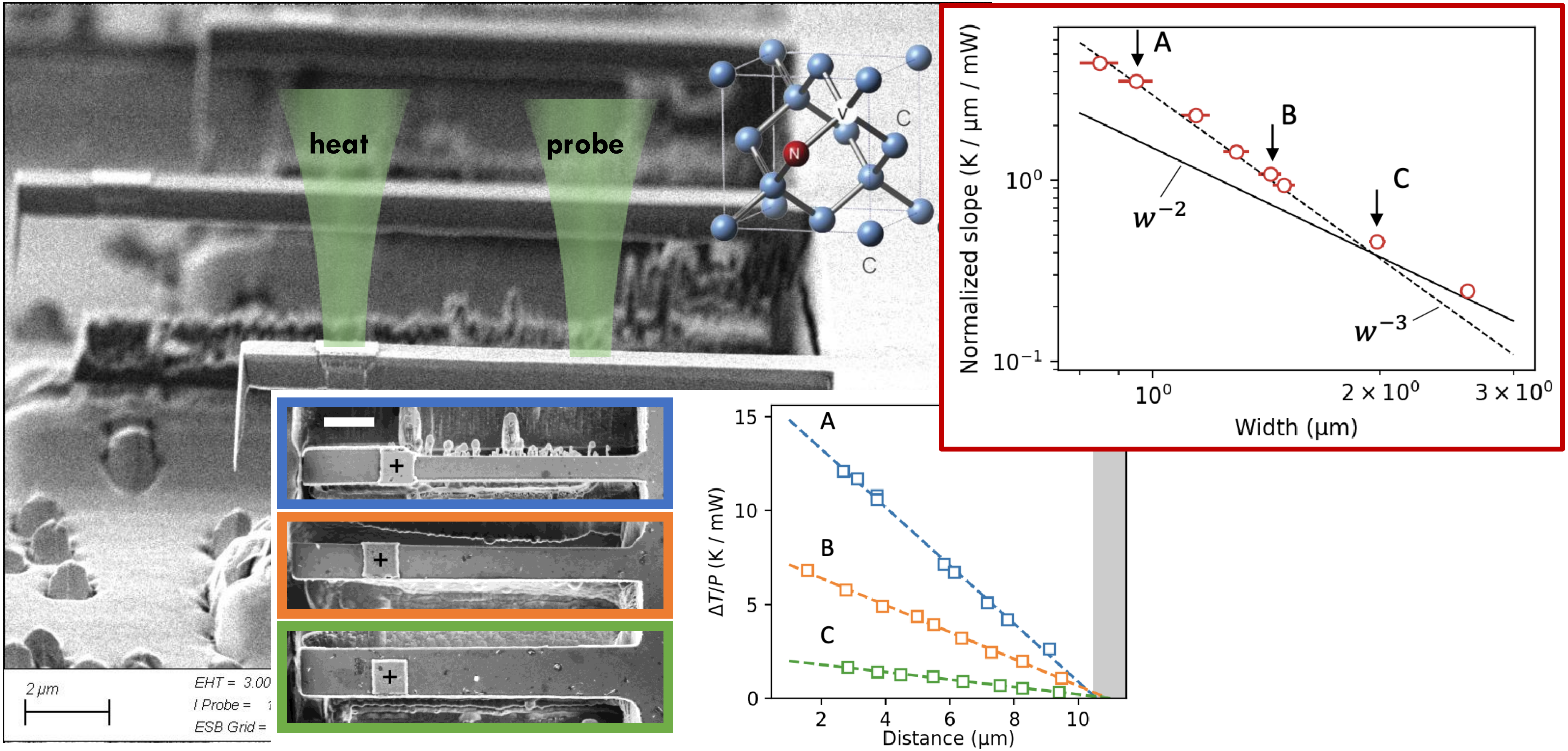




📖 B.J.M Hausmann … M. Loncar "Diamond nonlinear photonics" *Nature Photonics* **8**, 369–374 (2014)

Christophe Galland, Swiss Quantum Days 2025

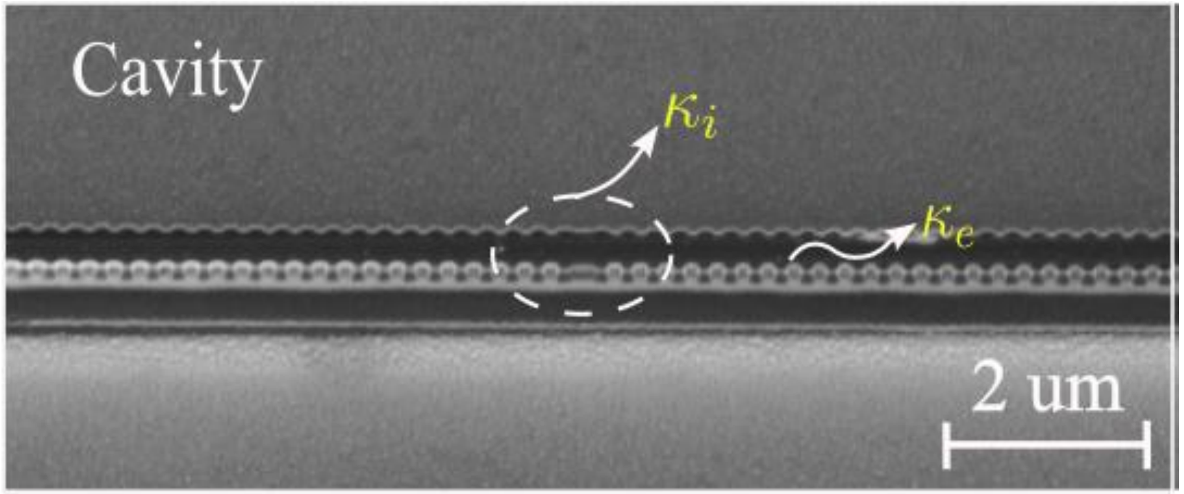
Extra: NV-based heat transport imaging

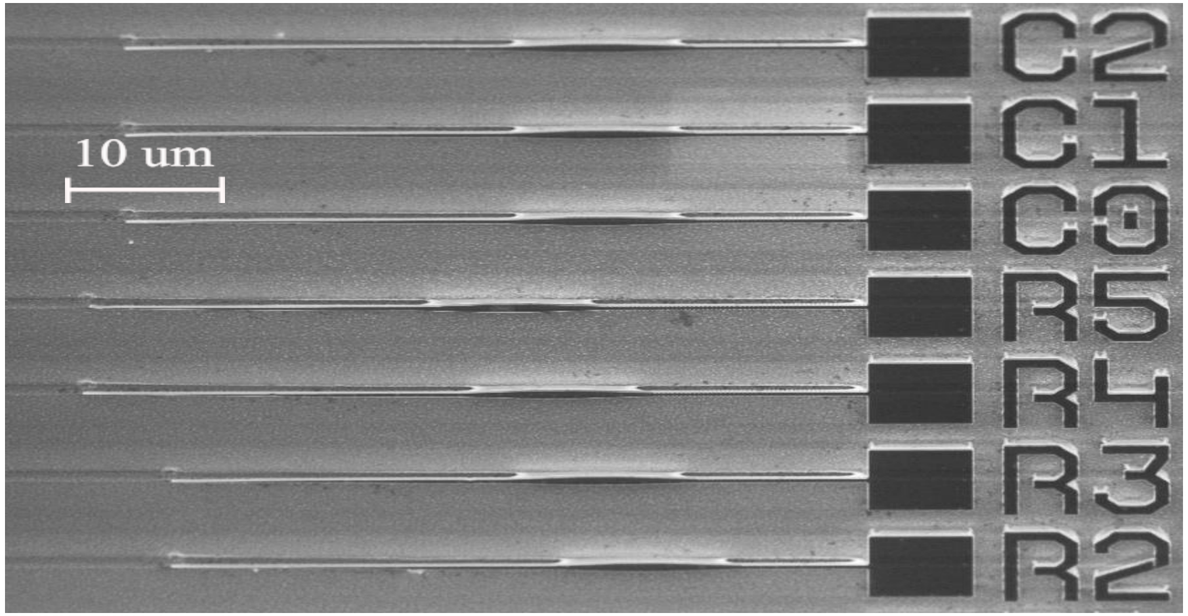


V. Goblot, K. Wu, Y. Zhu, arXiv:2411.04065 Collaboration: Michele Simoncelli (Cambridge), Nicola Marzari (EPFL)

Diamond photonics and color centers

Diamond nanophotonics



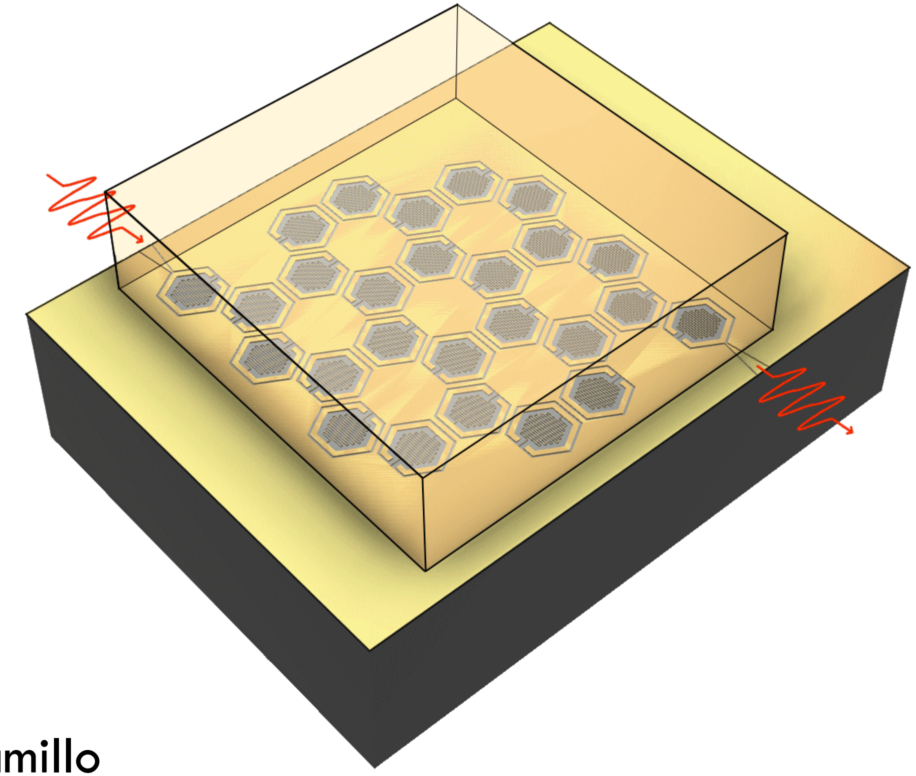


Yuchun Zhu
Claudio Jaramillo

NV coupling to superconducting resonators

Benedek Gaál, Valentin Goblot

(with Prof. Pasquale Scarlino)



Thank you!

May 23rd 2024