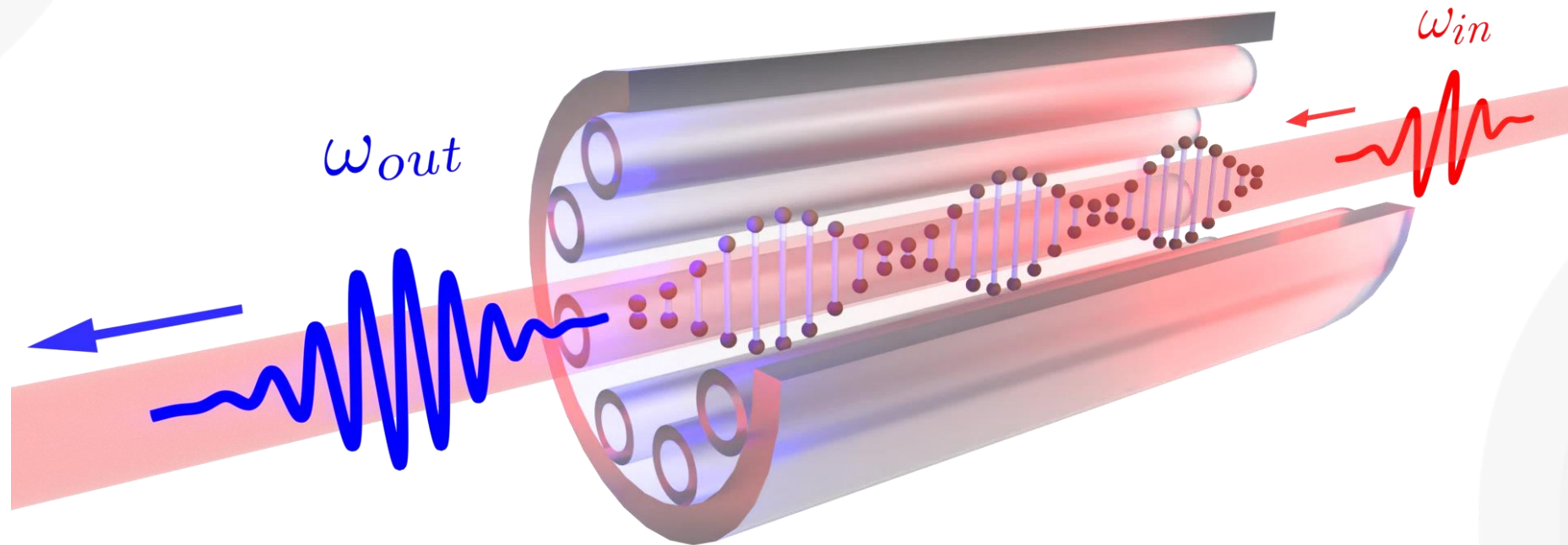


Coloring light quanta with synchronous molecular motion

David Novoa



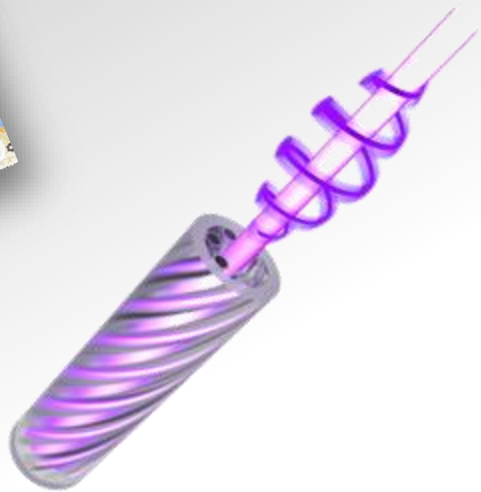
Bilbao & EFO-Lab



Bilbao & EFO-Lab



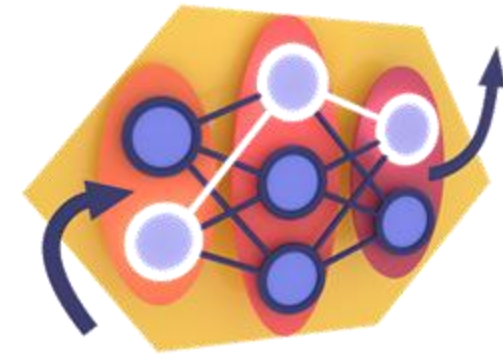
Extreme Fibre Optics Lab



Exotic light sources



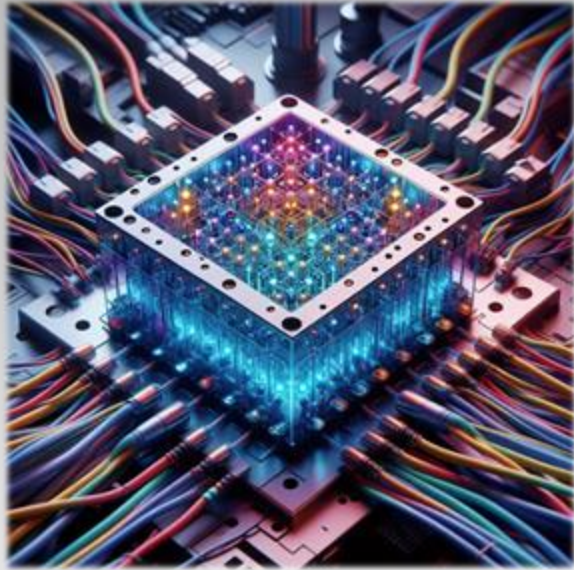
Nonlinear quantum optics



Smart photonics



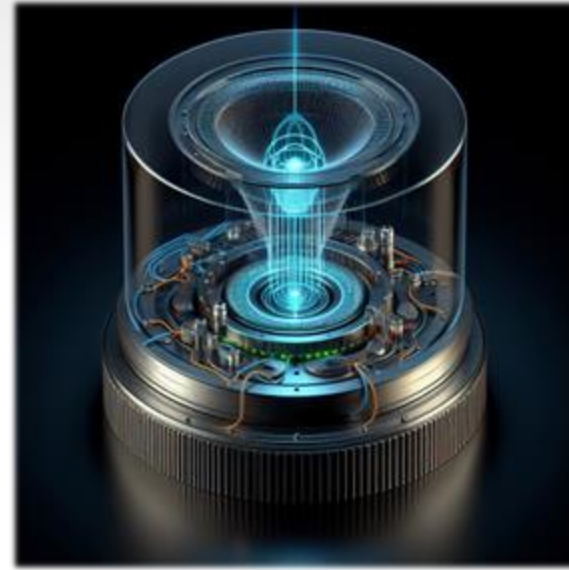
Quantum technologies



Q-Computing



Q-Simulation

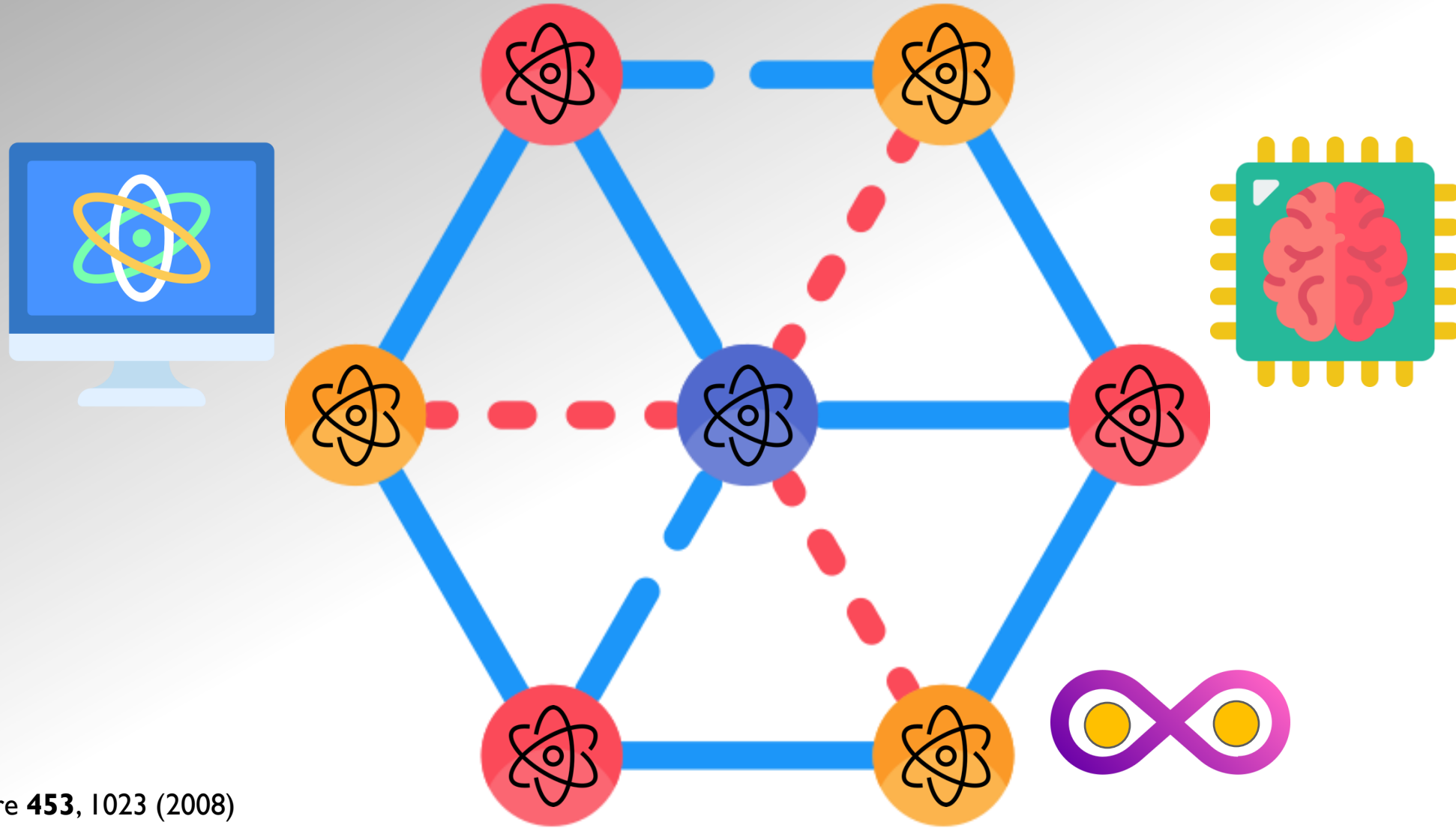


Q-Sensing



Q-Communications

Boosting hybrid quantum networks



Kimble, Nature **453**, 1023 (2008)

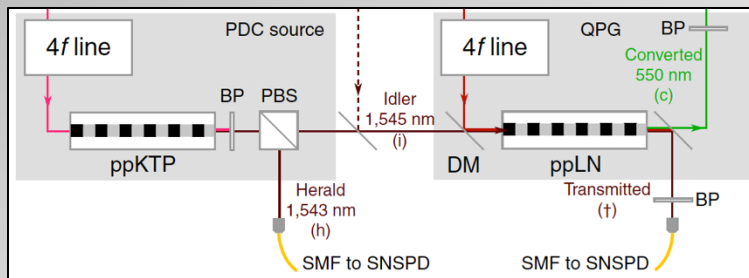
Awschalom *et al.*, PRX Quantum **2**, 017002 (2021)



Approaches to quantum frequency conversion

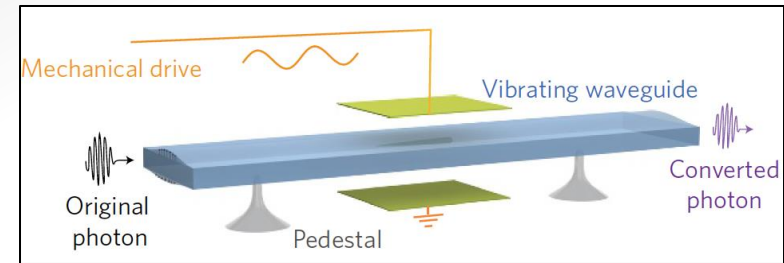
A good quantum convertor must (ideally) be

- Efficient
- Broadband
- Tunable
- Low noise & decoherence



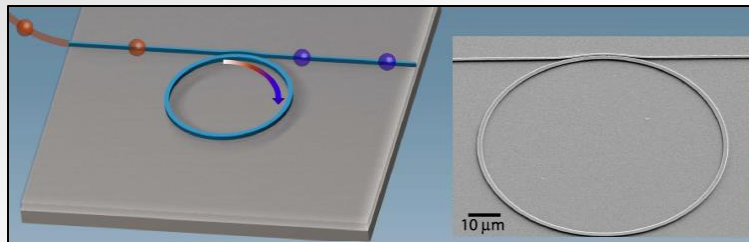
Nonlinear crystals

Allgaier *et al.*, Nat. Comms. **8**, 14288 (2017)



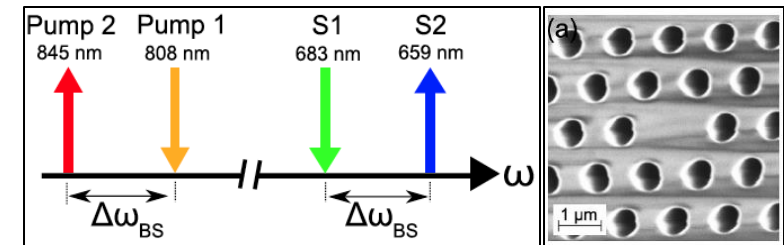
Opto-mechanical systems

Fan *et al.*, Nat. Photonics **10**, 766 (2016)



Microresonators on chip

Singh *et al.*, Optica **6**, 563 (2019)

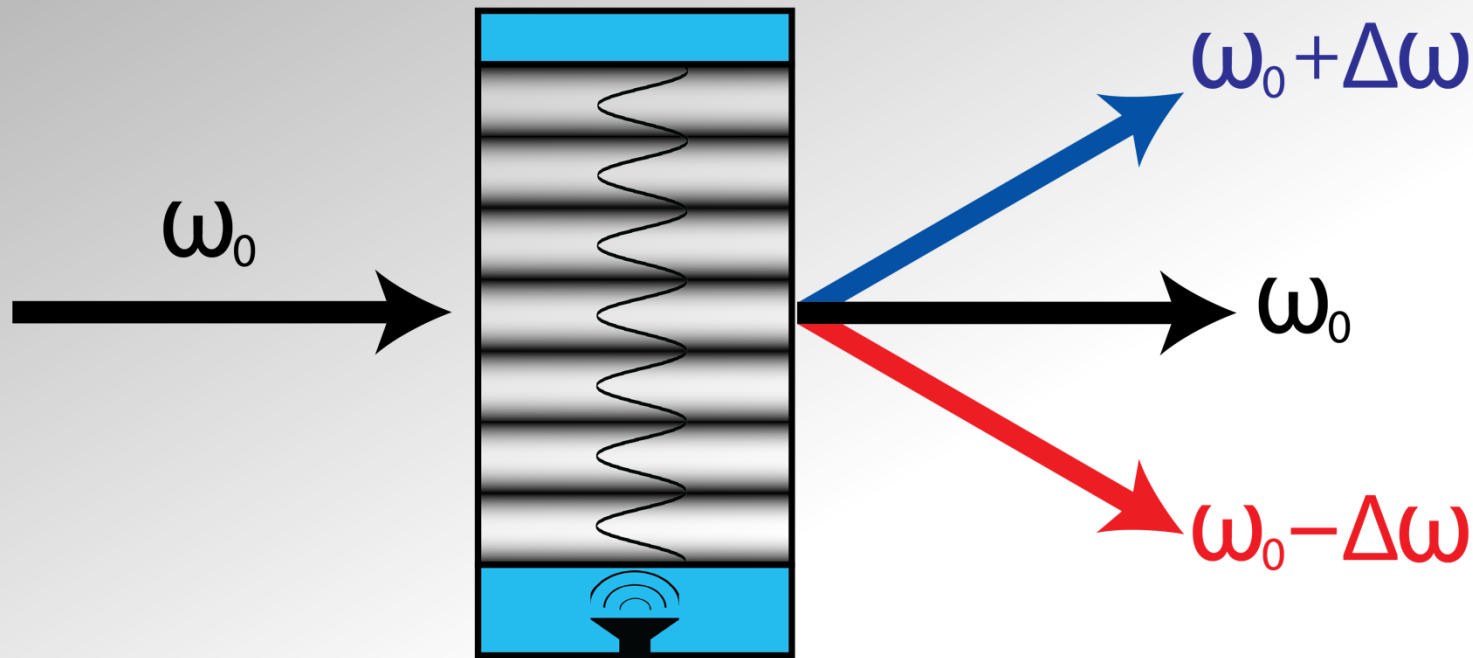


Optical fibers

McGuinness *et al.*, Phys. Rev. Lett. **105**, 093604 (2010)

Optical modulation of quantum light

“Acousto-optic” modulation at optical frequencies?



Wish list

- **Near-unity efficiency**
- **Tunable & Broadband**
- **Low (noise & loss)**
- **High spatial quality**
- **Thresholdless**

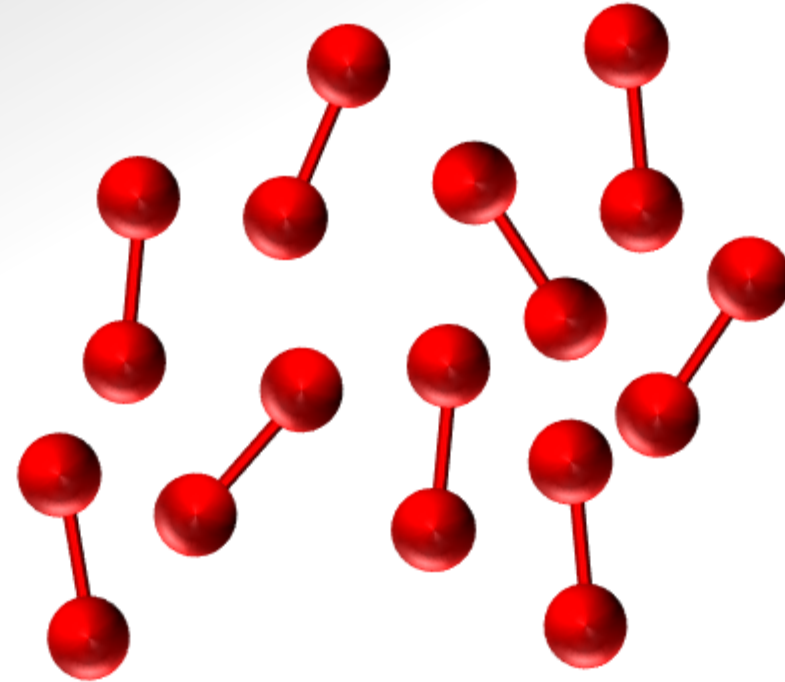


Hydrogen as a molecular modulator

- **Large shift ~ 125 THz**

- **Near-unity efficiency**
- **Tunable & Broadband**
- **Low (noise & loss)**
- **High spatial quality**
- **Thresholdless**

- **Highly dispersive**
- **Weak nonlinear response**

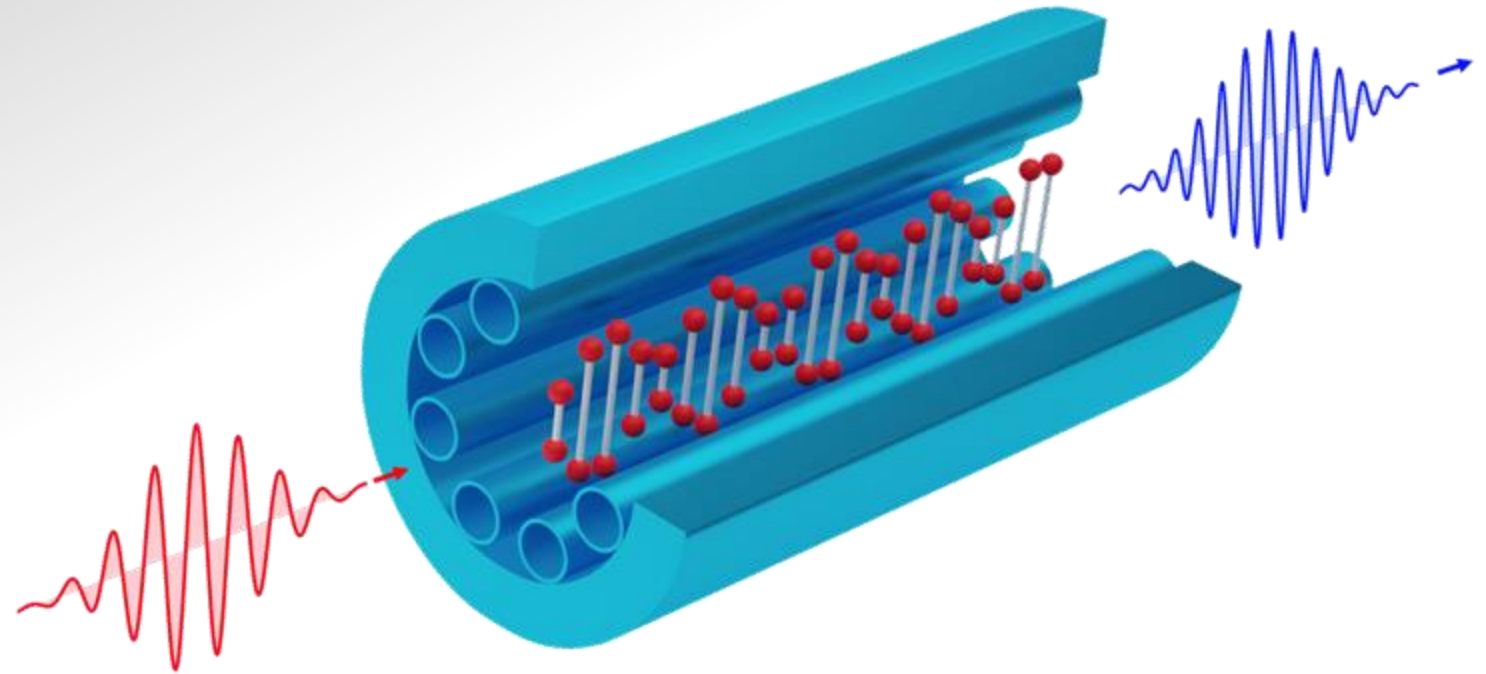


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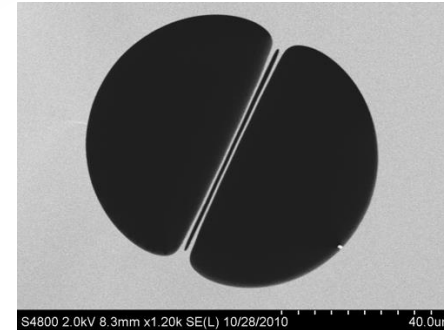
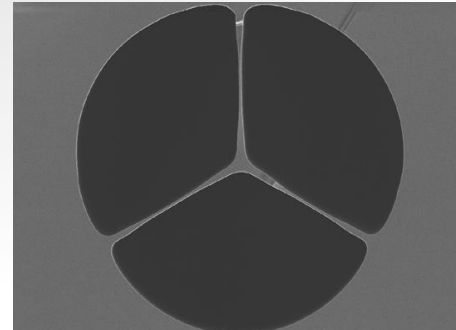
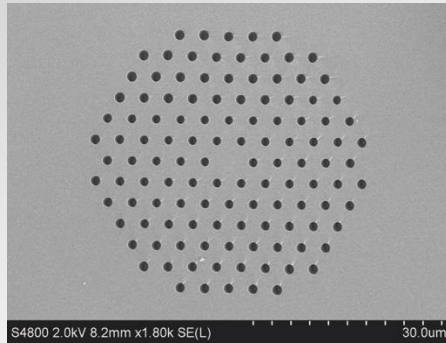


Photonic crystal fibers

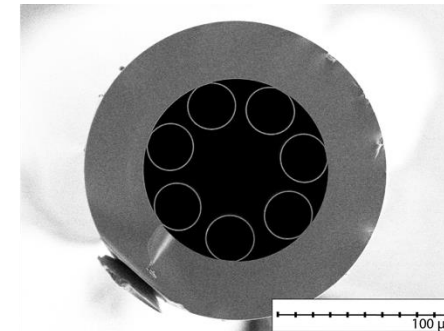
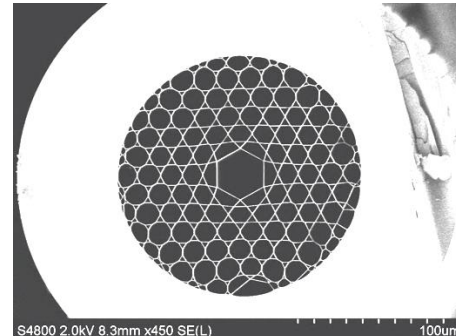
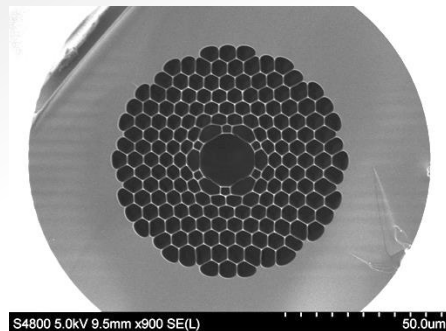


- ⊙ Philip Russell introduced the concept of PCF in the 90's
- ⊙ Micro-structured optical fibers with a periodic cladding
- ⊙ Capable of guiding light in either solid or hollow channels

Solid core



Hollow core

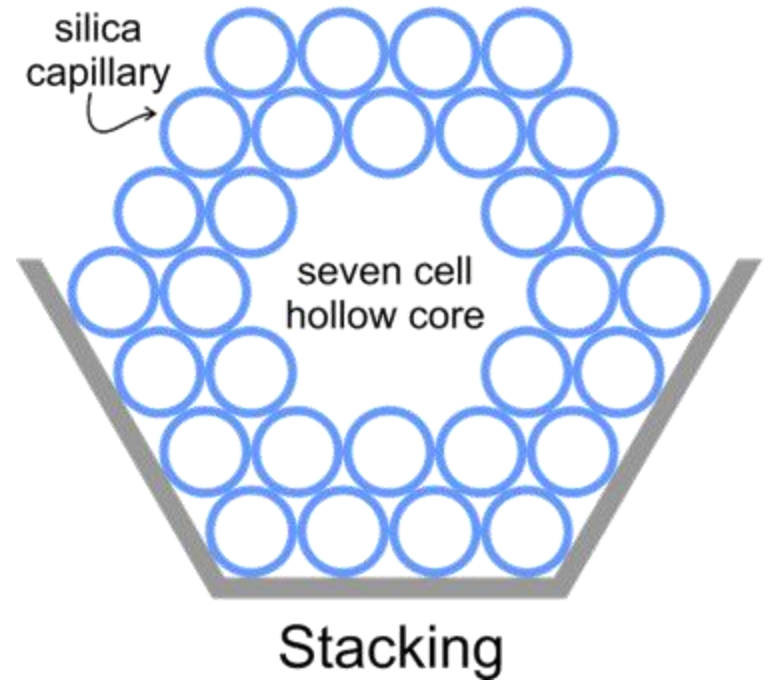
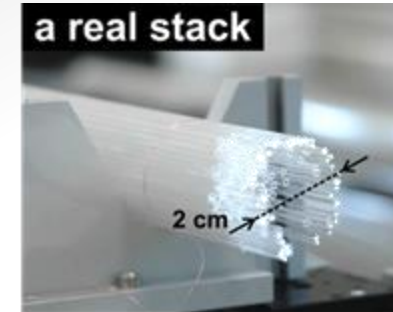
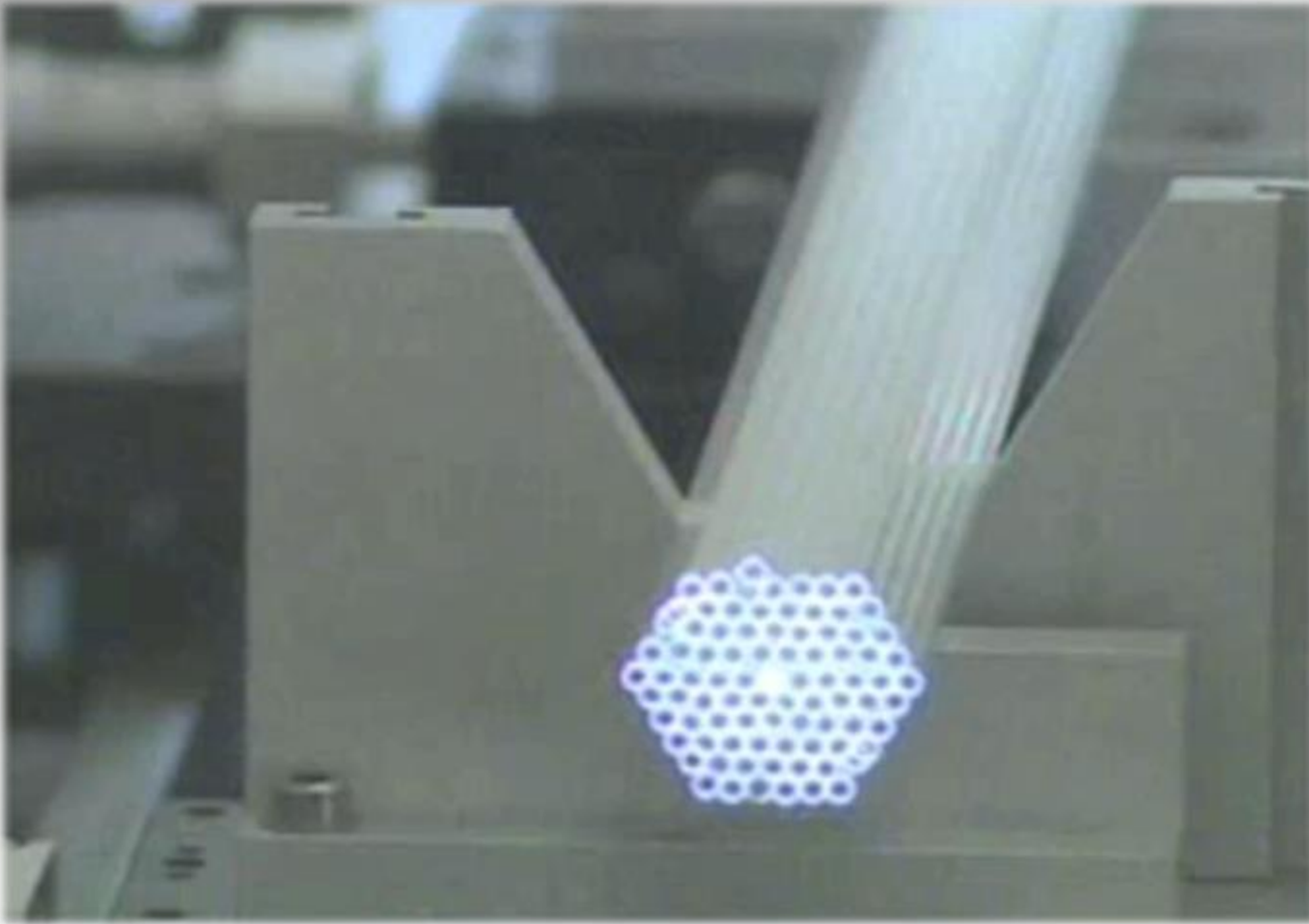


Knight *et al.*, *Opt. Lett.* **21**, 1547 (1996)

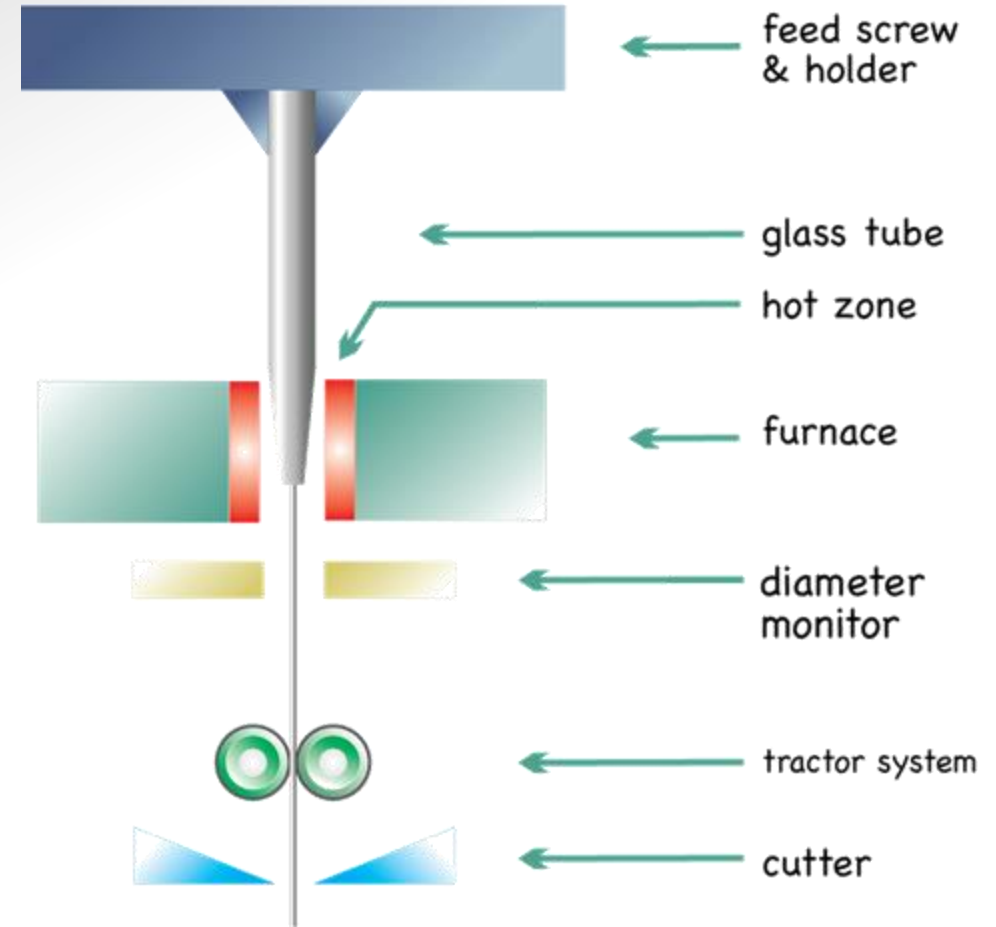
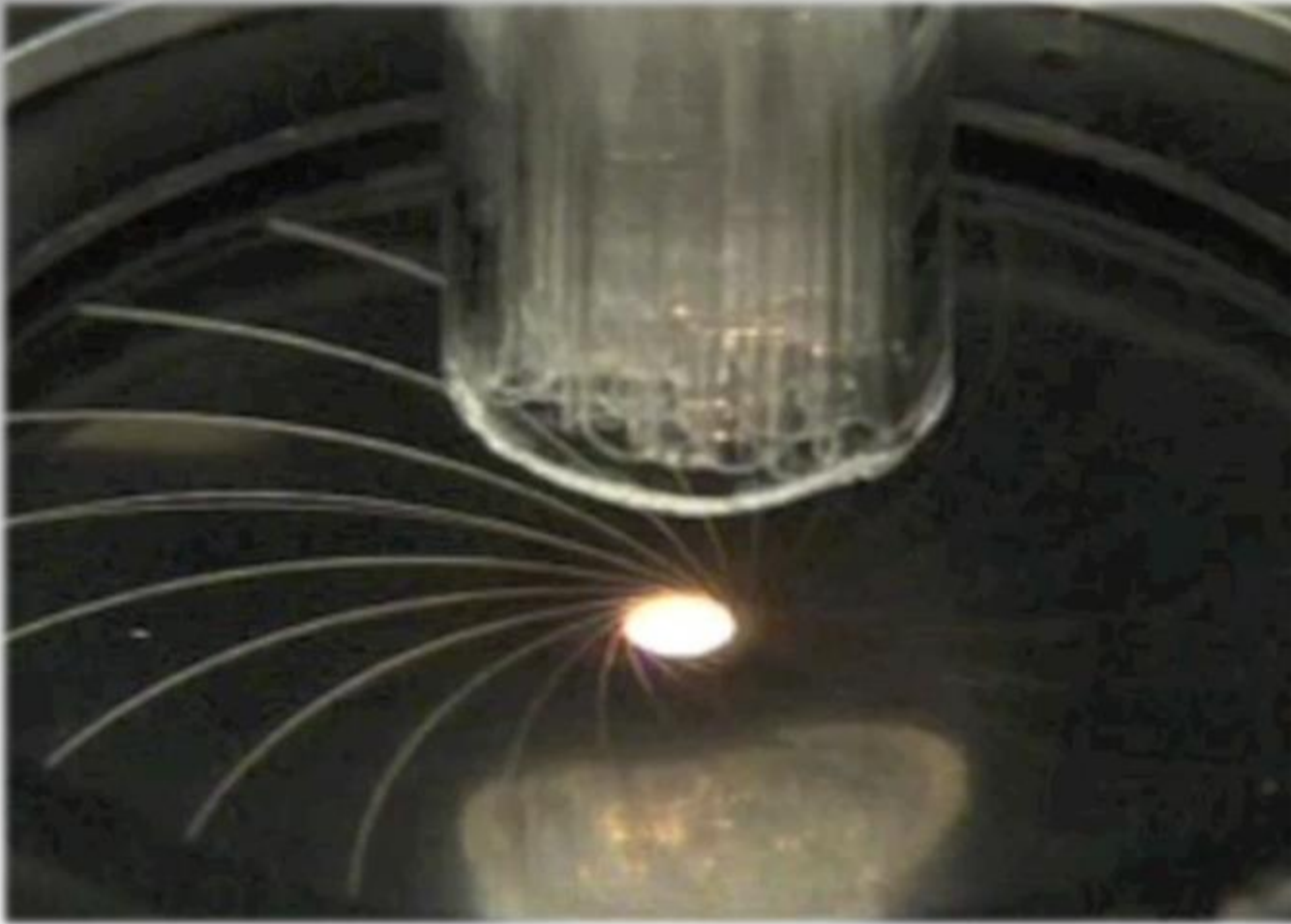
Russell, *Science* **299**, 358 (2003)



PCF Fabrication: Stack and Draw

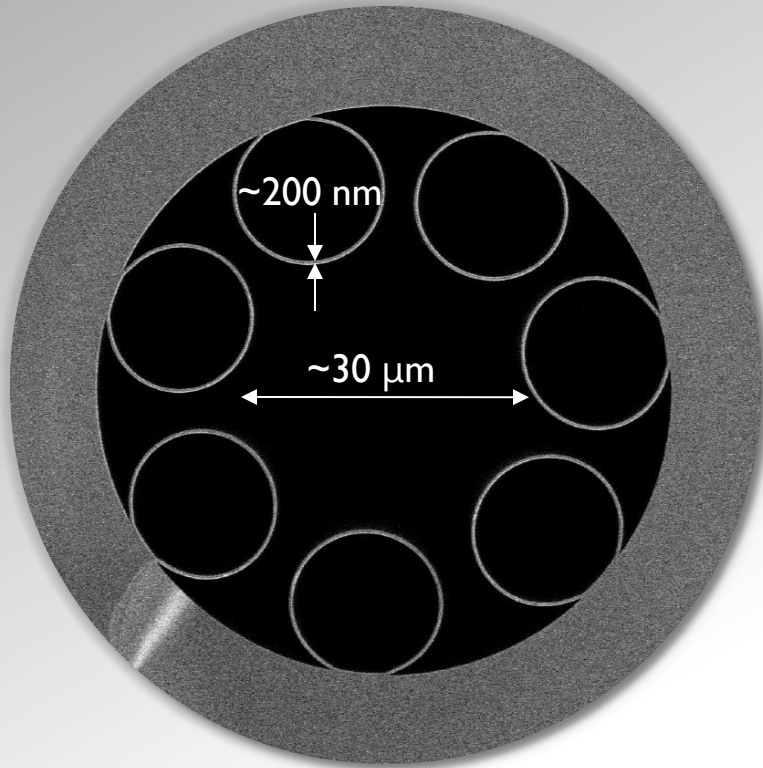


PCF Fabrication: Stack and Draw



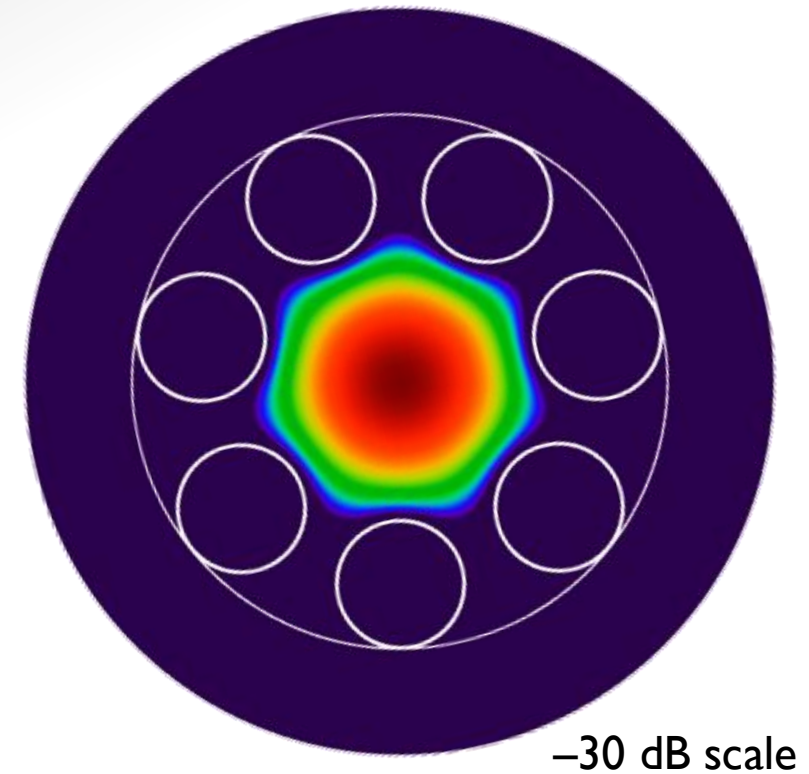
Hollow-core anti-resonant fibers

Scanning electron micrograph



- “Faster” speed of light
- Ultralow attenuation
- Broad transmission windows
- High damage threshold
- Adjustable dispersion & nonlinearity

Finite-element modelling



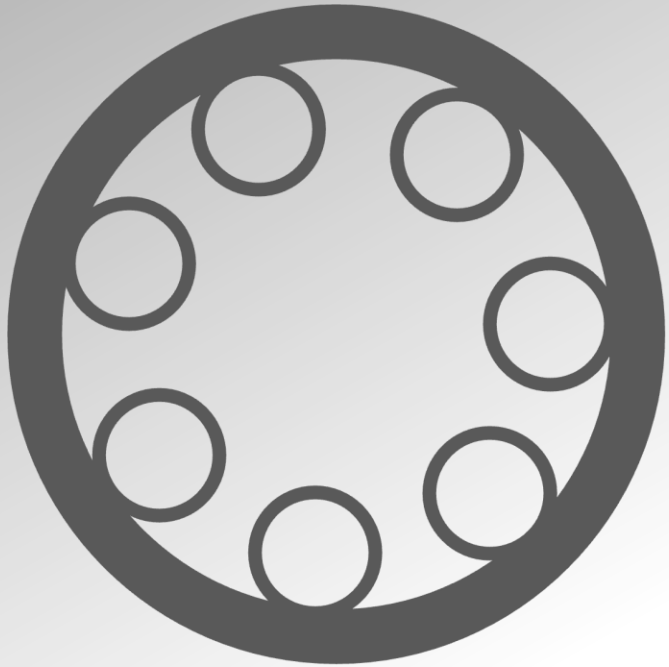
Benabid *et al.*, *Science* **298**, 399 (2002)

Pryamikov *et al.*, *Opt. Express* **19**, 1441 (2011)

Numkam-Fokoua *et al.*, *Adv. Opt. Phot.* **15**, 1 (2023)

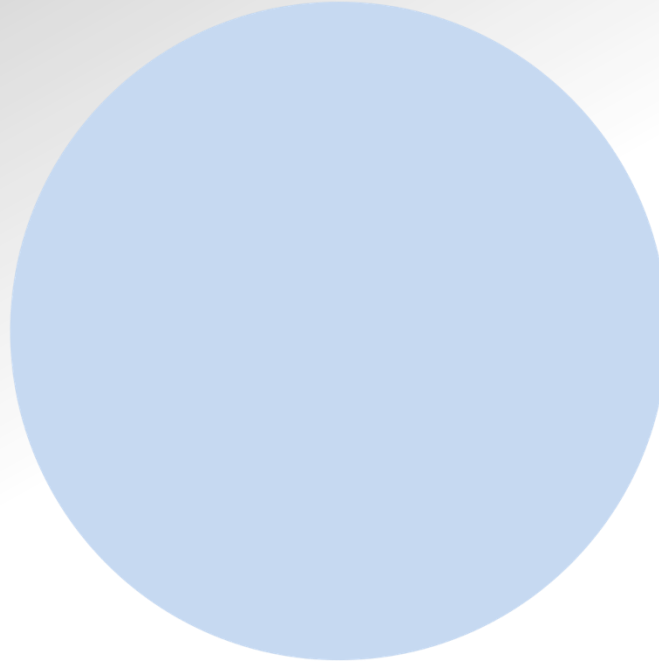
Pressure-tunable dispersion

Waveguide dispersion
(Anomalous)



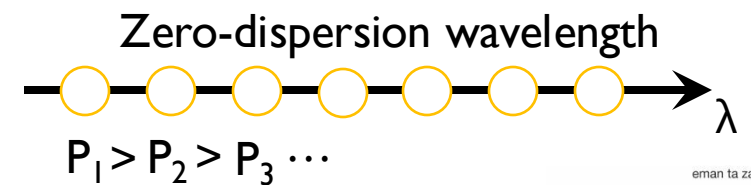
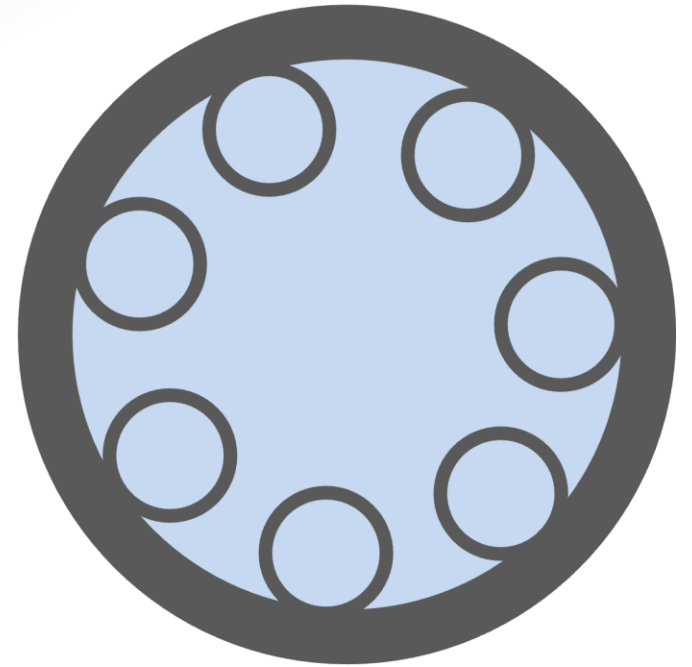
+

Gas dispersion
(Normal)



=

Combined dispersion
(Tunable)

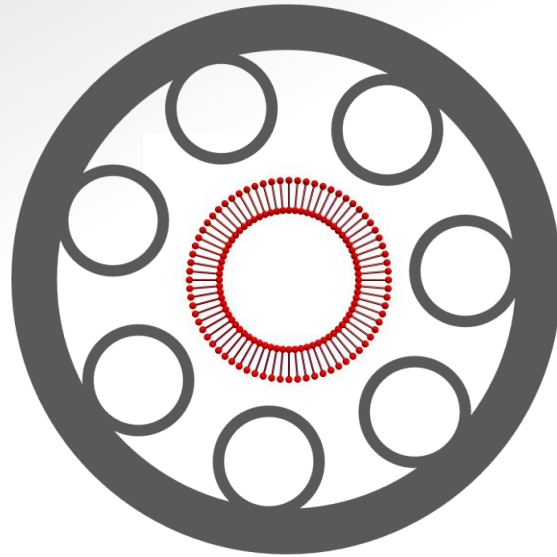
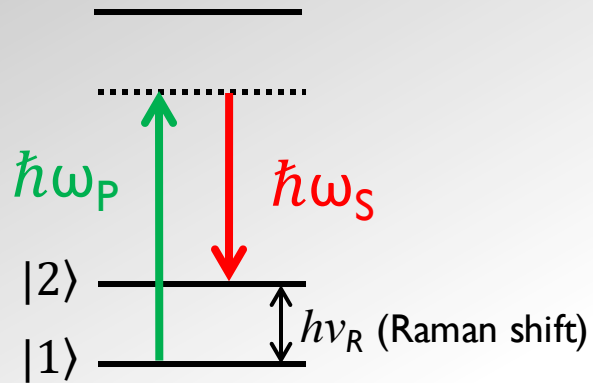
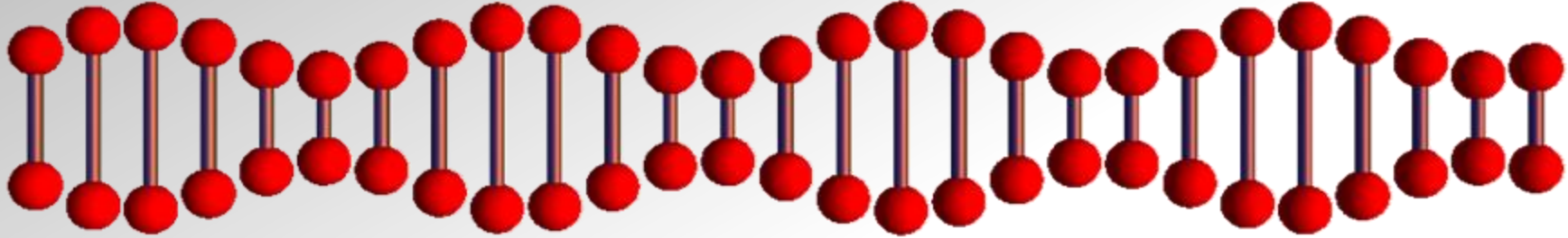
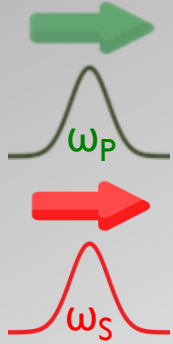


Marcatili *et al.*, Bell.Sys.Tech.J. **43**, 1783 (1964)

Russell *et al.*, Nat. Photonics **8**, 278 (2014)



Raman scattering & molecular modulation

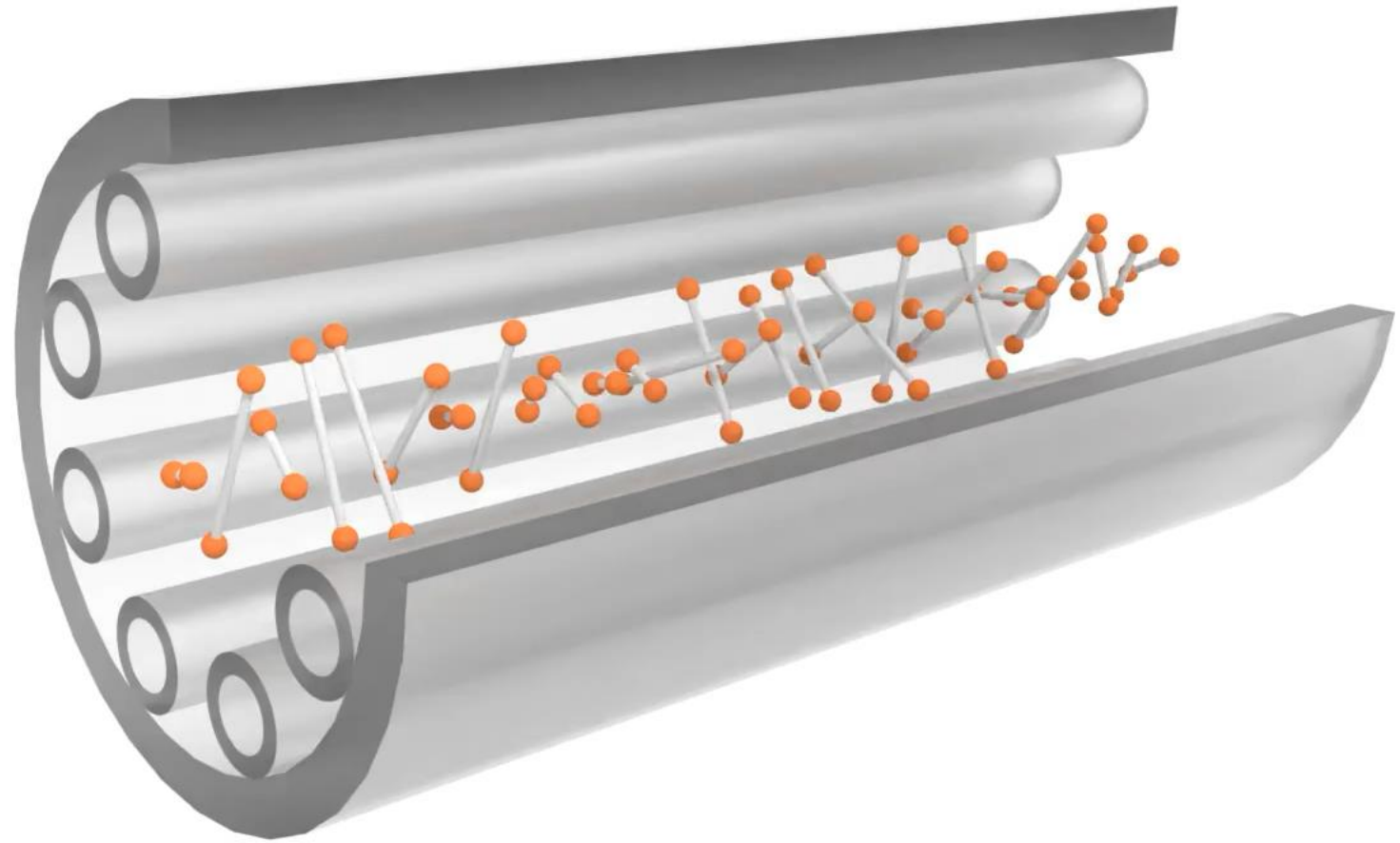


Synchronous molecular motion
 ↓
Travelling refractive-index pattern

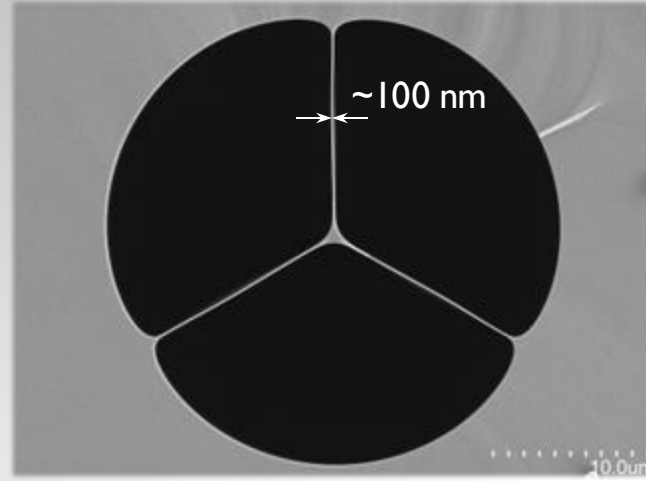
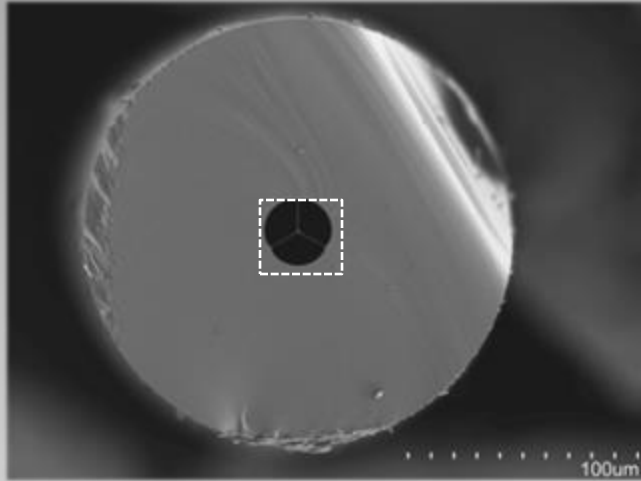
Hosseini *et al.*, *Phys. Rev. Lett.* **119**, 253903 (2017)
 Mridha *et al.*, *Optica* **6**, 731 (2019)
 Tyumenev *et al.*, *ACS Photonics* **7**, 1989 (2020)
 Arcos *et al.*, *EPL*, *in press* (2024)

Quantum frequency conversion of single photons

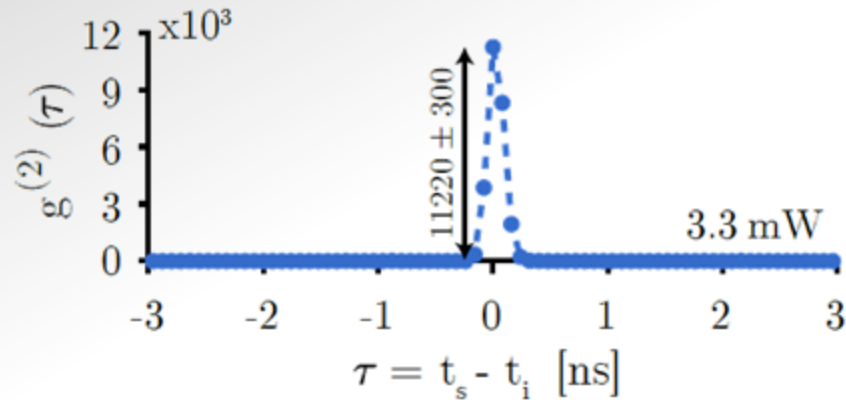
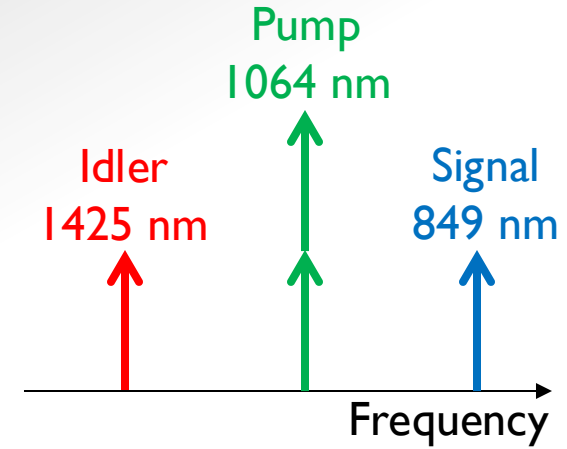
Tyumenev, Hammer, Joly, Russell, DN, Science **376**, 621 (2022)



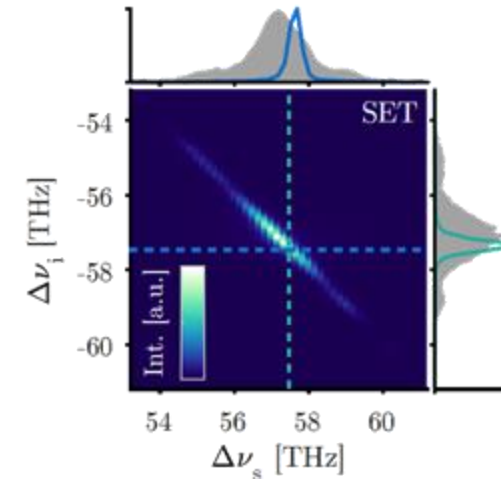
Source of entangled biphotons



Spontaneous four-wave mixing

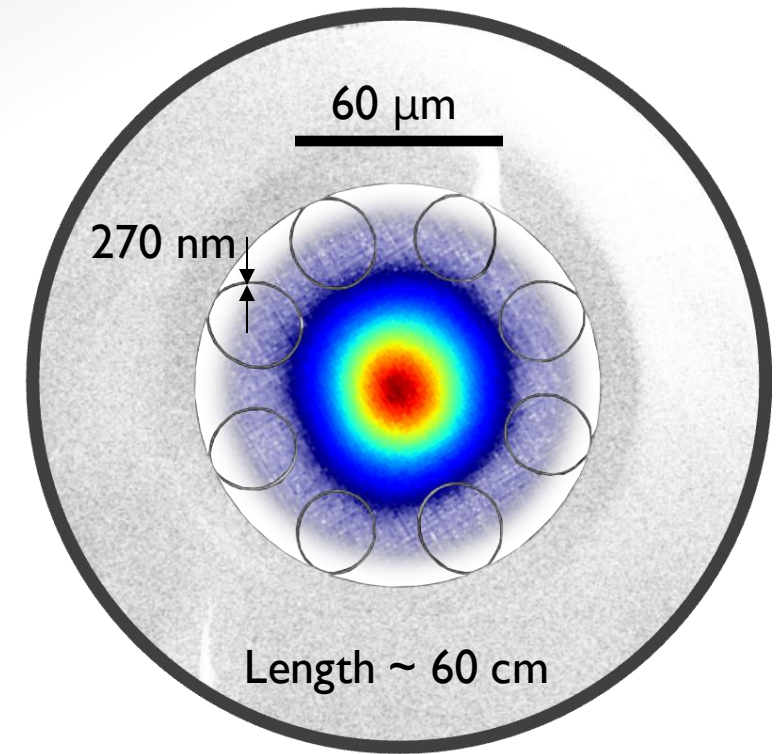
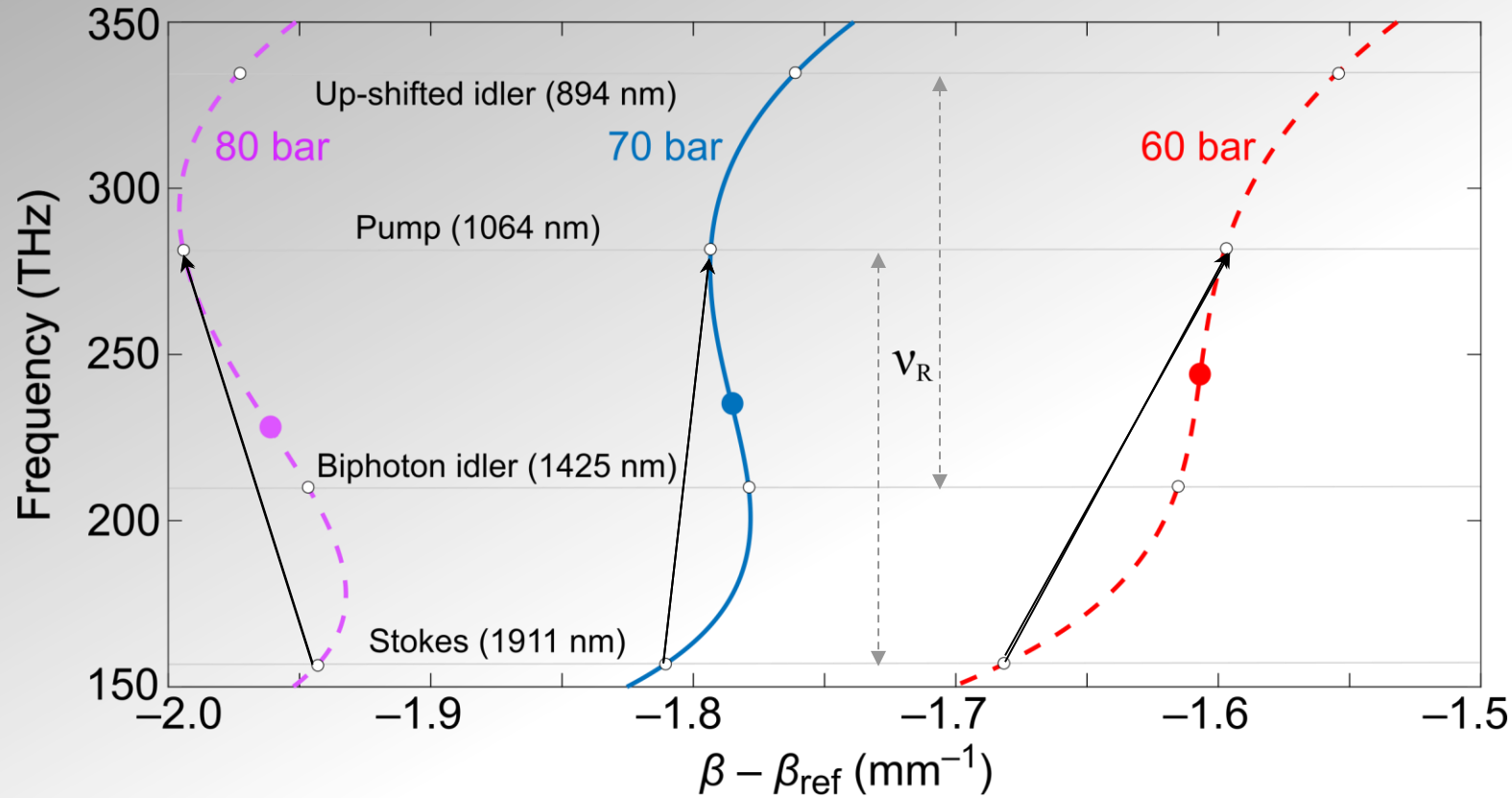


Stimulated emission tomography



Phase-matched molecular modulation

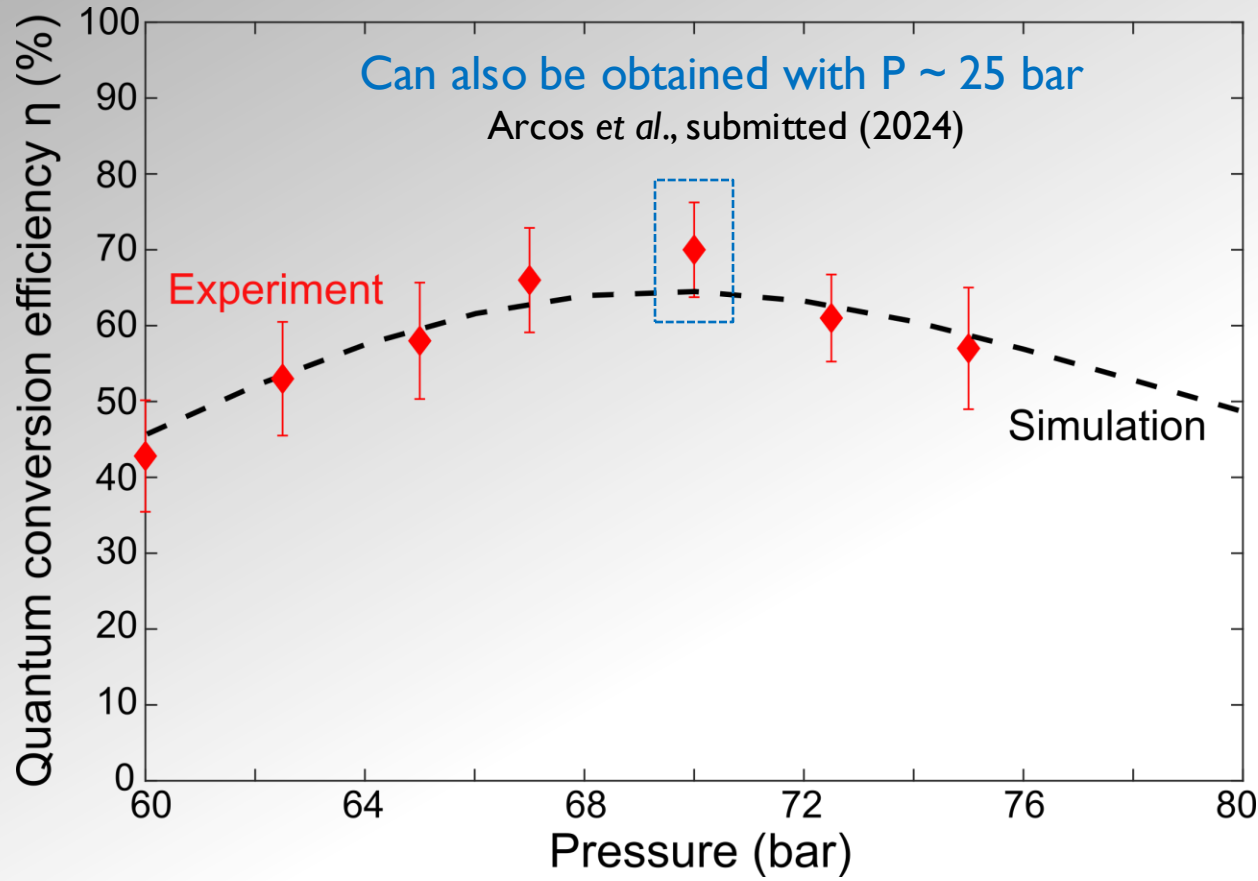
Energy & momentum must be conserved in the interaction



Record-high launching efficiency ~ **96%**

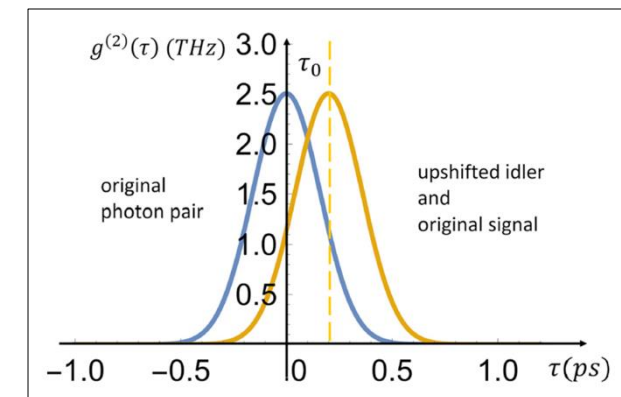
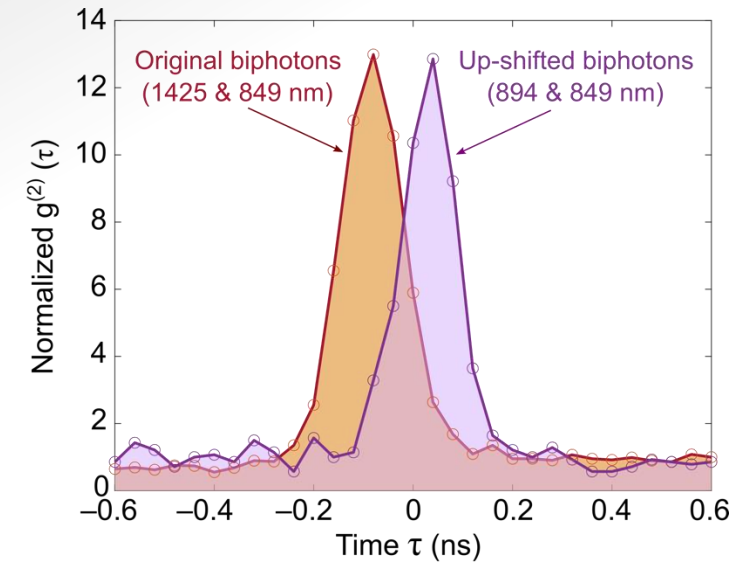
Quantum frequency conversion of single photons

Highly efficient conversion at the quantum level



Tyumenev *et al.*, Science **376**, 621 (2022)

Preservation of nonclassical correlations



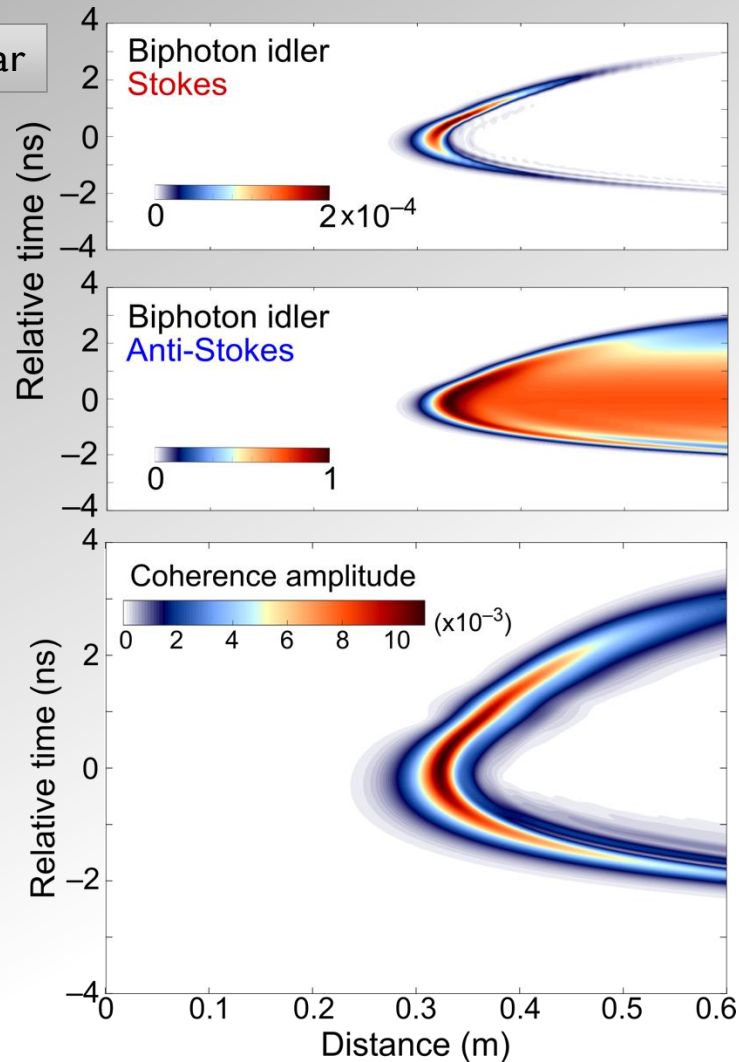
Wang *et al.*, Phys. Rev.A **108**, 063706 (2023)



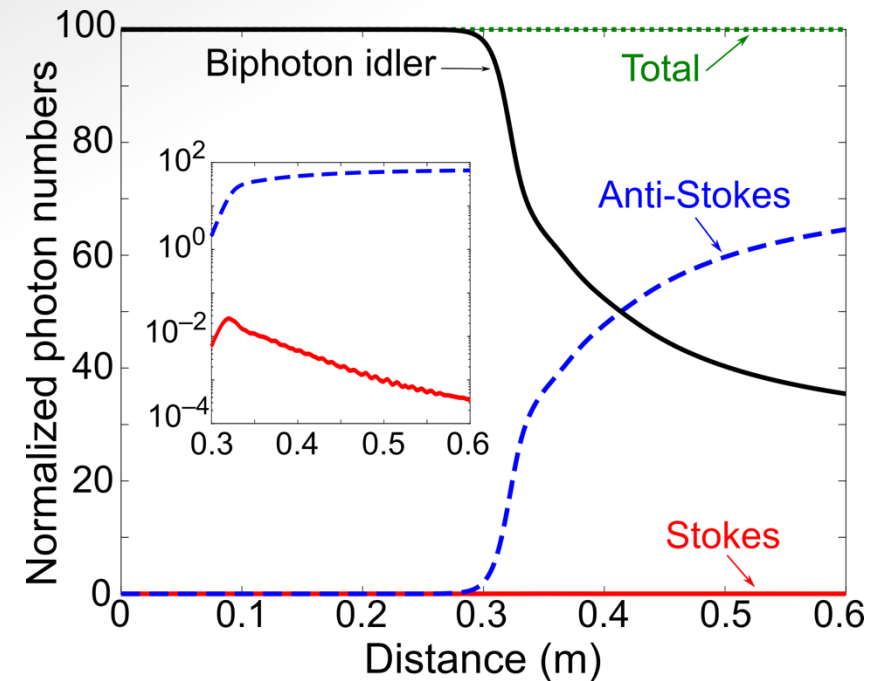
Modelling of the quantum conversion process

Modelling: Maxwell-Bloch equations

P = 70 bar







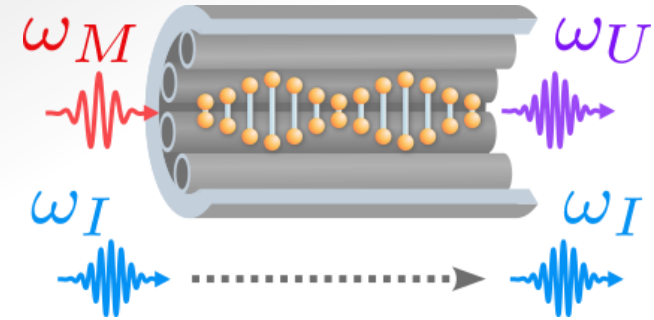
Selectivity: Down-conversion is 5 orders of magnitude weaker



Quantum model of fibre-based molecular modulation

Does this approach preserve other quantum properties such as entanglement?

Formalism	Light	Matter
Maxwell-Bloch		
G.Tavis-Cummings		



Two-stage model:

(I) Preparation of the molecular quantum coherence

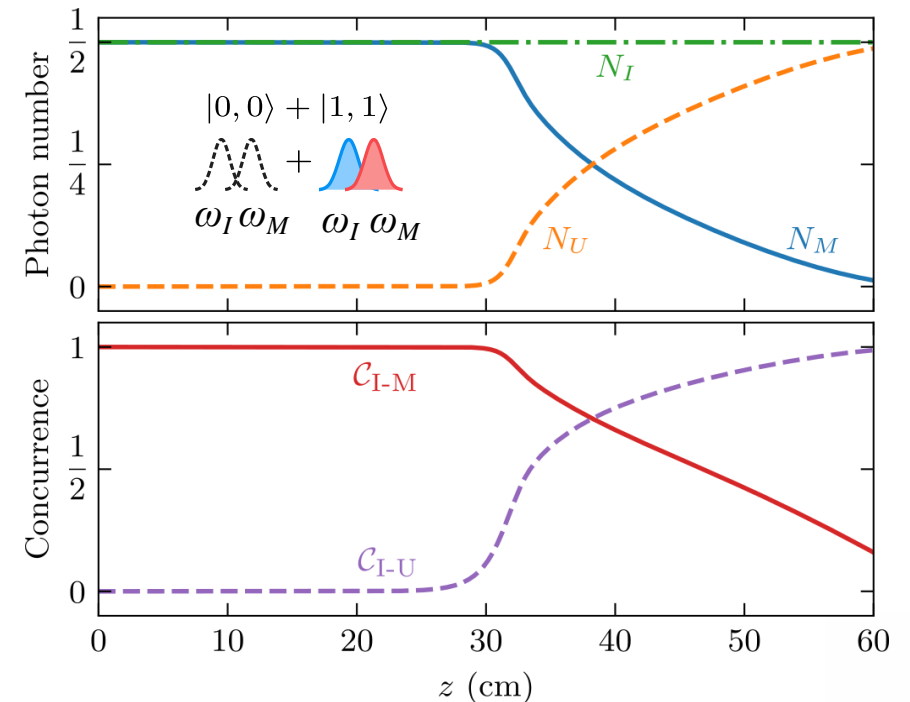
$$\hat{H}_\alpha^I = \hbar G_S (e^{i\Delta\beta z} \alpha_P \alpha_S^* \hat{J}^+ + e^{-i\Delta\beta z} \alpha_P^* \alpha_S \hat{J}^-)$$

(II) Molecular modulation of arbitrary quantum states

$$\hat{H}_\xi^I = \hbar G_U (\xi^* e^{i\Delta\beta z} \hat{a}_M^\dagger \hat{a}_U + \xi e^{-i\Delta\beta z} \hat{a}_M \hat{a}_U^\dagger)$$

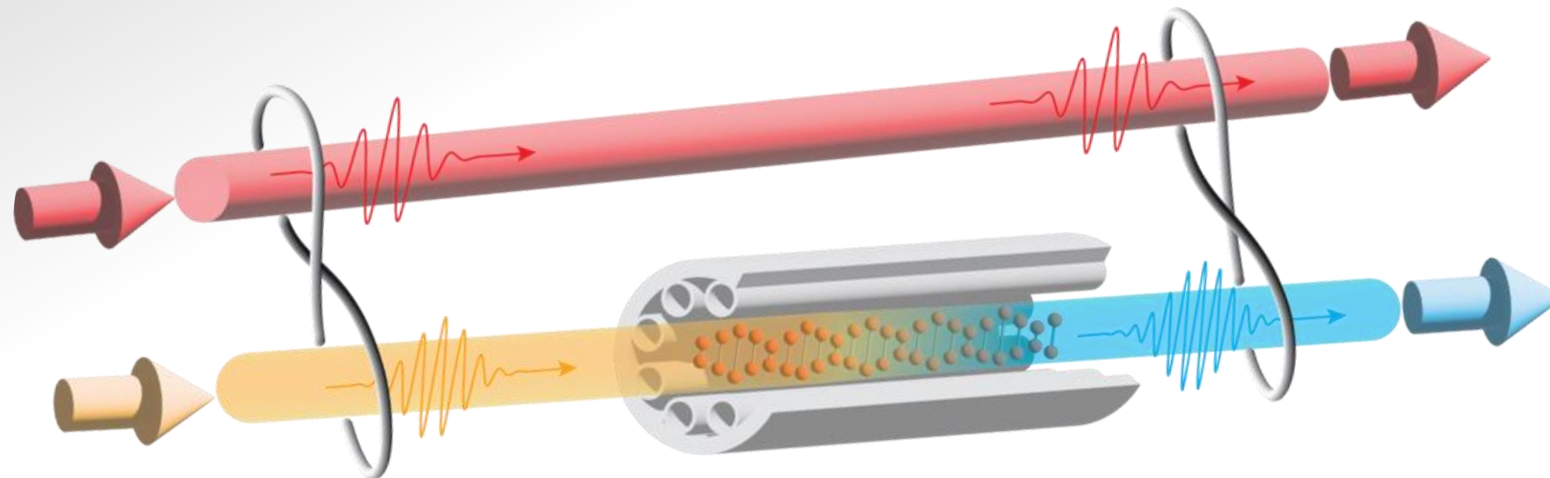
Tavis and Cummings, Phys. Rev. **170**, 379 (1968)

González-Raya *et al.*, submitted (2024)



Conclusions

- ✓ Anti-resonant fibers are excellent platforms for quantum nonlinear photonics
- ✓ Efficient quantum frequency conversion achieved in H₂-filled anti-resonant fibers
- ✓ A full quantum framework predicts the transfer of entanglement during molecular modulation



Coloring light quanta with synchronous molecular motion

Thank you for your attention!

