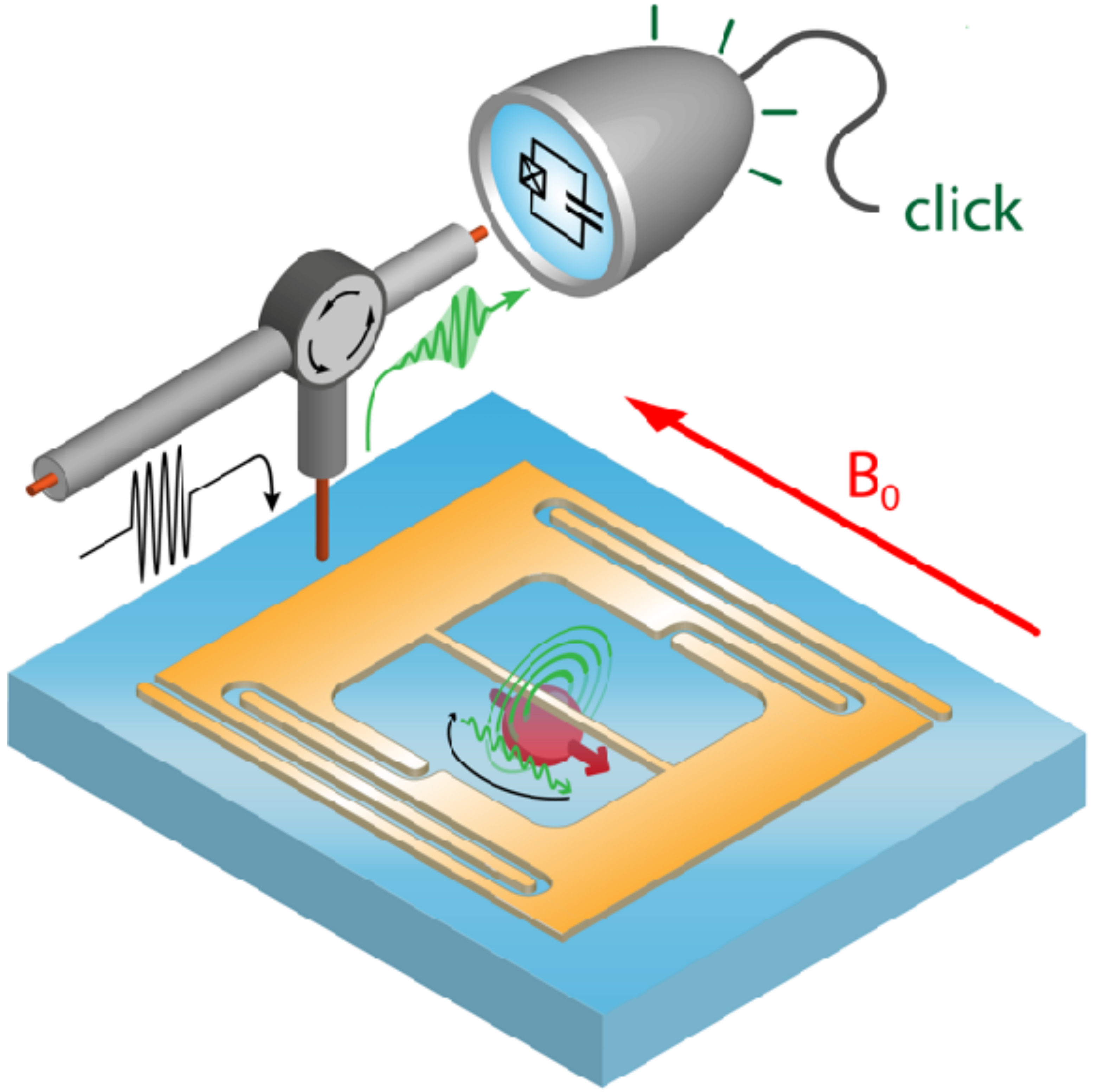
*Detecting **single** electron spin resonance with a single microwave photon counter*



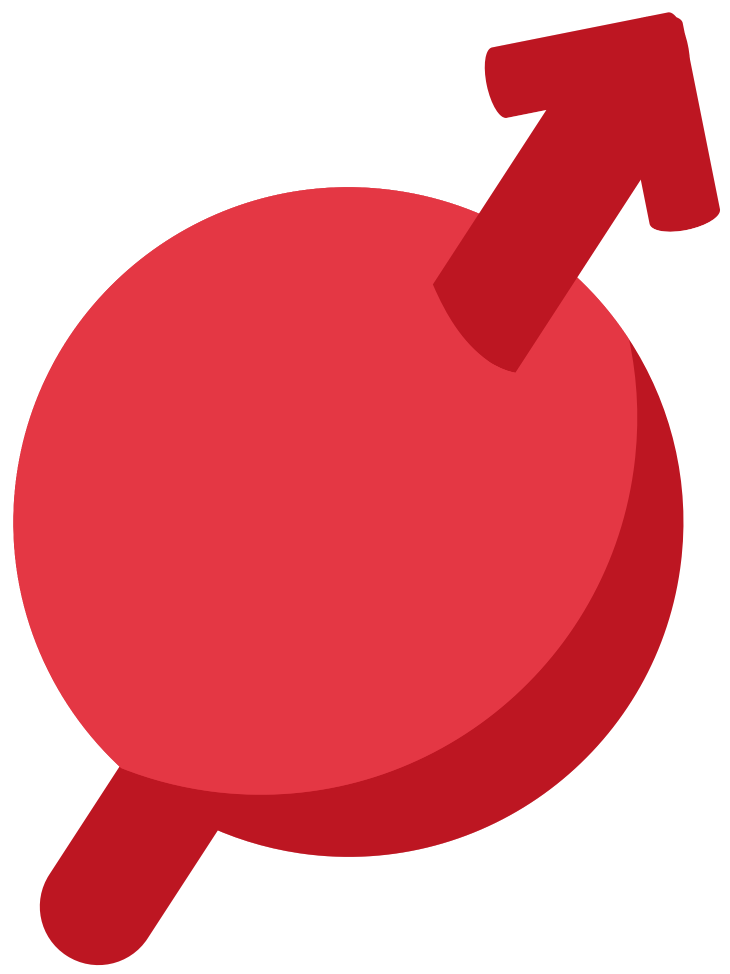
arXiv:2301.02653 (Nature *in press*)

Wang, Balembois, Billaud, Albertinale, Le Dantec, Rancic
Estève, Vion, Bertet, **Flurin**

Quantronics group / SPEC

SIR TEQ
DE LA RECHERCHE À L'INDUSTRIE cea
ANR AGENCE NATIONALE DE LA RECHERCHE
erc Starting Grant

Electron Spin Resonance



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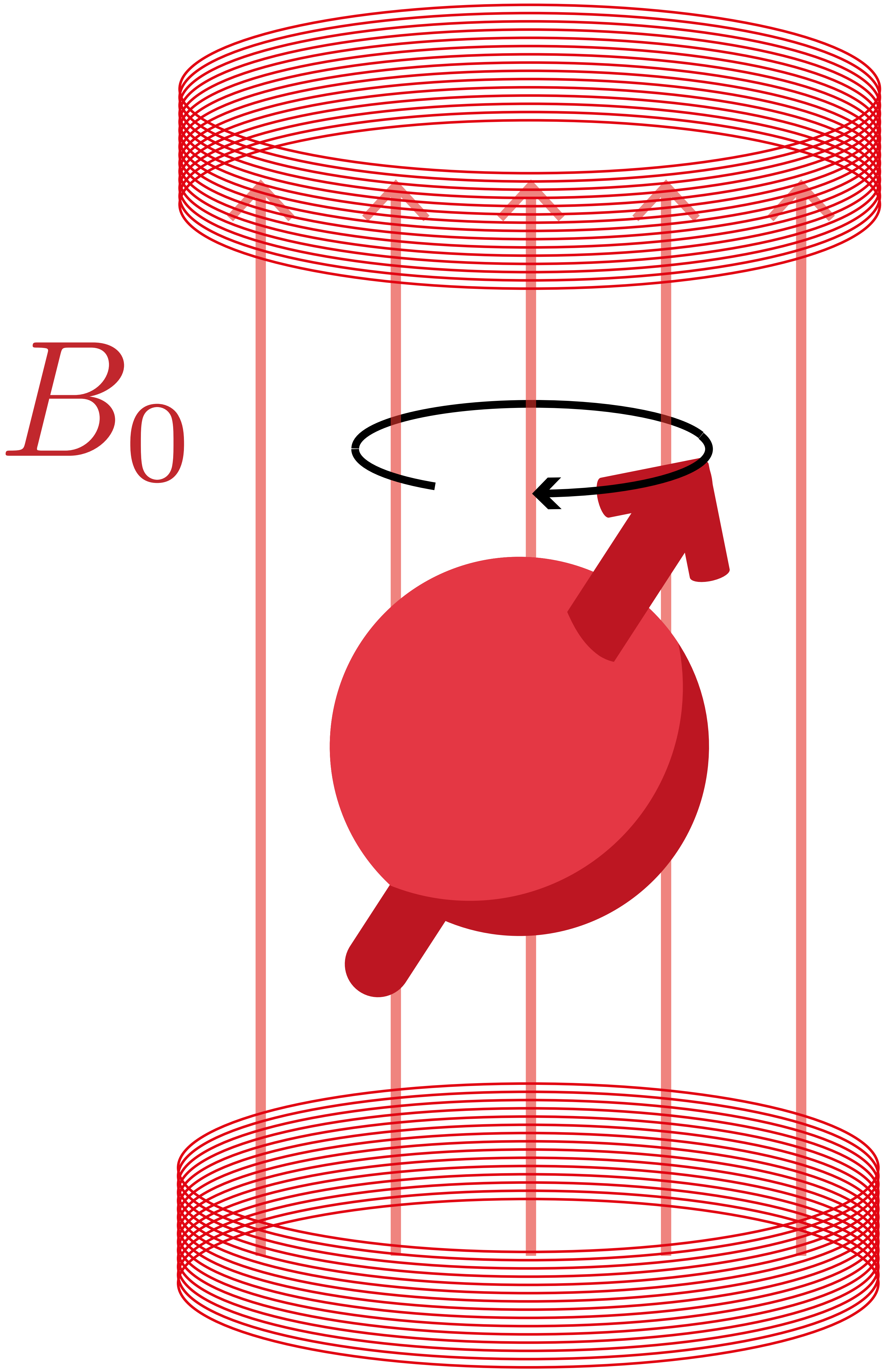

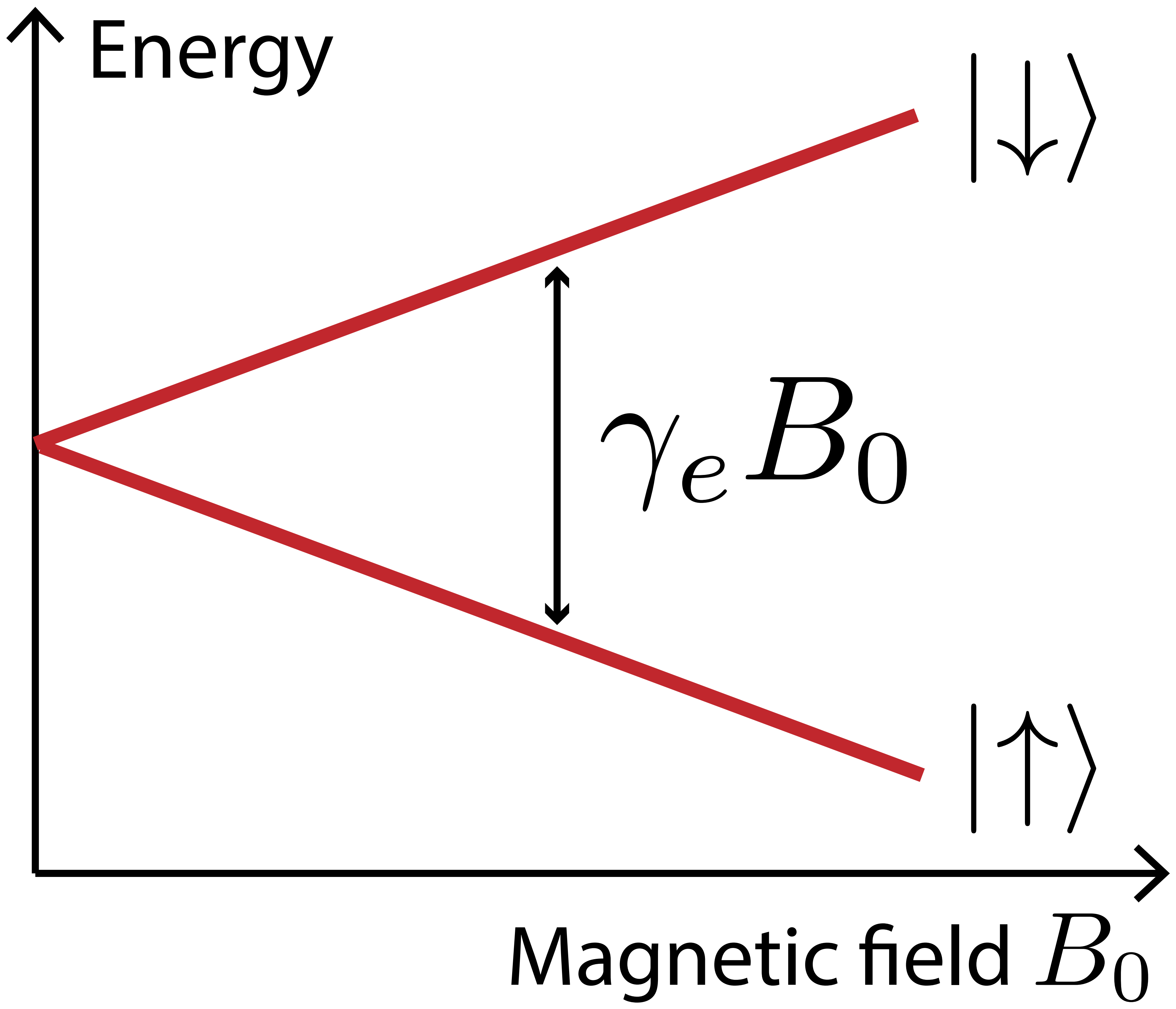

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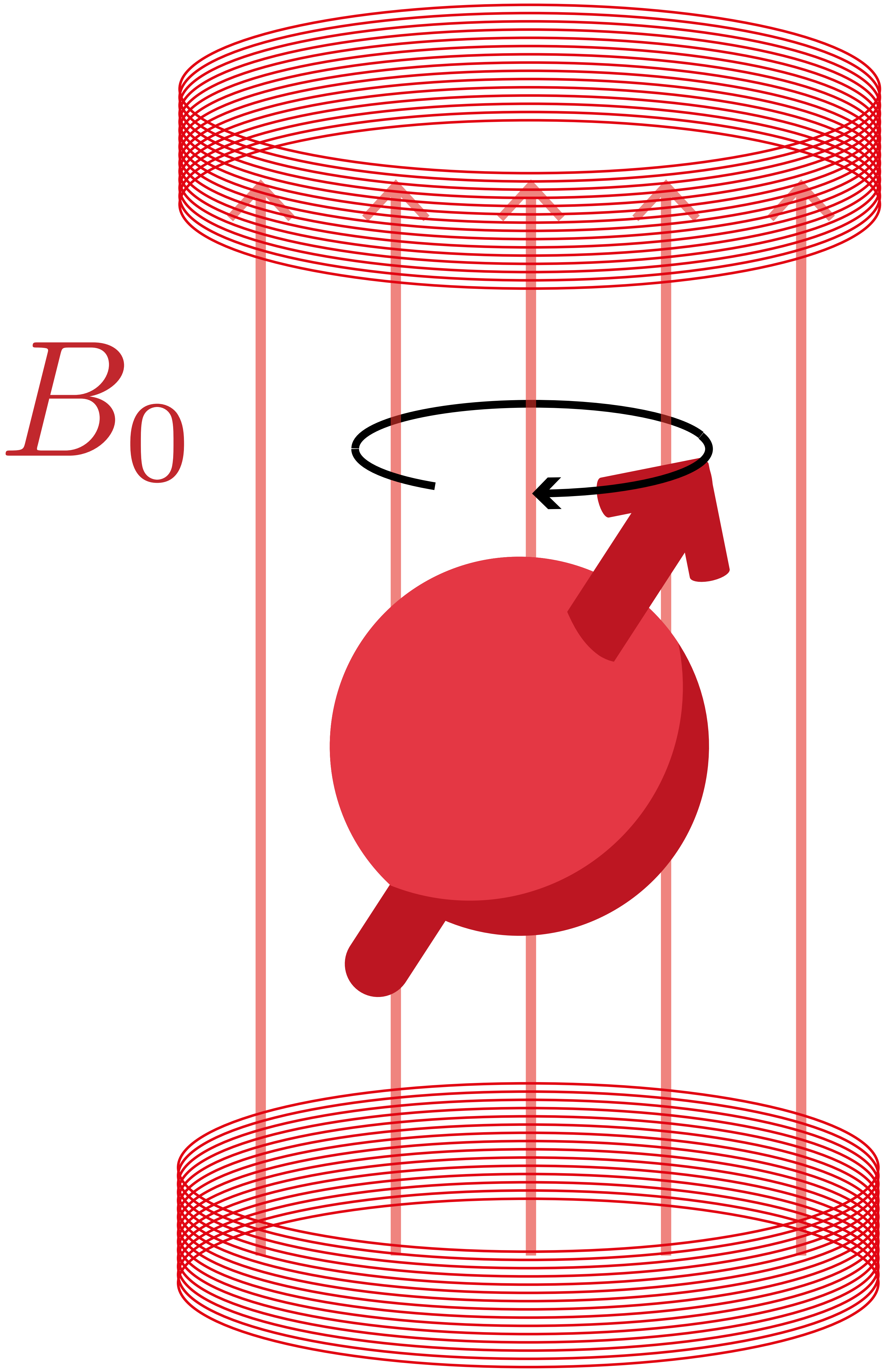
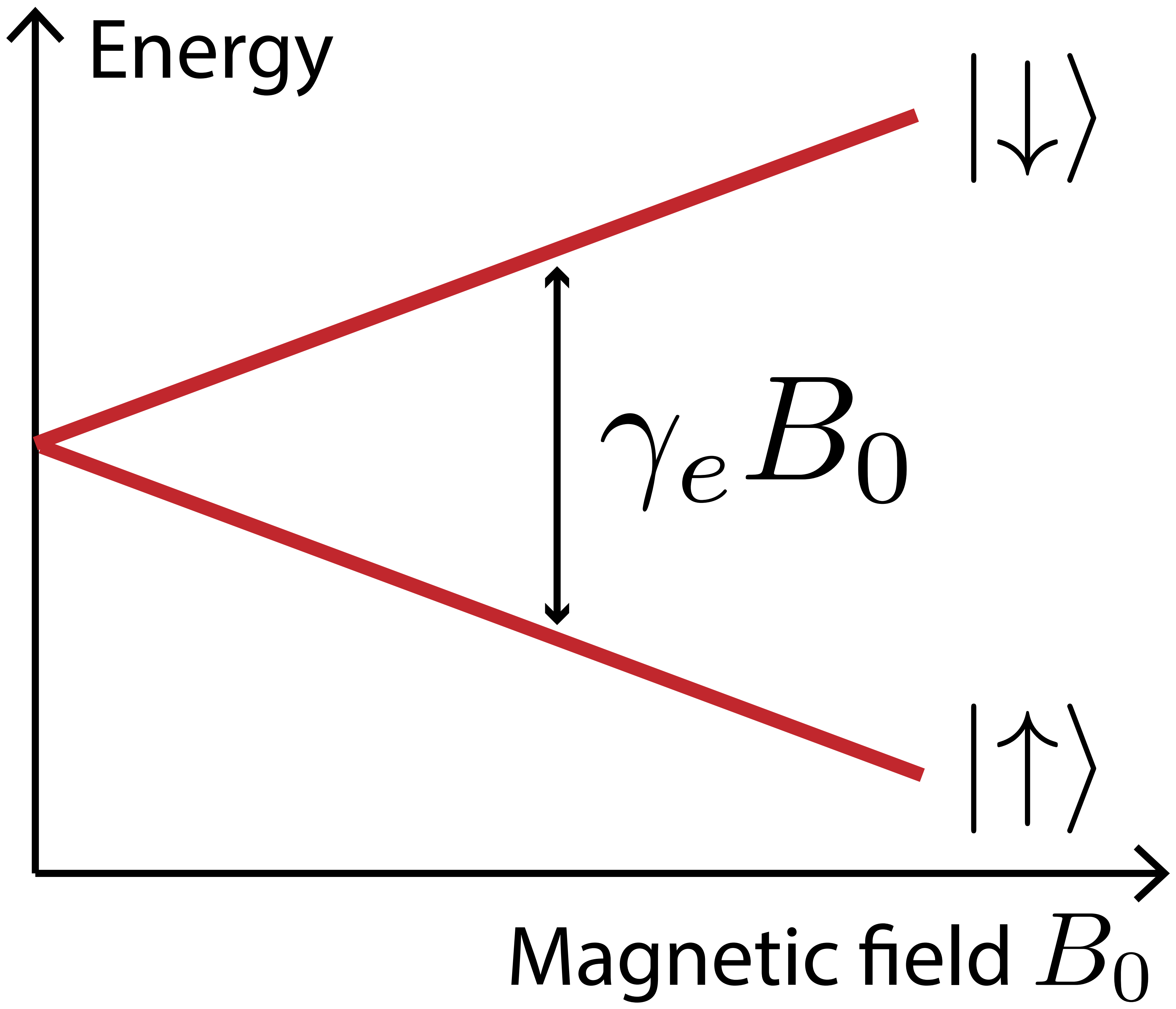
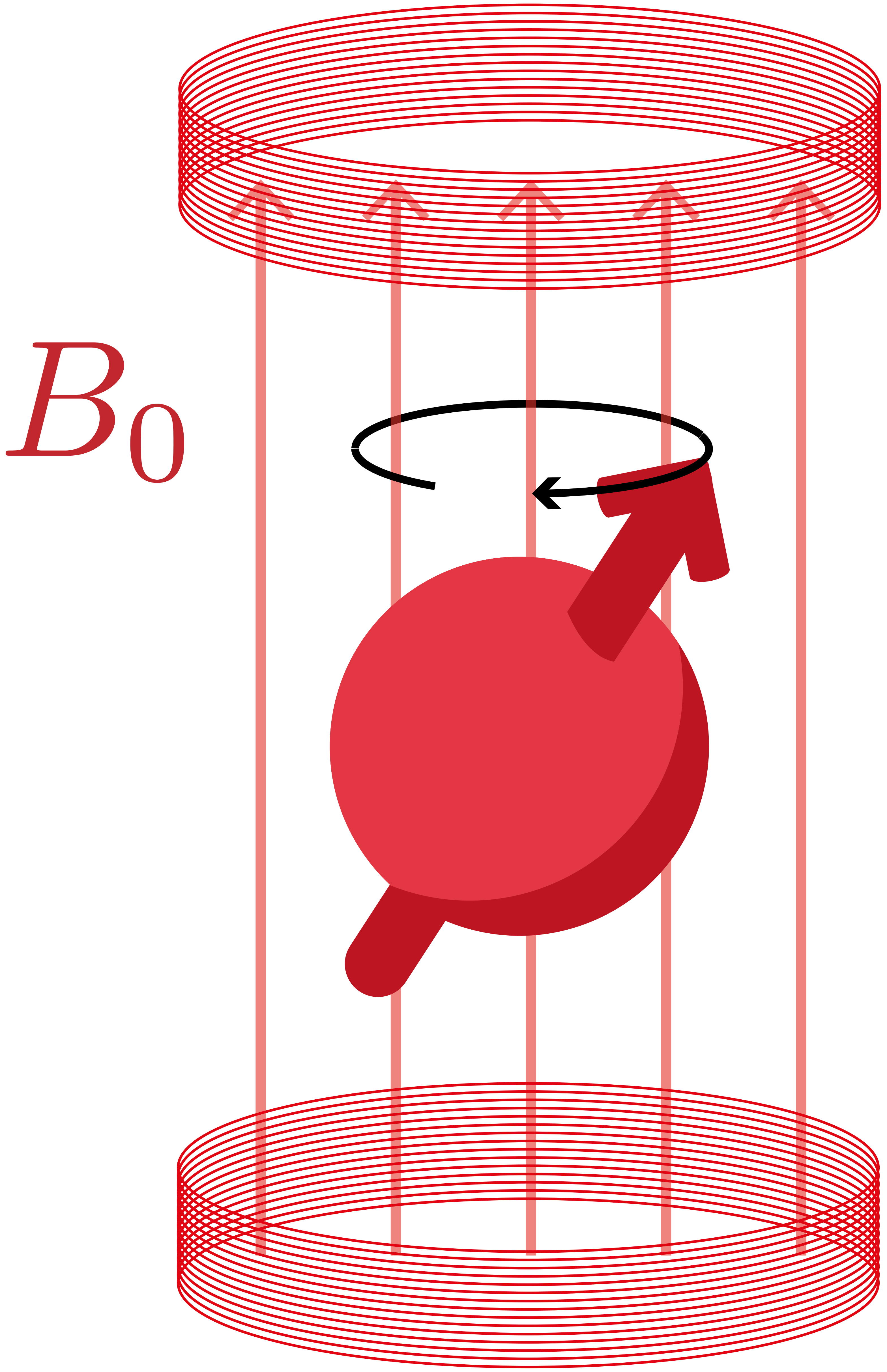


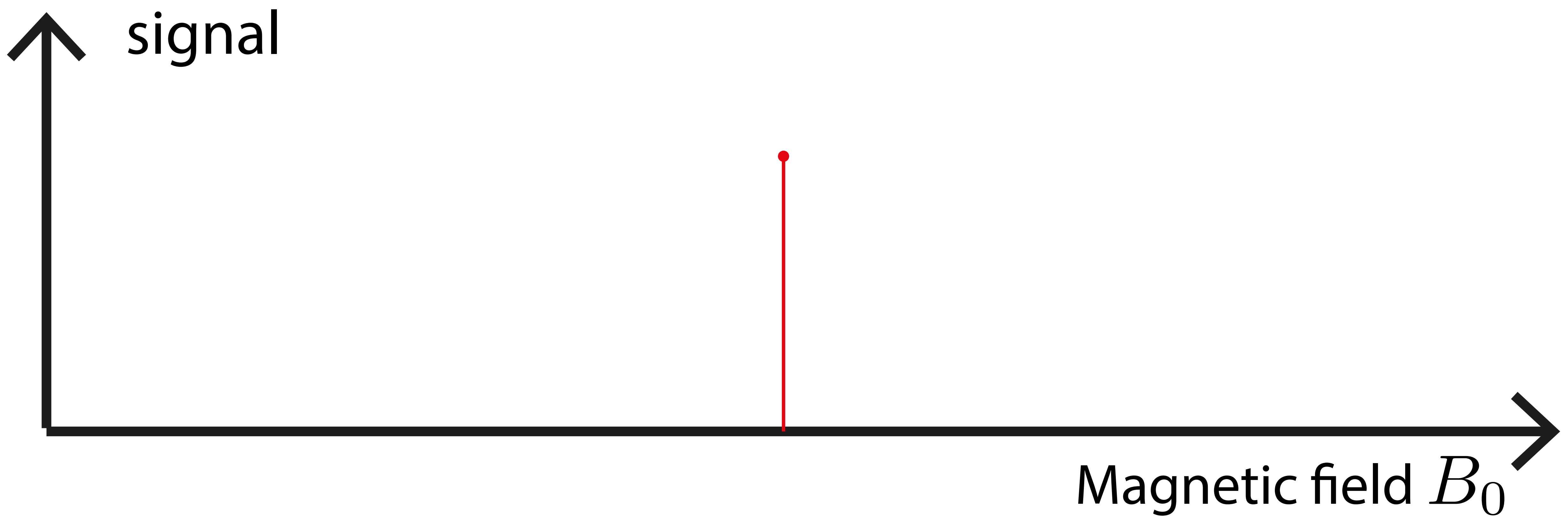


Free electron spin

$$\gamma_e = 28.0249514242(85)\ \mathrm{GHz/T}$$

Electron Spin Resonance





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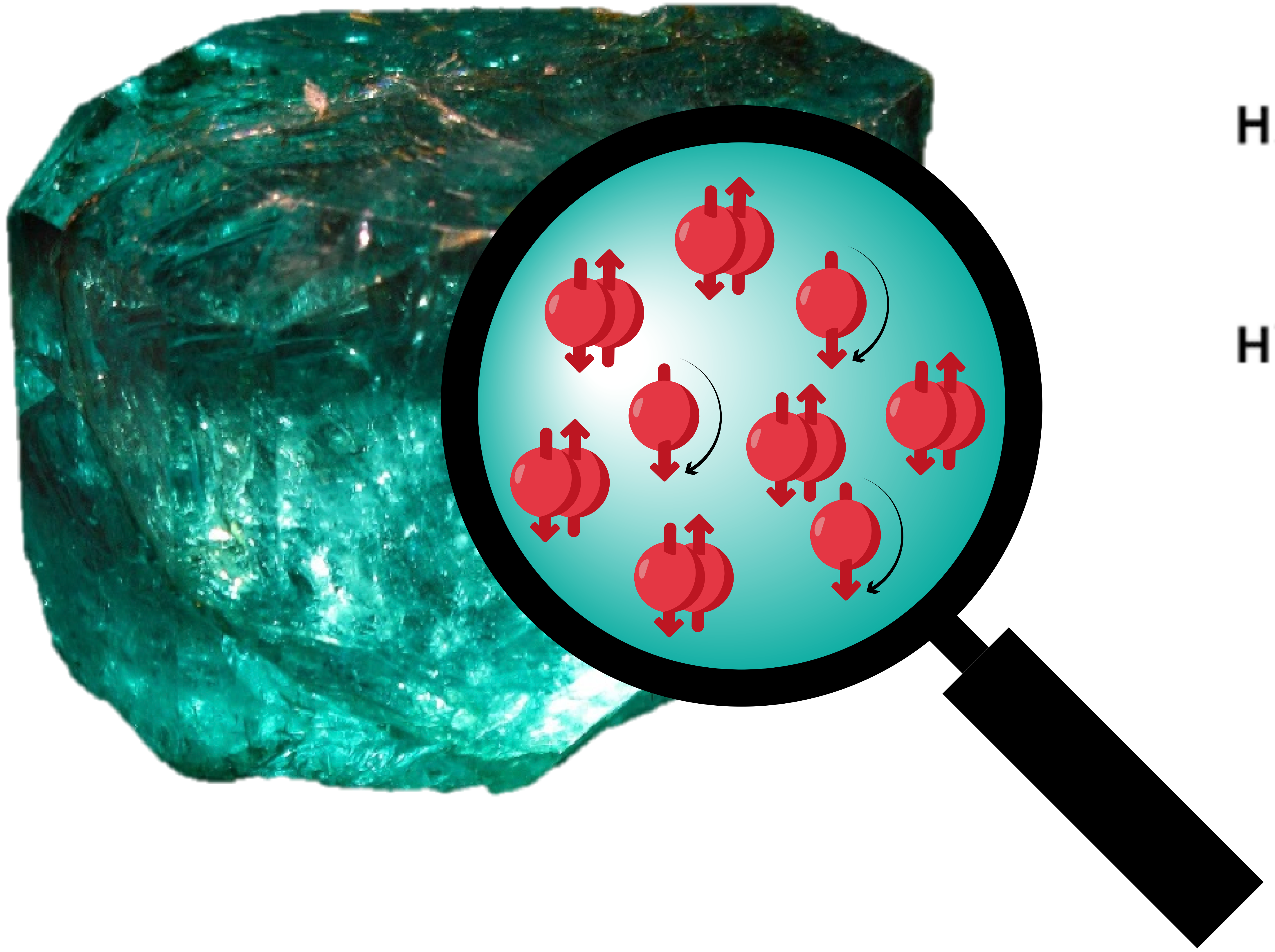
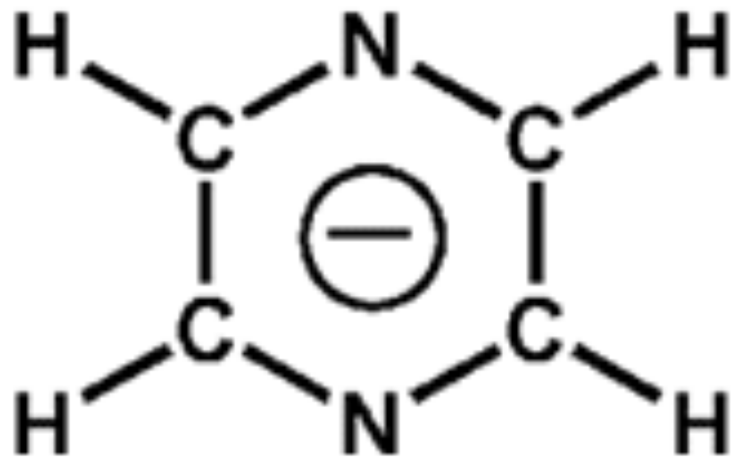
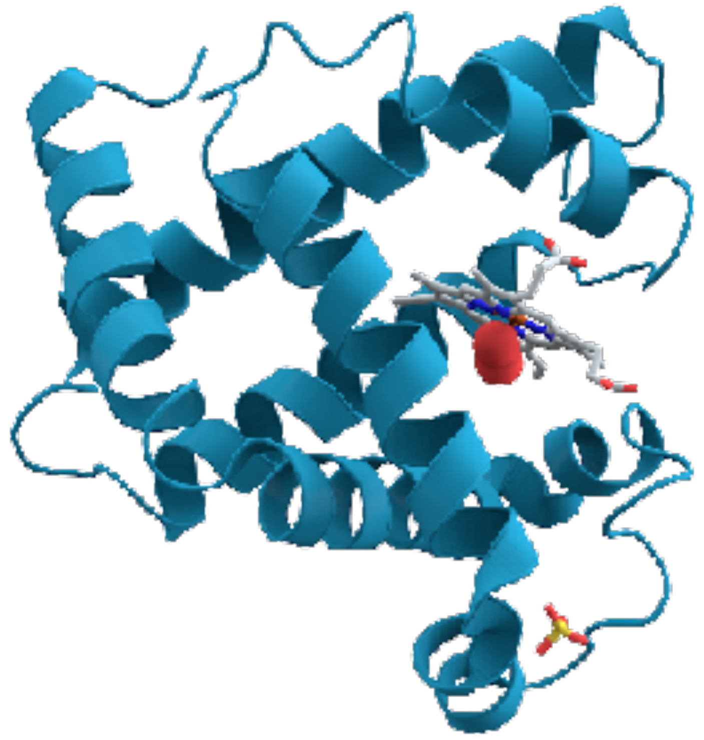
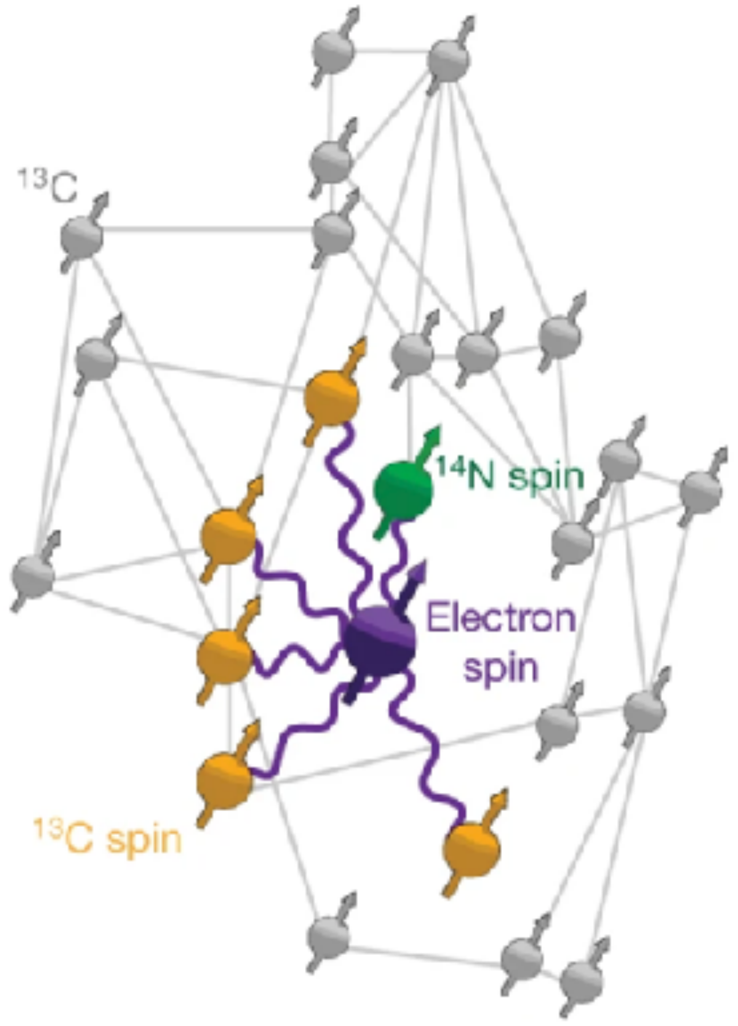




Chemistry

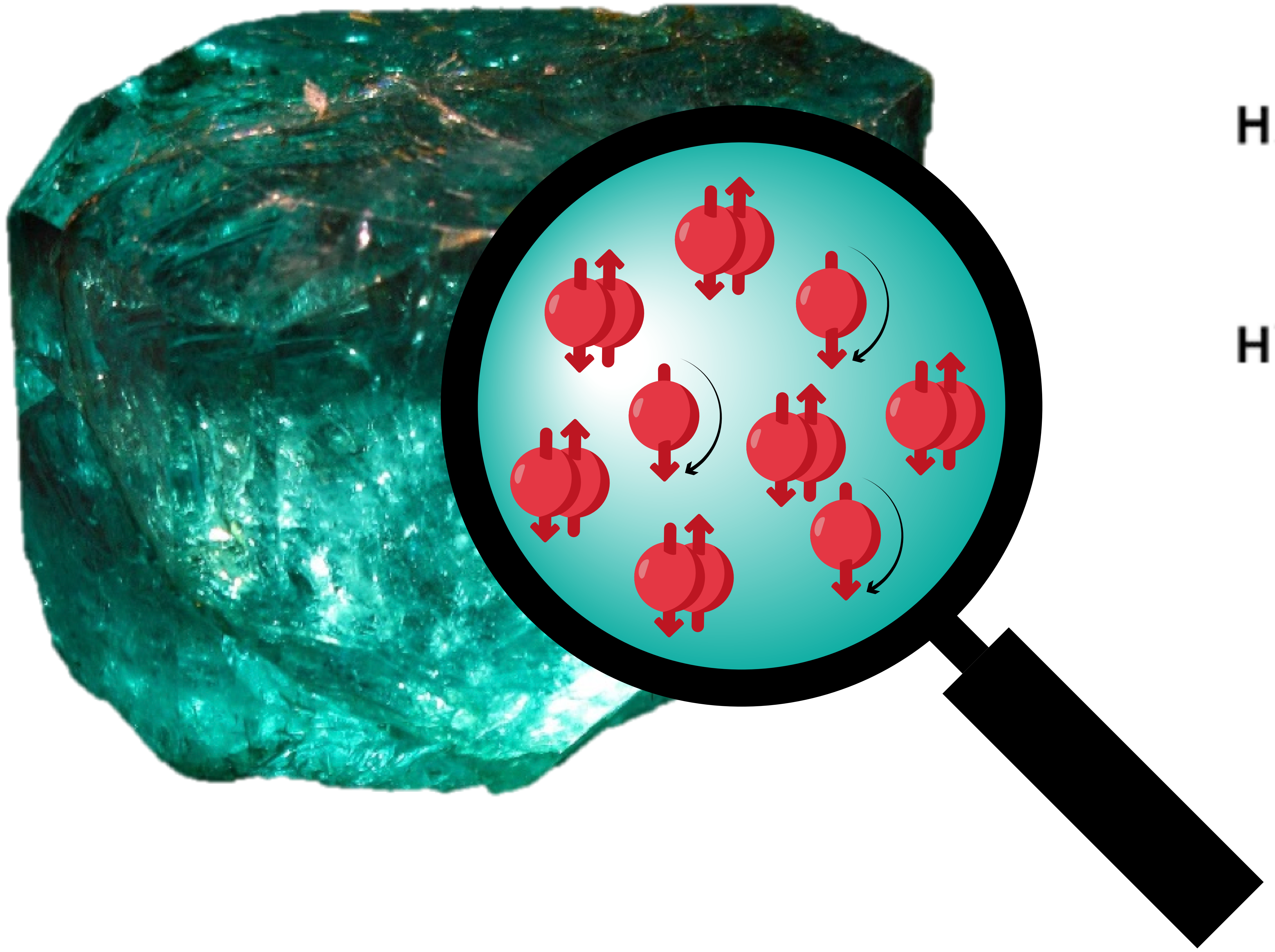


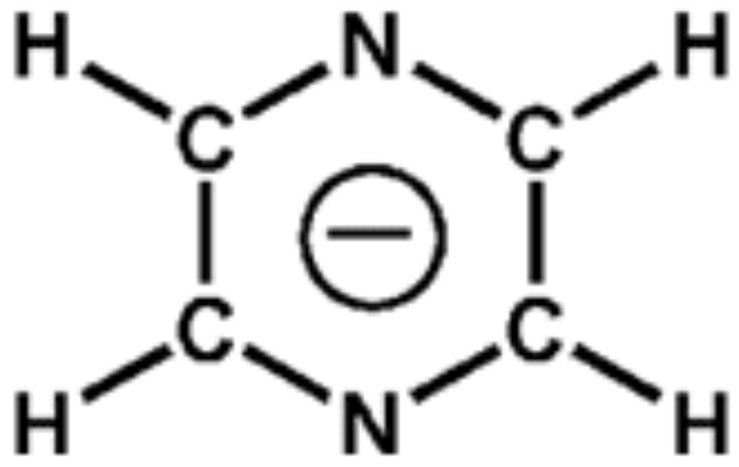
Biology



Quantum Computing

Electron Spin Resonance





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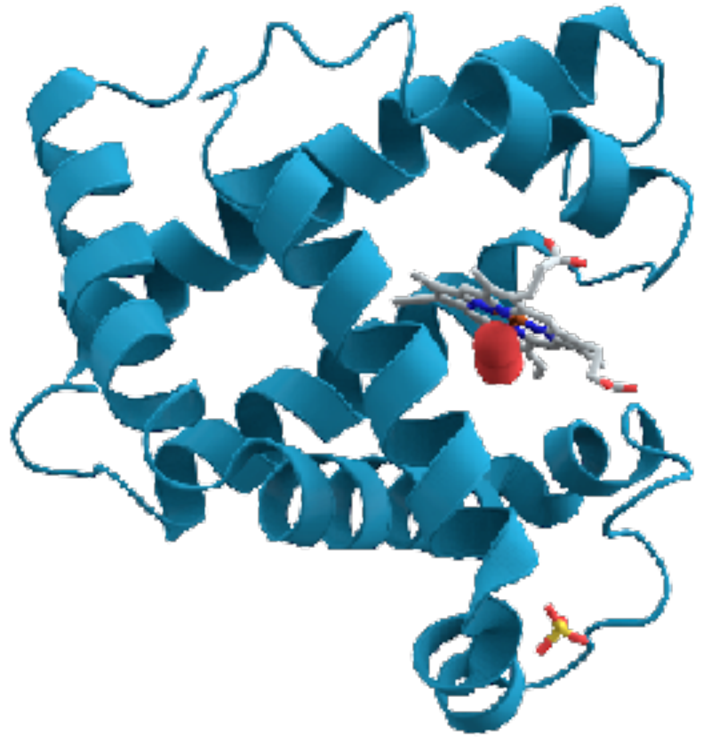

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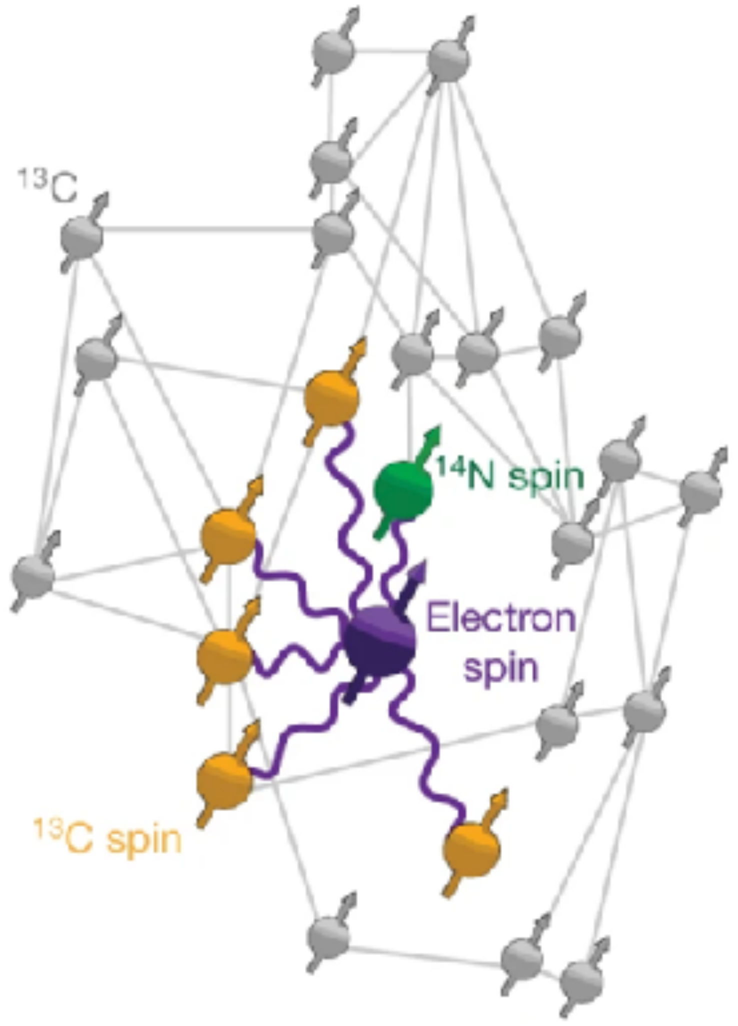

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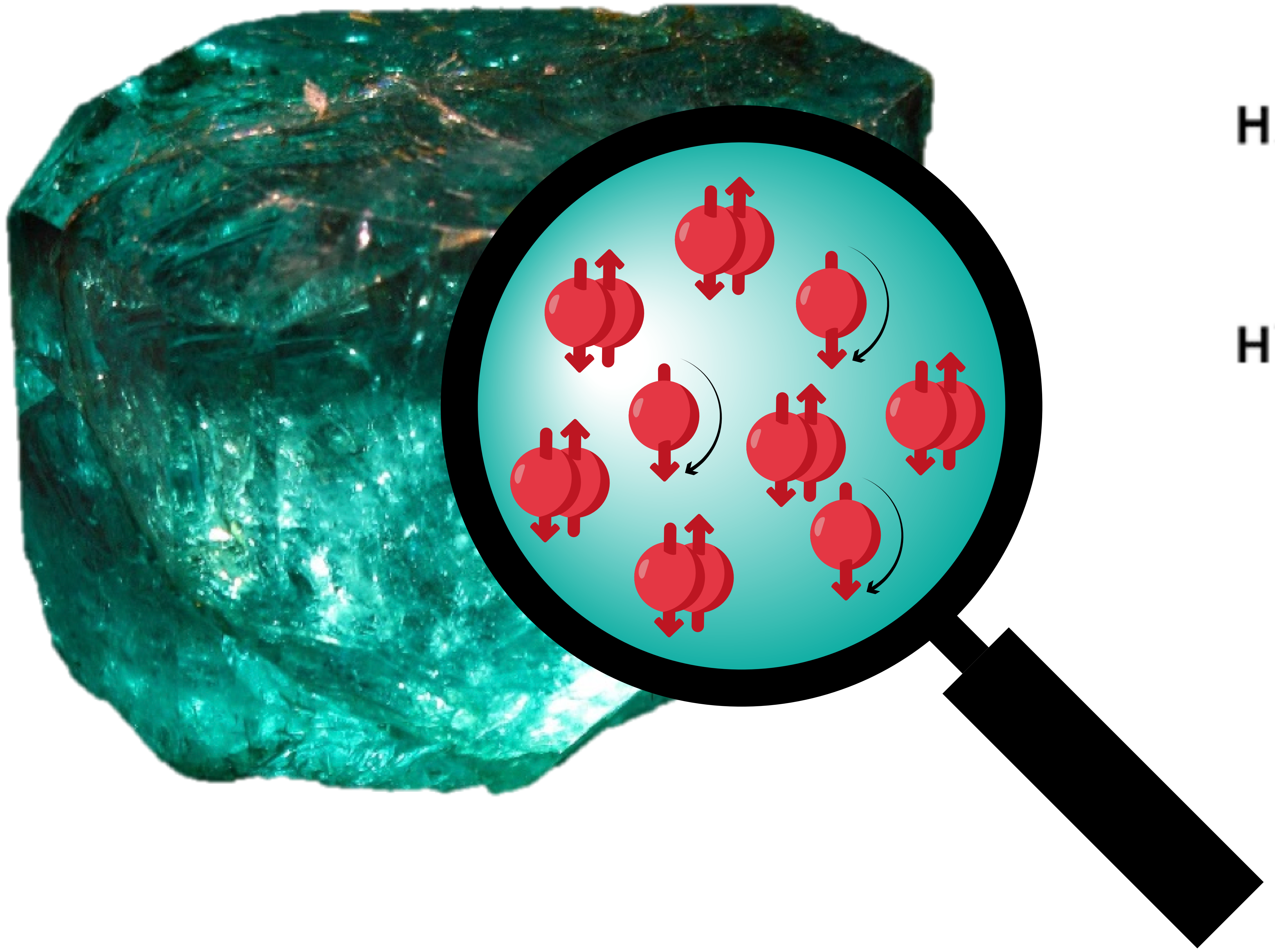

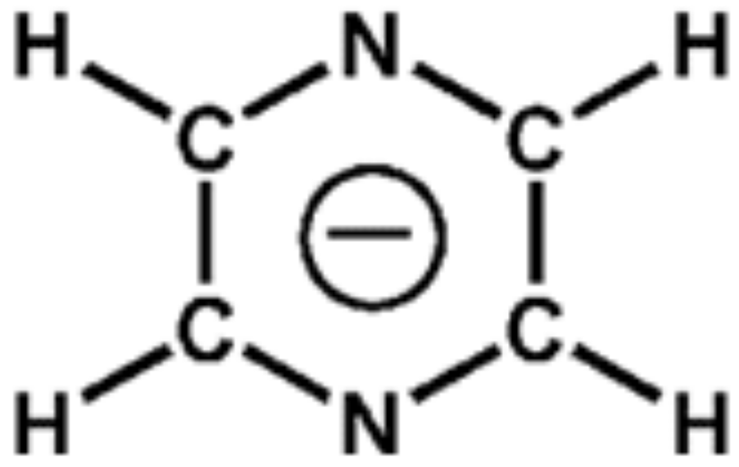

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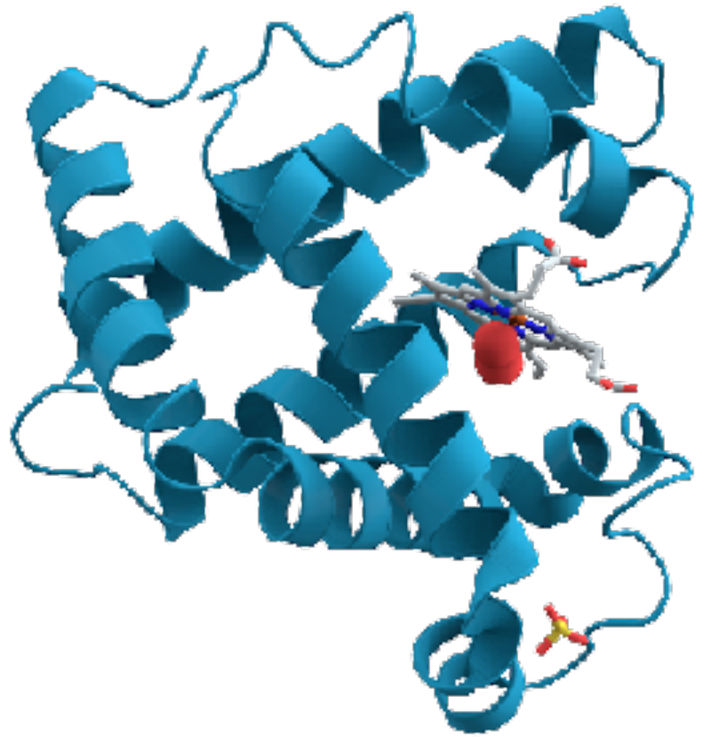

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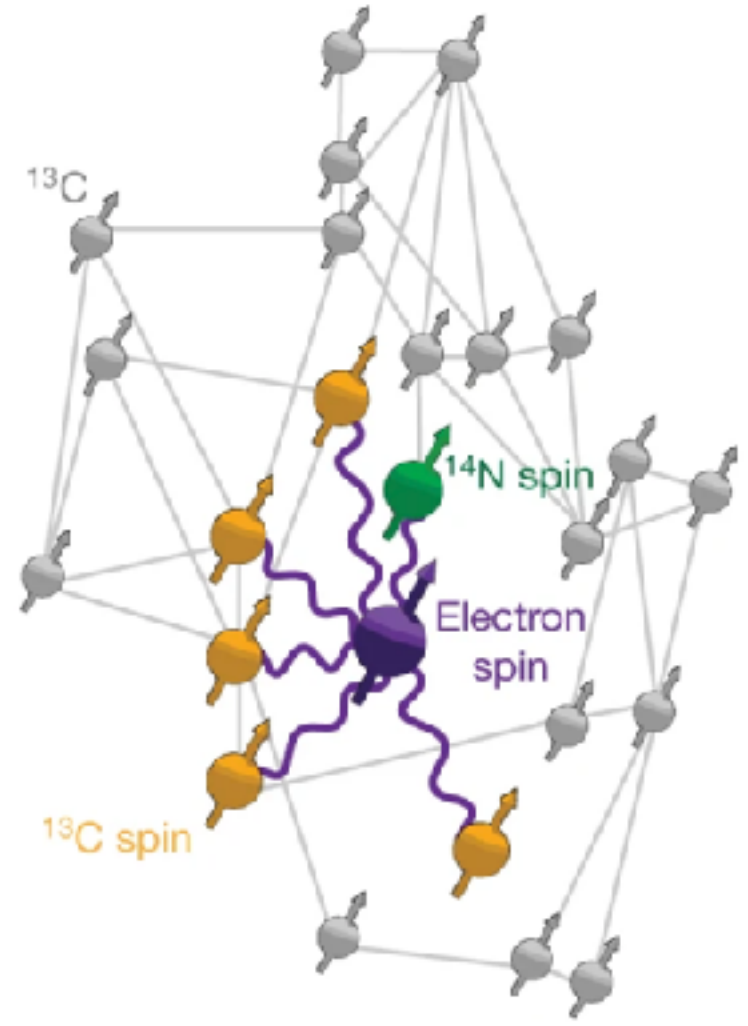
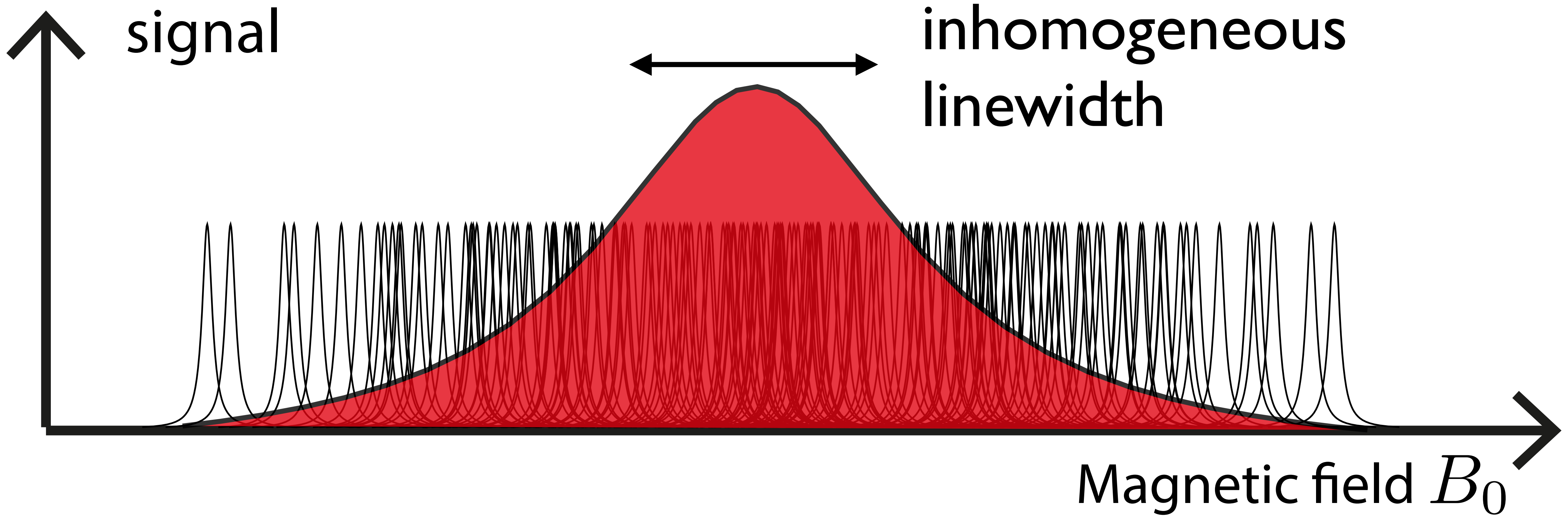


Quantum Computing



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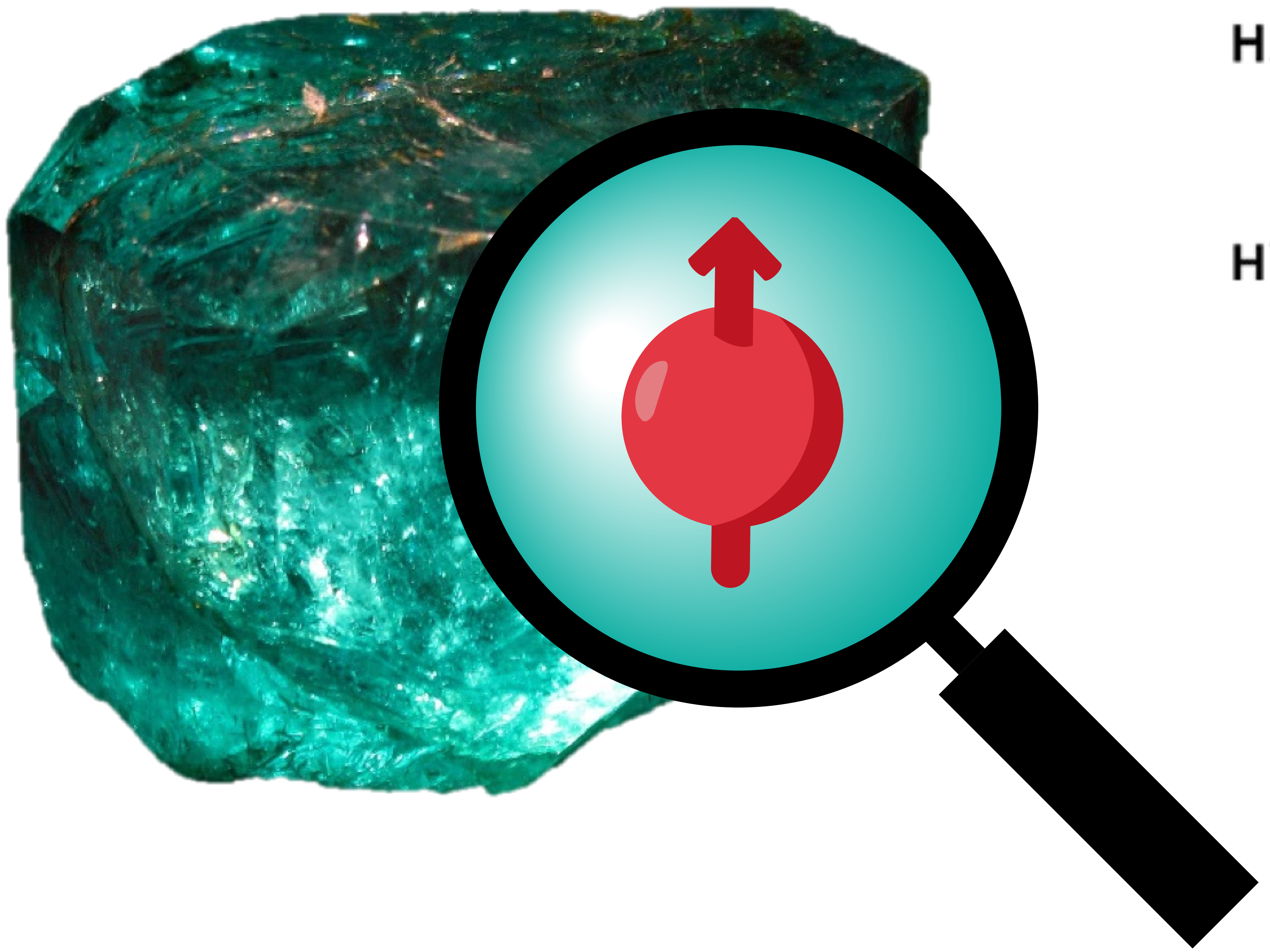

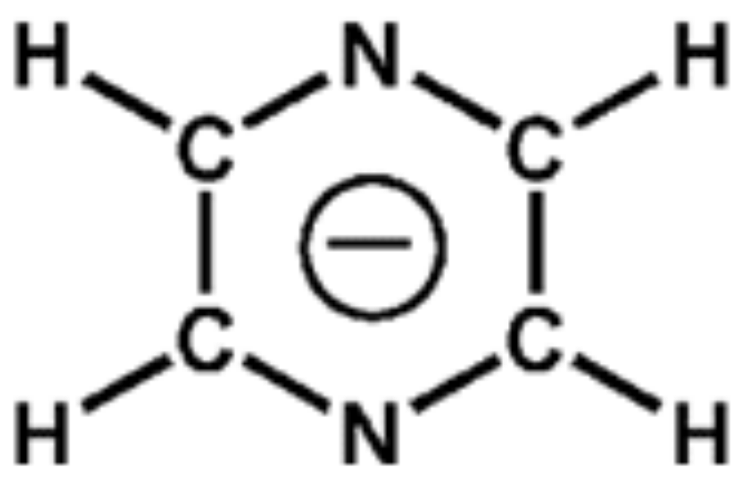

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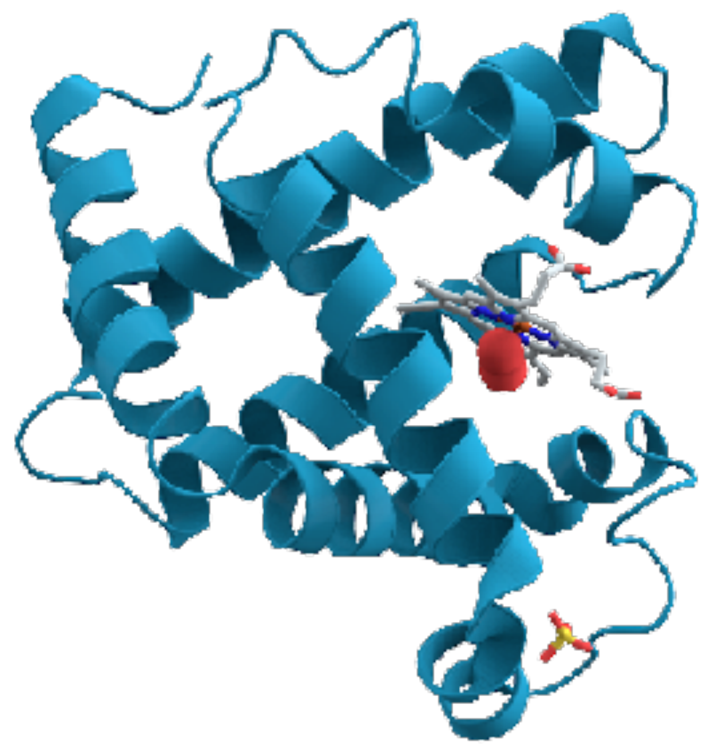

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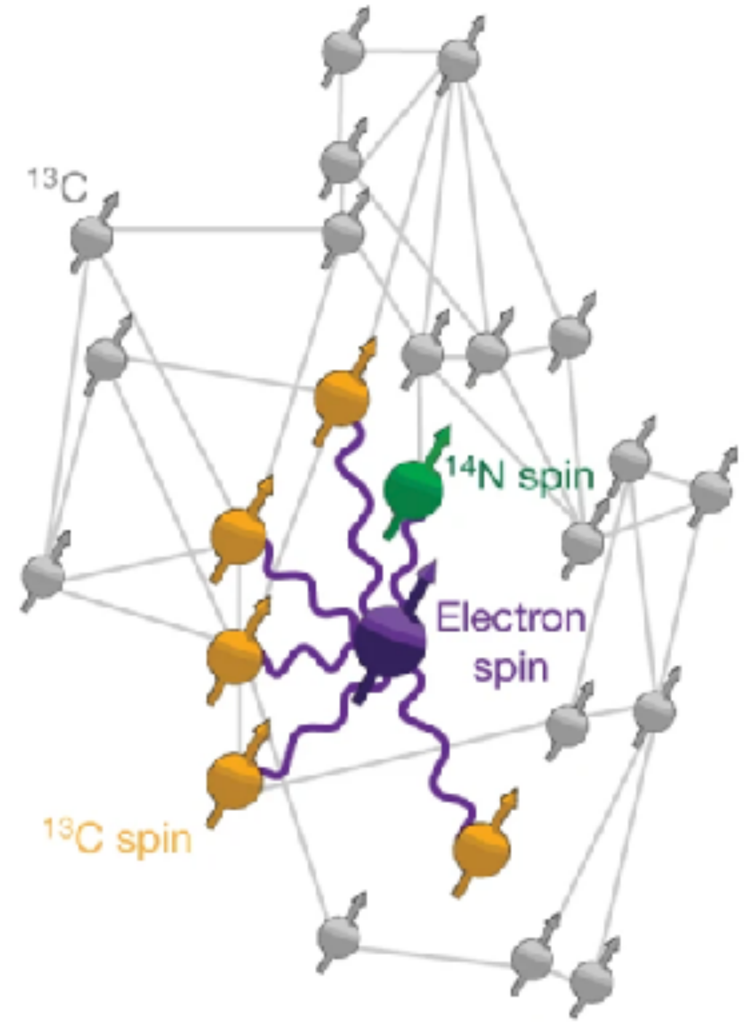
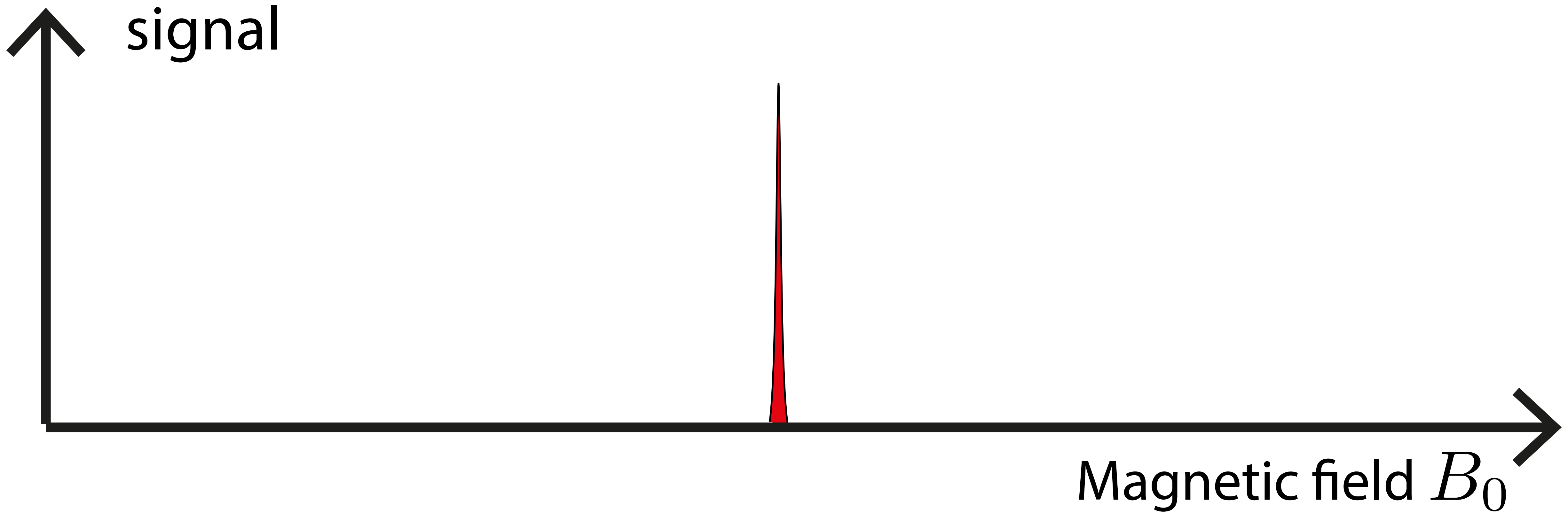


Quantum Computing

$$1\ \mathrm{spin}/\sqrt{\mathrm{Hz}}\ ?$$



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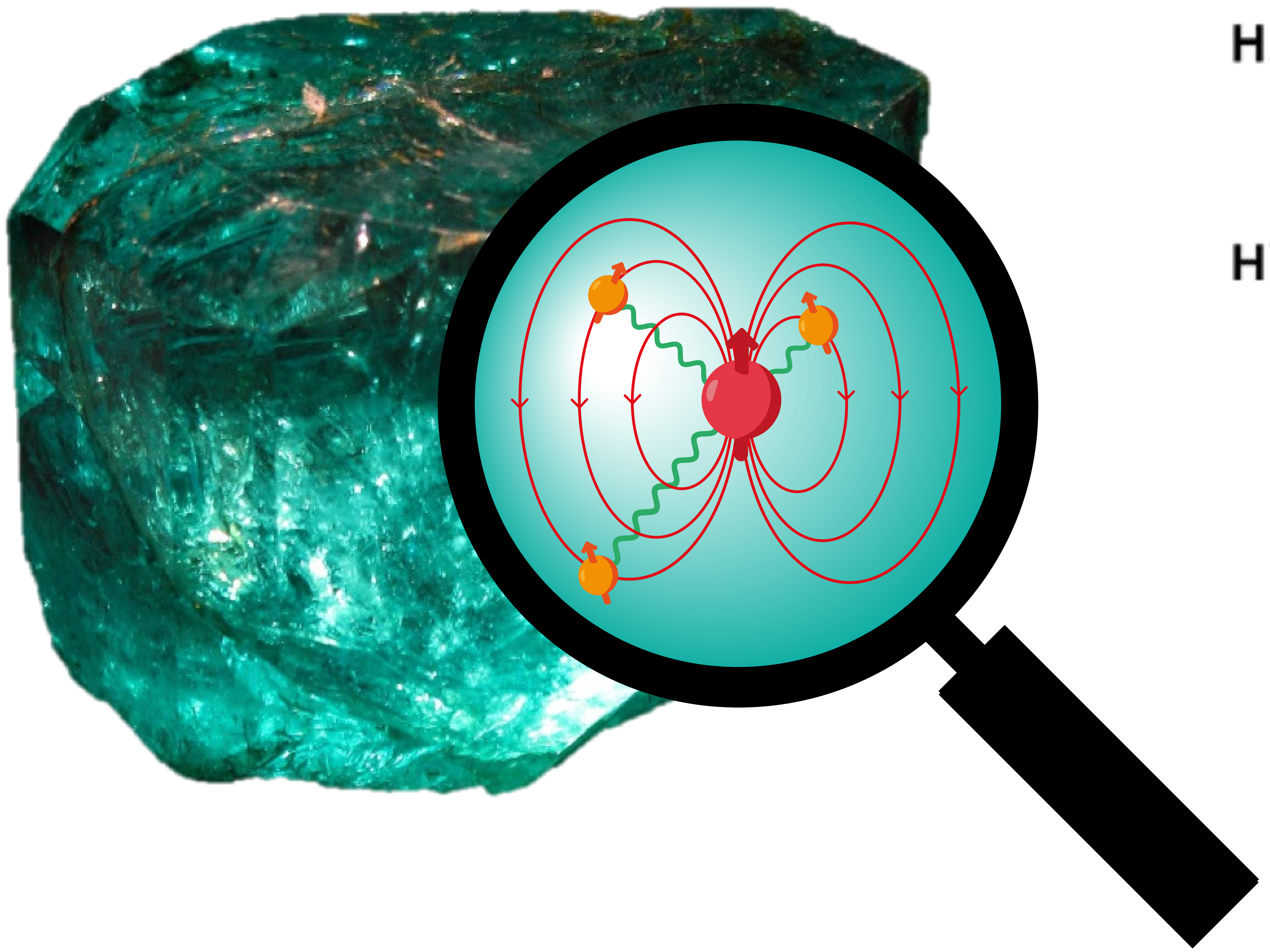

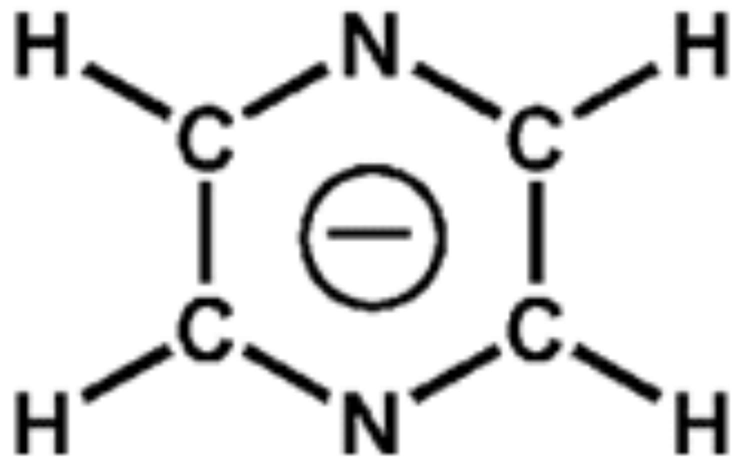

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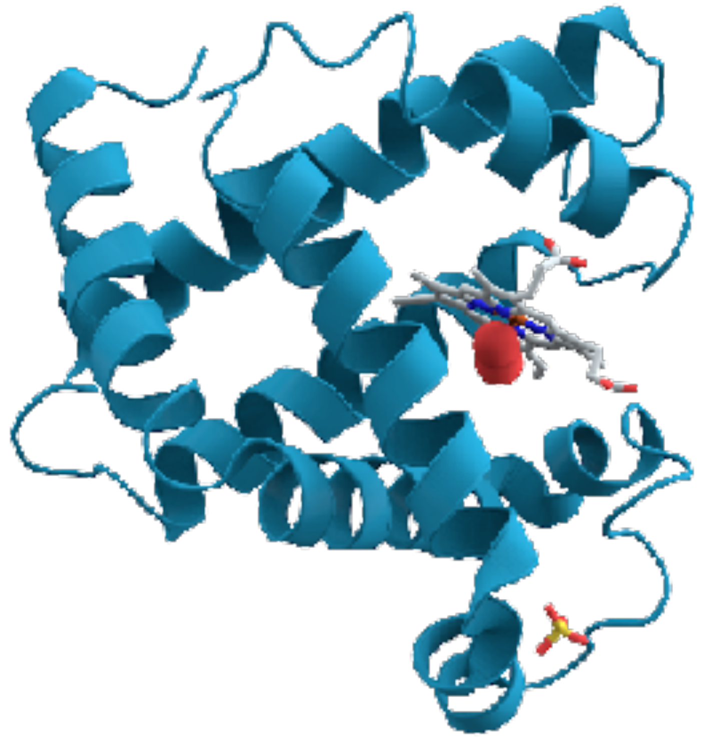
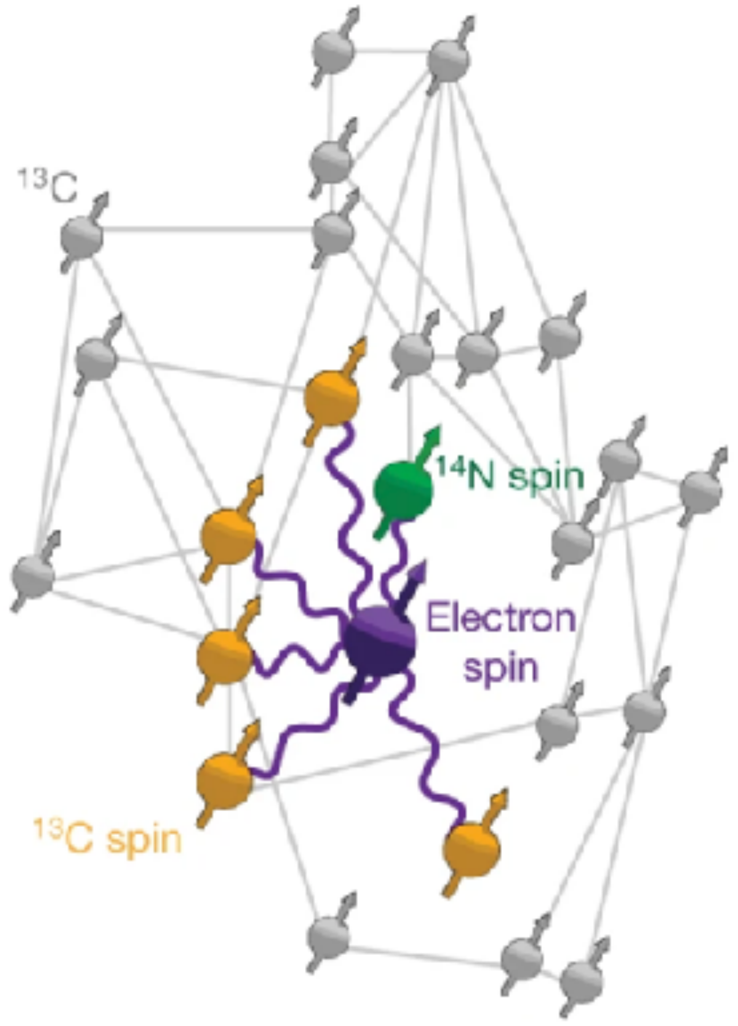


Biology



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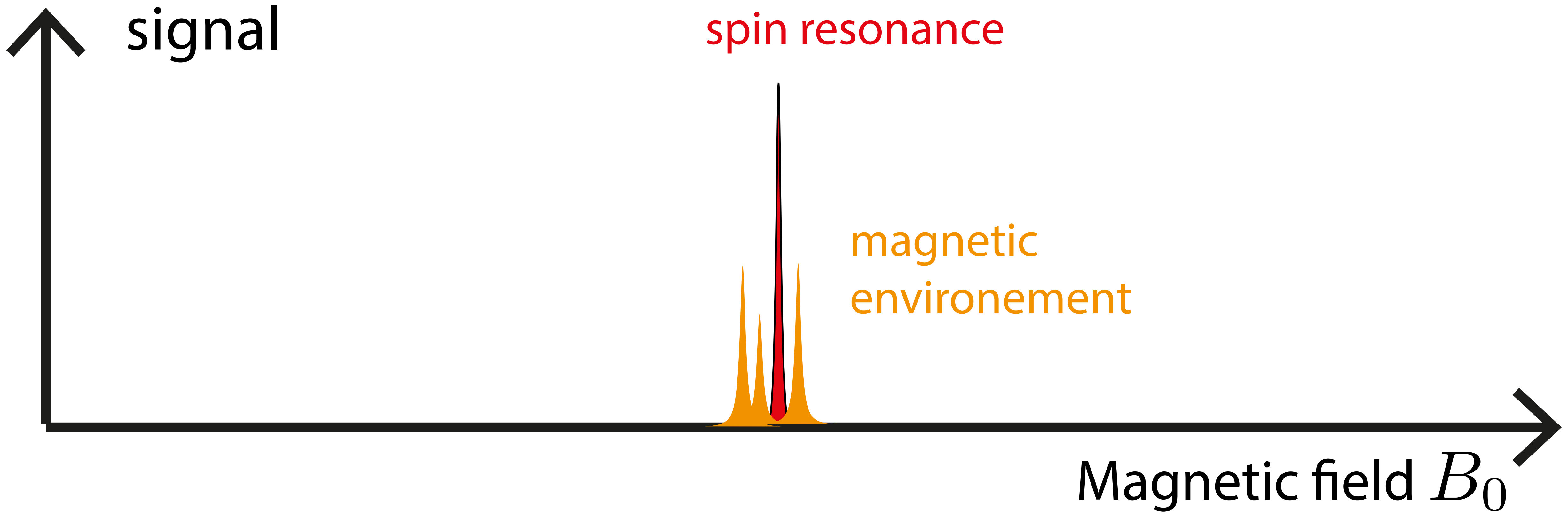

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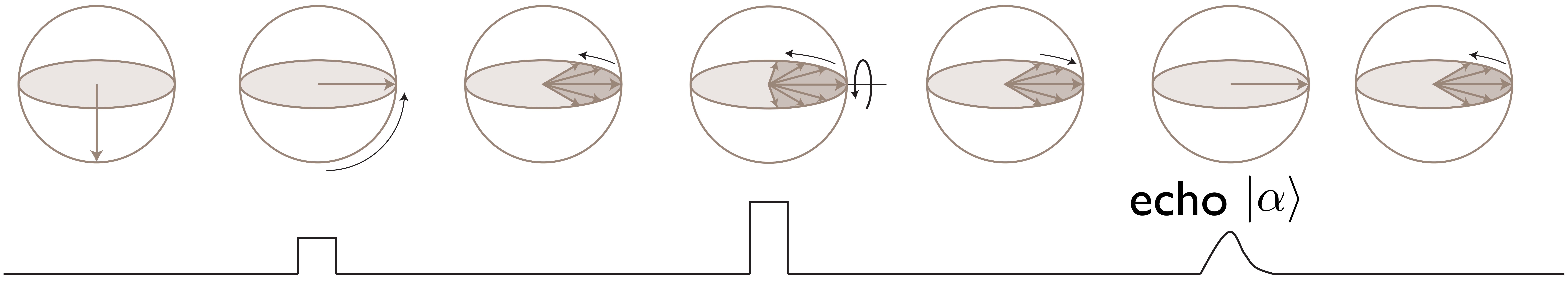
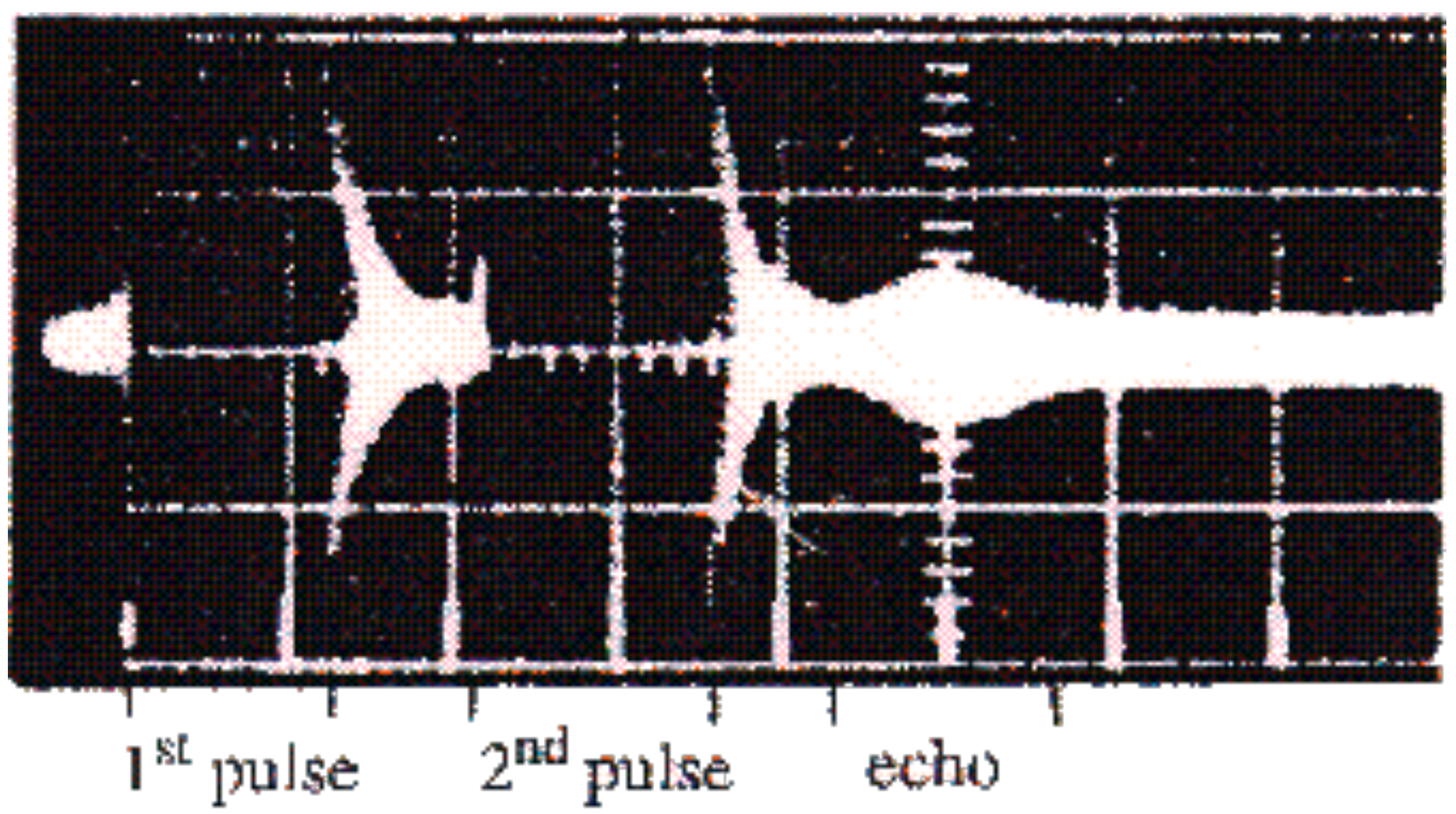




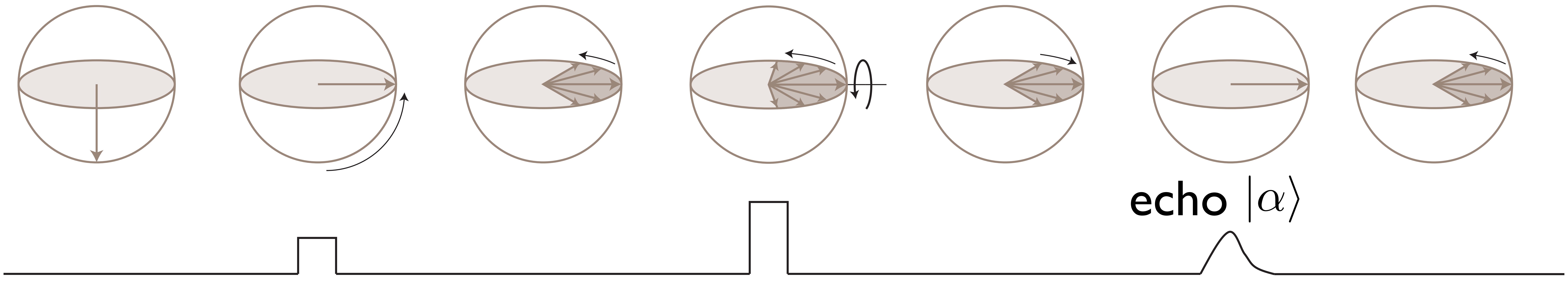
R.J. Blume, "Electron spin relaxation times in sodium- ammonia solutions", *Phys. Rev.* **109**, 1867 (1958).

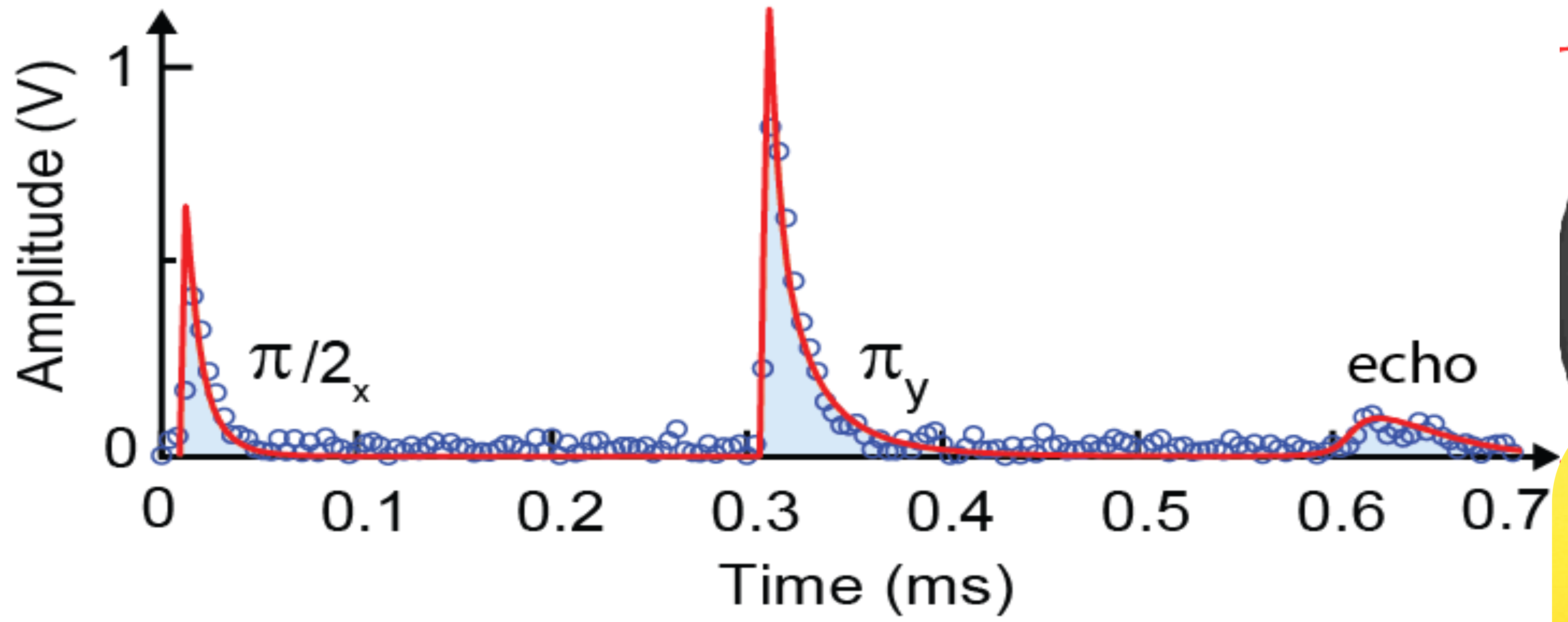
A. Schweiger and G. Jeschke, « Principles of pulse electron paramagnetic resonance » (Oxford University Press, 2001).

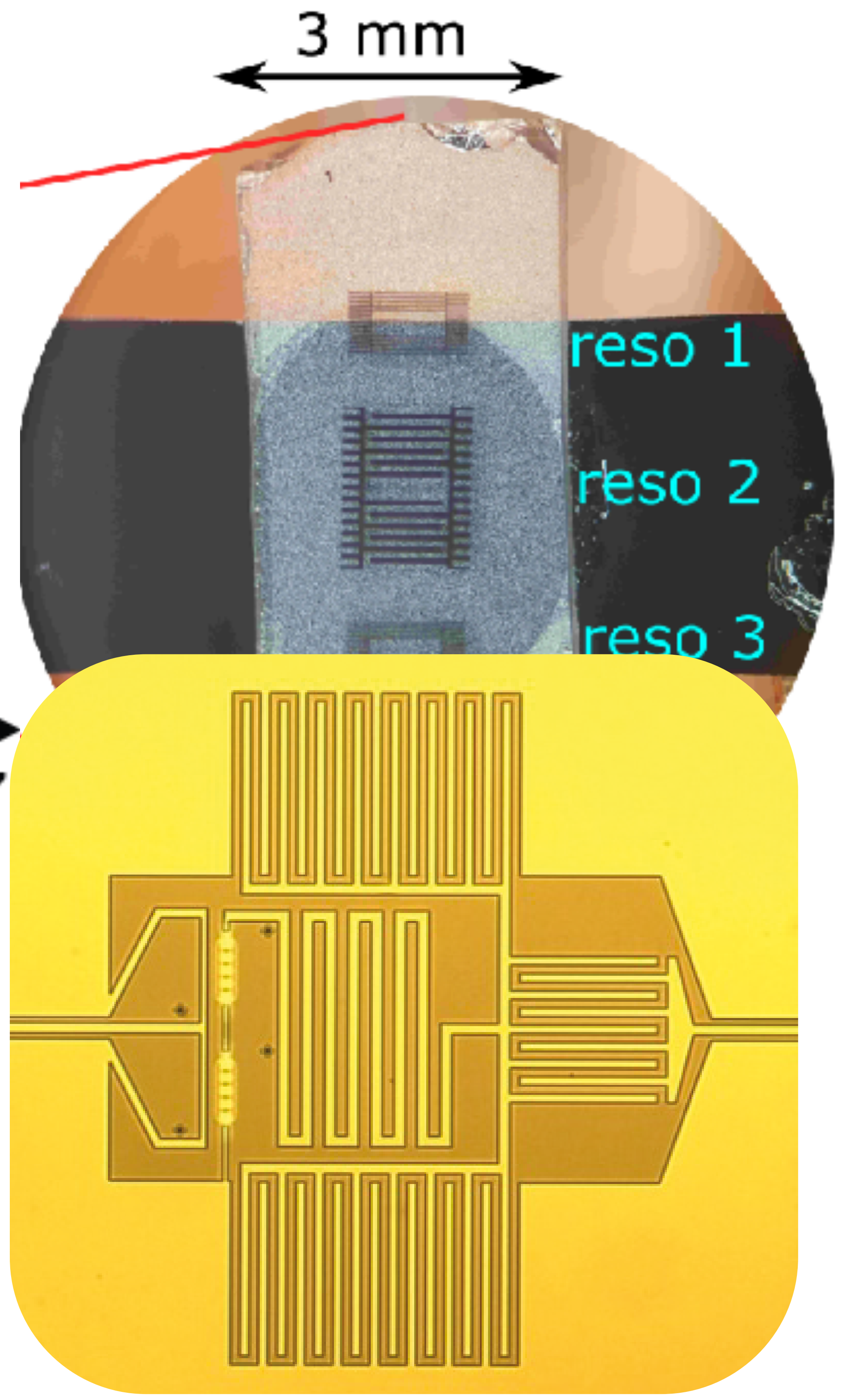
Electron Spin Resonance

10^{13} spins$/\sqrt{\text{Hz}}$

Electron Spin Resonance



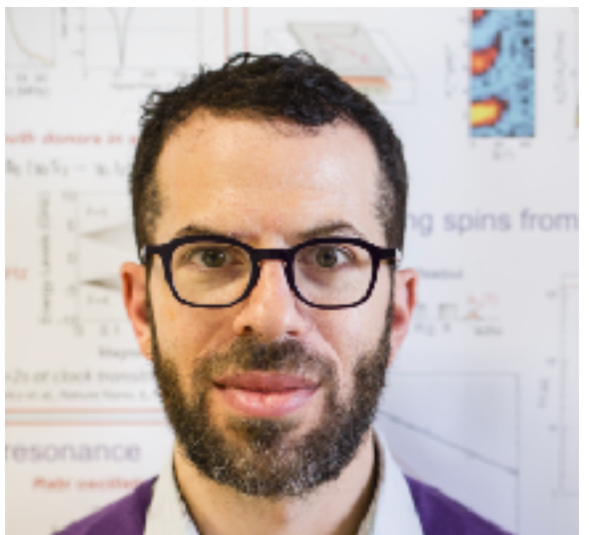




Superconducting Coupling Resonator & Quantum Limited Amplifier

$$100 \text{ spins}/\sqrt{\text{Hz}}$$

[Ranjan et al. Appl. Phys. Lett. **116**, 184002 (2020)]

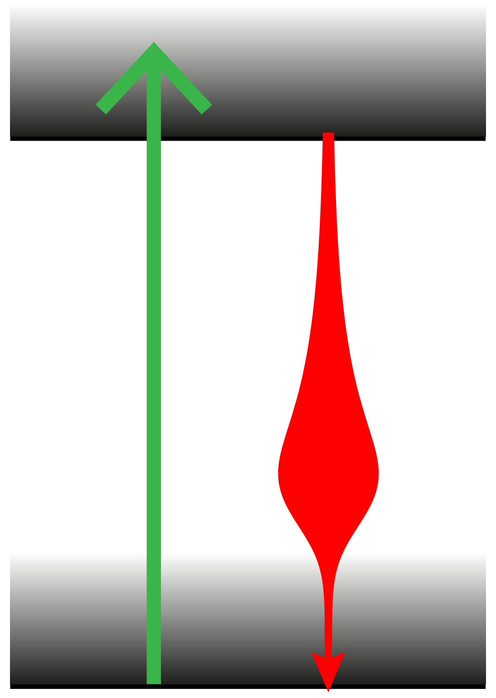


P. Bertet

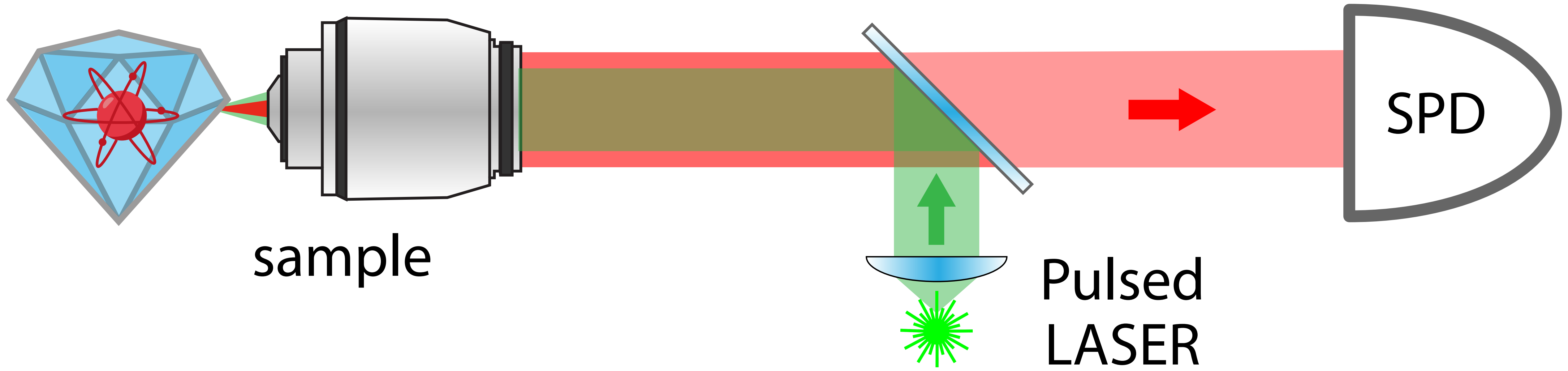
Fluorescence At Microwave Frequency

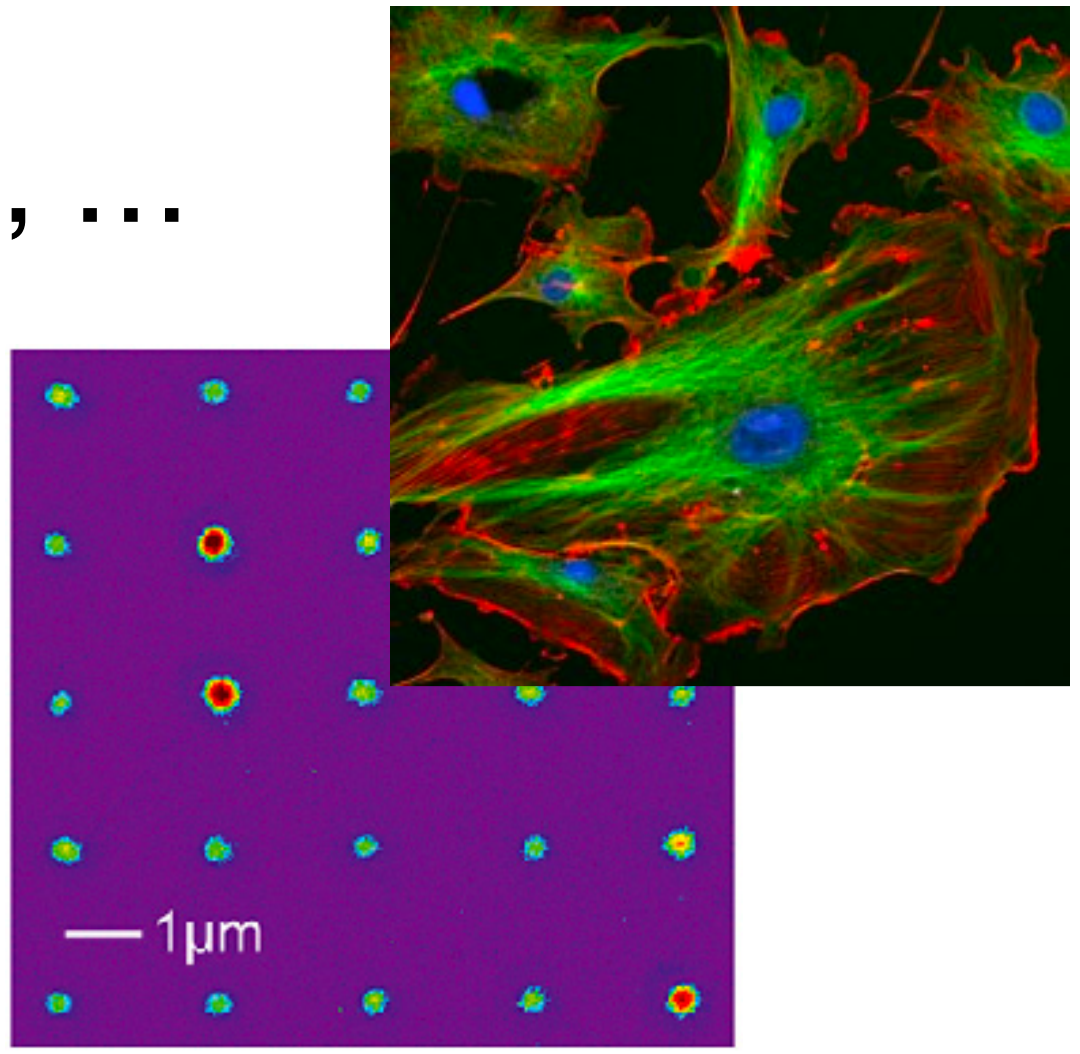
Fluorescence detection is ubiquitous in optics: atoms, molecules, NVs, …

700 THz



$\Gamma_r \sim 10^8$ s





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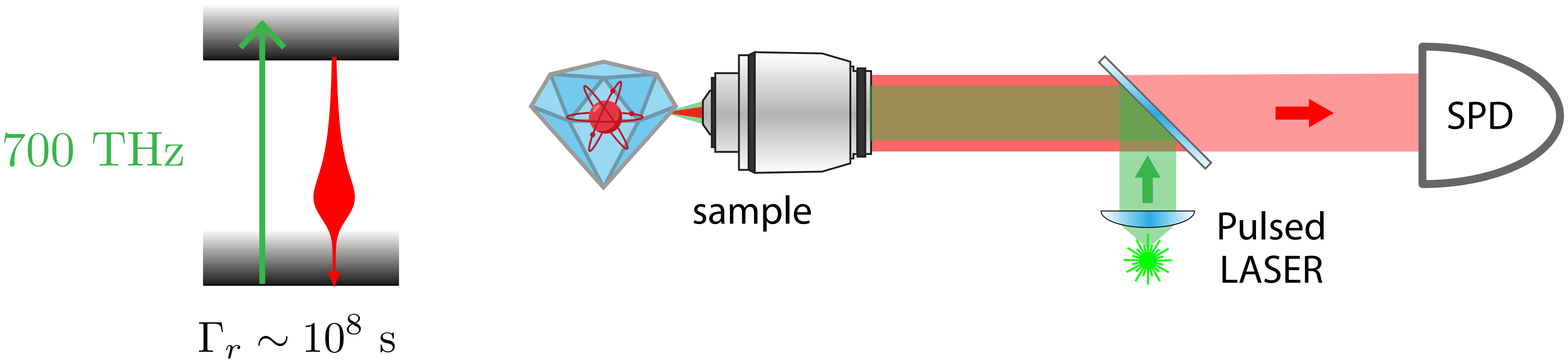
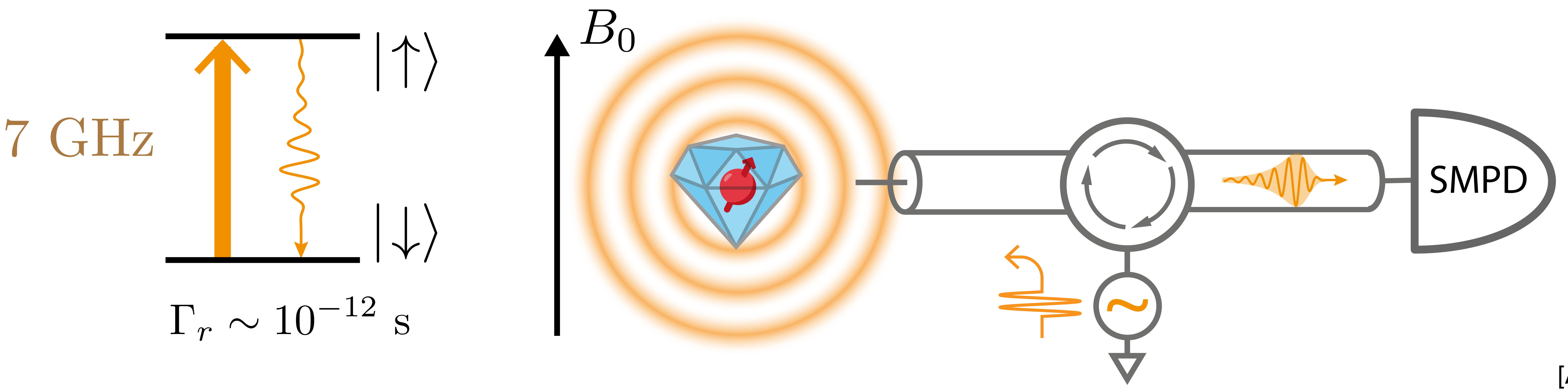


Can we detect spins by their microwave (MW) fluorescence ?



[Albertinale et al. Nature **600**, 434 (2021)]

Fluorescence At Microwave Frequency

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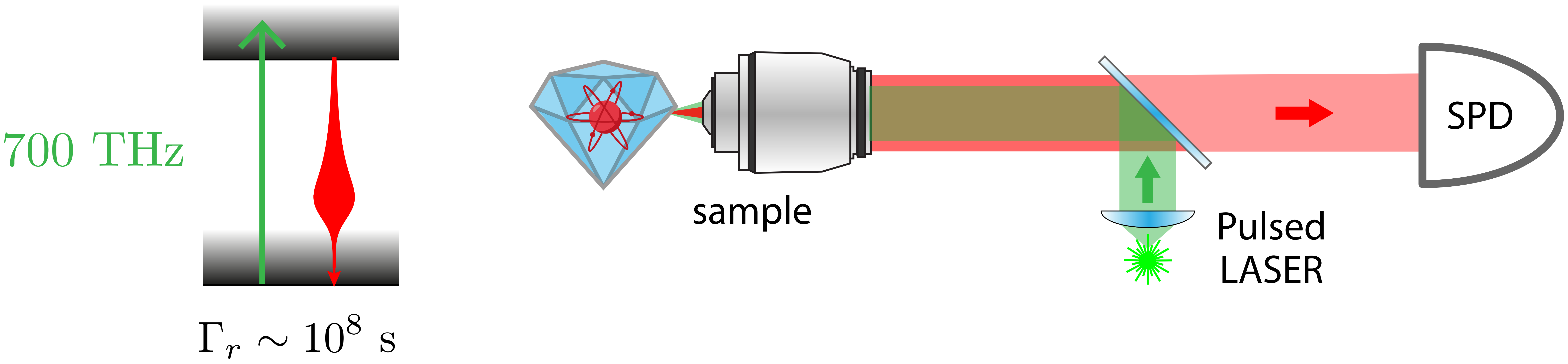

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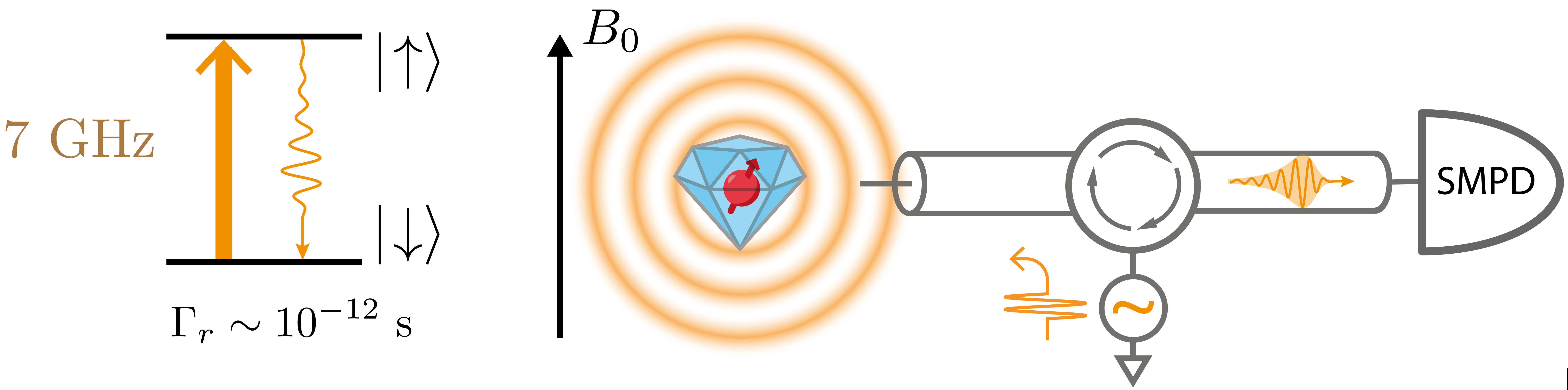
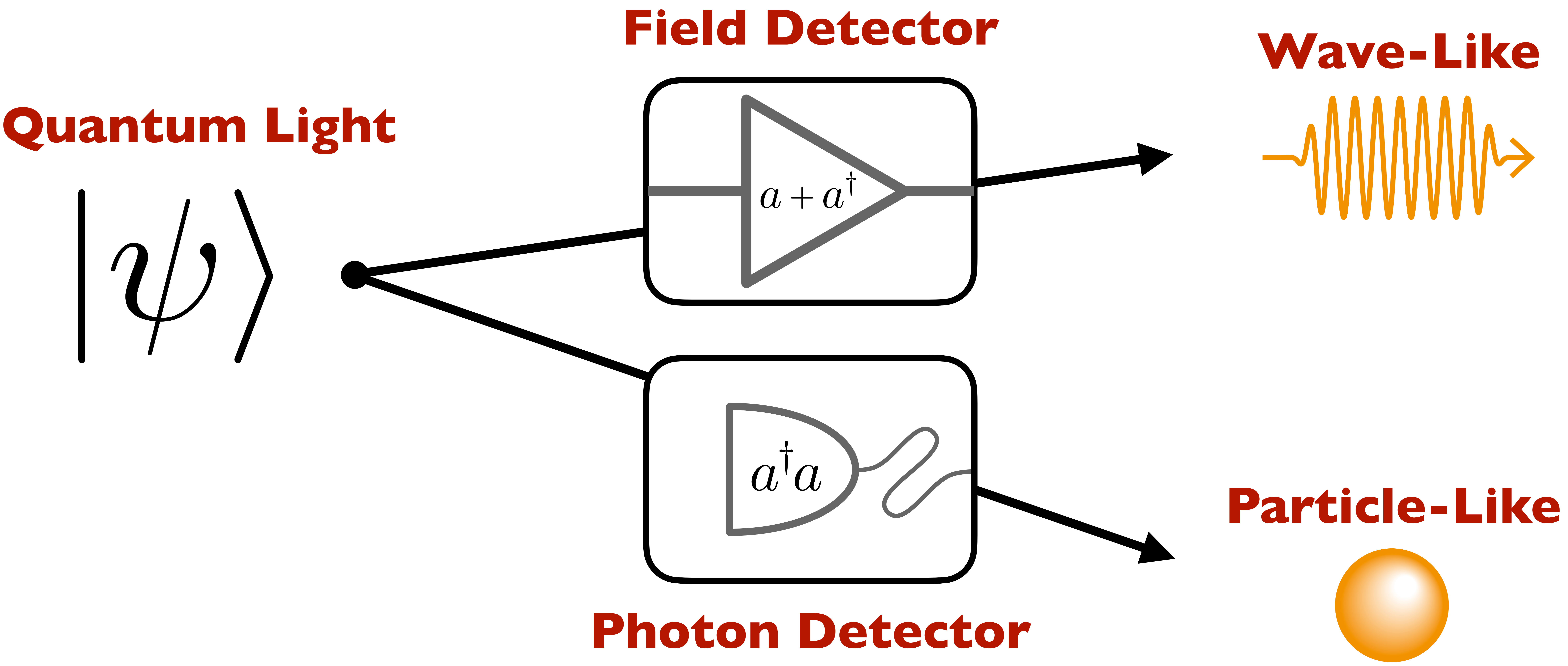


WANTED

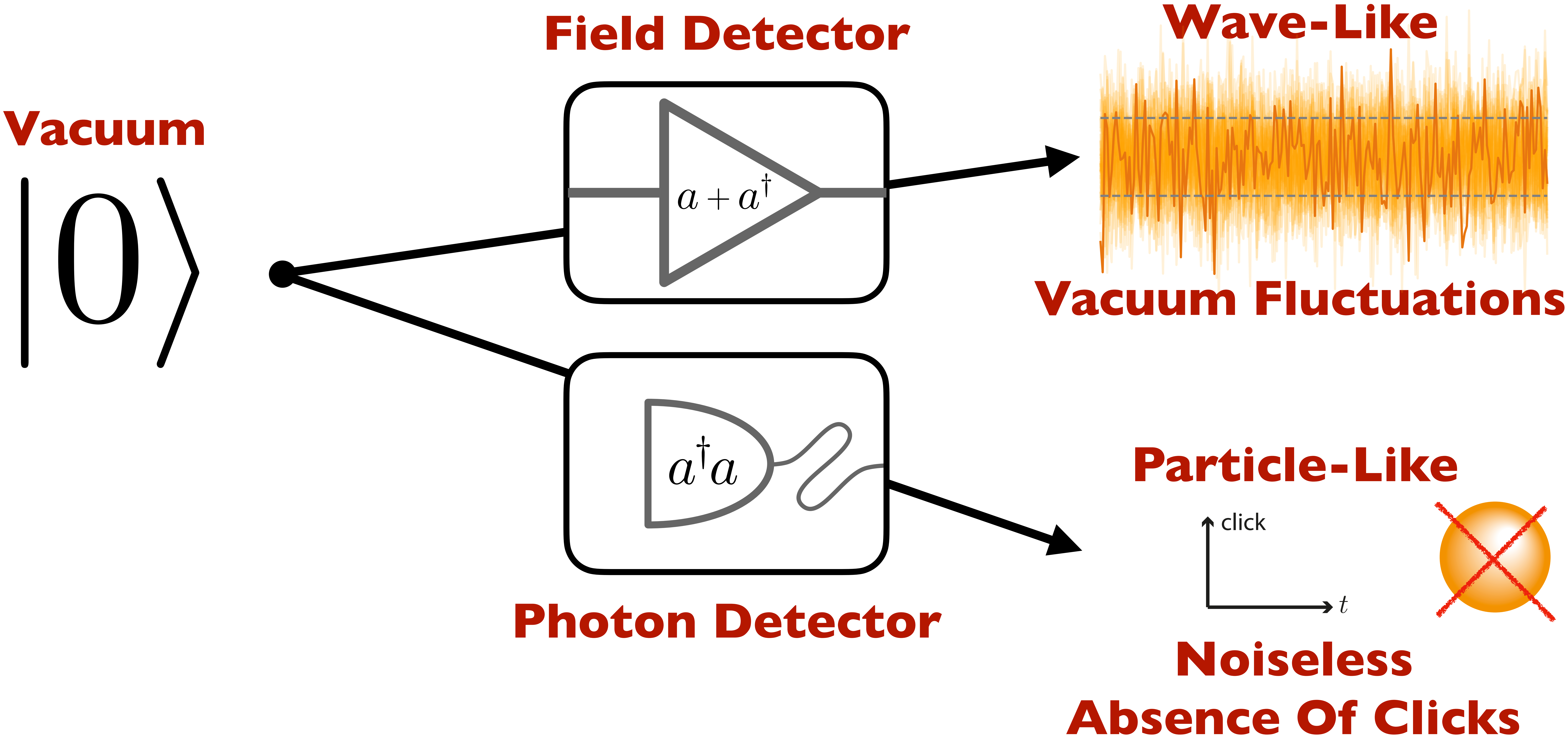
- Cryogenic temperature $\hbar\omega \gg kT$
- Large microwave radiative rate
- Single **Microwave** Photon Detector

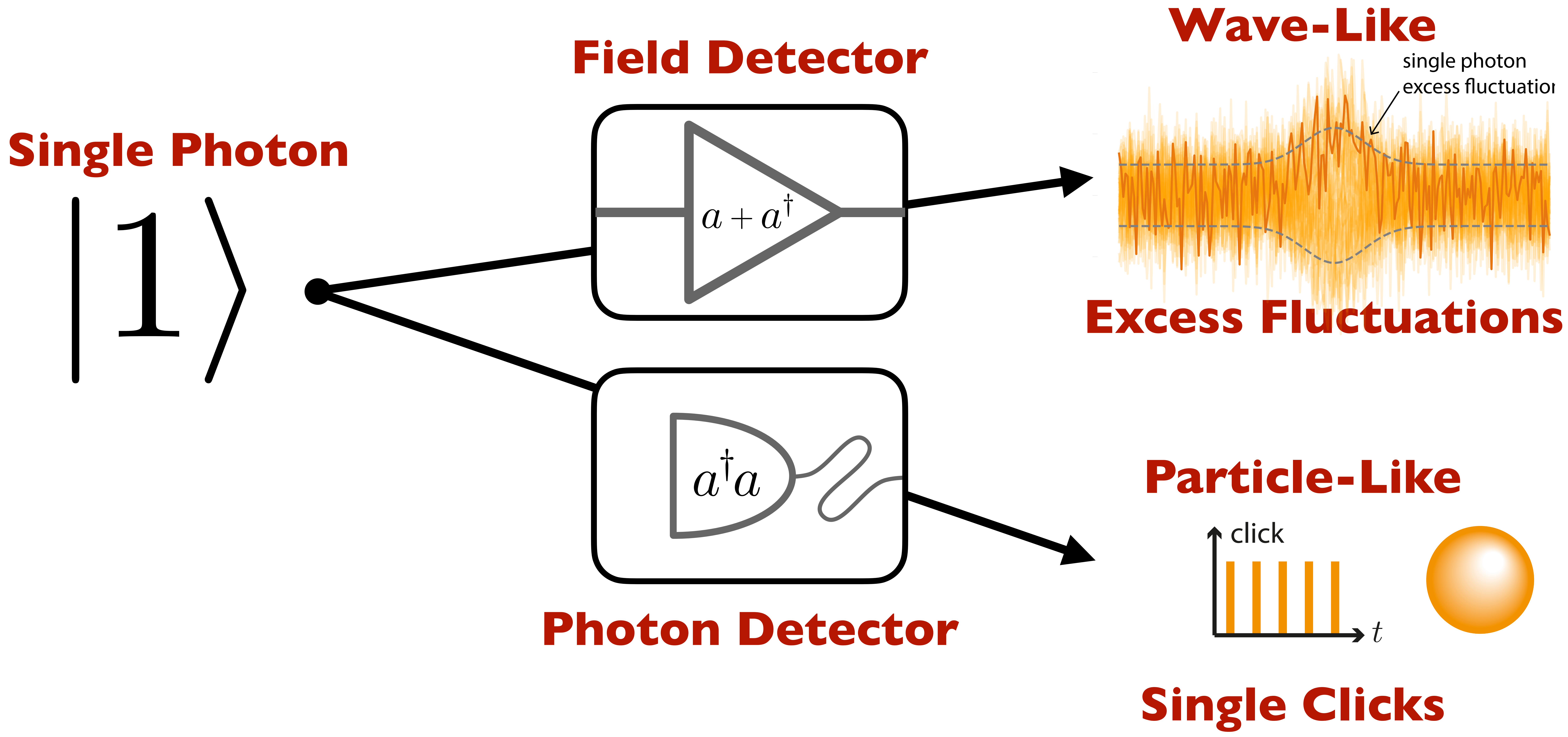
[Albertinale et al. Nature **600**, 434 (2021)]

Quantum Optics 101

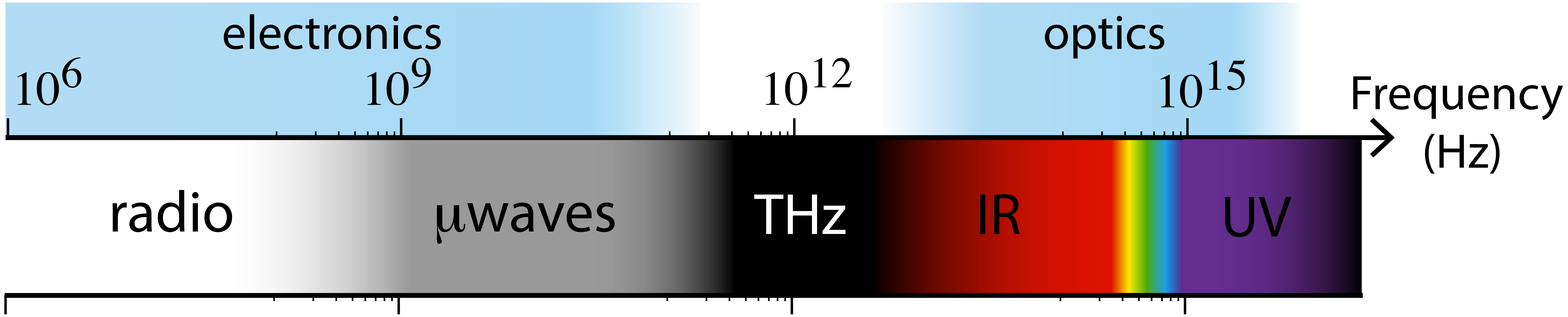


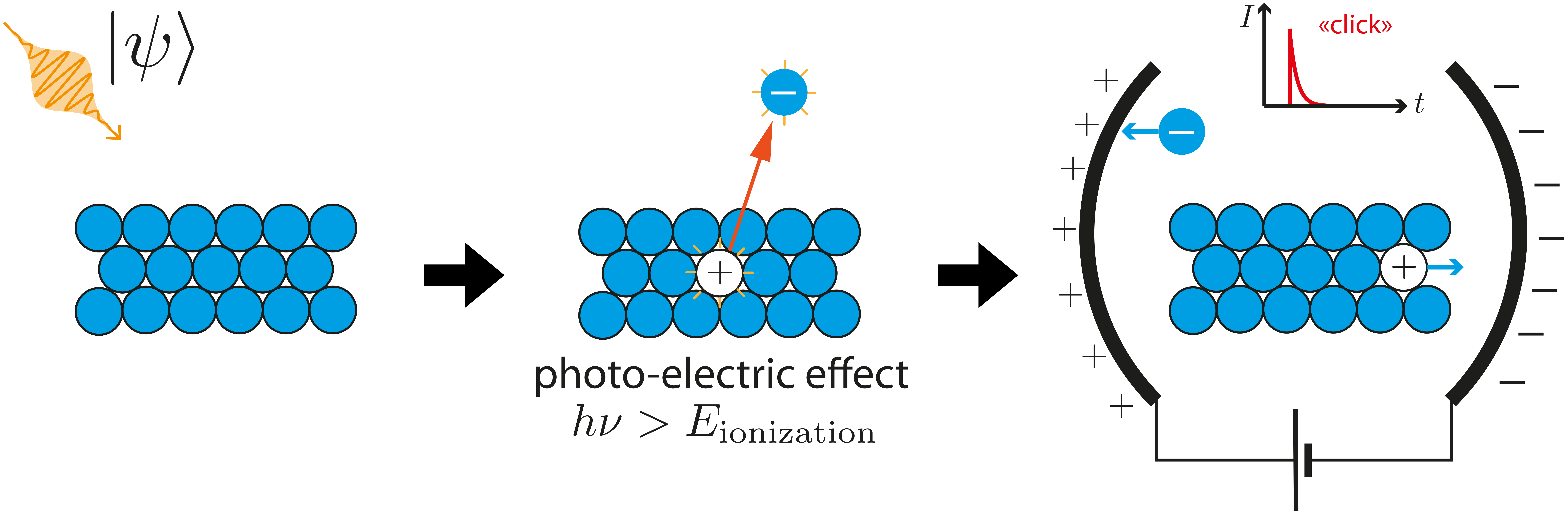
Quantum Optics 101




Single Photon
$|1\rangle$
Field Detector
$a + a^\dagger$
Photon Detector
$a^\dagger a$
Wave-Like
single photon
excess fluctuation
Excess Fluctuations
Particle-Like
click
t
Single Clicks

Detecting Photons At Microwave Frequency

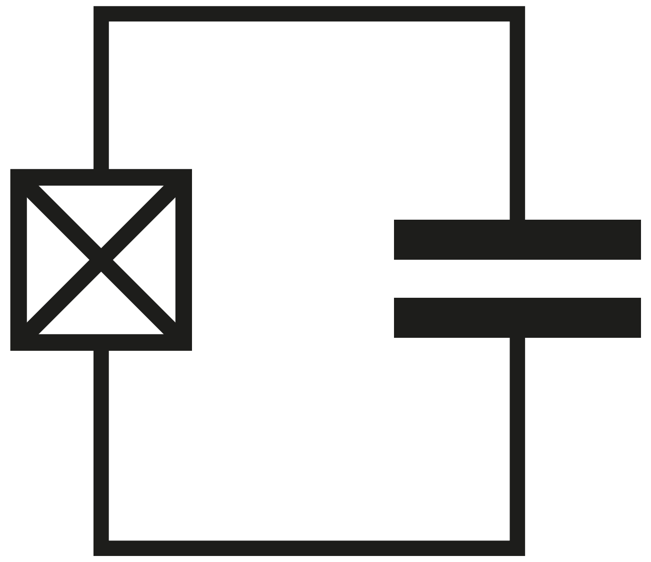


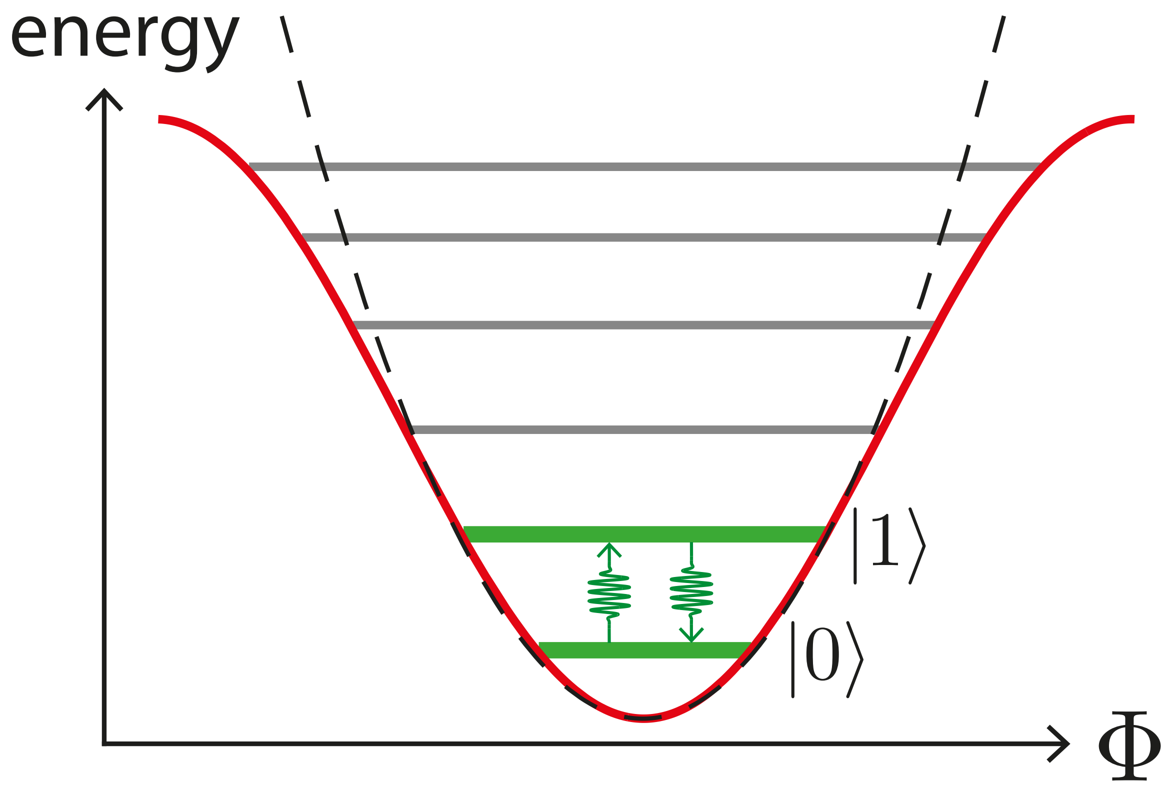


Circuits

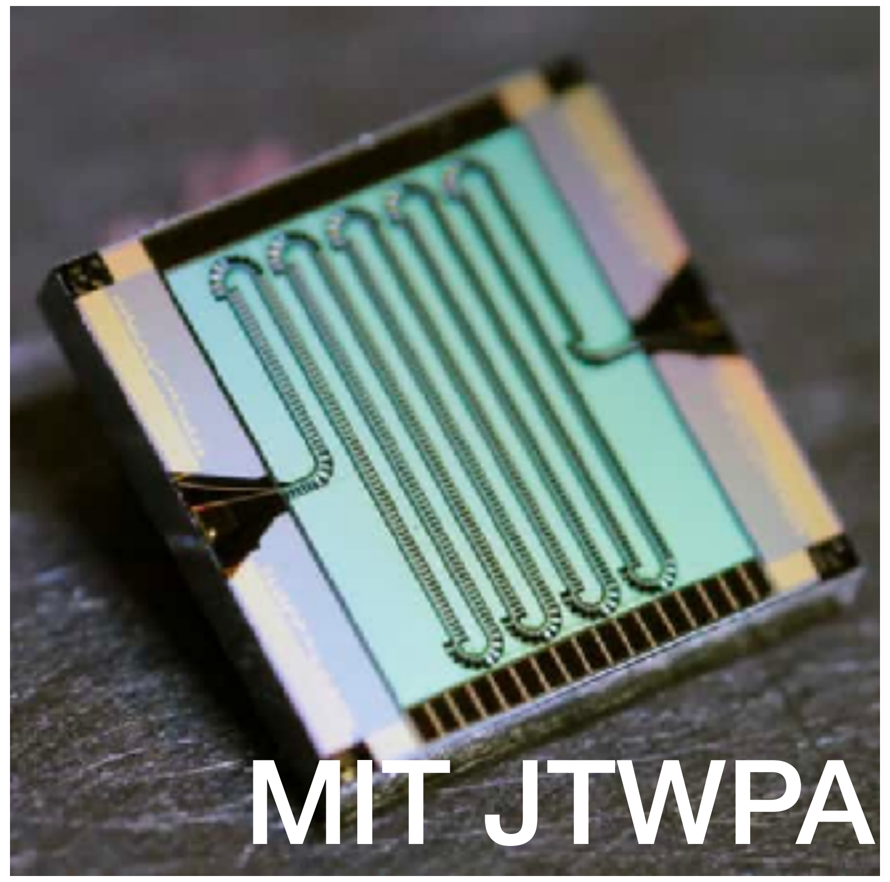
Artificial Atoms

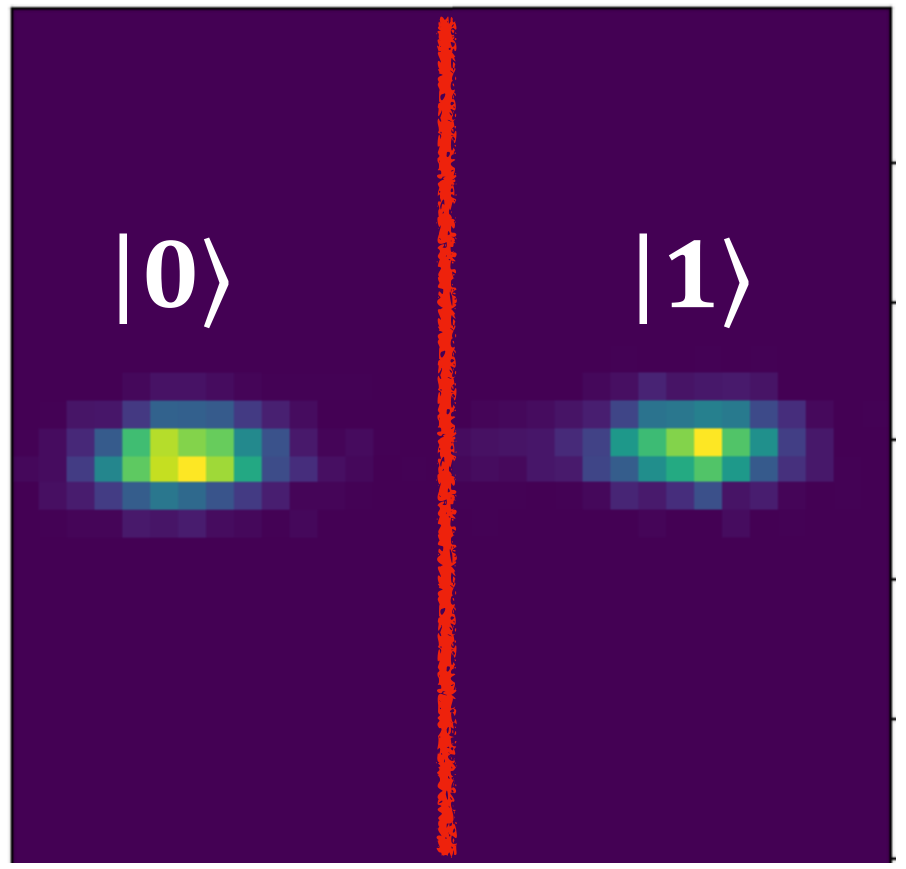
Transmon qubit



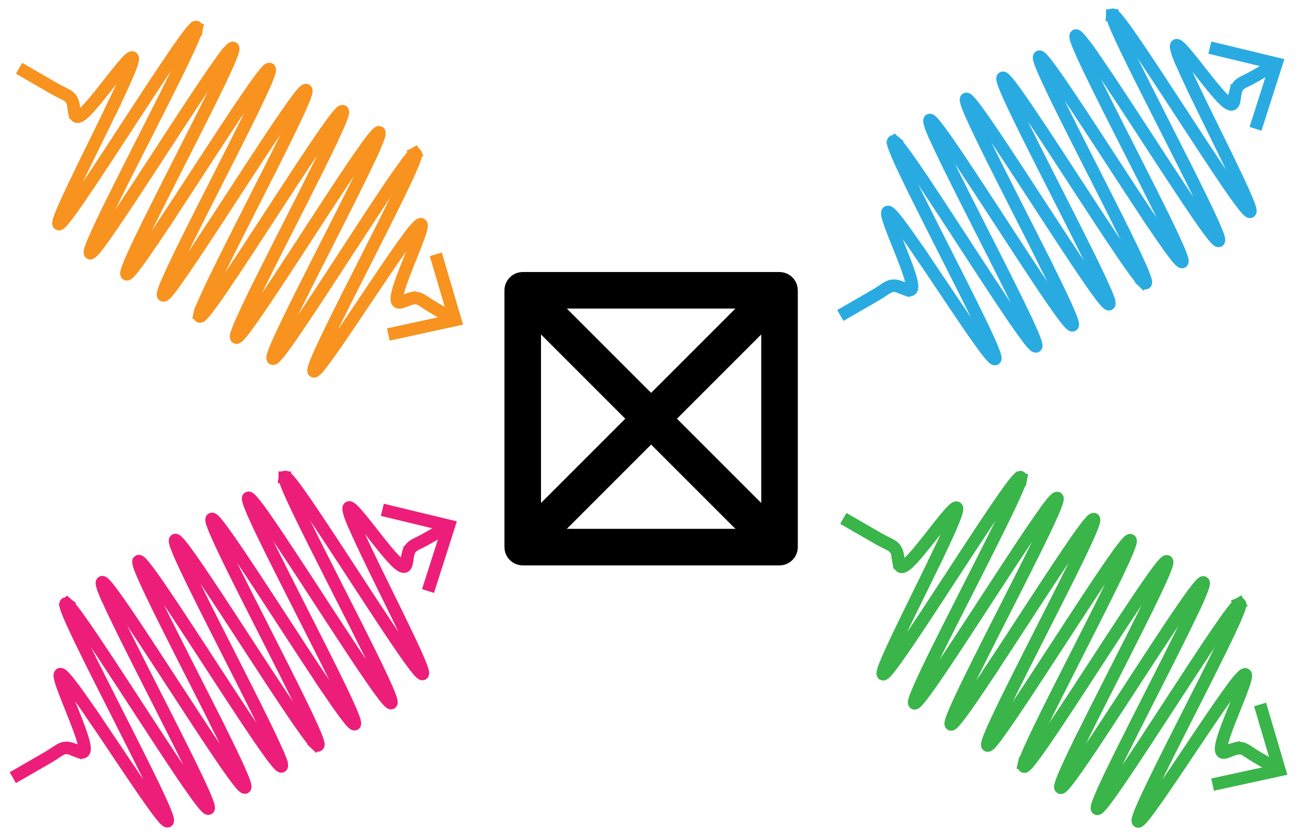


Quantum non-demolition measurement



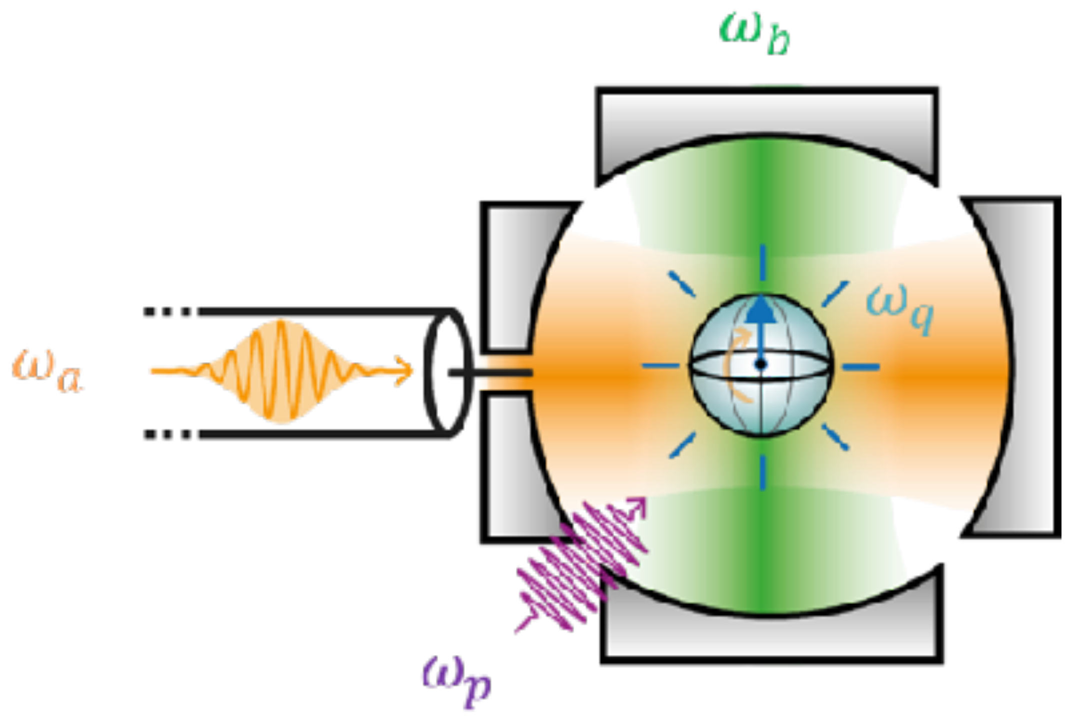


Built-in 4-wave-mixing



[1] R. Lescanne et al., PRX (2020)
[2] K. Inomata et al. Nature com (2016)
[3] J.-C. Besse et al. arXiv:1711.11569v1 (2017)
[4] Kono, S, et al. *Nature Physics* 14.6 (2018)
[5] Opremcak, A., et al., *Science* 361.6408 (2018)

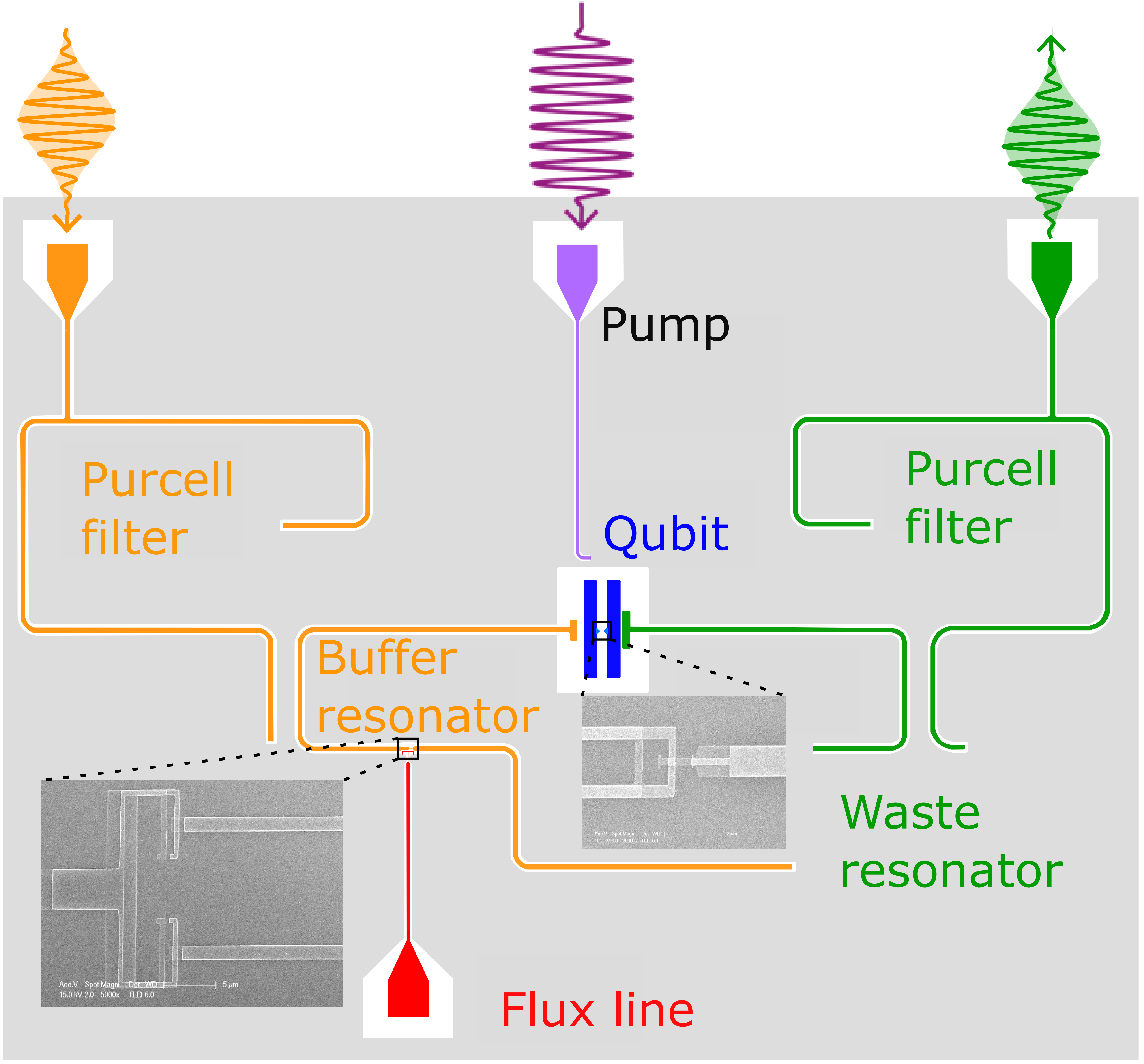
Sample design



$$\frac{\omega_a}{2\pi} = 7\ \mathrm{GHz}$$
$$Q_a = 5 \cdot 10^4$$
$$\chi_a = 5.4\ \mathrm{MHz}$$

$$\frac{\omega_q}{2\pi} = 6,2\ \mathrm{GHz}$$
$$T_1 = 30\ \mu s$$

$$\frac{\omega_b}{2\pi} = 7,7\ \mathrm{GHz}$$
$$Q_a = 4 \cdot 10^3$$
$$\chi_a = 19\ \mathrm{MHz}$$



Léo Balembois

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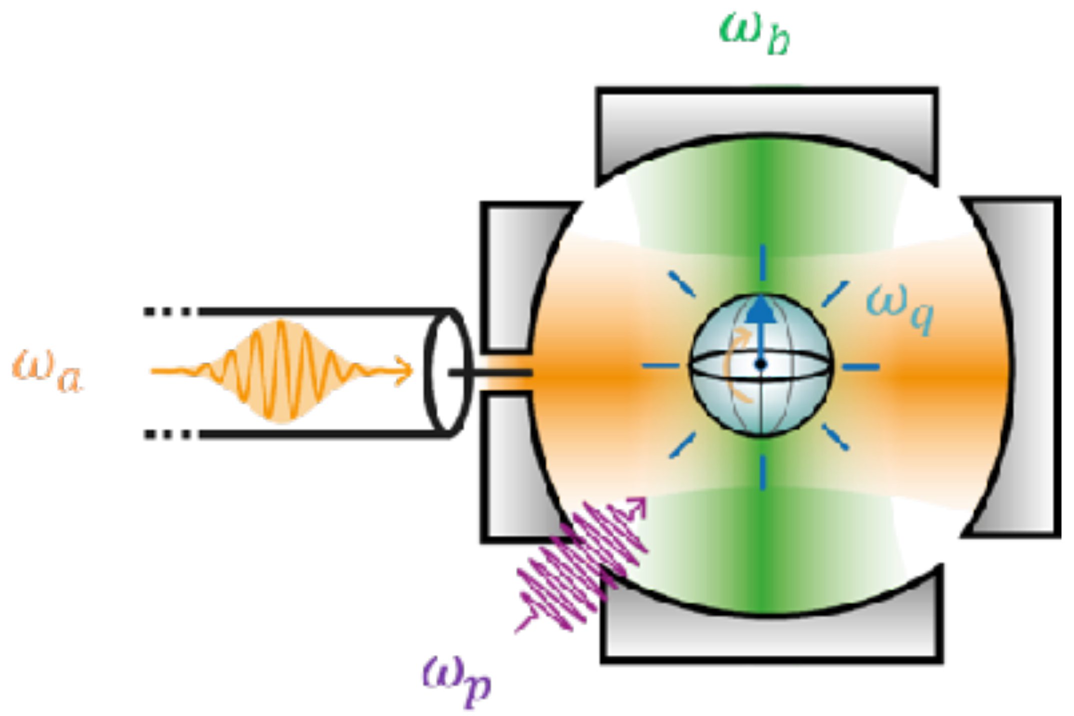

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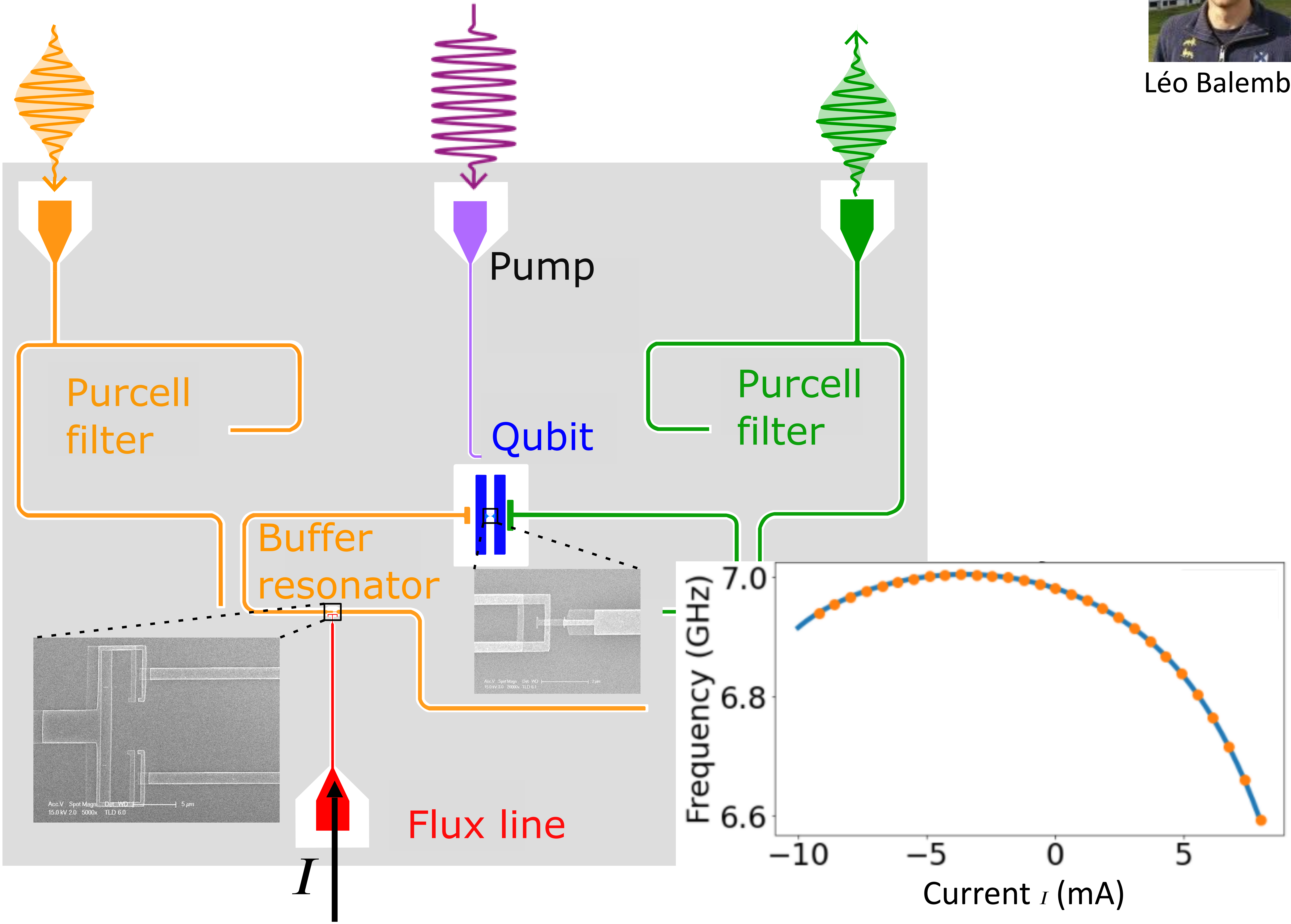
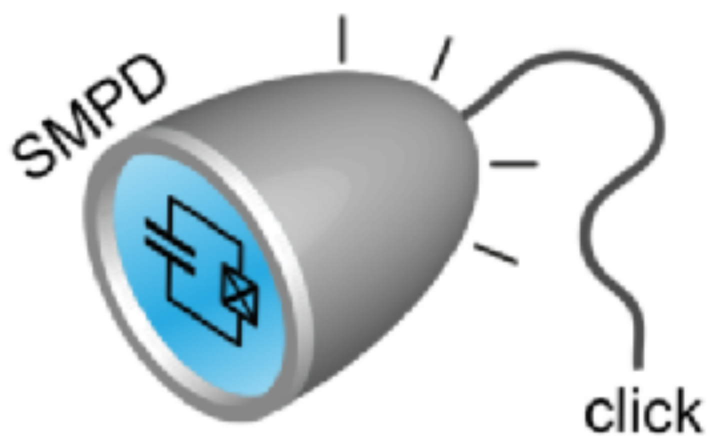
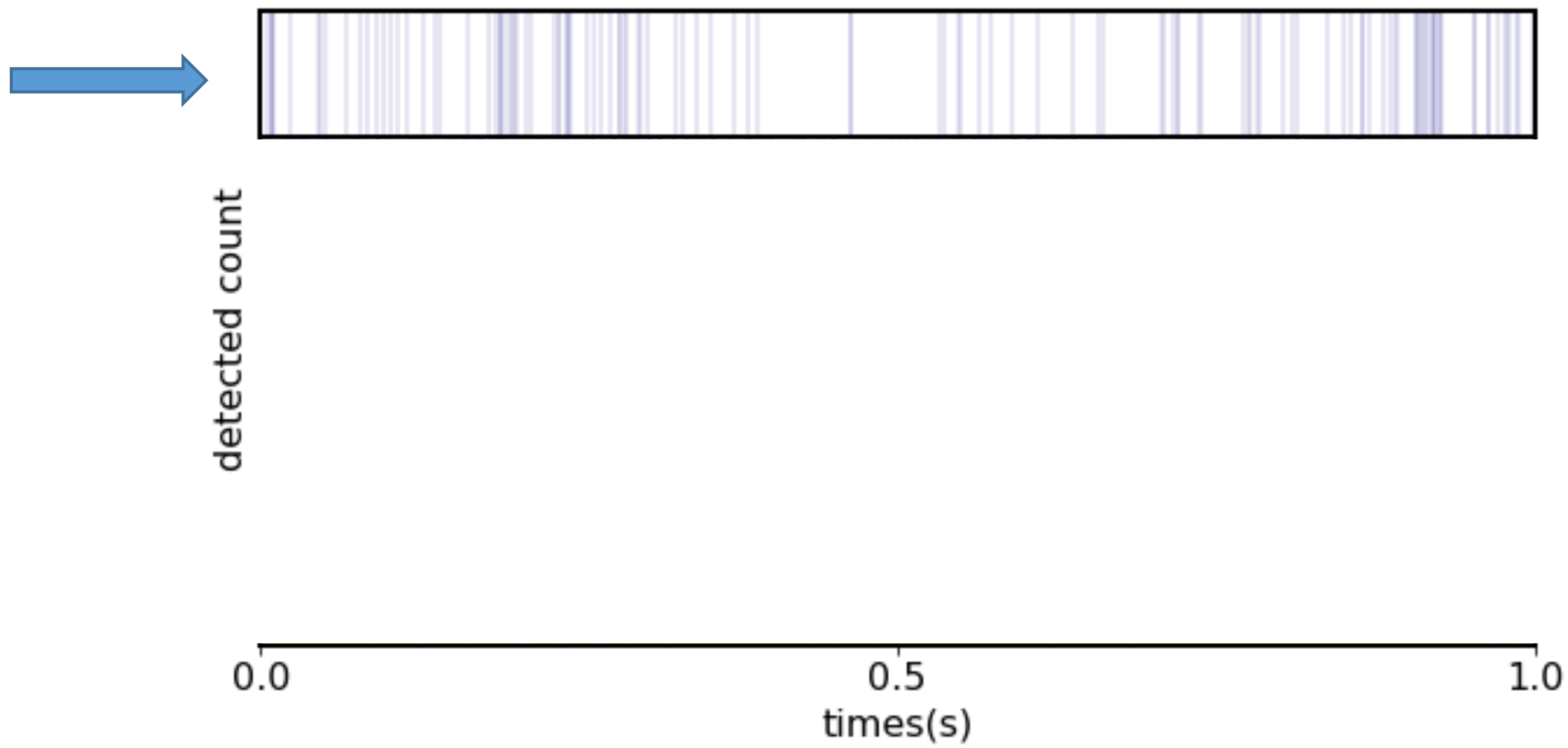


Quantum efficiency and darkcount rate

Darkcount rate $\alpha = 85\ s^{-1}$





Efficacité quantique et taux de compte d'obscurité

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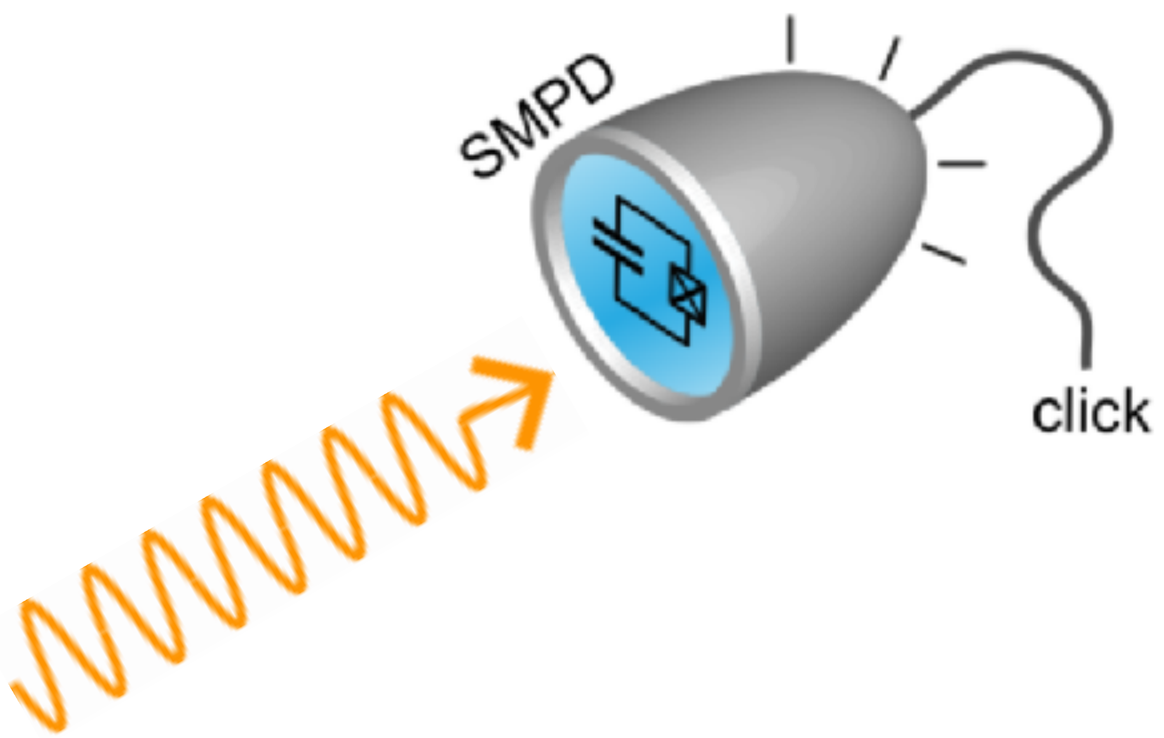

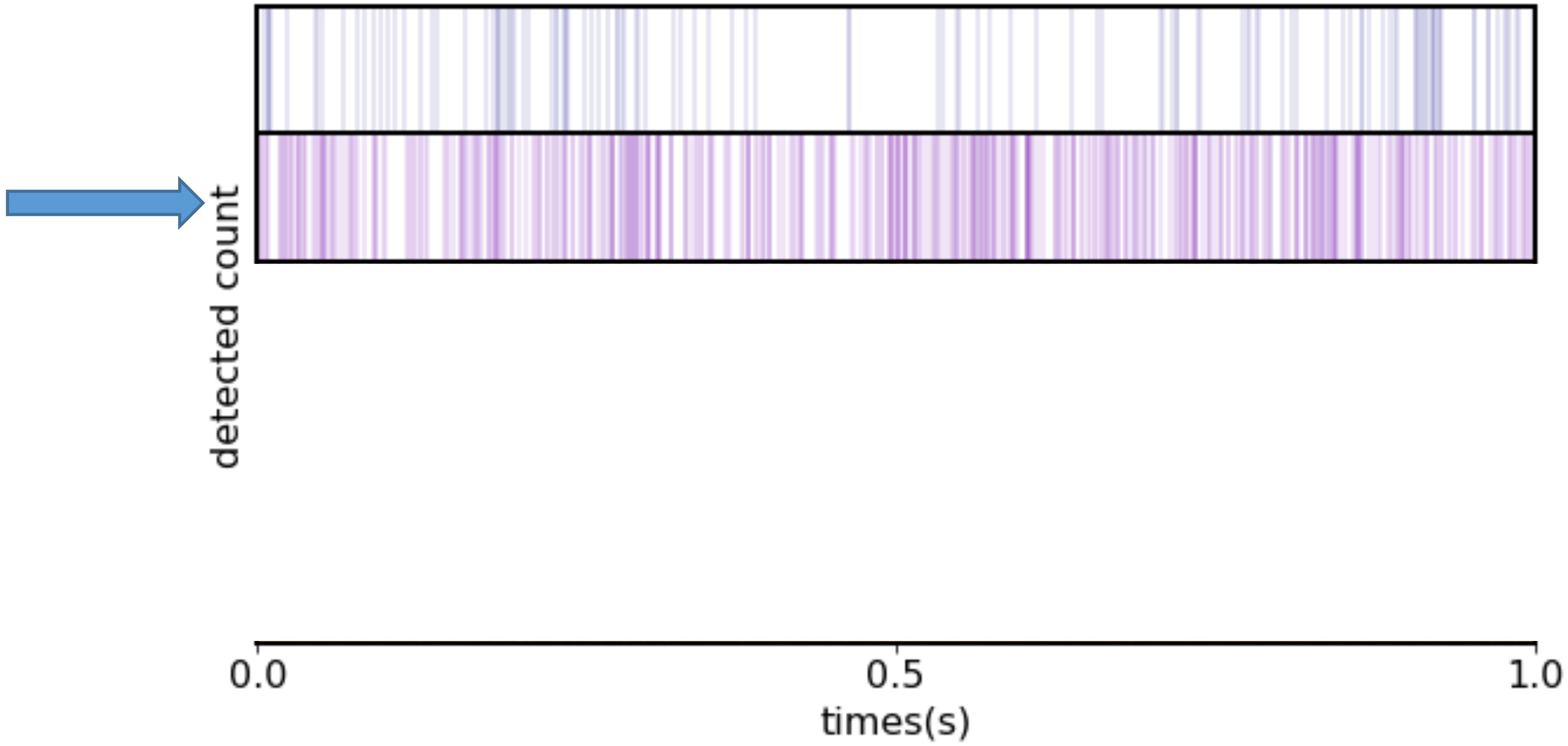

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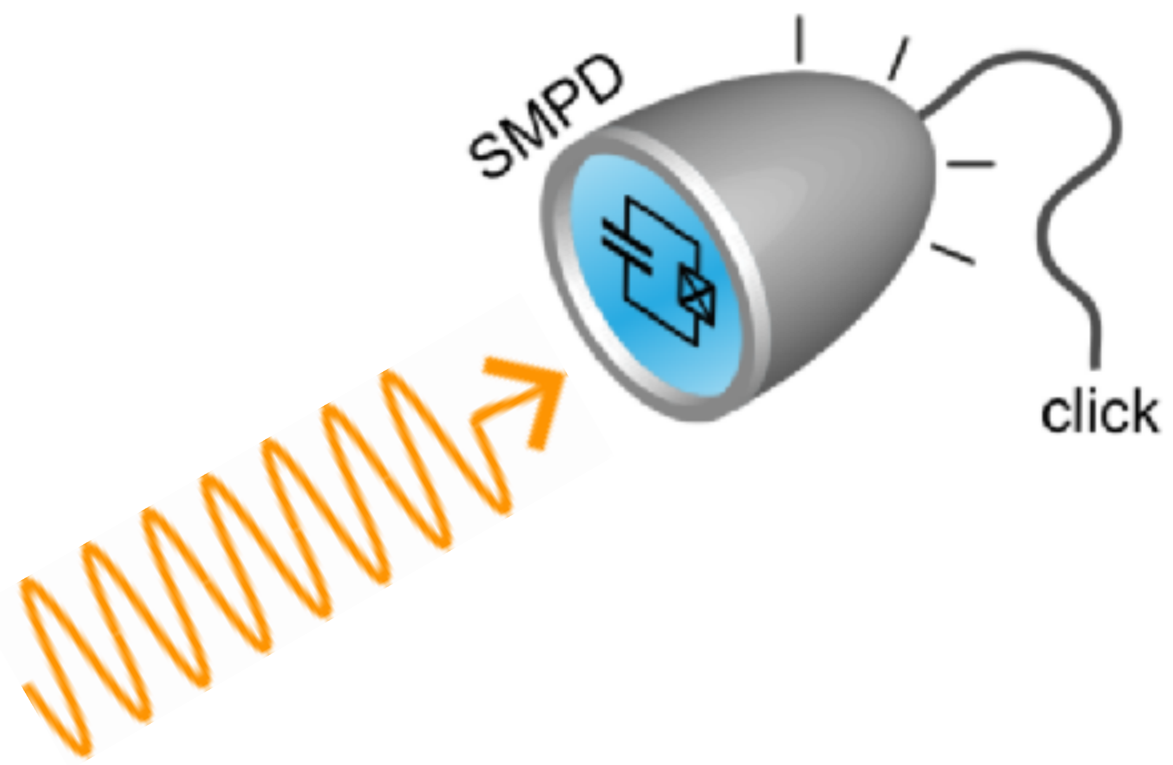

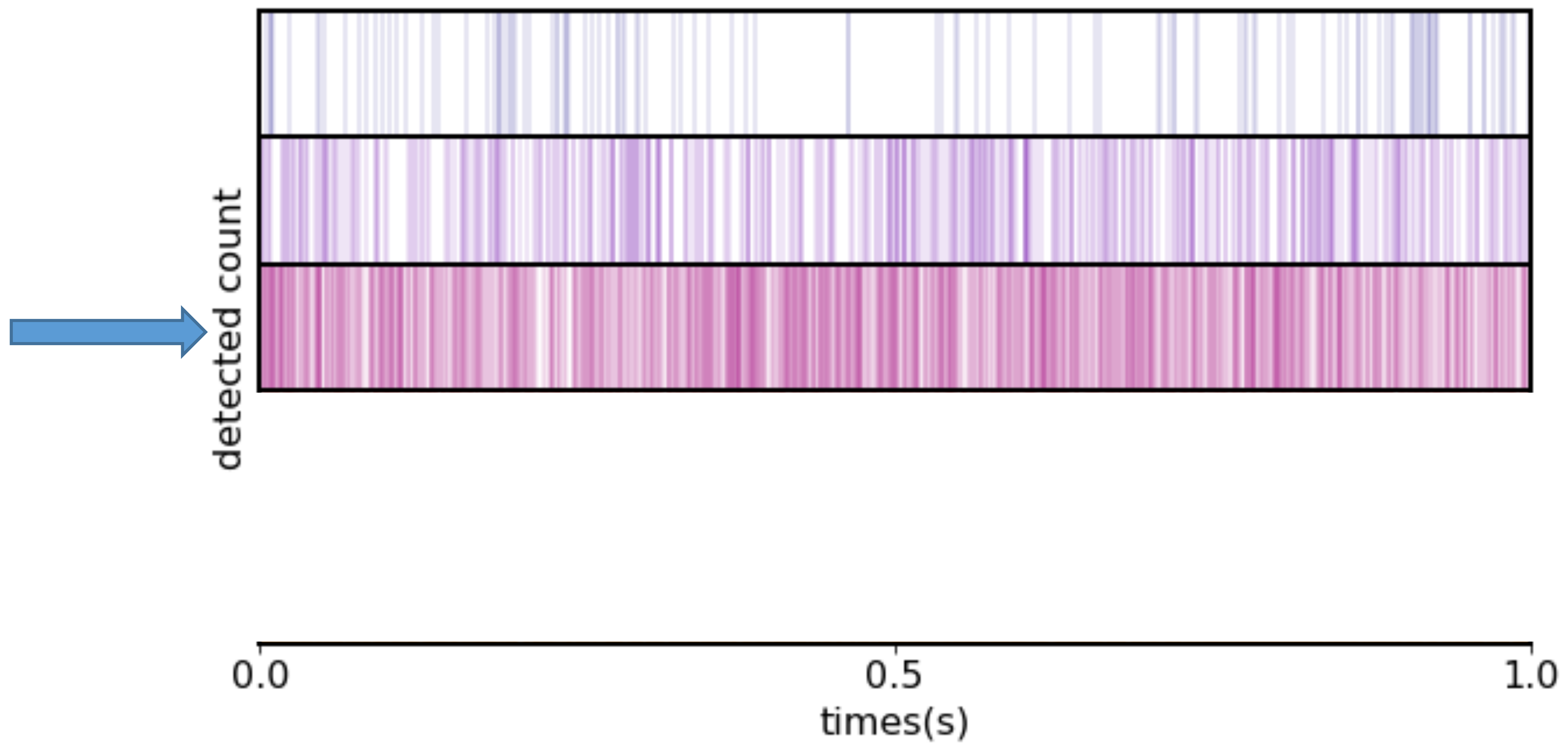

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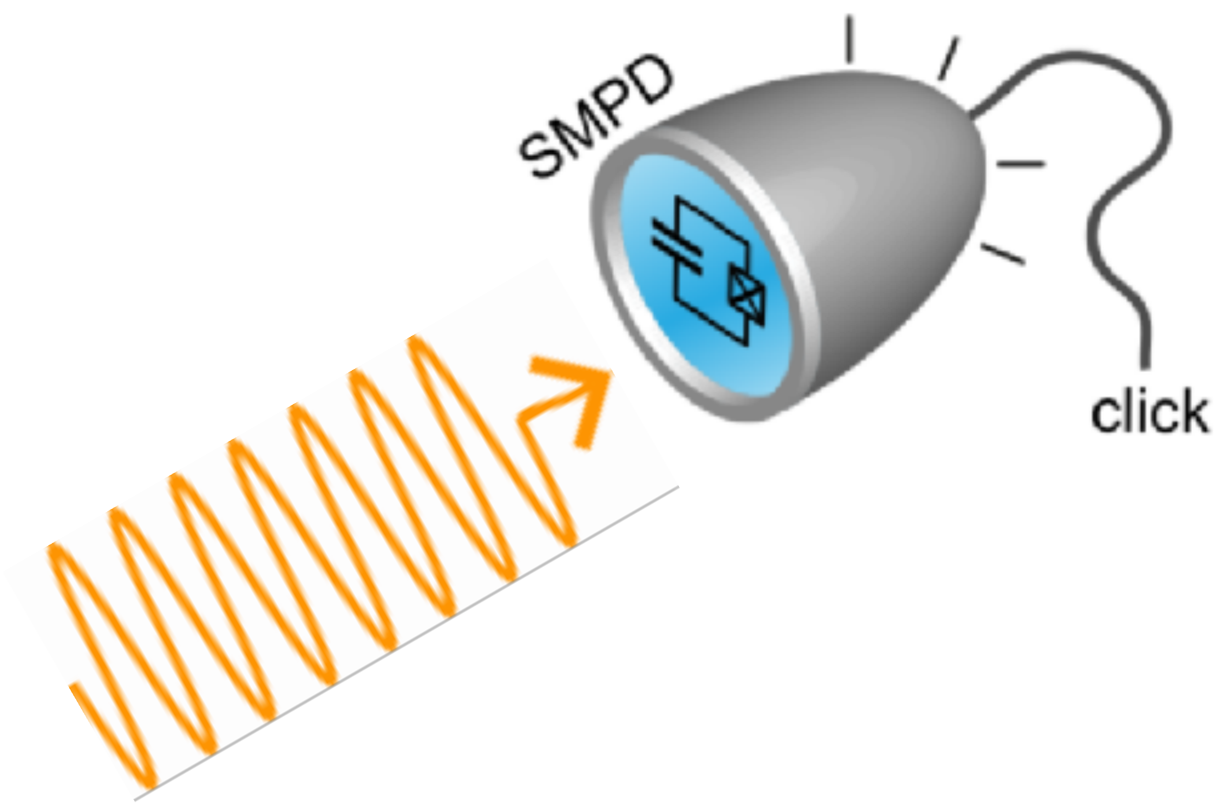
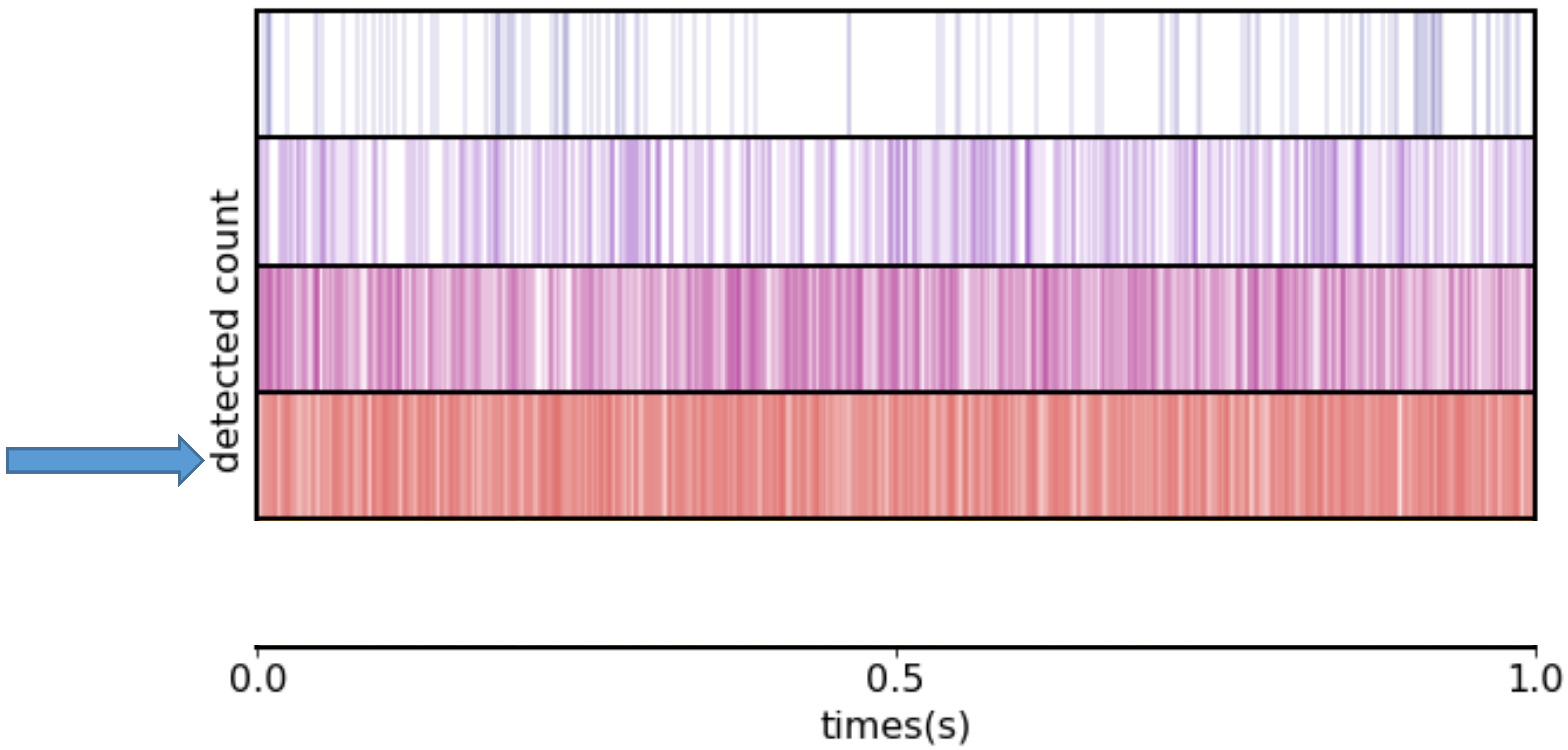
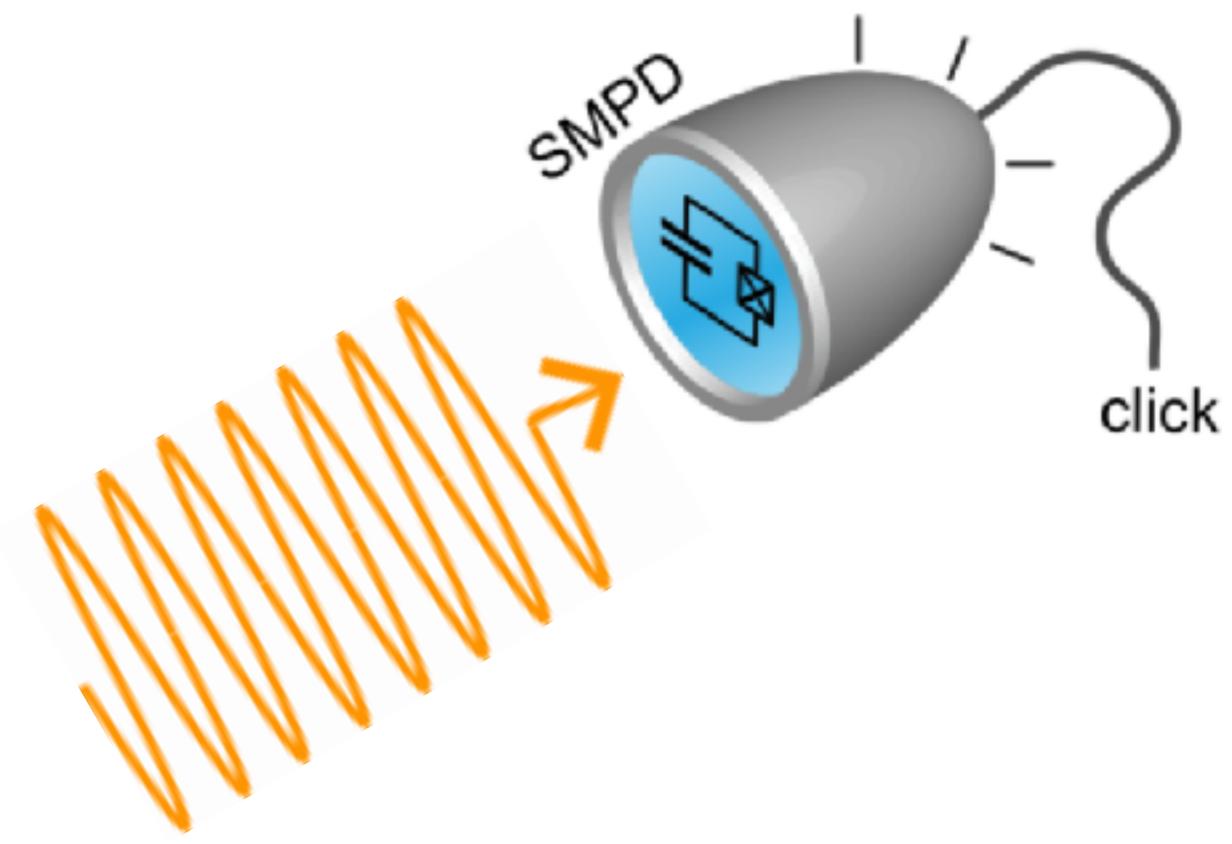
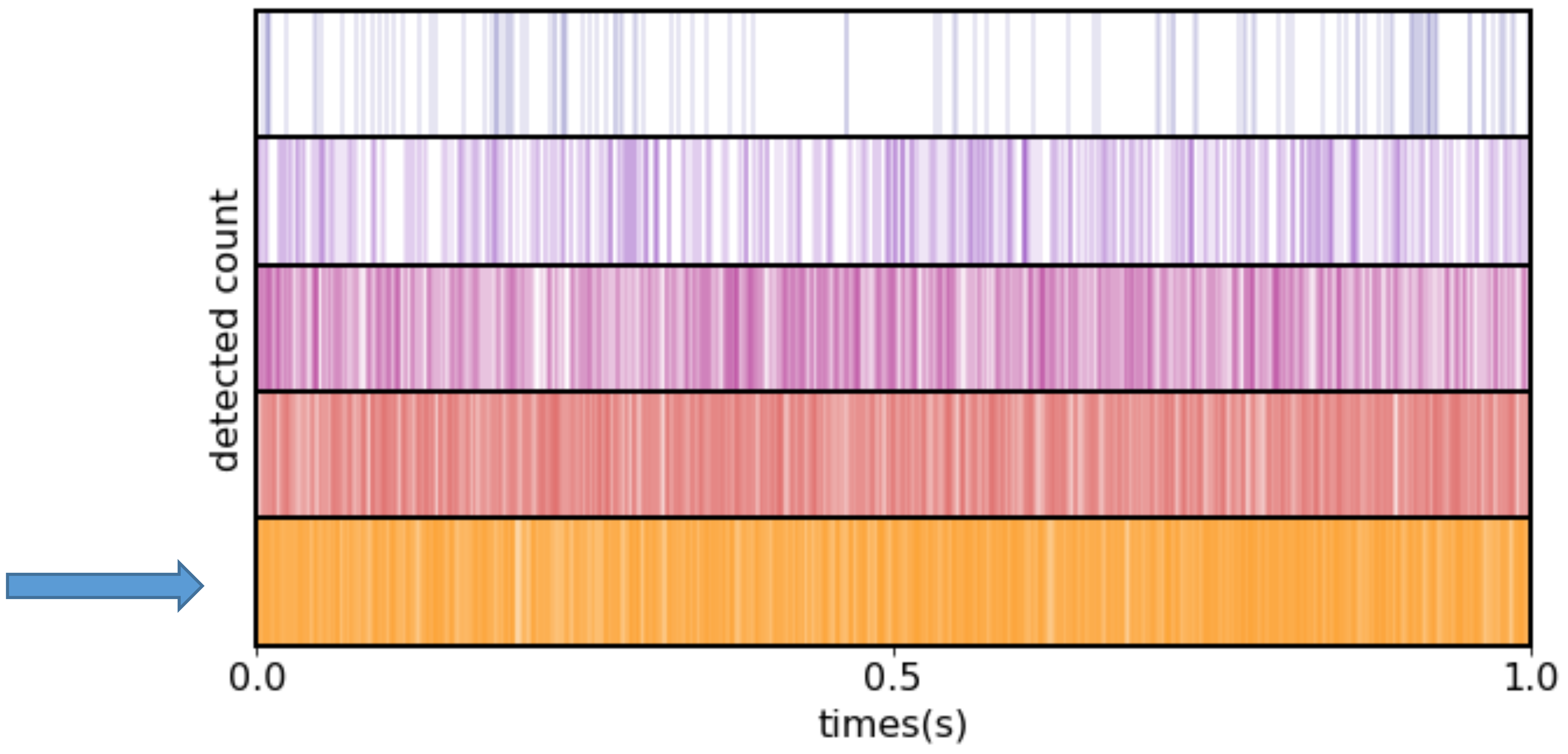




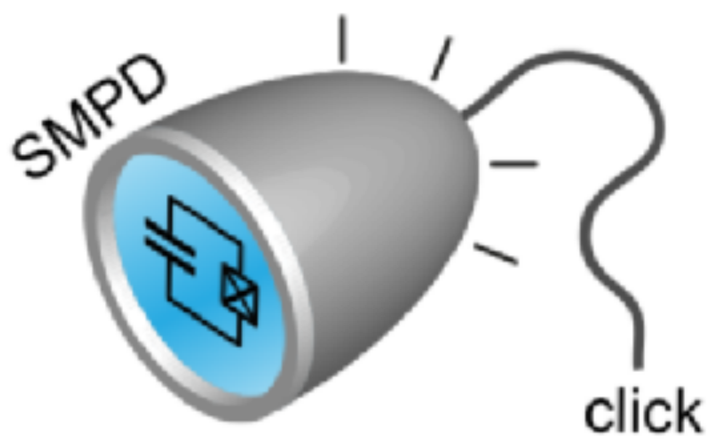
Efficacité quantique et taux de compte d'obscurité

Darkcount rate $\alpha = 85\ s^{-1}$



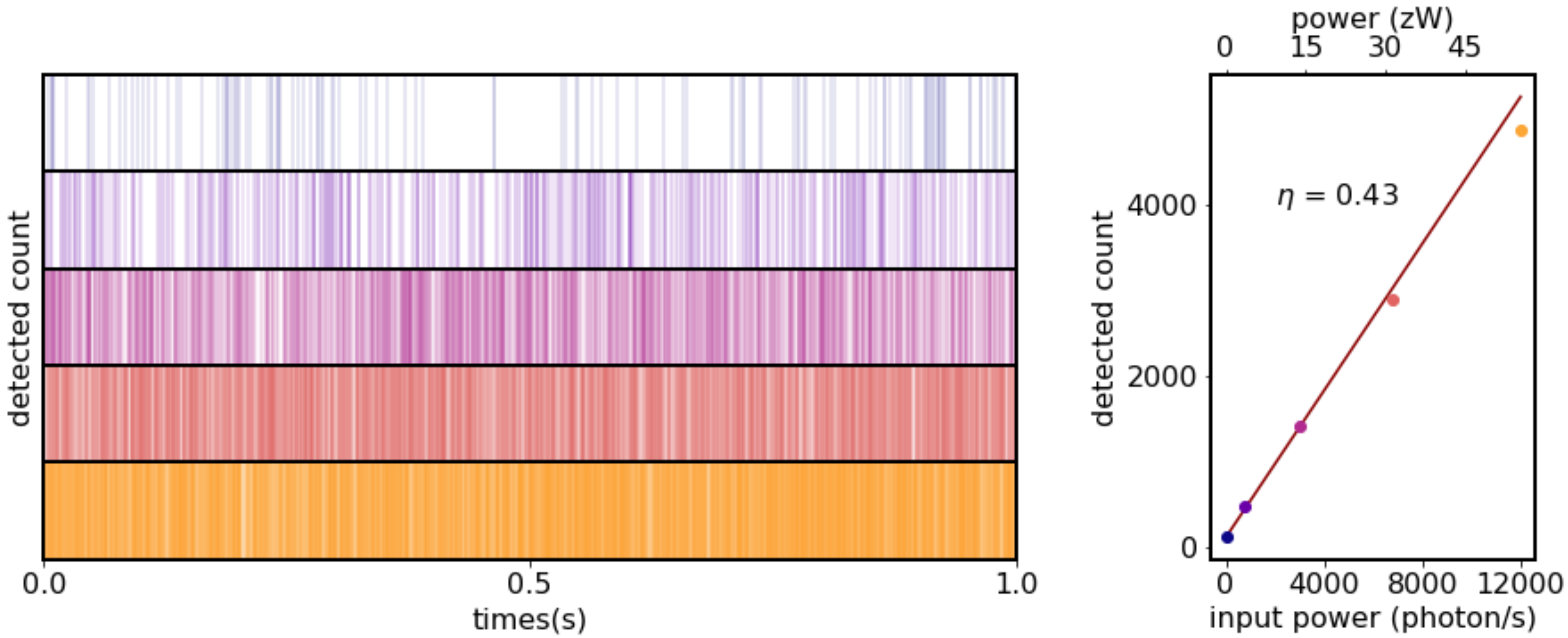


Efficacité quantique et taux de compte d'obscurité

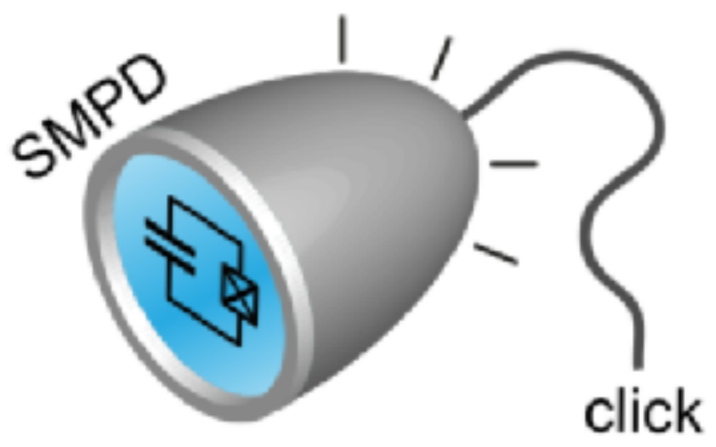


Darkcount rate $\alpha = 85\ s^{-1}$

Operational Efficiency : $\eta = 45\%$



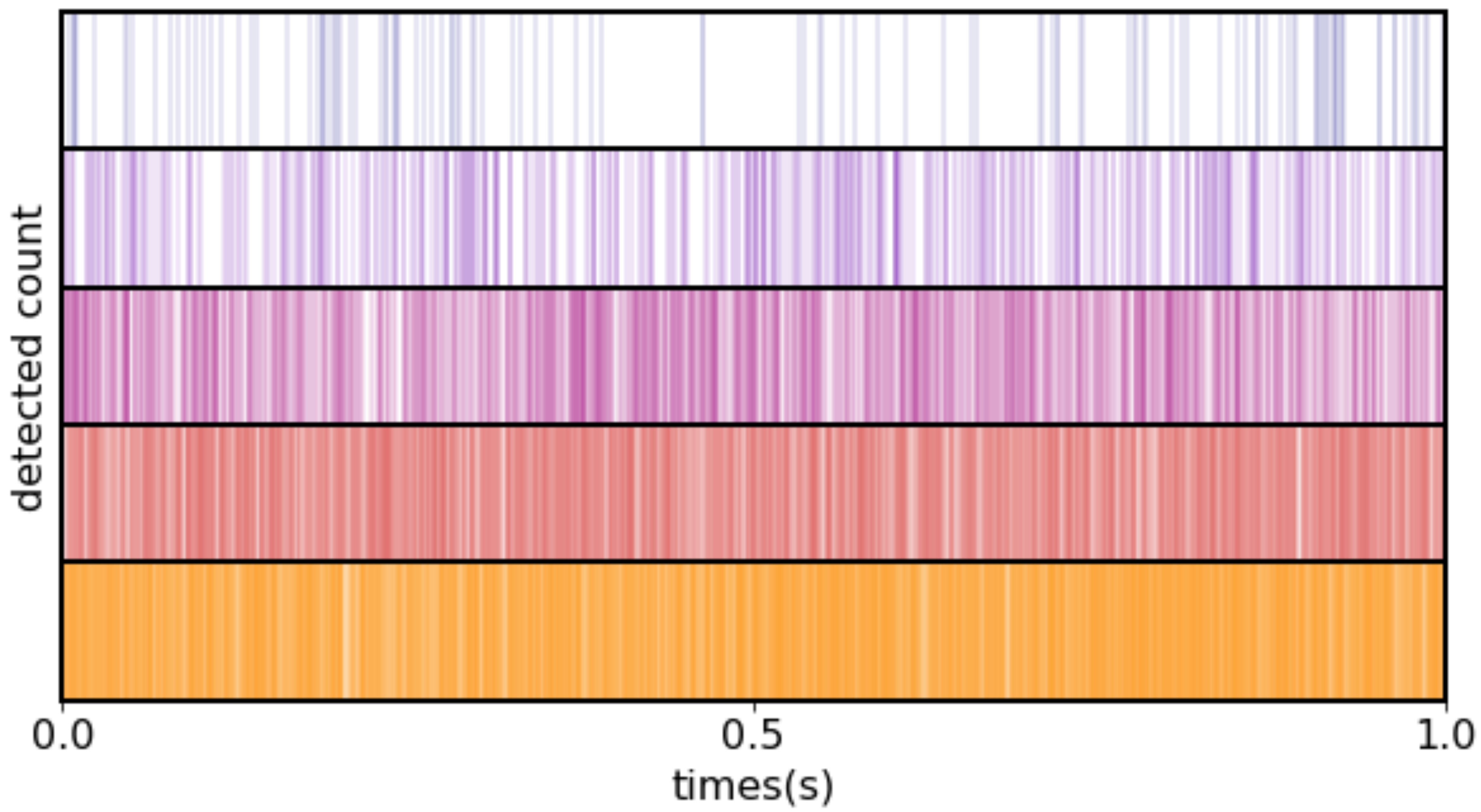
Efficacité quantique et taux de compte d'obscurité

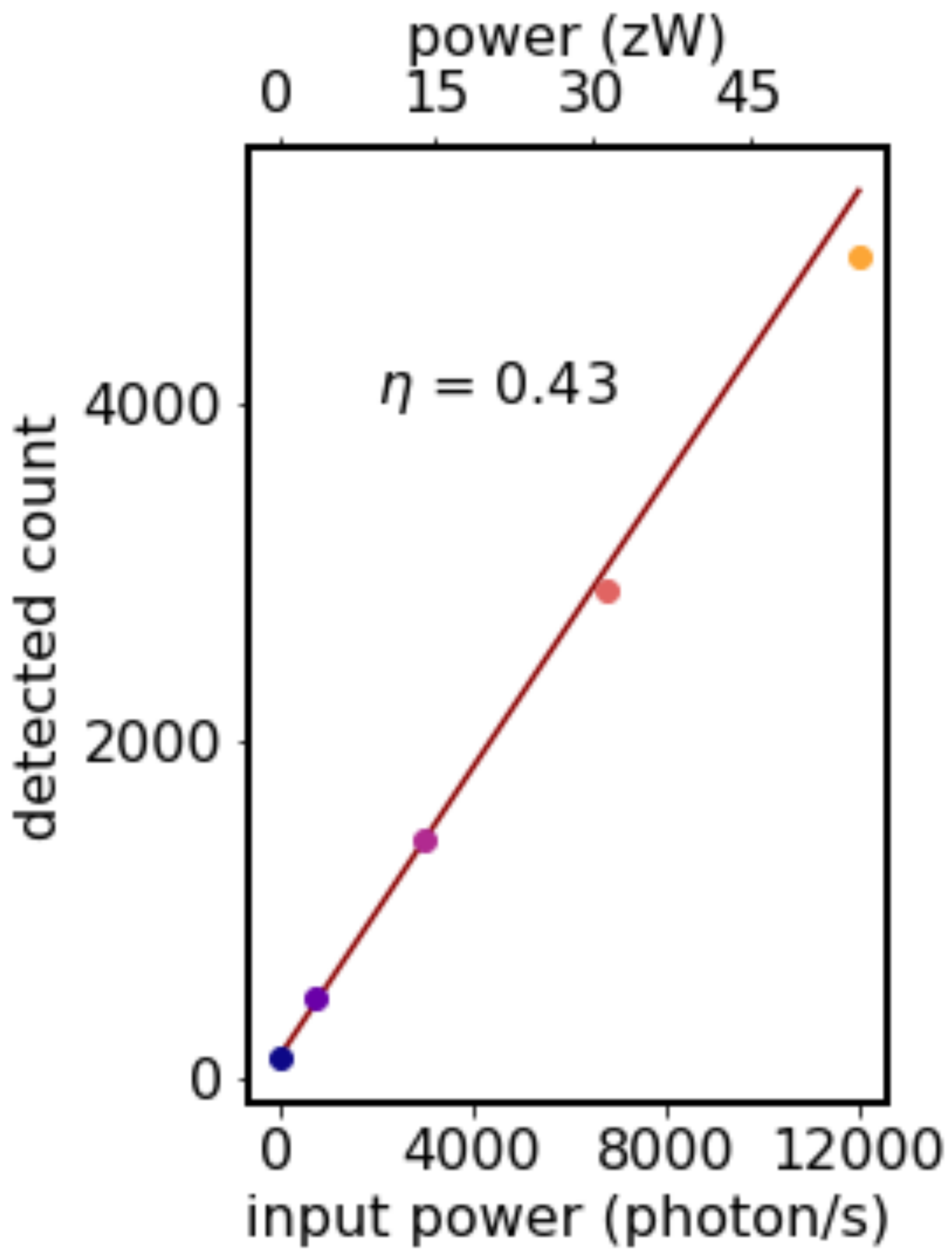


Darkcount rate $\alpha = 85\ s^{-1}$

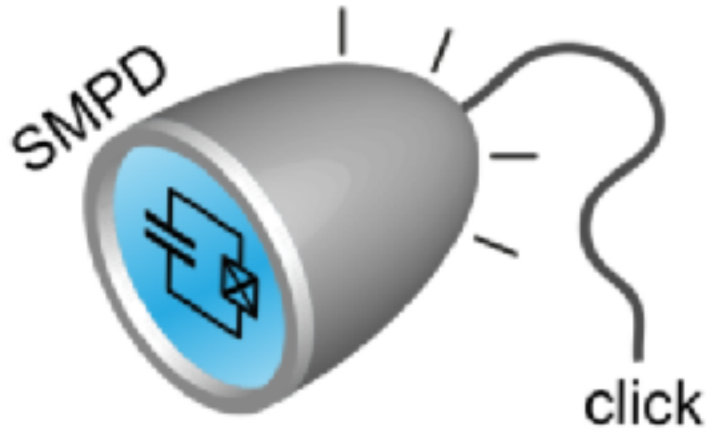
Operational Efficiency : $\eta = 45\%$

Power sensitivity: $\hbar\omega \frac{\sqrt{\alpha}}{\eta} = 10^{-22}\ \mathrm{W}/\sqrt{\mathrm{Hz}}$





Efficacité quantique et taux de compte d'obscurité

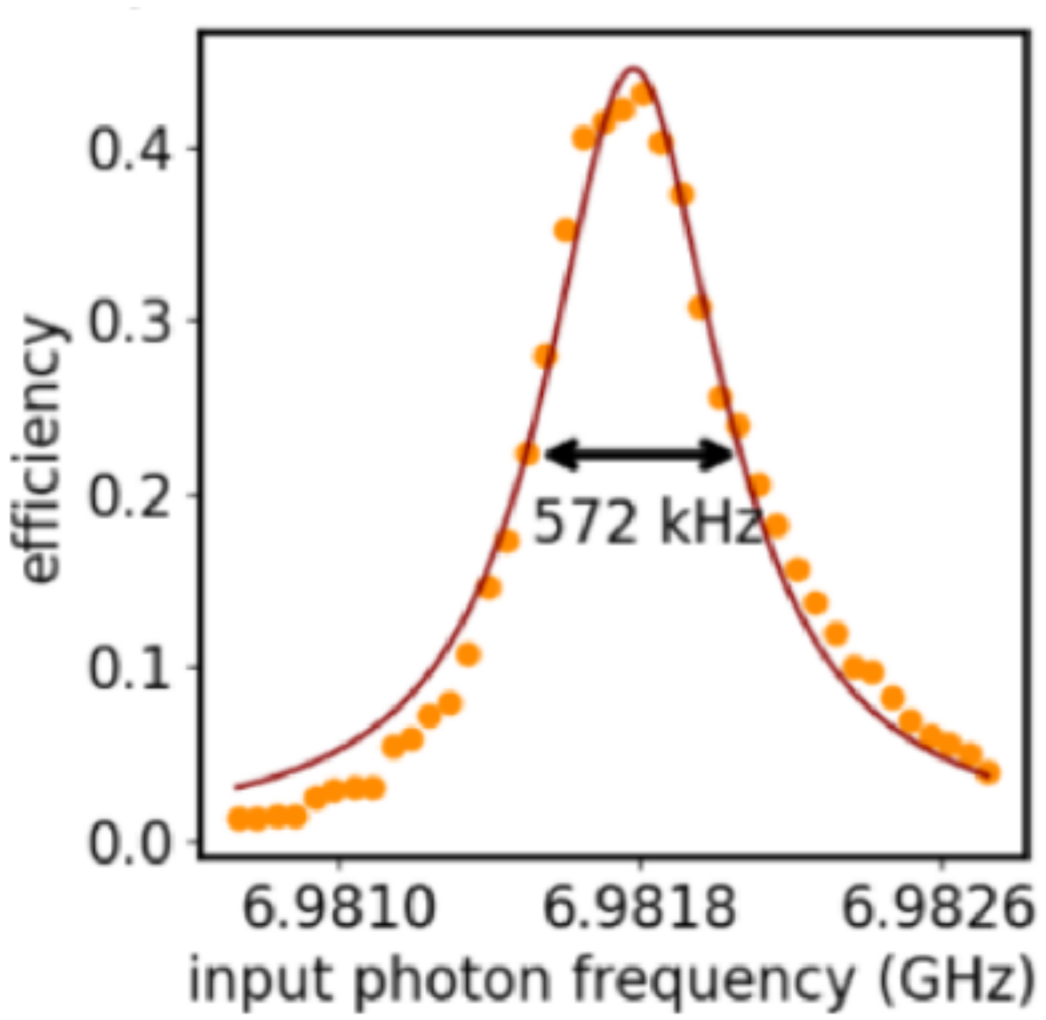


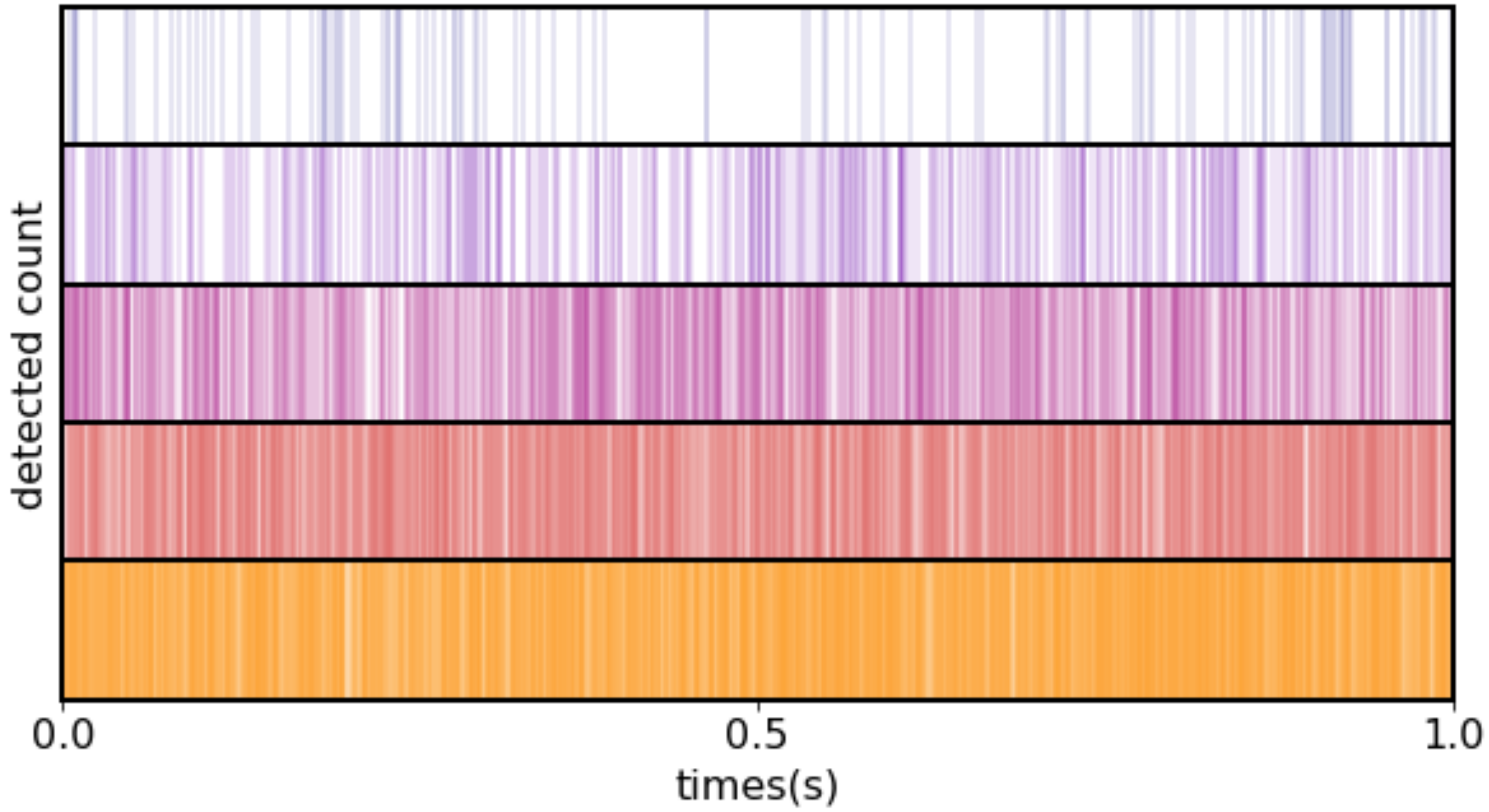
Darkcount rate $\alpha = 85\ s^{-1}$

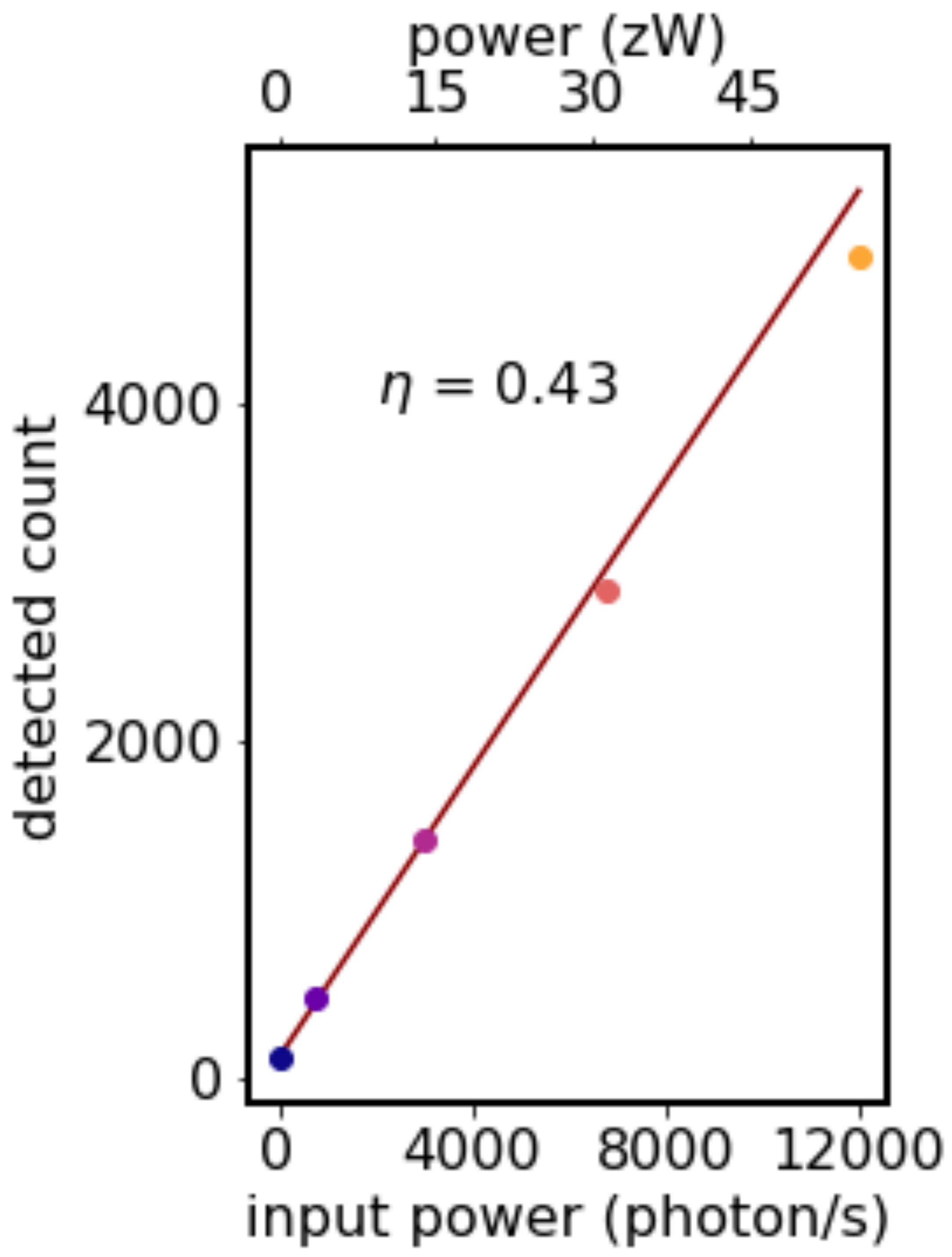
Operational Efficiency : $\eta = 45\%$

Power sensitivity: $\hbar\omega\frac{\sqrt{\alpha}}{\eta} = 10^{-22}\ \mathrm{W}/\sqrt{\mathrm{Hz}}$

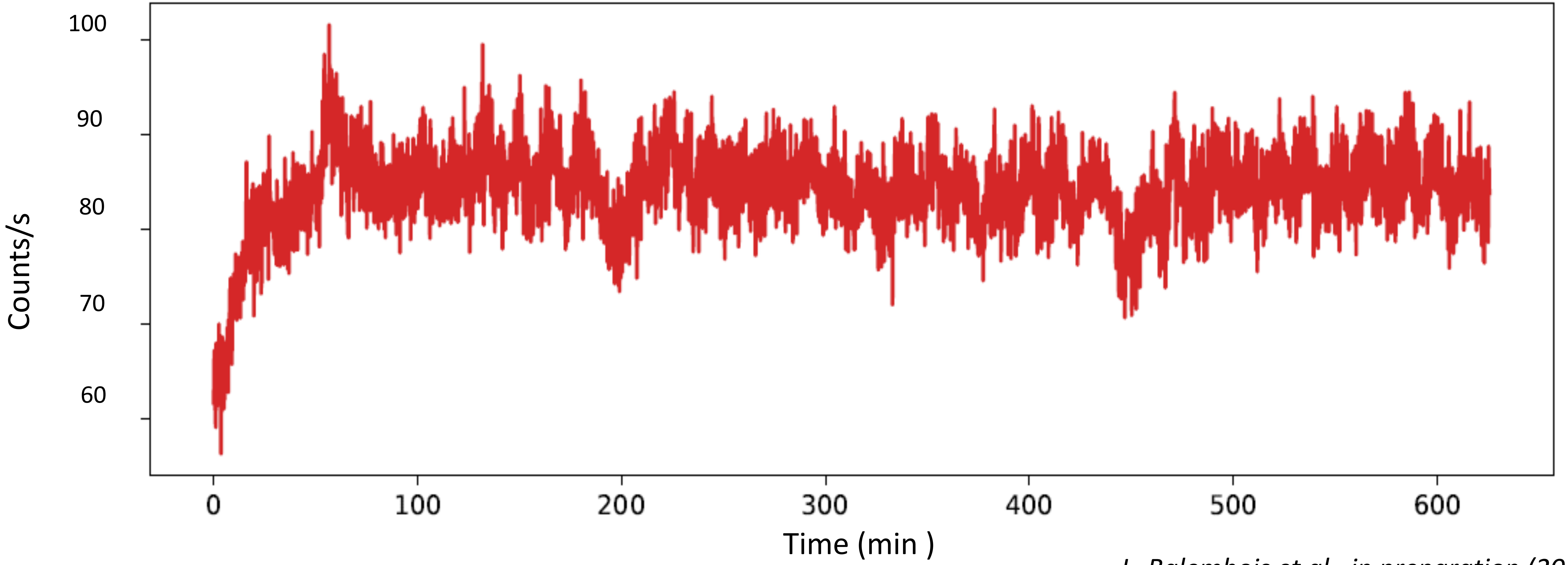
Bandwidth: $\mathrm{BW} = 0.5\ \mathrm{MHz}$







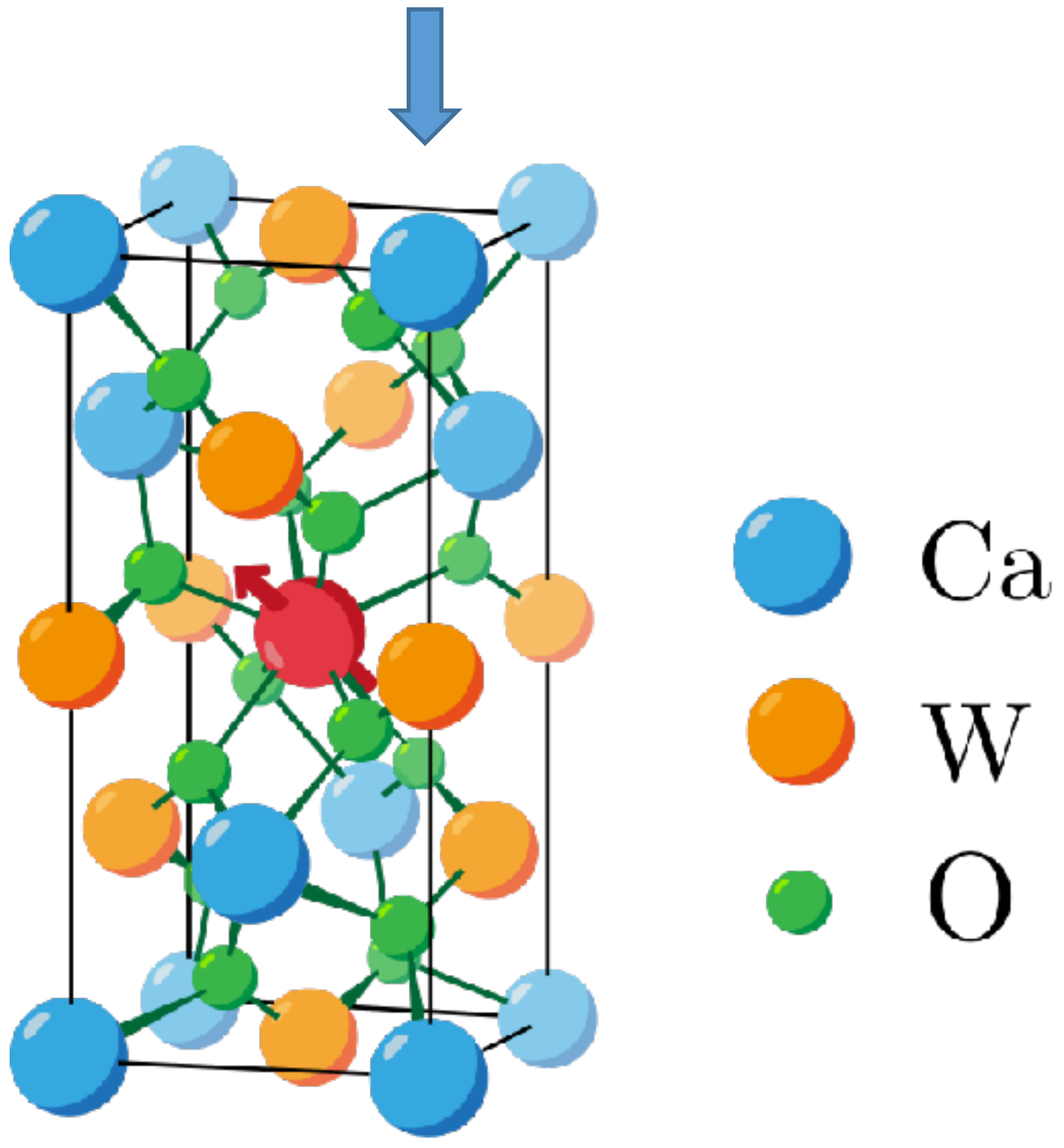
Long-term SMPD operation



L. Balembois et al., in preparation (2022)

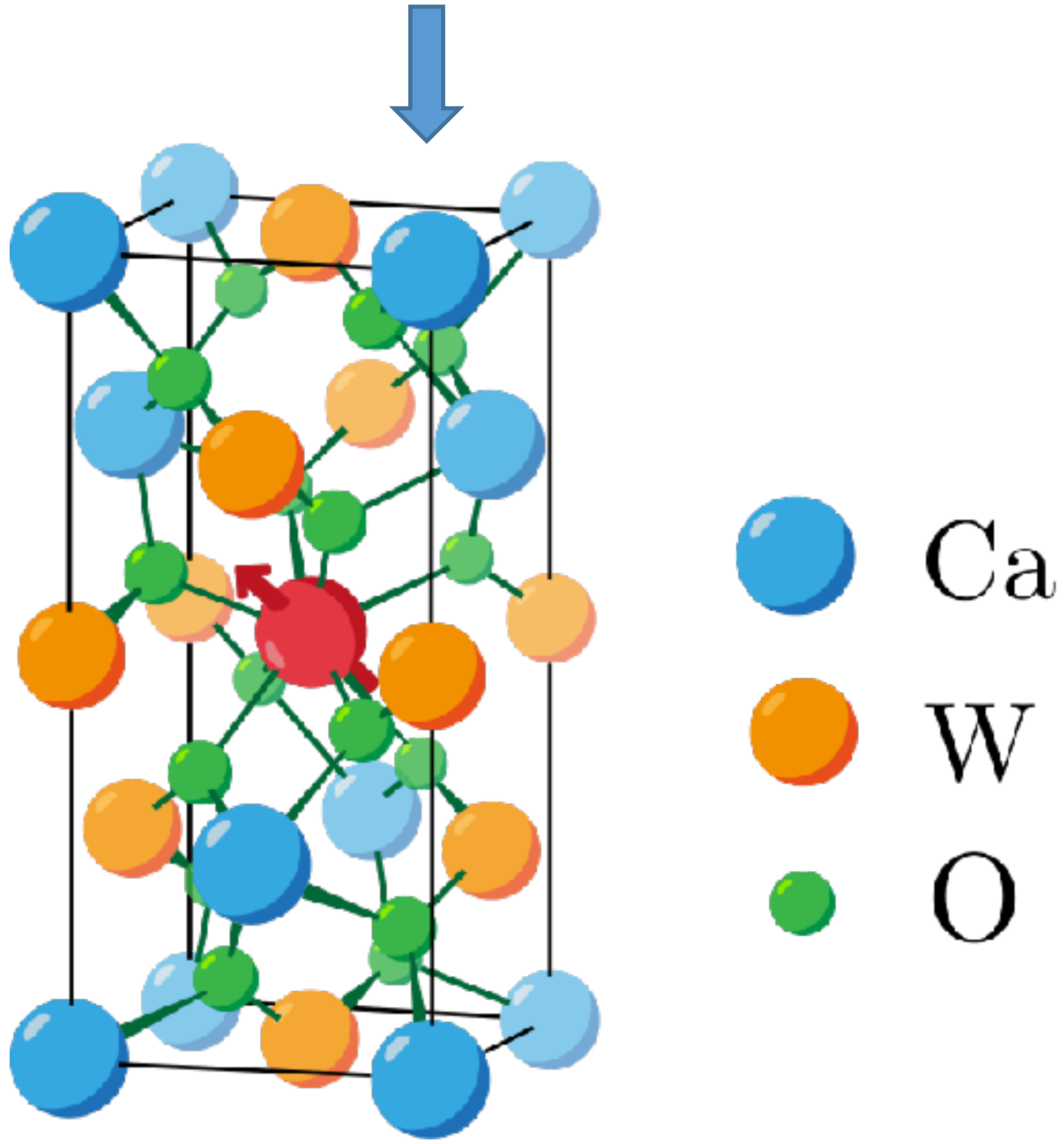
Er : $CaWO_4$

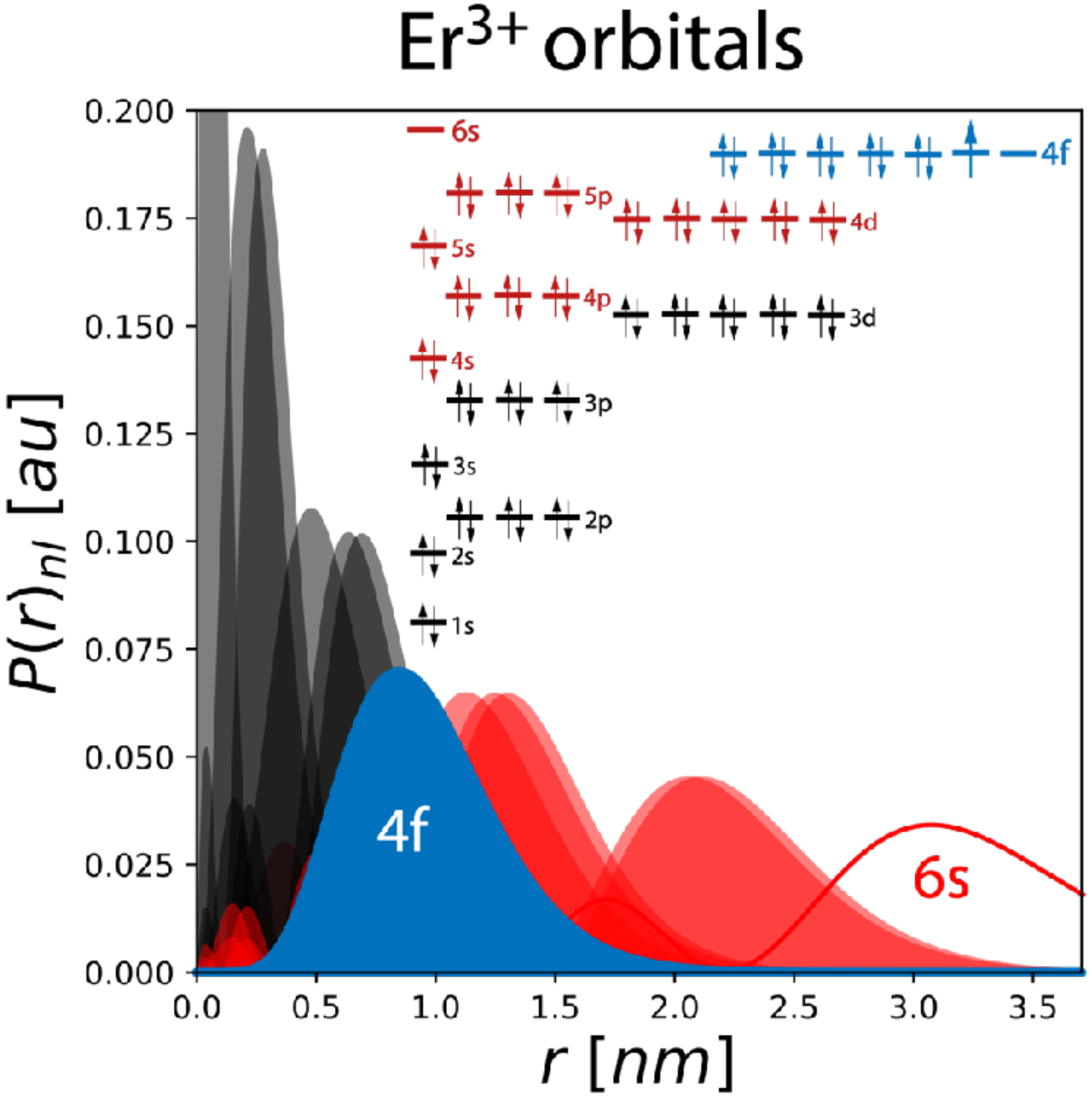
Scheelite



Er : $CaWO_4$

Scheelite

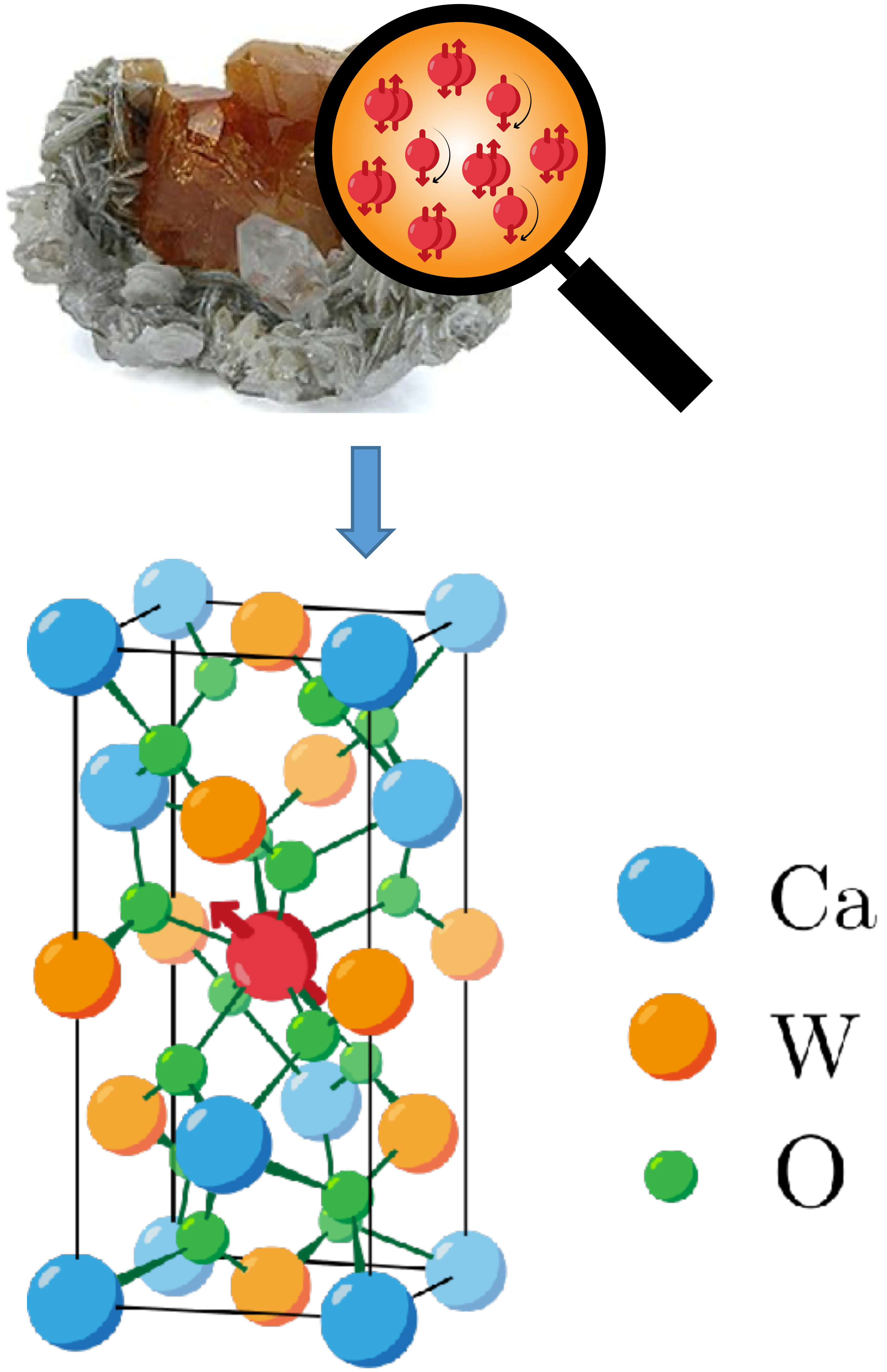


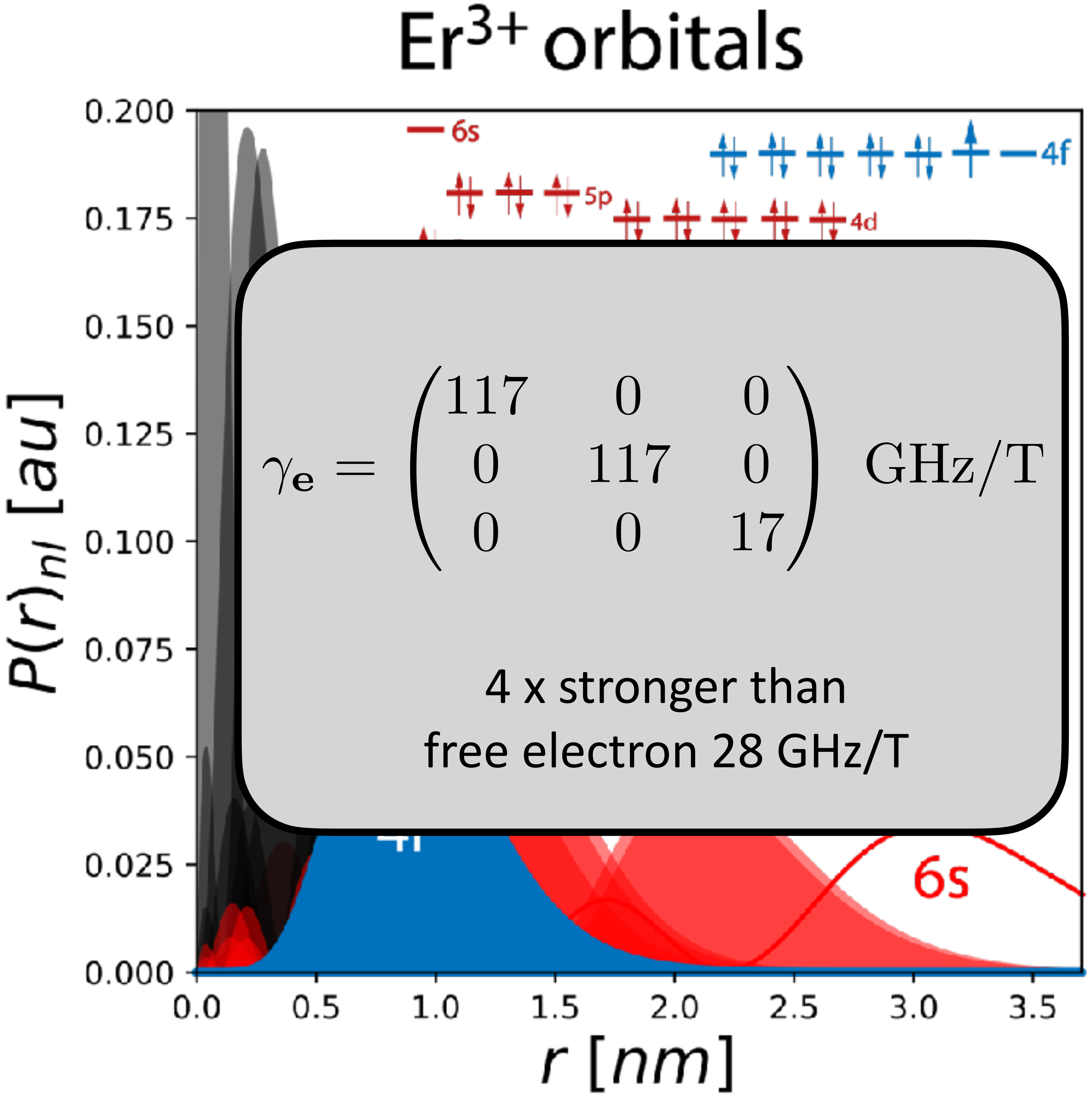


Spins

Er : $CaWO_4$

Scheelite

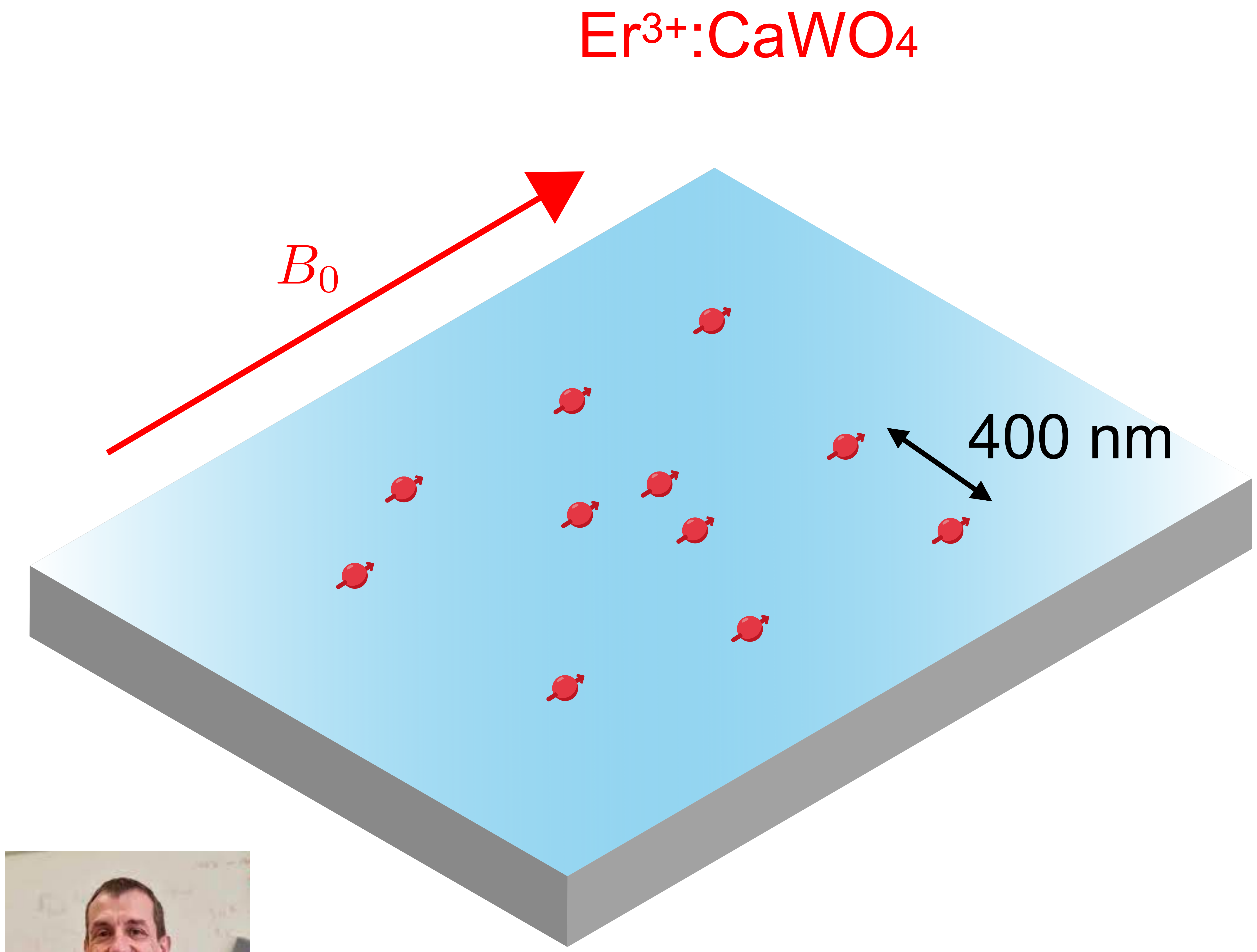


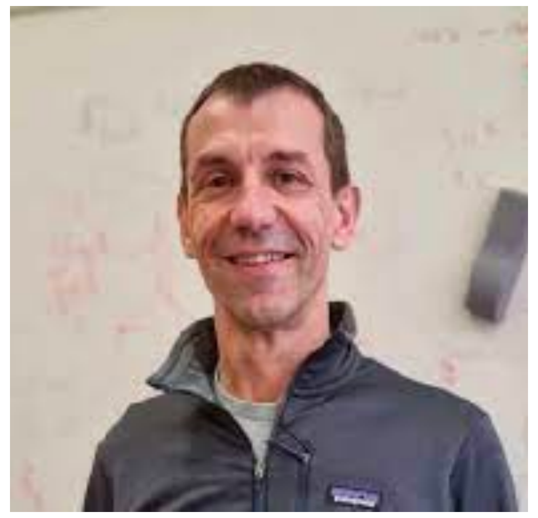


$$\gamma_{\mathbf{e}} = \begin{pmatrix} 117 & 0 & 0 \\ 0 & 117 & 0 \\ 0 & 0 & 17 \end{pmatrix} \ \mathrm{GHz/T}$$

4 x stronger than
free electron 28 GHz/T

Coupling spins to a microwave resonator





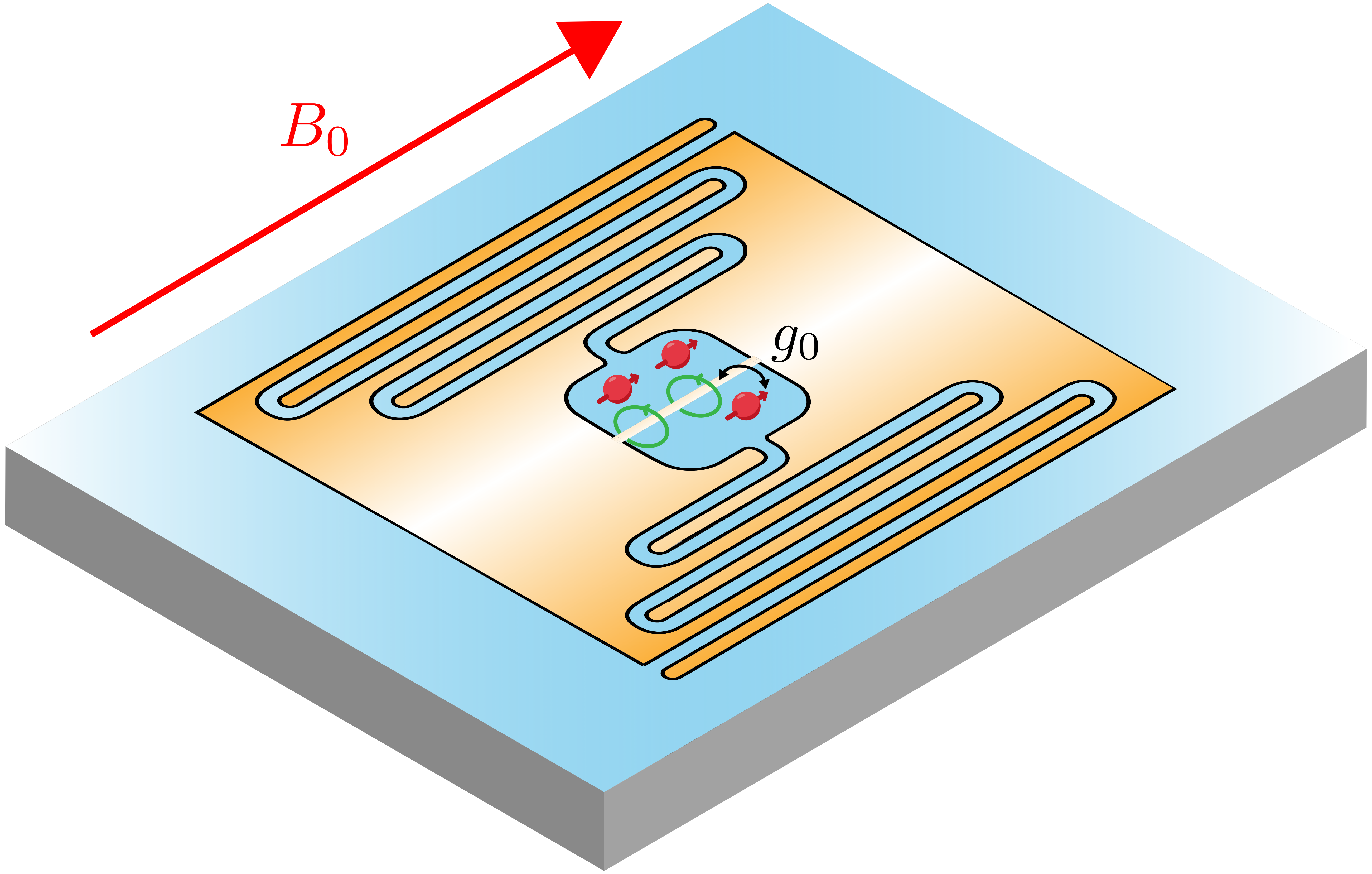
P. Goldner

Spin relaxation in the microwave domain

Photons in free space
~1000 years

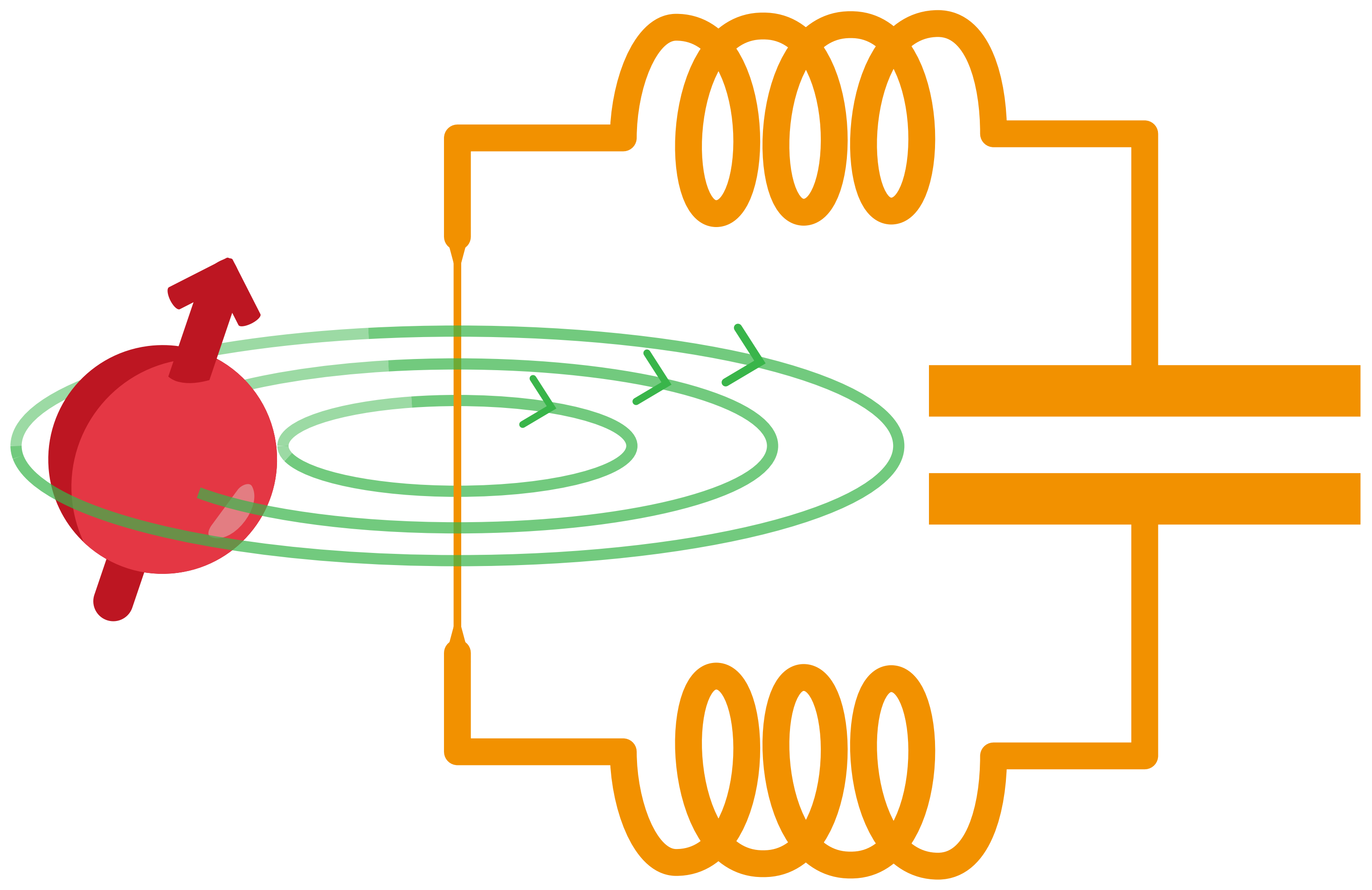
Phonons in lattice
~1 s

Coupling spins to a microwave resonator



Spin – microwave photon coupling

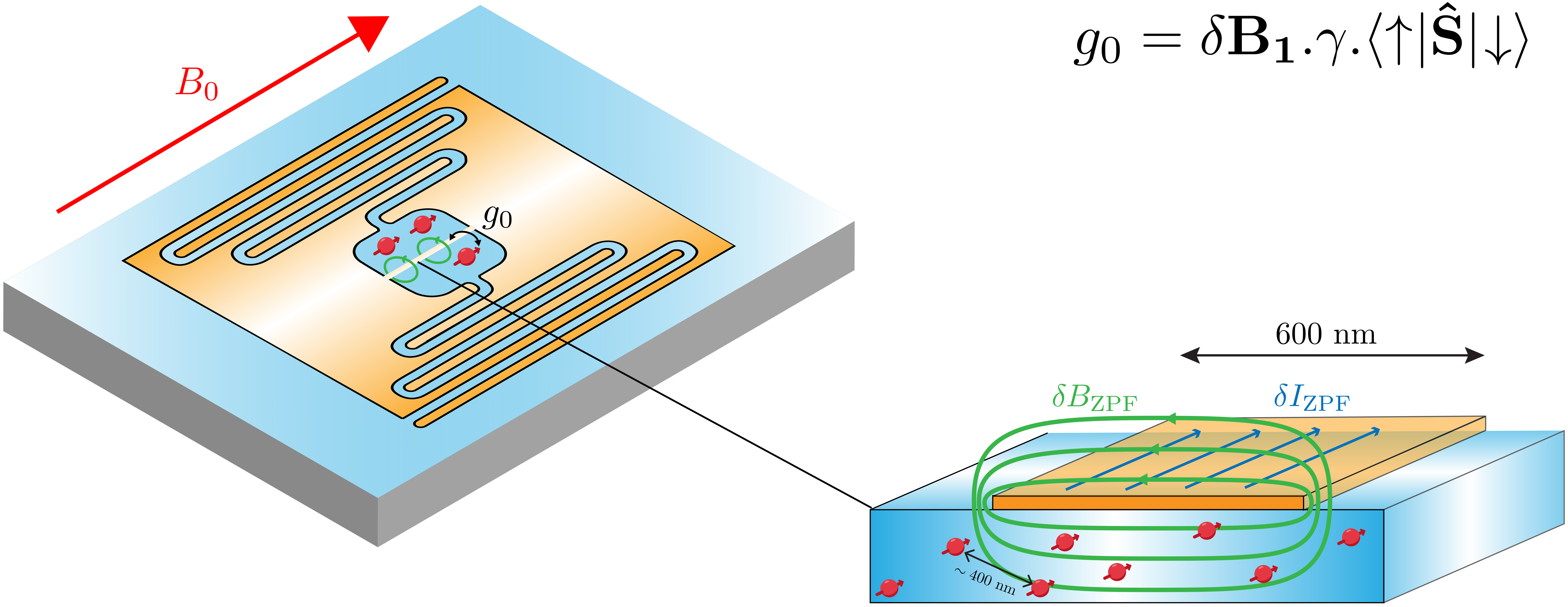
$$g_0 = \delta \mathbf{B_1} . \gamma . \langle \uparrow | \hat{\mathbf{S}} | \downarrow \rangle$$



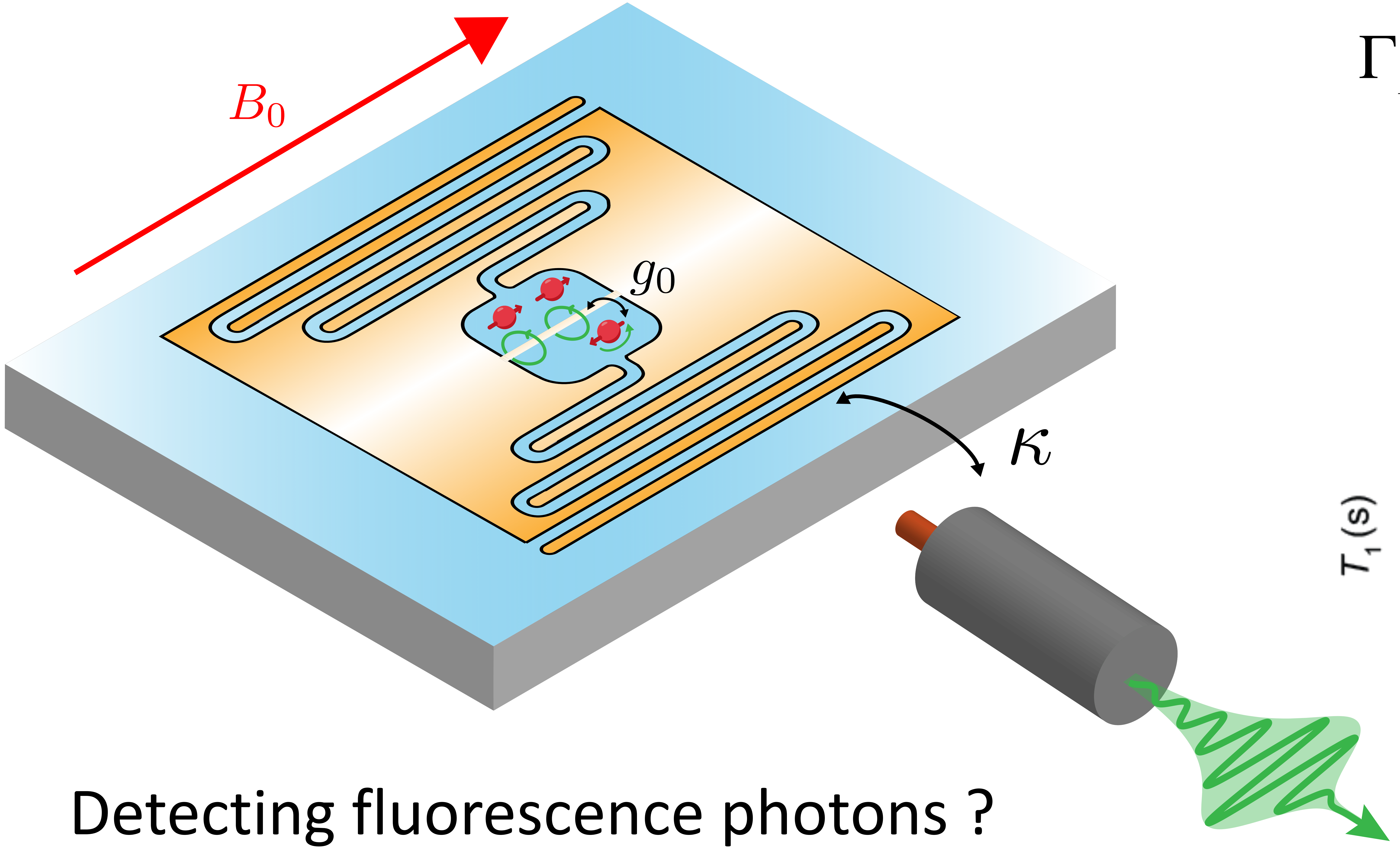
Coupling spins to a microwave resonator

Spin – microwave photon coupling

$$g_0 = \delta \mathbf{B_1} . \gamma . \langle \uparrow | \hat{\mathbf{S}} | \downarrow \rangle$$

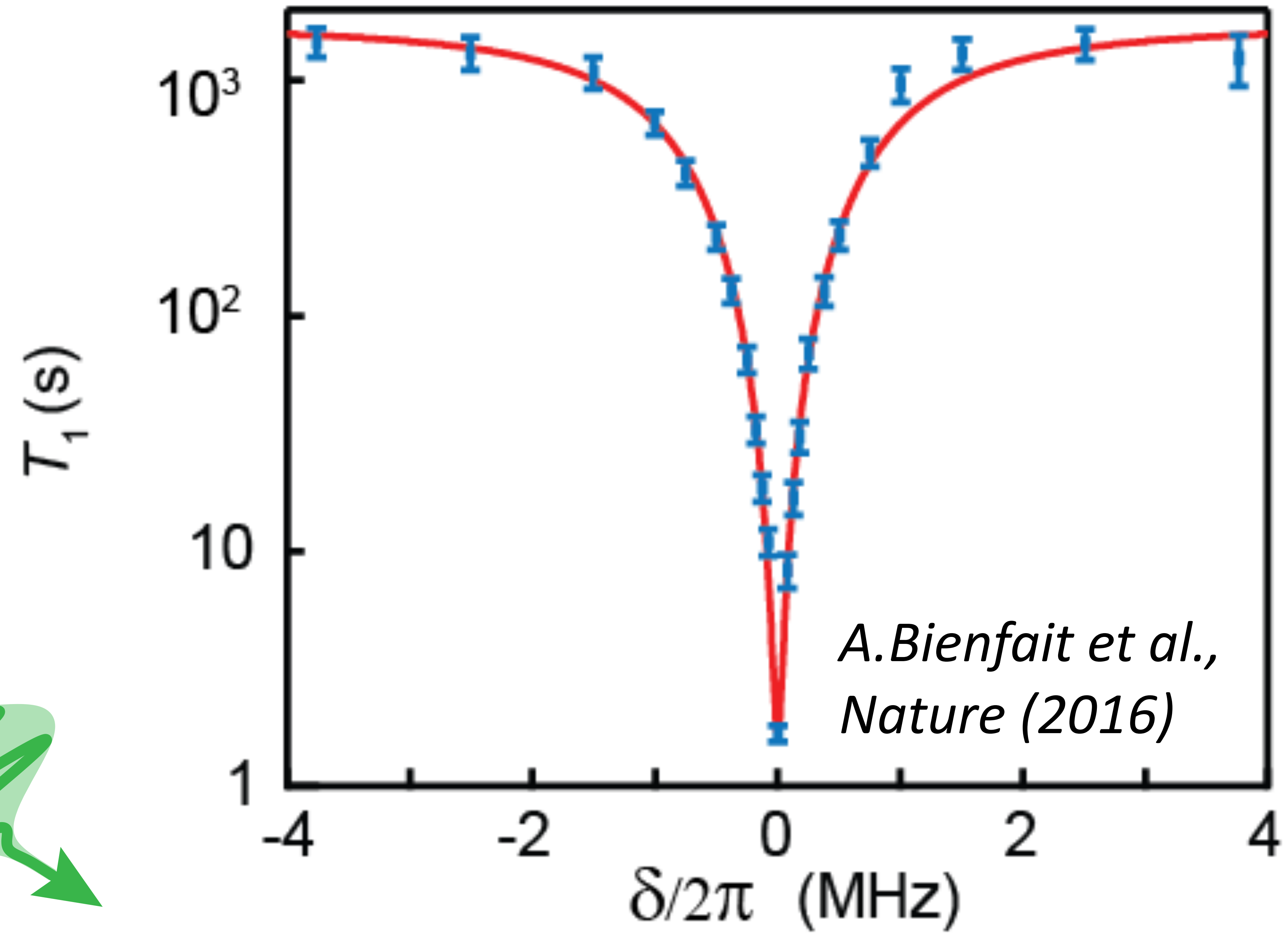


Purcell Effect



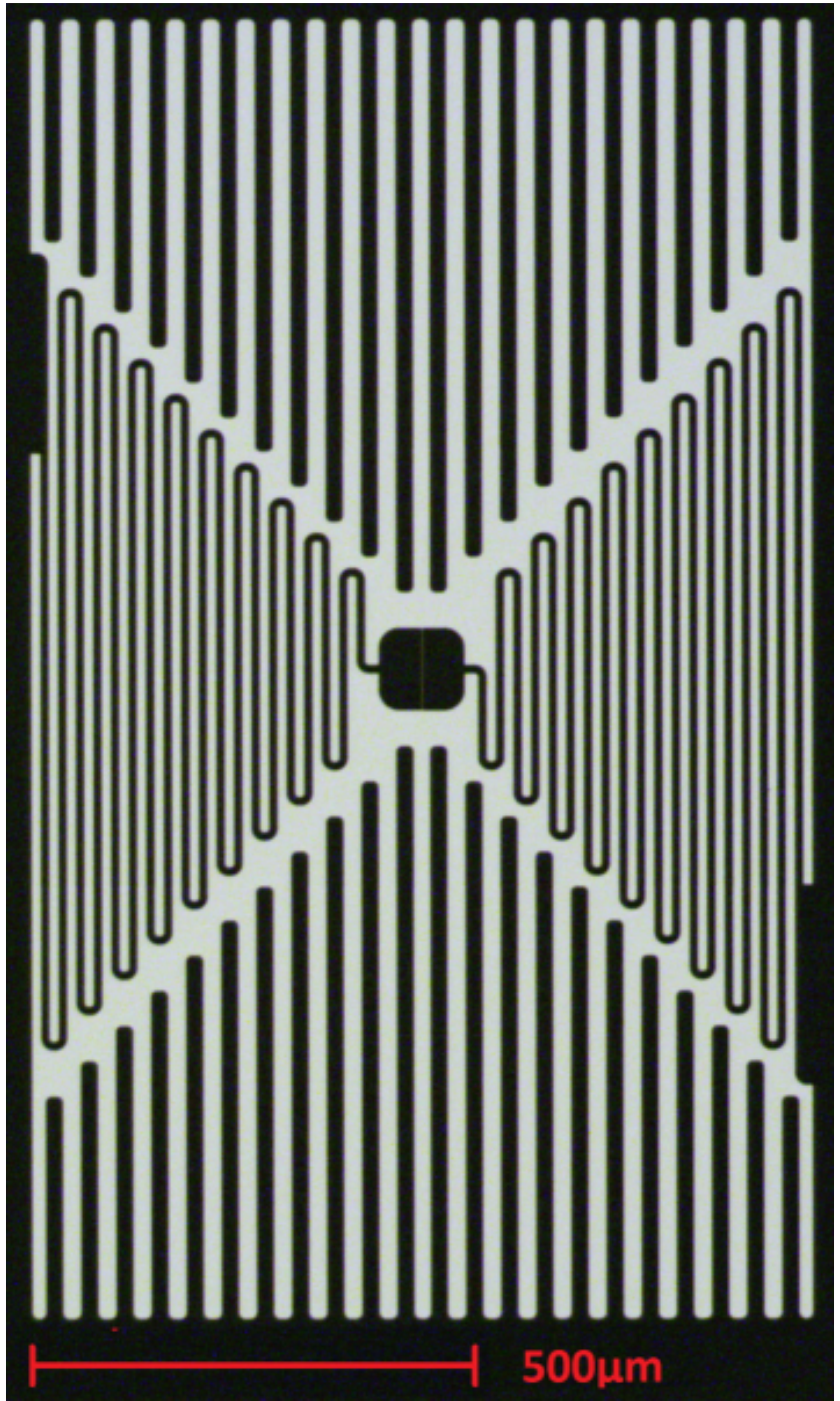
$$\Gamma_R = \frac{4g_0^2}{\kappa}\frac{1}{1+4\left[\frac{\delta}{\kappa}\right]^2}$$

Audrey Bienfait

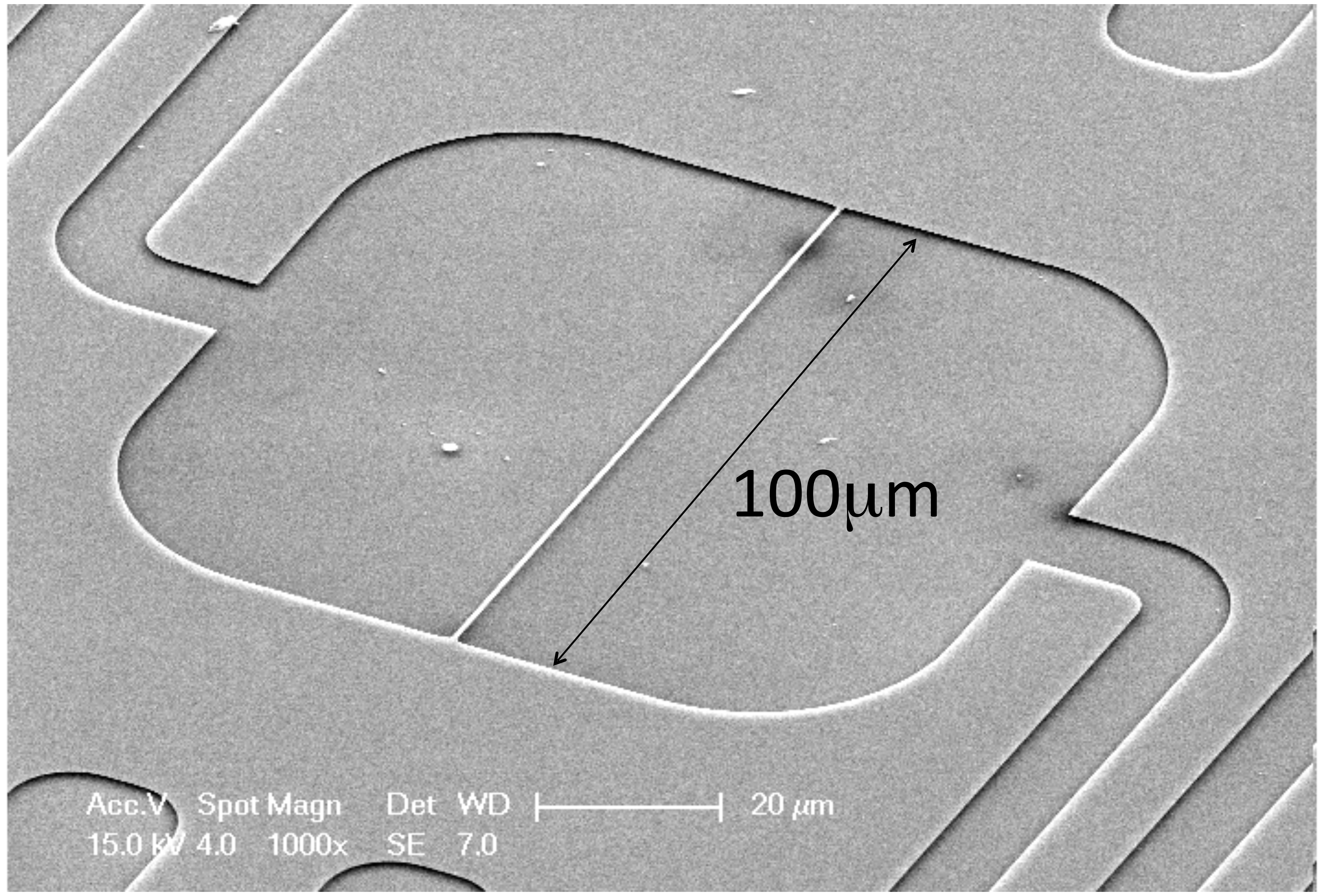


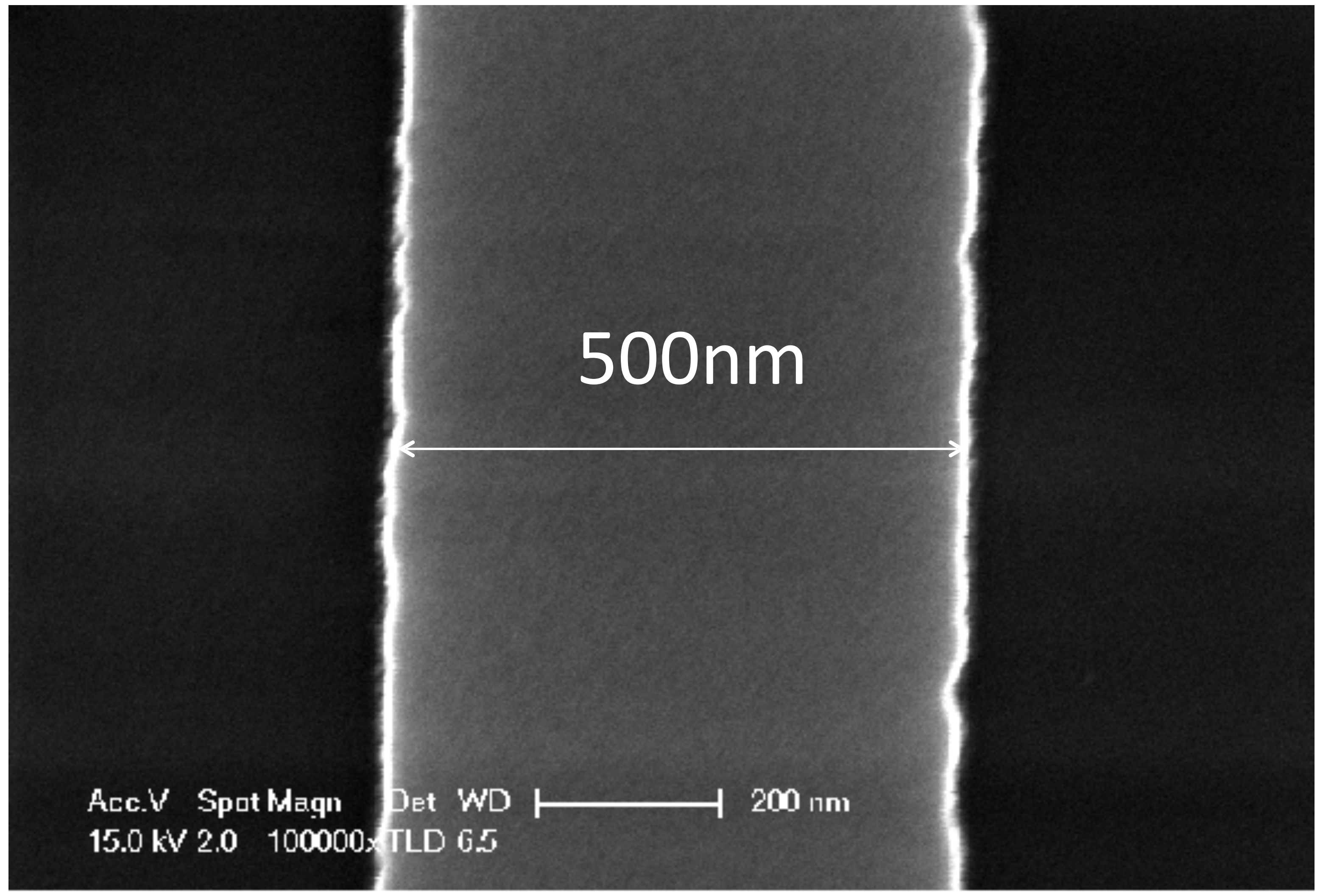
Detecting fluorescence photons ?

Single Spin Detection



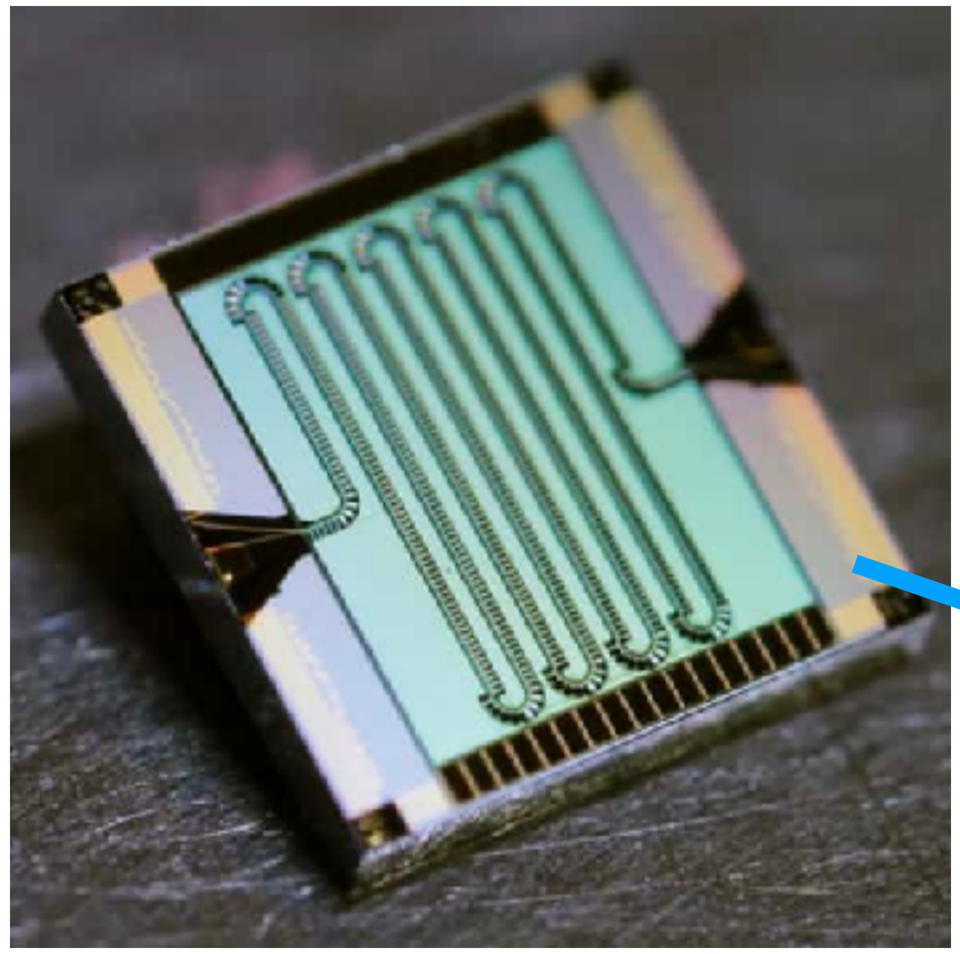
B_0// c-axis

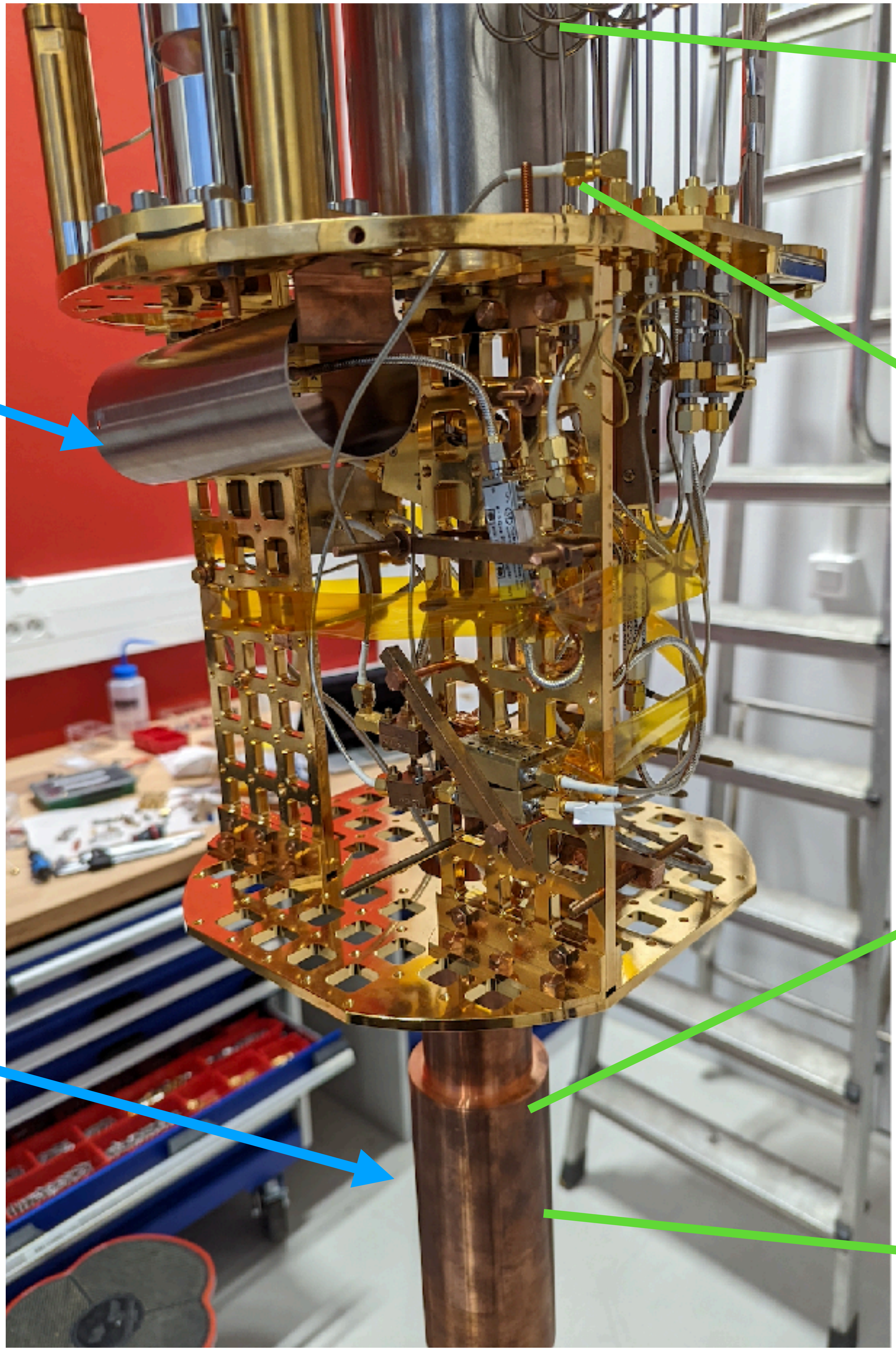




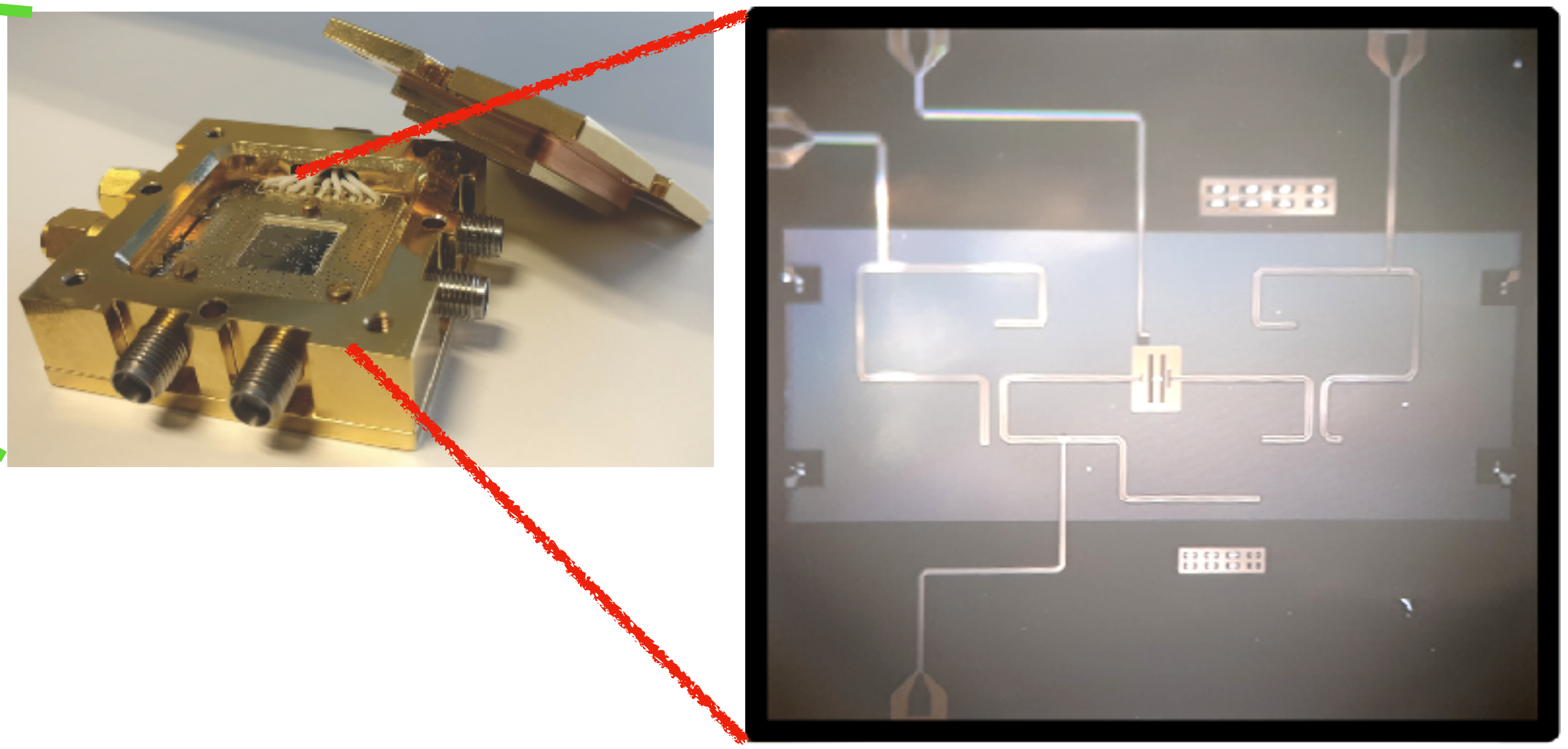
Experiment — Nanowire resonator on Er:CaWO4

MIT JTWPA

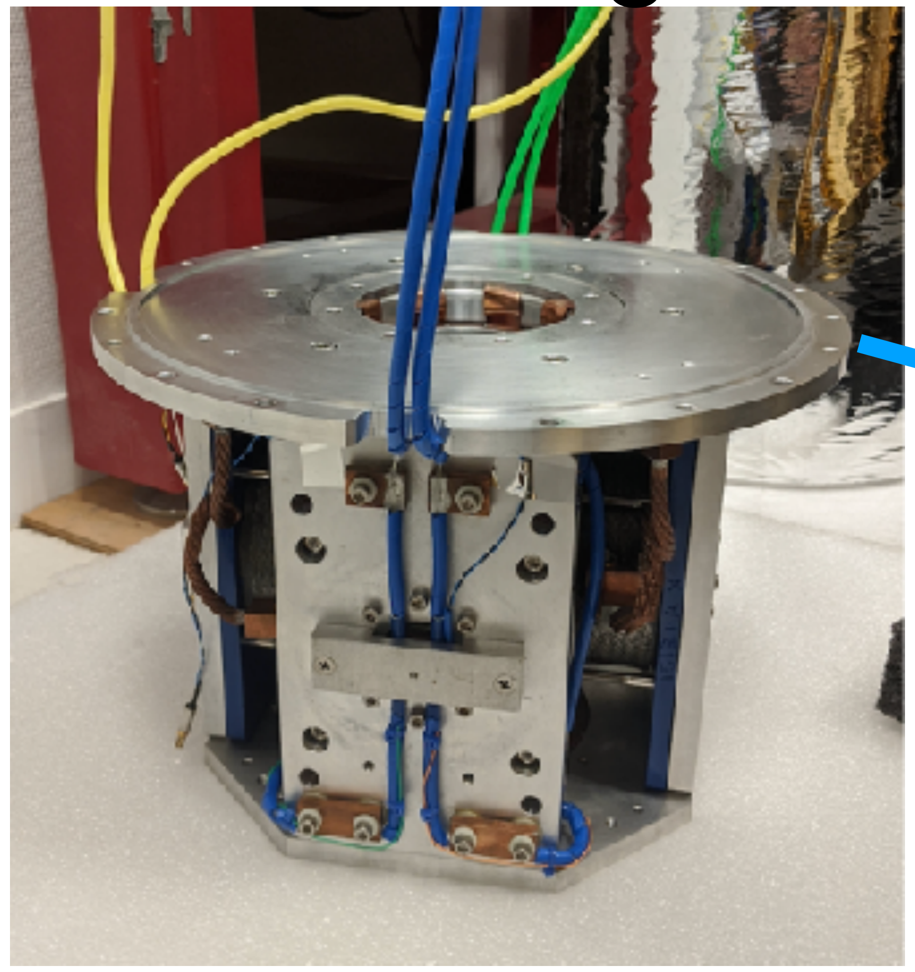


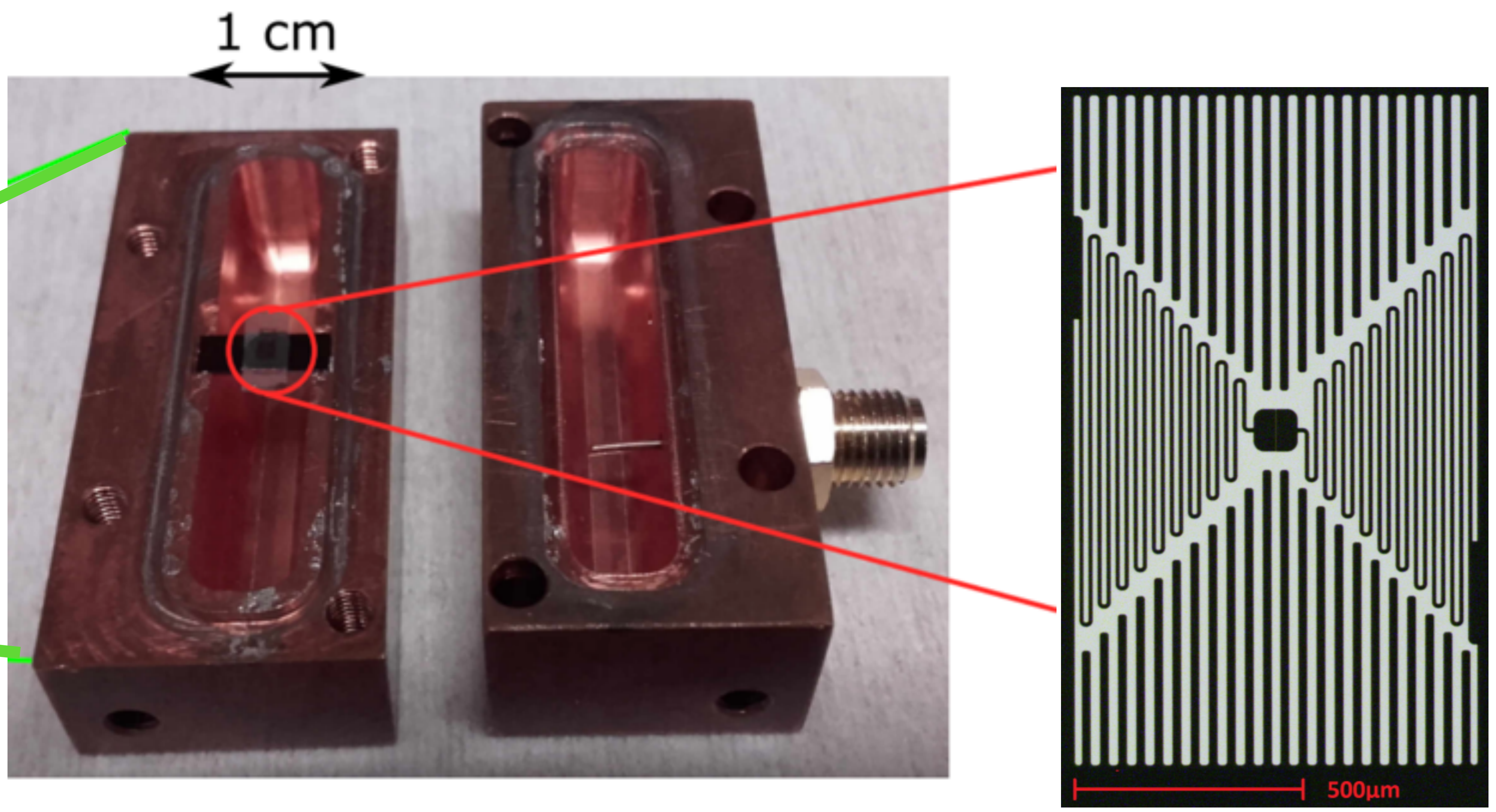


Single Microwave Photon Detector

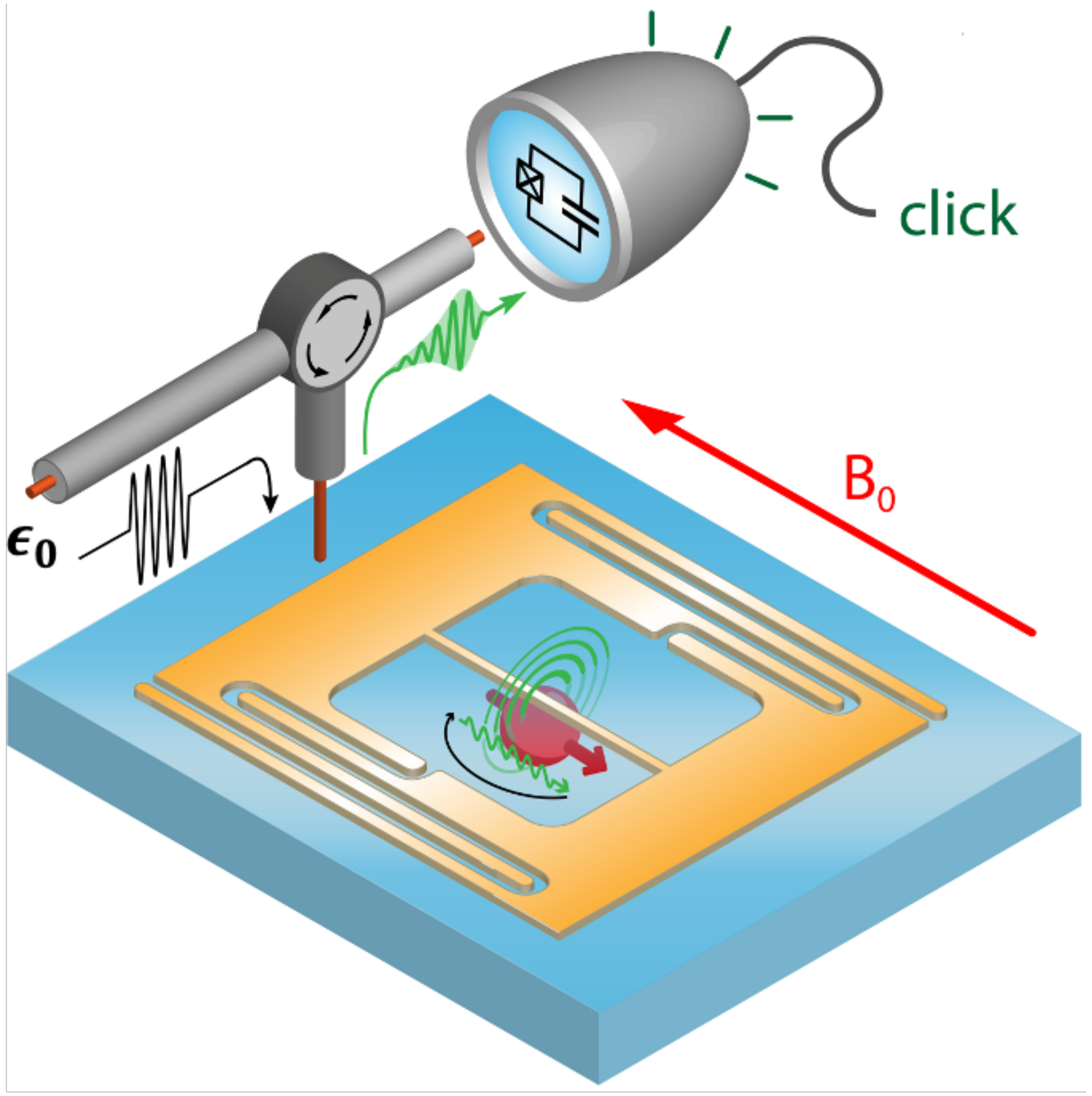


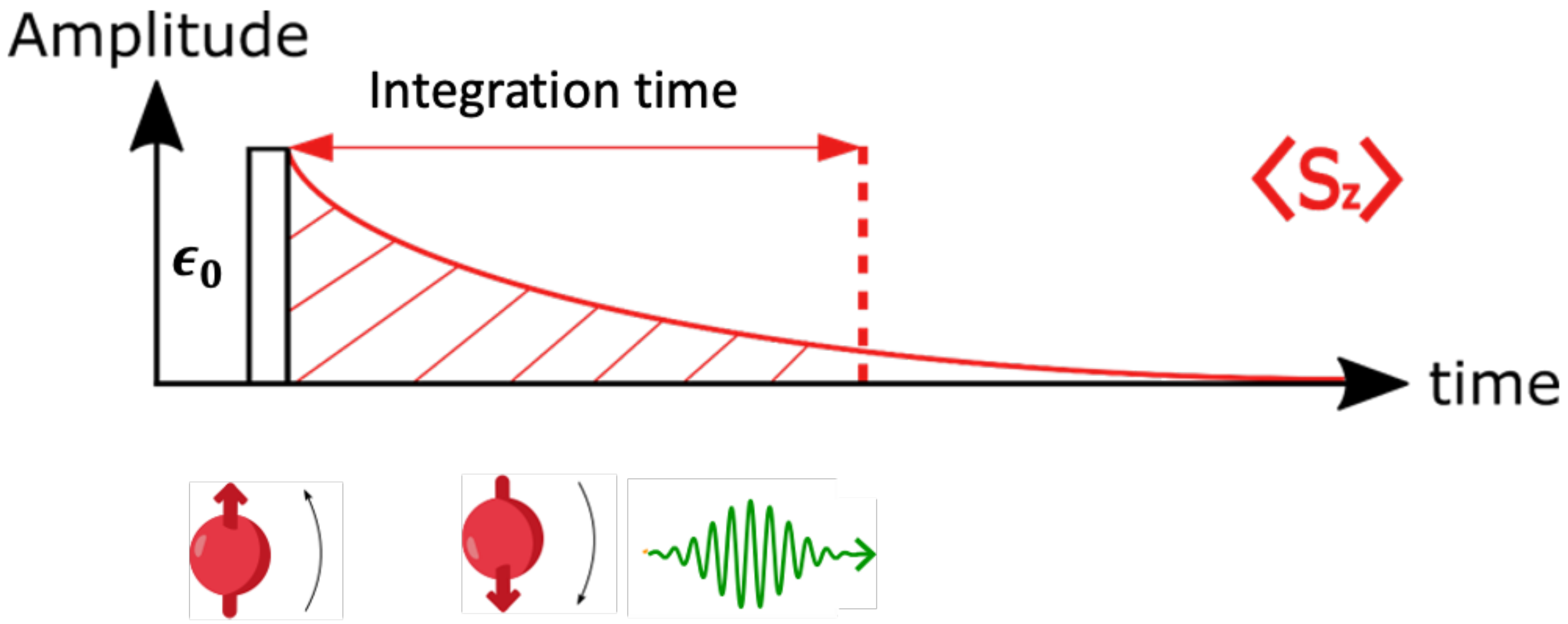
1T/1T/1T
vector magnet



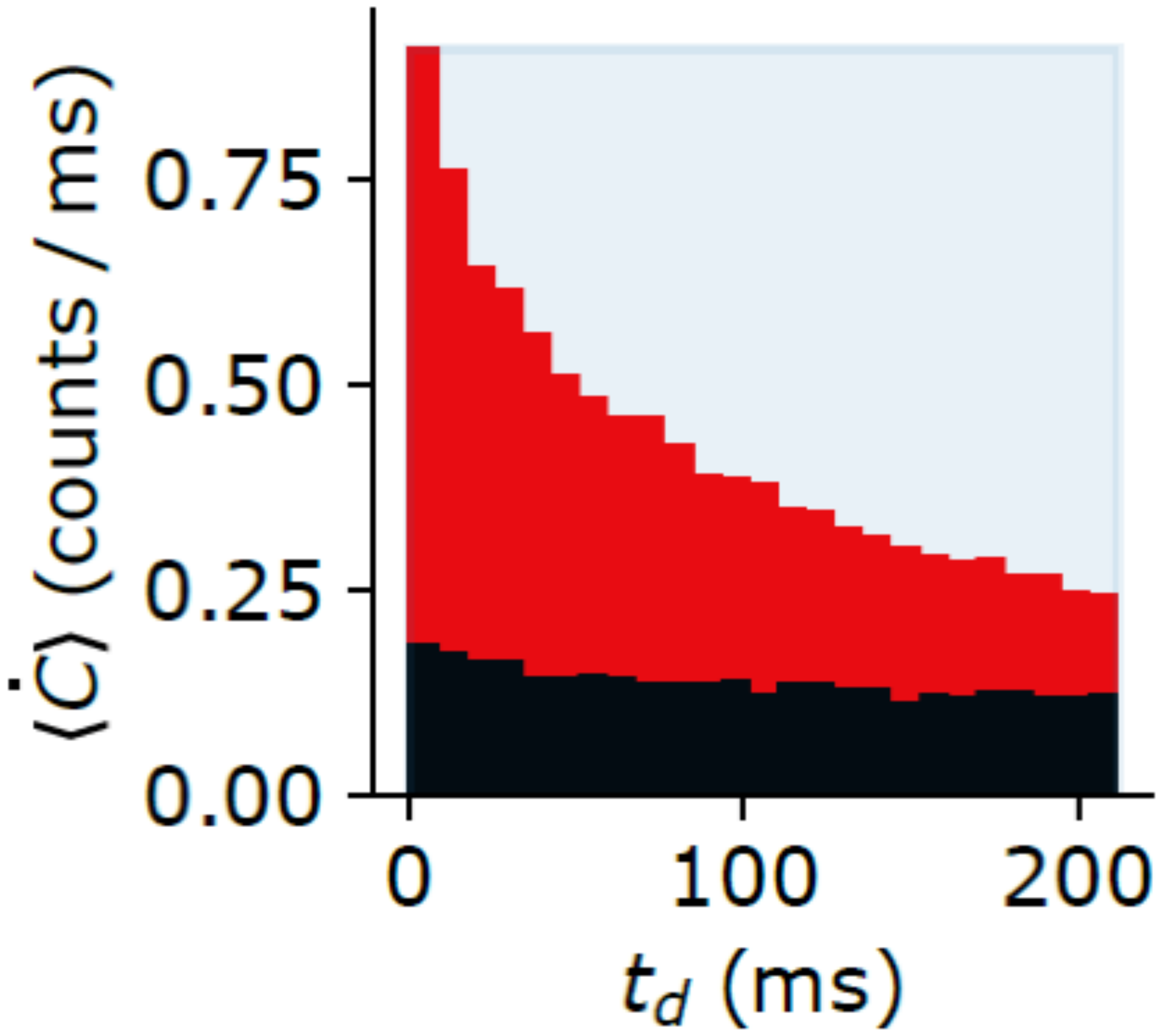


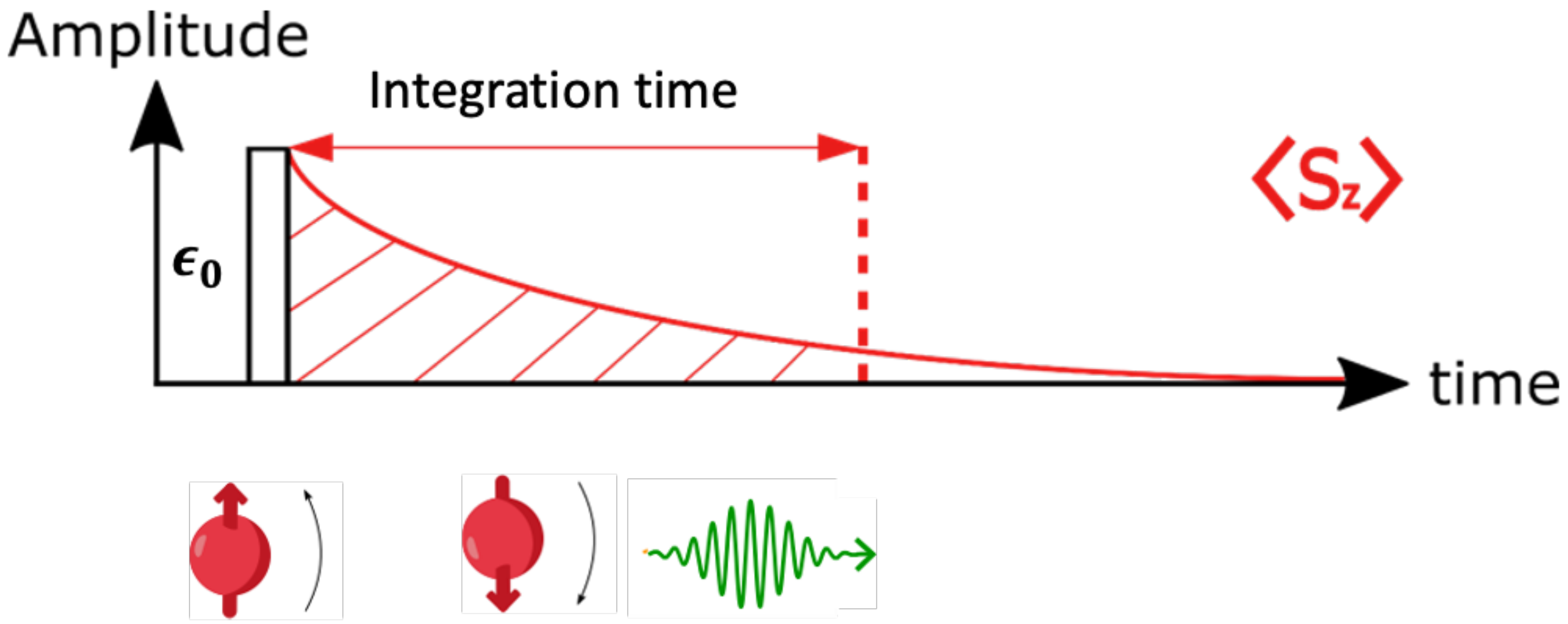
Microwave fluorescence detection of spins





Microwave fluorescence detection of spins

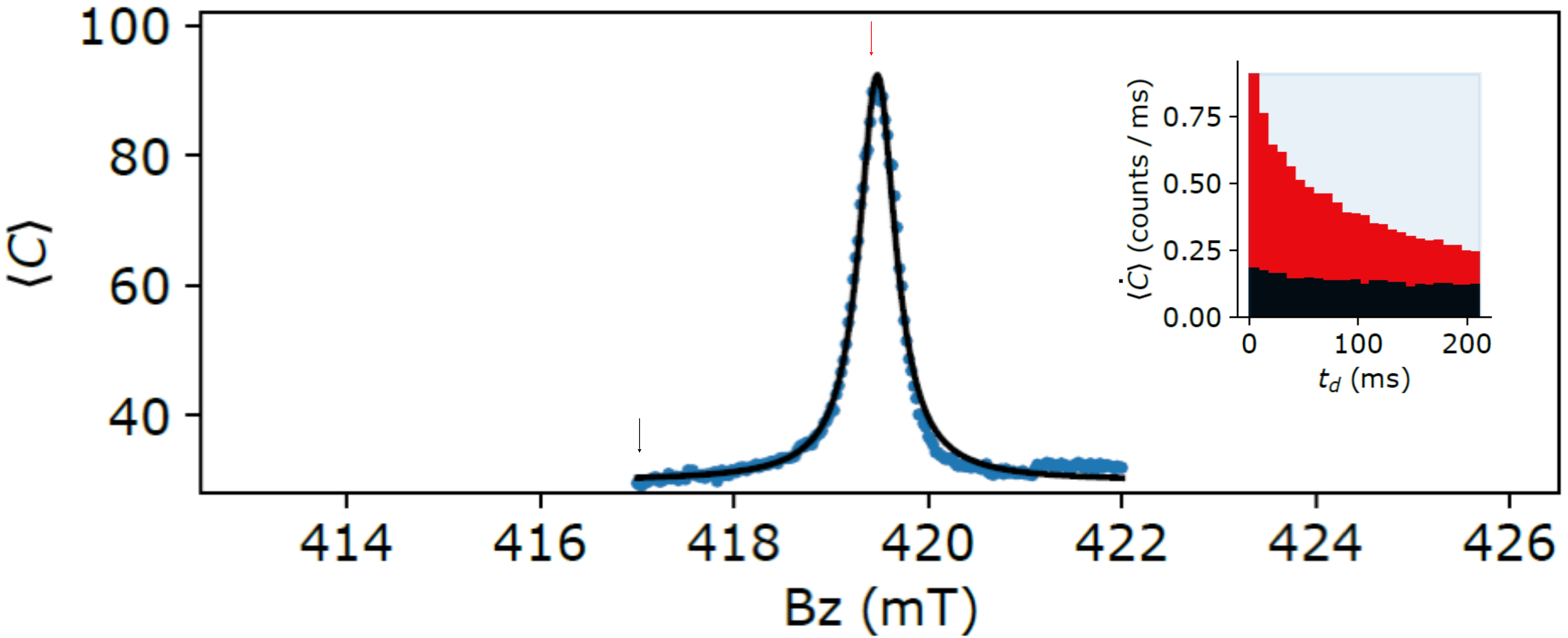




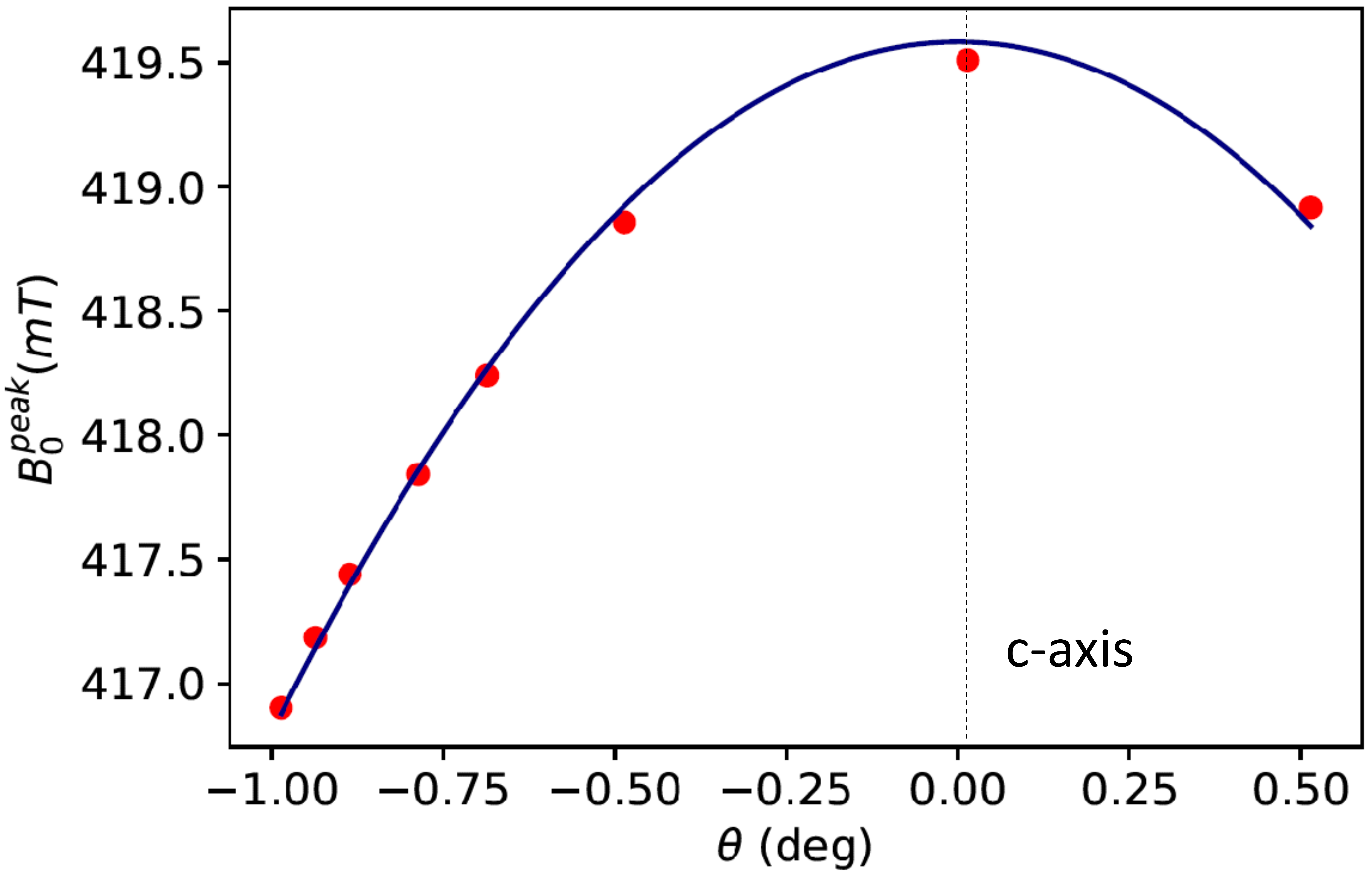
Detecting spins by their fluorescence
with a microwave photon counter
Nature **600,** 434-438 (2021)

High-power spectroscopy

Integration time : 200ms



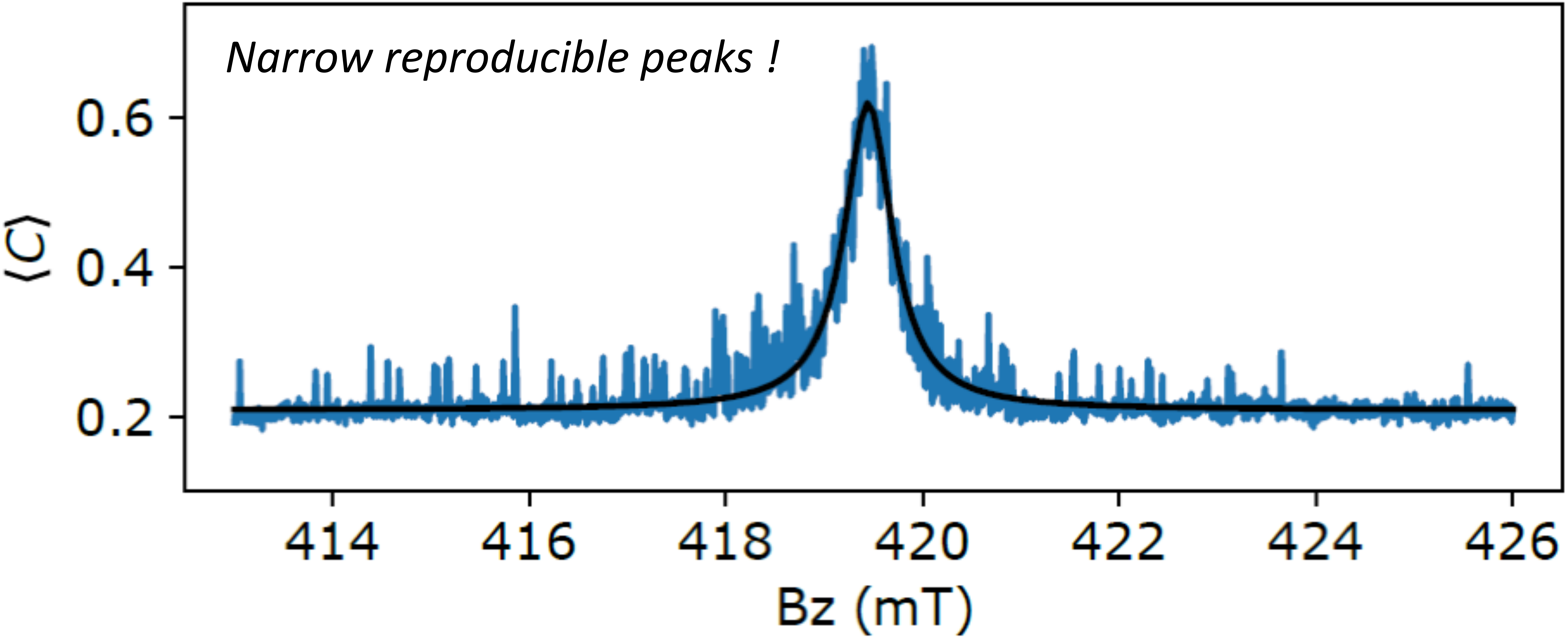
High-power spectroscopy : angular dependence



$$\gamma_{\mathbf{e}} = \begin{pmatrix} 117 & 0 & 0 \\ 0 & 117 & 0 \\ 0 & 0 & 17 \end{pmatrix} \text{ GHz/T}$$

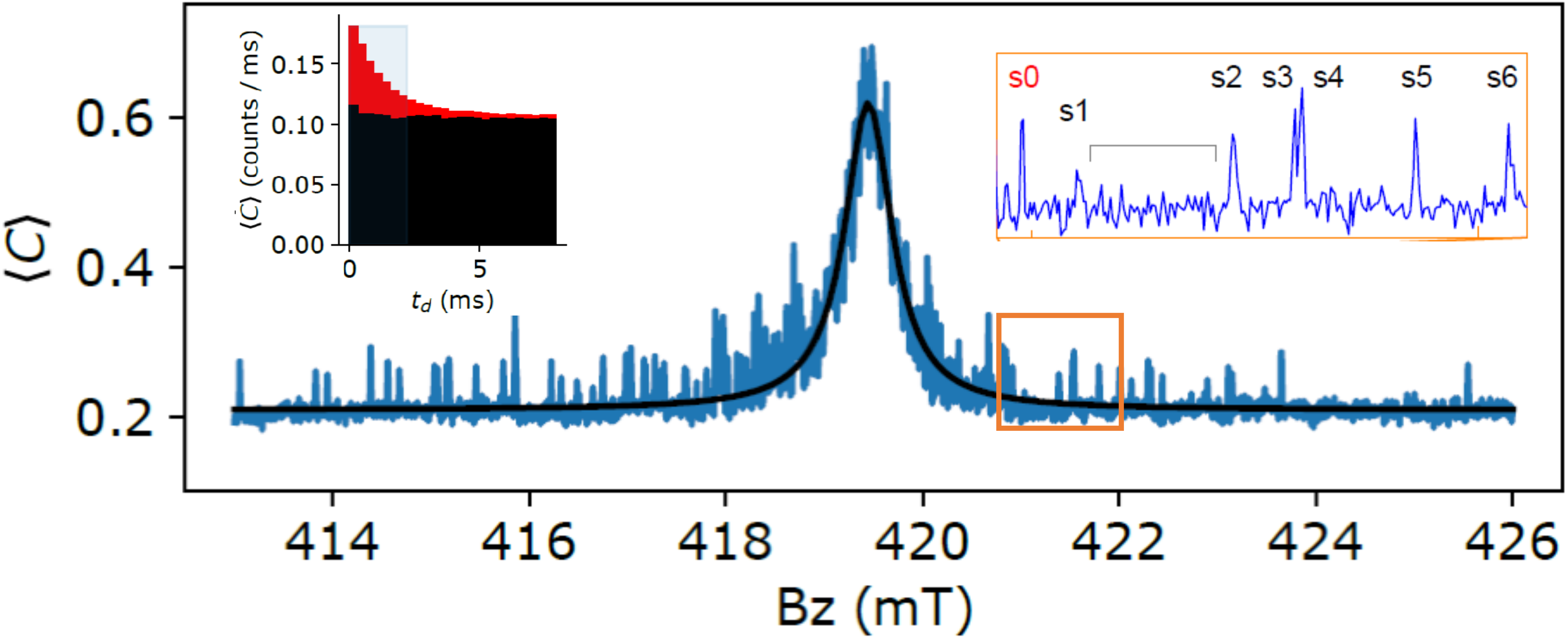
Low-power spectroscopy ($\theta = 0^{\circ}$)

Integration time : 2ms



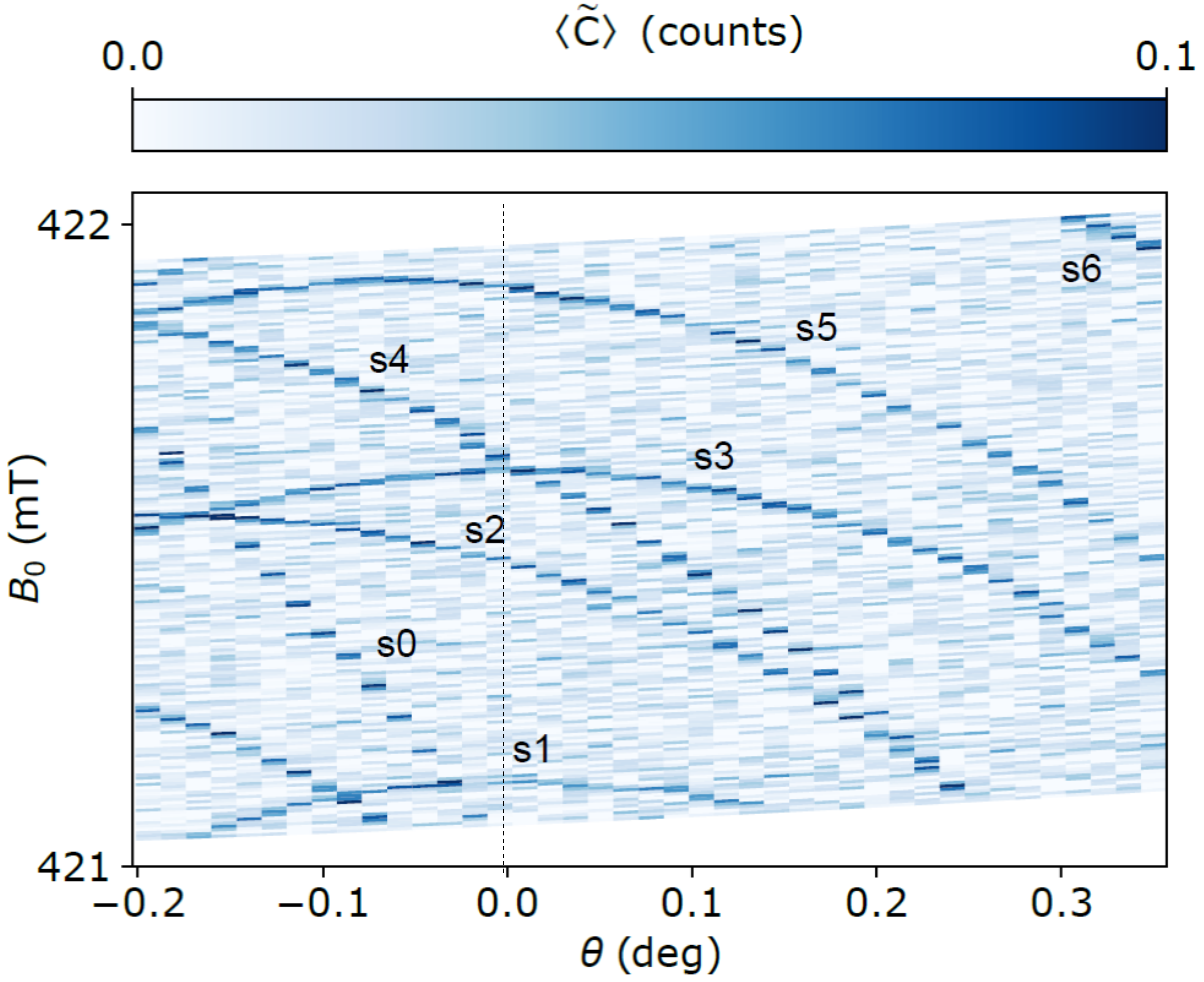
Z. Wang et al., arxiv (2023)

Low-power spectroscopy ($\theta = 0^\circ$)



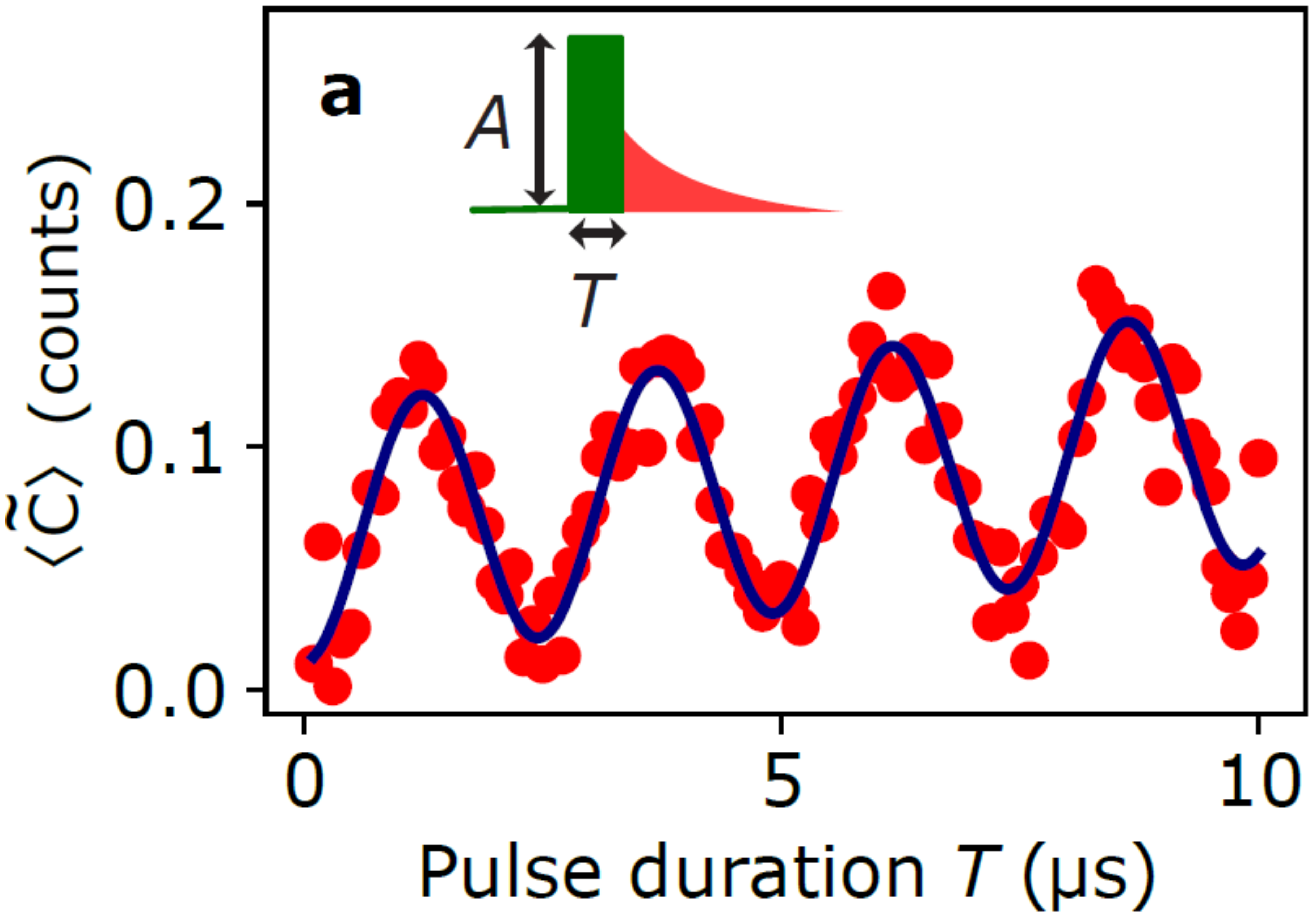
Z. Wang et al., arxiv (2023)

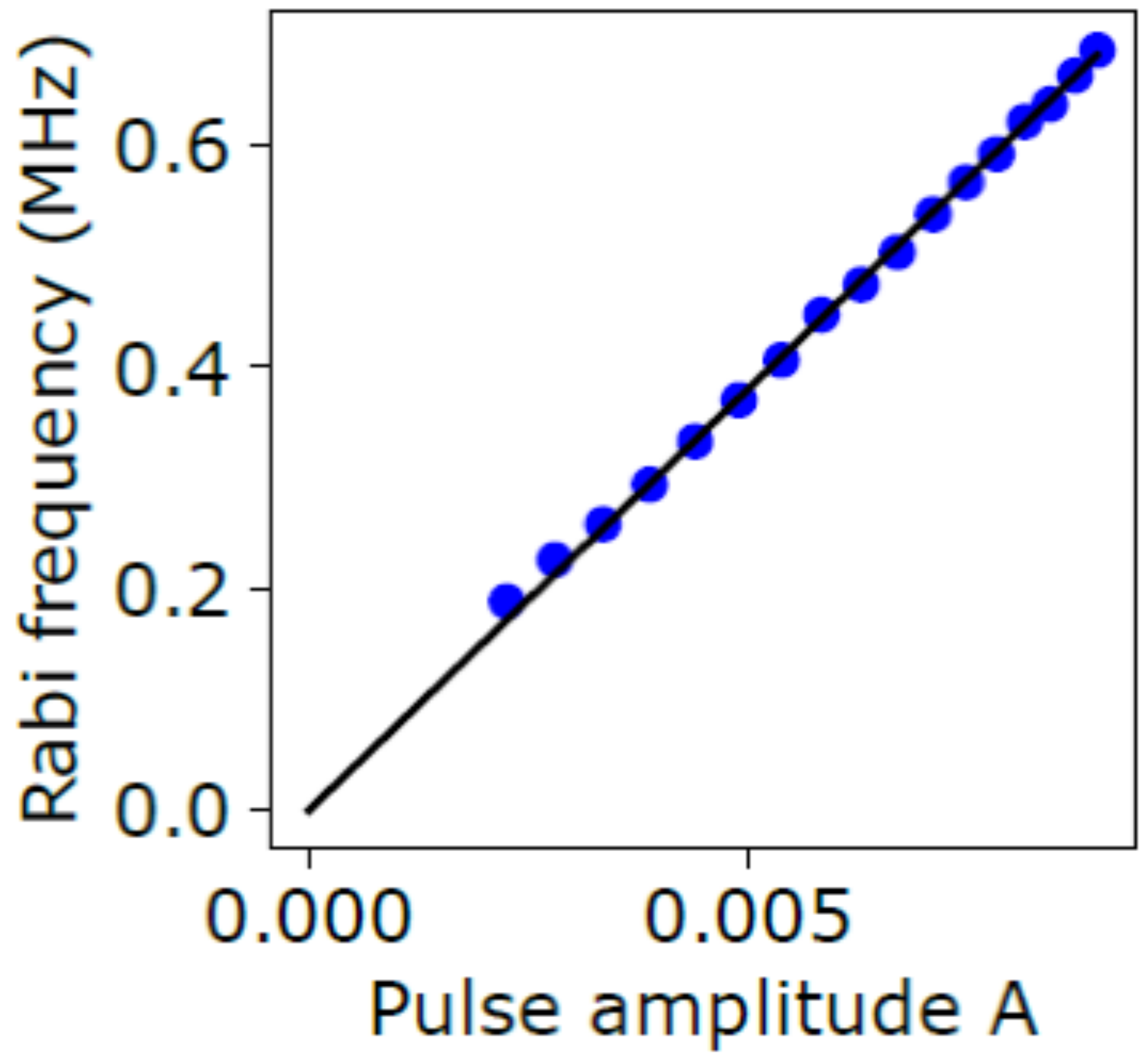
Low-power spectroscopy : angular dependence



Z. Wang et al.,arxiv (2023)

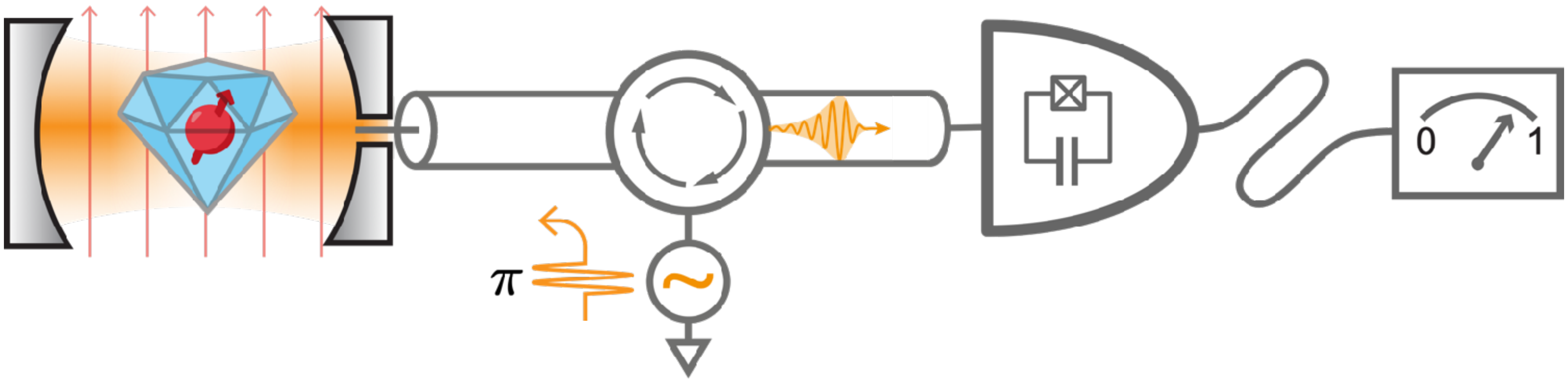
Rabi oscillations





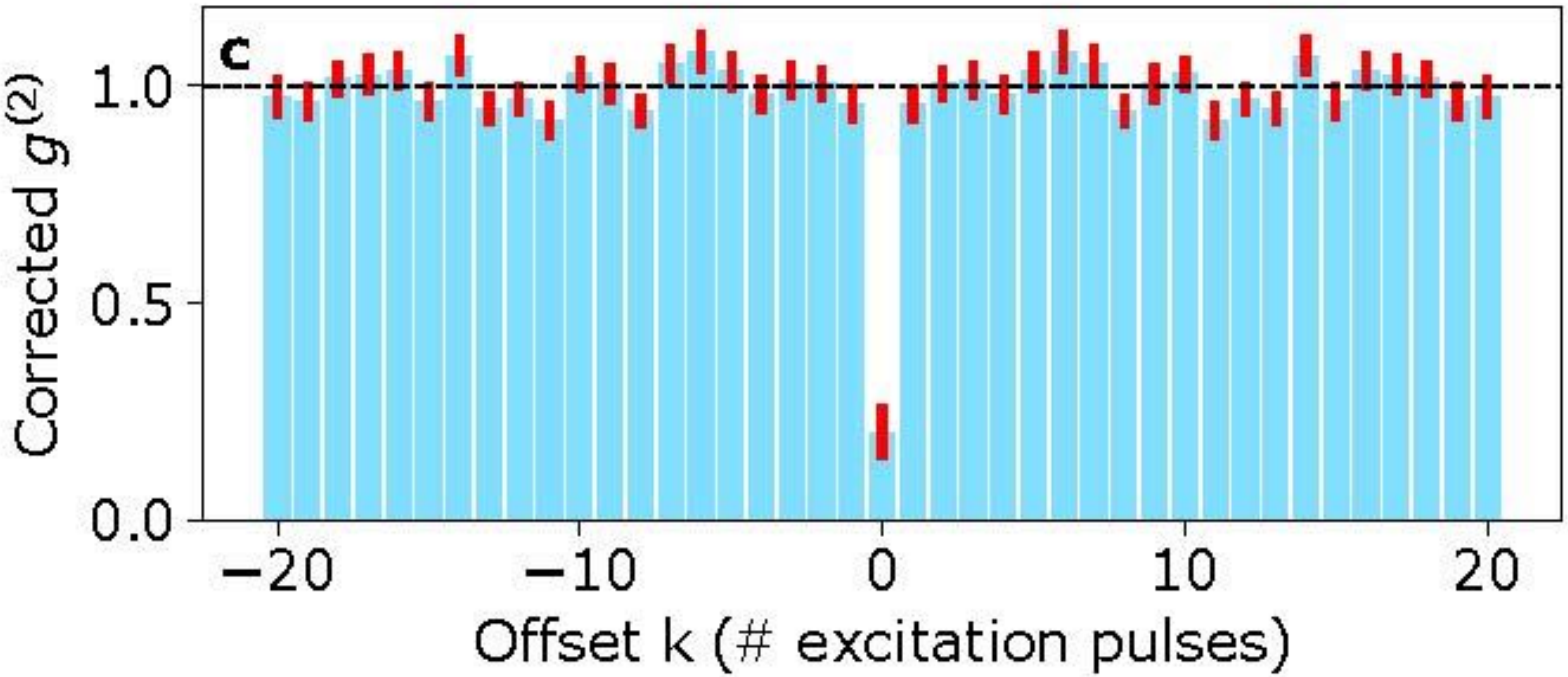
Z. Wang et al., arxiv (2023)

Intensity-intensity correlations



$$g^{(2)}(\tau) = \frac{\langle C(0)C(\tau)\rangle}{\langle C(0)\rangle\langle C(\tau)\rangle}$$

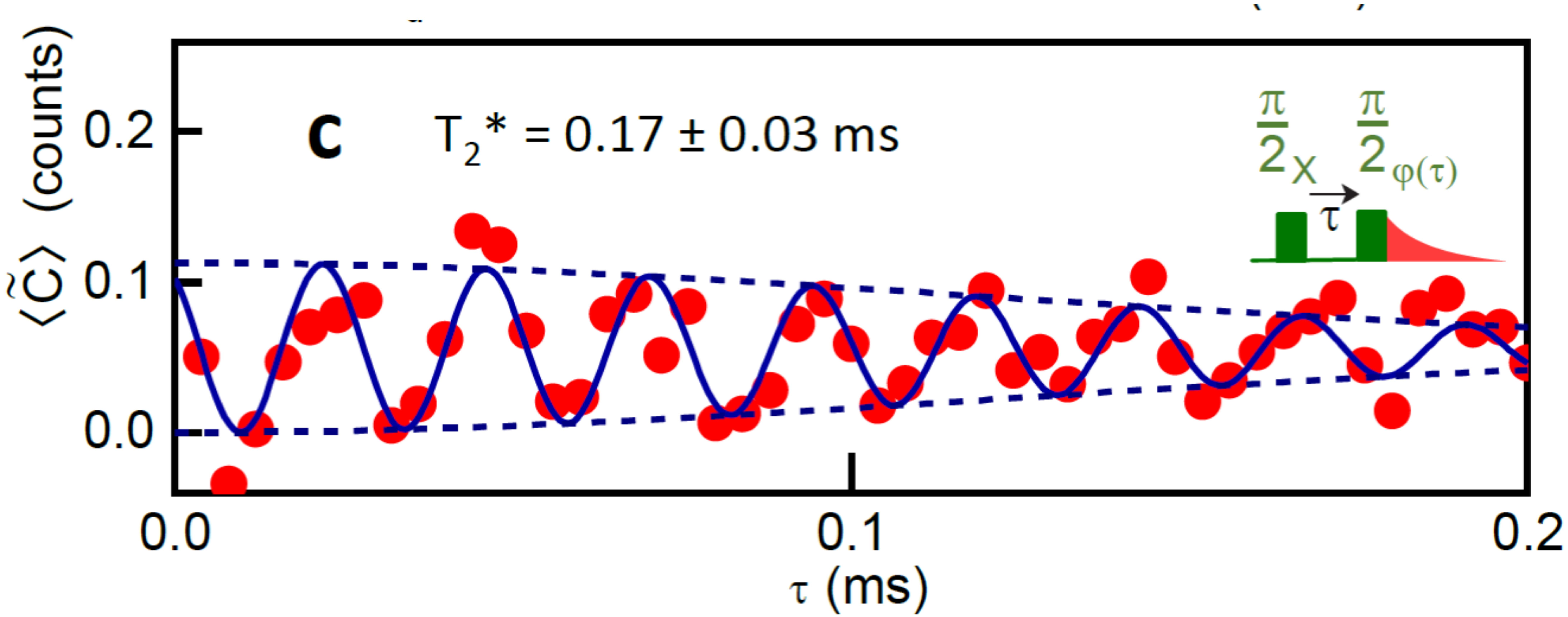
Intensity-intensity correlation function



$g^2(0) = 0.23 \pm 0.06 < 0.5$ → Single-spin emision

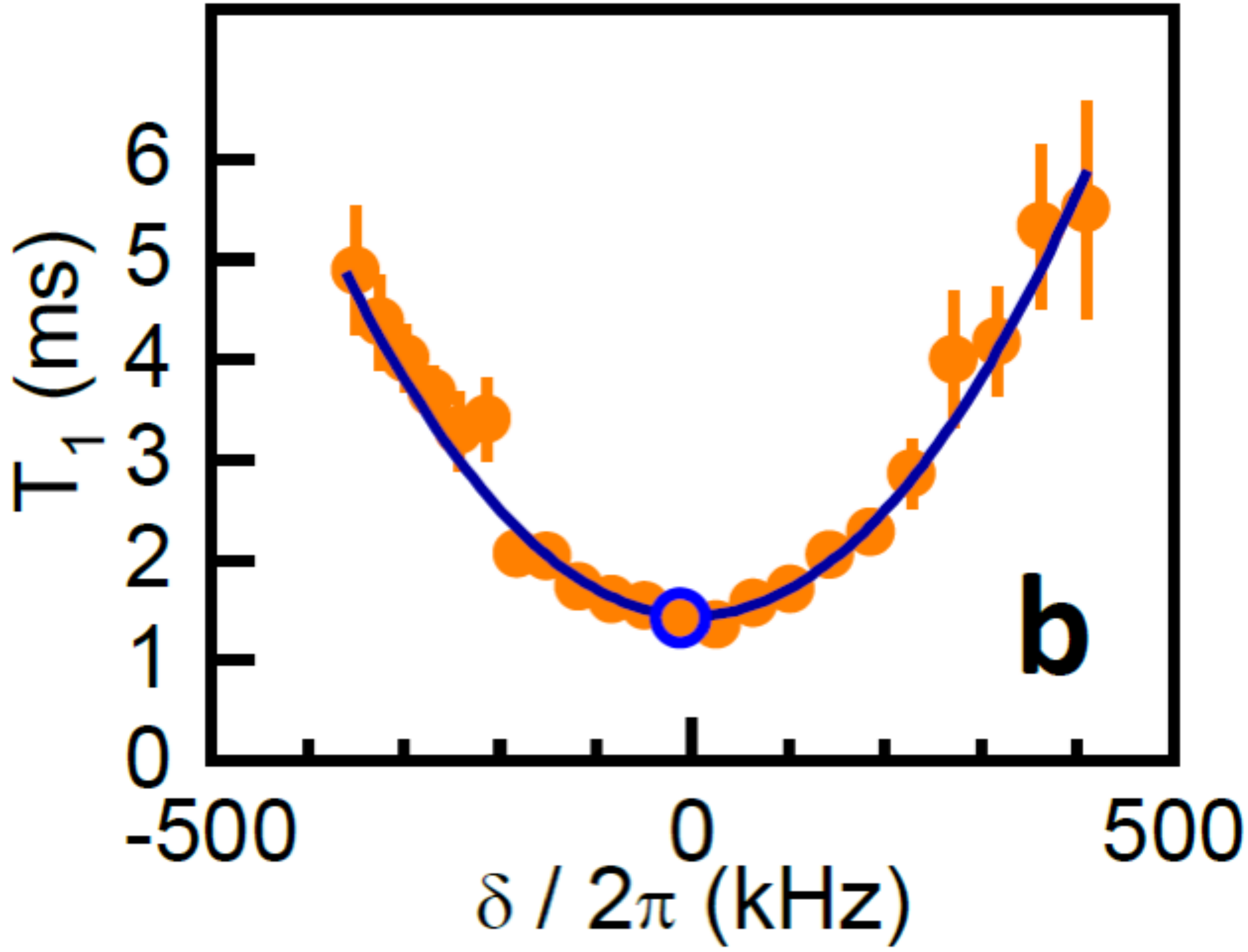
Z. Wang et al., arxiv (2023)

Single-ion coherence time (1) : Ramsey



Z. Wang et al., arxiv (2023)

Purcell effect on a single spin



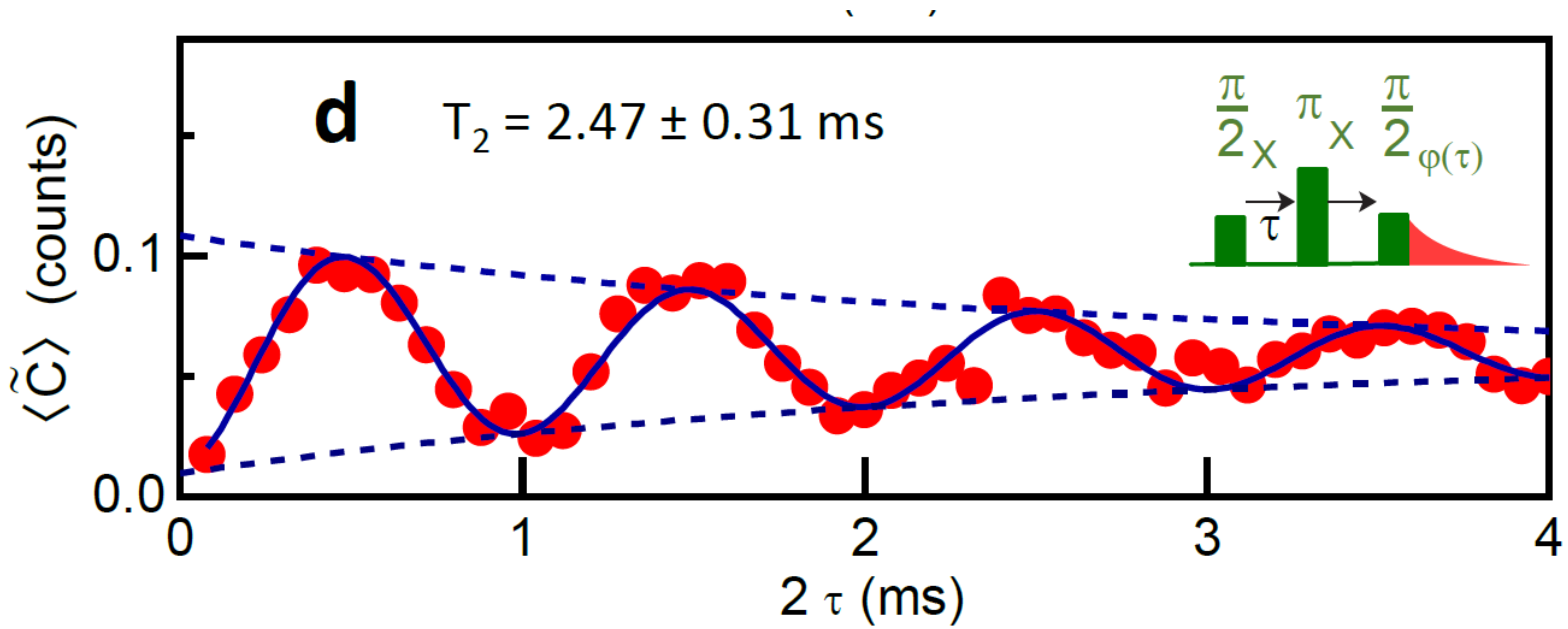
$$\Gamma_R = \frac{4g_0^2}{\kappa}\frac{1}{1+4\left[\frac{\delta}{\kappa}\right]^2}$$

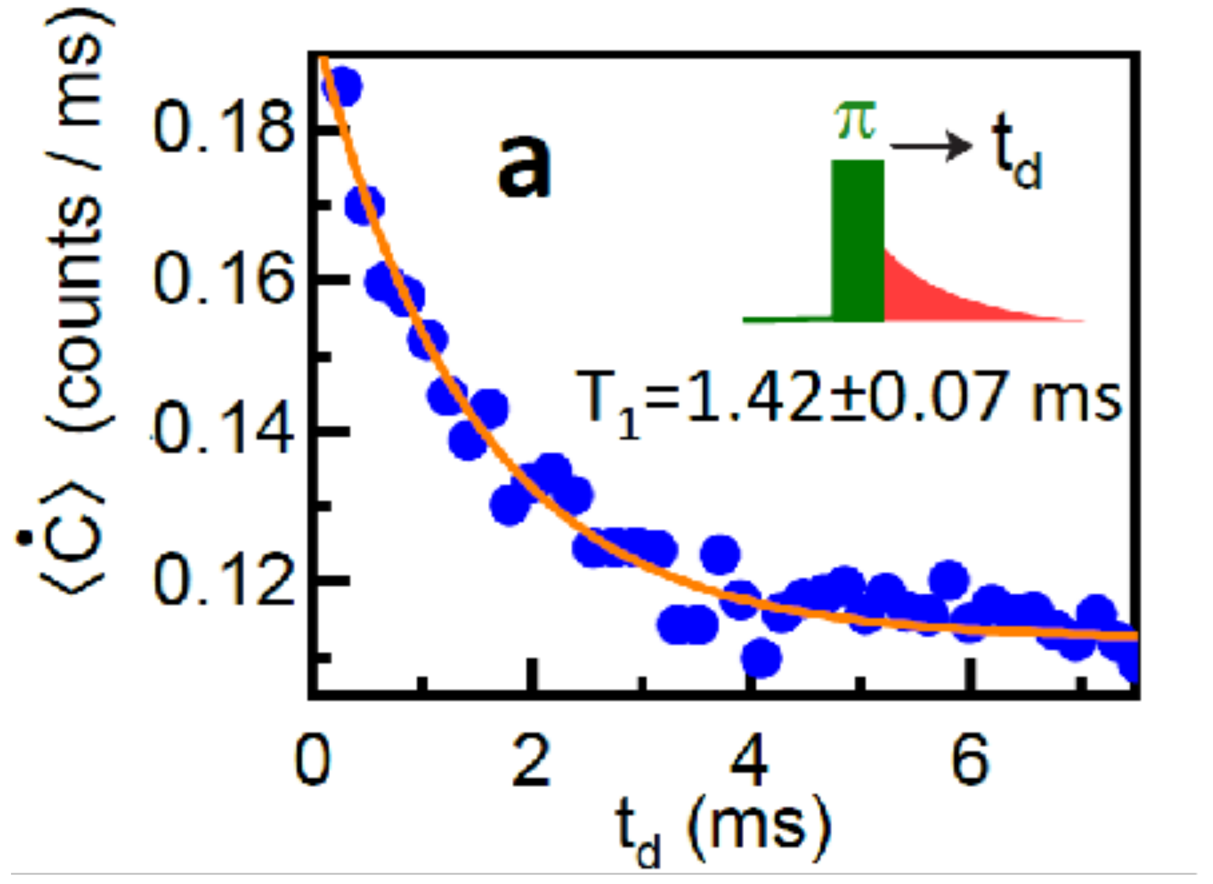
$$T_1 \cong \Gamma_R^{-1}$$

(Er^{3+} spin in the Purcell regime)

Z. Wang et al., arxiv (2023)

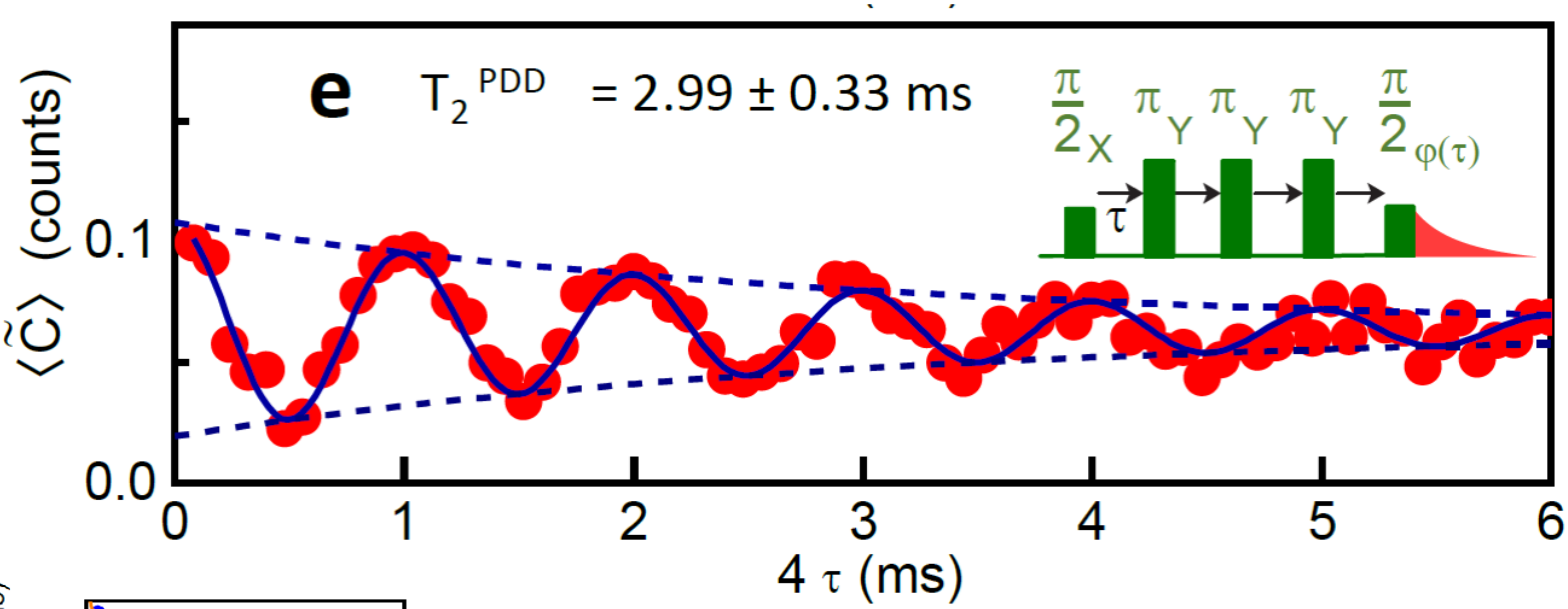
Single-ion coherence time (2) : Hahn echo

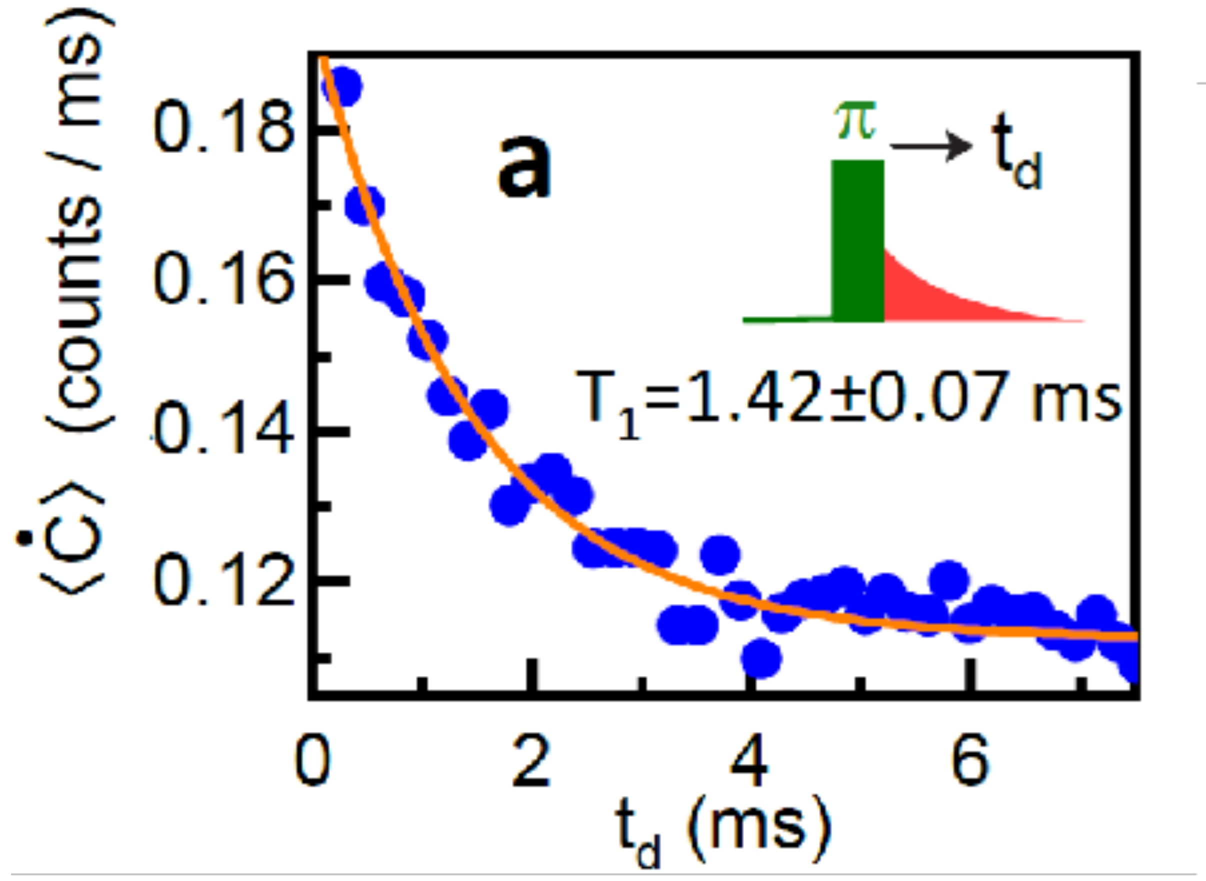




Z. Wang et al., arxiv (2023)

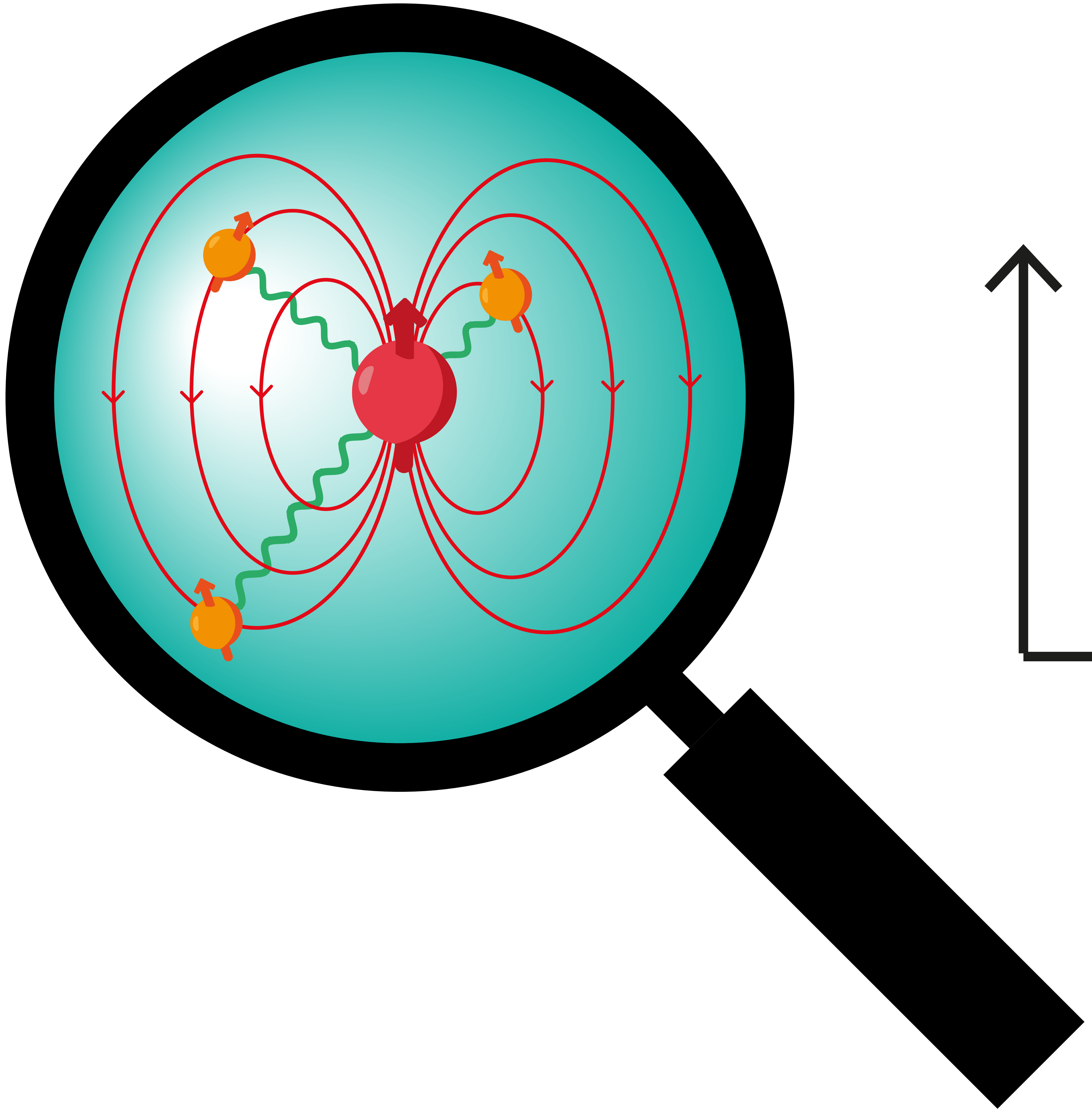
Single-ion coherence time (3) : 3-Pulse Dynamical Decoupling

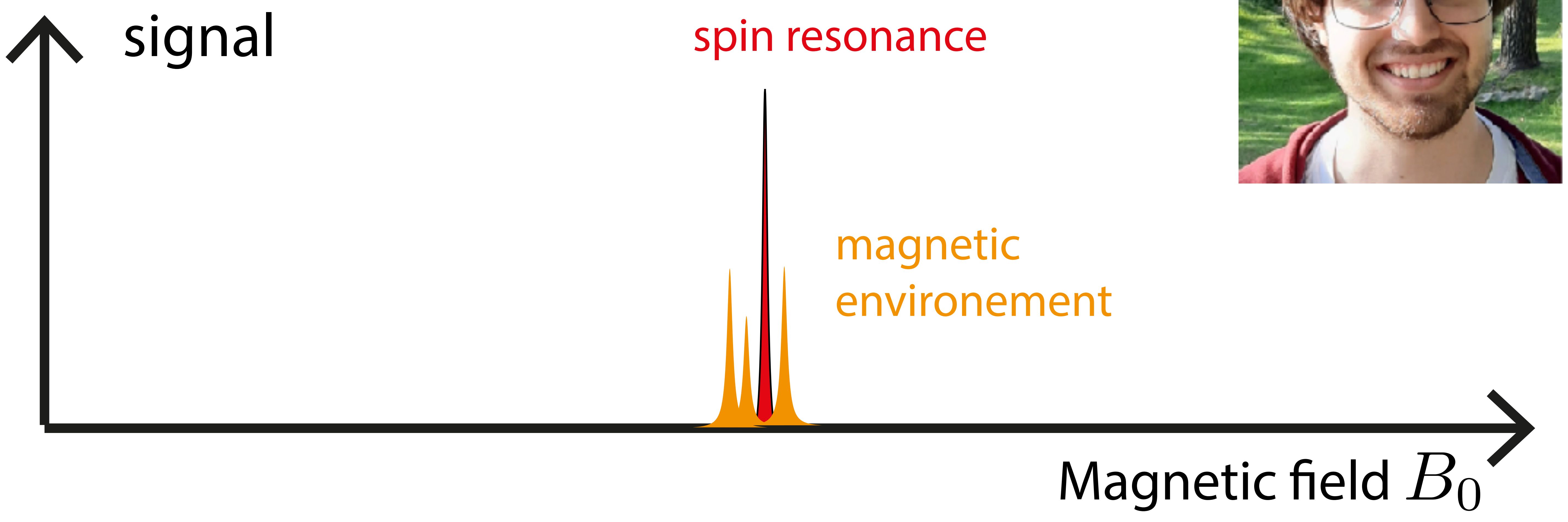




Z. Wang et al., arxiv(2023)

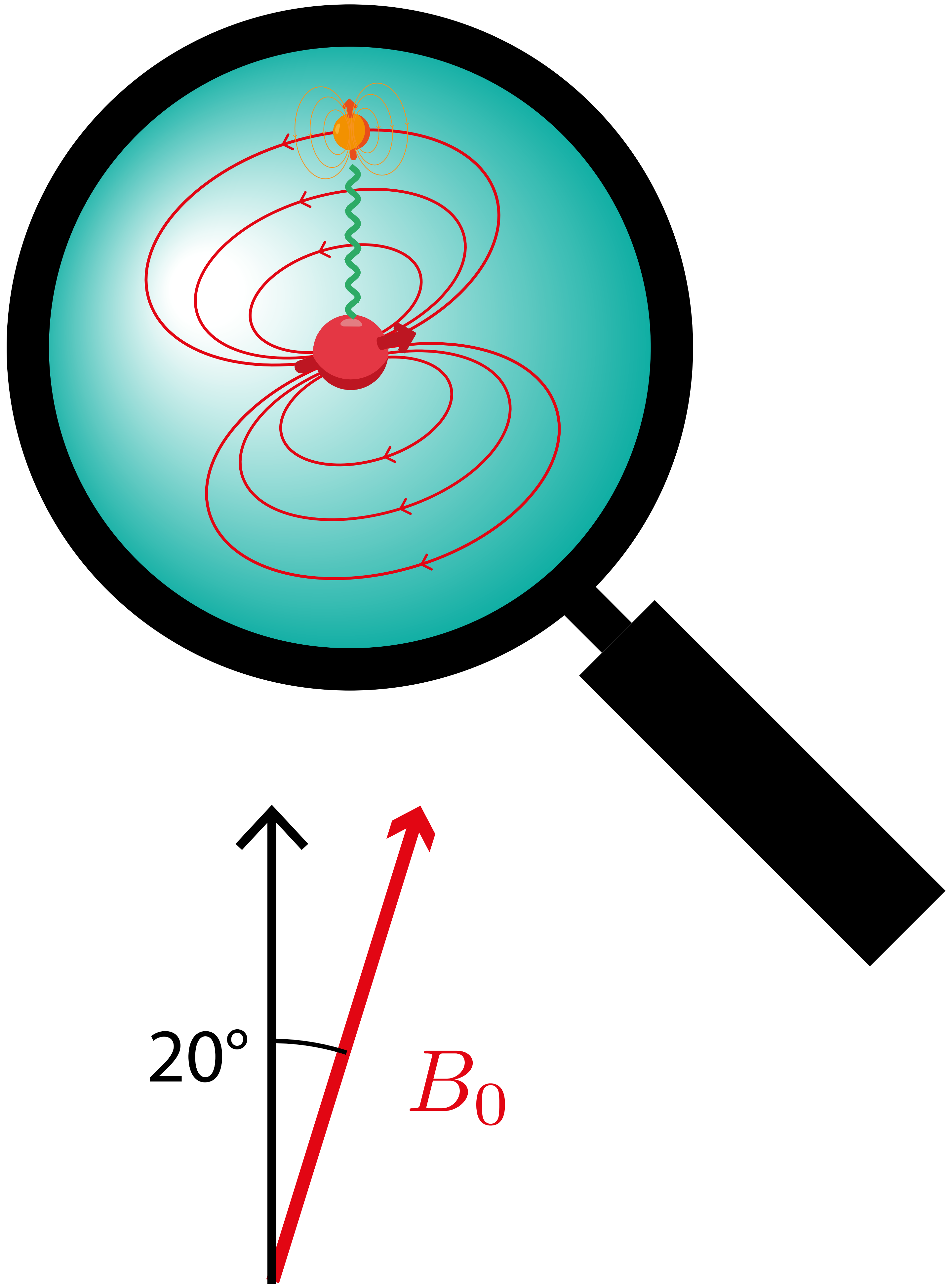
Probing the nuclear environment

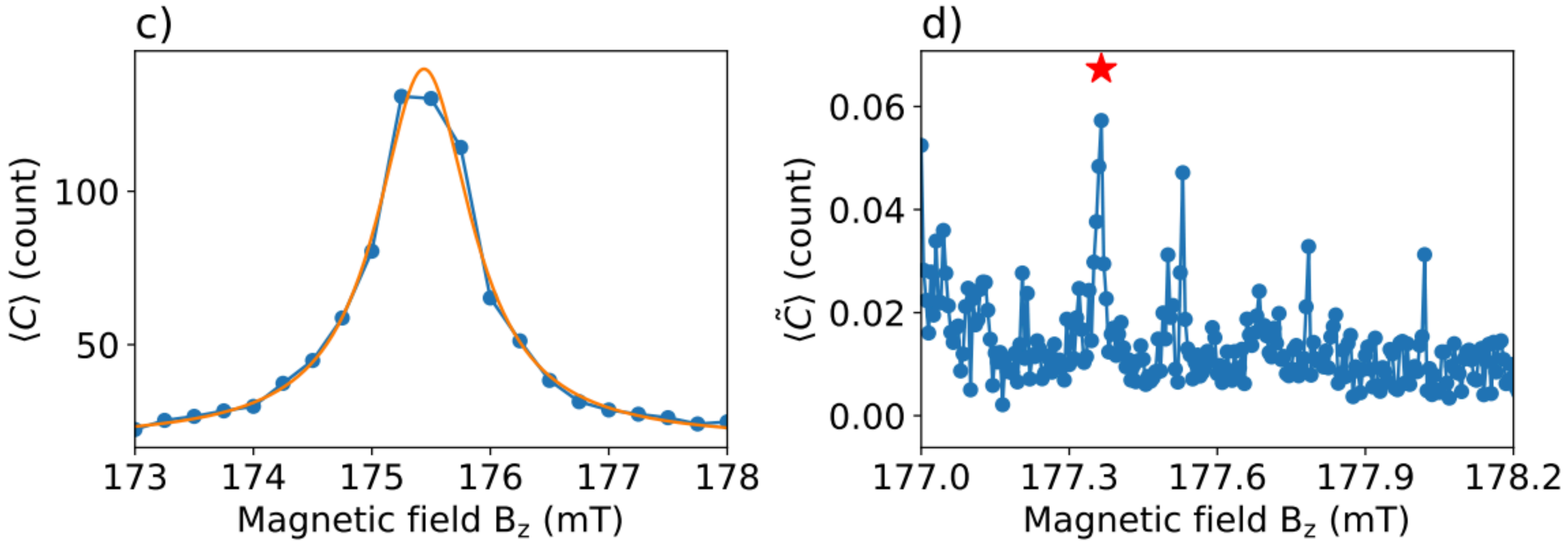




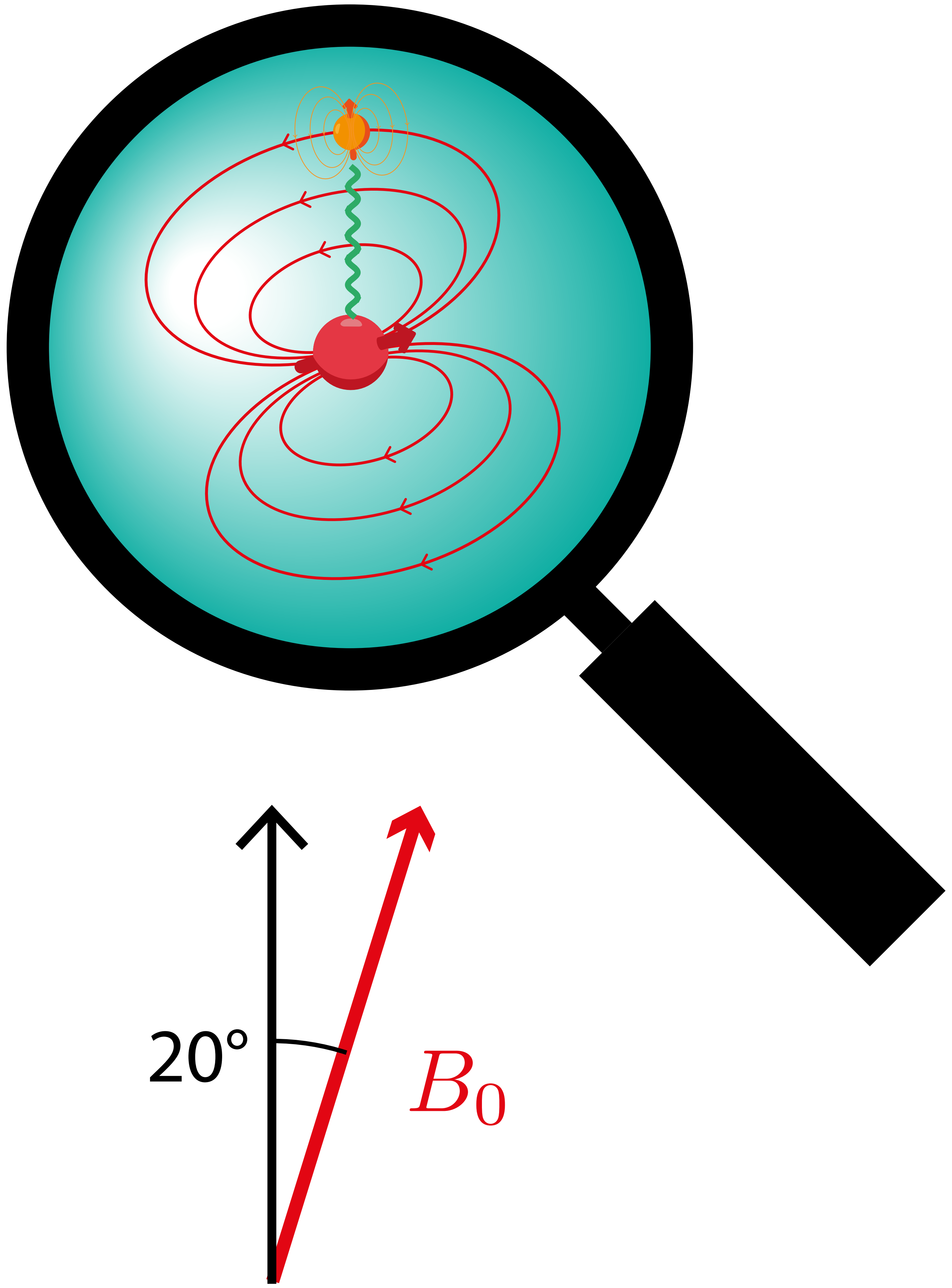
Jaime Travesedo

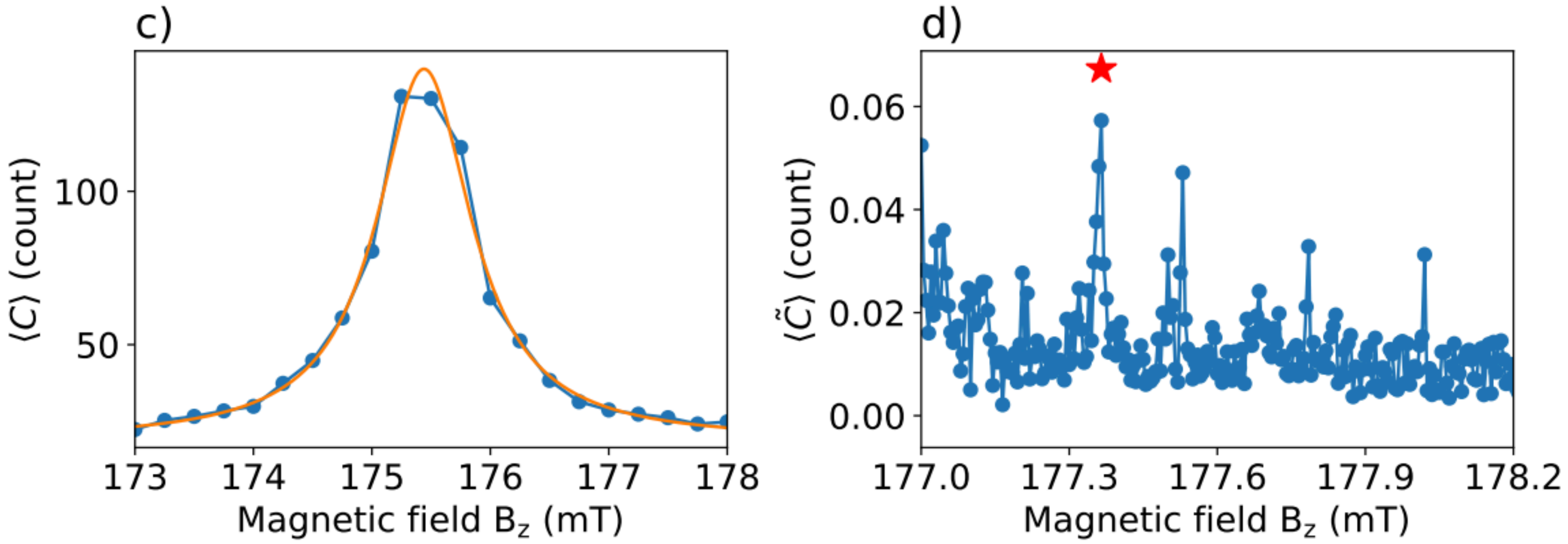
Probing the nuclear environment

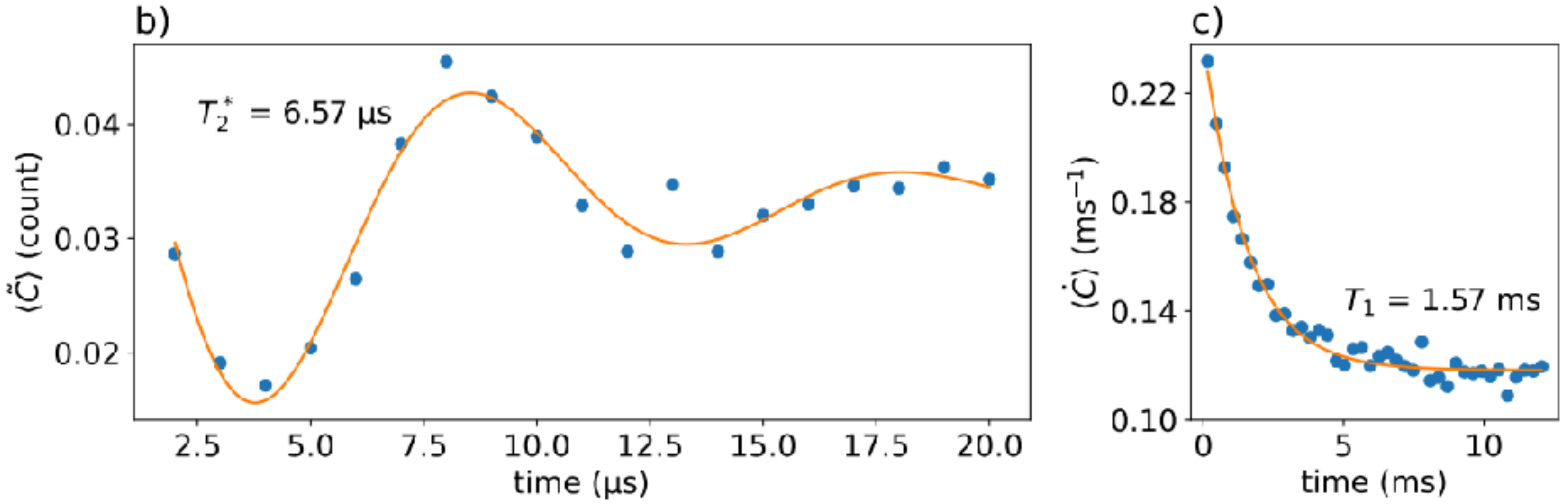




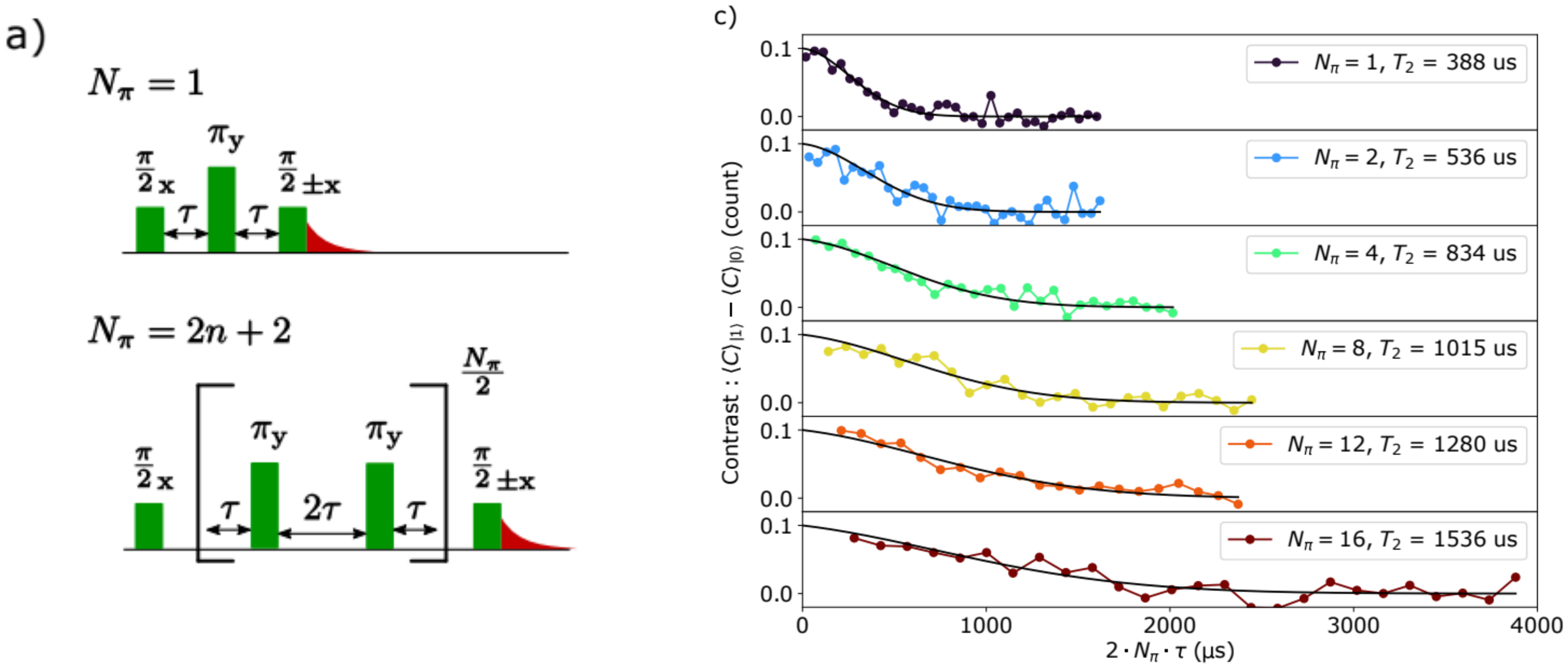
Probing the nuclear environment





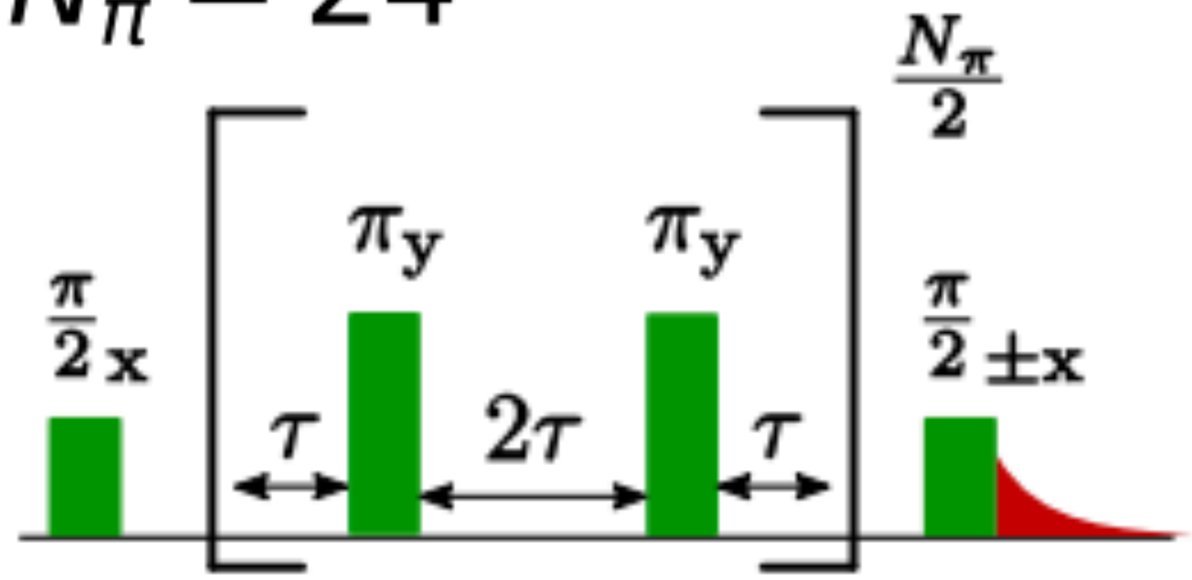


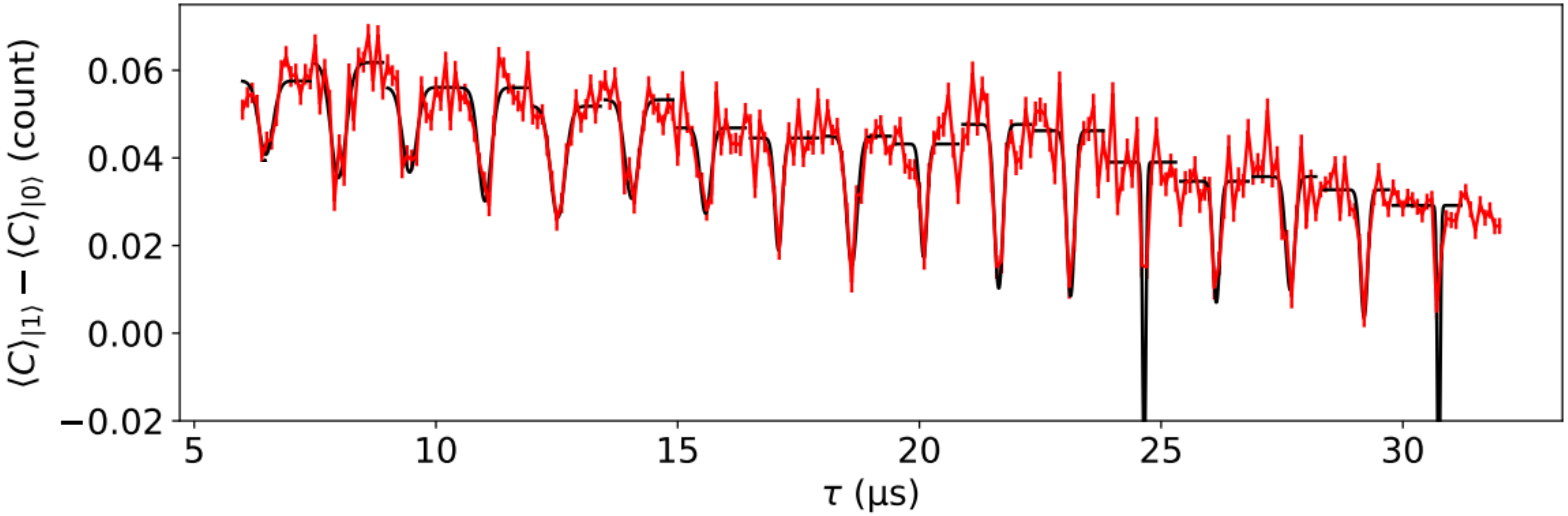
Dynamical decoupling



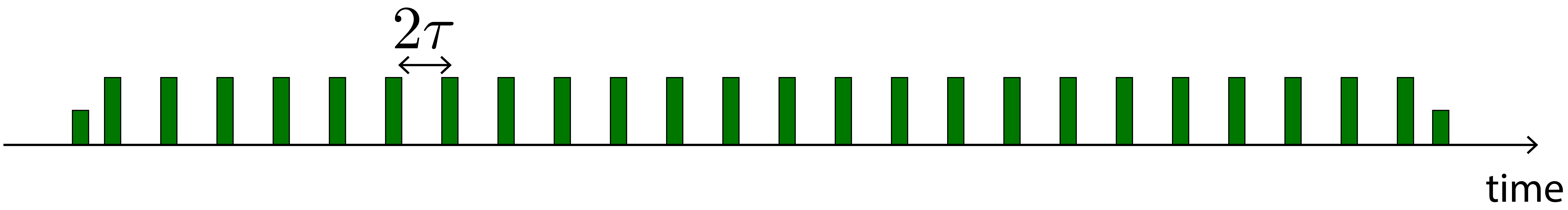
Dynamical decoupling

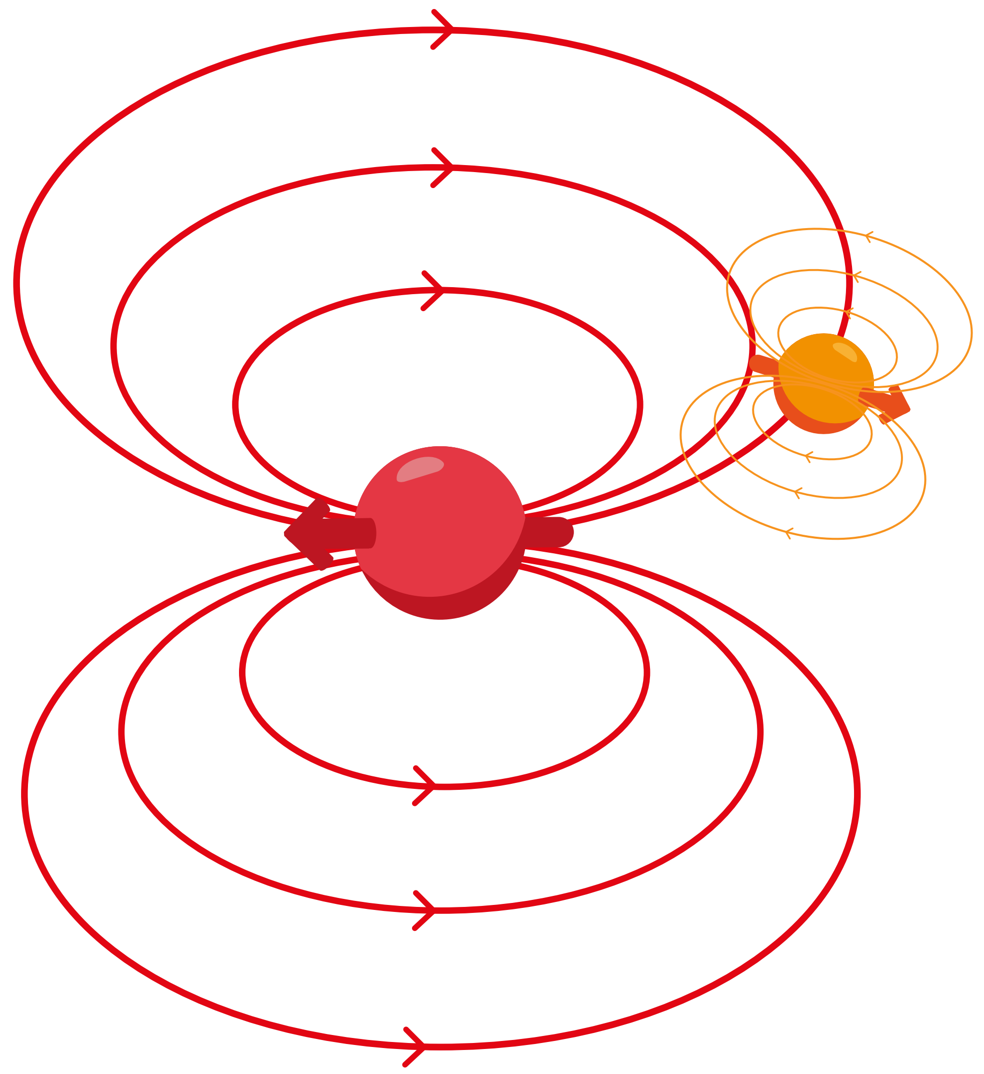
$N_\pi = 24$



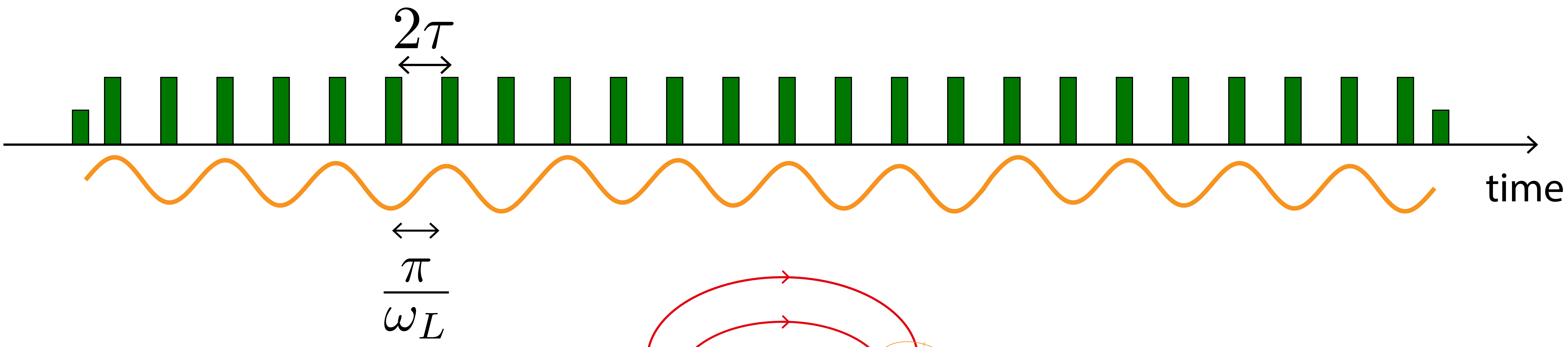


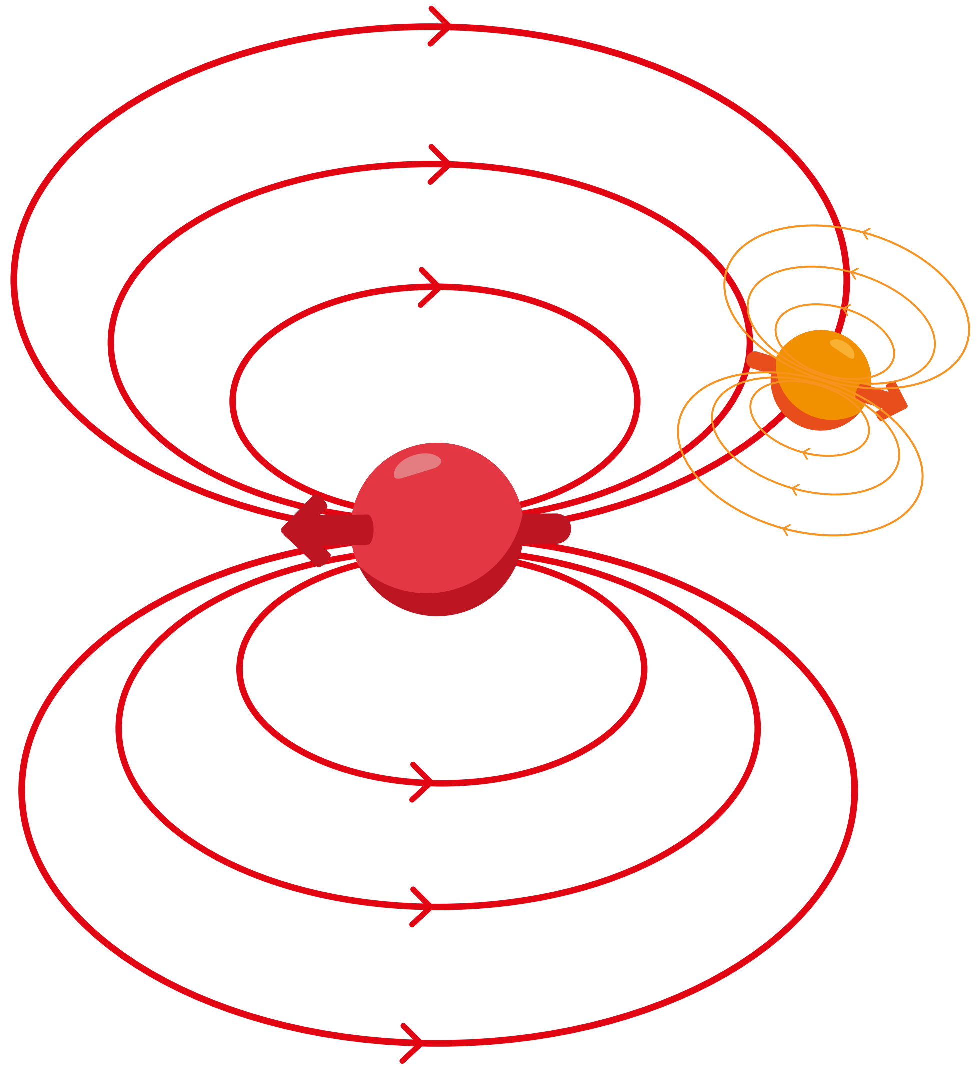
Dynamical decoupling as a nuclear spin probe





Dynamical decoupling as a nuclear spin probe



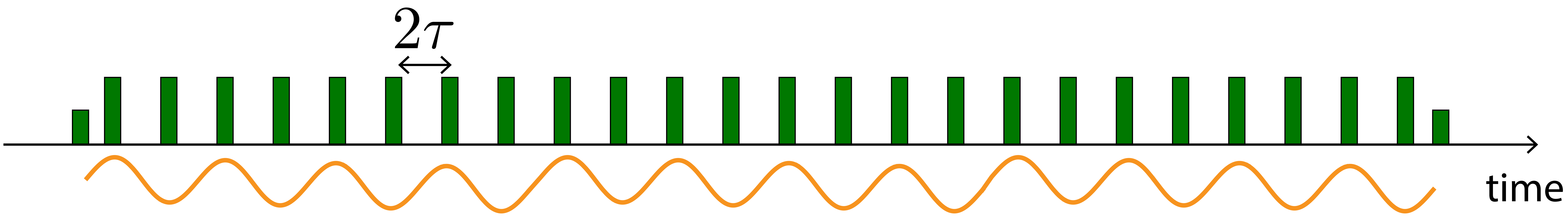


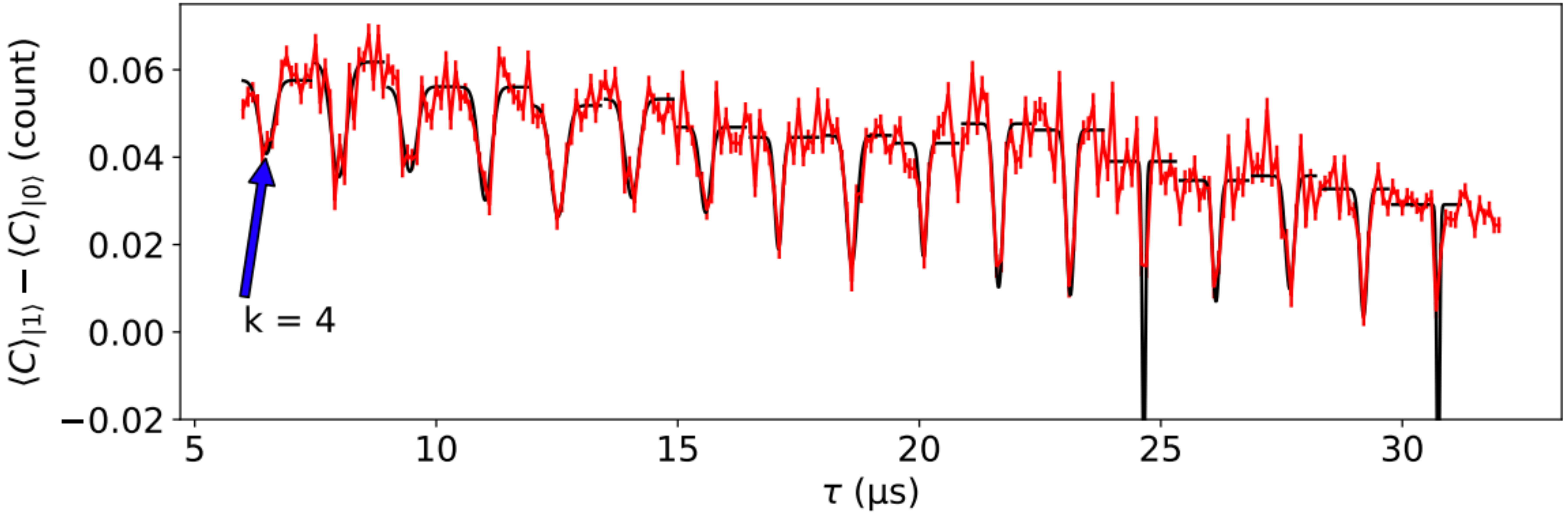
Nuclear resonance condition :

$$2\tau = (2k+1)\frac{\pi}{\omega_L}$$

Taminiau et al. PRL 2012
Kolkowitz et al. PRL 2012

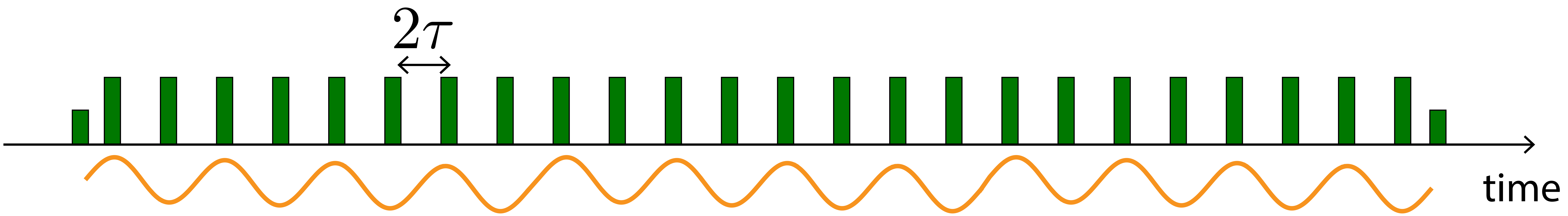
Dynamical decoupling as a nuclear spin probe

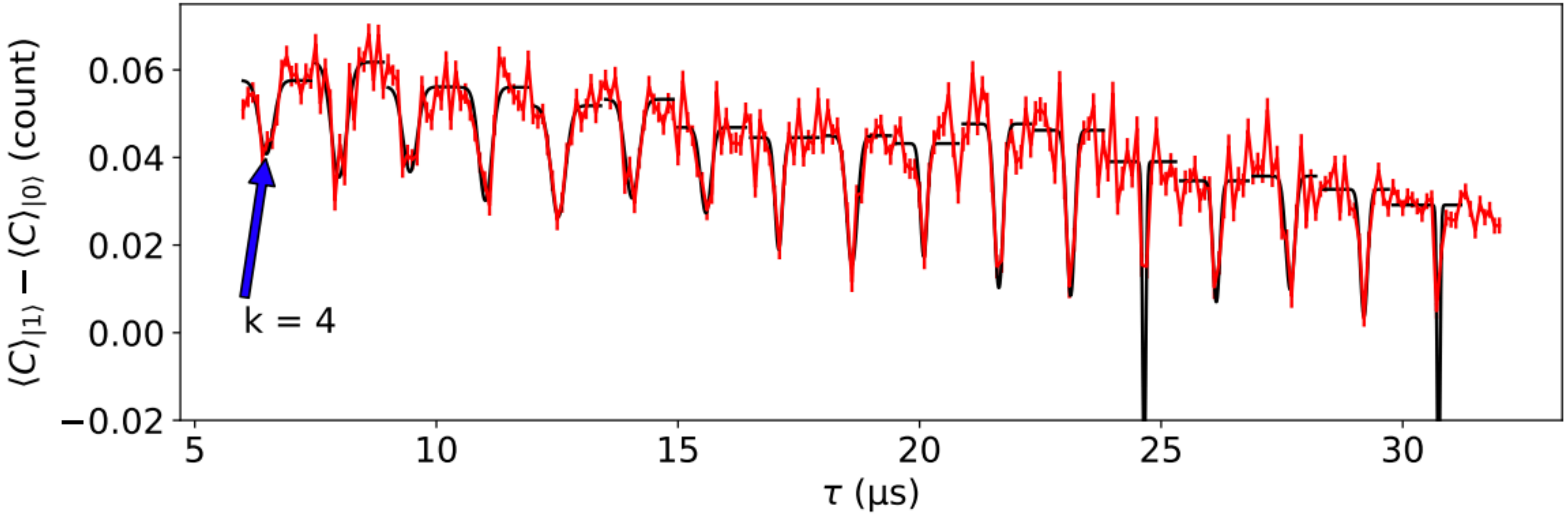


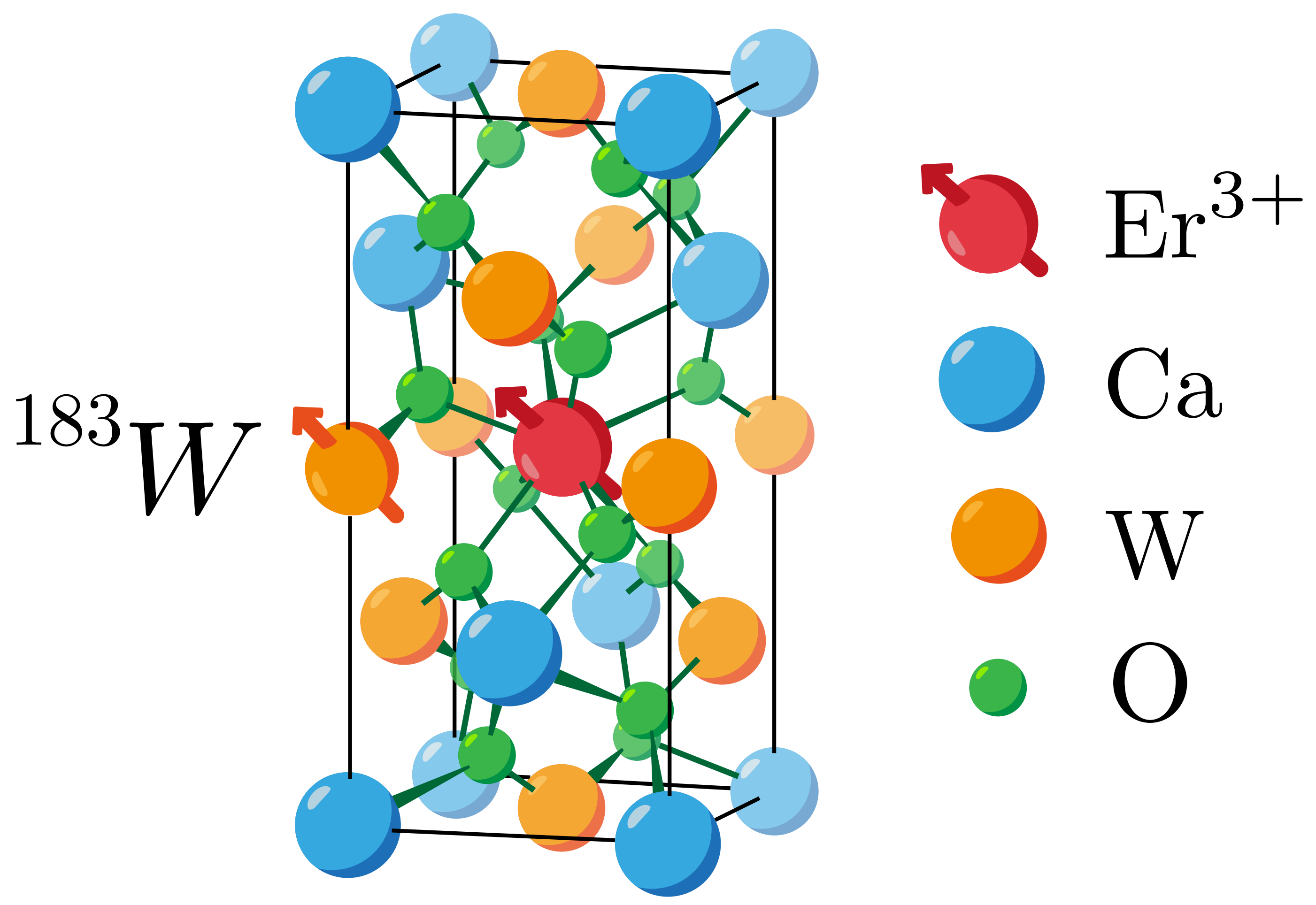


$$\Delta\tau = \frac{\pi}{\omega_L} = 1.52\ \mu s \quad \rightarrow \quad \boxed{\frac{\omega_L}{2\pi} = 332\ \text{kHz}}$$

Dynamical decoupling as a nuclear spin probe

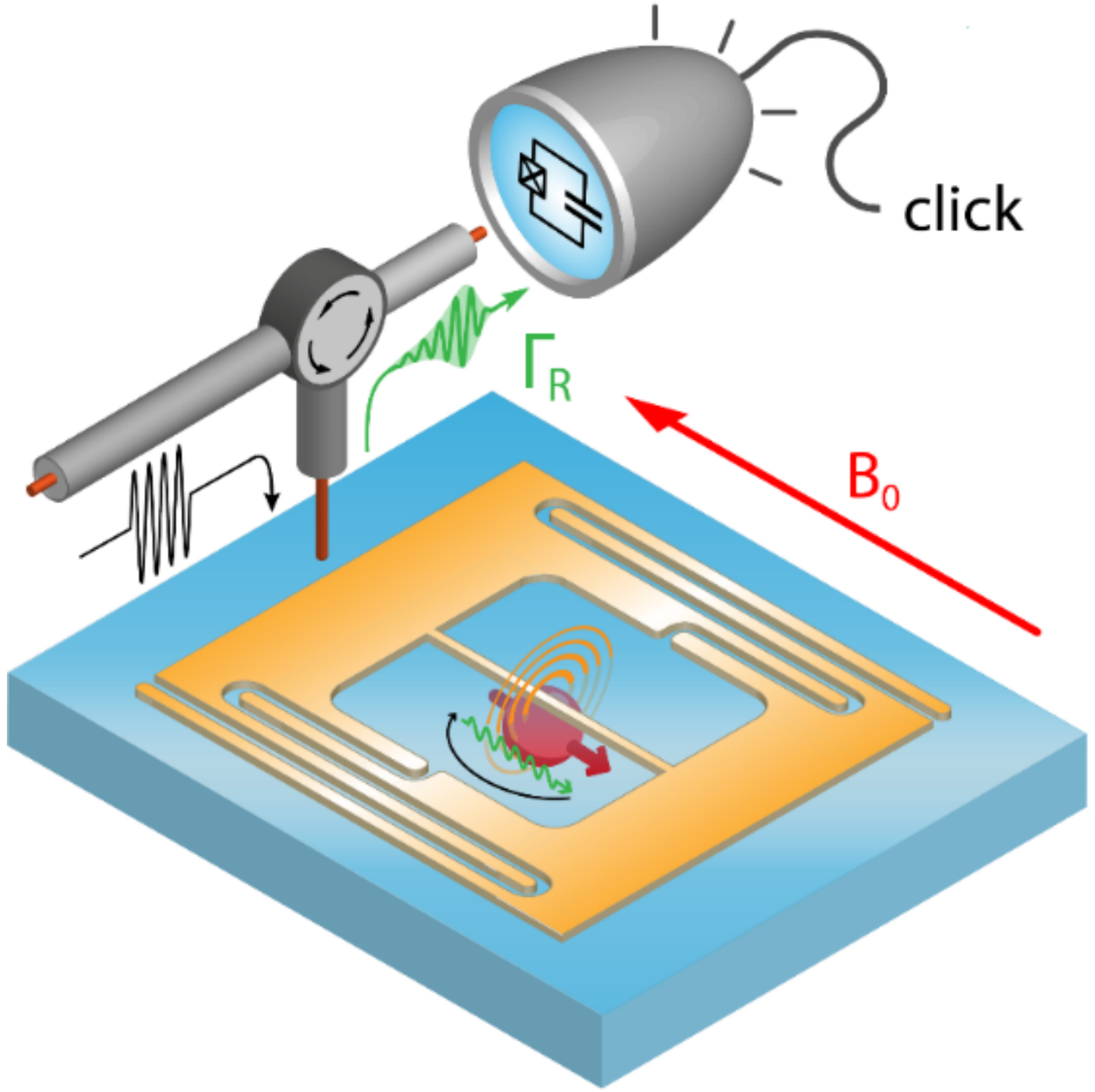




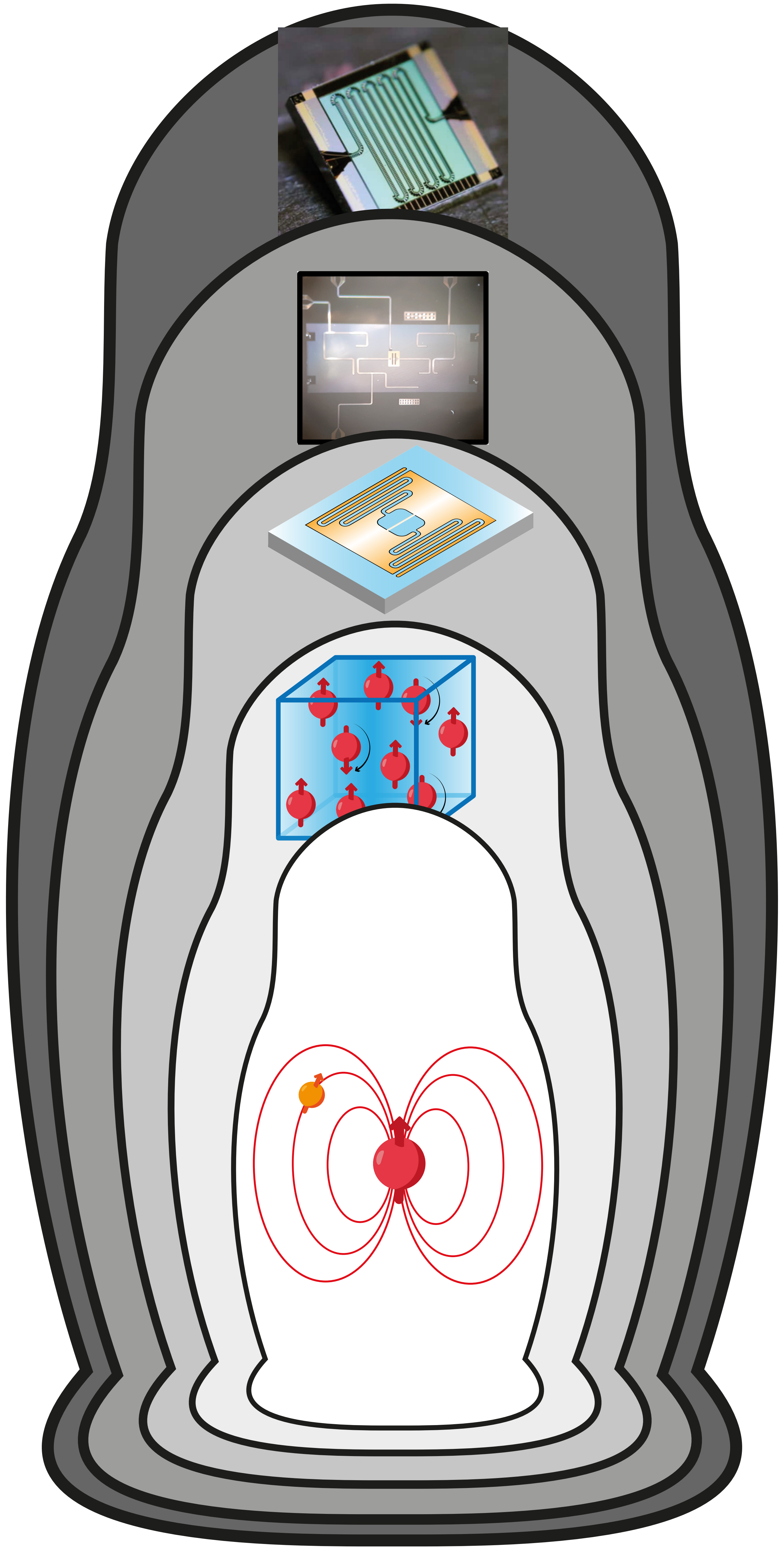


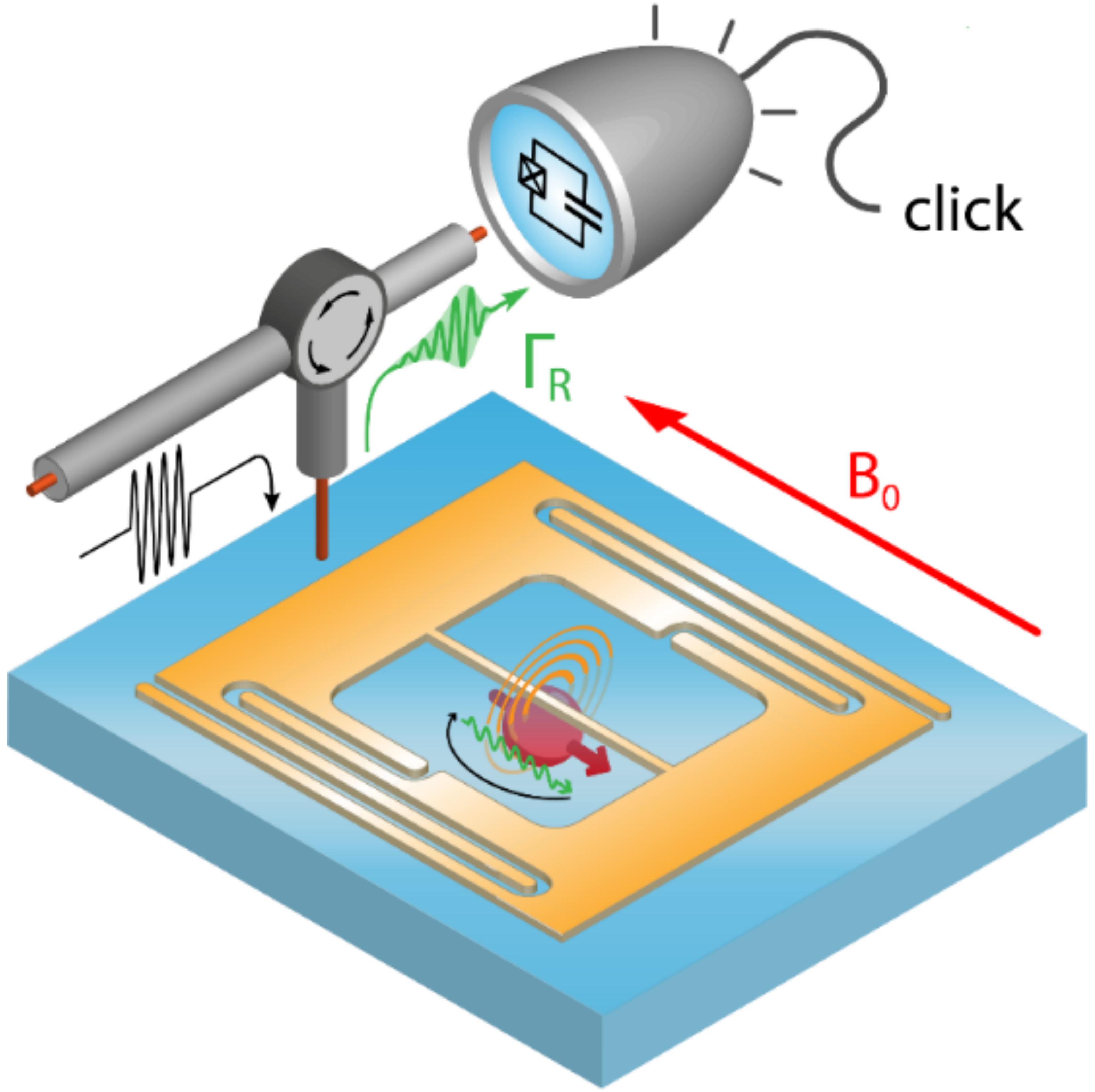
$$\Delta\tau = \frac{\pi}{\omega_L} = 1.52\ \mu s \quad \Rightarrow \quad \frac{\omega_L}{2\pi} = 332\ \text{kHz} \quad \Rightarrow \quad \frac{\omega_L}{2\pi\, B_0} = 1.78\ \text{MHz/mT} = \gamma_W\ !$$

Conclusion



Conclusion

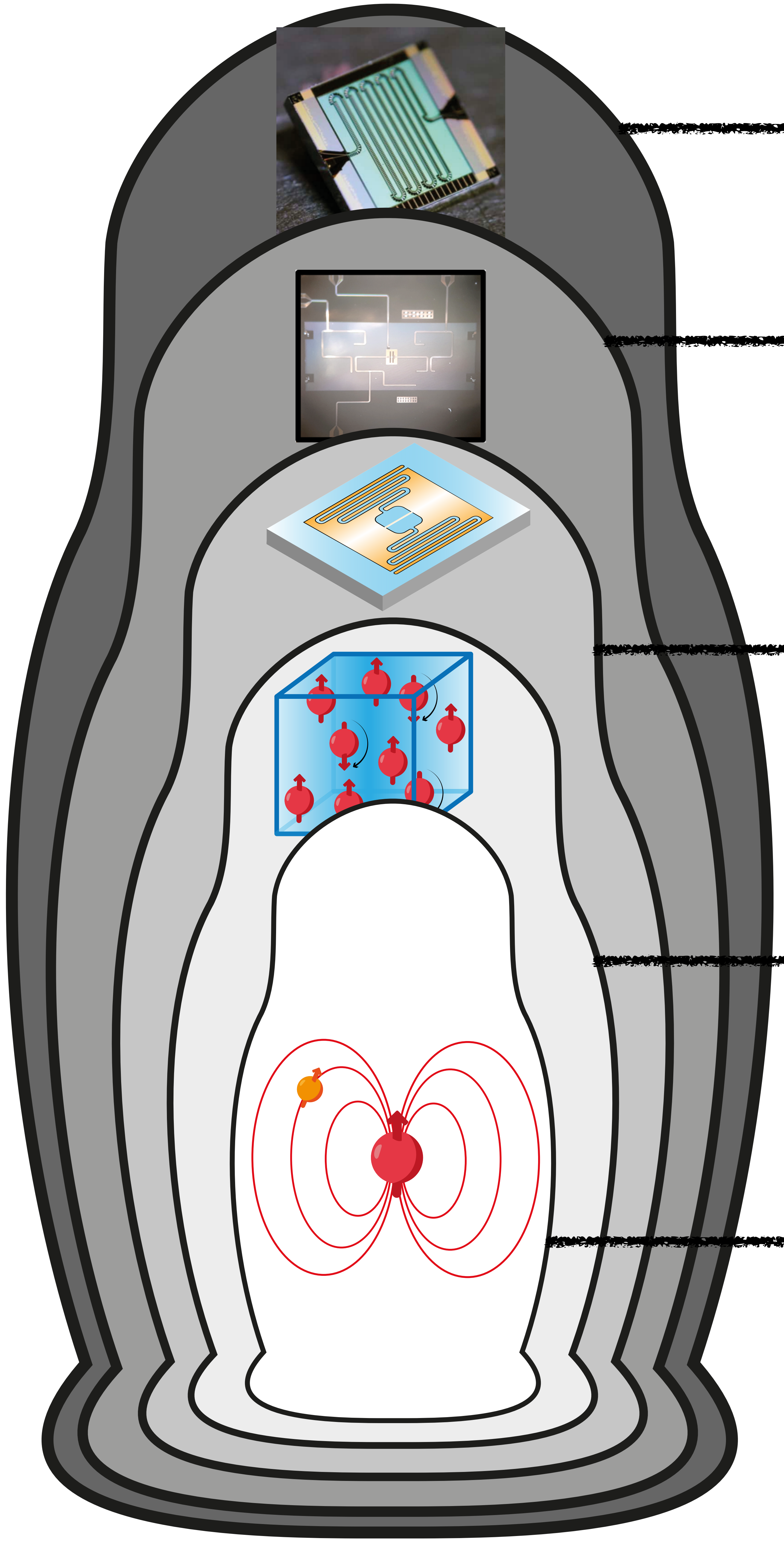




Single Spin Fluorescence Detection

- Universal
- Large detection volume ($\sim 10\mu m^3$)
- 1 $Er^{3+}/\sqrt{Hz}$, large improvements possible
- Does not require long coherence time

Perspective



Strong academic & industrial effort for circuits

SMPD development $\hbar\omega\frac{\sqrt{\alpha}}{\eta} = 10^{-23}\ \mathrm{W}/\sqrt{\mathrm{Hz}}$

x10 sensitivity = x100 integration time

Advanced antenna design for enhanced magnetic focusing

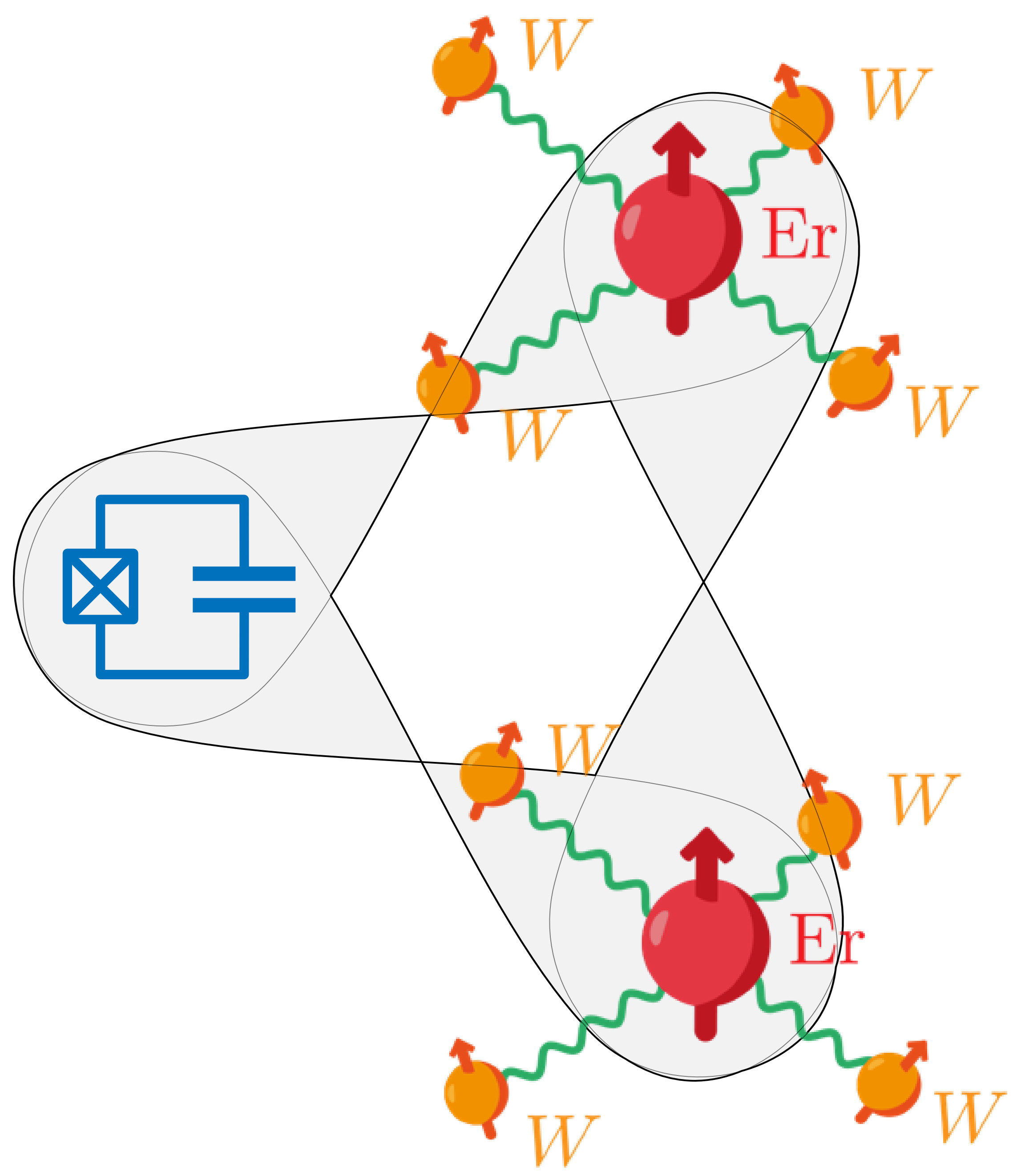
New spin species in new substrates

RF control for single nuclear spin manipulation

Perspective

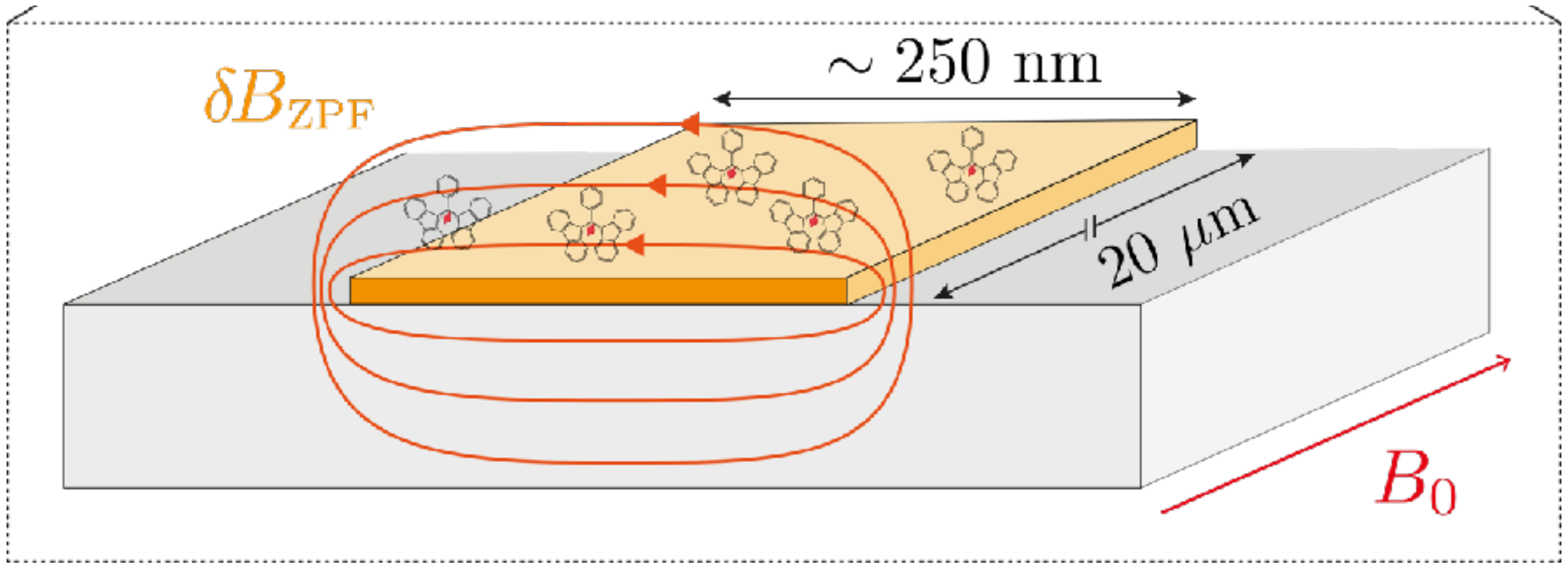
Architecture for spin-based quantum computing

- Coherence times up to second
- Interfacing with microwave photons, superconducting circuits, and nuclear spins



- Practical single-spin EPR spectroscopy

at millikelvin temperature
(transition ions & radicals in molecules, …)

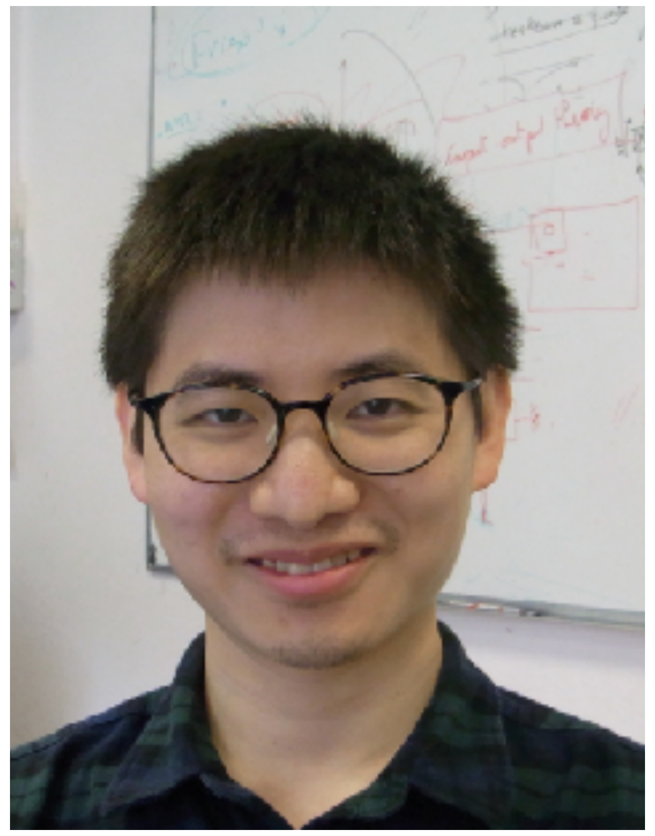


Thanks!

J. Travesedo

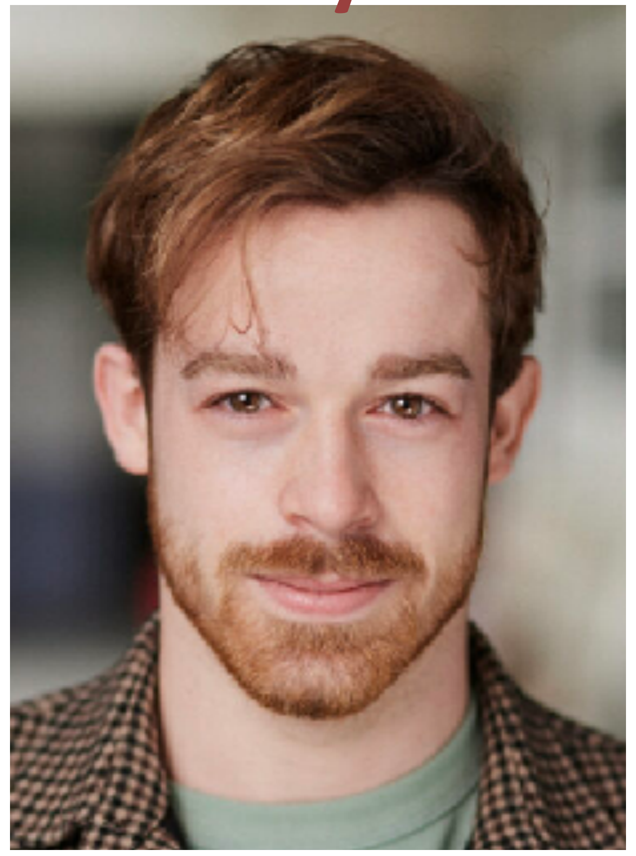
L. Balembois

Z. Wang

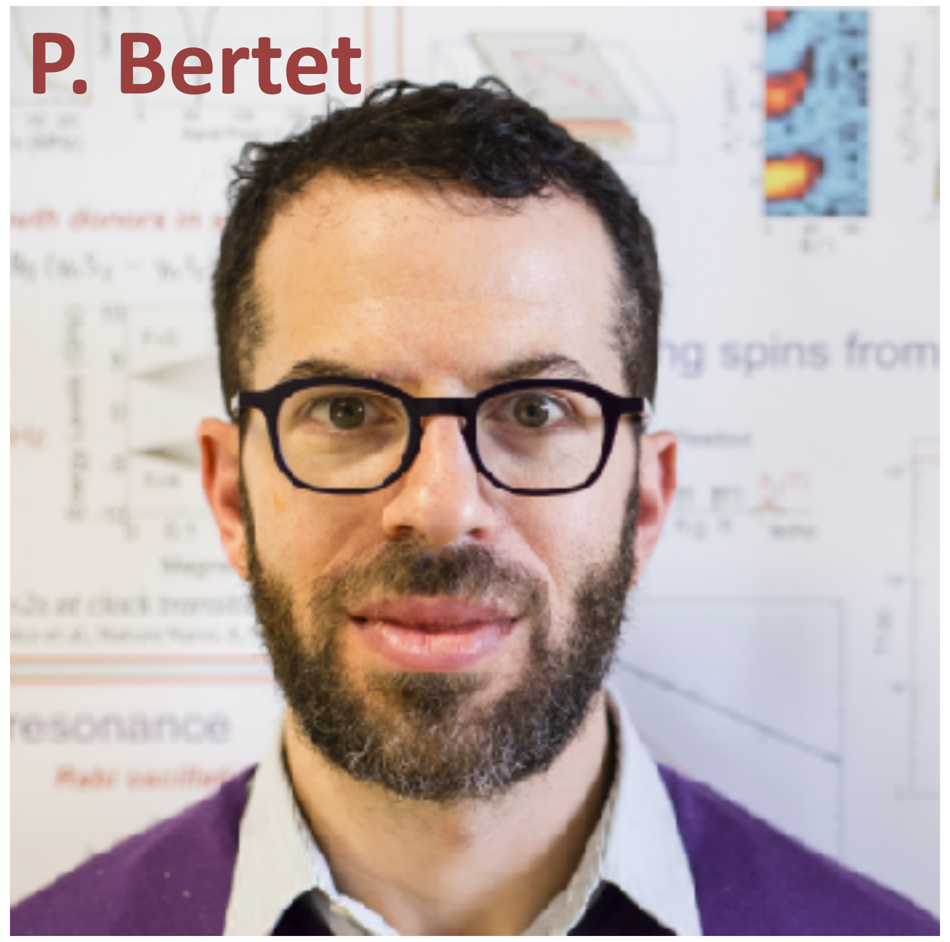


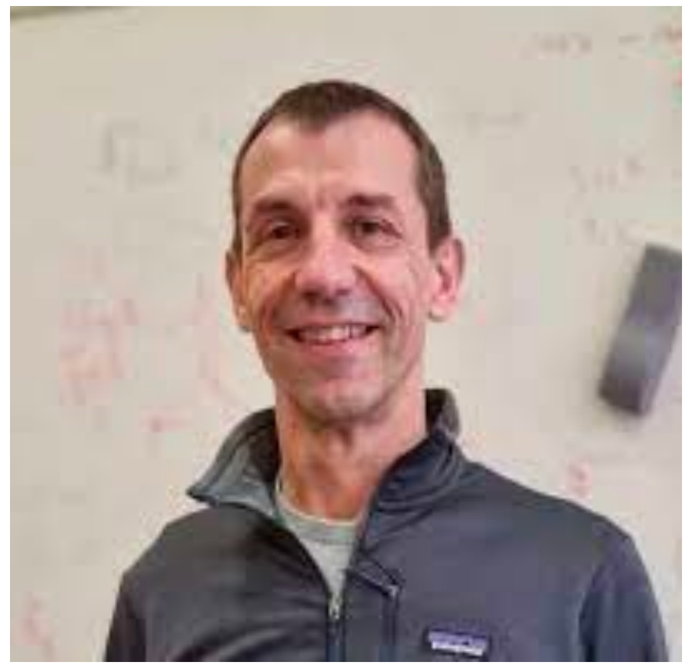
L. Pallegoix

A. May



P. Bertet





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Quantronics group

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