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Cavendish Laboratory



taming a nuclear spin ensemble with quantum optics

Mete Atatüre

Cavendish Laboratory
University of Cambridge

Quantum Optical Materials and Systems Group, Cambridge UK



European
Research
Council



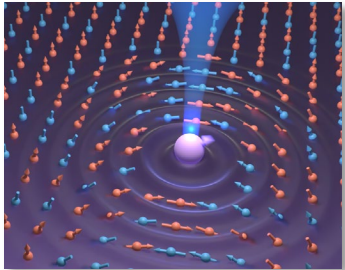
HENRY
ROYCE
INSTITUTE



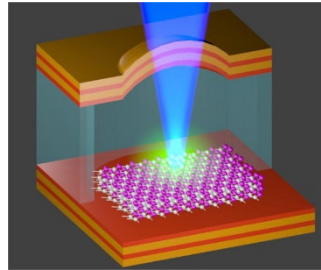
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Quantum photonics and spin physics with novel material platforms

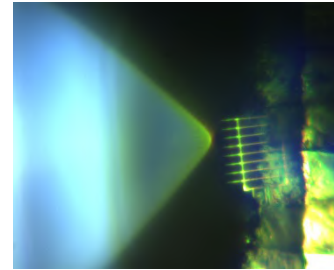
III-Vs



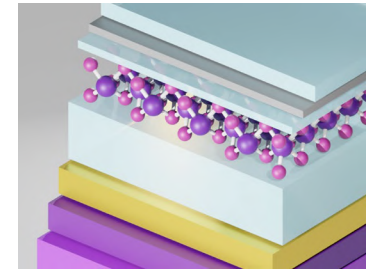
hBN



Diamond

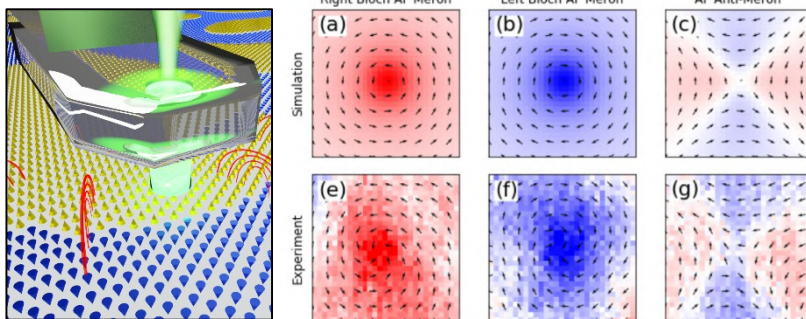


2d vdWs

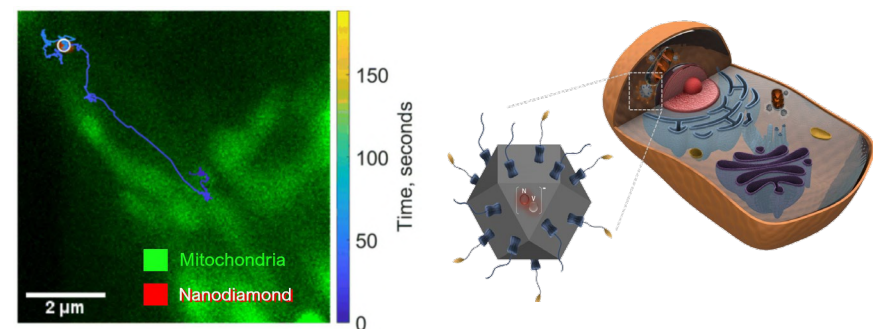


Quantum sensing of emergent magnetism and complexity of life sciences

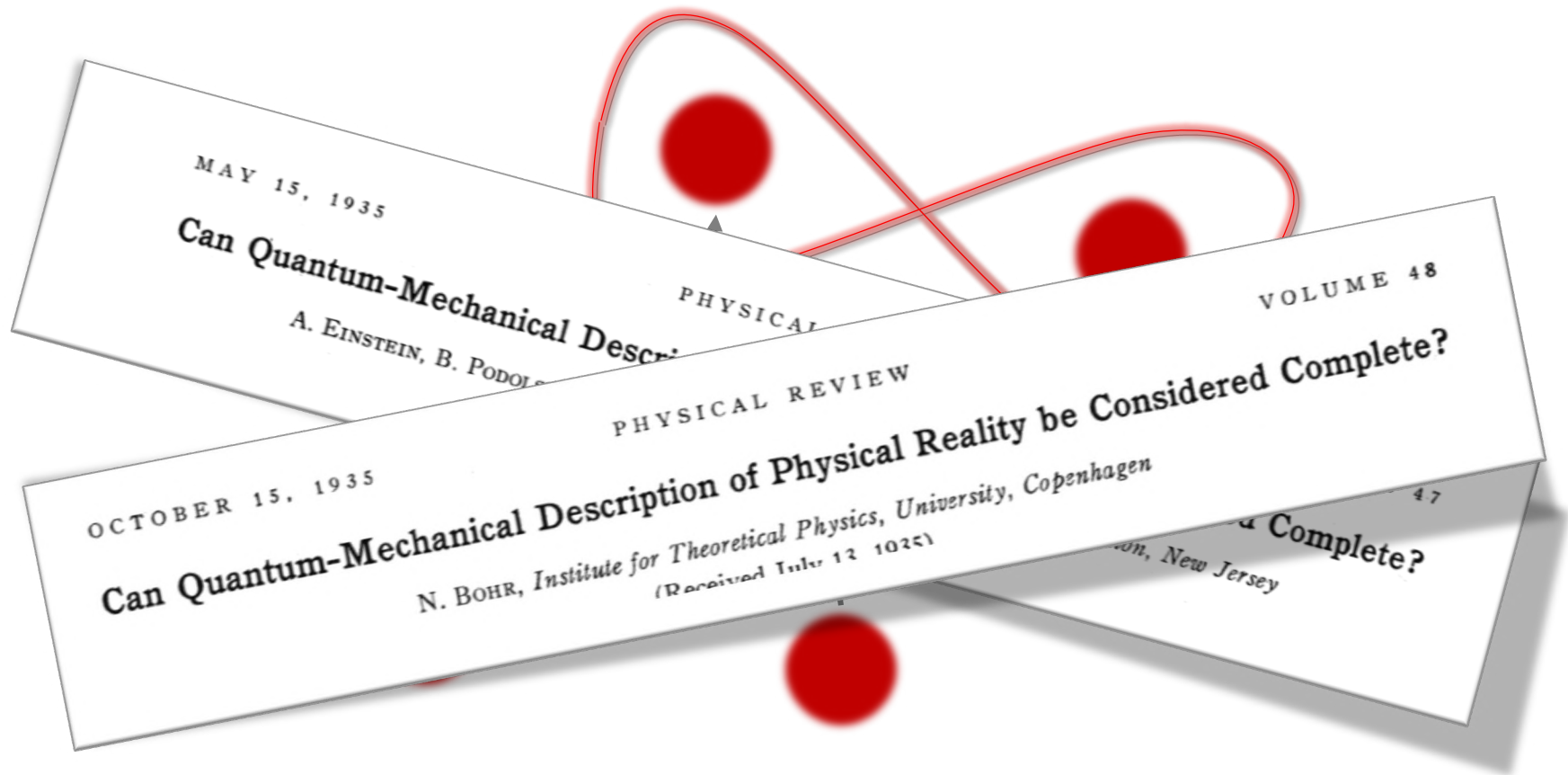
Diamond magnetometry



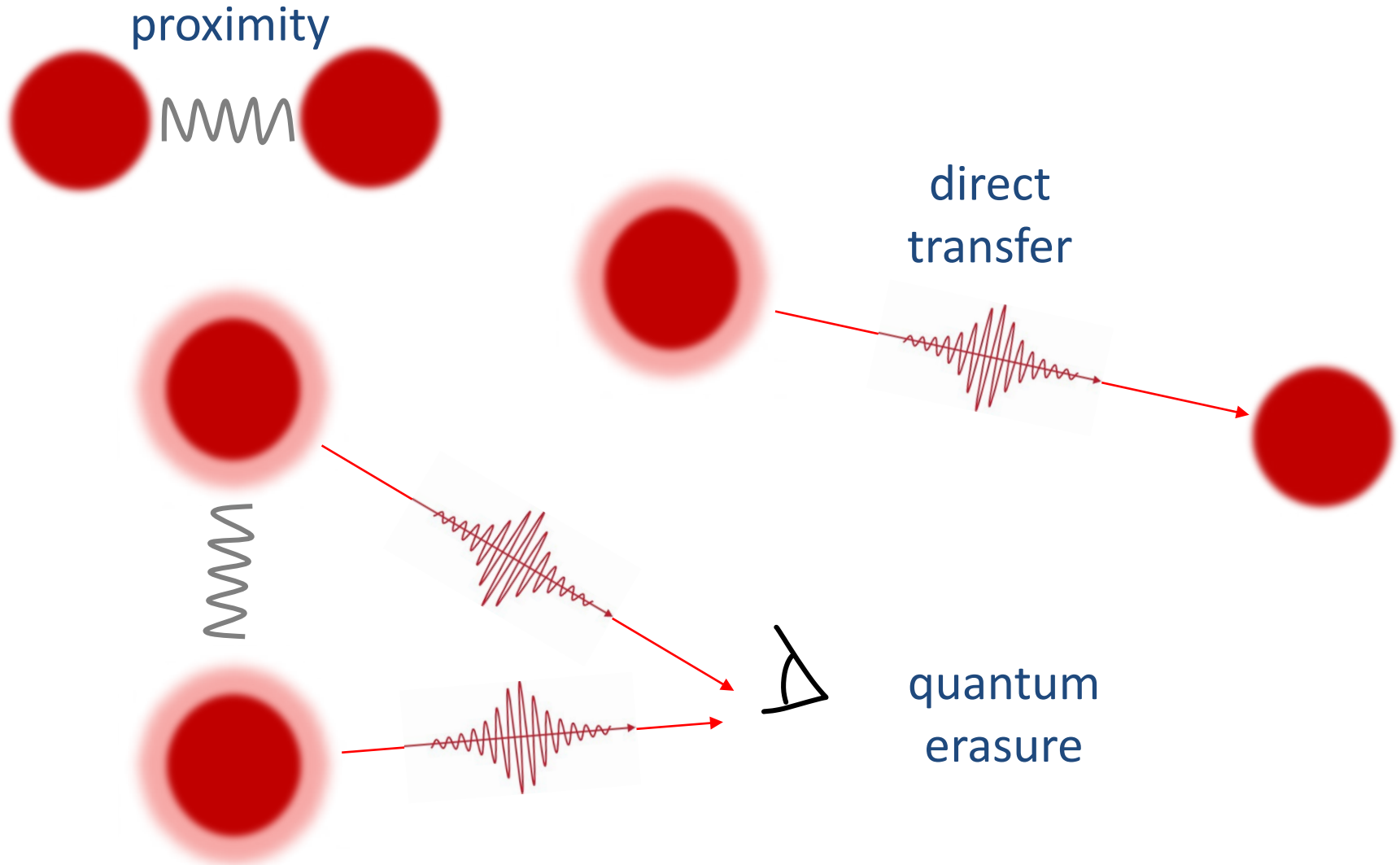
Nanodiamond biosensing



creating correlations



creating correlated particles



creating correlated matter



creating correlated matter – atoms & interactions

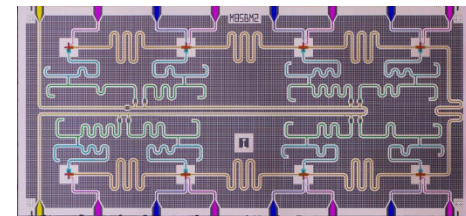
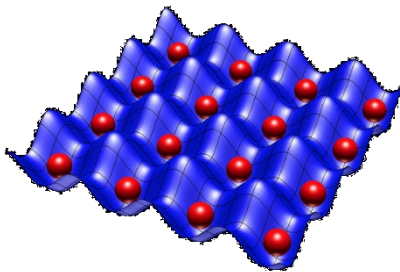


Real atoms
Real matter

Synthetic atoms
Real interactions

Real atoms
Synthetic matter

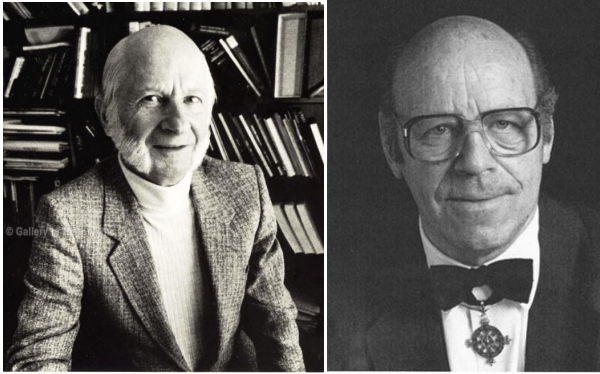
Synthetic atoms
Synthetic matter



our question...

Can we capitalize on quantum optics, light-matter interaction and engineered material properties to create useful and exotic collective states?

isolating the quantum from the classical



Jean Dehmelt

W. Pauli

Dehmelt & Paul (Nobel 1989)

"for the development of the ion trap technique"



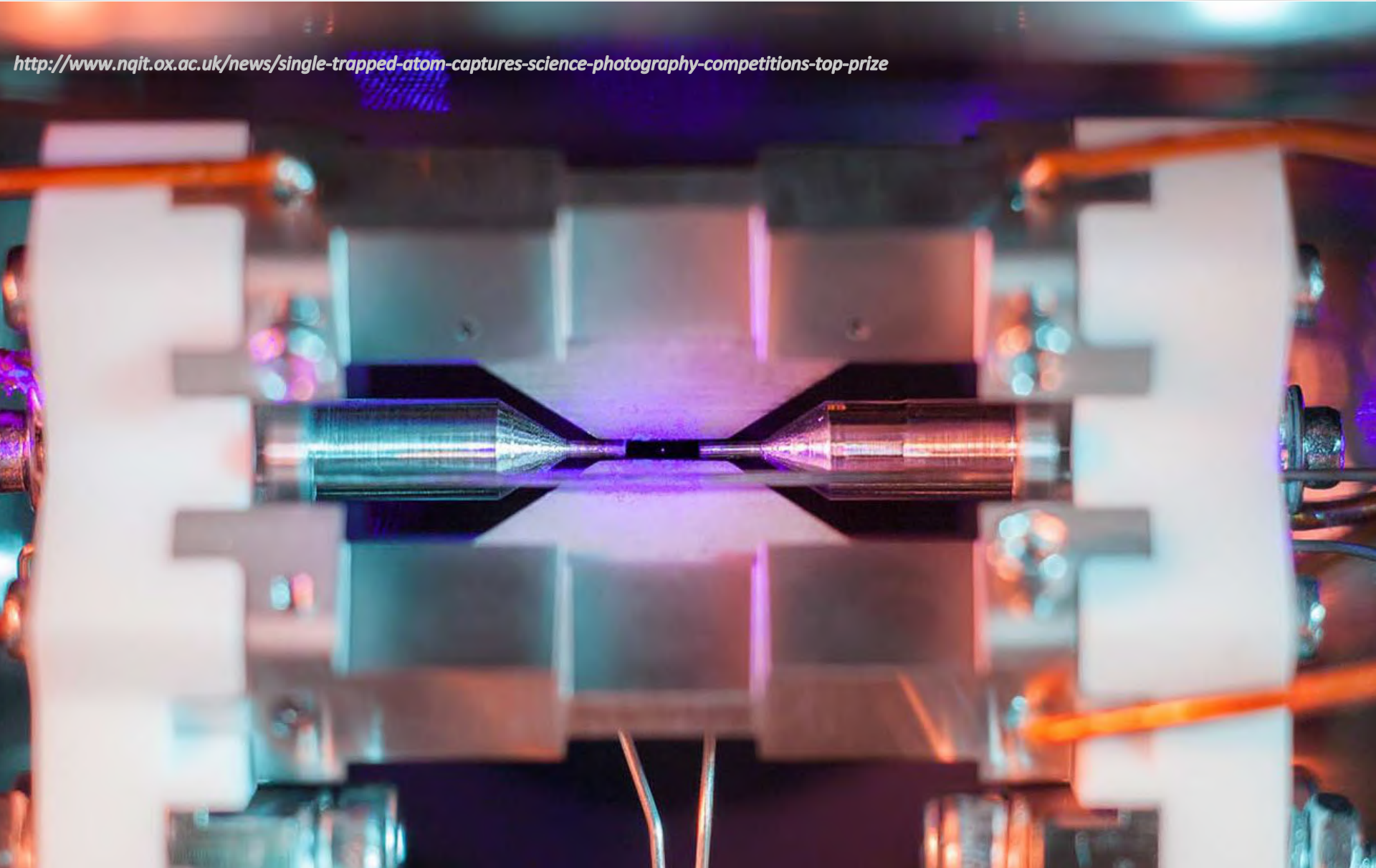
Wineland & Haroche (Nobel 2012)

"for ground-breaking experimental methods that enable measuring and manipulation of individual quantum systems"

Singular quantum objects & entanglement [quantum 2.0]

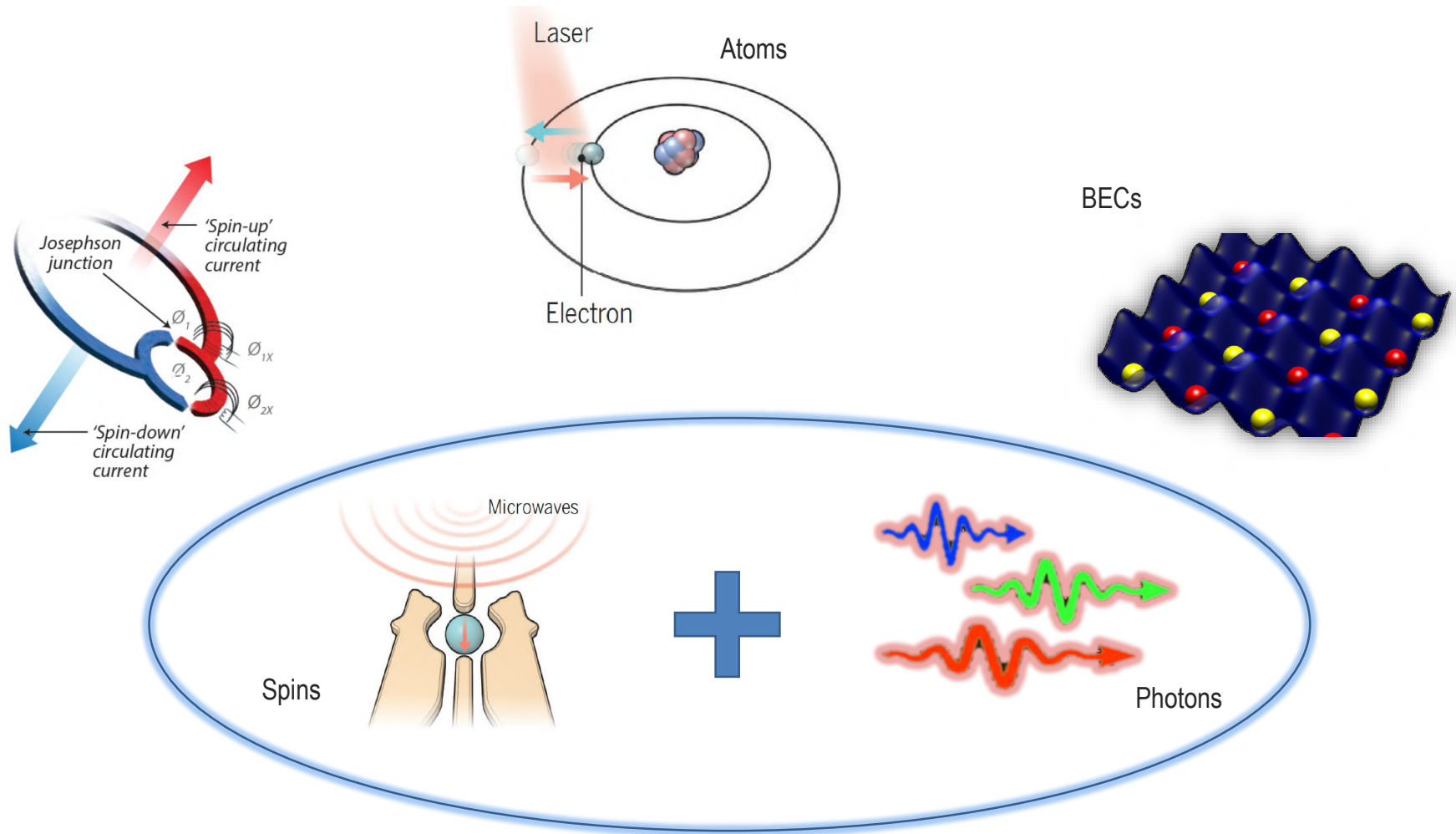
a single ion trapped in vacuum

<http://www.nqit.ox.ac.uk/news/single-trapped-atom-captures-science-photography-competitions-top-prize>

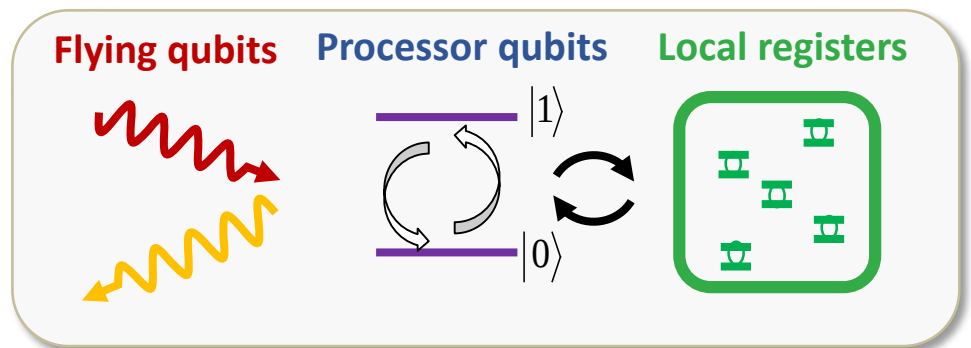
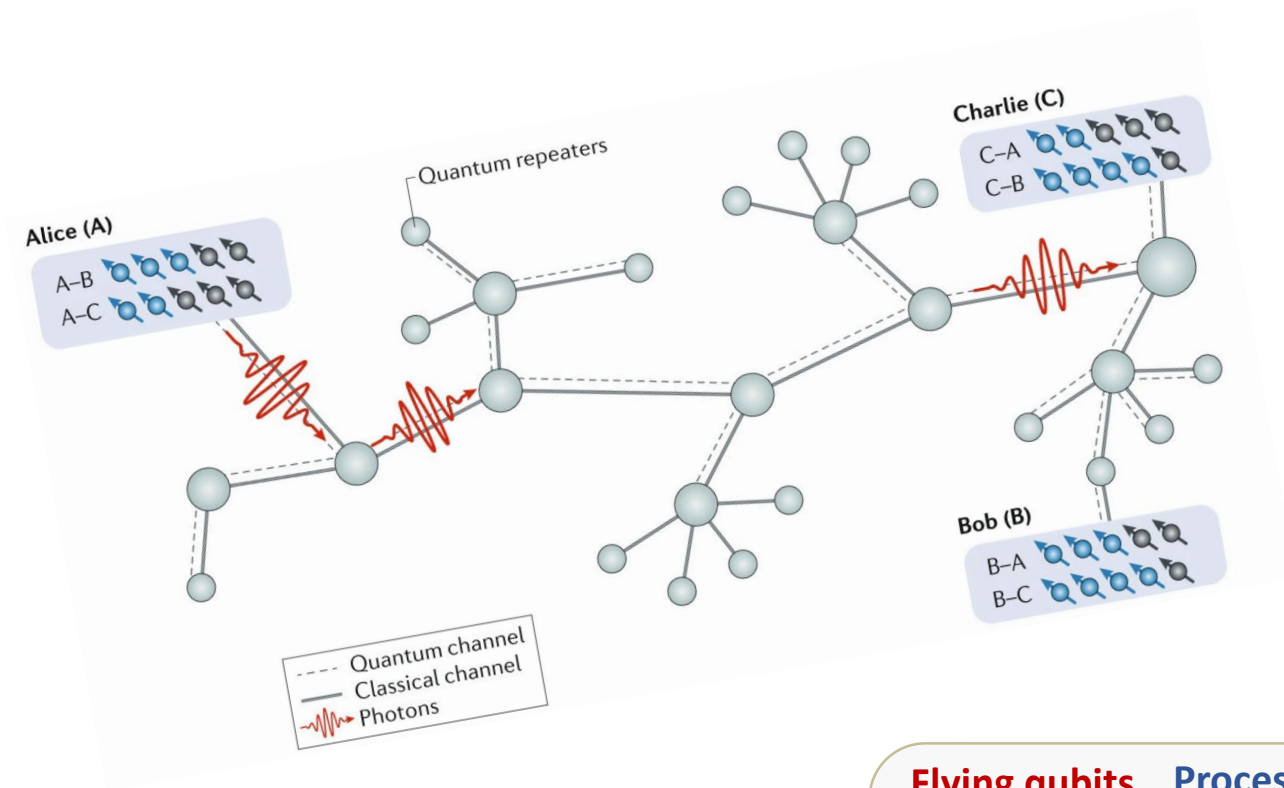


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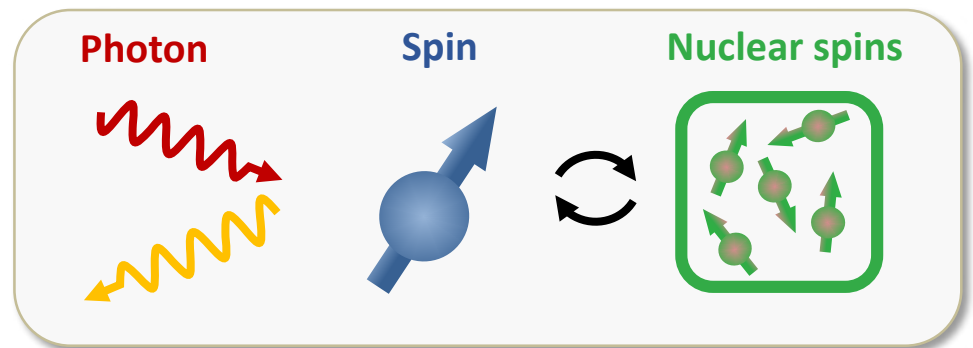
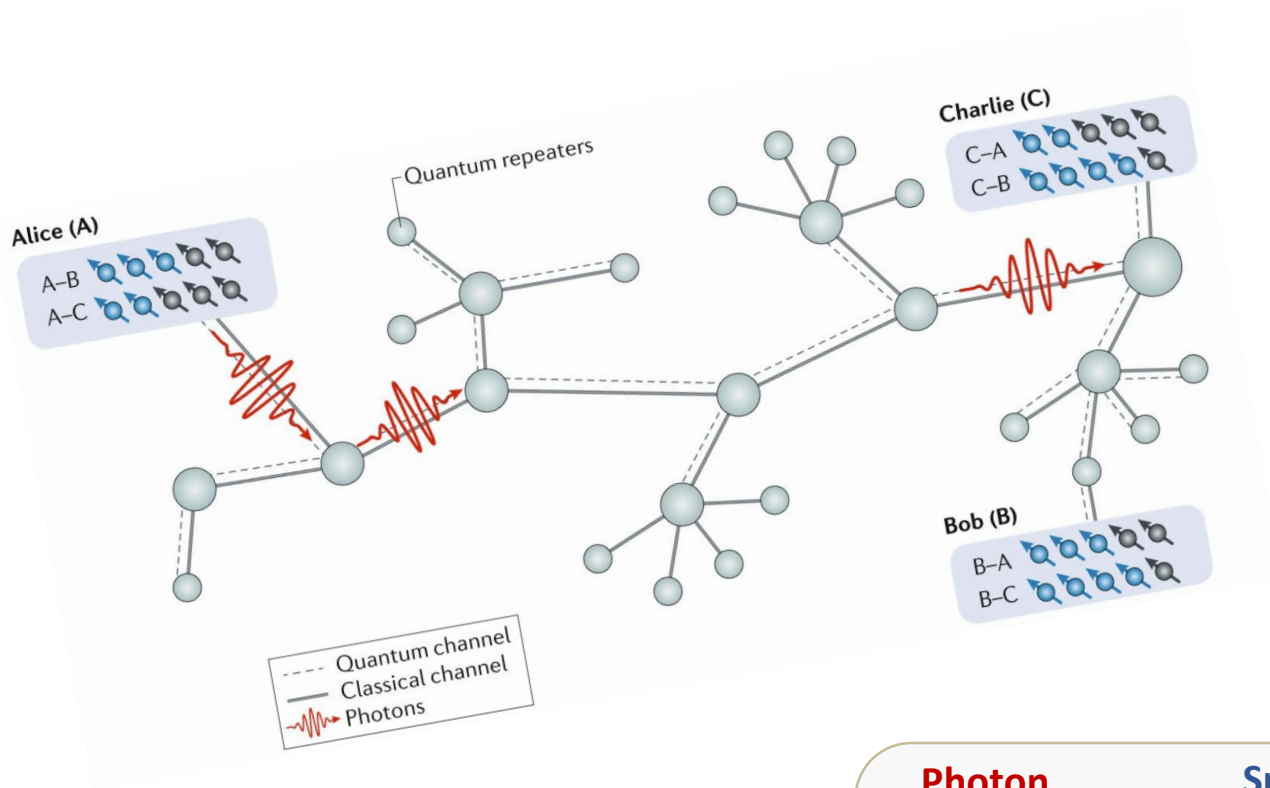
a catalogue of quantum objects



quantum nodes in a network



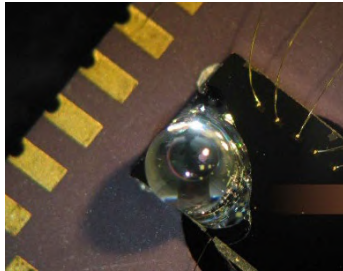
quantum nodes in a network



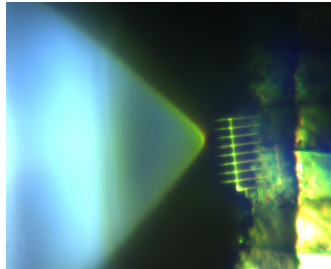
our solid-state sandbox

Quantum photonics and spin physics with many material platforms

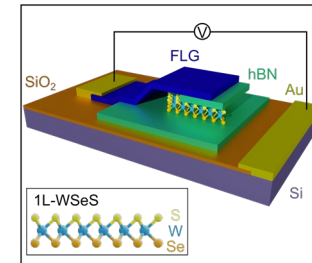
III-Vs



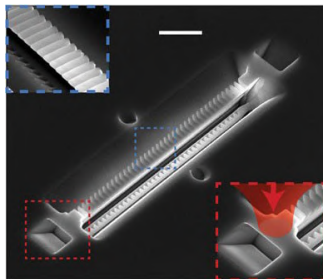
Diamond



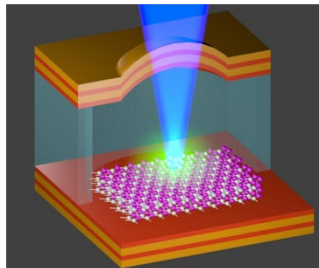
2d vdWs



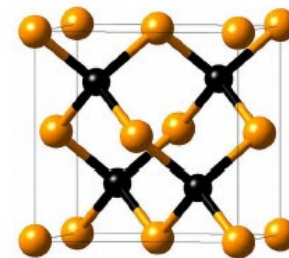
Rare Earth



hBN

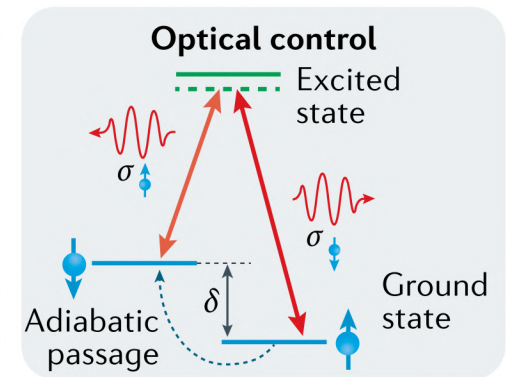
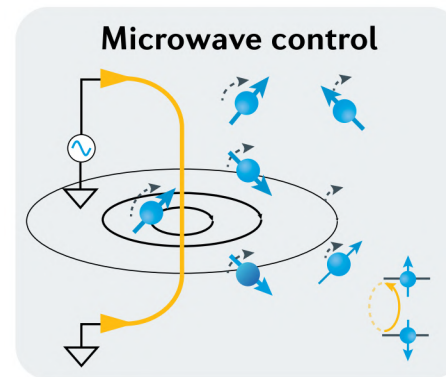
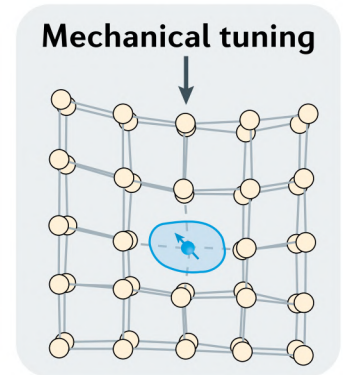
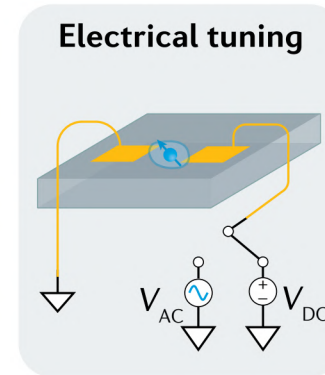
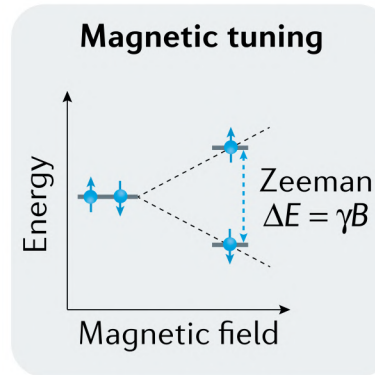
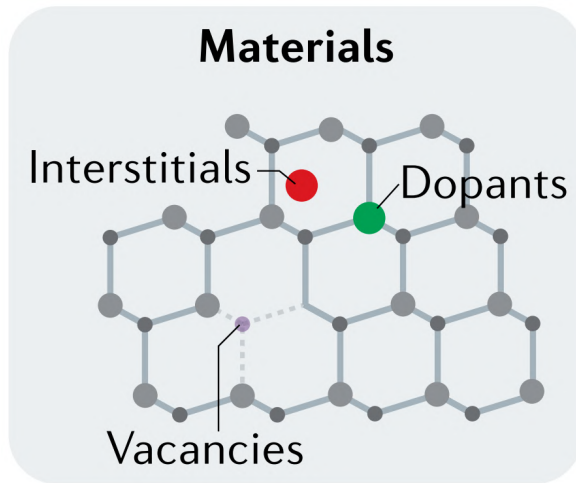


SiC



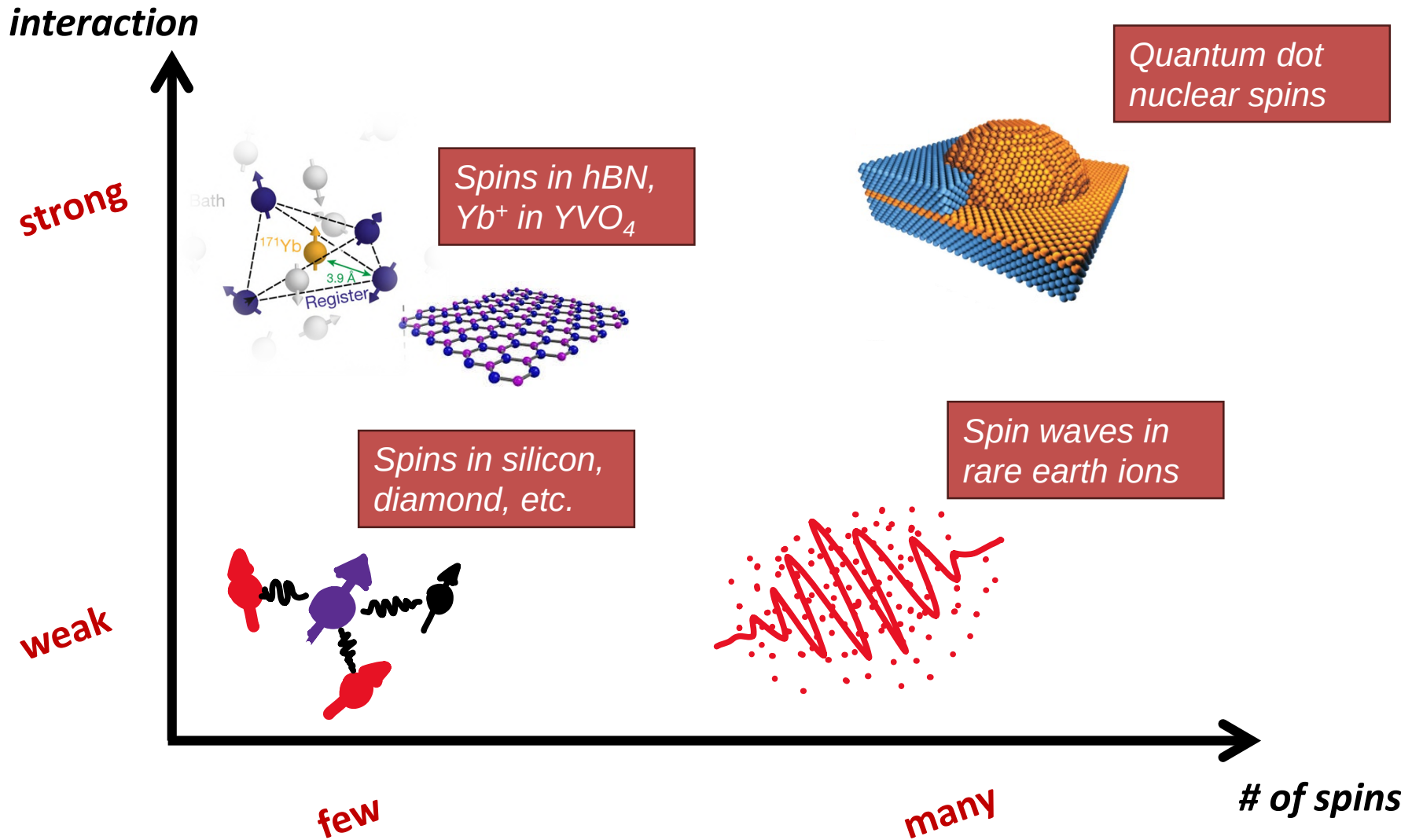
...and more!

working with optics and spin

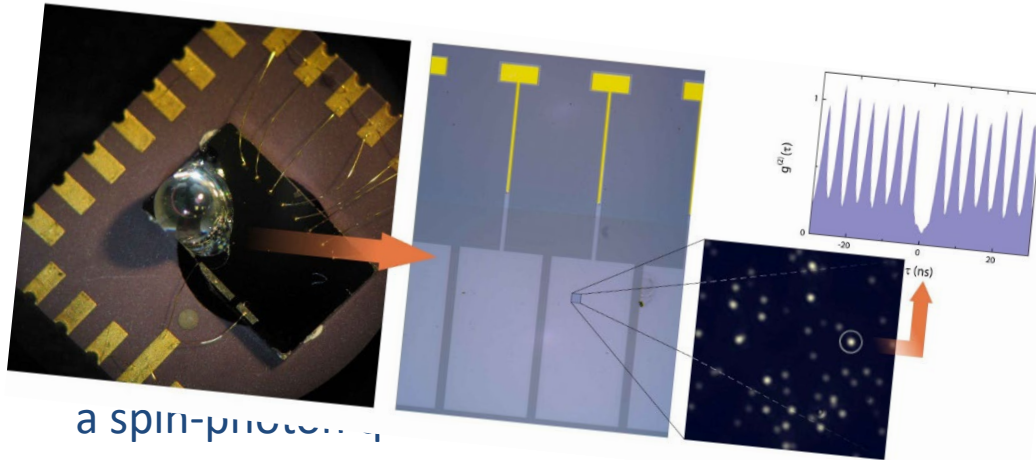
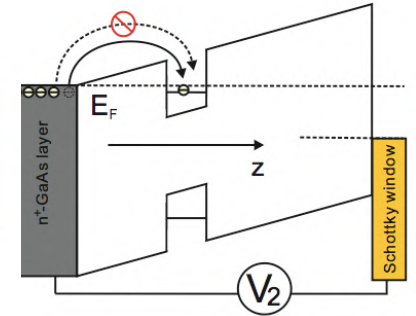
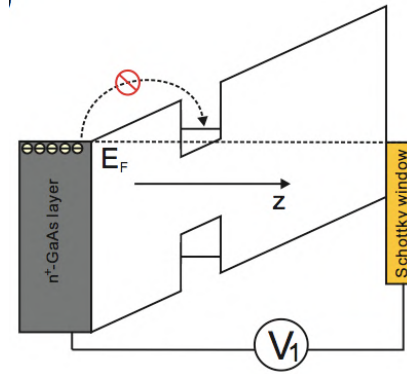
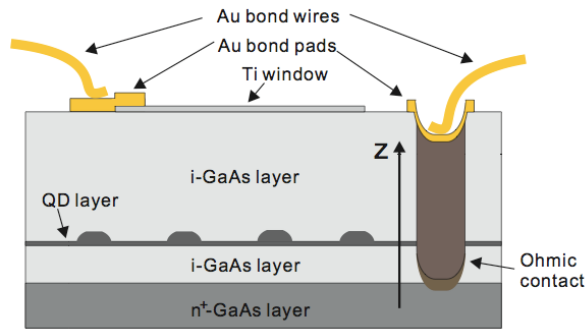
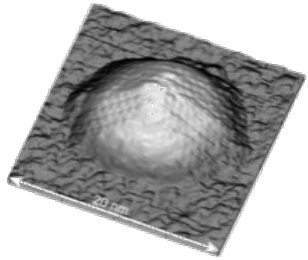


Wolfowicz et al., Nat Rev Mat, 6, 906 (2021)

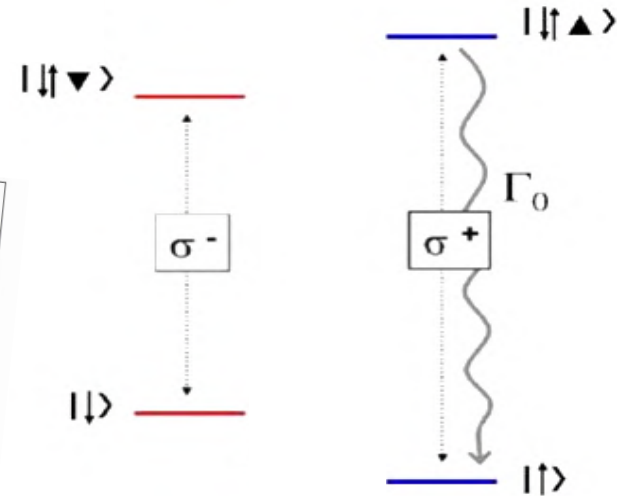
the 'parameter space' for central spin



semiconductor quantum dot devices (InGaAs)



a spin-photonic

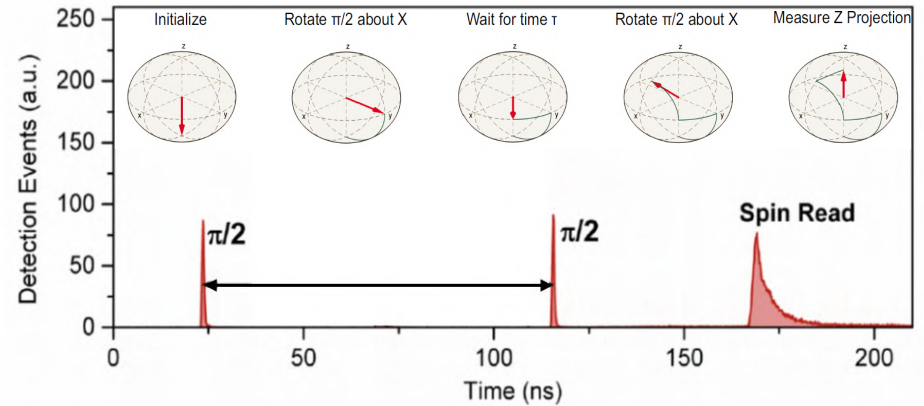
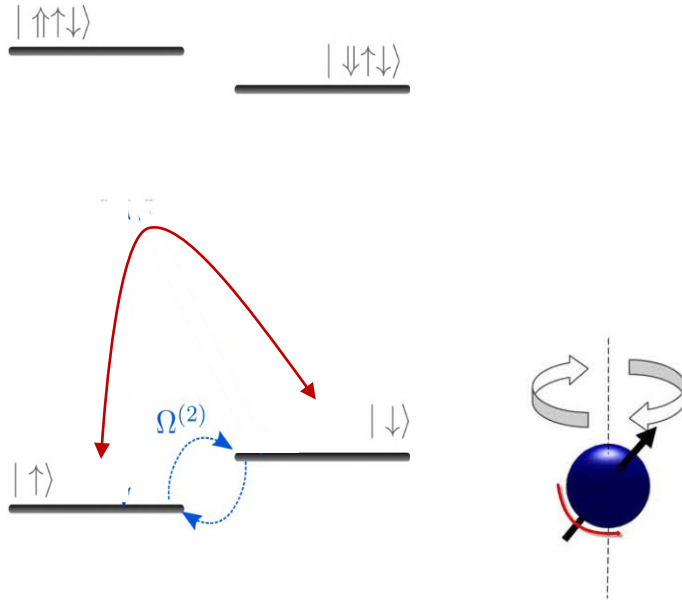


Charged Exciton
(optically accessible spin)

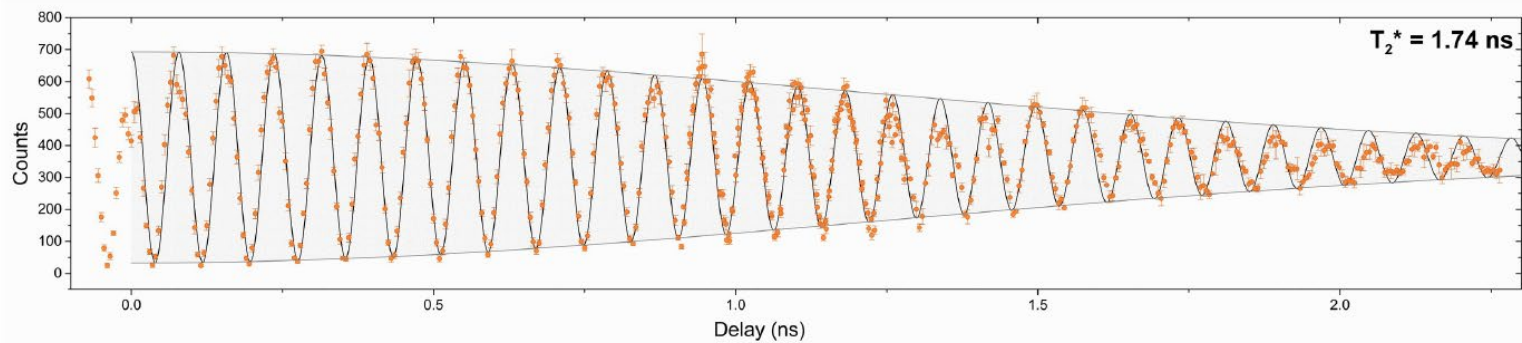
“optics” of semiconductor quantum dots



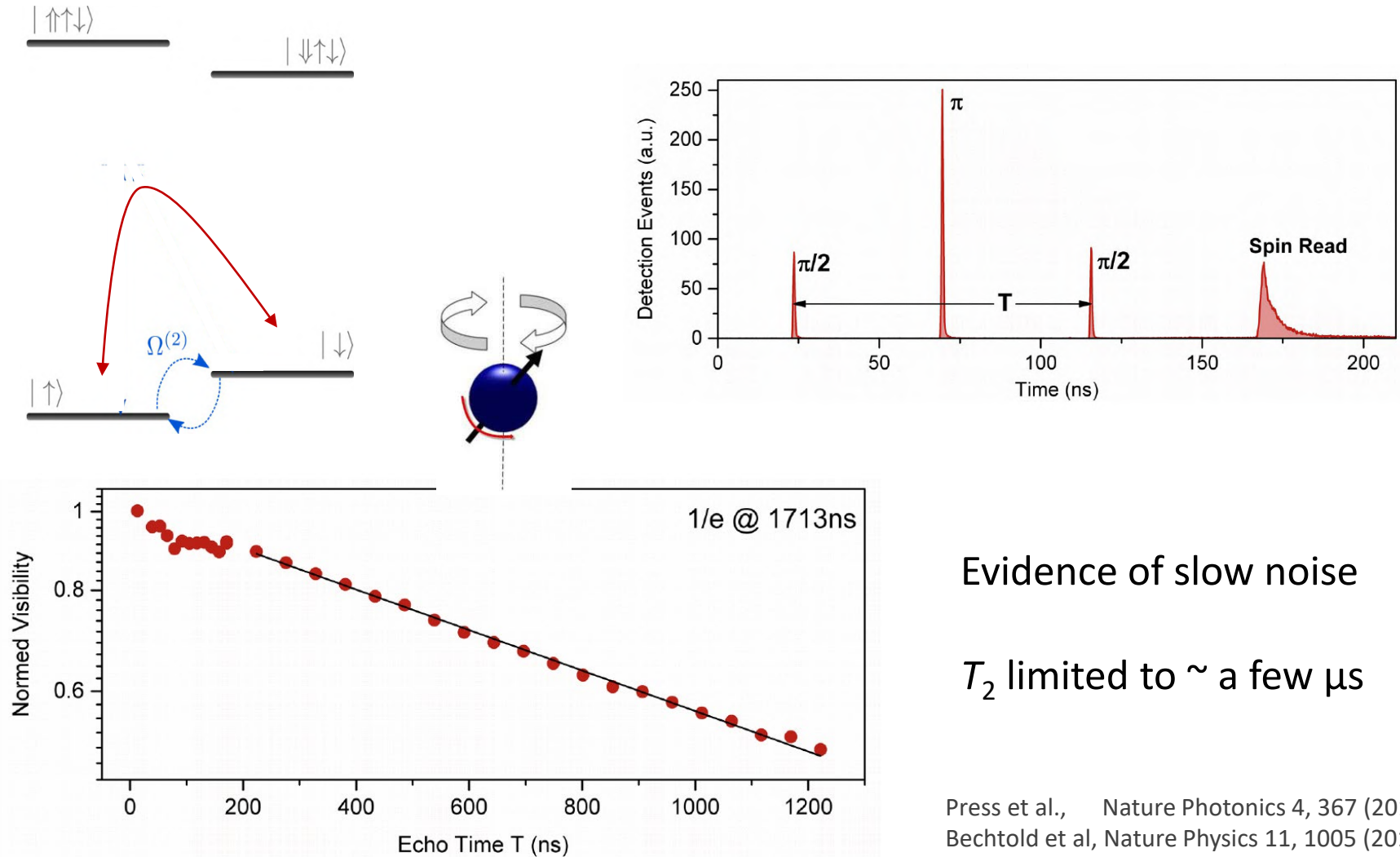
all-optical quantum control and spin coherence



Ramsey



all-optical quantum control and spin coherence

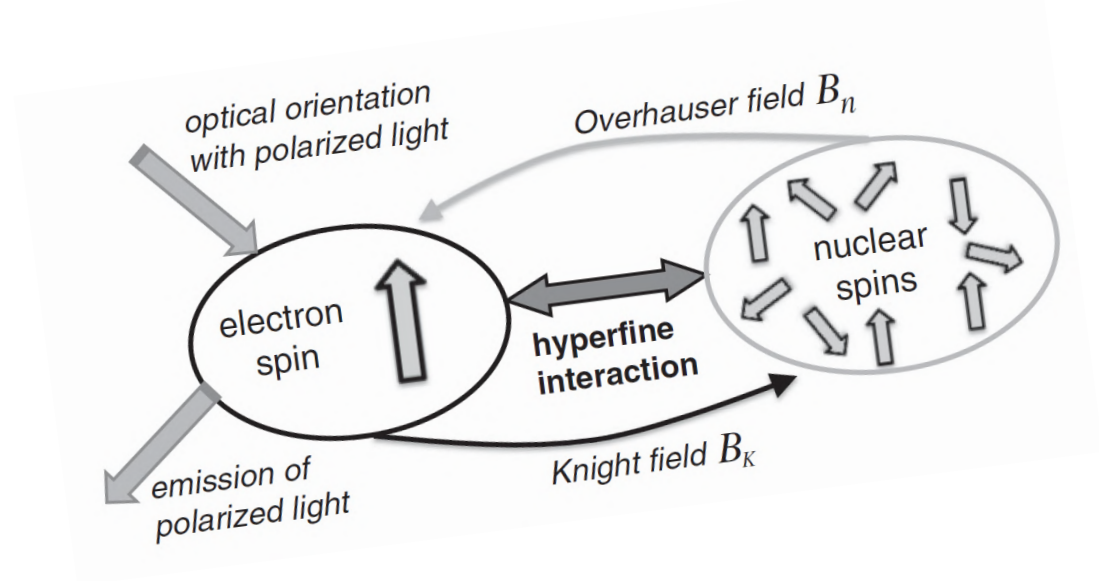
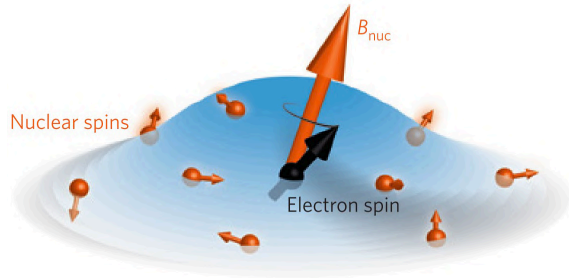


Evidence of slow noise

T_2 limited to $\sim$ a few μs

Press et al., Nature Photonics 4, 367 (2010)
Bechtold et al, Nature Physics 11, 1005 (2015)
Stockill et al., Nature Comms 7, 12745 (2016)

the central spin problem: getting rid of the problem



Dial-up your I_z :

In addition to a quasistatic (classical) magnetic noise,
hyperfine interaction allows Dynamical Nuclear Spin Polarisation!

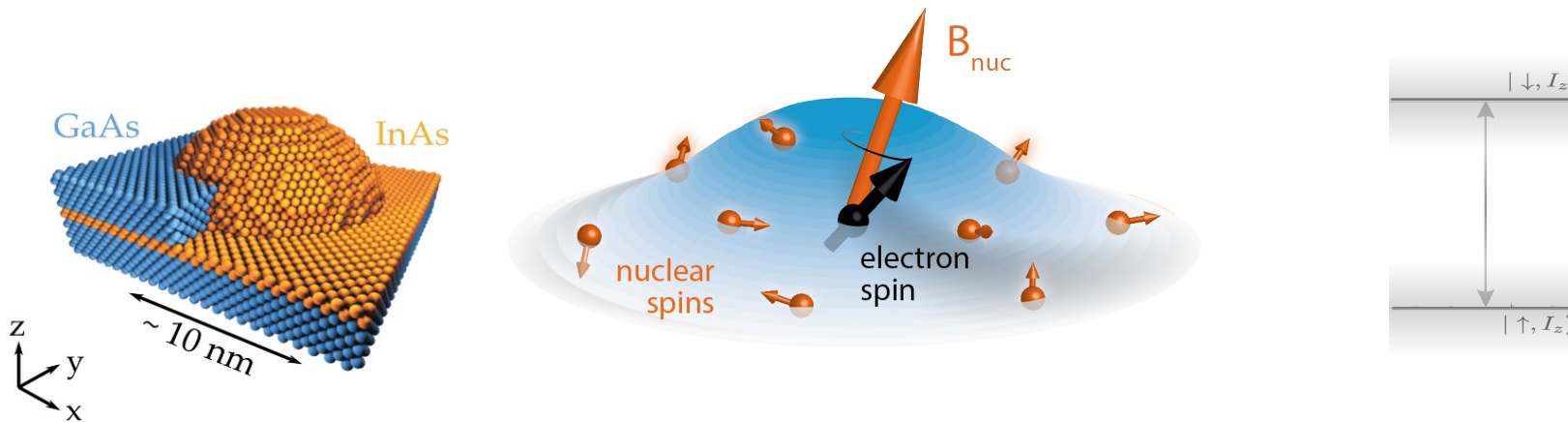
This can polarise nuclear spins! (~70% achieved)

There is an Option B!

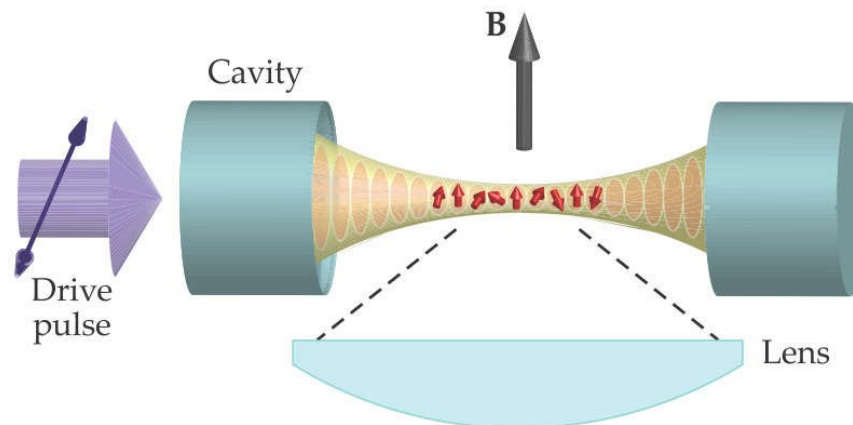
Urbaszek et al., *Reviews of Modern Physics* **85**, 79 (2013)
Chekhovich et al., *Nature Materials* **12**, 494 (2013)

the central spin problem: a many-body physics toolbox

There is strong interaction between the electron and the nuclear spins ($\sim 50,000!$)

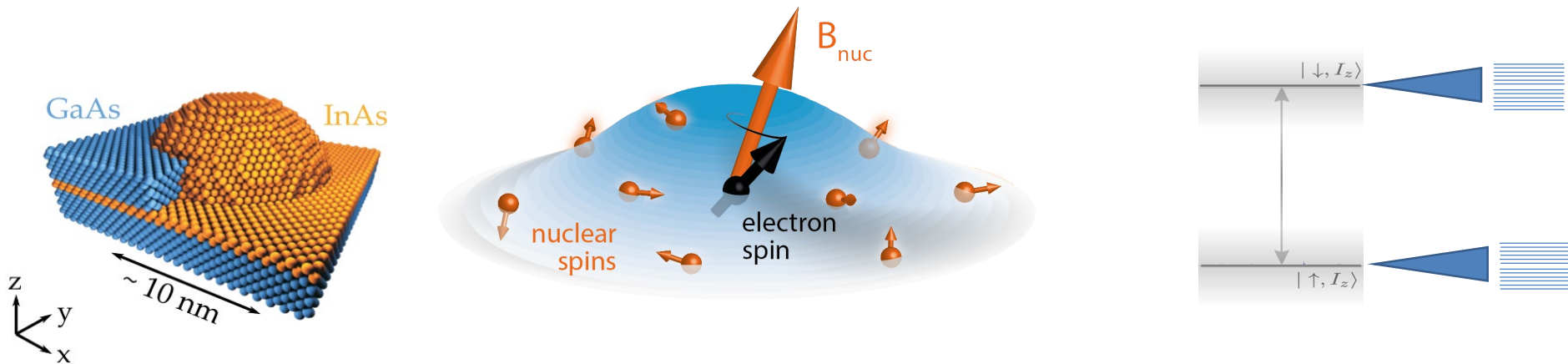


Analogies with an atomic ensemble coupled strongly to a cavity mode



the central spin problem: a many-body physics toolbox

There is strong interaction between the electron and the nuclear spins ($\sim 50,000!$)

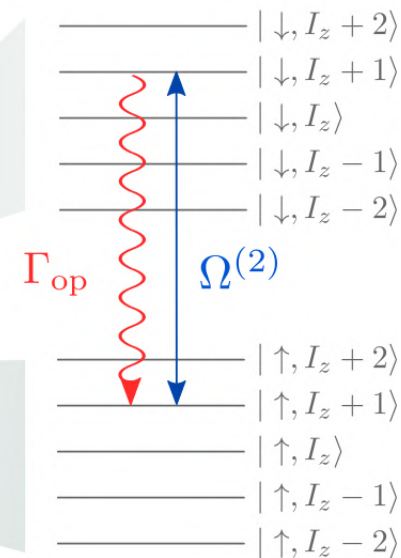
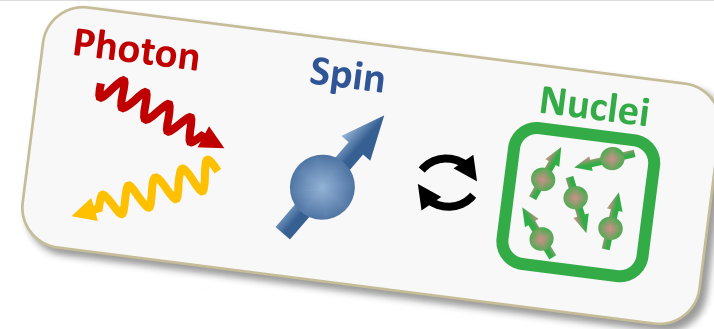
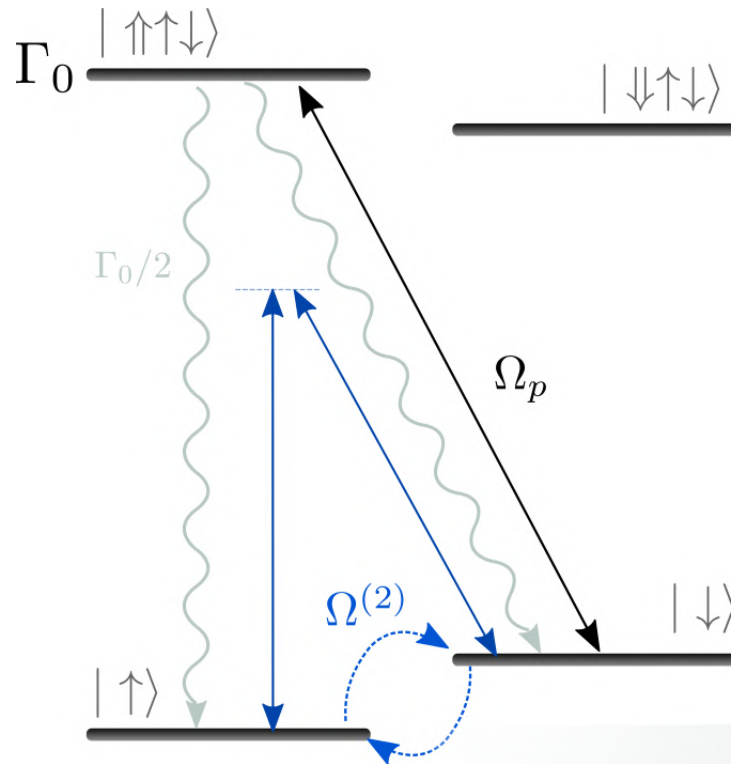


Analogies with an atomic ensemble coupled strongly to a cavity mode

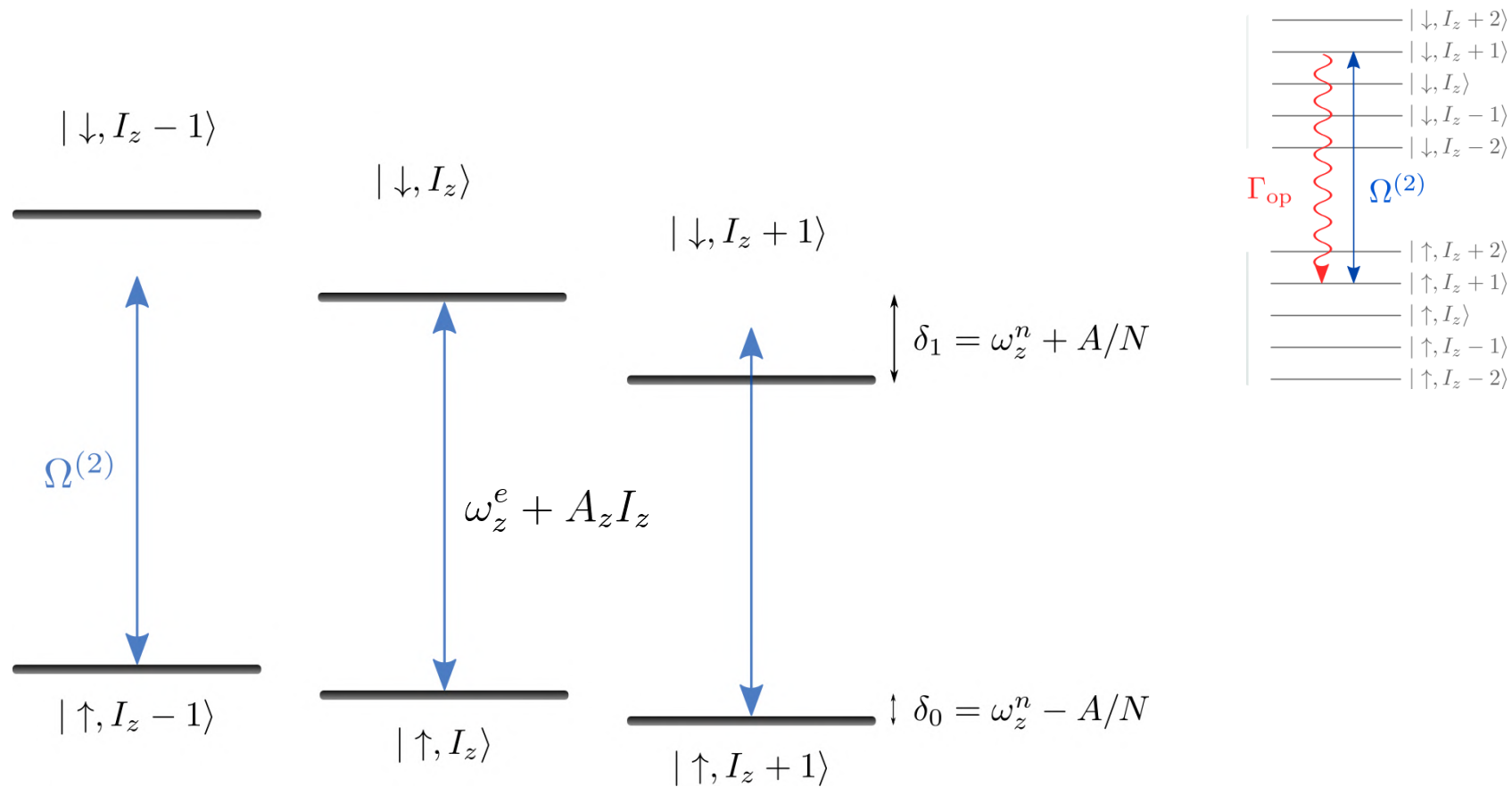
theorists got excited...

- Robust quantum memory [Taylor et al., PRL **90**, 206803 (2005)]
- Collective dark states [Christ et al., PRB **75**, 155324 (2007)]
- Squeezed nuclear states [Rudner et al., PRL **107**, 206806 (2011)]
- Dissipative phase transitions [Kessler et al., PRA **86**, 012116 (2012)]

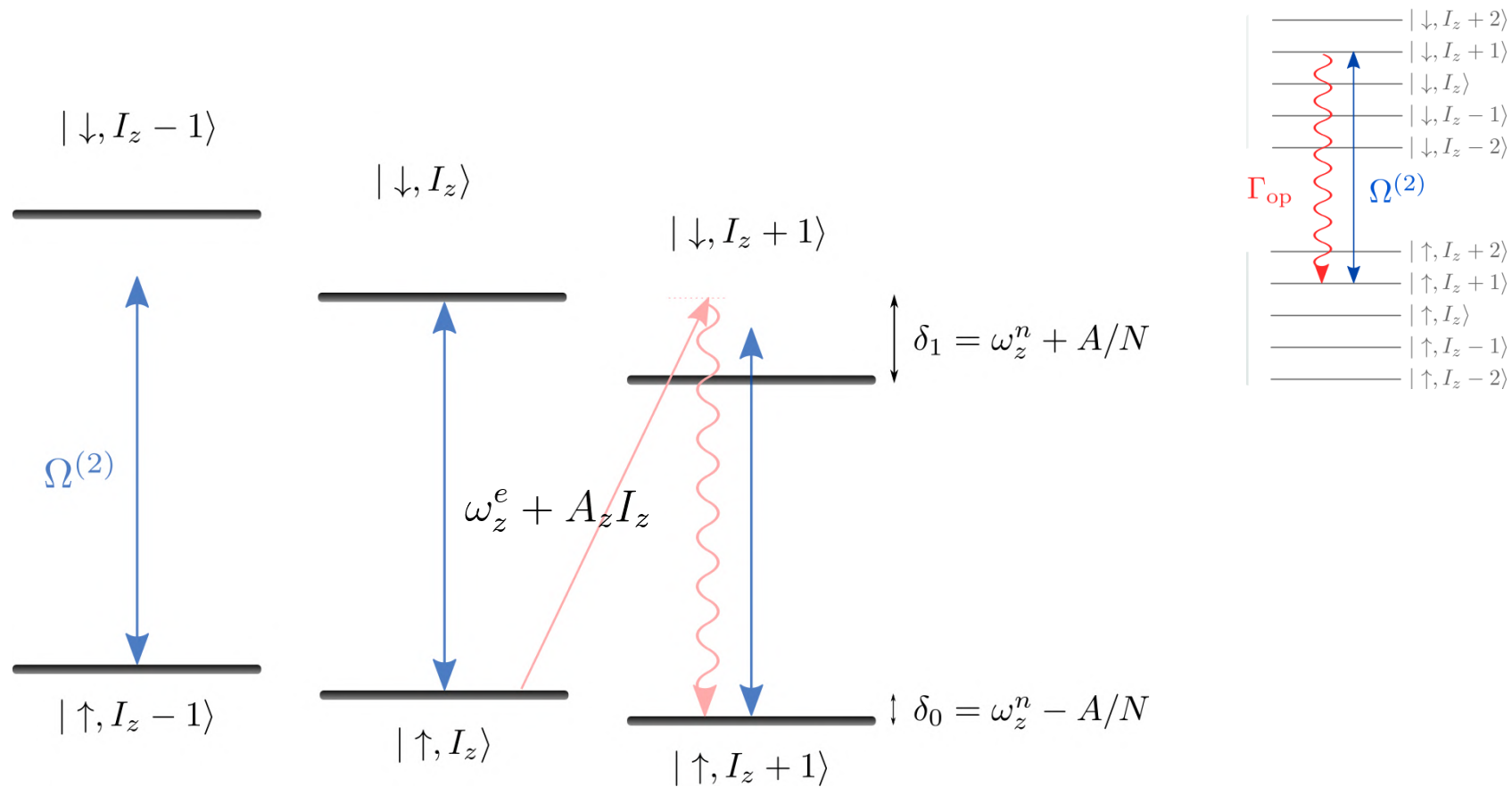
the electronuclear spin-photon coupled system



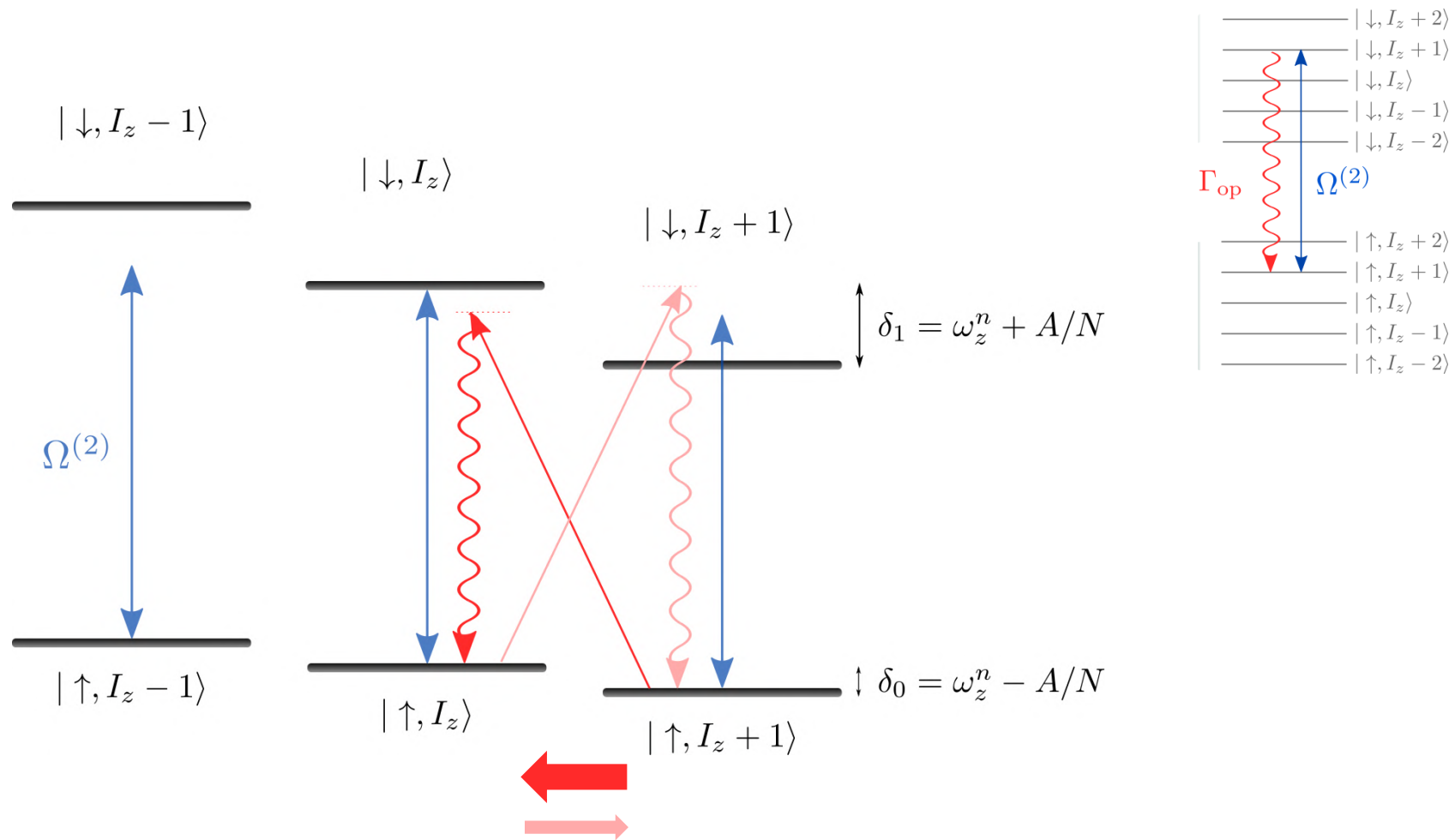
optical orientation of nuclear spins



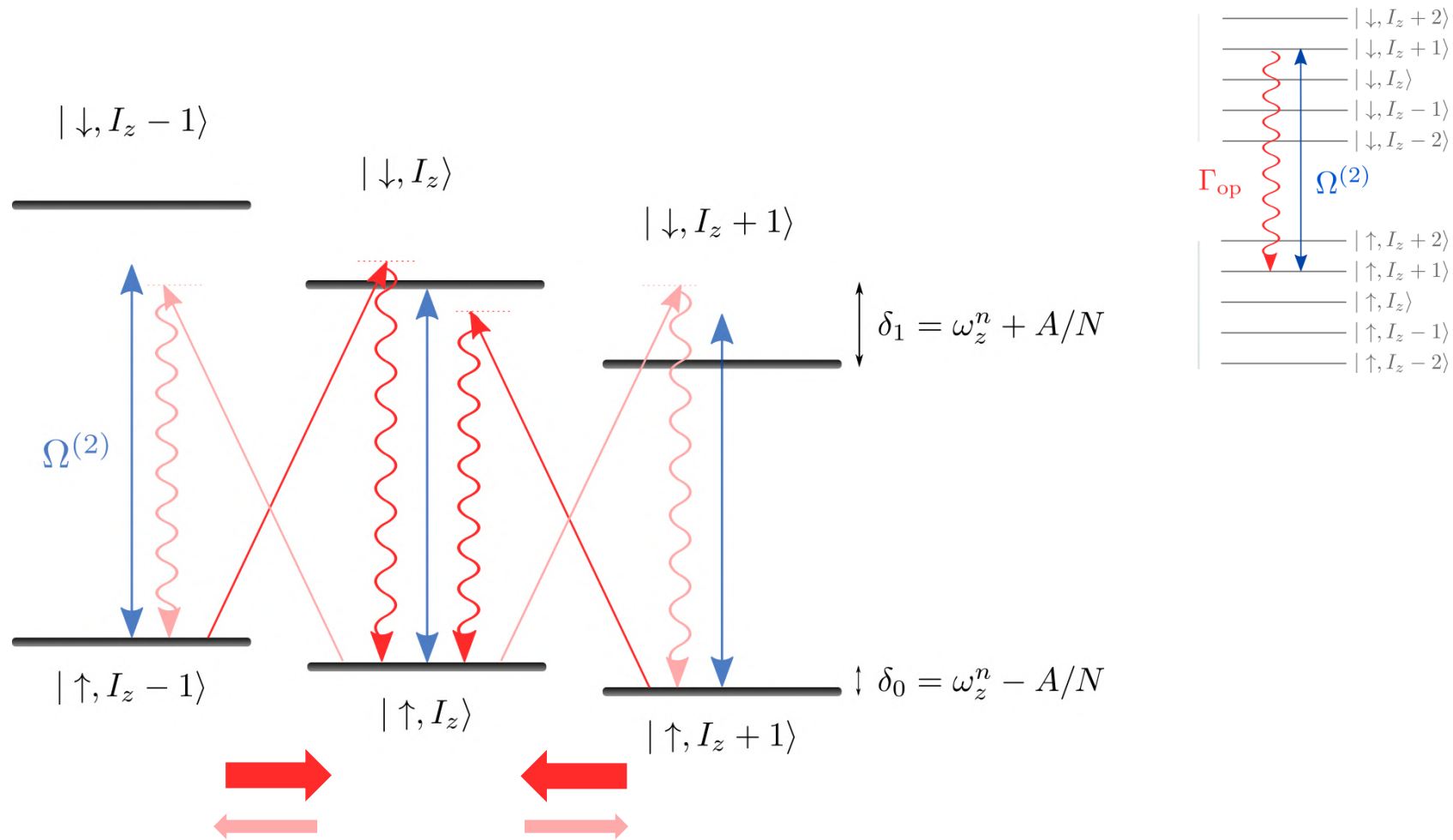
optical orientation of nuclear spins



optical orientation of nuclear spins

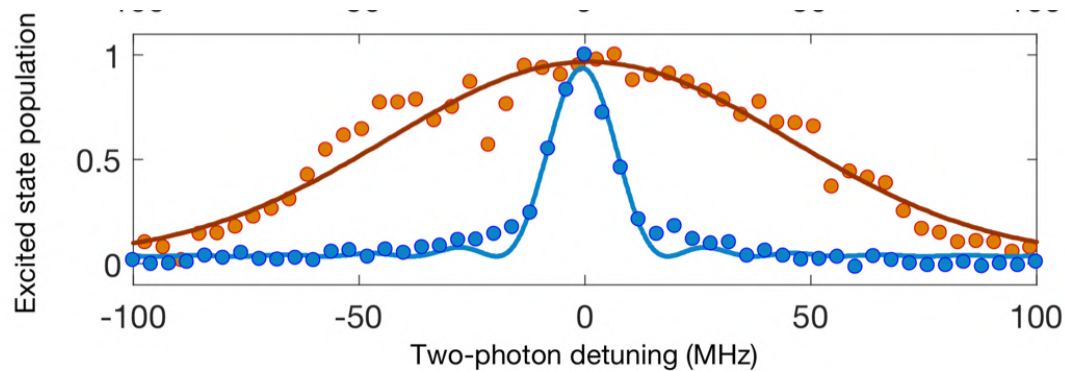
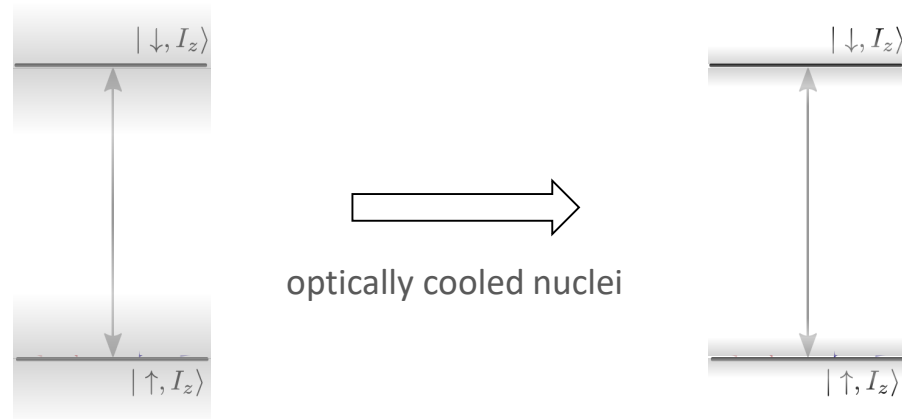
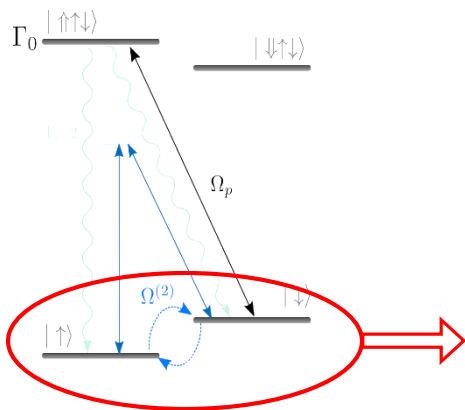


optical orientation of nuclear spins



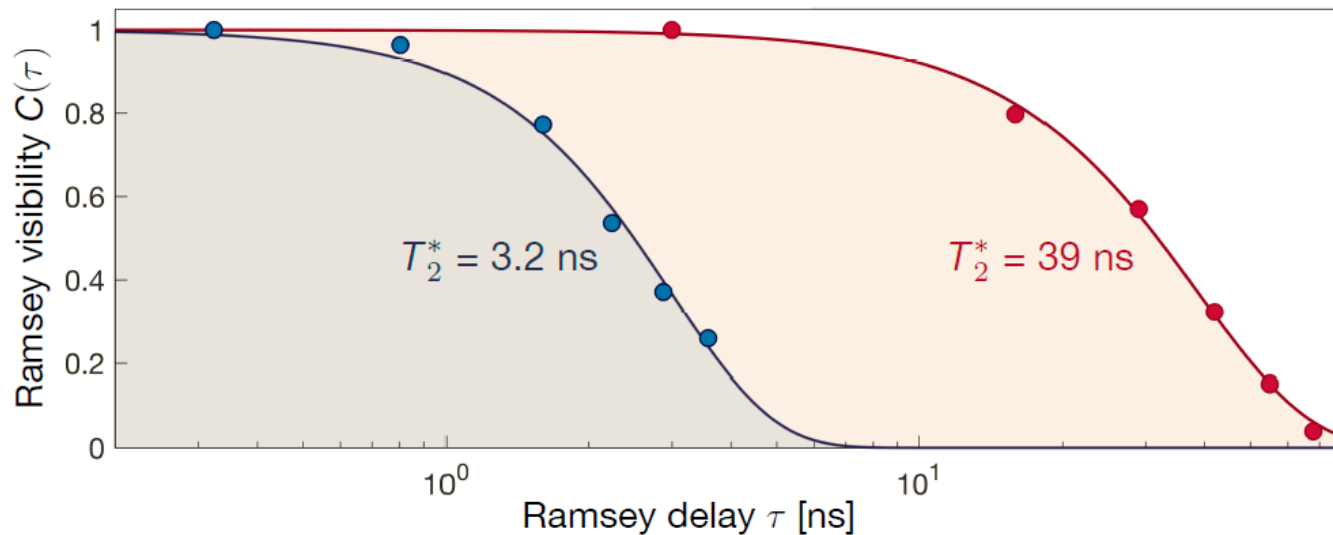
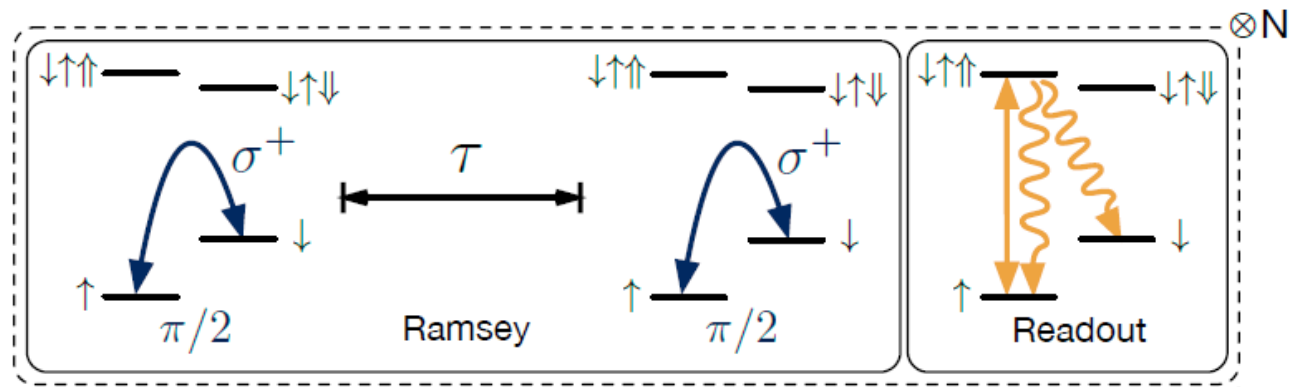
The anharmonic ladder of states leads to a single I_z determined by two-photon detuning.

how cool are those cooled nuclei?



Measured ESR linewidth is ~ 15 MHz $\rightarrow 400$ μ K

electron coherence after suppressing nuclear magnetic noise

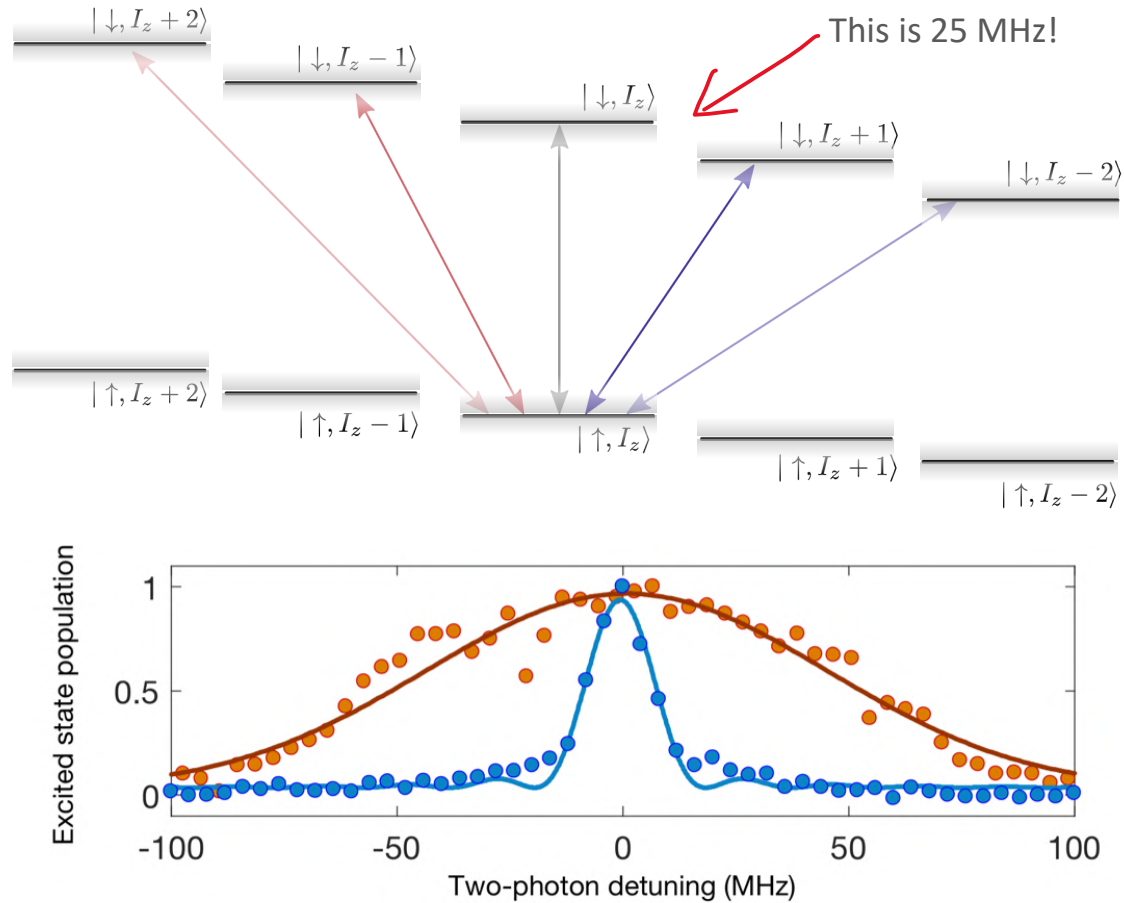


Ethier-Majcher et al., PRL **119** 130503 (2017)



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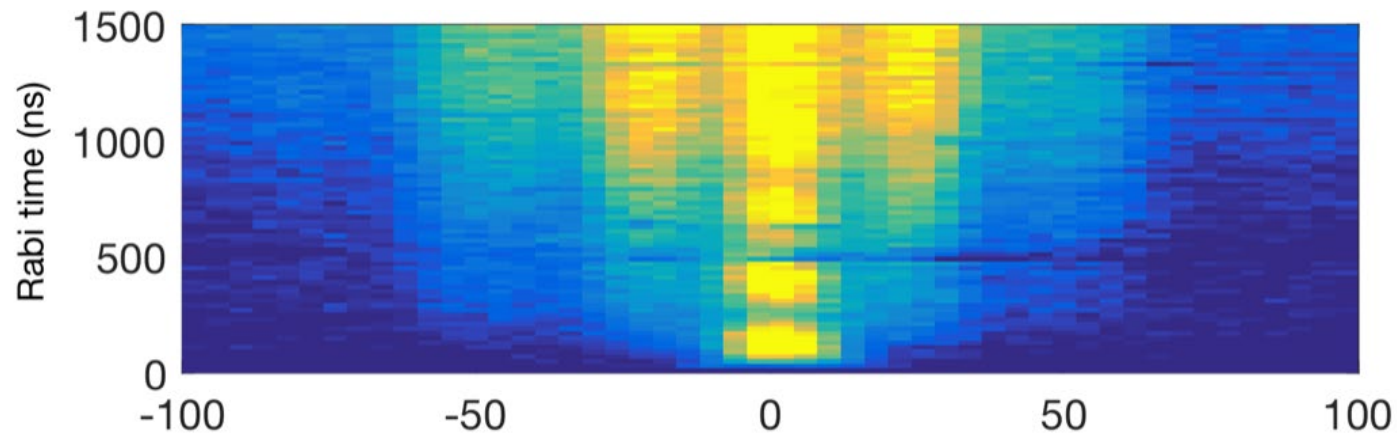
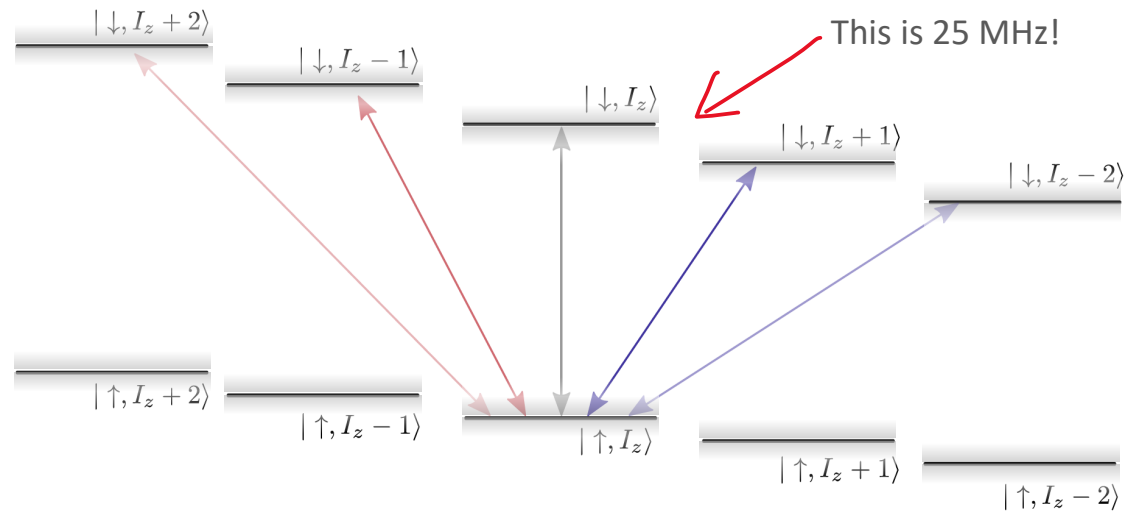
measuring nuclear spin variance via optical ESR



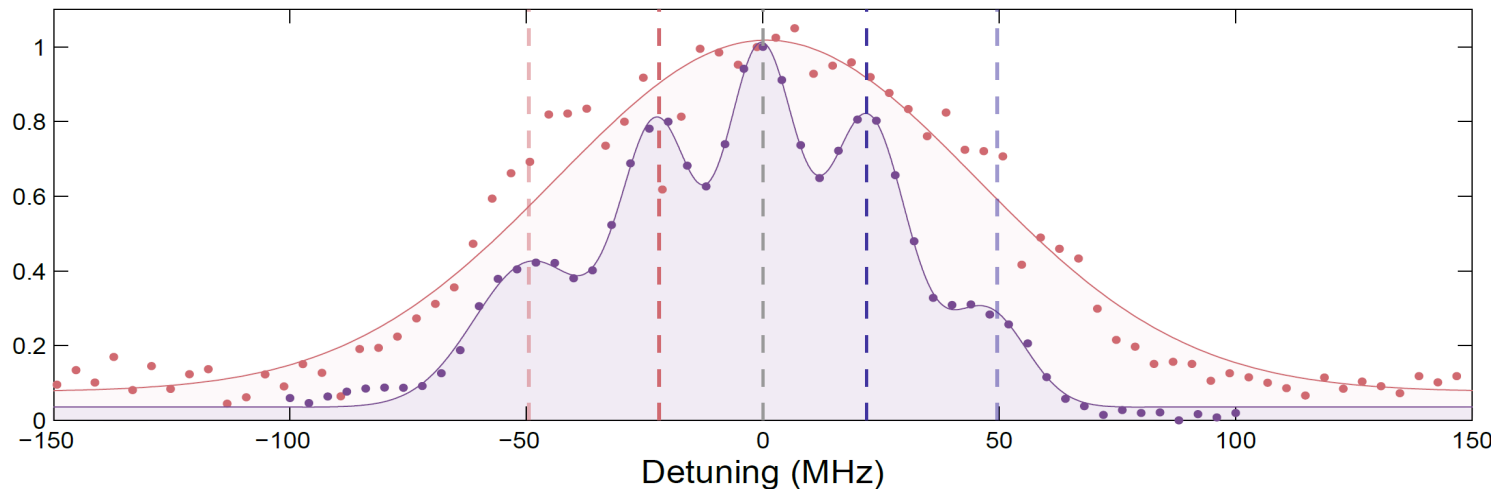
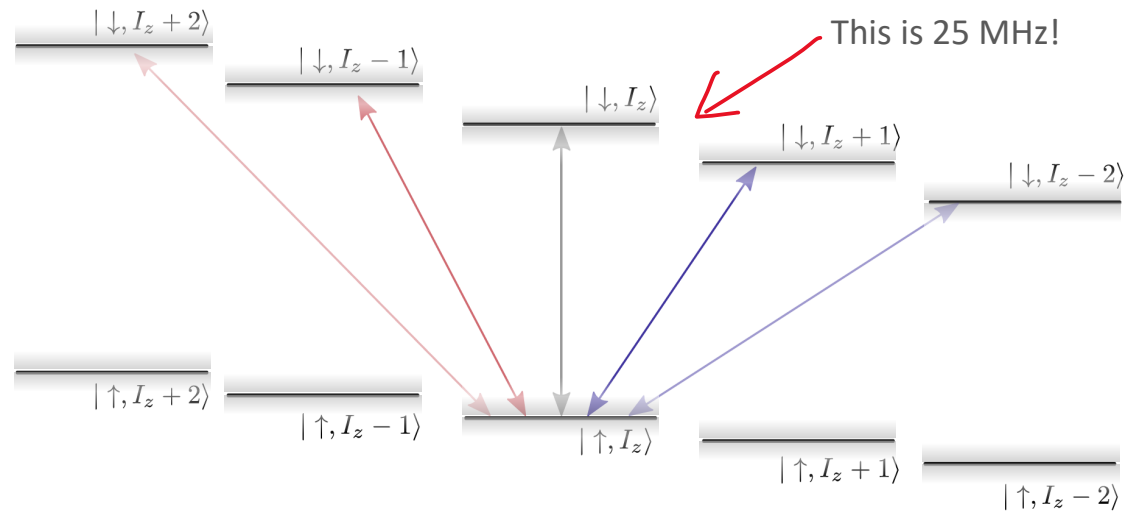
Measured ESR linewidth is ~ 15 MHz $\rightarrow 400$ μ K



entering the sideband-resolved regime

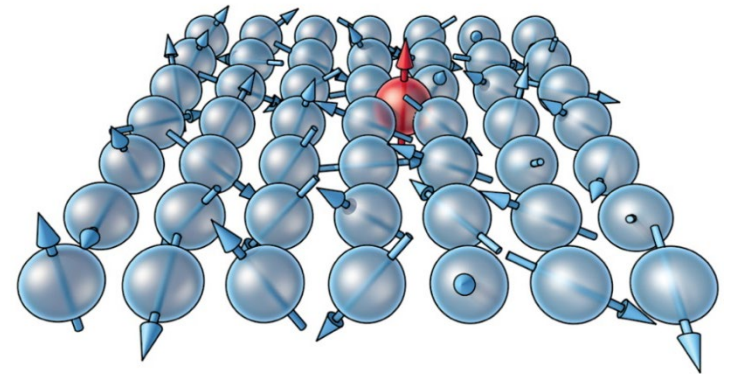
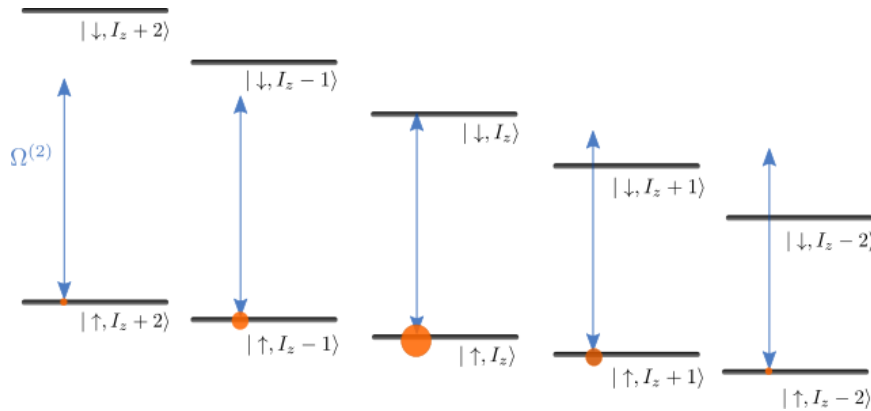


entering the sideband-resolved regime

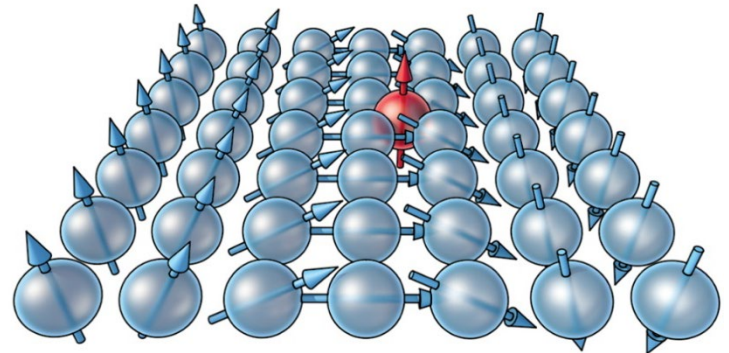
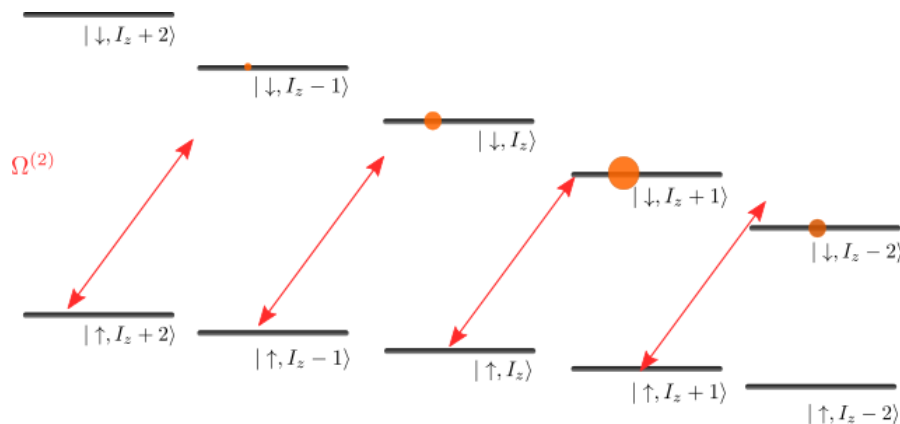


driving of a collective nuclear excitation

1. Prepare the nuclear bath in a cooled state

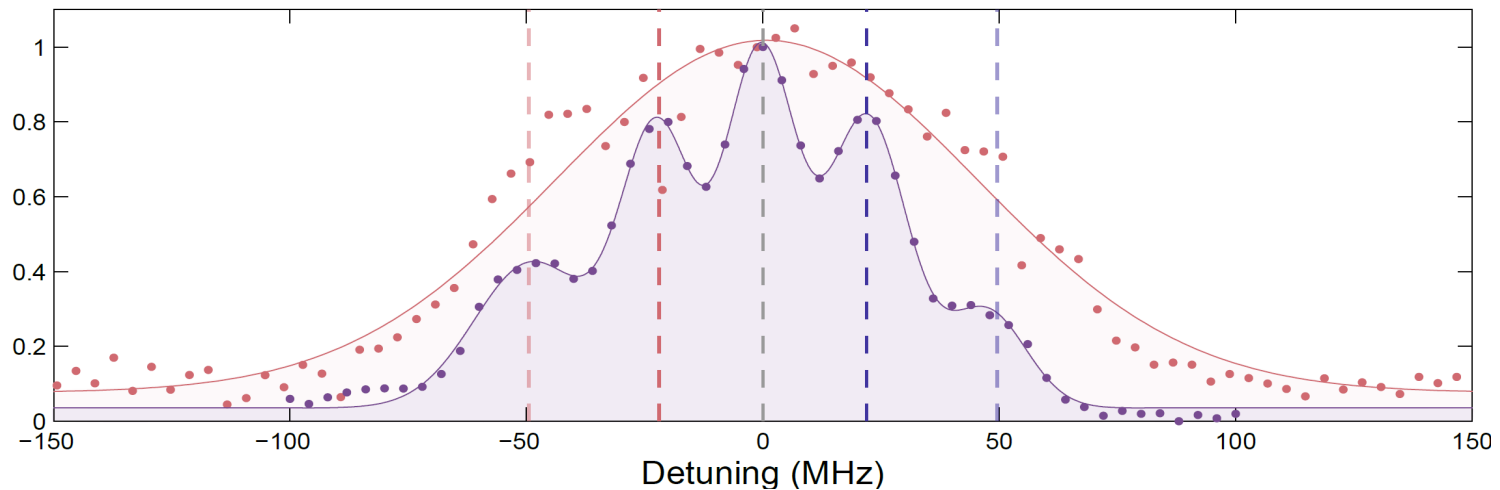
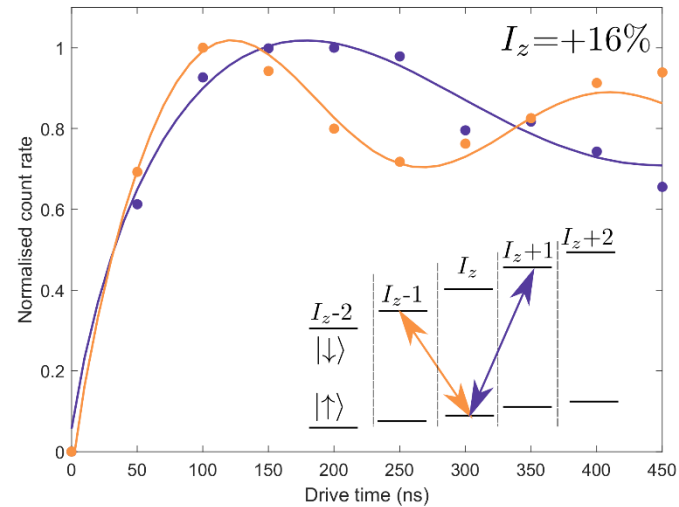
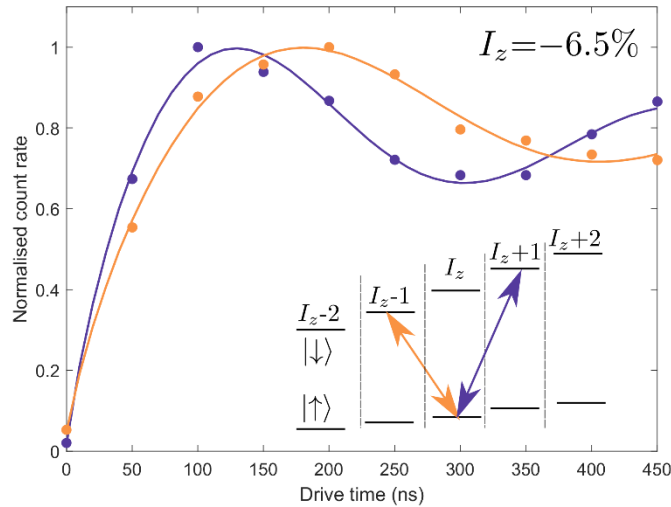


2. Coherently drive a nuclear excitation sideband



collective coherence of nuclear magnons

Collectively enhanced spin-excitation: $\Omega_{e-nuc} \propto \sqrt{N} A_i^{nc}$

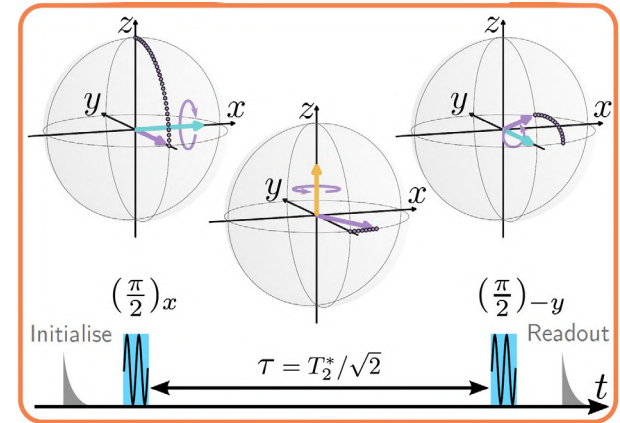
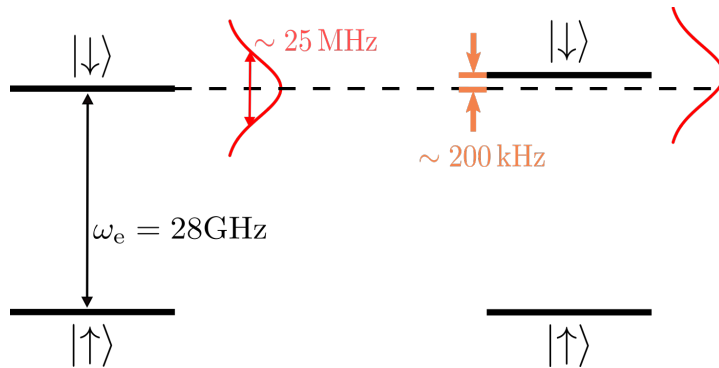
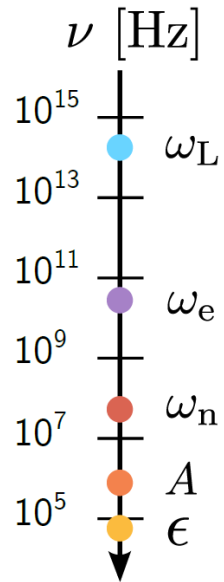


Gangloff *et al.*, Science 364, 62 (2019).

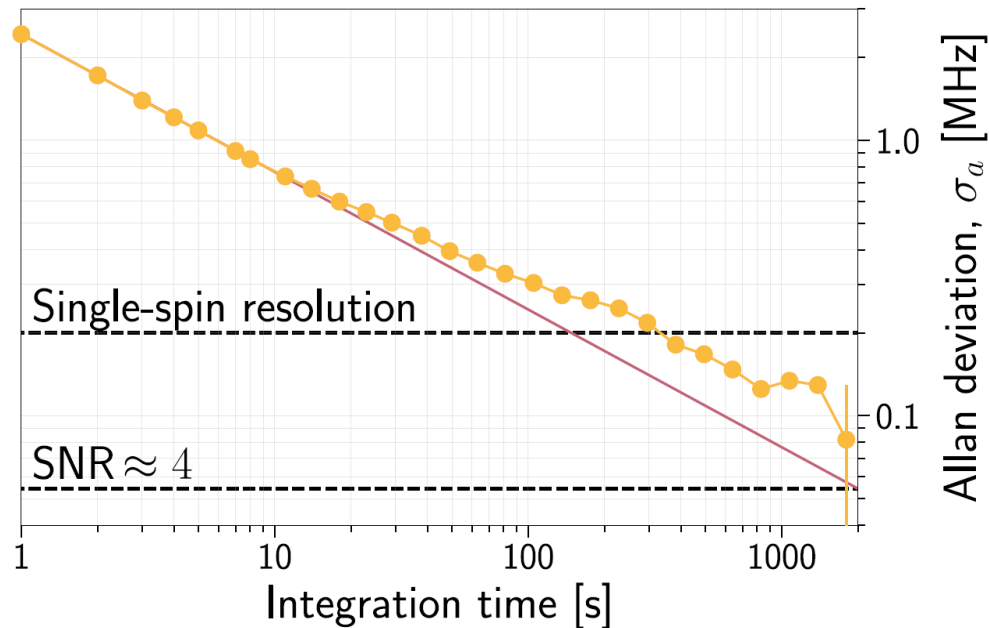


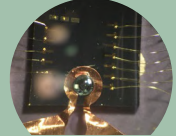
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Sensing: full experimental sequence

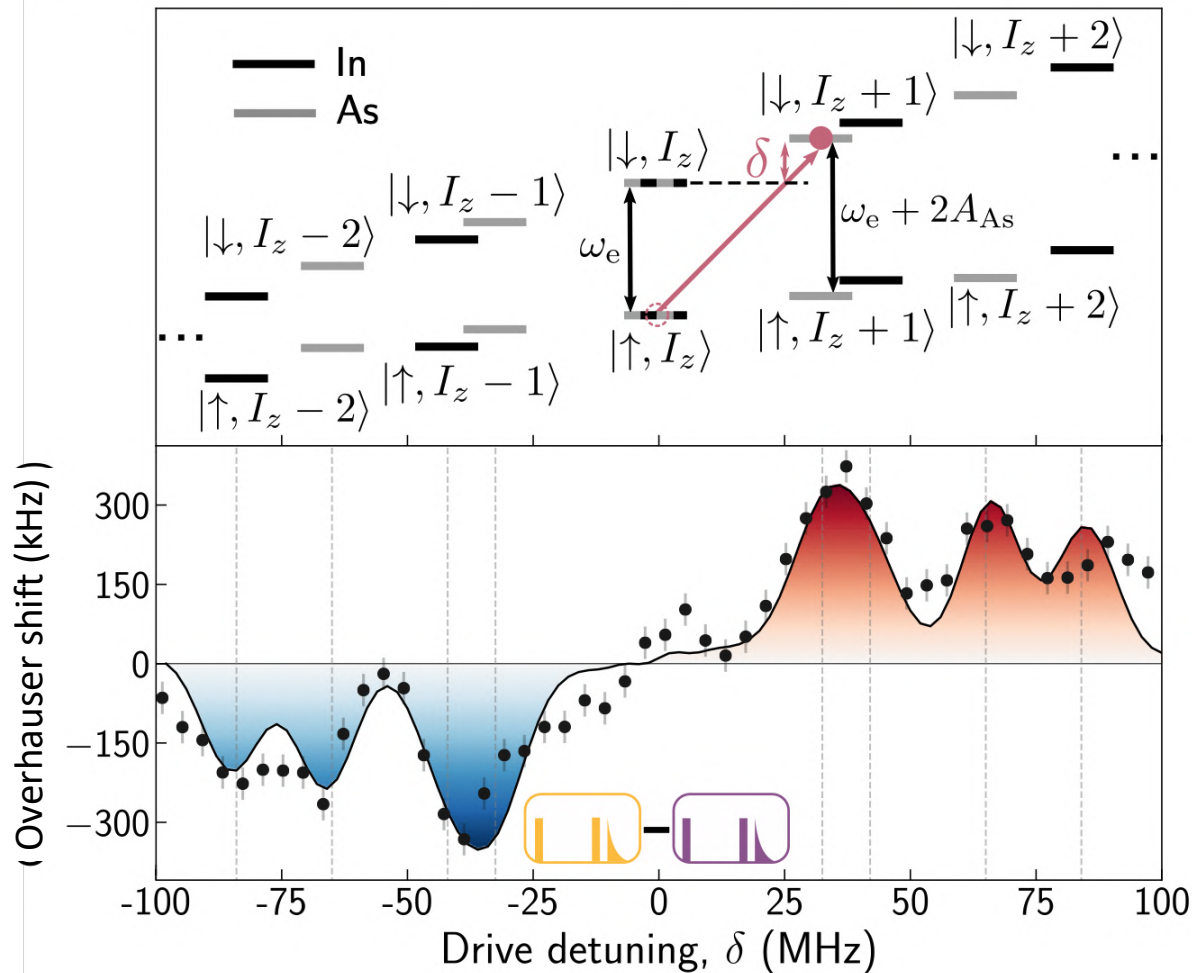
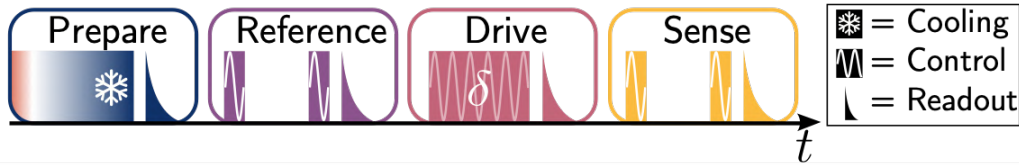


Frequency shift detected via Ramsey Interferometry





Magnon sensing: Spectrum

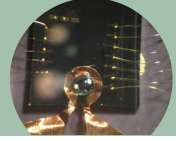


(+) electron phase

Spin-up magnon

Spin-down magnon

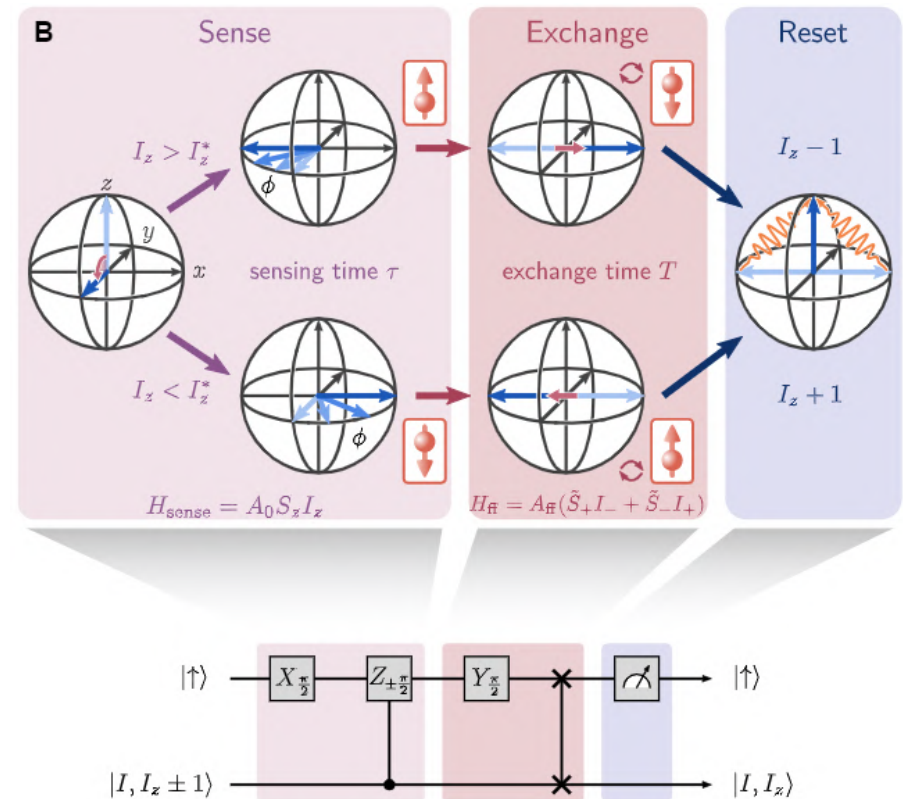
(-) electron phase



Pushing the limits of cooling

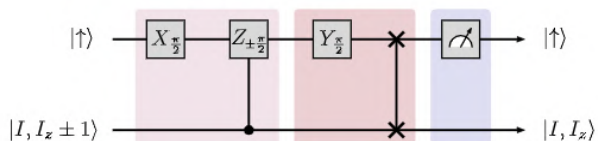
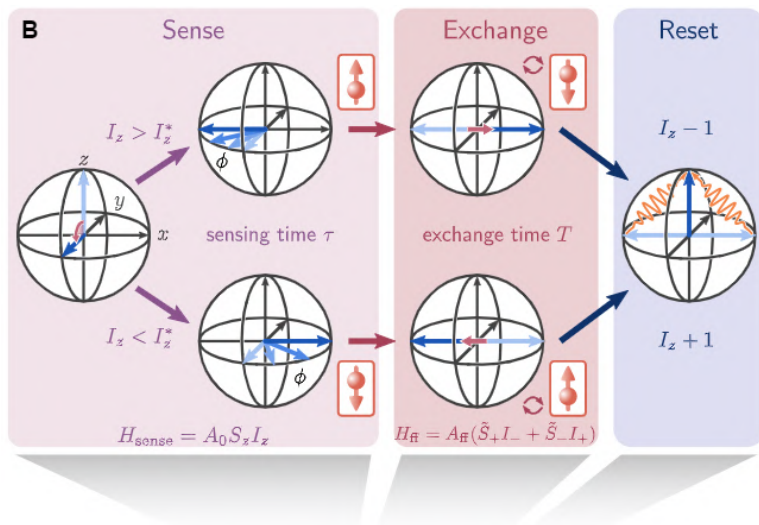
Cooling the nuclei via electro-nuclear coupling in the steady-state

Sensing the writing of a single magnon into the nuclei

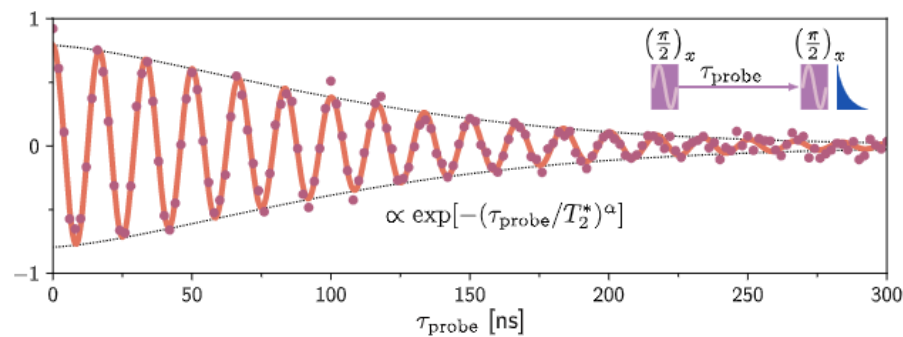
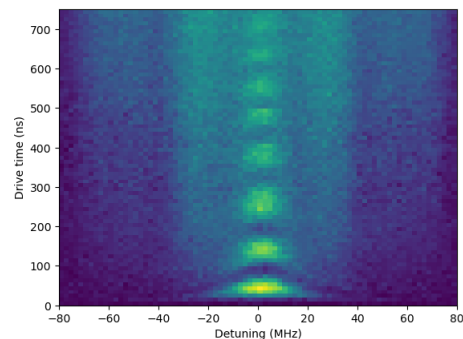




Pushing the limits of cooling



Raman cooling



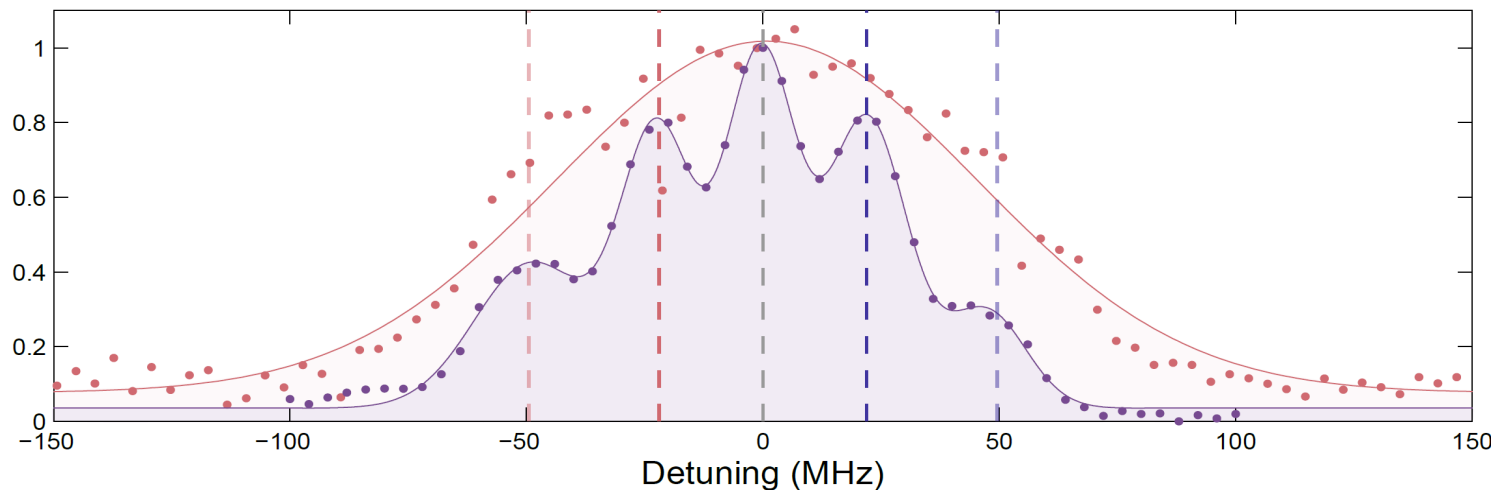
an inhomogeneous ensemble of species (^{113}In , ^{115}In , ^{69}Ga , ^{71}Ga , ^{75}As)

Strain is good!
(quadrupolar enhancement)

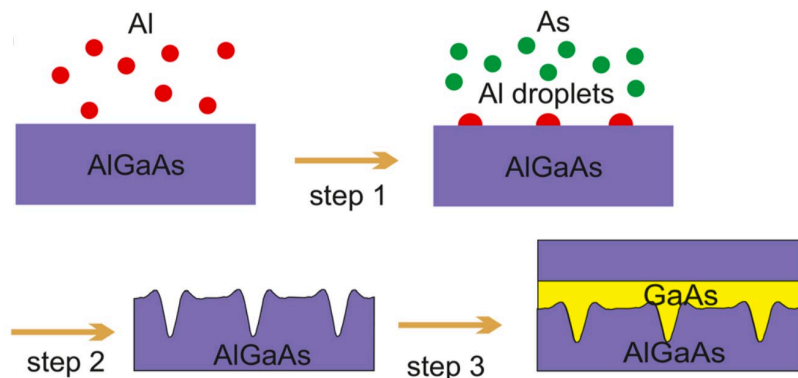
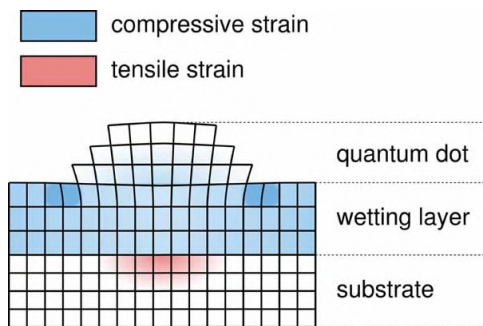
Species have slightly different
gyromagnetic ratios

Strain *dispersion* is bad!
(inhomogeneity of nuclei)

Heterogeneity contributes to
the modest magnon coherence



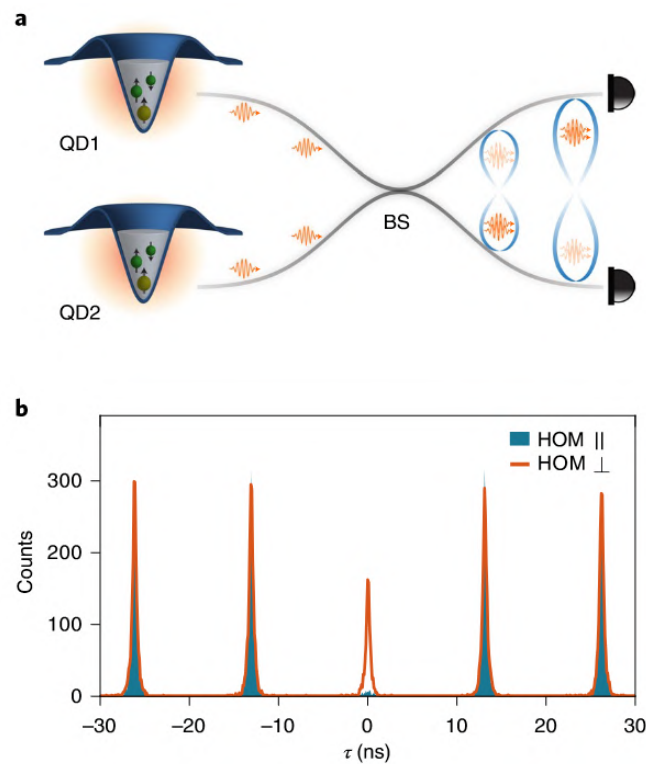
next generation quantum-dot devices



Negligible difference in lattice constant

Quantum interference of identical photons from remote GaAs quantum dots

Liang Zhai¹✉, Giang N. Nguyen¹, Clemens Spinnler¹, Julian Ritzmann², Matthias C. Löbl¹, Andreas D. Wieck², Arne Ludwig², Alisa Javadi¹ and Richard J. Warburton¹

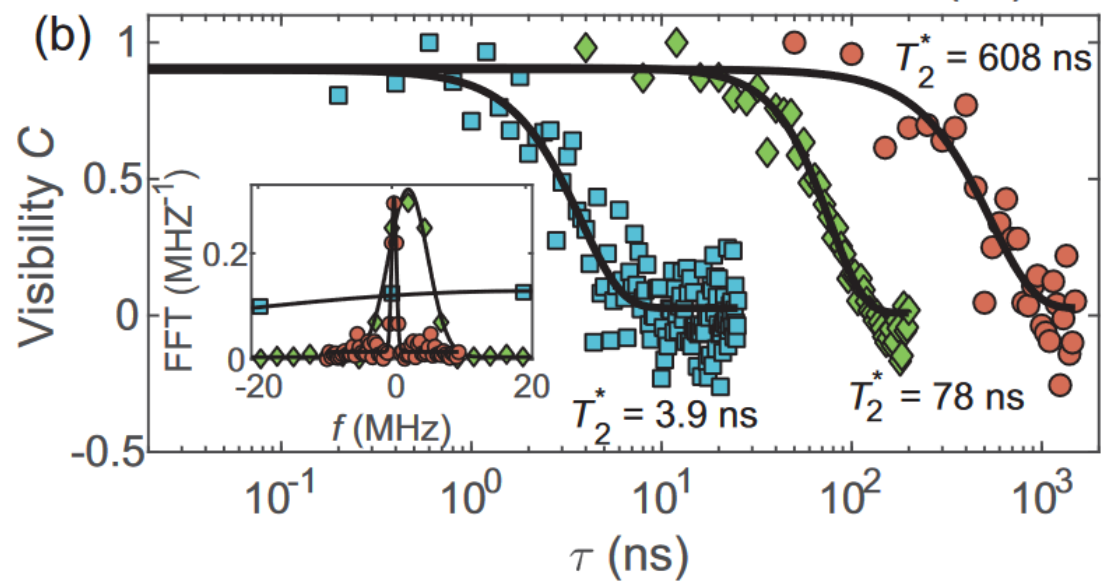
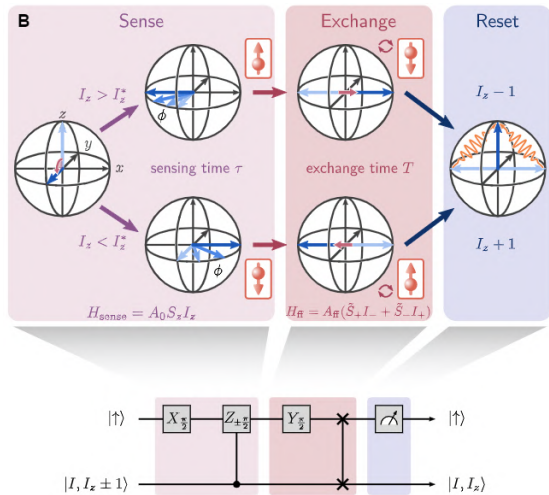


93% remote HOM visibility

Covre da Silva et al., Appl. Phys. Lett. **119**, 120502 (2021)

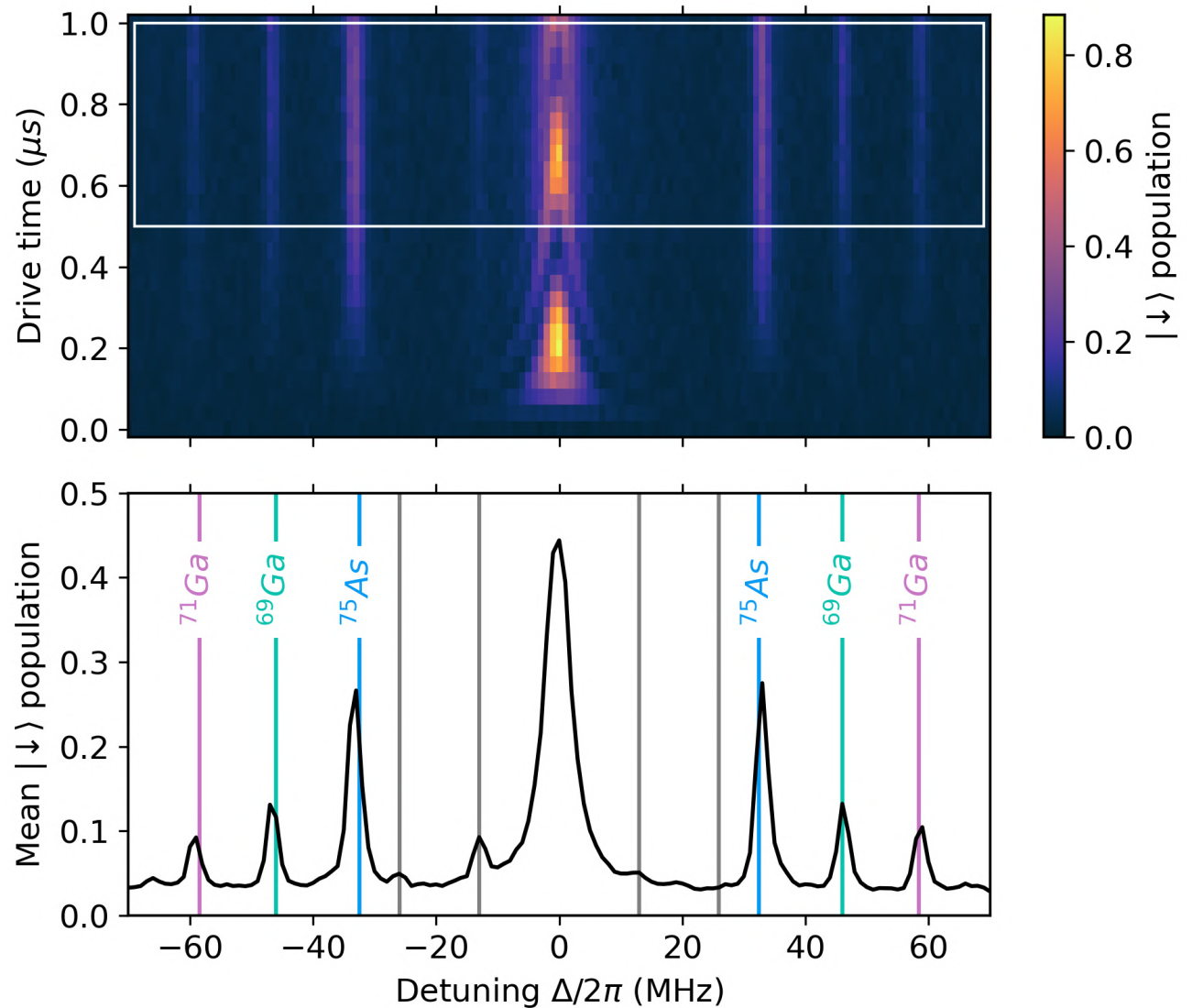
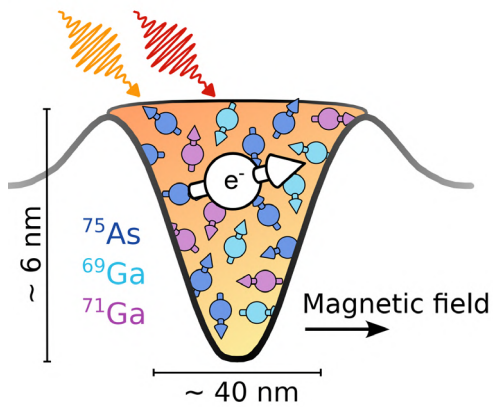
Zhai et al., Nature Nanotechnol. **17**, 829 (2022)

first evidence of optically cooled nuclei in GaAs QDs



150-fold improvement from 4 ns

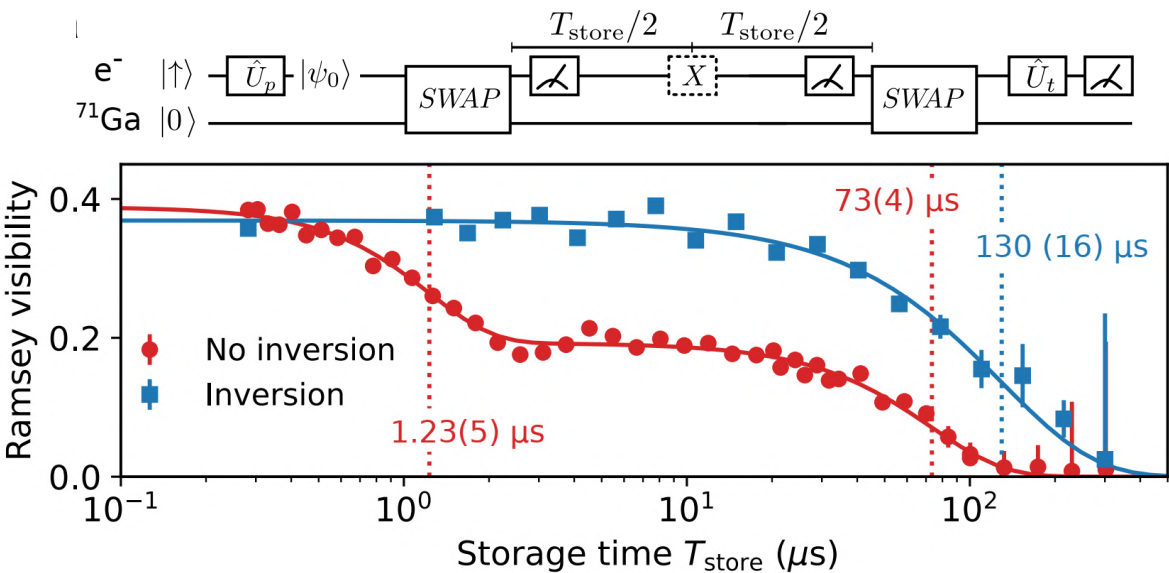
nuclear magnon spectrum for GaAs QD spins



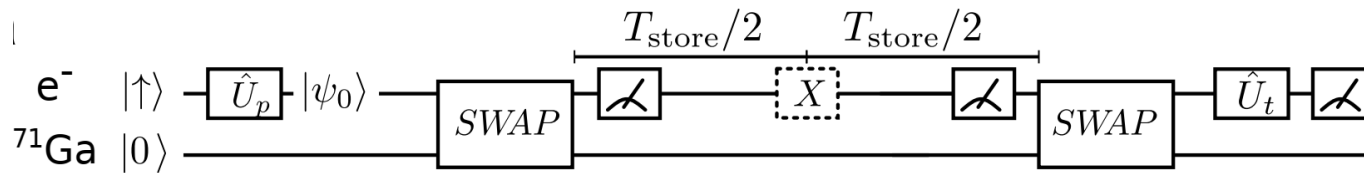
Long-Lived Memory for Mesoscopic Quantum Bits

J. M. Taylor, C. M. Marcus, and M. D. Lukin
Department of Physics, Harvard University, Cambridge, Massachusetts 02138, USA
(Received 17 January 2003; published 20 May 2003)

We describe a technique to create long-lived quantum memory for quantum bits in mesoscopic systems. Specifically we show that electronic spin coherence can be reversibly mapped onto the collective state of the surrounding nuclei. The coherent transfer can be efficient and fast and it can be used, when combined with standard resonance techniques, to reversibly store coherent superpositions on the time scale of seconds. This method can also allow for “engineering” entangled states of nuclear ensembles and efficiently manipulating the stored states. We investigate the feasibility of this method through a detailed analysis of the coherence properties of the system.

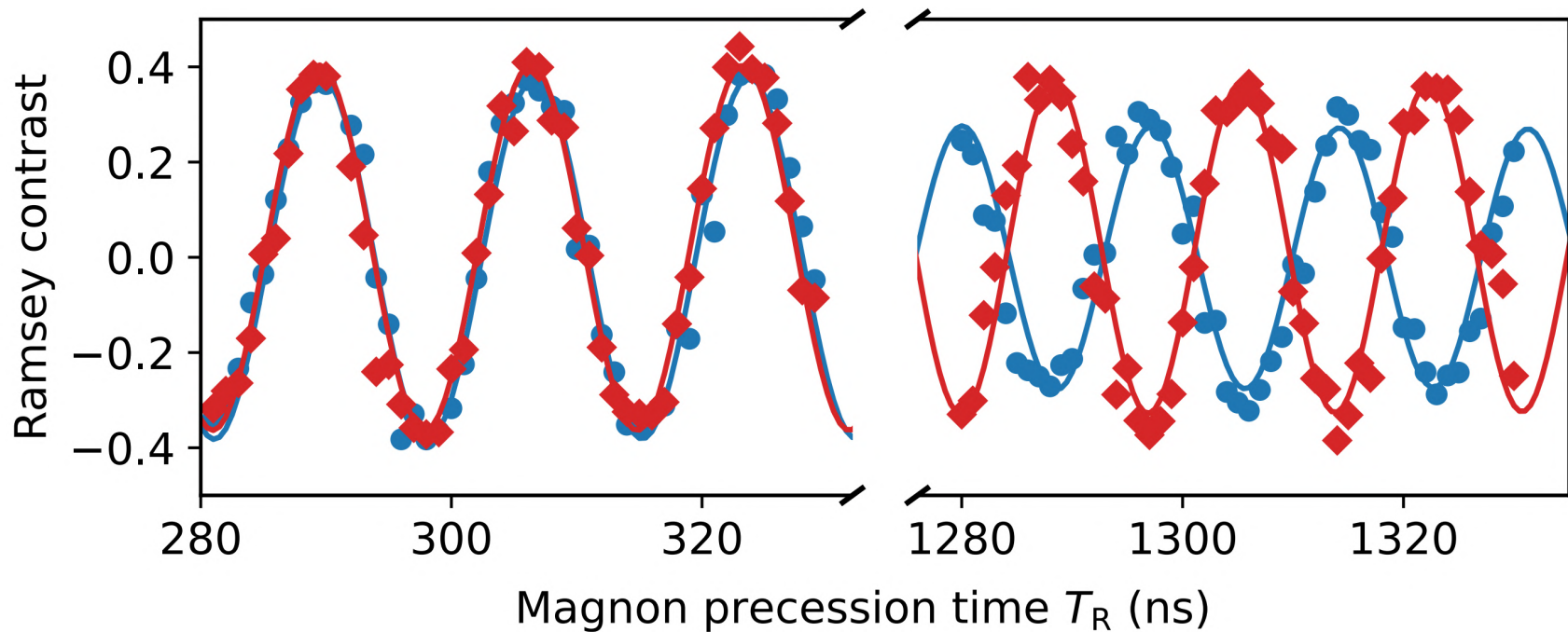


A nuclear magnon spin sensor (roles reversed...)

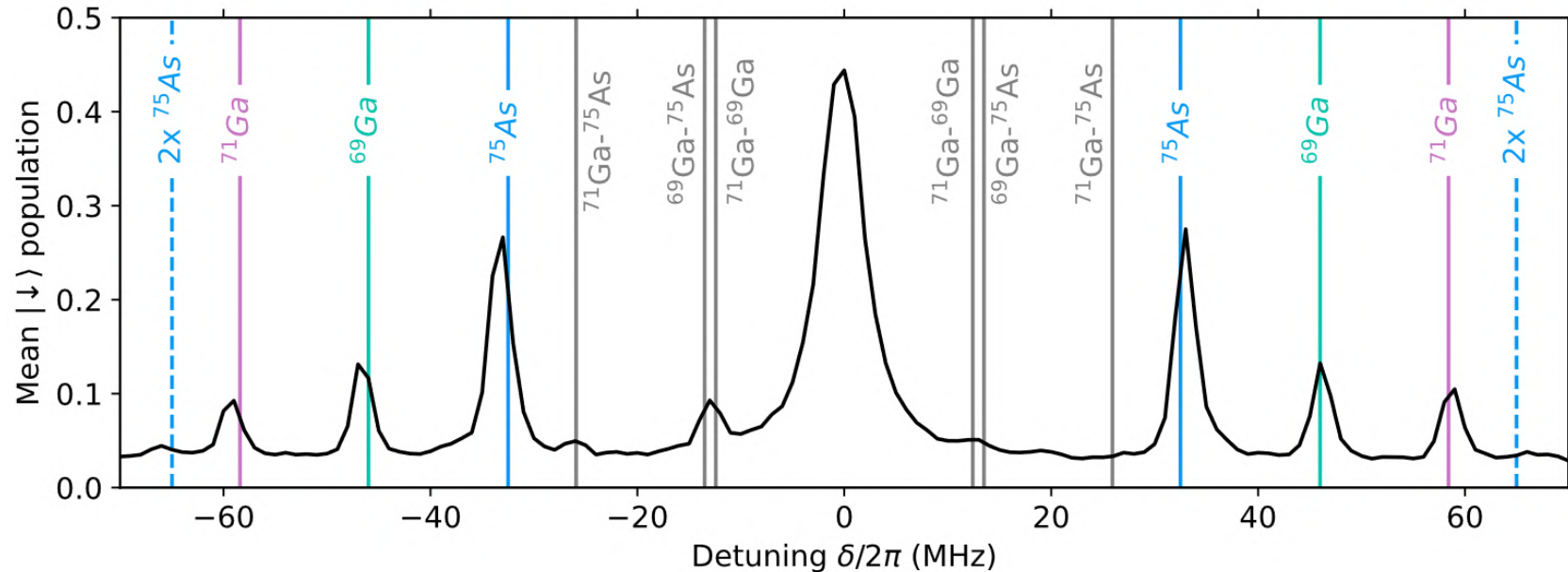


● $|\uparrow\rangle$ during precession

◆ $|\downarrow\rangle$ during precession



closer look at the magnon spectrum...3-body interactions



Three-body interactions at nuclear difference frequencies

A direct route to creating correlations

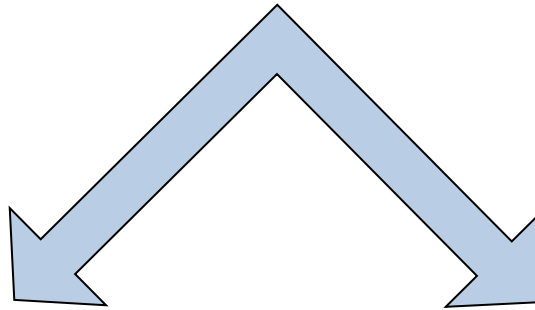
our storyline in the last decade (building on almost two decades)



a coherent ensemble...to play with

A long-lived coherent spin ensemble

with independently tuneable $\langle I_z \rangle$ and ΔI_z



the team (and friends)



QD team:

Martin Appel
Clara Bachorz
Jonny Bodey
Gabriel Ethier-Majcher
Dorian Gangloff
Claire Le Gall
Alex Ghorbal
Urs Haeusler
Lukas Huthmacher
Daniel Jackson
Constantin Lang
Clemens Matthiesen
Noah Shofer
Megan Stanley
Rob Stockill
Leon Zaporski
Christian Schrimpf
Isabel Parke
Yusuf Karli
...

and special thanks to:

Rastelli Group (GaAs QD growth – Linz)
National III-V Centre (InAs QD growth – Sheffield)

EPSRC (NQIT, QUES²T), EU (S³NANO, spinNANO, QLUSTER, QUDOT-TECH, Quantum Flagship, MEEDGARD), ERC, ONRG



UNIVERSITY OF
CAMBRIDGE

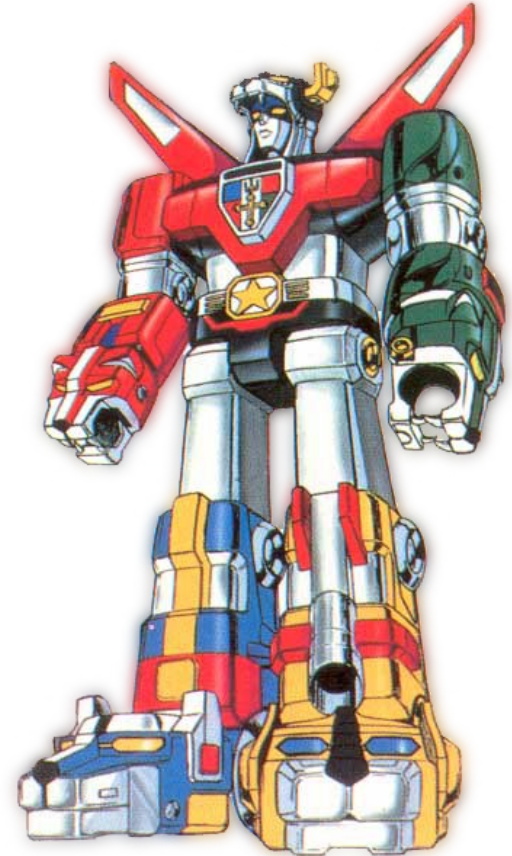
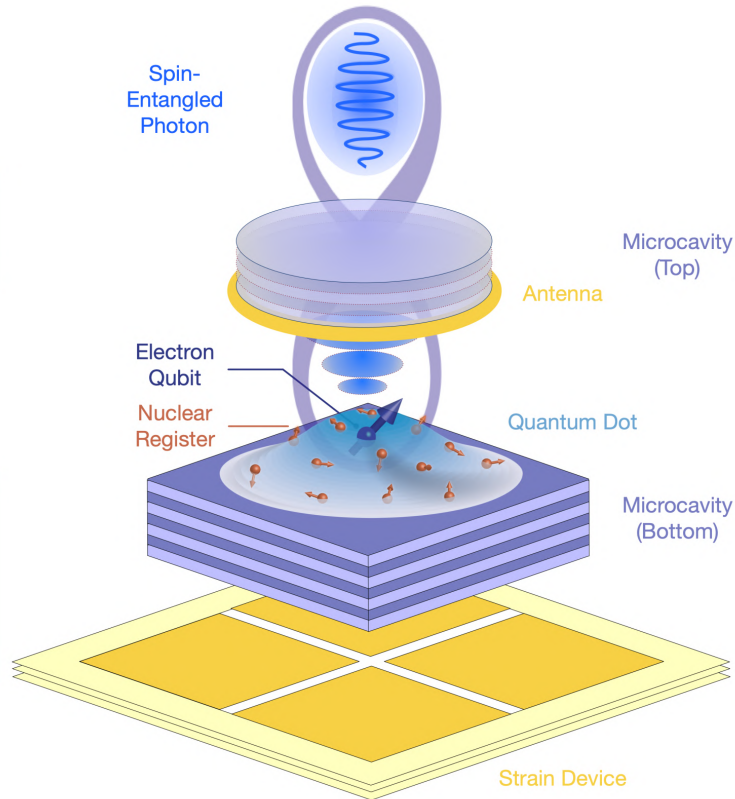


MEEDGARD – a ‘Voltron’ of quantum-dot devices

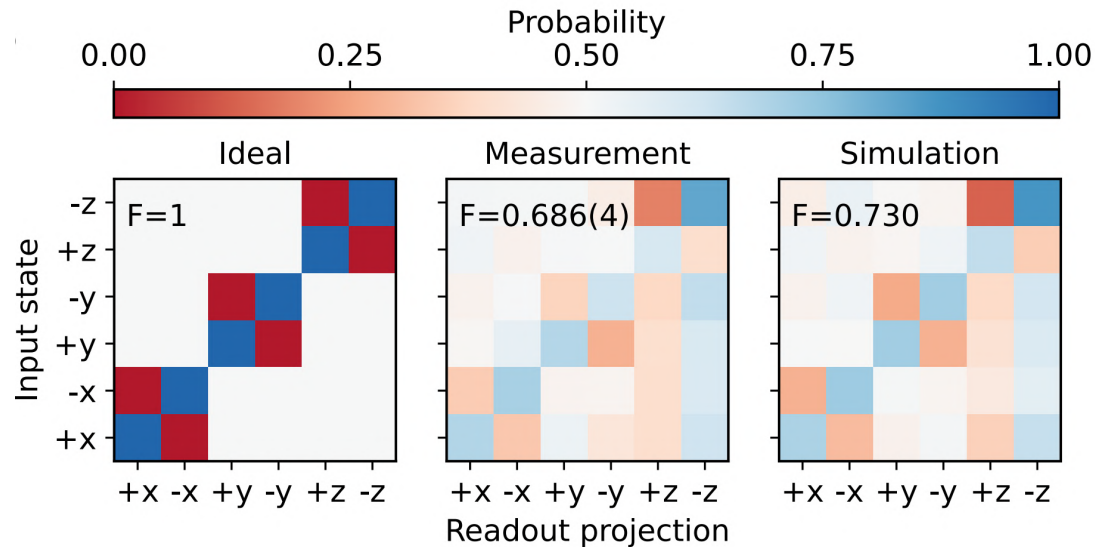
<https://meedgard.phy.cam.ac.uk>

Exciton-polaritons
(charged and neutral)

**Electro-nuclear
magnons**
(anisotropy or strain
activated)



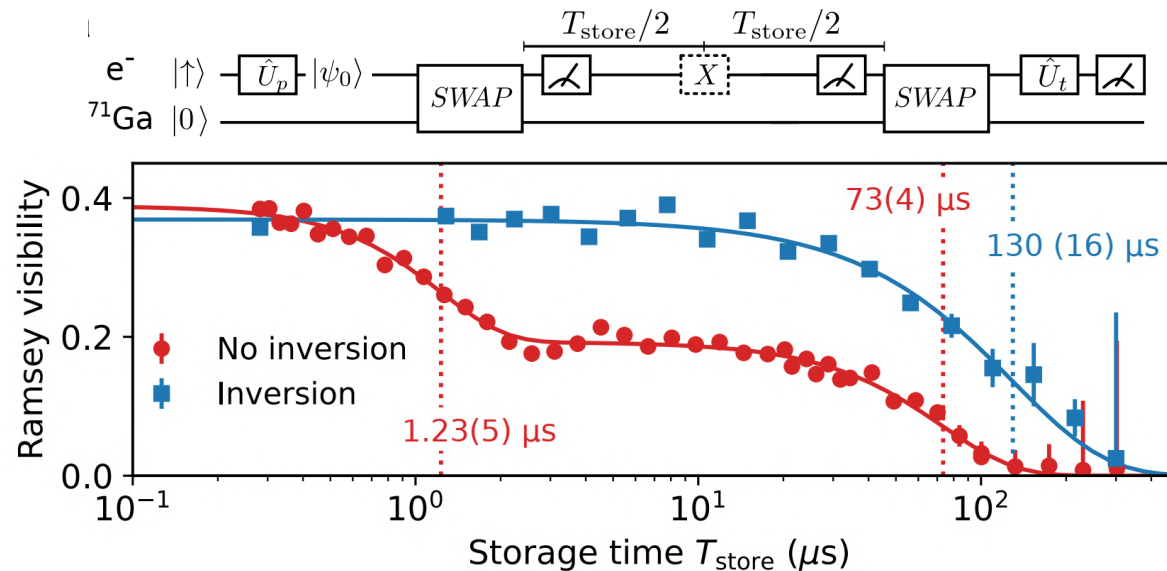
a many-body quantum register

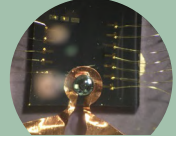


$$F = (1 + \langle C \rangle) / 2$$

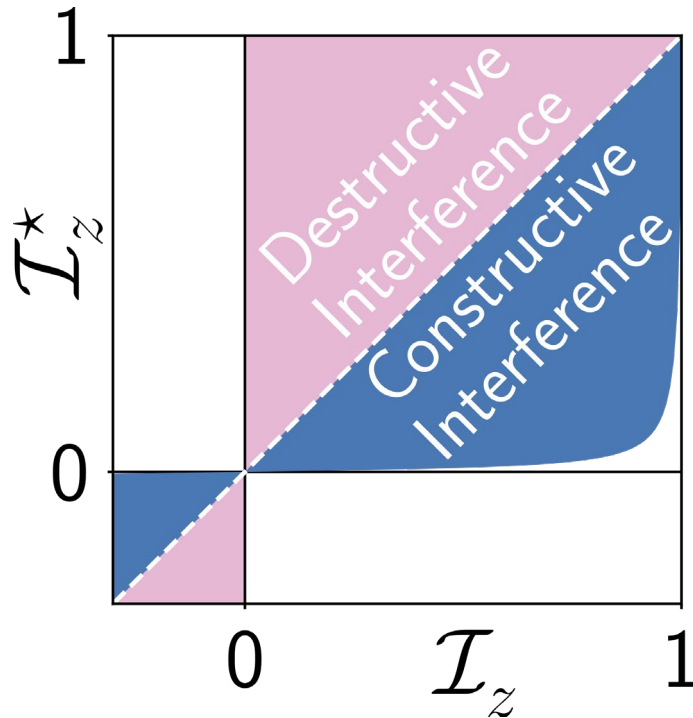
$$F_{\text{limit}} = 2/3$$

$$F_{\text{exp}} = 0.69$$





Compare $\mathcal{I}_z$ from Overhauser shift to $\mathcal{I}_z^*$ from sideband asymmetry

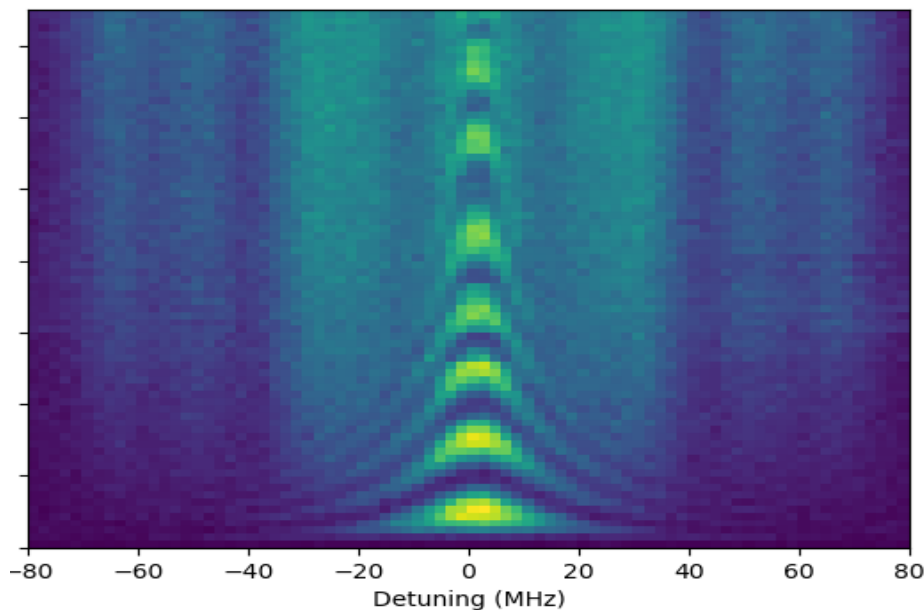
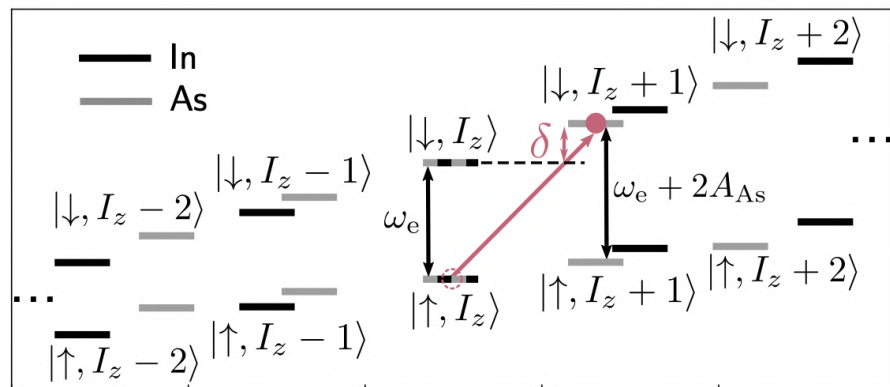


Christ et al., PRB **75**, 155324 (2007)

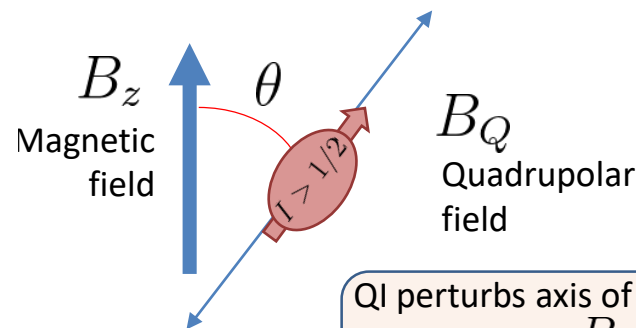
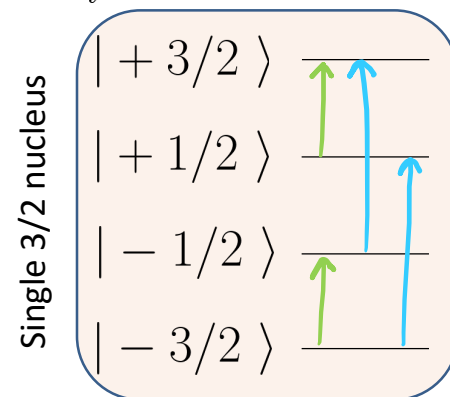
Taylor et al., PRL, 91, 246802 (2003)

D.A. Gangloff et al., *Nature Physics*, 17, 1247 (2021)

resolving nuclear species via magnons



$$H_{e-nuc}^{nc} = \sum_i A_1^{nc} (S_z (I_x^i I_z^i + I_z^i I_x^i)) + \sum_i A_2^{nc} (S_z (I_x^{i2} - I_y^{i2}))$$



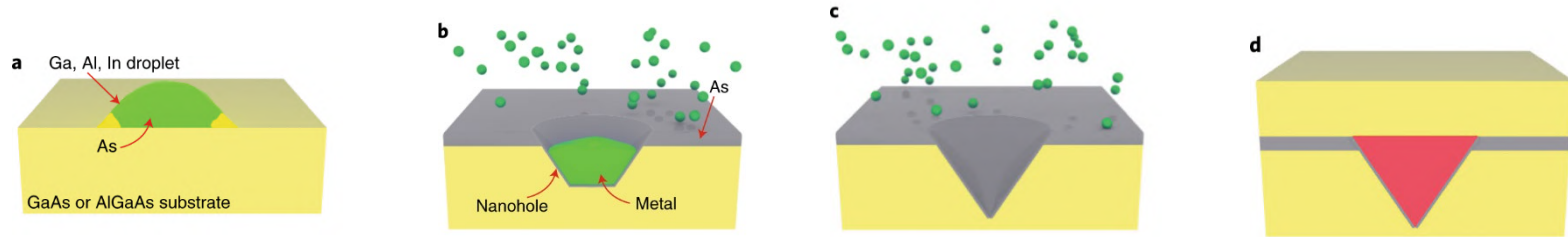
QI perturbs axis of the e-nuc HI

$$A_1^{nc} = A_z \frac{B_Q}{B_z} \sin(2\theta)$$

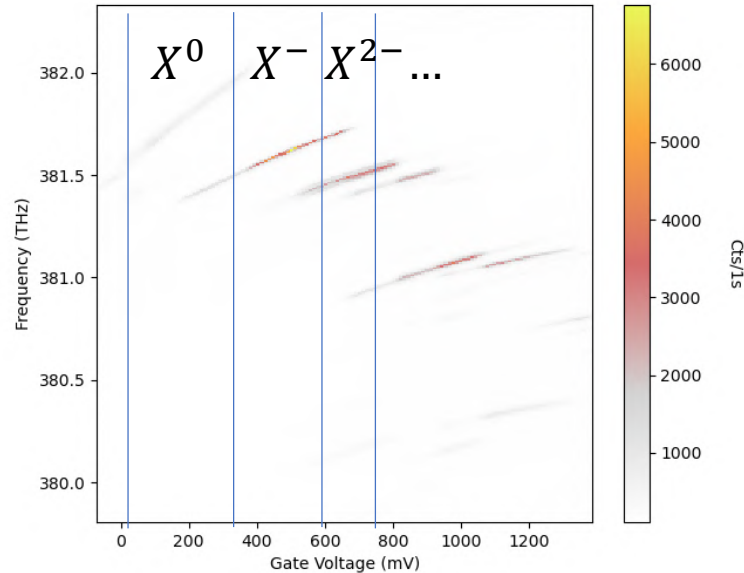


how to avoid the strain dispersion problem (avoid dispersion!)

Strain-free GaAs QDs

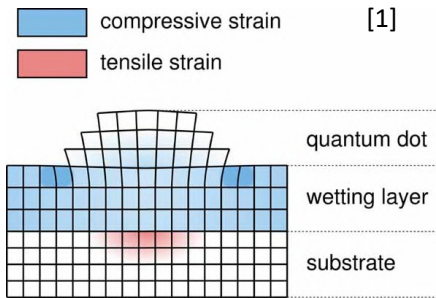


Gate controlled GaAs QDs



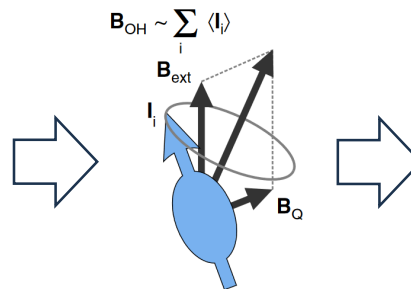
The problem with InGaAs

Stranski-Krastanow growth:

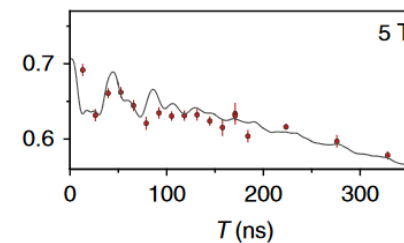


7% lattice mismatch

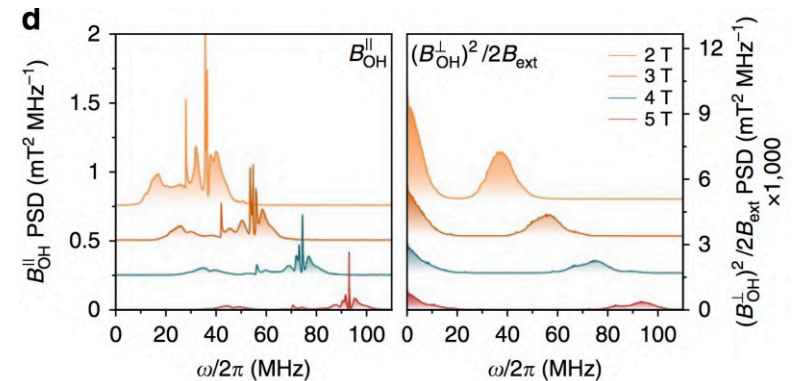
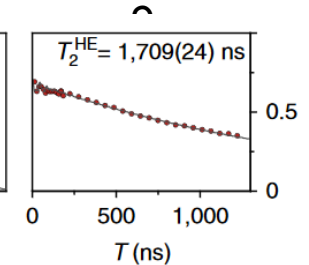
- Strain: **Non-collinear coupling**
- Strain dispersion: **5 MHz nuclear broadening**
- Broadening limits electron coherence, electro-nuclear coupling, NMR control, etc...



Spin echo [2]:



Limited spin
coherence: T_2



[1] Figge et al., Phys. Status Solidi B 248, 8 (2011)

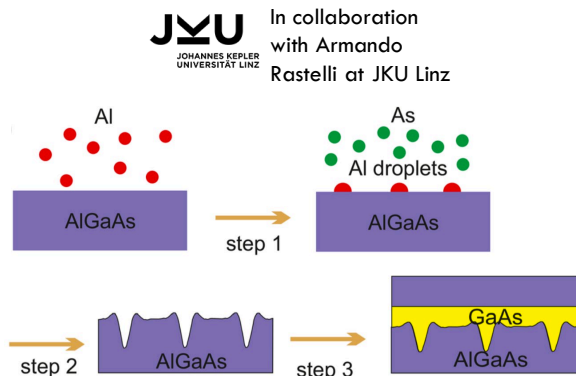
[2] Stockill et al, Nat. Commun., 7, 12745 (2016)

GaAs droplet quantum dots

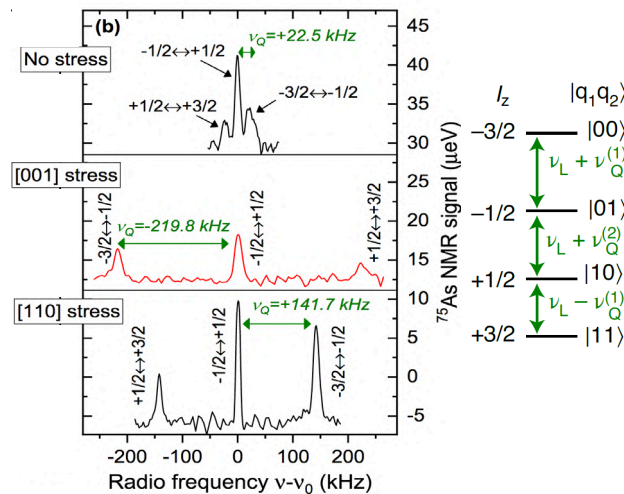
Droplet epitaxy growth[1]:

QD detected NMR on 75Ga

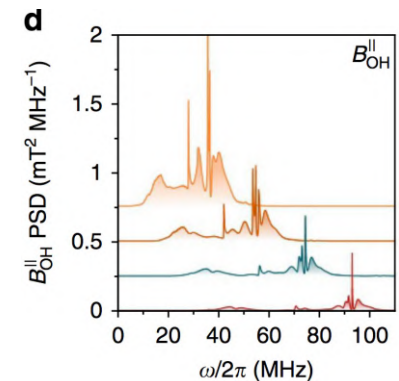
Comparison with InGaAs



Negligible difference in lattice constant



10-100 KHz broadening [2]



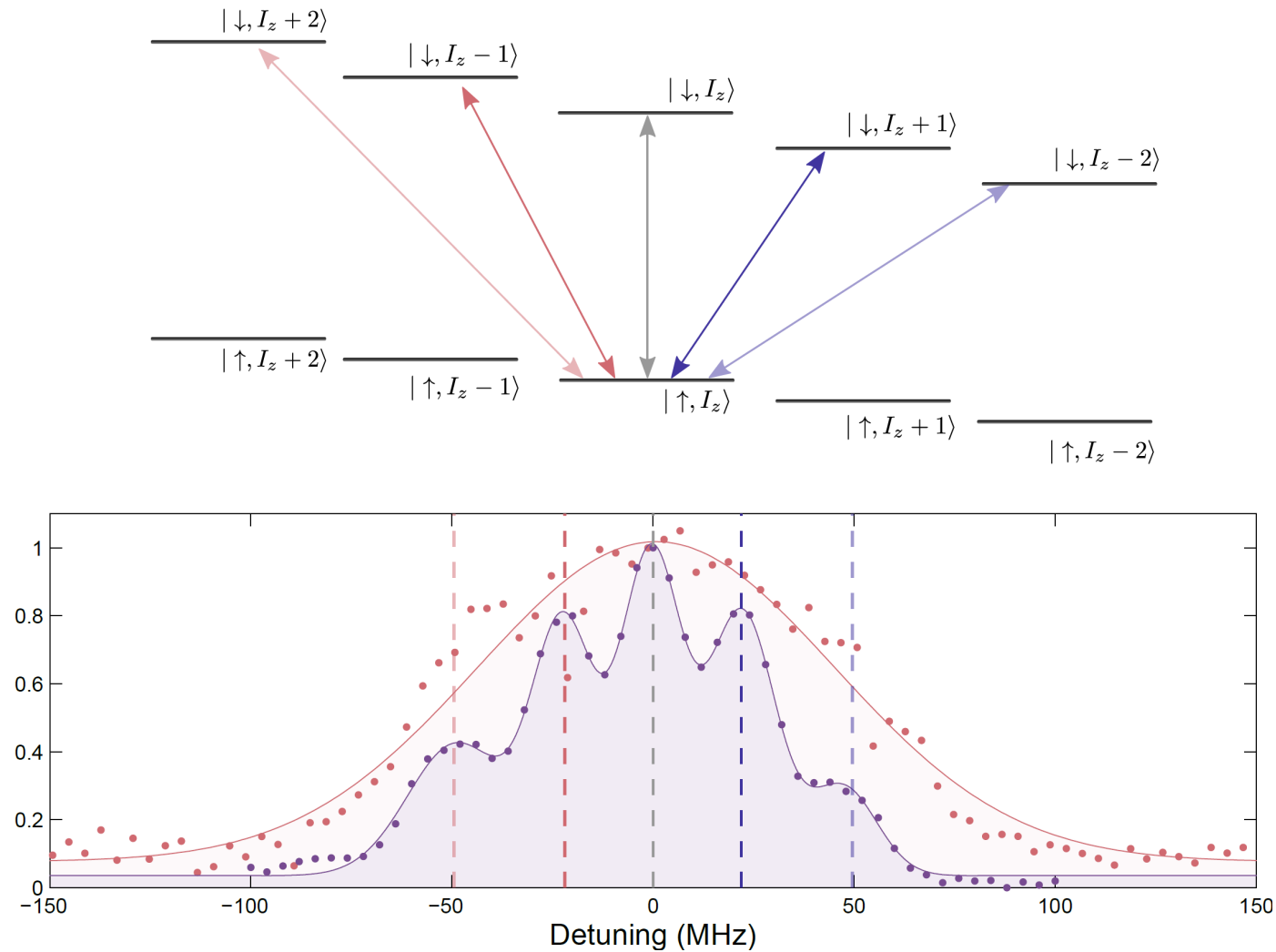
~ 10 MHz broadening

How often do you win two orders of magnitude?!?

[1] Covre da Silva et al., Appl. Phys. Lett. 119, 120502 (2021)

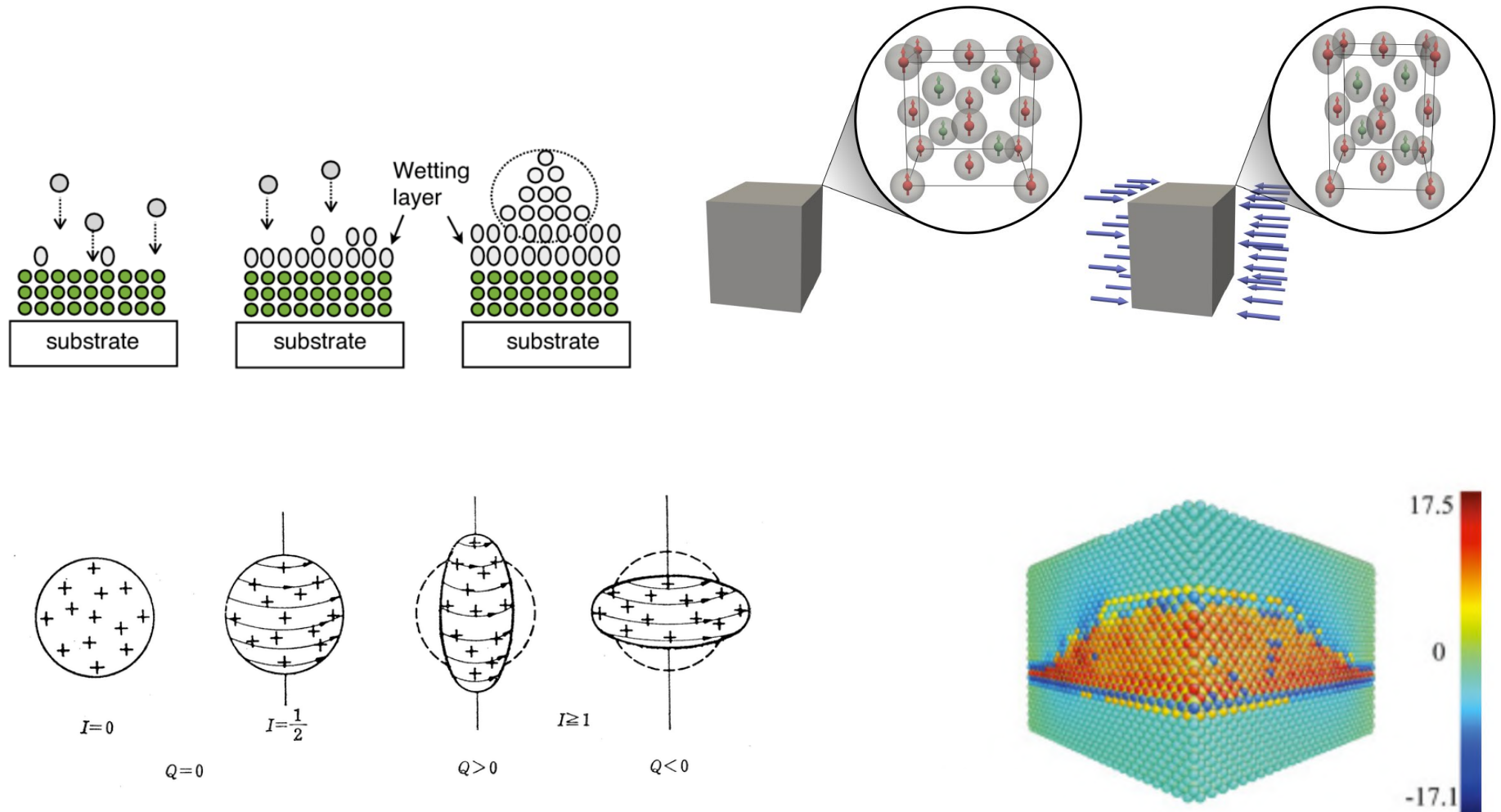
[2] Chekhovich et al, Phys. Rev. B 97, 235311 (2018)

probing the single-spin excitation spectrum of the nuclei

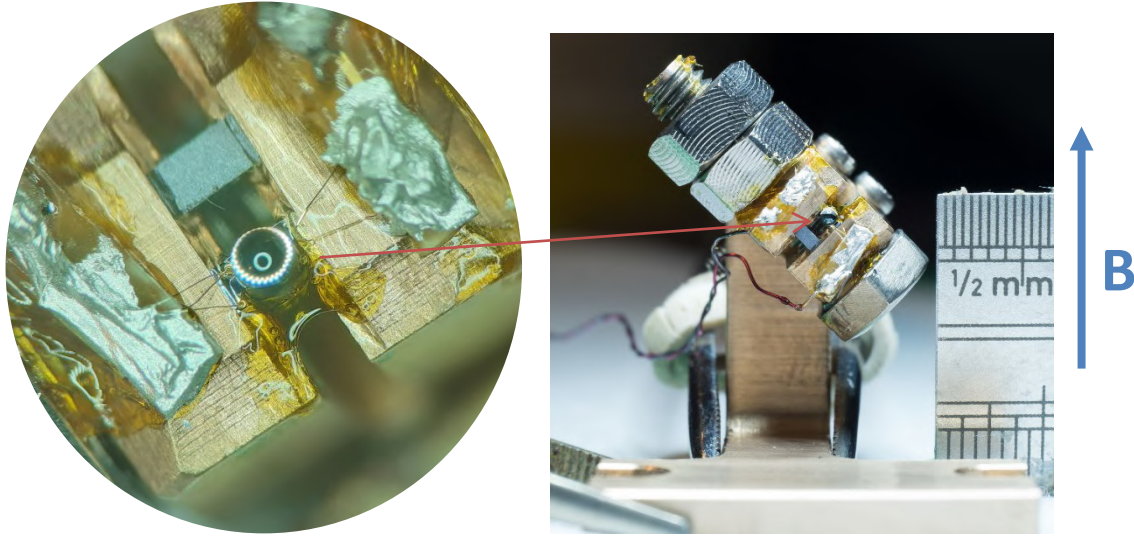


Gangloff *et al.*, Science 364, 62 (2019).

the role of strain on electro-nuclear interactions



first optical cooling of the nuclei in GaAs QDs

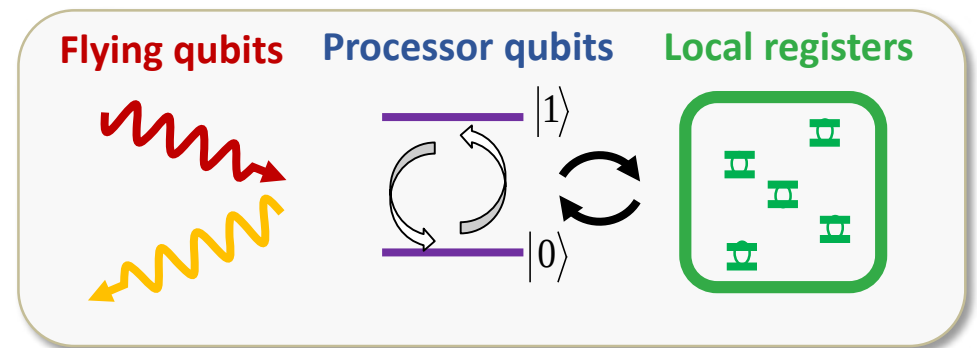
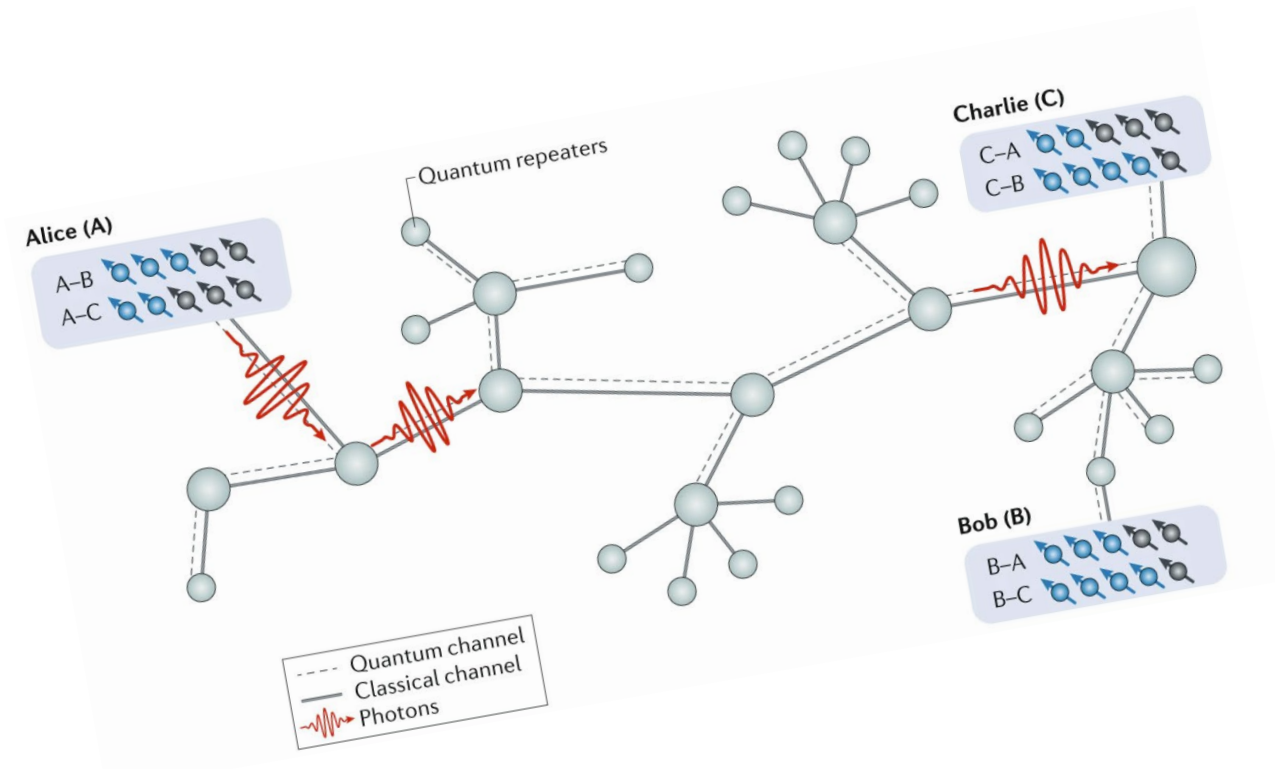


Attempt to induce homogeneous strain.

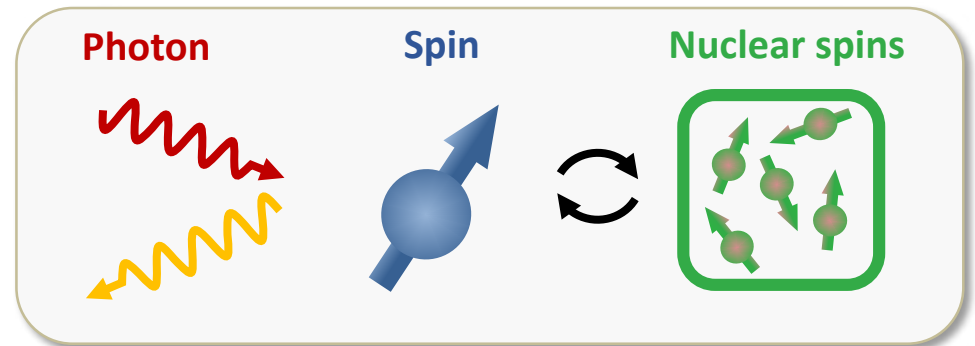
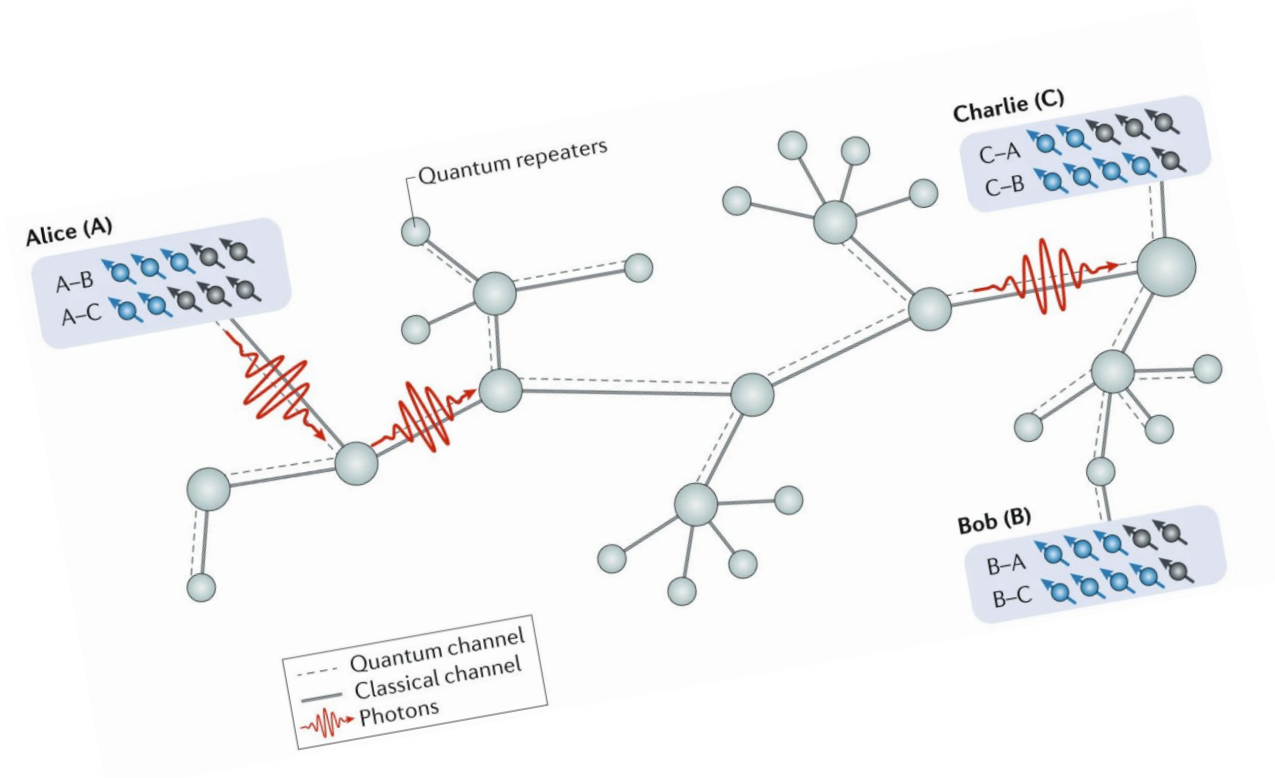
Up to 120 kHz quadrupolar shift

(many GaAs QD devices were harmed in the making of this research)

quantum nodes in a network



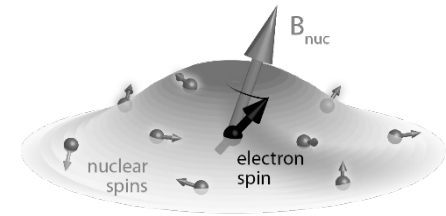
quantum nodes in a network



electronuclear interactions

Zeeman

$$\hat{H} = \omega_e \hat{S}_z + \sum_i \omega_i \hat{I}_z^{(i)}$$



Collinear hyperfine

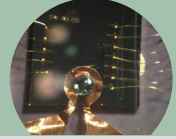
$$+ \hat{S}_z \sum_i a_{\parallel}^{(i)} \hat{I}_z^{(i)} \quad (\text{T2* limit})$$

Non-collinear hyperfine

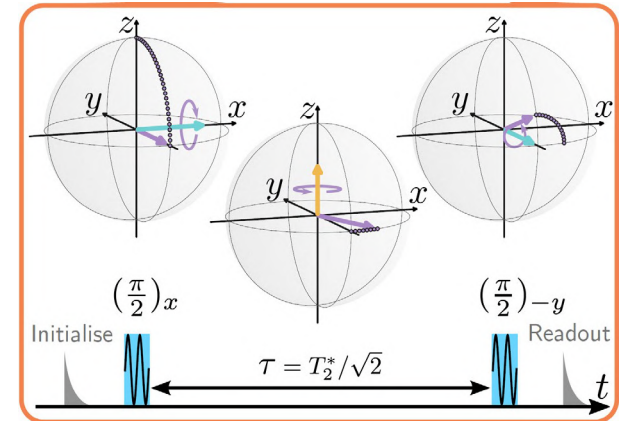
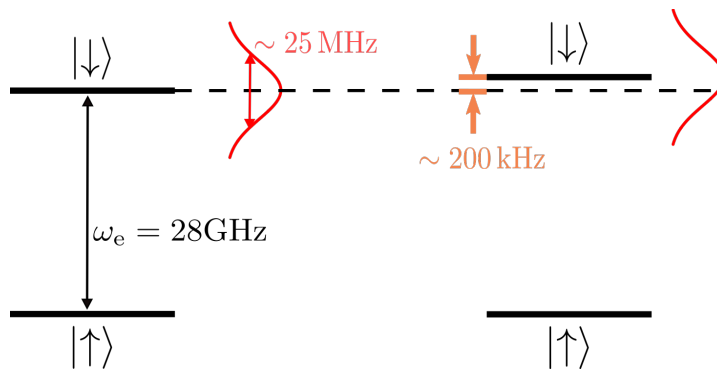
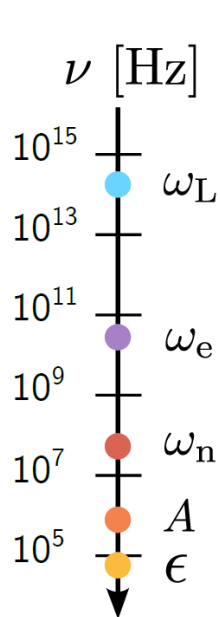
$$+ \hat{S}_z \sum_i a_{\perp}^{(i)} \hat{I}_x^{(i)} = \hat{S}_z \sum_i a_{\perp}^{(i)} (\hat{I}_+^{(i)} + \hat{I}_-^{(i)})/2$$

Three-body interaction

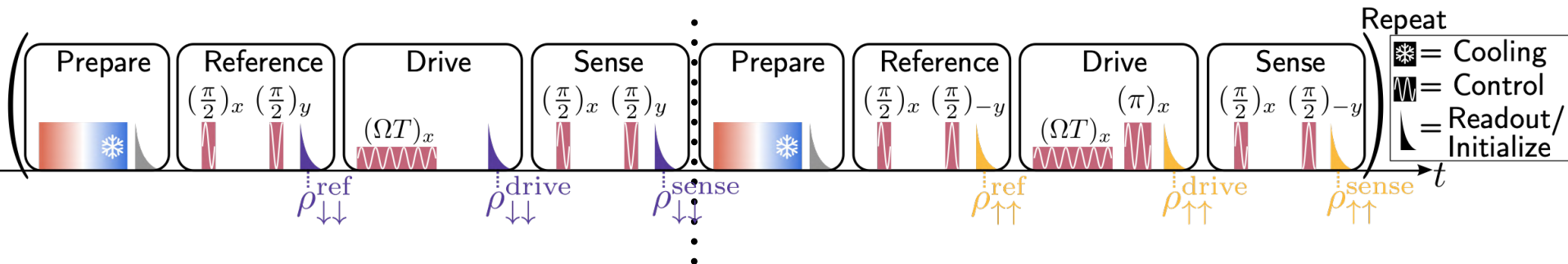
$$+ \hat{S}_z \sum_{i,j} \frac{a_i a_j}{2\omega_e} \left(\hat{I}_x^{(i)} \hat{I}_x^{(j)} + \hat{I}_y^{(i)} \hat{I}_y^{(j)} \right) = \hat{S}_z \sum_{i,j} \frac{a_i a_j}{4\omega_e} \left(\hat{I}_+^{(i)} \hat{I}_-^{(j)} + \hat{I}_-^{(i)} \hat{I}_+^{(j)} \right)$$



Sensing: Full experimental sequence

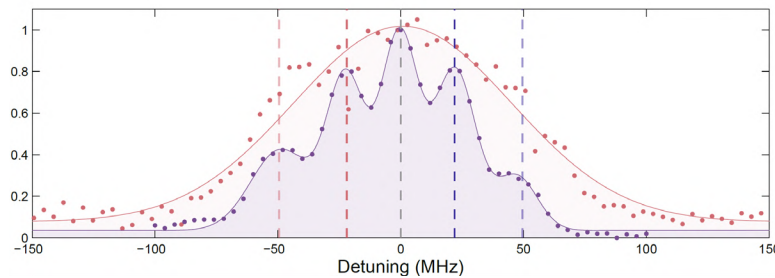
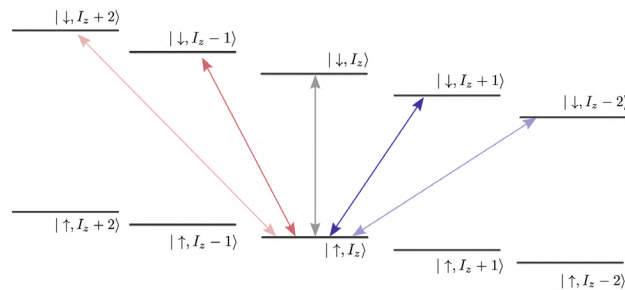


Frequency shift detected via Ramsey Interferometry
Electron spin becomes a sensor with 1.9 ppm sensitivity

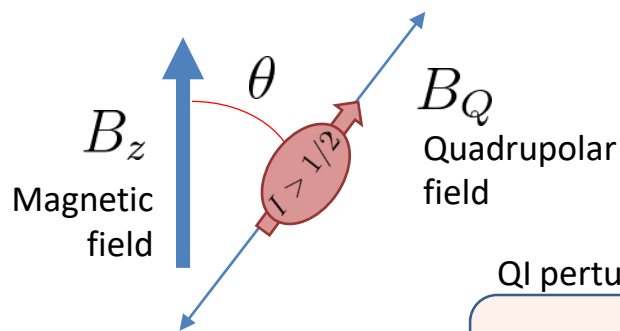


$$\Delta\omega_D = \Delta\omega_{\text{sense}} - \Delta\omega_{\text{ref}}$$

strain-enabled non-collinear hyperfine interaction



$$H_{\text{e-nuc}}^{nc} = \sum_i A_1^{nc} (S_z (\underbrace{I_x^i I_z^i + I_z^i I_x^i}_{\text{green}})) + \sum_i A_2^{nc} (S_z (\underbrace{I_x^i{}^2 - I_y^i{}^2}_{\text{blue}}))$$



QI perturbs axis of the e-nuc HI

$$A_1^{nc} = A_z \frac{B_Q}{B_z} \sin(2\theta)$$

Single 3/2 nucleus

