Magnetic strong coupling in a spin-photon system

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Collaborators

- Graduate students: Lei Chen ($\rightarrow$Cornell), Nick Groll ($\rightarrow$Argonne)
- Sylvain Bertaina (former postdoc, now $\rightarrow$ CNRS-Marseille, France)

Chemistry: Prof. N. Dalal (FSU), Prof. G. Christou (UF)

The EPR group at NHMFL: Prof. S. Hill, Dr. J. van Tol

SQUIDs: Dr. W. Wernsdorfer (CNRS-Grenoble)

Applied Superconductivity Center, NHMFL-FSU: Prof. Alex Gurevich

Dept of Physics, University of Tokyo: Prof. Seiji Miyashita

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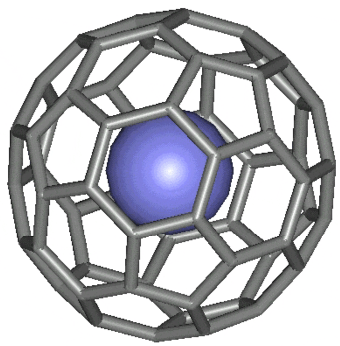
Outline

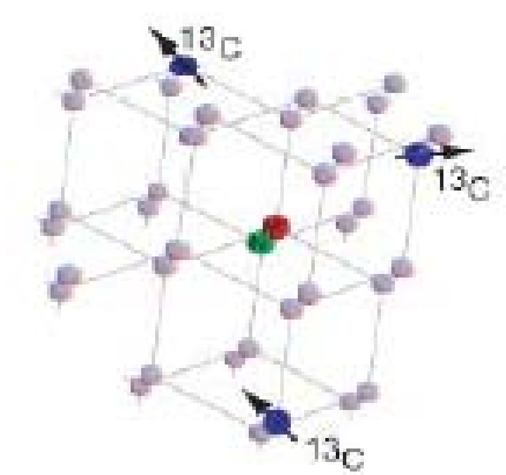
- Spin systems (molecular magnets, diluted spins)
- Single & multi-photon manipulation of diluted spins
- Spin-photon coupling
- On-chip superconducting cavity in high-fields
- Conclusions

Quantum spins, towards QC at room T



$N@C_{60}$ **S = 3/2**

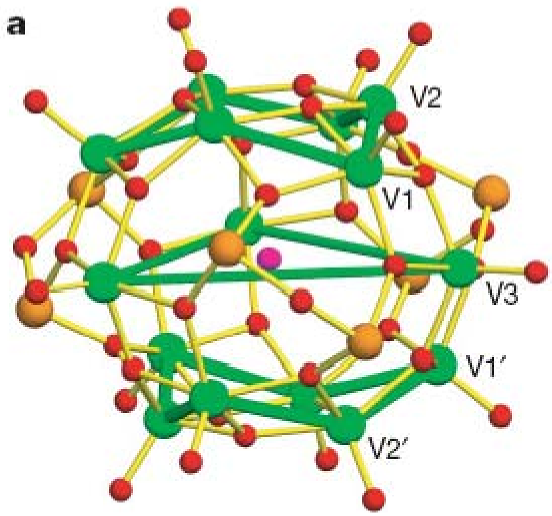
- neutral, non-metallic N: *S=3/2*
- pulsed EPR on powder
- coherence at room temperature (Rabi oscillations)



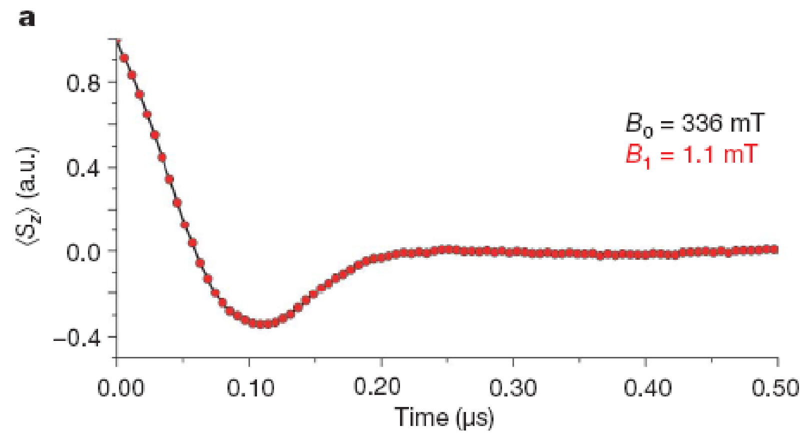
- Nitrogen vacancies color centers in ultra-pure diamond
- Axial symmetry, *S=1*, large crystal field
- Optical detection on *SINGLE SPINS*
- room temperature Rabi oscillations

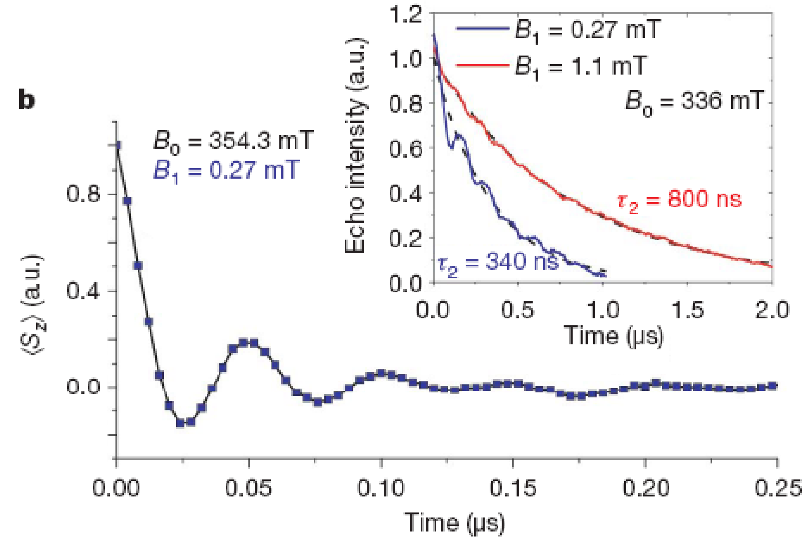
Spin coherence in molecular magnets

- Coherent oscillations in V_{15} molecular magnet S=1/2



S. Bertaina, S. Gambarelli, T. Mitra, B. Tsukerblat, A. Mueller & B. Barbara
"Quantum oscillations in a molecular magnet"
Nature **453**, 203 (2008)





Cr_7Ni,Mn: A. Ardavan *et al, Phys. Rev. Lett.* **98**, 220501 (2007)
Fe_8 molecule: S. Takahashi *et al*, *Phys. Rev. Lett.* **102**, 087603 (2009)
Fe_4 molecule: Schlegel *et al*, *Phys. Rev. Lett.* **101**, 147203 (2008)

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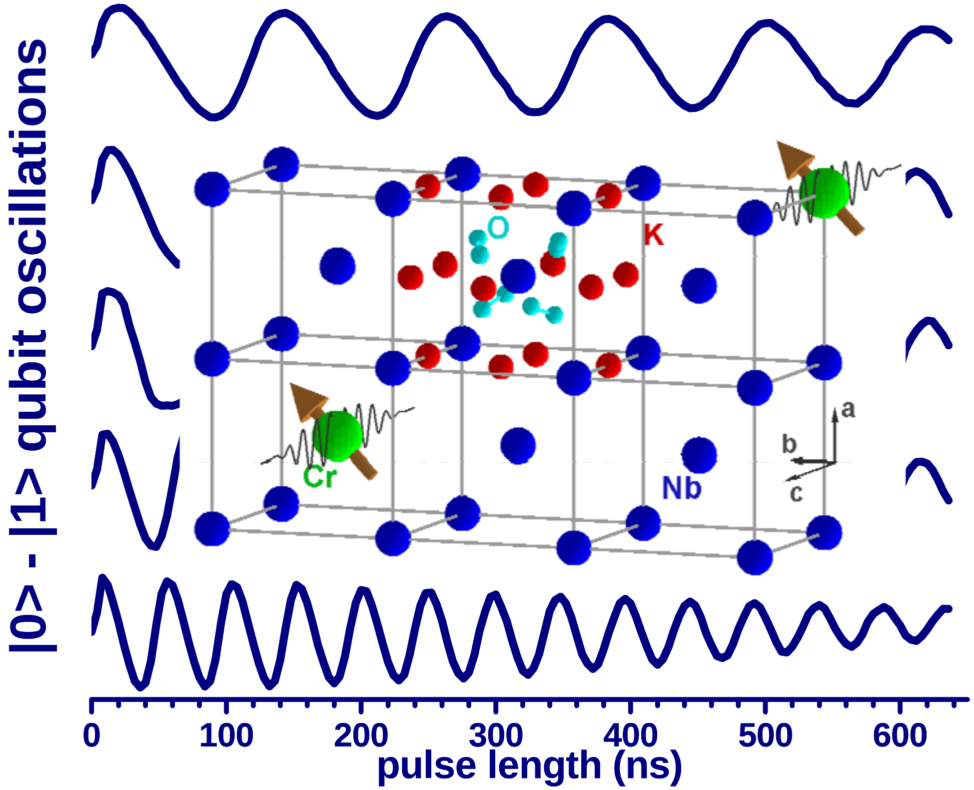
Diluted spin systems: Cr (*S=1/2*), Mn (*S=5/2*)

Cr:K_3NbO_8
S = 1/2



S. Nellutla *et al*, *PRL* **99**, 137601 (2007)

- Coherent manipulation of two or multi-level systems
- Compared with quantum dots: *spins interact with a very small number of nuclei* (less decoherence)
- *symmetry vs. decoherence*

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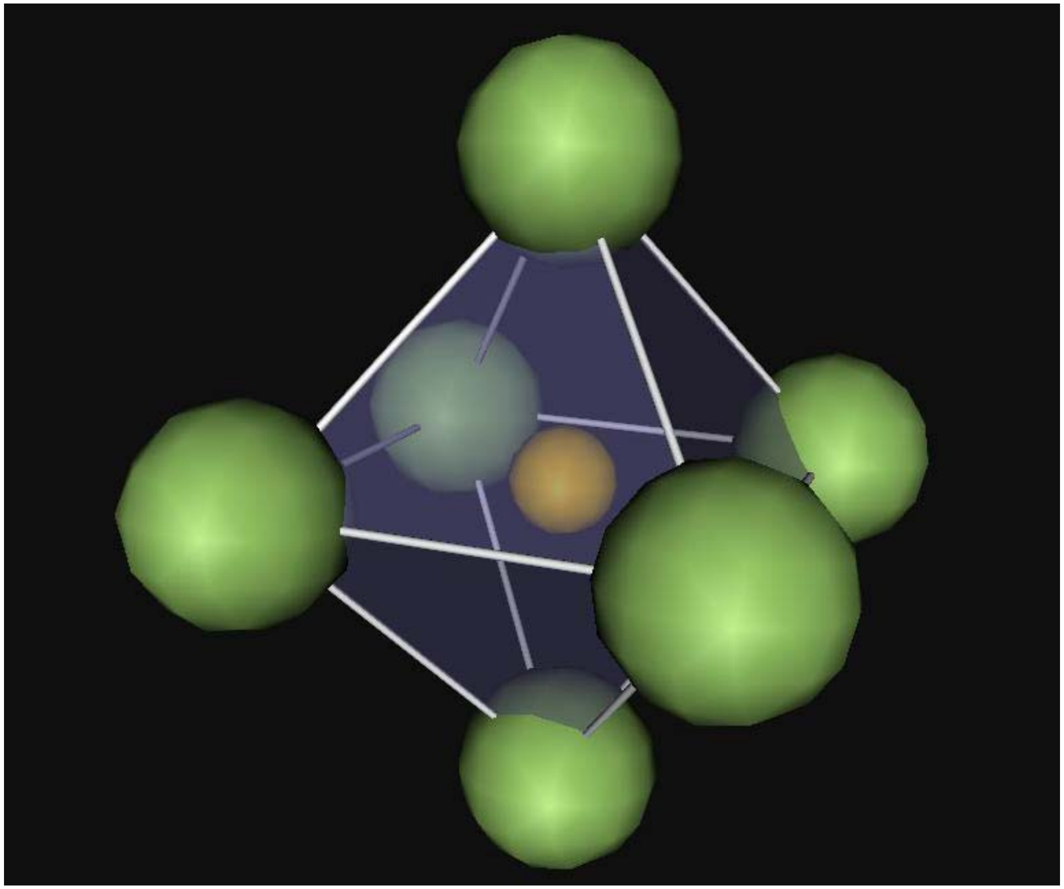
Spin coherence with Mn spins

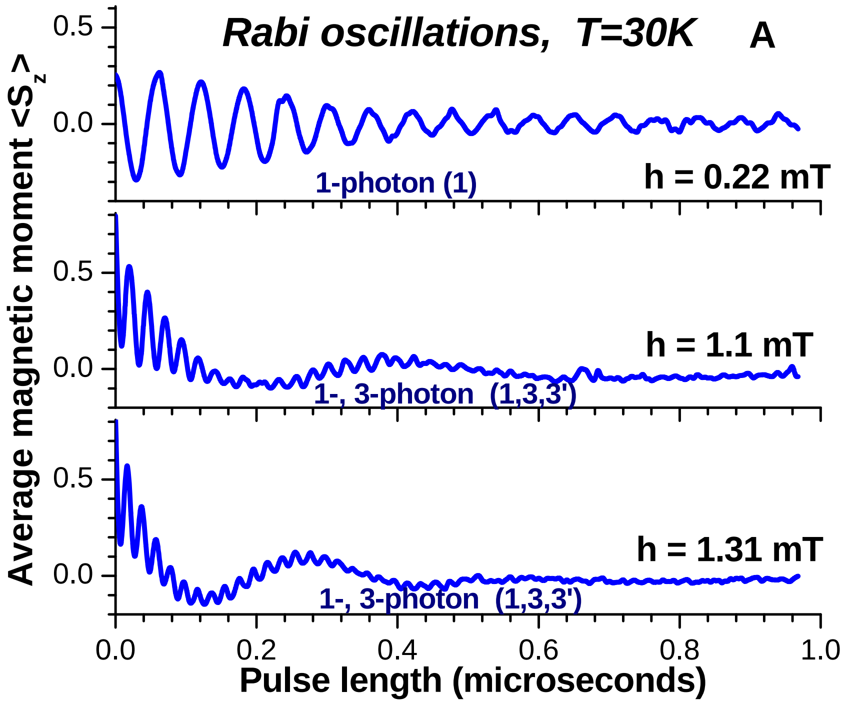
- Mn diluted in MgO single crystal
- Spin Hamiltonian

$$H = a/6\,[S_x^4+S_y^4+S_z^4- S(S+1)(3S^2-1)/5] +g\mu_B \vec{H}_0\cdot\vec{S}-A\vec{S}\cdot\vec{I}$$

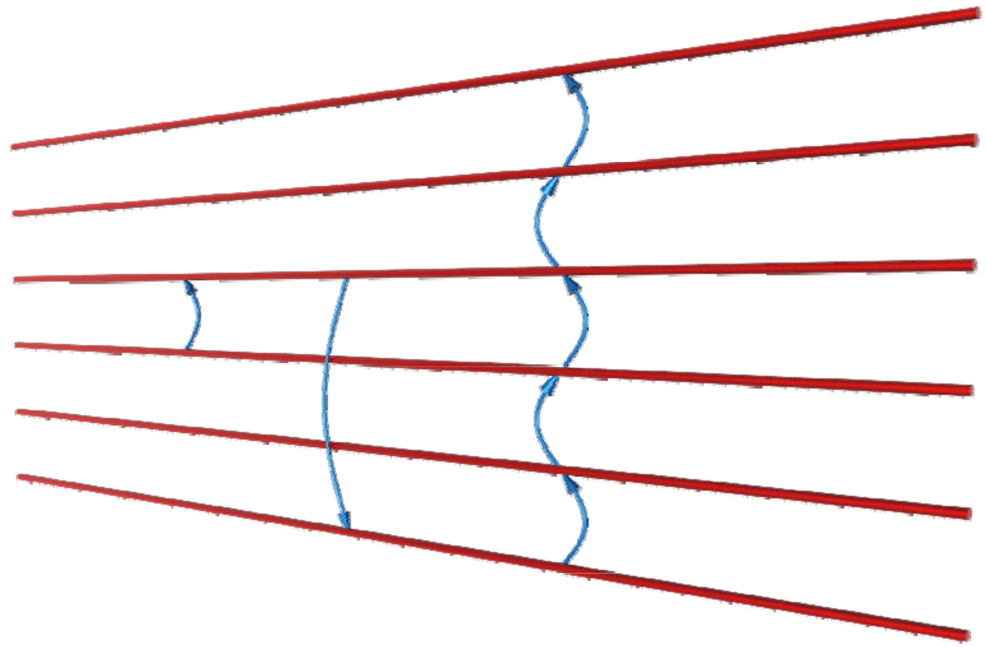
g = 2.0025, a = 55.7 MHz, A = 244 MHz, S = I = 5/2



Multi-photon/multi-level spin control



S. Bertaina et al, *Phys. Rev. Lett.* **102**, 050501 (2009)



Rabi oscillations showing $\langle S_z \rangle$ as a function of MW pulse length for different MW powers.

Single or multi-photon coherent drive

Pseudo-harmonic

H_0 || (1,0.8,1), m_I=-3/2

Non-harmonic ↔ quasi-harmonic

Rabi frequency between two consecutive levels, in a multi-level system:

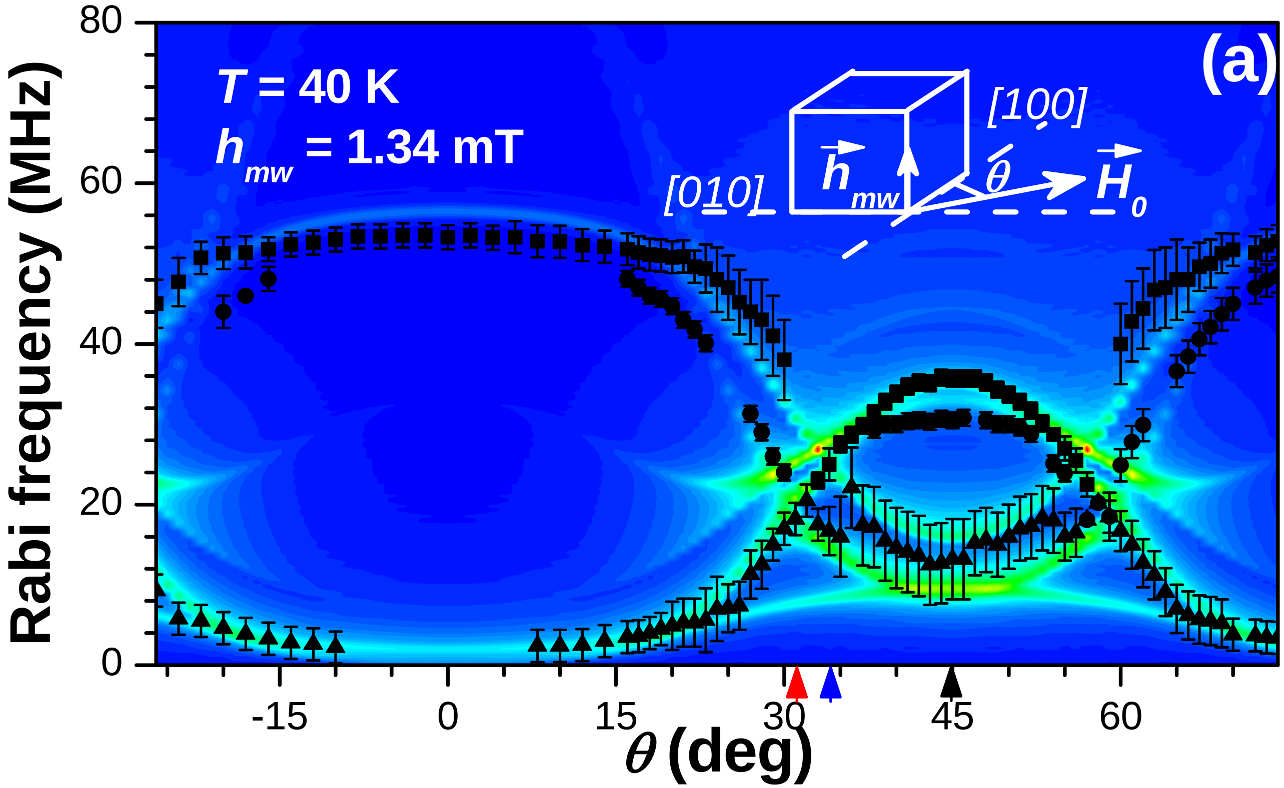
$hF_R^1 = 1/2\ g\mu_B h_{mw} \sqrt{S(S+1)-S_z(S_z+1)}$

$S_z = -1/2 \leftrightarrow 1/2$, for θ around 0°

Rabi frequency in a two level system:

$hF_R^{comp} = 1/2\ g\mu_B h_{mw}$

F_R^1/F_R^{comp} = 53.5 MHz / 18.07 MHz =2.96

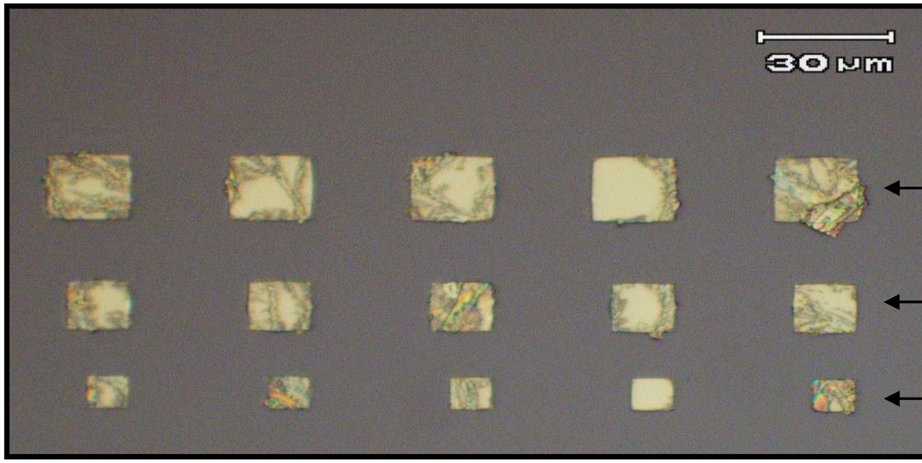


At θ=θ^{comp} : *non-harmonic* → *harmonic* multi-level system

Towards on-chip applications

- work done by grad students L. Chen, N. Groll

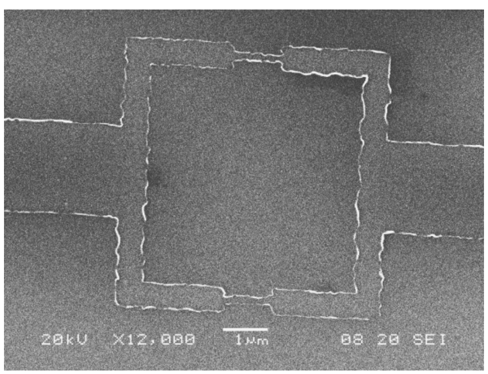
N. Groll et al, *J. Appl. Phys.* **106** (4), 046106 (2009)



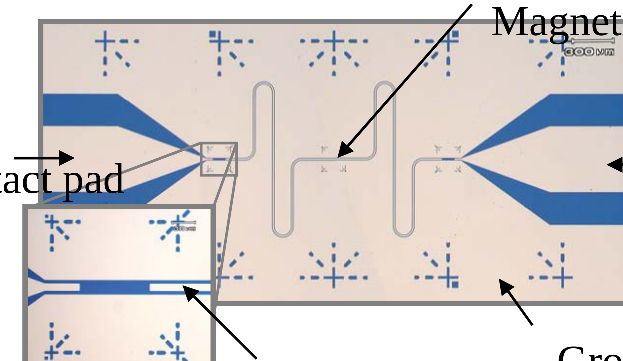
25 micron squares

15 micron squares

10 micron squares



SQUID, L. Chen



Magnetic molecule deposition region

Contact pad

Contact pad

Ground plane

Centerline Nb conductor

On-chip cavity, N. Groll

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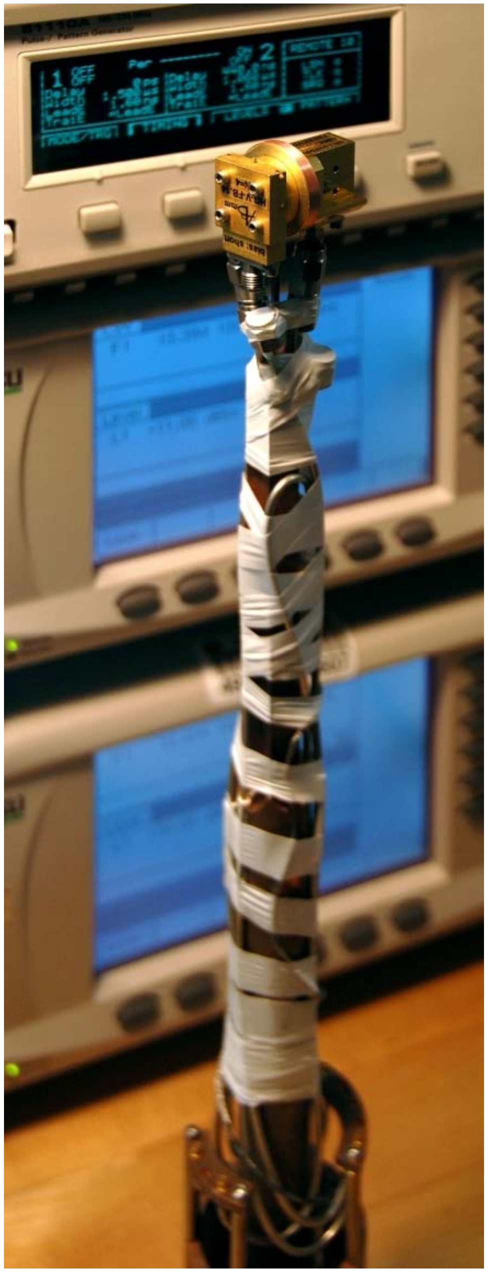
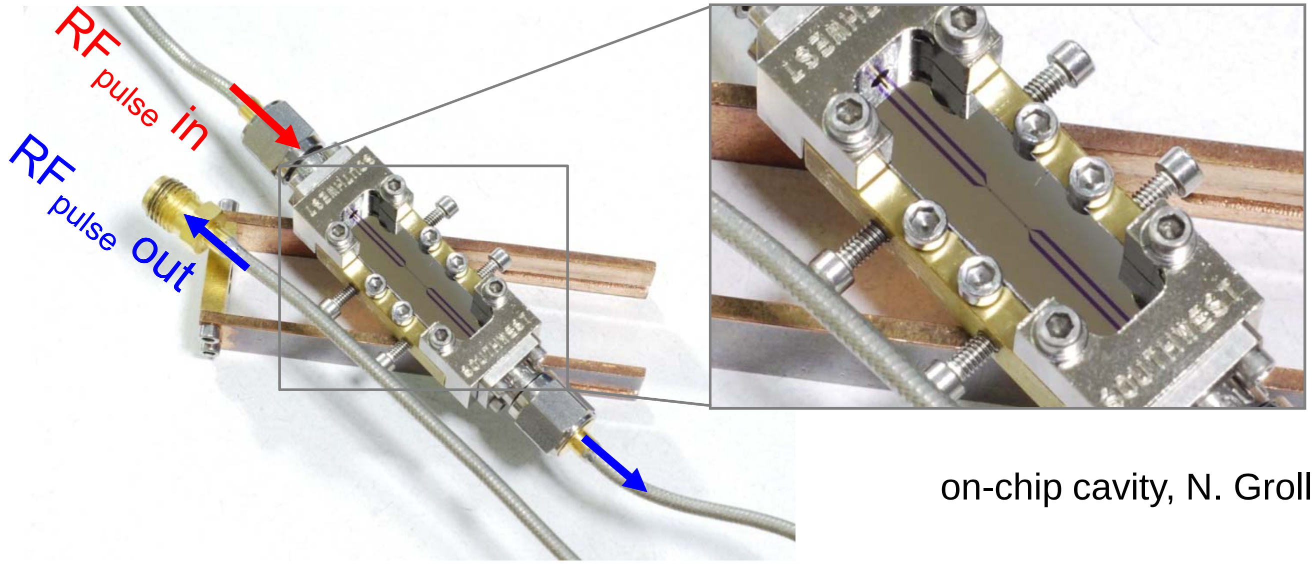
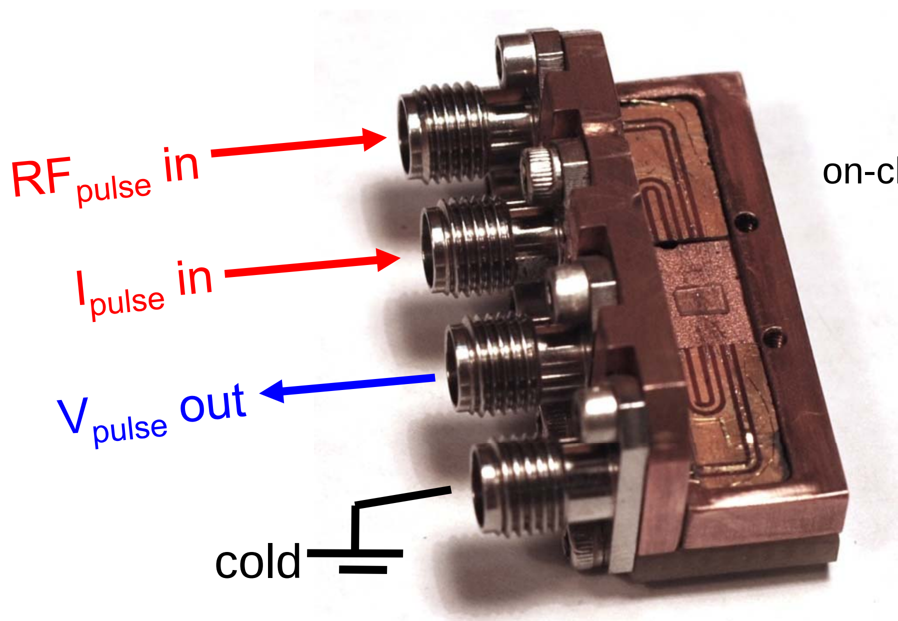
Probe head and exchangeable holders





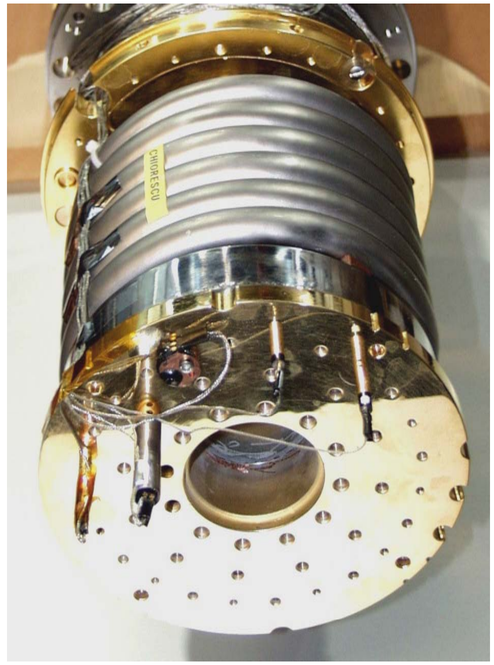
on-chip cavity, N. Groll

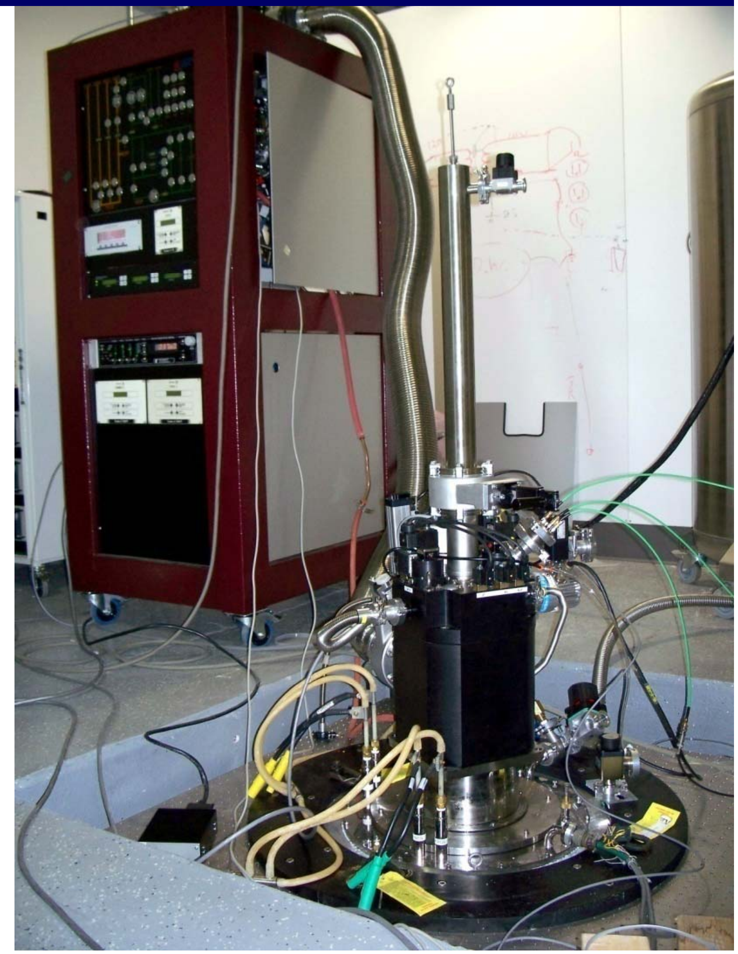


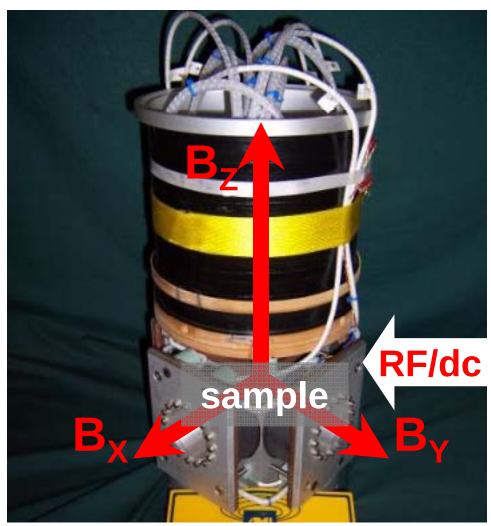
on-chip SQUID, L. Chen

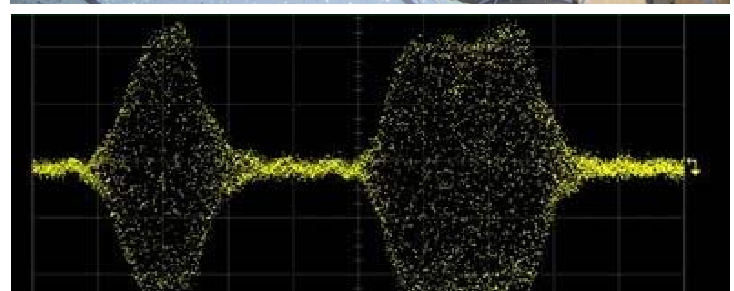
Experimental Setup

- 3-axis magnet, 2 inches bore: 7T on Z, 1T on X & Y
- dilution refrigerator, top loader
  cooling power 5mW @ 10mK
- non-magnetic vibration isolators
- microwave equipments:
  fast scope, CW, pulses
- fast current pulses:
  30ps rise/fall time
- arbitrary shaped I pulses





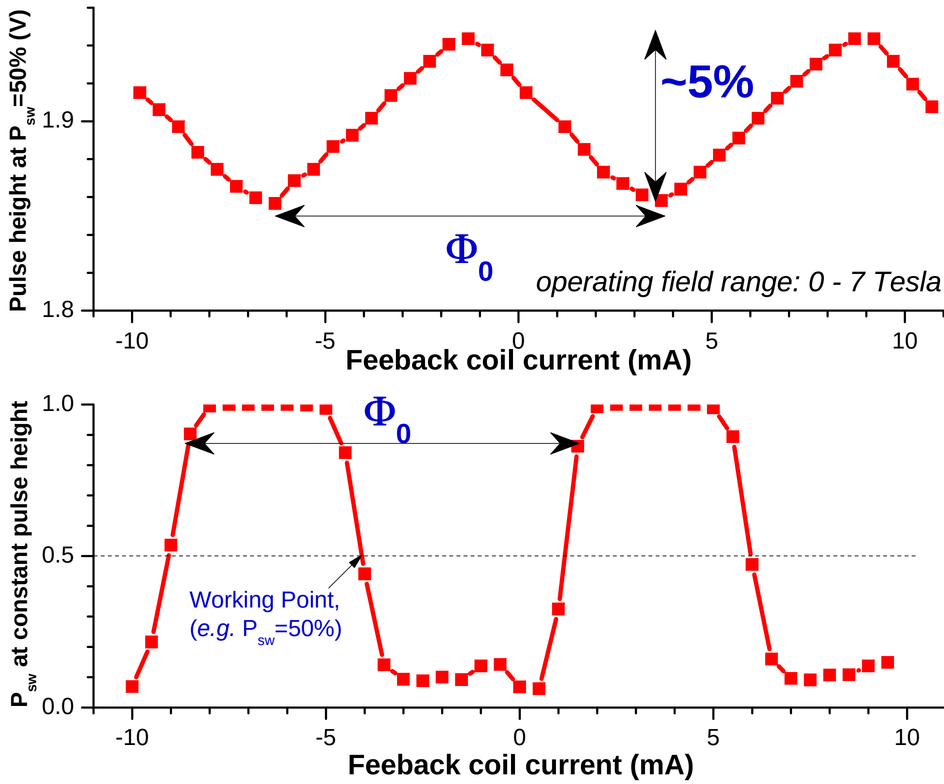


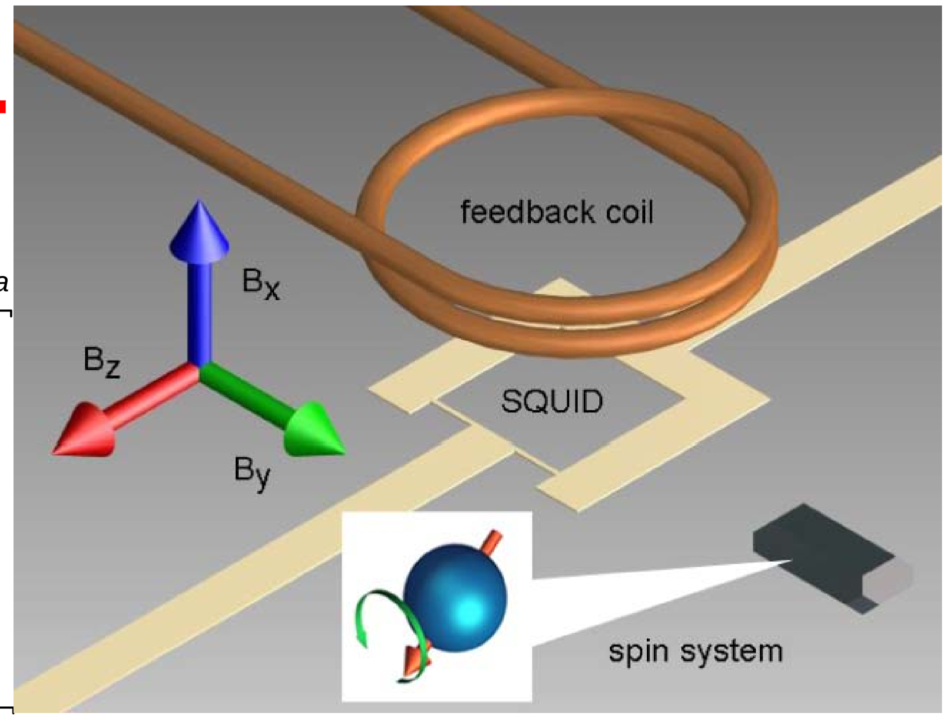


RF pulses: t_{rise}~100ps, frequency: up to 50GHz

SQUID setup

- The variable magnetic flux generated by the studied sample is compensated by the current sent into a feedback coil atop of the SQUID.





L. Chen, W. Wernsdorfer, C. Lampropoulos, G. Christou, I.C., *Nanotechnology* **21**, 405504 (2010)

Featured on: http://nanotechweb.org/cws/article/lab/44029

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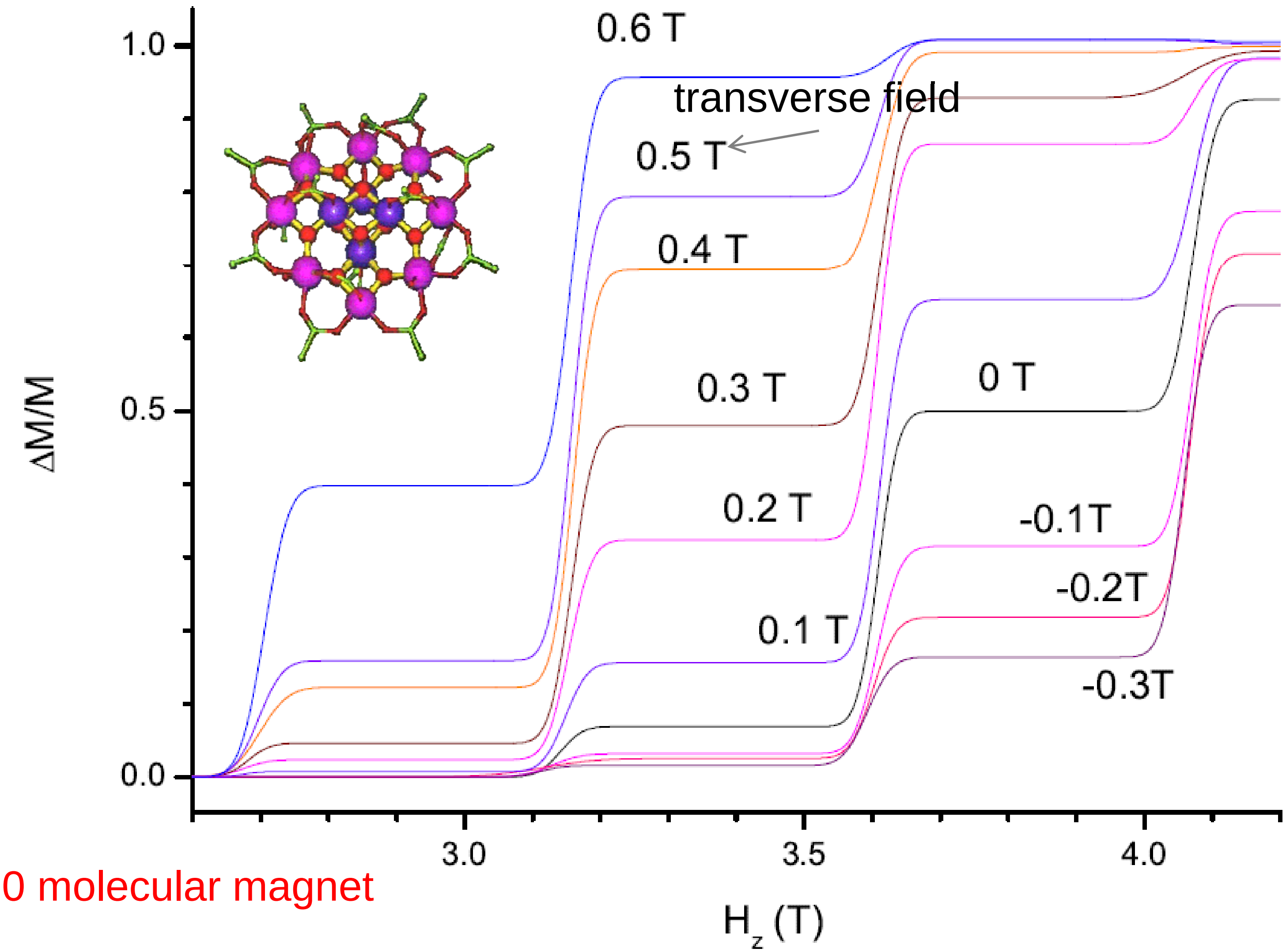
SQUIDs in high fields

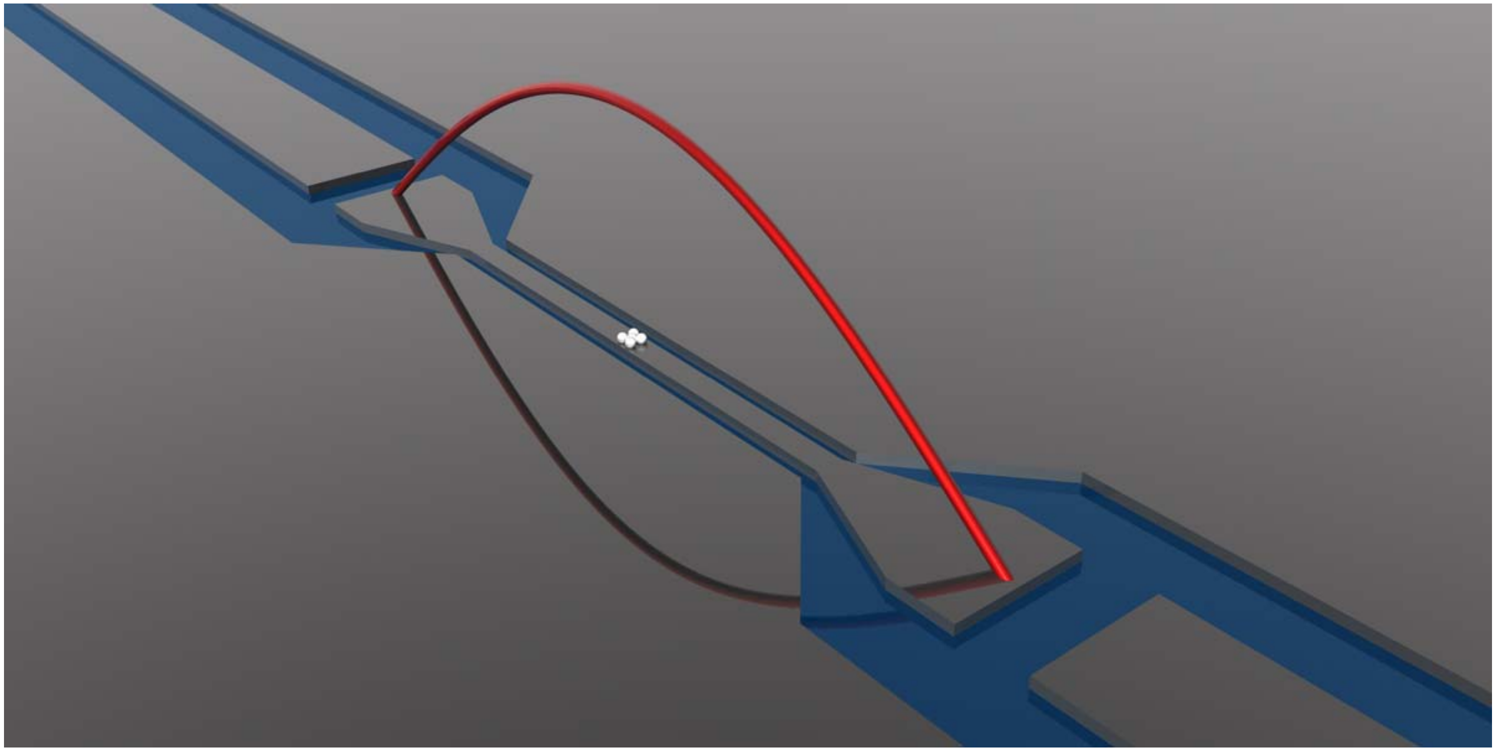
- Samples from Prof. G. Christou, University of Florida
- first micro-squid measurements up to 5.5 Tesla



Mn_{12}, S=10 molecular magnet

On-chip superconducting cavities

- RF field confined: large E and B fields
- Provide significant coupling between one photon and an ensemble of spins



Molecules or diluted spins, placed in the anti-node of an on-chip cavity

Spin-photon cooperative phenomena

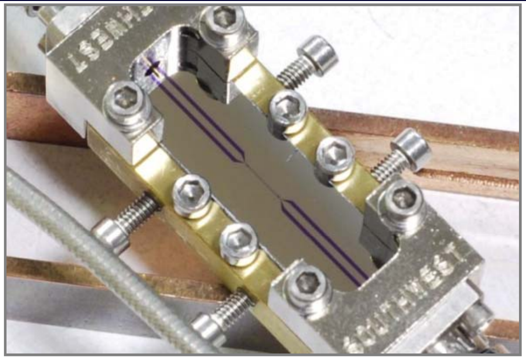
- Assume *a single two-level system* coupled to an RF field:

$$H = H_{TLS} + H_r + H_{JC} = h\nu_q \sigma_z / 2 + h\nu_r \left(a^\dagger a + 1/2\right) + hg\left(a^\dagger \sigma_- + a\sigma_+\right)$$

Zeeman term e.m. field spin-photon J.C. coupling

- When coupling *g* > cavity decay and spin dephasing rates, "strong coupling regime": spin-cavity photon exchange
- The photon exchange rate is the Rabi oscillation $\propto\sqrt{n}$ where $n=\langle a^\dagger a\rangle$ is the RF field energy
- Effects seen with E-field coupling:
  - atomic physics
  - superconducting charge qubits
  - quantum dots
- B-field effects much smaller.
  Solution: increase # of spins

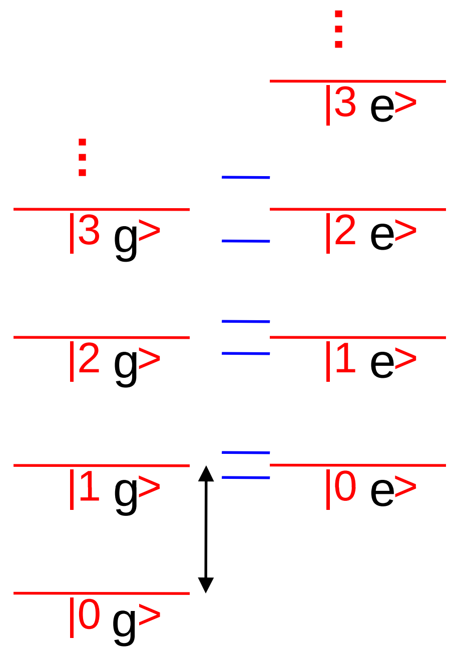
Prerequisites



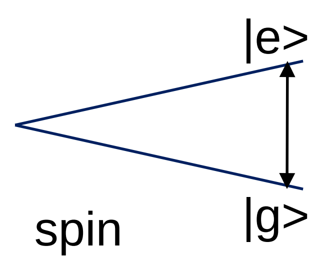
Cavity: $E = \hbar\omega(n + \frac{1}{2})$

$n = \langle a^{\dagger} a \rangle$

$$H = \hbar\omega(a^{\dagger}a + 1/2) + \hbar\omega_0\sigma_z/2 + \hbar g(a^{\dagger}\sigma_- + a\sigma_+)$$



- Jaynes-Cummings coupling *g*, from $E_{n=0} = \hbar\omega/2$
- For spins: with $\hbar g = g_S \mu_B S B_1$



1 atom, 1 photon

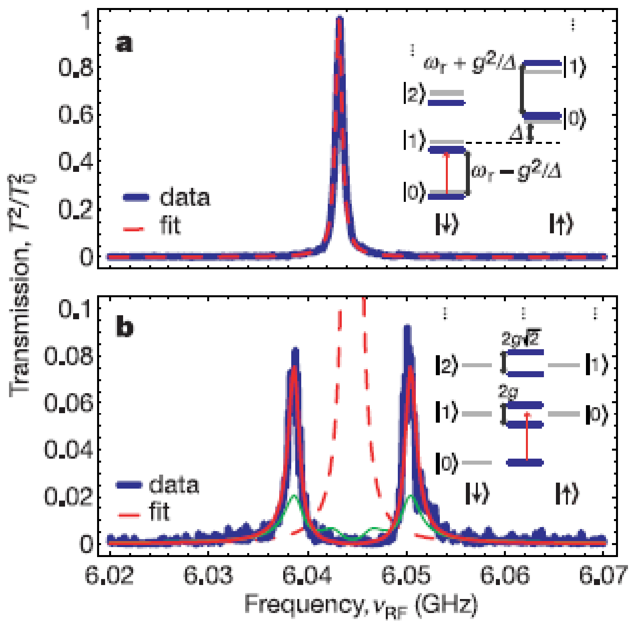
ω_0: Zeeman splitting (atom $\rightarrow$ spin)
ω : cavity resonance

2x2 H matrix:

$$\begin{pmatrix} \frac{-\hbar\omega_0}{2} + \hbar\omega & \hbar g \\ \hbar g & \frac{\hbar\omega_0}{2} \end{pmatrix}$$

"dispersive" regime:
QND measurement →

At resonance:
Vacuum Rabi splitting →



$$E_\pm = \hbar\omega/2 \pm \hbar\frac{\delta}{2}$$

$$\delta = \sqrt{\Delta^2 + 4g^2}$$

$$\Delta = \omega - \omega_0$$

E-field:
superc. qubit in an
on-chip cavity

A. Wallraff, D. I. Schuster, A. Blais, L. Frunzio, R. S. Huang, J. Majer, S. Kumar, S. M. Girvin and R. J. Schoelkopf, Strong coupling of a single photon to a superconducting qubit using circuit quantum electrodynamics, *Nature* **431**, 162-167 (2004).

I. Chiorescu, P. Bertet, K. Semba, Y. Nakamura, C. J. P. M. Harmans and J. E. Mooij, Coherent dynamics of a flux qubit coupled to a harmonic oscillator, *Nature* **431**, 159-162 (2004).

B-field with a MacroSpin:
superc. qubit in an LC oscillator

1 atom, n photons

ω_0: Zeeman splitting (atom $\rightarrow$ spin)
ω: cavity resonance

2x2 H *blocks* for each pair $\{|n,g\rangle, |n-1,e\rangle\}$

$$\begin{pmatrix} \frac{-\hbar\omega_0}{2} + n\hbar\omega & \hbar g \\ \hbar g & \frac{\hbar\omega_0}{2} + (n-1)\hbar\omega \end{pmatrix}$$

The eigenstates are given by

$$E_\pm = \hbar\omega(n - \frac{1}{2}) \pm \hbar\frac{\delta}{2}$$

$$|\Psi_\pm\rangle = \frac{1}{\sqrt{2}}\begin{pmatrix} \sqrt{1 \mp \Delta/\delta} \\ \pm\sqrt{1 \pm \Delta/\delta} \end{pmatrix}$$

where $\Delta = \omega - \omega_0$ and $\delta = \hbar\sqrt{\Delta^2 + 4ng^2} = E_+ - E_-$

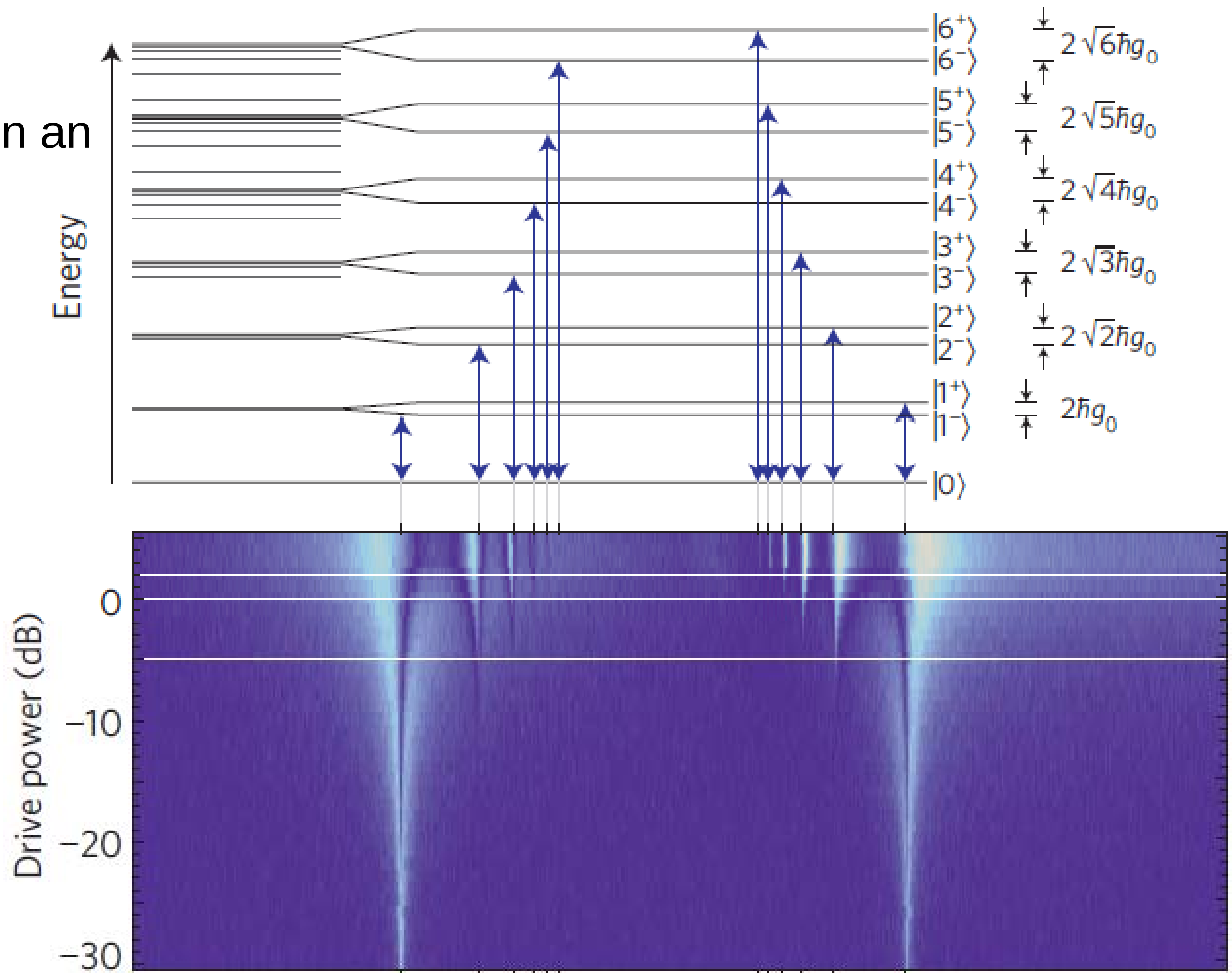
At resonance $\omega = \omega_0$

$$\boxed{E_\pm = (n - \frac{1}{2})\hbar\omega_0 \pm \hbar g\sqrt{n}}$$

1 atom, n photons

Experiments

E-field:
superc. qubit in an on-chip cavity



Blue arrows: non-linear multi-photon processes

L.S. Bishop, J. M. Chow, J. Koch, A. A. Houck, M. H. Devoret, E. Thuneberg, S. M. Girvin, R. J. Schoelkopf, “Nonlinear response of the vacuum Rabi resonance”, *Nature Physics* **5**, 105 (2009).

N atoms

- Assume *N* non-interacting spins coupled to an RF field:

$$H = h\nu_0 \sum_i \sigma_z^i / 2 + h\nu\left(a^\dagger a + 1/2\right) + \hbar g \sum_i \left(a^\dagger \sigma_-^i + a\sigma_+^i\right)$$

$S=\sum_i s_i$ a large Macrospin

$$H = h\nu_0 S_z / 2 + h\nu\left(a^\dagger a + 1/2\right) + \hbar g\left(a^\dagger S_- + aS_+\right)$$

Same formalism, but spectroscopy peaks separation goes up $g \rightarrow g\sqrt{N}$!!

M. Tavis and F.W. Cummings, “Exact Solution for an N-Molecule-Radiation-Field Hamiltonian”, *Phys. Rev.* **70**, 379 (1968).

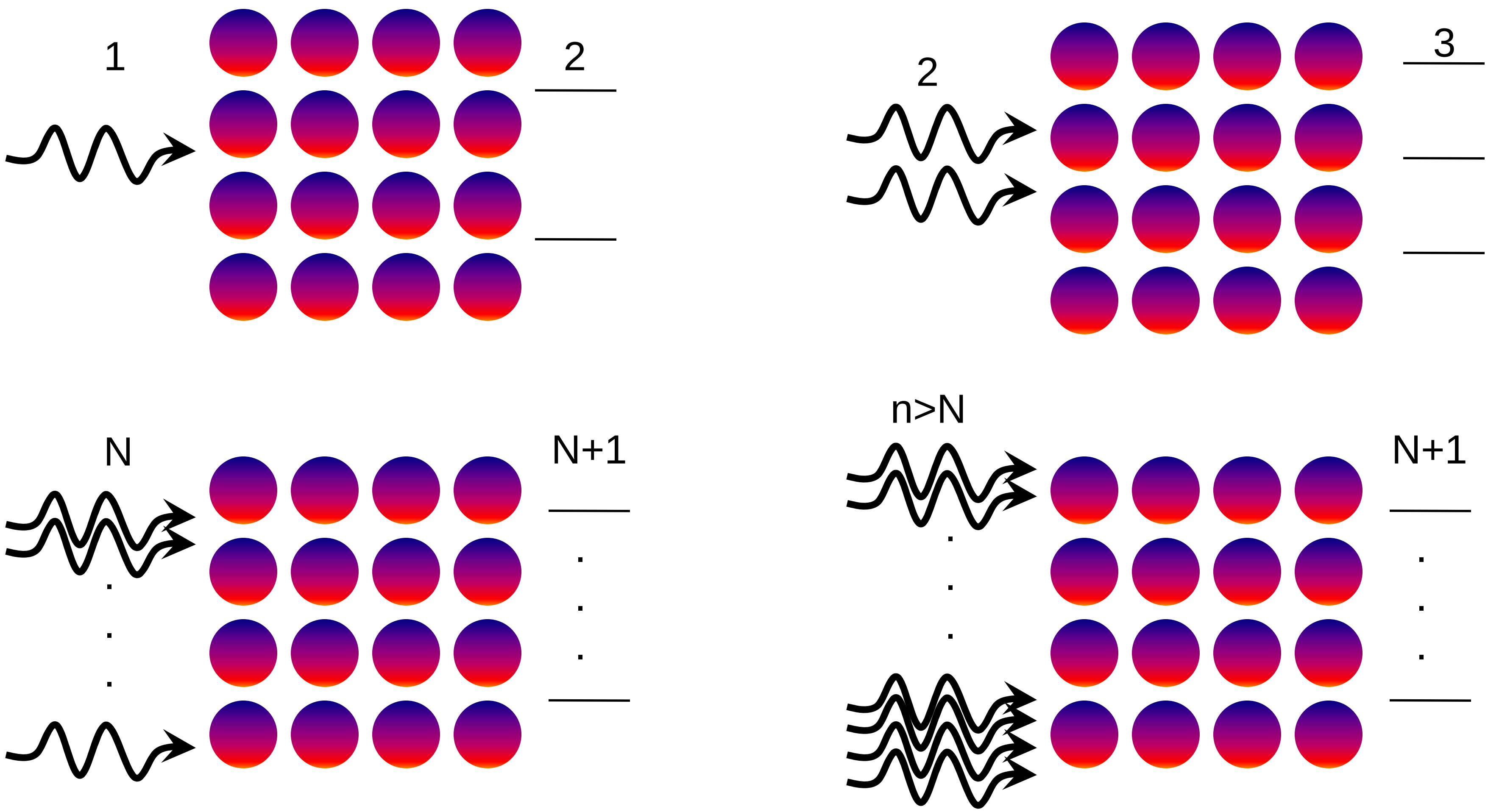
G. S. Agarwal, “Vacuum-Field Rabi Splittings in Microwave Absorption by Rydberg Atoms in a Cavity”, *Physical Review Letters* **53**, 1732 (1984).

- Seen in quantum optics for up to ~10^3-10^4 atoms (E-field effect): Kimble group (Caltech), Haroche group (ENS-Paris)
- Superconducting charge qubits

Multi-spin/photon cooperative effects

- consider a system of *N* spins and *n* photons

*I. Chiorescu, N. Groll, S. Bertaina, T. Mori, S. Miyashita, Phys. Rev. B, **82**, 024413 (2010)*



N atoms, *1* photon

- *N* spins and 1 photon: the resonance case

$$\begin{pmatrix} \hbar\omega_0(1-N/2) & 0 & \dots & 0 & \hbar g \\ 0 & \hbar\omega_0(1-N/2) & \dots & 0 & \hbar g \\ \vdots & \vdots & \ddots & 0 & \hbar g \\ 0 & 0 & \dots & \hbar\omega_0(1-N/2) & \hbar g \\ \hbar g & \hbar g & \dots & \hbar g & -N\hbar\omega_0/2+\hbar\omega \end{pmatrix}$$

⇓ Unitary transformation (think H_2 molecule, S-T states)

$$\begin{pmatrix} \hbar\omega_0(1-N/2) & 0 & \dots & 0 & 0 \\ 0 & \hbar\omega_0(1-N/2) & \dots & 0 & 0 \\ \vdots & \vdots & \ddots & 0 & 0 \\ 0 & 0 & \dots & \hbar\omega_0(1-N/2) & \hbar g\sqrt{N} \\ 0 & 0 & \dots & \hbar g\sqrt{N} & -N\hbar\omega_0/2+\hbar\omega \end{pmatrix}$$

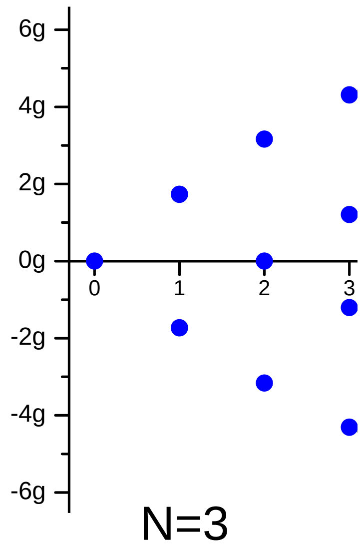
- *only* the case S=N/2

$$E_1^{(1)} - E_0^{(1)} = \hbar\sqrt{\Delta^2 + 4Ng^2}$$

- the basis is $|1, N\rangle \text{and} |0, \mathrm{N}-1\rangle$

N atoms, n photons

- n photons, at resonance



$$E_{interaction} = E_{eigenstate} - nh\nu - E_0^0$$

n=1 $\Rightarrow \pm g\sqrt{N}$

n=2 $\Rightarrow 0, \pm 2g\sqrt{N-1/2}$

n=3 $\Rightarrow \pm g\sqrt{5(N-1) \pm \sqrt{(4N-5)^2 + 8N}}$

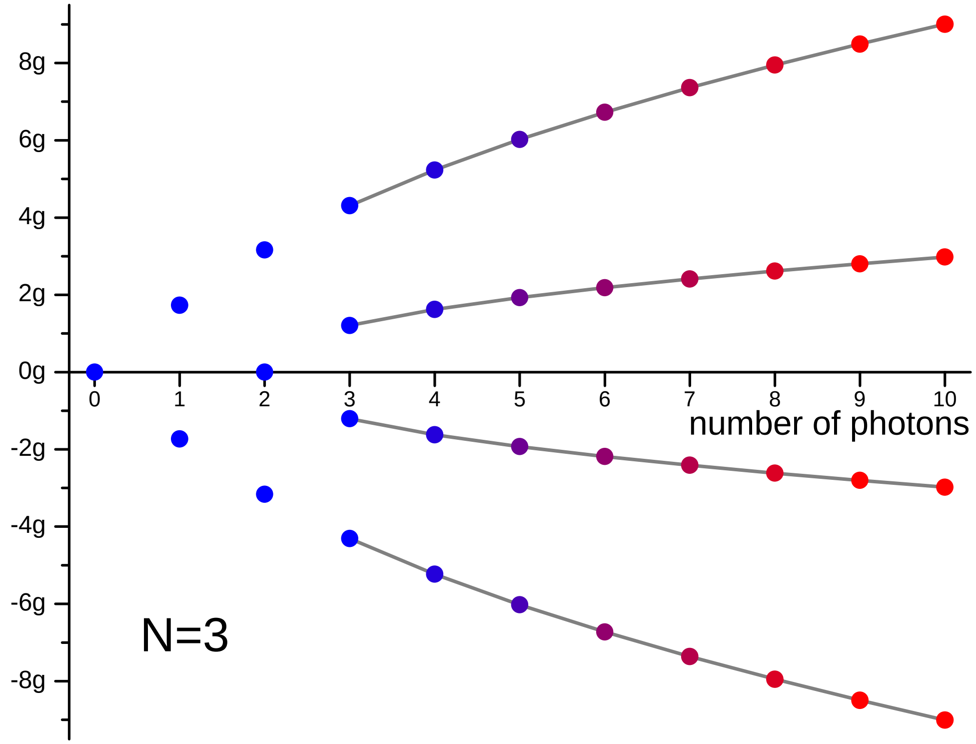
New levels appears, until $n=N$

E_0^0 (empty cavity with N spins in |g>)

N atoms, *n* photons

- *n* photons, at resonance
- Ex: N=3

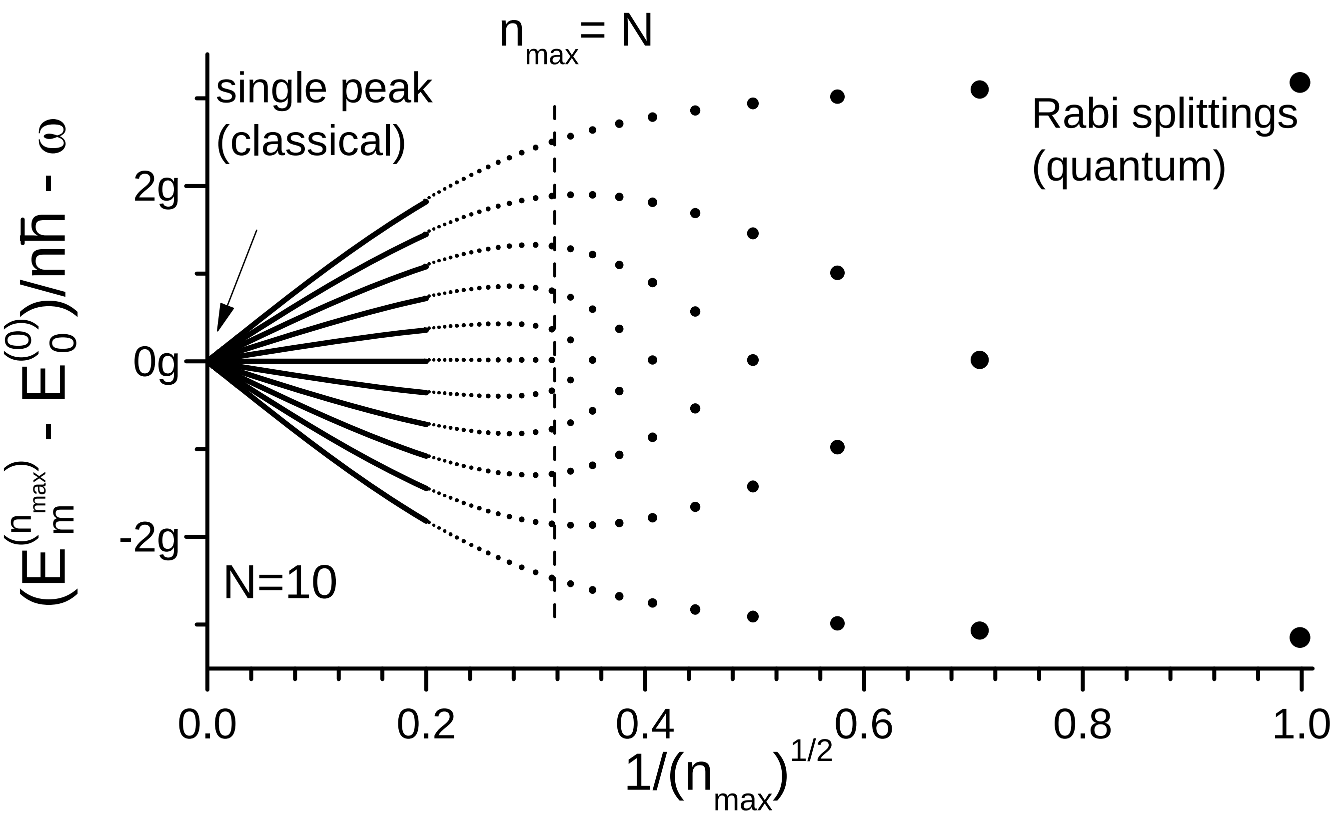
$$\pm g\sqrt{5(N-1)\pm\sqrt{(4N-5)^2+8N}} \xrightarrow{N\to n\gg 1} \pm 3g\sqrt{n}, \pm g\sqrt{n}$$



- no new levels are generated
- the "oversaturation" is adjusting the *N+1* existing levels

Rabi splittings & ESR

- *nonlinear multi-photon* excitations
- *divide by n* the energy distance to E_0^0 (empty cavity with *N* spins in |g>)



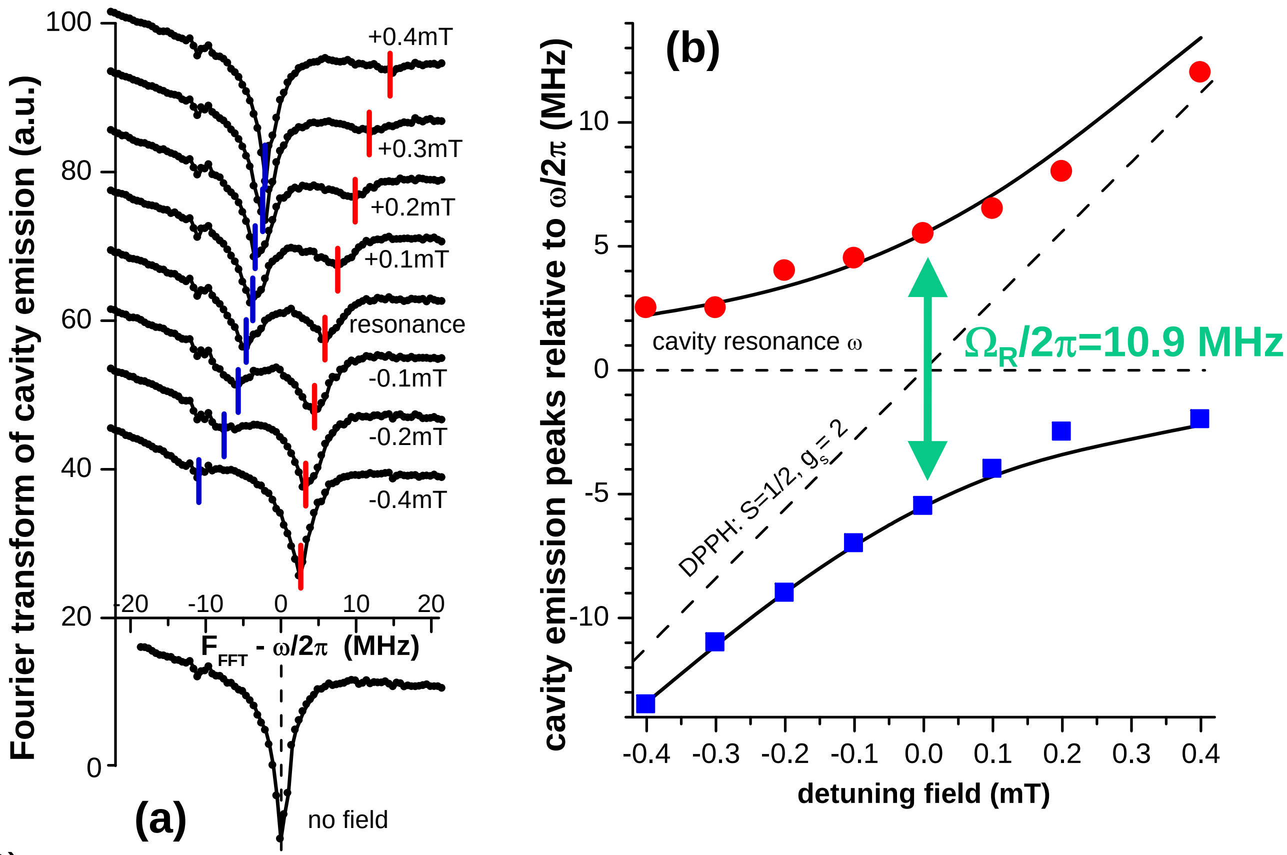
Spins 1/2 coupled to photons

- S=1/2 sample (DPPH), home-build heterodyne detector
  (UCGP-NHMFL online report 2007)

$$\omega_{e0,1} - \omega = -\frac{\Delta}{2} \pm \frac{1}{2}\sqrt{\Delta^2 + \Omega_R^2}$$

- cavity emission (ringing)
- not only one photon in the cavity (ok, b/c at RT)
- *N* spins and *1* photon

  $\Omega_R = 2g\sqrt{N}$
- rough estimation:
  N/V=4e18 cm^{-3}



<u>NV in diamonds:</u>

Yale: PRL, 105, 140501 (2010)

Saclay: PRL, 105, 140502 (2010)

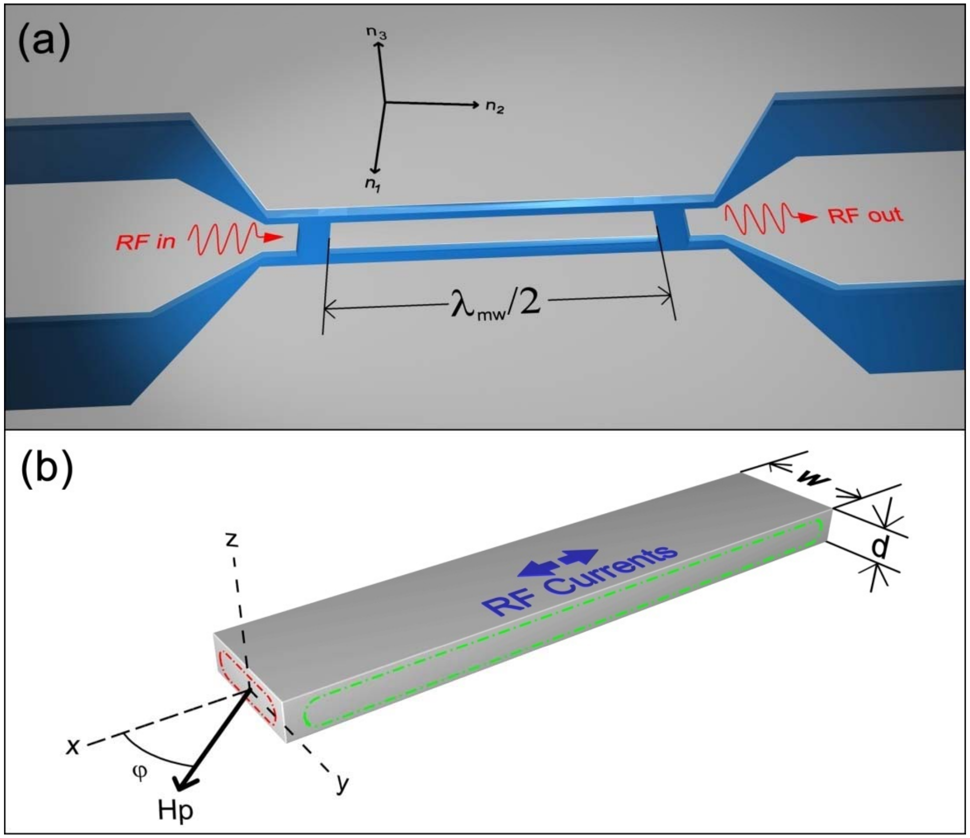
Featured in *Nature*, **468**, 44 (2010)

Superconducting cavity in high fields

N. Groll, A. Gurevich, I. Chiorescu, *PRB Rap Comm* **81**, 020504 (2010)

- Meissner effect: $J = -en_s v_s$ at low fields
- Nonlinear Meissner effect (NLME): $J = -en_s(v_s)v_s$ at high fields

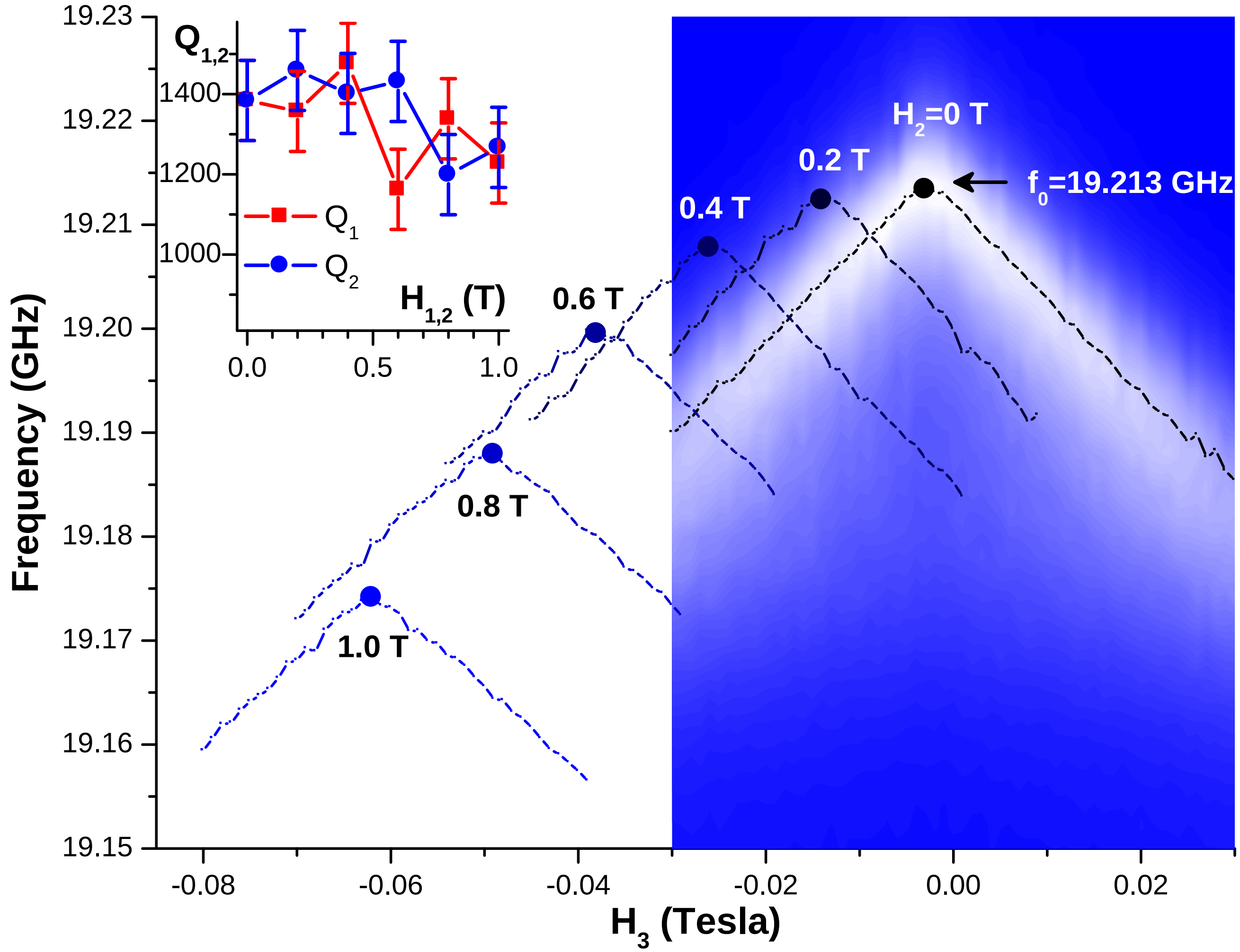
- very week effect: needs large fields, but is easily masked by vortices!
- needs a true Meissner state: thin films & precise field orientation



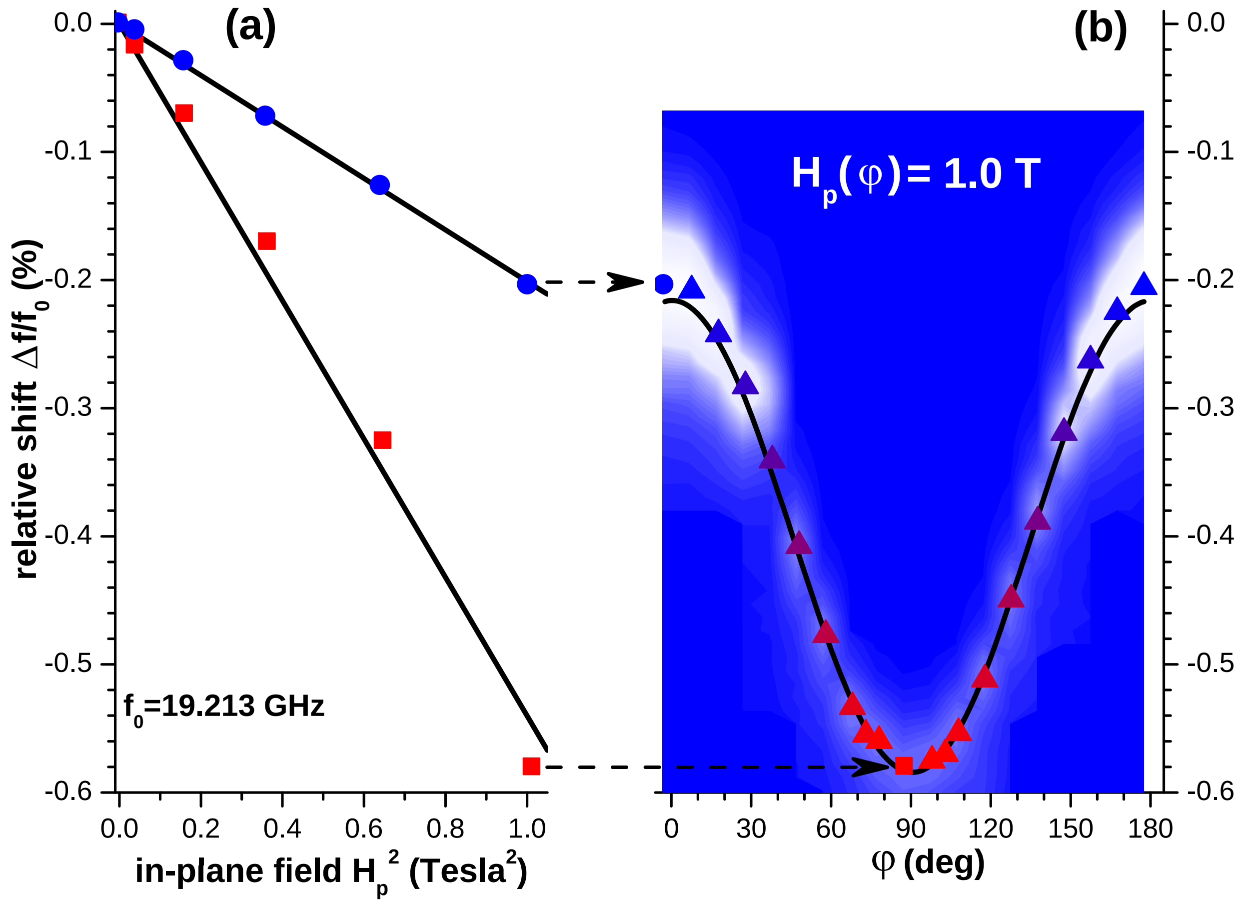
- in a linear cavity, there is a well defined orientation between J and the RF current
- NLME can probe unconventional pairing symmetries of moving condensates (s-,p-,d-waves or multiband superconductivity in pnictides)

Nb: vortex dissipation *vs.* NLME

- resonance frequency vs. "axial" fields
- resonance frequency decreases: linearly (vortices) and quadratically (cusp, NLME)



NLME: angular spectroscopy



- Same method can be applied to study other superconductors
- Current collaborations: T. Prolier (Argonne), C.-B. Eom (Wisconsin)

Conclusions

- *On-chip SQUID, a highly sensitive flux detector:*
  - aspects of spin dynamics in molecular materials,

- *On-chip superconducting cavity:*
  - low-number photon field coupled to independent spins
  - sensitive magnetic resonance, on-chip

- *Quantum Optics with solid-state systems*
  - based on atomic physics, well-established field
  - important applications (quantum computing)
  - fundamental aspects of Quantum Mechanics in solids