The Abdus Salam
International Centre for Theoretical Physics

United Nations
Educational, Scientific
and Cultural Organization

International Atomic
Energy Agency

SMR 1760 - 19

COLLEGE ON

PHYSICS OF NANO-DEVICES

10 - 21 July 2006

Endohedral fullerenes and electron spin resonance quantum information processing

Presented by:

Arzhang Ardavan

University of Oxford, United Kindgdom

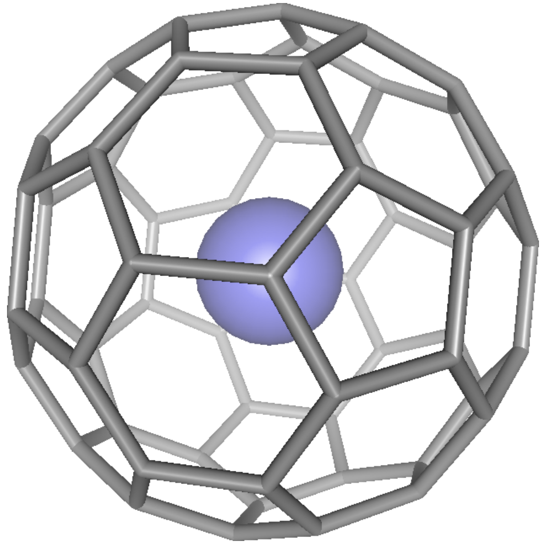
Endohedral fullerenes and electron spin resonance quantum information processing

Arzhang Ardavan
Department of Physics, University of Oxford

Oxford: **John Morton**, Gavin Morley (now at NHMFL)
Simon Benjamin, Kyriakos Porfyrakis, Andrew Briggs

Princeton: **Alexei Tyryshkin**, Steve Lyon

$N@C_{60}$



"Atomic" N is located at high symmetry point in C_{60} cage:

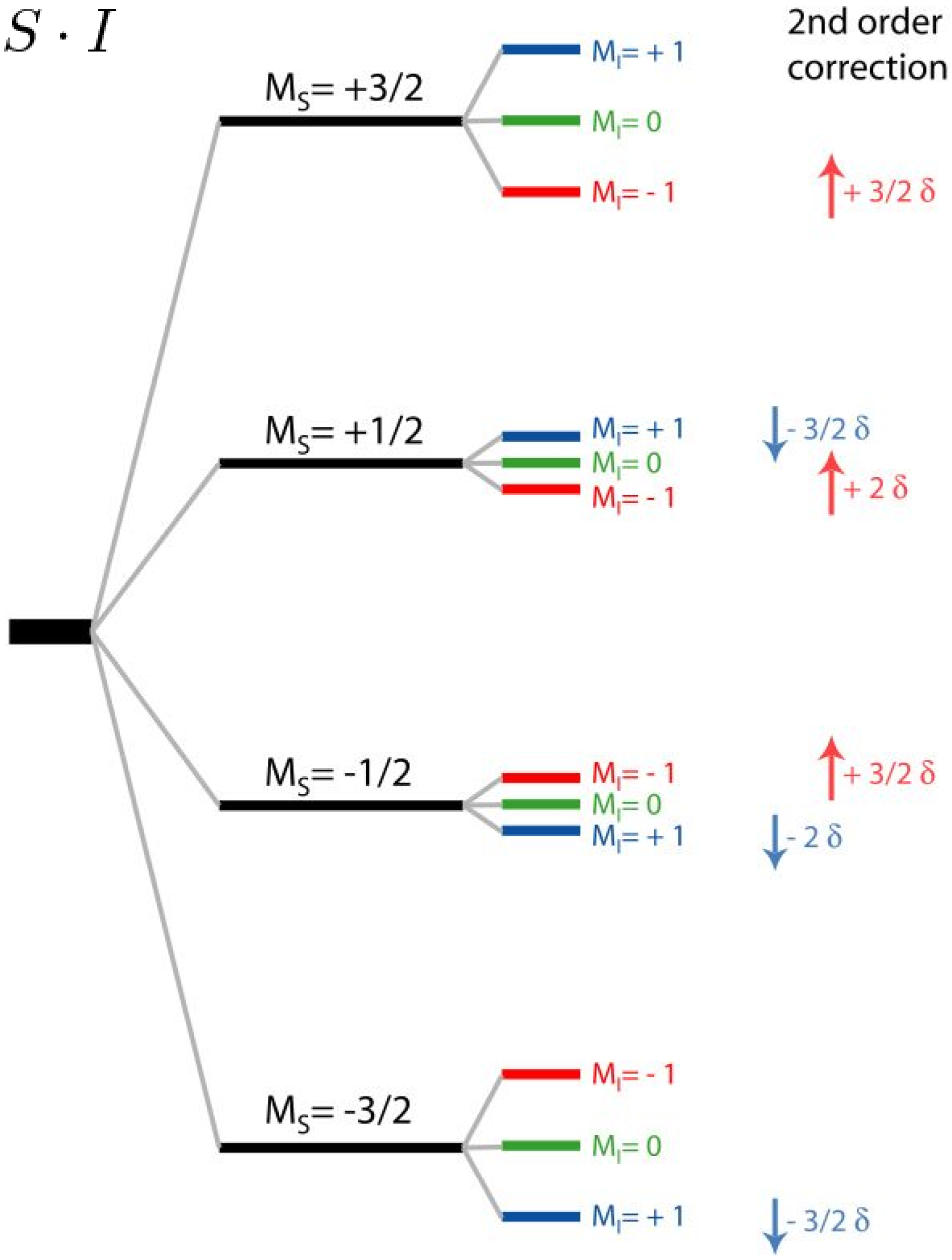
- N retains its S=3/2 spin,
- ^{14}N and ^{15}N varieties can be manufactured,
- very little interaction between N and cage,
- $N@C_{60}$ is almost indistinguishable chemically from C_{60}.

Synthesis by ion implantation or plasma discharge yields ~ 1 $N@C_{60}$ molecule per 10^5 C_{60} molecules:

- purification is a challenge!

N@C$_{60}$

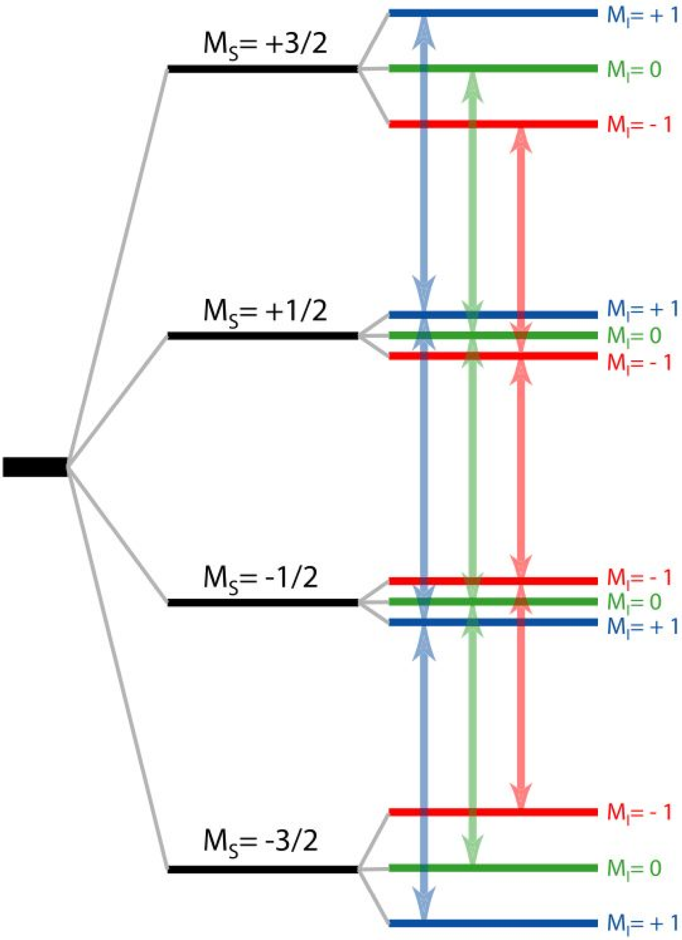
$$\mathcal{H} = \omega_S S_z + \omega_I I_z + aS \cdot I$$

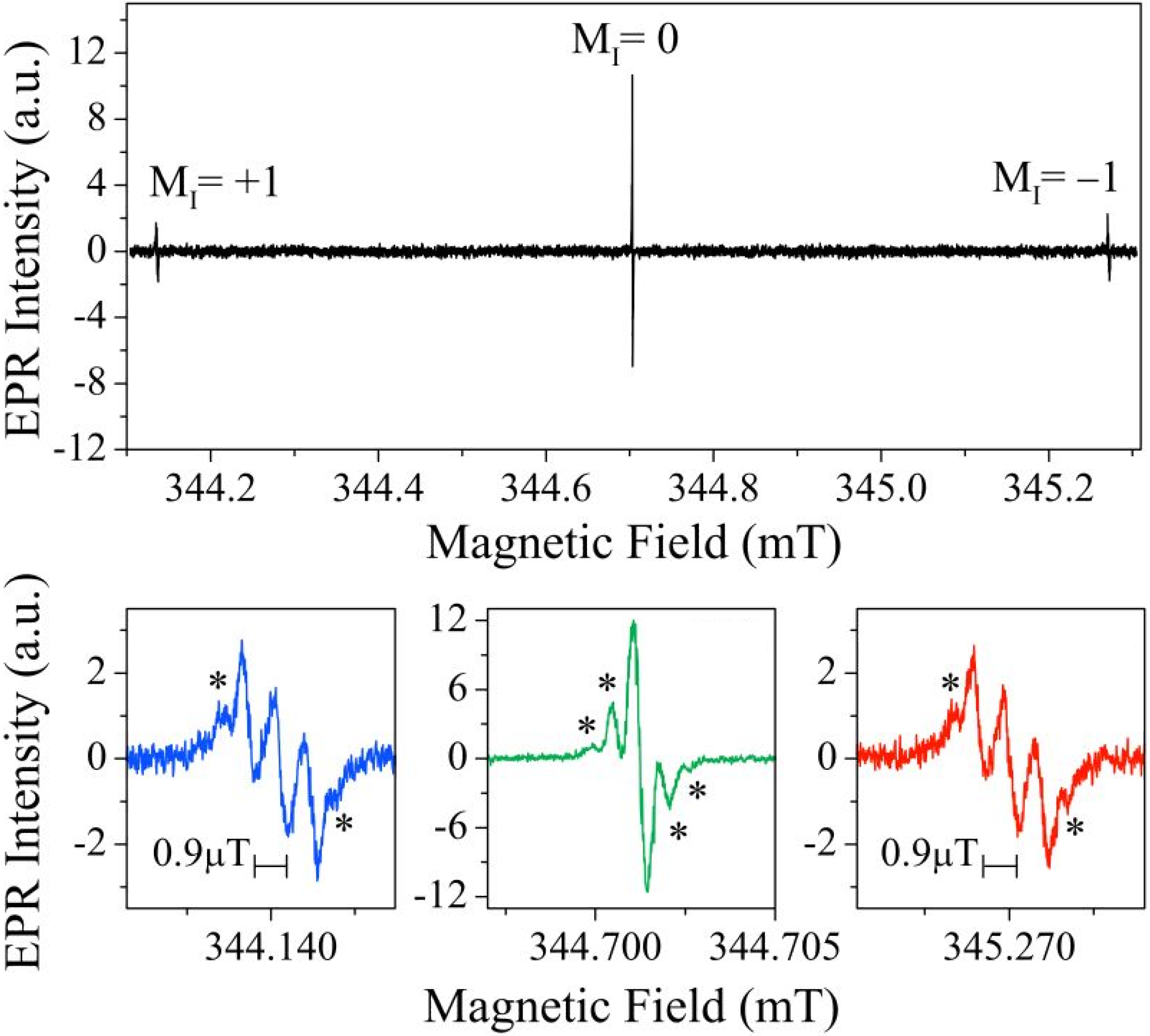


$$\delta = \frac{a^2}{B}$$

$N@C_{60}$

$$\mathcal{H} = \omega_S S_z + \omega_I I_z + a S \cdot I$$

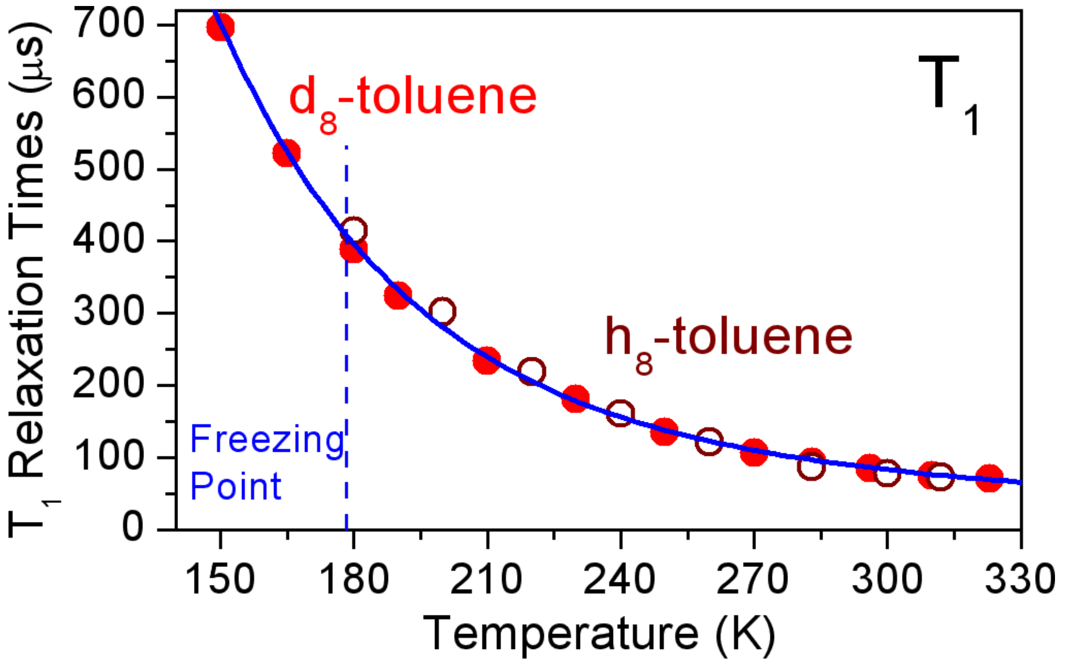




N@C_{60} lifetimes in Toluene

T_1 has bi-exponential dependence.
Mechanisms: collision induced

- zero-field splitting fluctuations?
- rotation – spin coupling?

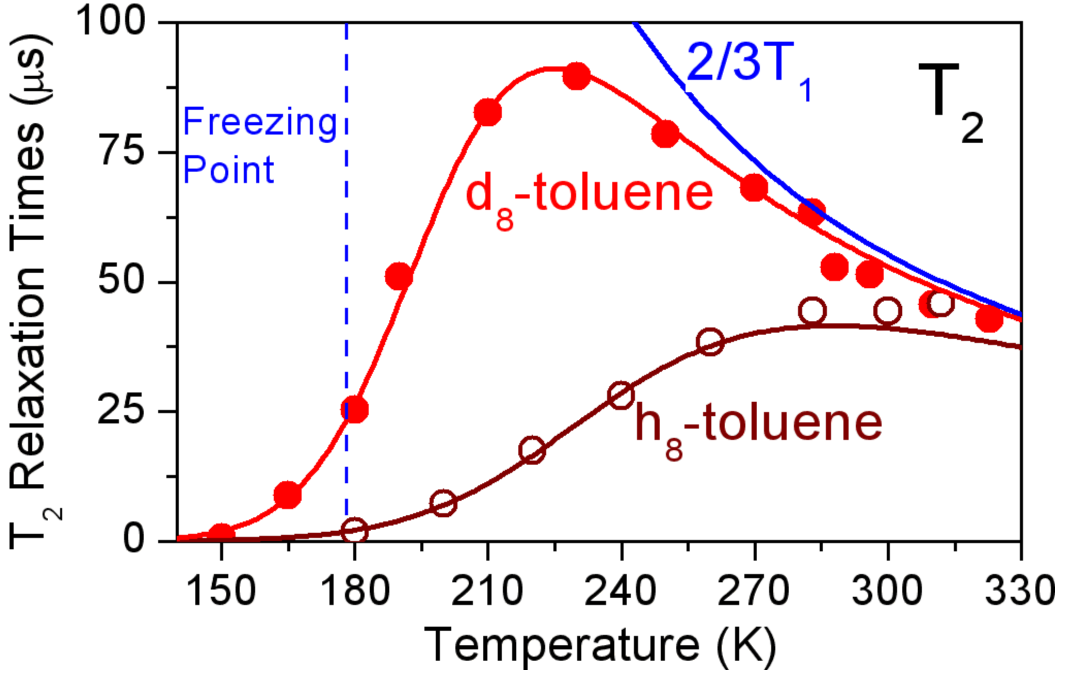


T_2 depends on magnetic nuclei in solvent,

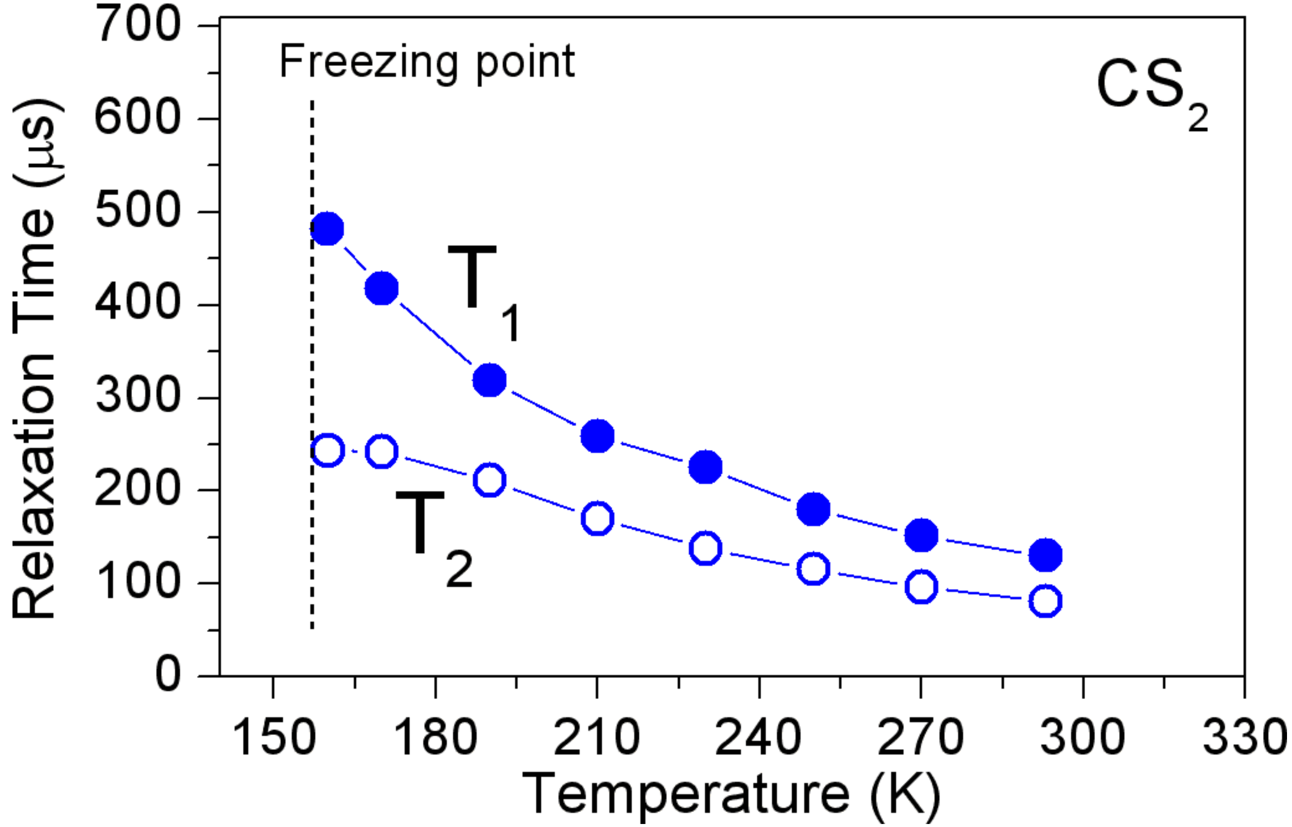
$$\frac{1}{T_2} = \frac{3}{2T_1} + \frac{1}{T(\text{diffusion})}$$

Diffusion relaxation depends on:

- diffusion rate of C_{60} in toluene
- H/D concentration
- $d_{min\ \mathrm{N\text{-}H/D}} = 4.5$ Å



N@C_{60} lifetimes in CS_2



High temperature:

- $T_2 \approx 2/3\ T_1$ (intrinsic?)
- Maximum T_2 = 243 μs at 157 K.

Low temperature:

- T_2 drops when solvent freezes
- Maximum $T_1 \sim$ 1 minute at 4K

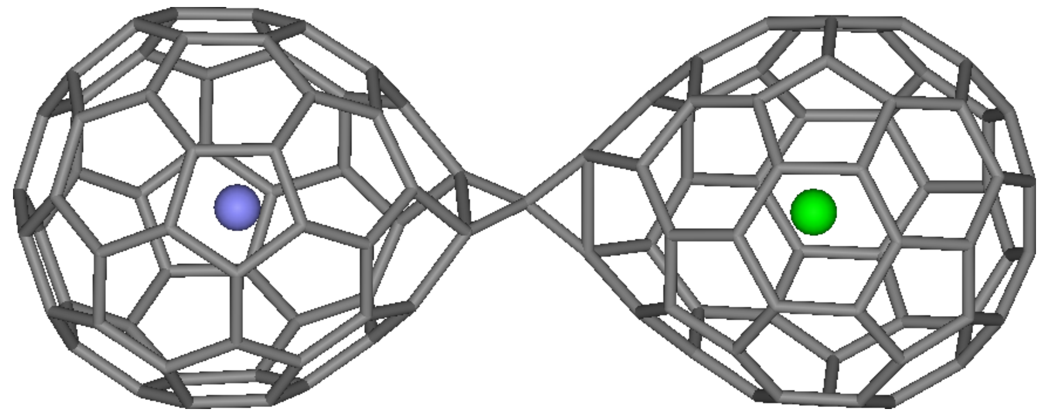
N@C_{60} as a “bottom-up” qubit

Useful properties:

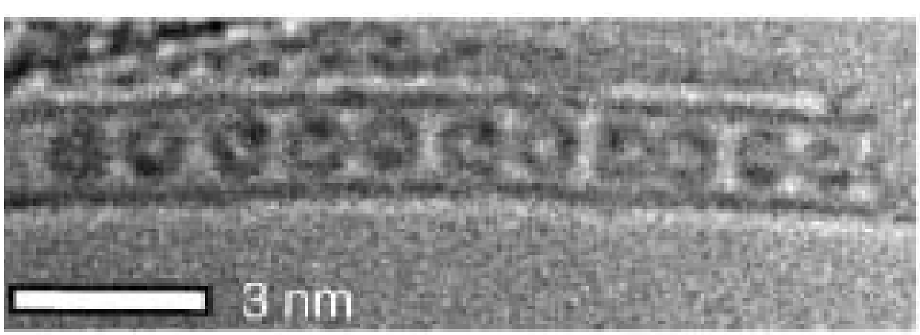
- Well-defined molecular building-block.
- Extremely long spin lifetimes,
  - Single qubit figure-of-merit: (coherence time)/(time for π-pulse) > 10000.
- Pulsed electron spin resonance provides the necessary single-qubit unitary transformations.
- Fullerene chemistry is mature – comp multi-qubit structures are possibl

Multi-qubit structures

Asymmetric endohedral fullerene dimer:



Globally addressed linear array,
S.C. Benjamin, J. Levy



Ce@C_{82} peapods:
A.N. Khlobystov et al.,
Angew. Chem. Int Ed. **24** 1386 (2004)

Electron moments in QIP

INSTITUTE OF PHYSICS PUBLISHING NANOTECHNOLOGY

Nanotechnology 1

TUTORI

Reci
comp

Veronica Ce

Department of P
4056 Basel, Swit

Received 1 D
Published 25
Online at stac

Abstract
Technological
measured by t
microchip. U
growth in the
of the transist
mechanics tak
for progress (
stimulated ext

NATURE | VOL 410 | 12 APRIL 2001

Qua
in m

Michael

Departm
4056 Ba

Shor a
outper
searchi
mechan

INSTITUTE OF PHYSICS PUBLISHING SEMICONDUCTOR SCIENCE AND TECHNOLOGY

Semicond. Sci. Technol. 17 (2002) 355–366 PII: S0268-1242(02)33606-X

Electron spins in artificial atoms and molecules for quantum computing

Vitaly N Golovach and Daniel Loss

Department of Physics and Astronomy, University of Basel, Klingelbergstrasse 82, CH-4056 Basel, Switzerland

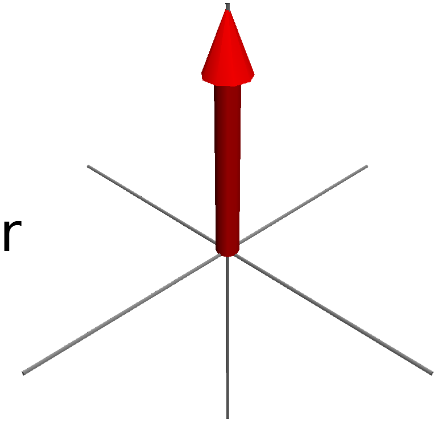
Received 7 February 2002
Published 20 March 2002
Online at stacks.iop.org/SST/17/355

Single-spin rotations can be achieved by ESR techniques [39]. One applies a static local magnetic field B for the qubit(s), which should be rotated. An ac magnetic field is then applied perpendicular to the first field with the resonant frequency that matches the Larmor frequency $\omega_L = g\mu_B B/\hbar$. Due to paramagnetic resonance [40], this causes spin flips in the quantum dots with the corresponding Zeeman splitting.

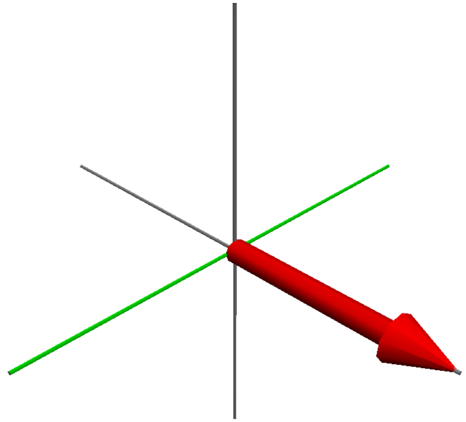
Single qubit gate fidelities

How well can we perform unitary transformations in an ESR spectrometer?

Doing a $\pi/2$-pulse or

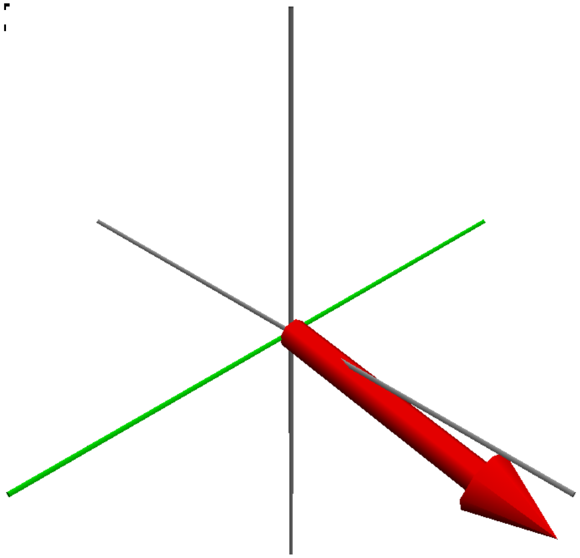


should

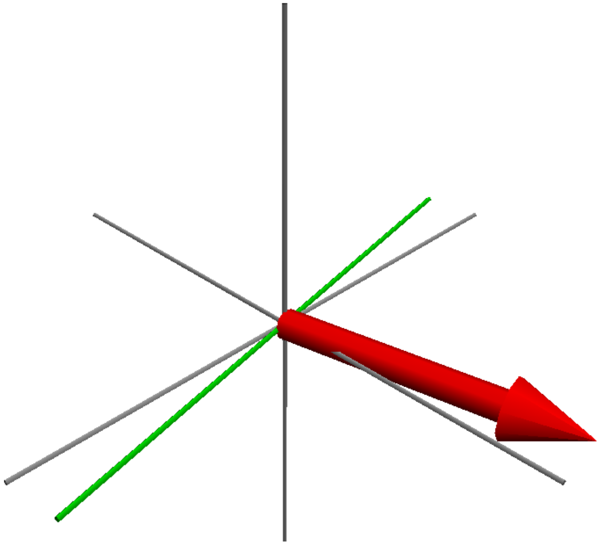


give

But we might get rotation angle errors:

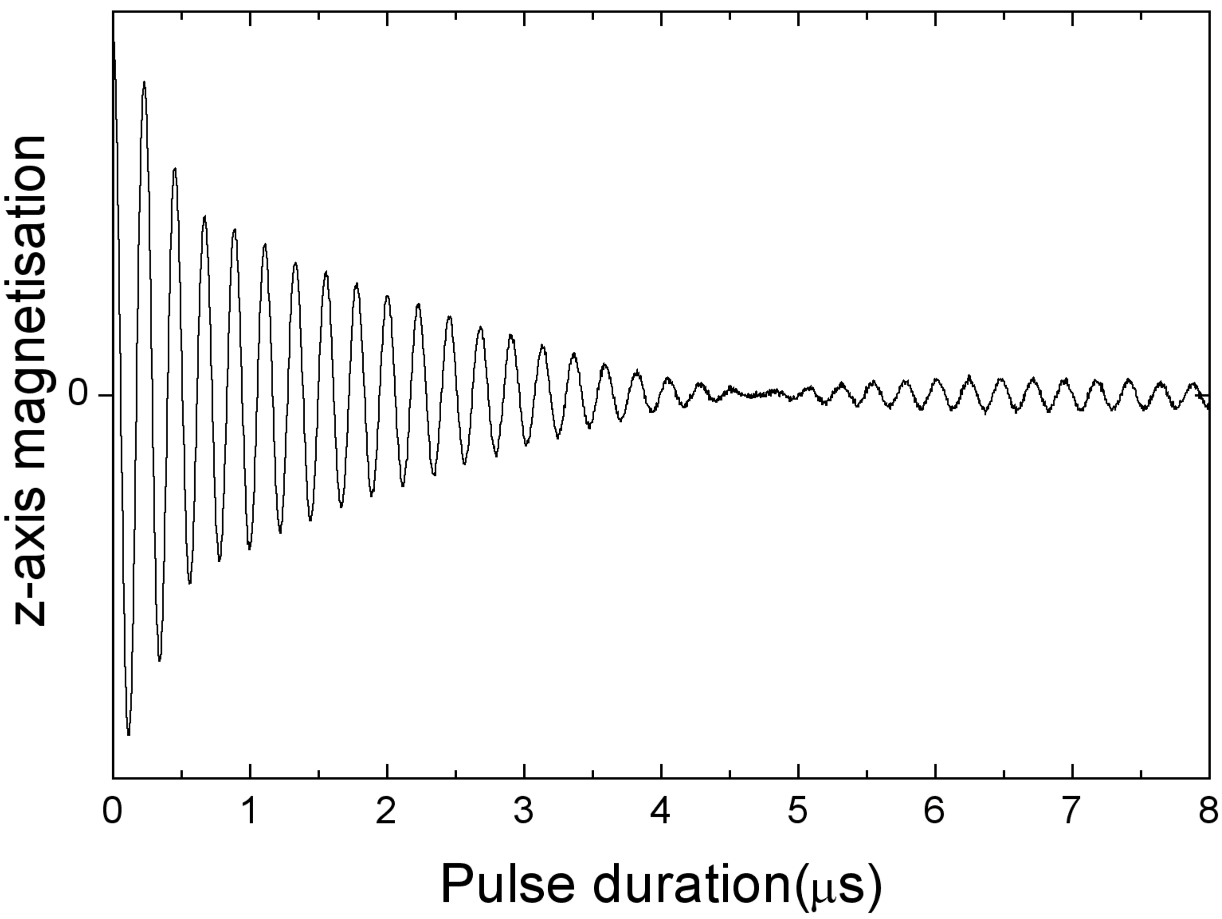


or rotation axis errors:



And can we perform complicated pulse sequences?

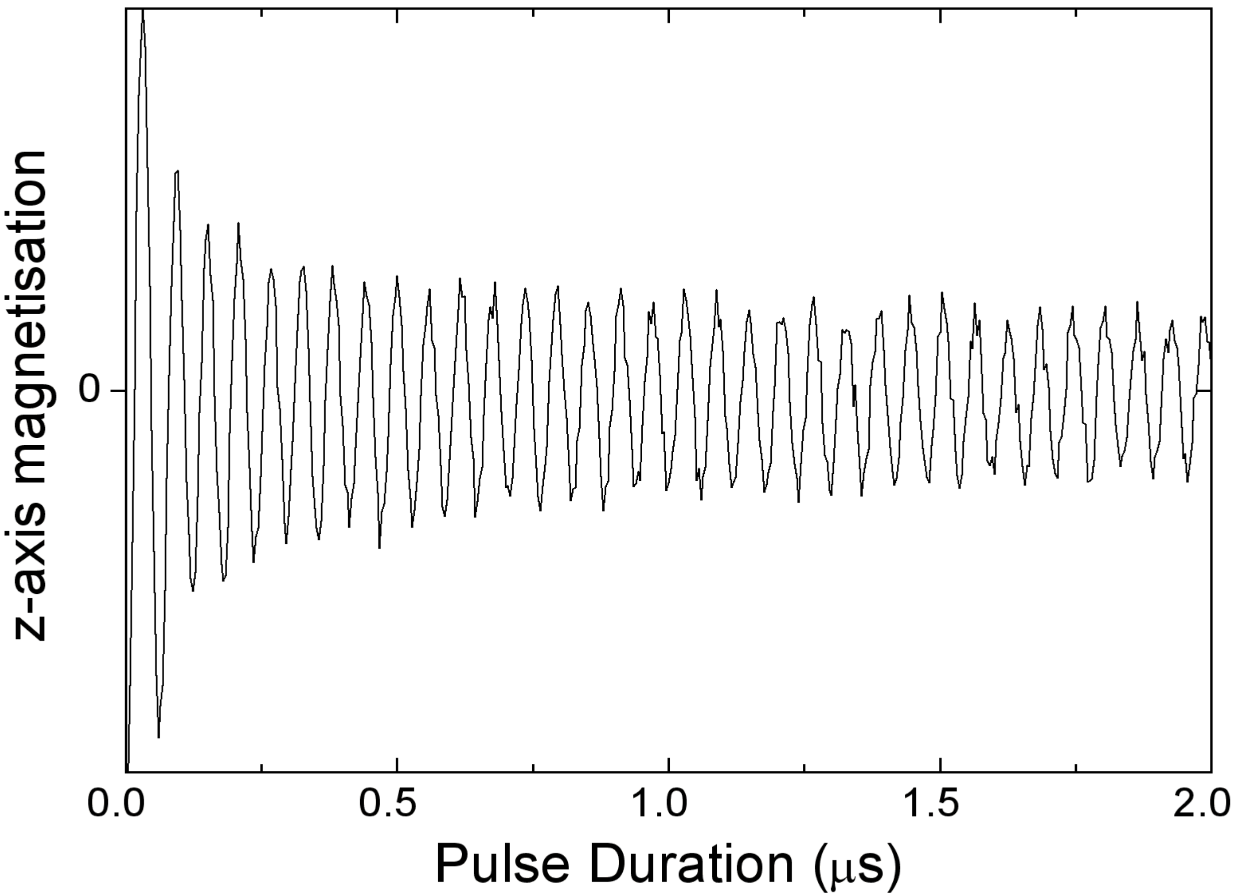
Rabi oscillations



Microwave field inhomogeneity → Rabi oscillation envelope

Rabi oscillations

With a smaller sample:



Testing rotation angle error: Carr – Purcell (CP) and Carr – Purcell – Meiboom – Gill (CPMG)

Borrowing from NMR:

Carr – Purcell:

$\pi/2_x - \tau - \pi_x - \tau - \text{echo} - \tau - (\ \pi_x - \tau - \text{echo} - \tau\ )_n$

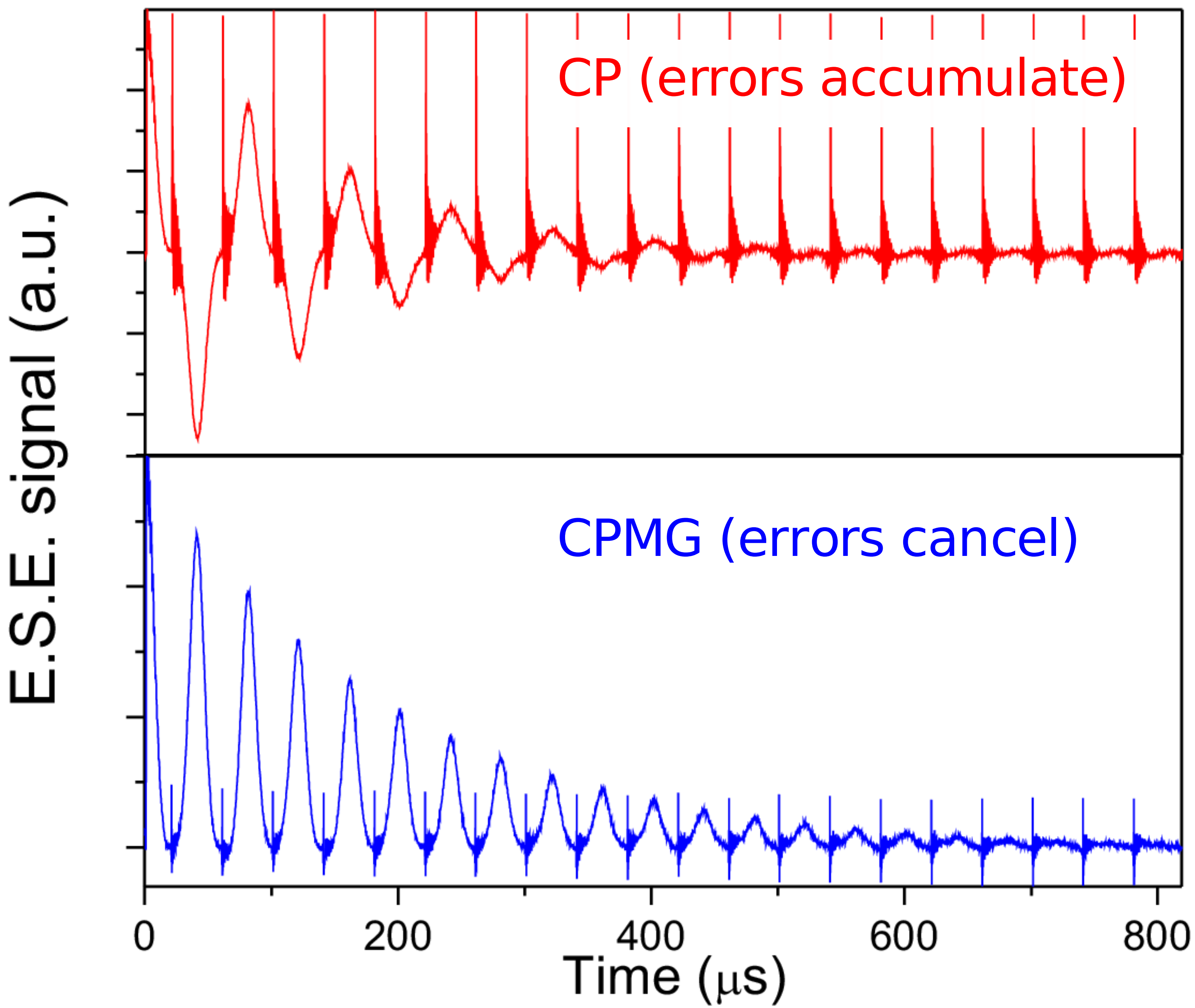
- Errors accumulate

Carr – Purcell – Meiboom – Gill:

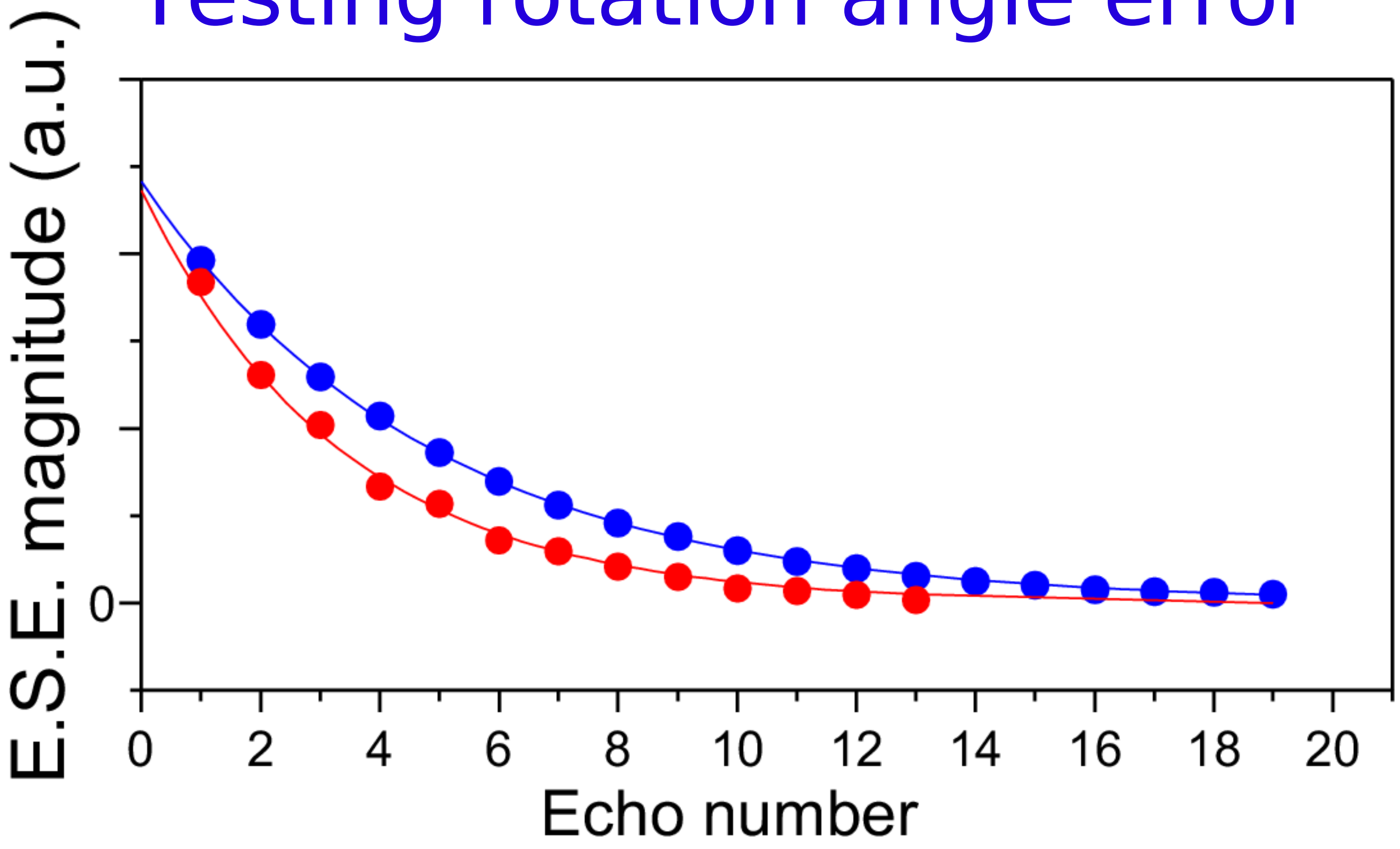
$\pi/2_x - \tau - \pi_y - \tau - \text{echo} - \tau - (\ \pi_y - \tau - \text{echo} - \tau\ )_n$

- Errors cancel out

Testing rotation angle error



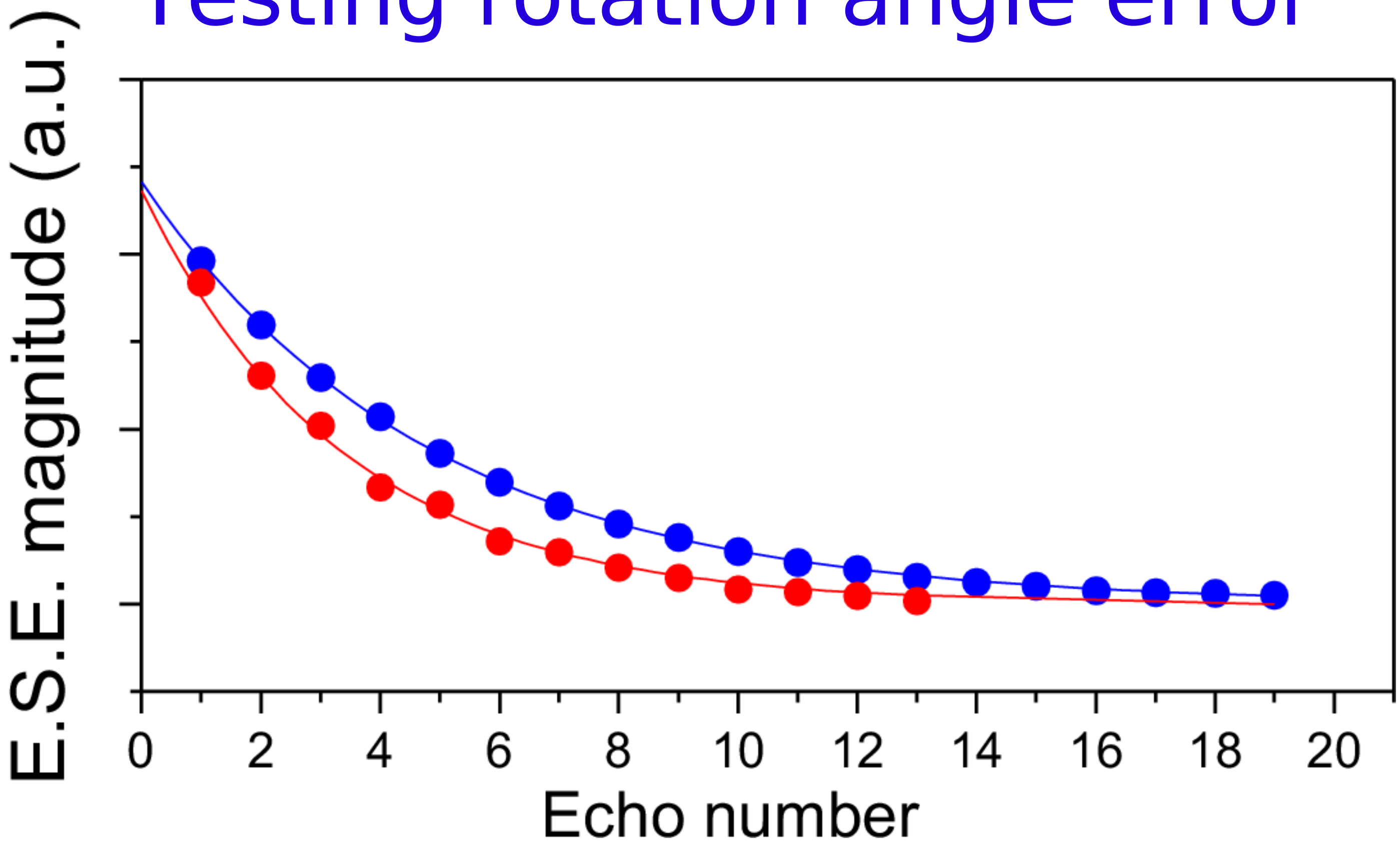
Testing rotation angle error



CPMG: $A_{ESE}(n) = A_0 \exp\left(-\frac{2n\tau}{T_2}\right)$

T_2 = 190 μs

Testing rotation angle error



CP: $$A_{ESE}(n) = A_0 \left[\int_{-\infty}^{\infty} \frac{1}{\sigma\sqrt{2\pi}} \exp\left(-\frac{\delta^2}{2\sigma^2}\right) \cos(n\delta)\mathrm{d}\delta \right] \exp\left(-\frac{2n\tau}{T_2}\right)$$
$$= A_0 \exp\left(-\frac{n^2\sigma^2}{2} - \frac{2n\tau}{T_2}\right)$$

for a gaussian distribution of rotation angle errors, δ.
Rotation angle error, σ = 18 degrees, or 10%...

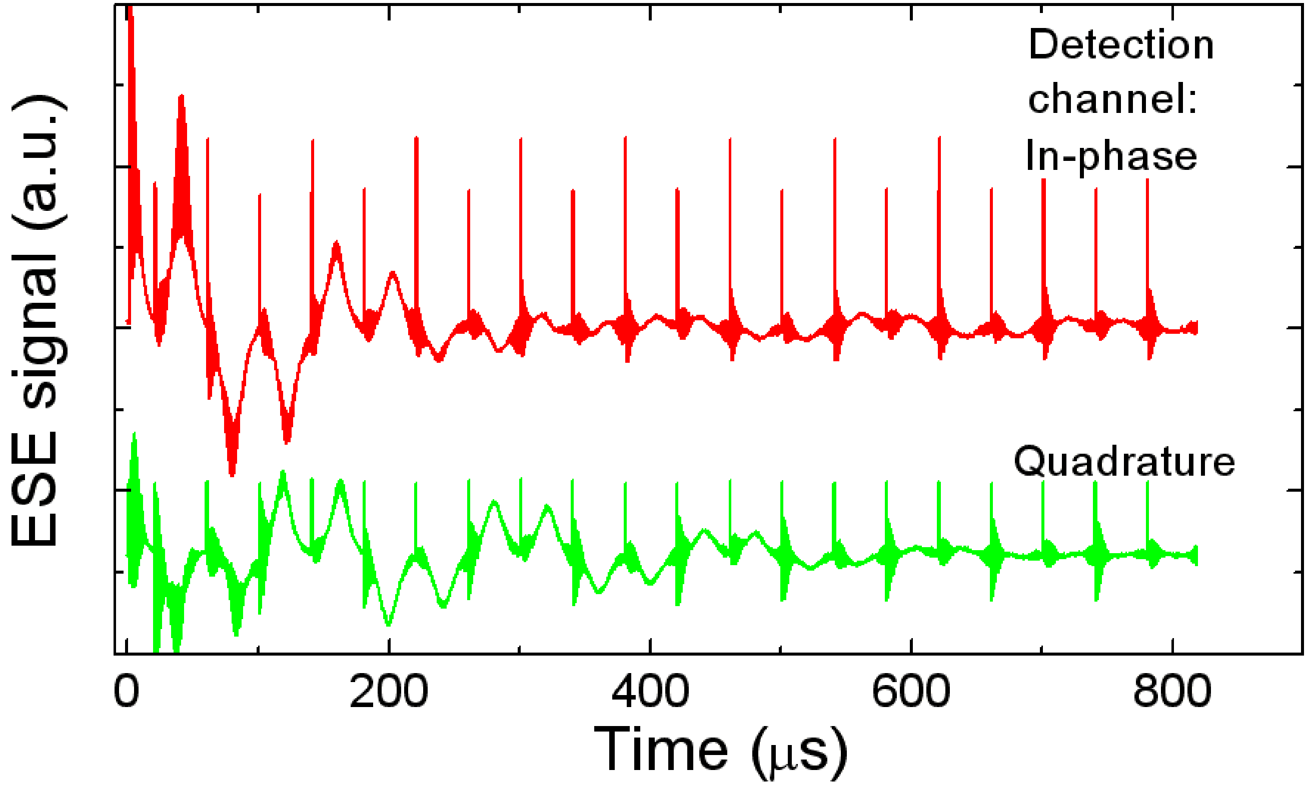
Testing axis error:
Sequence for Phase-error AMplification

SPAM:

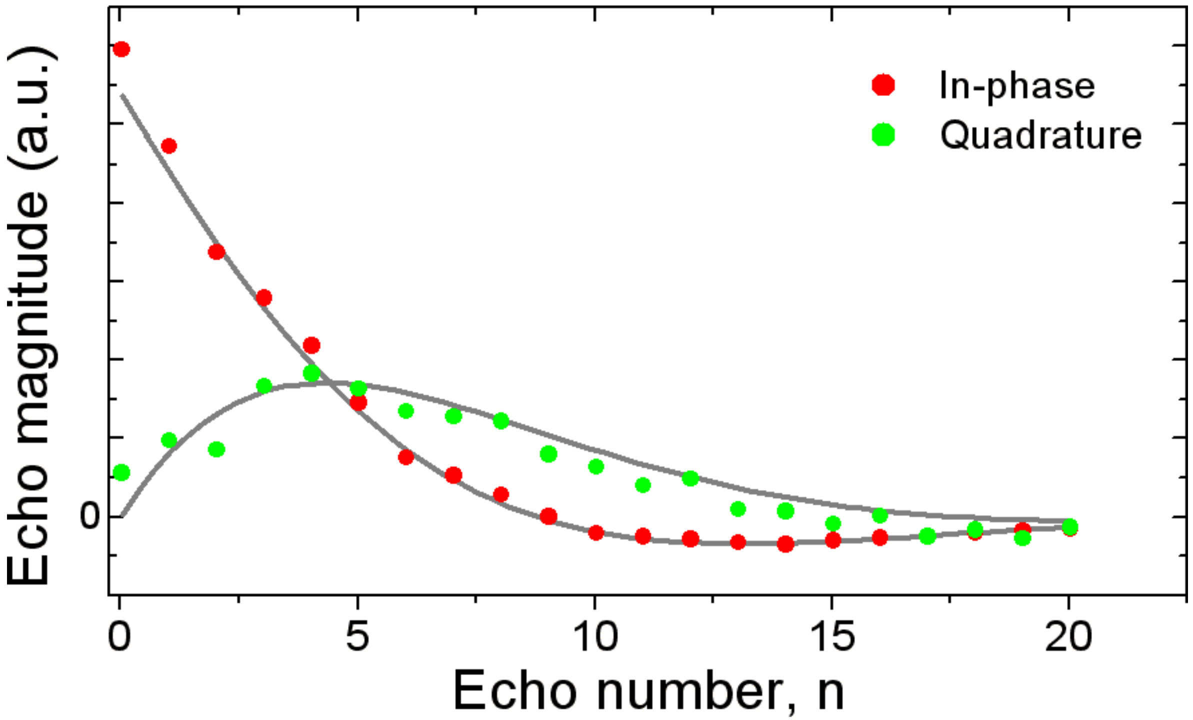
$\pi/2_x - \tau - \pi_y - \tau - \text{echo} - \tau - (\ \pi_x - \tau - \text{echo} - \tau\ - \pi_y - \tau - \text{echo} - \tau)_n$

- accumulates axis error

With ~10 degrees error introduced intentionally:



Testing axis error:
Sequence for Phase-error AMplification



Fitting to

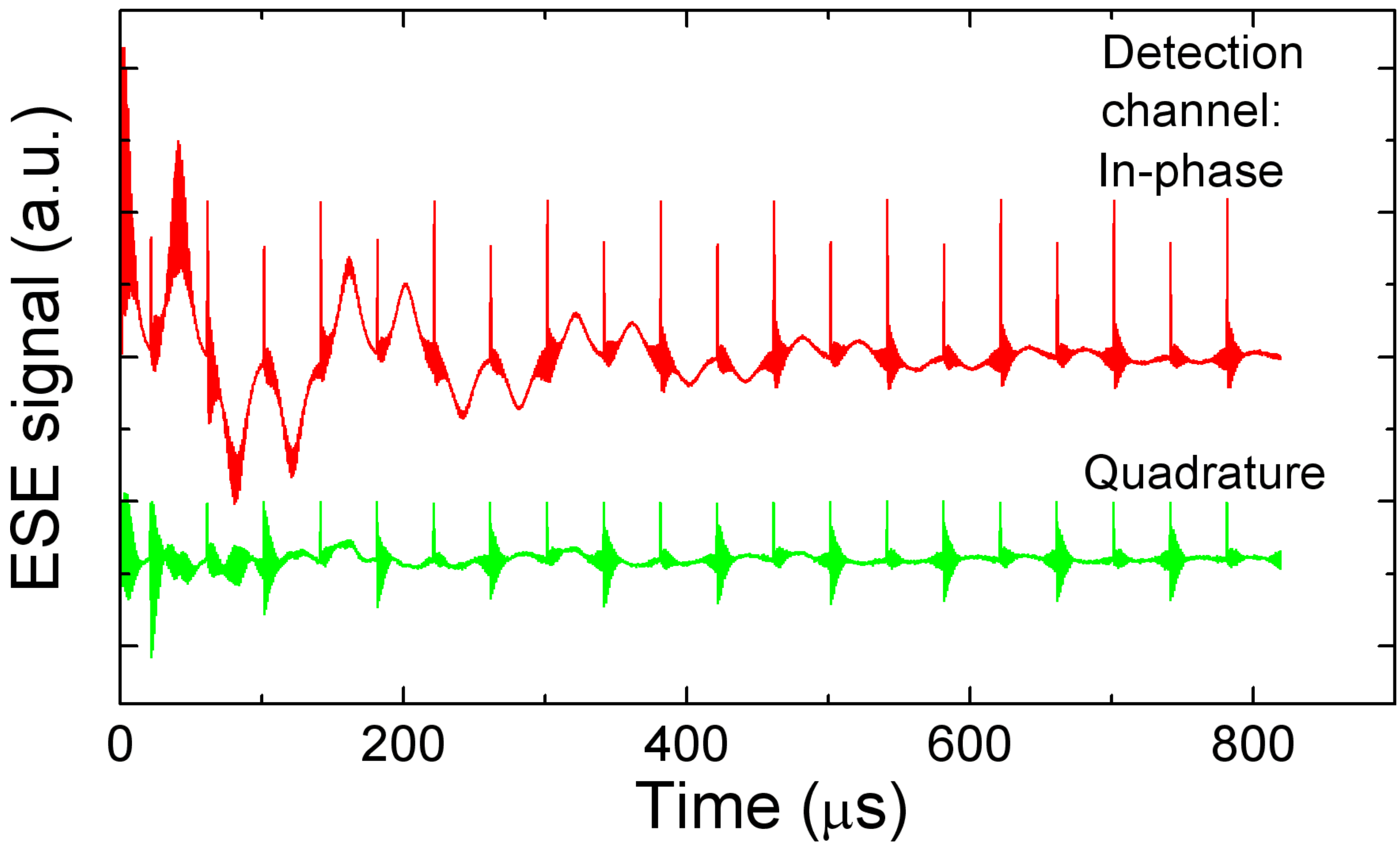
$$A_{In-phase}(n) = A_0 \cos(n\delta) \exp\left(-\frac{2n\tau}{T_2}\right)$$

$$A_{Quadrature}(n) = A_0 \sin(n\delta) \exp\left(-\frac{2n\tau}{T_2}\right)$$

gives δ = 10.3±0.5 degrees

Testing axis error: Sequence for Phase-error AMplification

With optimized phase settings:



$\delta < 0.3$ degrees

Morton *et al.*, Phys Rev A **71** 012332 (2005)

BB1 correction pulses

The situation according to Donald Rumsfeld:

"There are known knowns. These are things we know that we know. There are known unknowns. That is to say, there are things that we know we don't know..."

- Rotation axis (phase) error is small
- Rotation angle error is big

The BB1 composite sequence:

$(1+\epsilon)\pi_{x} - (1+\epsilon)\pi_{104.5°} - (1+\epsilon)2\pi_{313.4°} - (1+\epsilon)\pi_{104.5°}$

is a better π_{x} rotation.

Review of composite correction pulses:
S. Wimperis, J. Magn. Reson. **A 109** 221 (1994)

Brown *et al.*, Phys. Rev. A **72**, 039905 (2005)

BB1 correction sequence

$$\mathcal{R}_{\mathrm{composite}} = \mathcal{R}^{\alpha}_{(1+\epsilon)\pi}\mathcal{R}^{\beta}_{2(1+\epsilon)\pi}\mathcal{R}^{\alpha}_{(1+\epsilon)\pi}\mathcal{R}^{0}_{(1+\epsilon)\theta}$$

Defining the fidelity of two unitary operators *A*, *B*, as

$$F(A,B) = \frac{1}{2}\mathrm{Tr}(A.B^{-1})$$

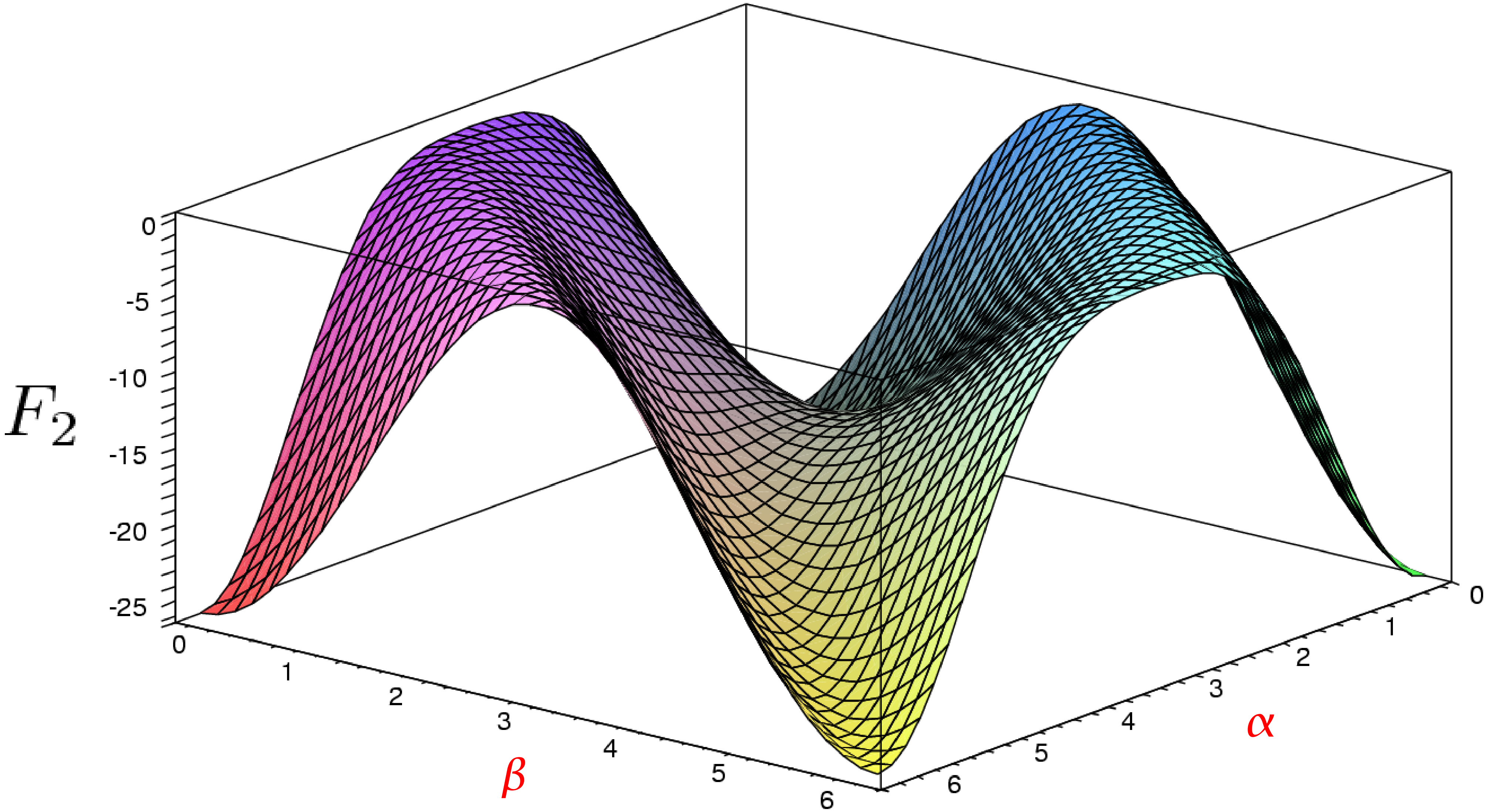
expand fidelity of composite operator in powers of the error,

$$F(\mathcal{R}_{\mathrm{composite}}, \mathcal{R}^{0}_{\theta}) = 1 + F_2\epsilon^2 + F_4\epsilon^4 + \ldots$$

where $F_i = F_i(\theta, \alpha, \beta)$

Choose α and β to kill off leading terms in fidelity expansion.

BB1 correction sequence



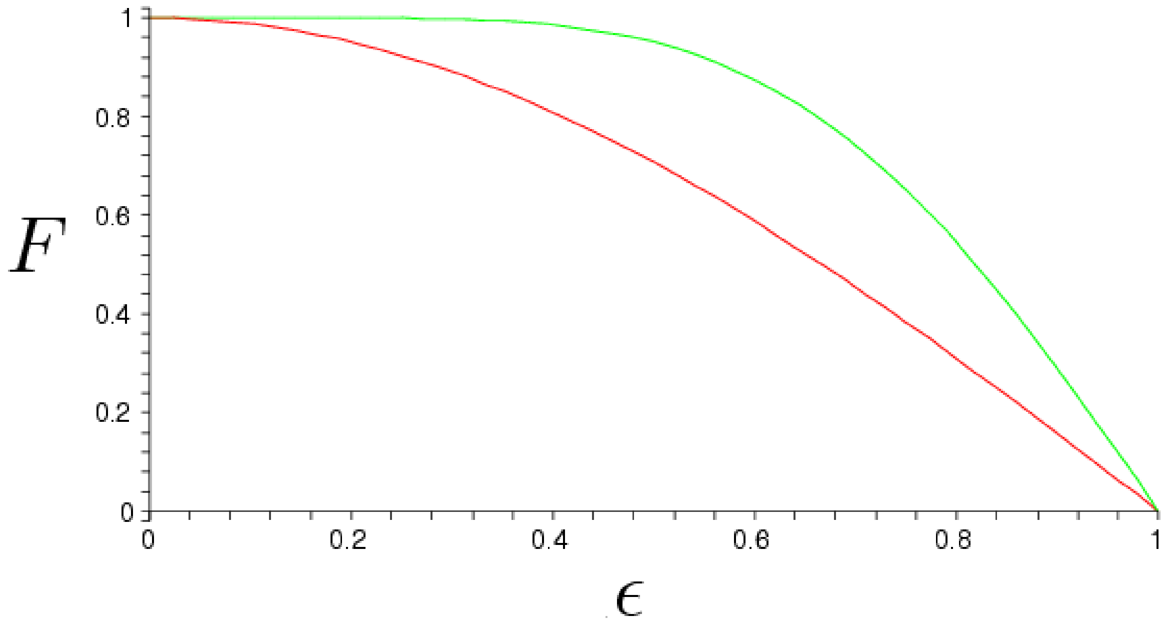
BB1 correction sequence

Fidelity of naïve pulses is

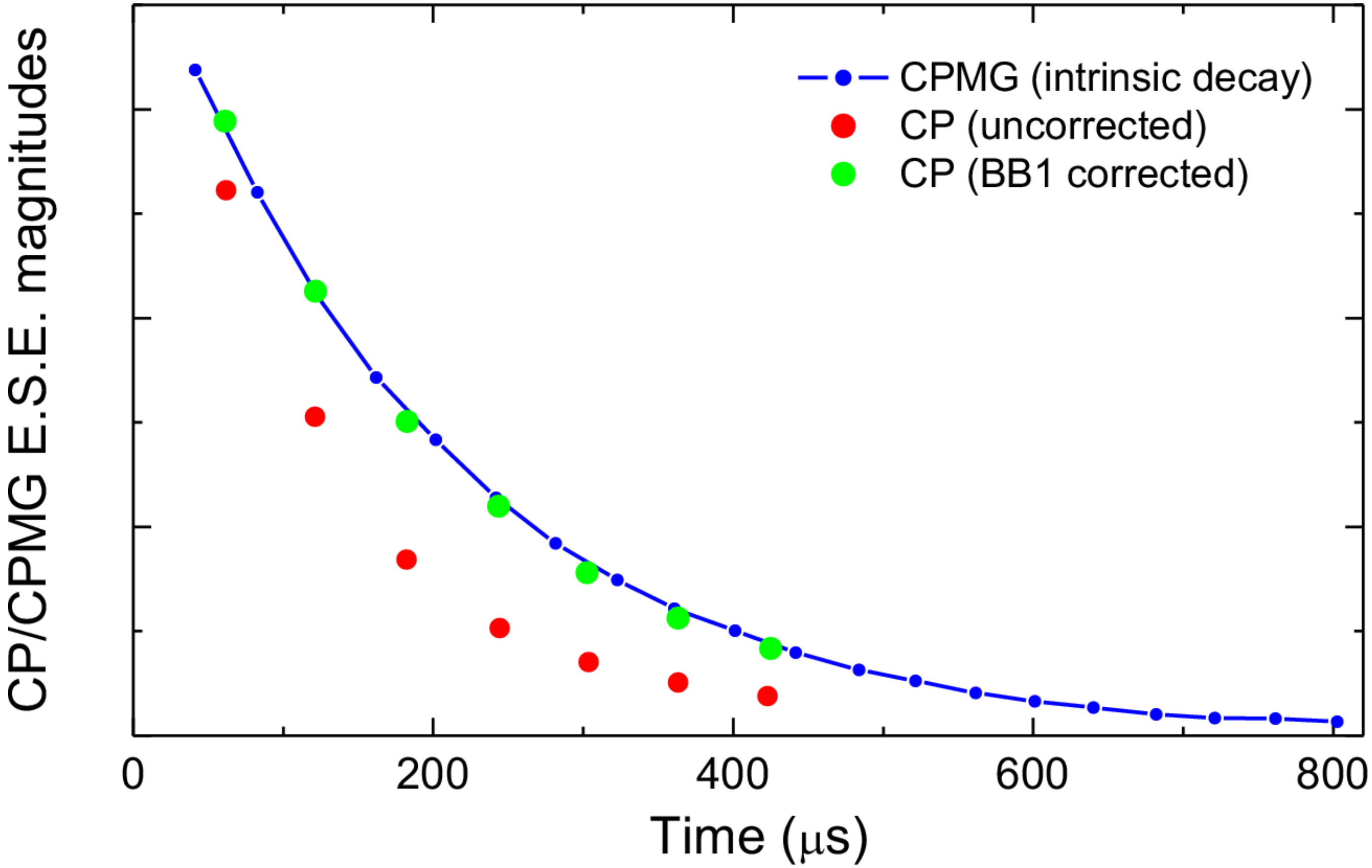
$$F\Big(R, (1+\epsilon)R\Big) - 1 \sim \epsilon^2$$

but fidelity of corrected pulses is

$$F(R, R_{composite}) - 1 \sim \epsilon^6$$



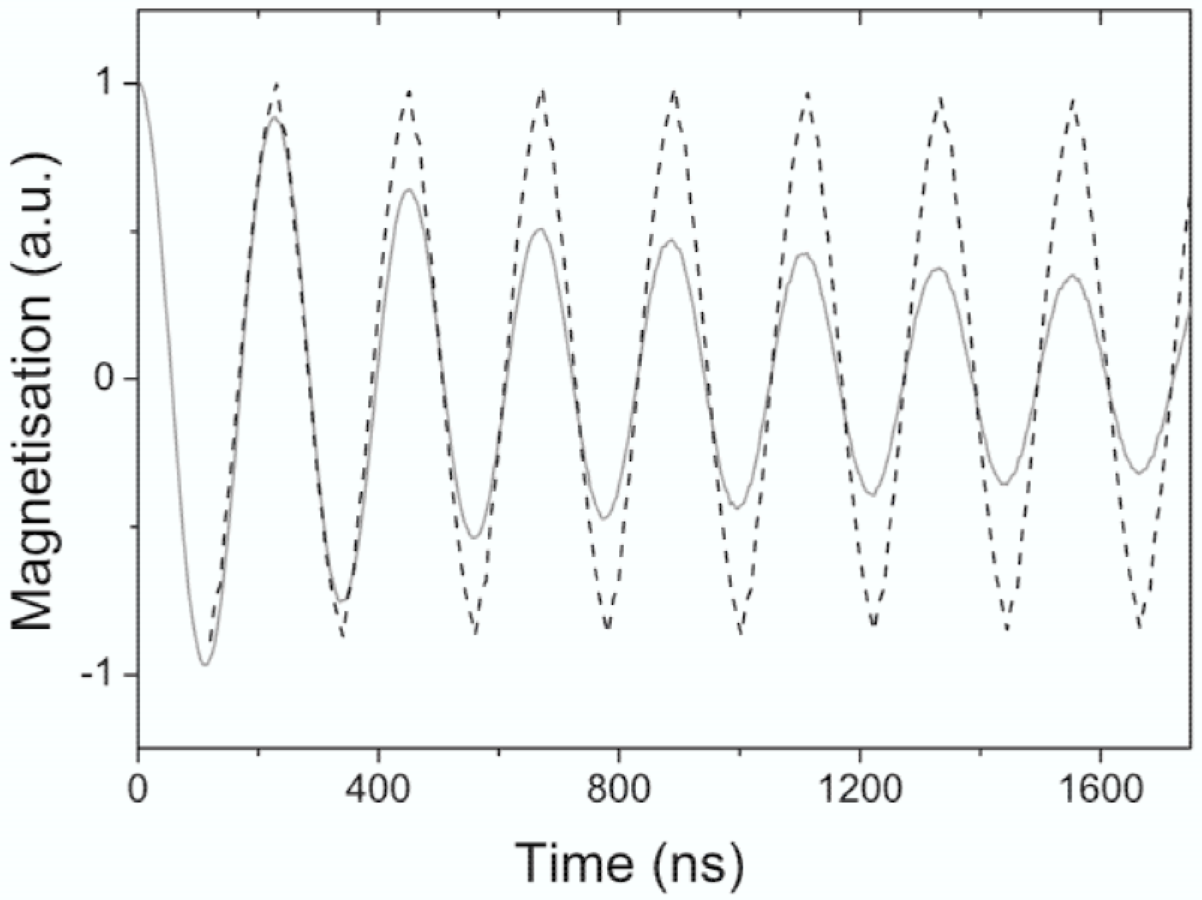
BB1 corrected π – pulses



BB1 corrected π – pulses are indistinguishable from perfect.

Morton *et al.*, Phys. Rev. Lett. **95**, 200501 (2005)

BB1 corrected Rabi oscillations



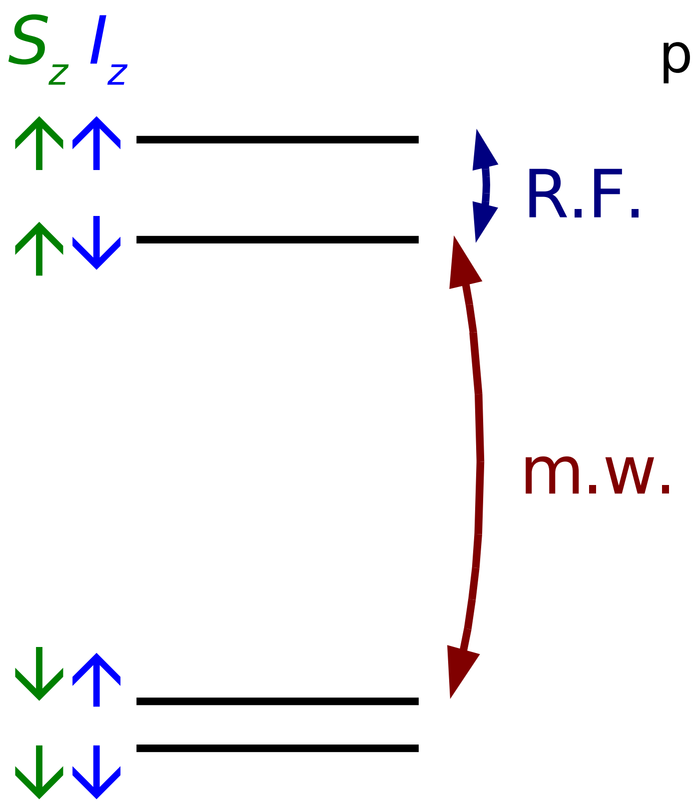
Morton *et al.*, Phys. Rev. Lett. **95**, 200501 (2005)

Application of composite sequences in other contexts

Can we use the nucleus too?

Dynamic nuclear polarization:

$$\mathcal{H} = \omega_e S_z + \omega_n I_z + aS \cdot I \quad \text{and} \quad \hbar\omega_S > k_B T >> \hbar\omega_I$$



population

αp_0

αp_0

αp_0

p_0

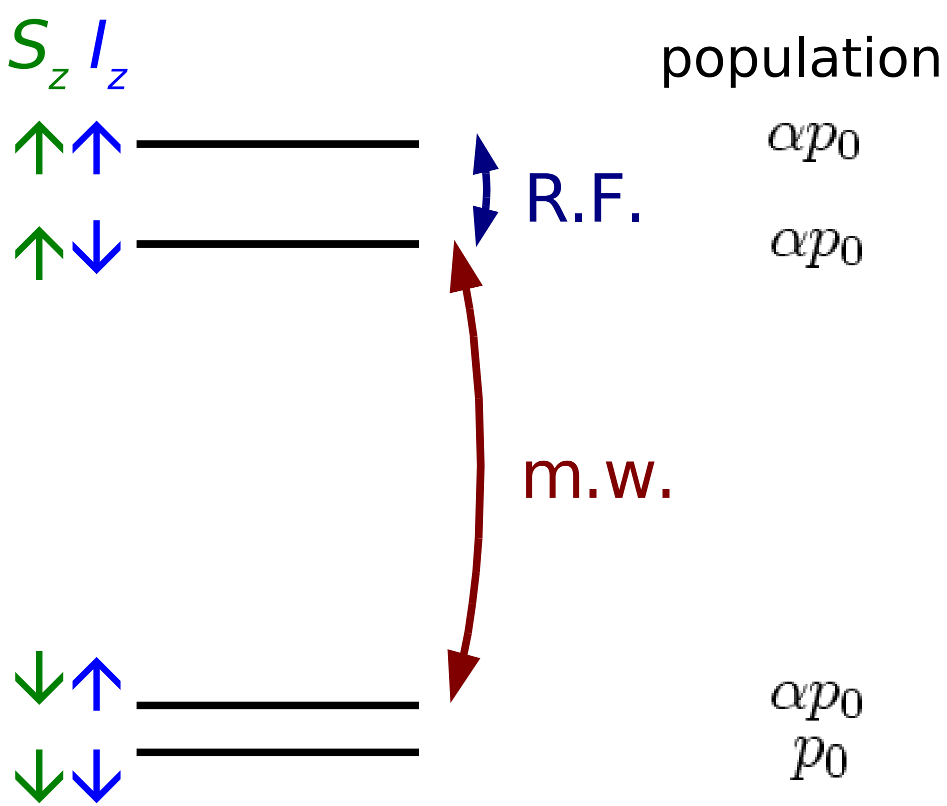
In steady state, with $T_1^I >> T_1^S$,

$$\alpha = \exp\left(-\frac{\hbar\omega_S}{k_B T}\right)$$

Can we use the nucleus too?

Dynamic nuclear polarization:

$$\mathcal{H} = \omega_e S_z + \omega_n I_z + aS \cdot I \quad \text{and} \quad \hbar\omega_S > k_B T >> \hbar\omega_I$$



In steady state, with $T_1^I >> T_1^S$,

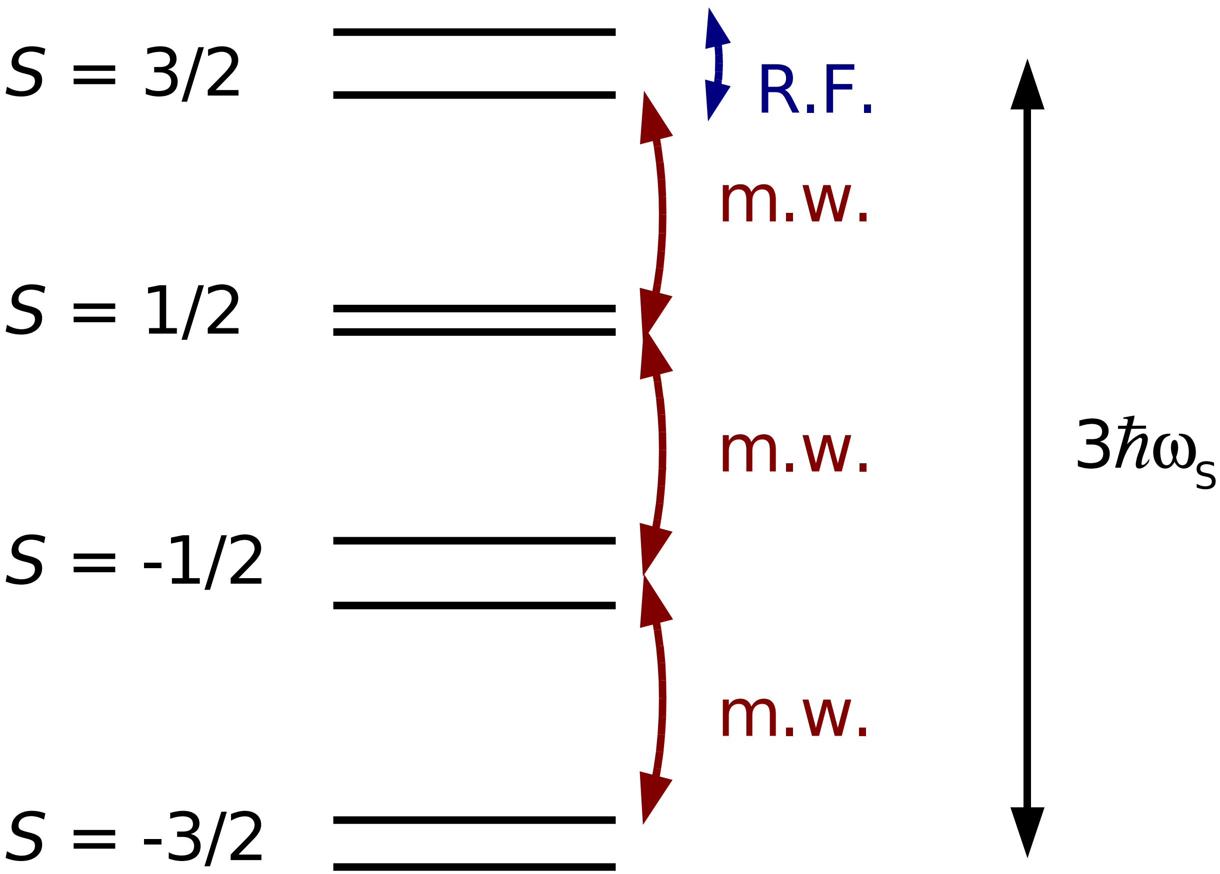
$$p_0 = \frac{1}{1+3\alpha}$$

In w-band (95 GHz) ESR at 0.5 K,

$\alpha = 10^{-4}$, $p_0 = 0.9997$

Can we use the nucleus too?

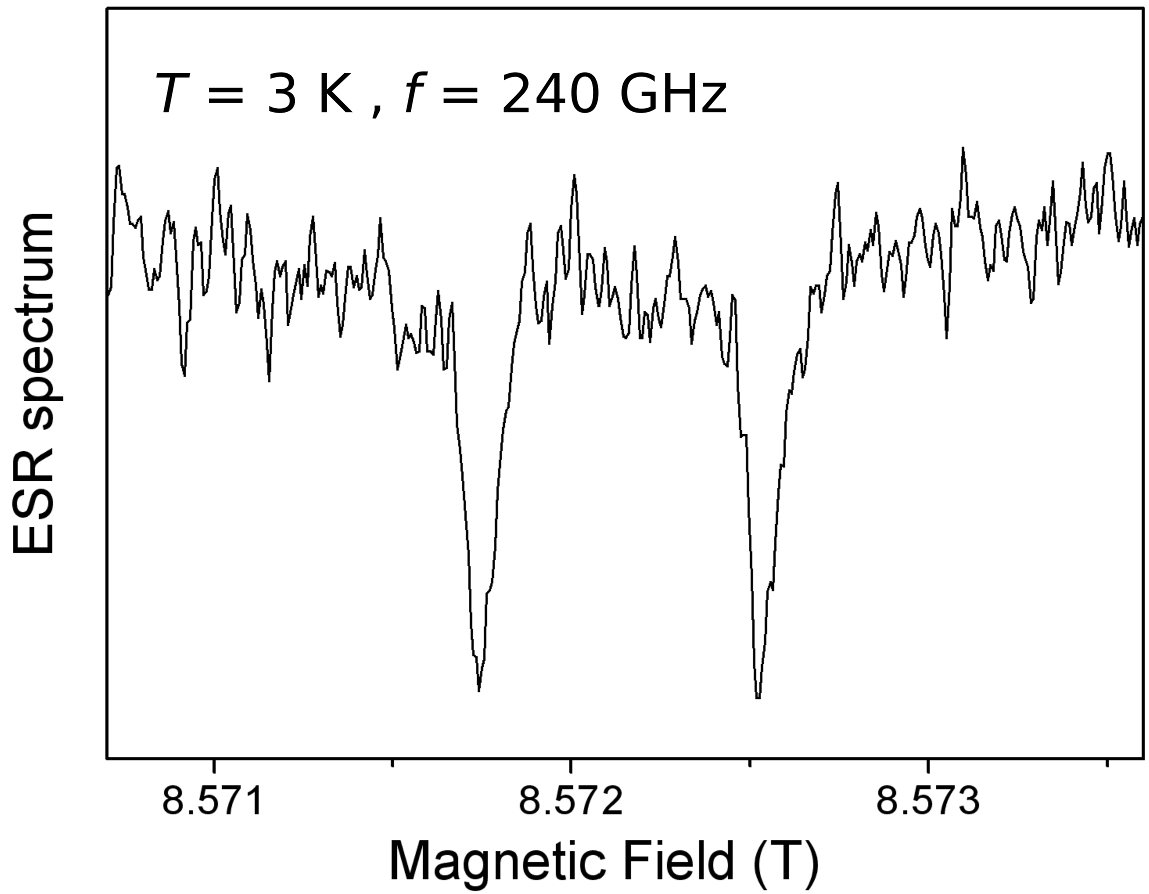
What about an S = 3/2, I=1/2 system like $^{15}N@C_{60}$?



Polarization is more effective than for S = 1/2 case!

Dynamic nuclear polarization in $^{15}N@C_{60}$

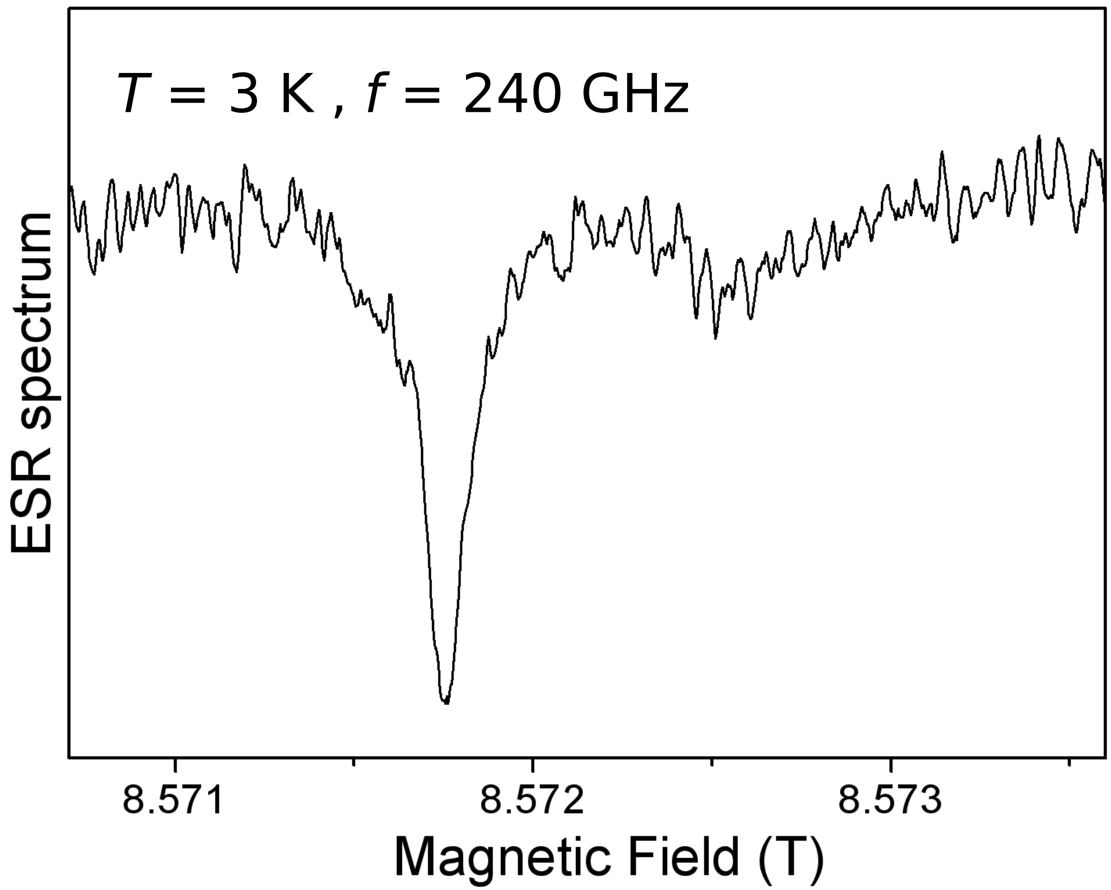
ESR spectrum before preparation:



- Two ^{15}N hyperfine-split lines,
- Lineshape distorted by saturation.

Dynamic nuclear polarization in $^{15}N@C_{60}$

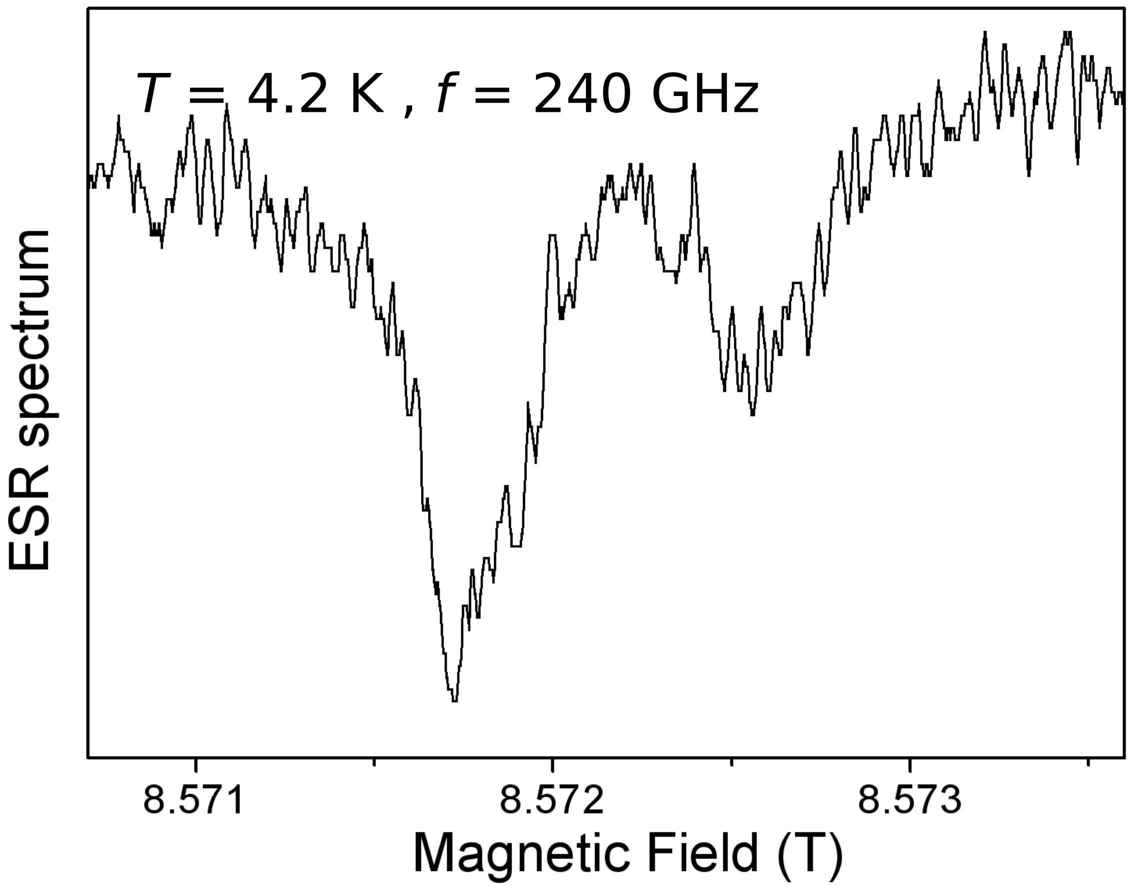
ESR spectrum after 20 minutes preparation:



- Upper ^{15}N hyperfine line strongly suppressed,
- nuclear polarization > 80%

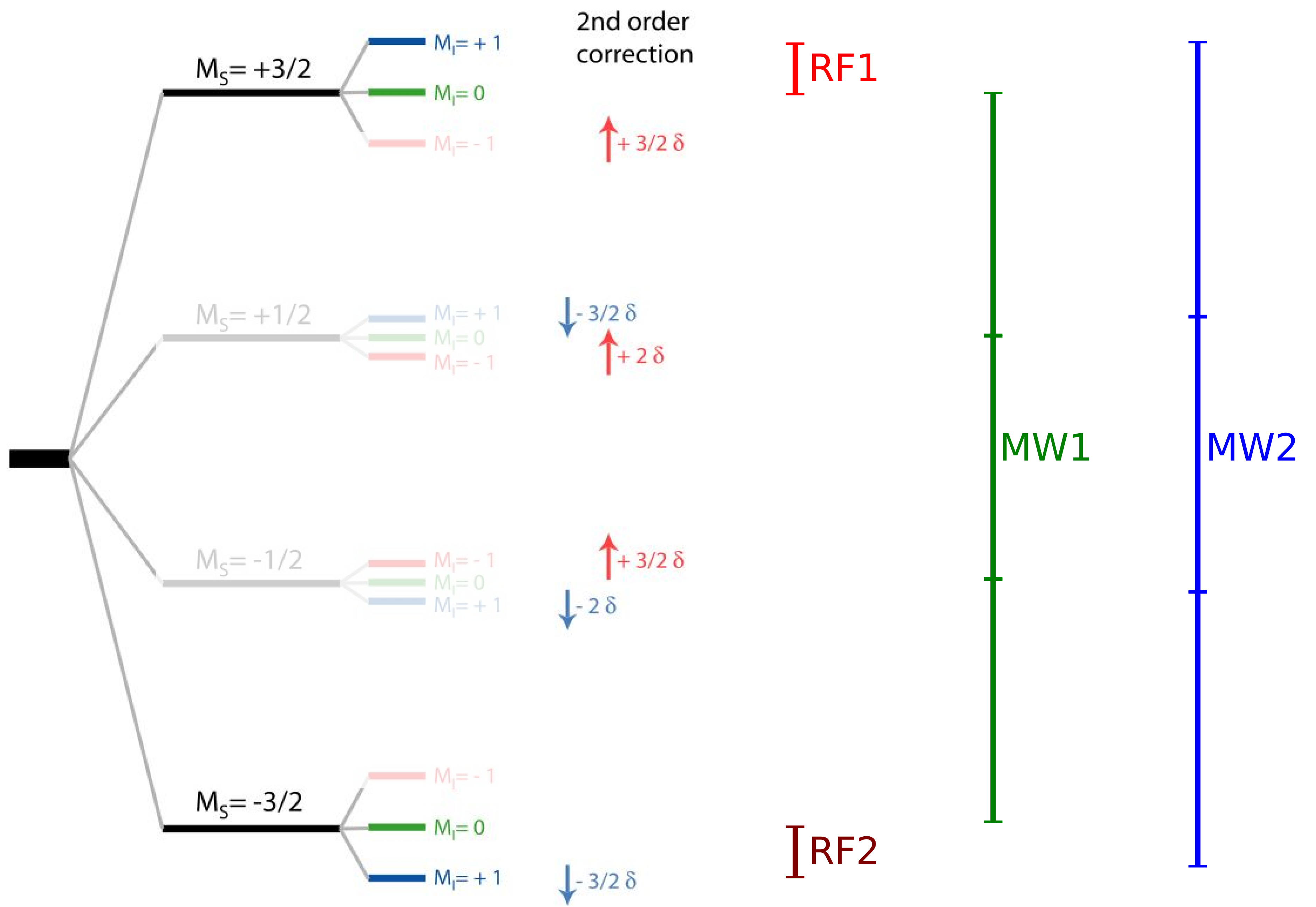
Dynamic nuclear polarization in $^{15}N@C_{60}$

ESR spectrum 11.5 hours later:

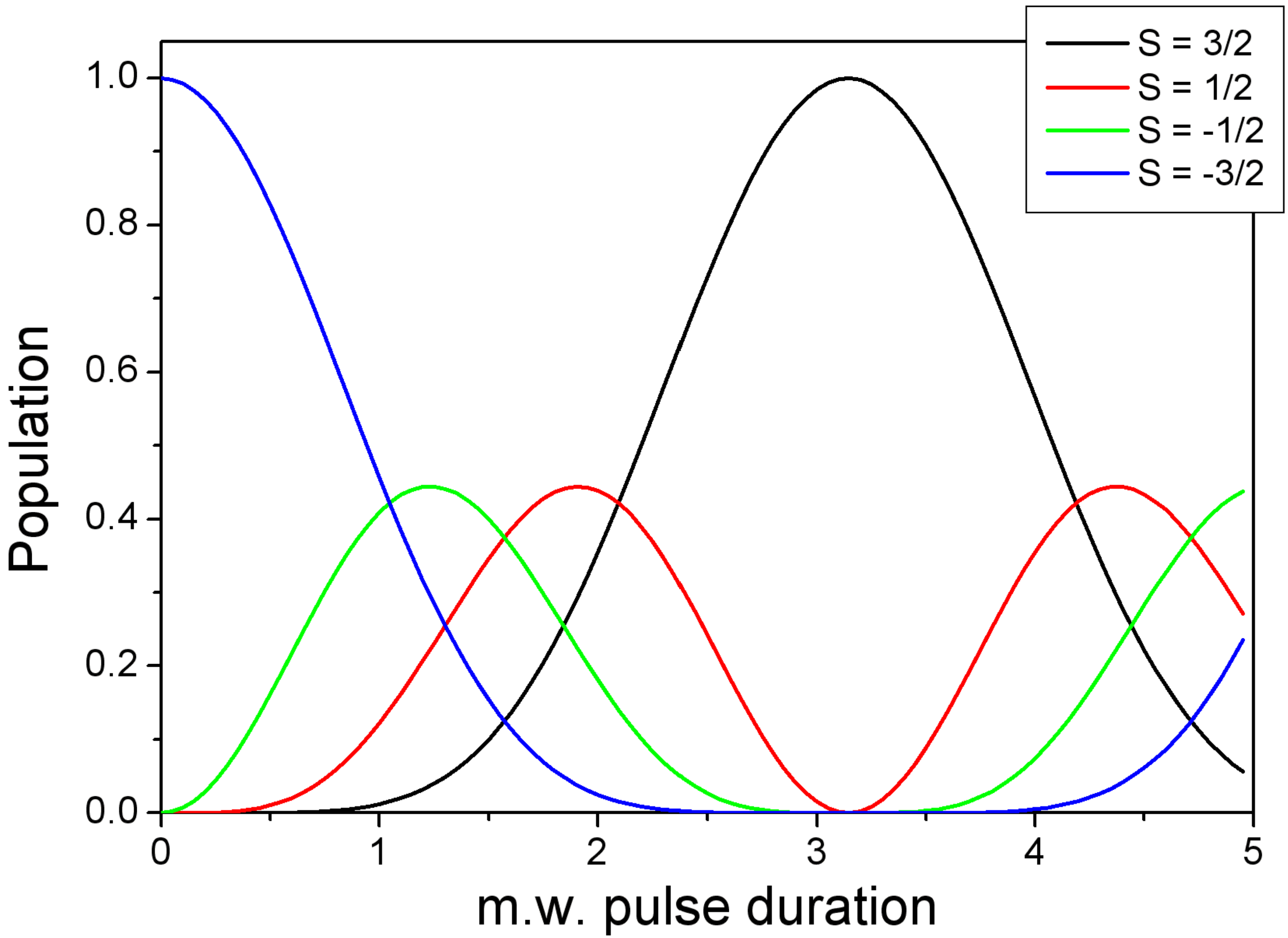


- Nuclear polarization ~ 1/*e*
- T_1^N ~ one day!

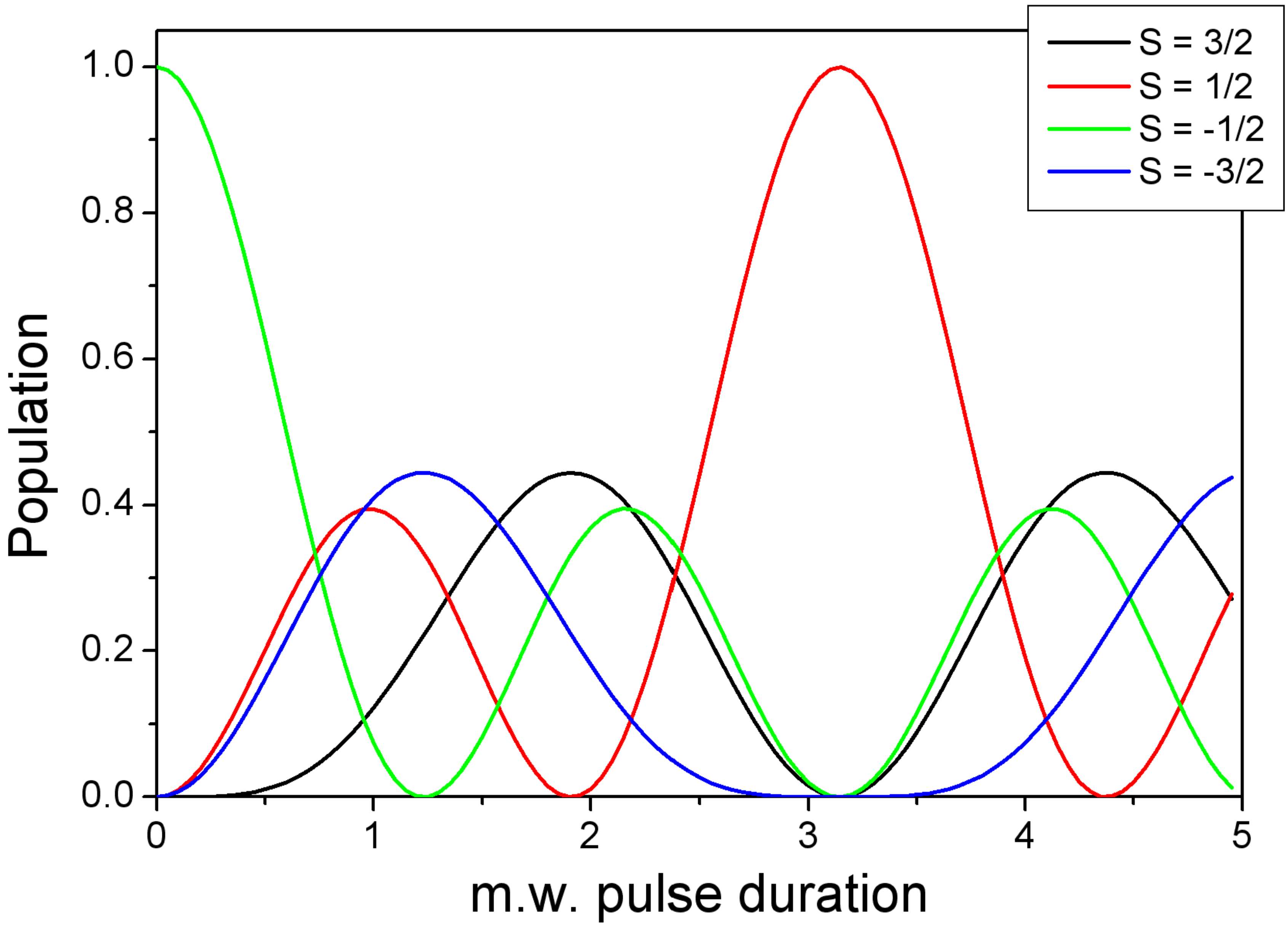
Two qubits on $^{14}N@C_{60}$



π-pulse on $S=3/2$



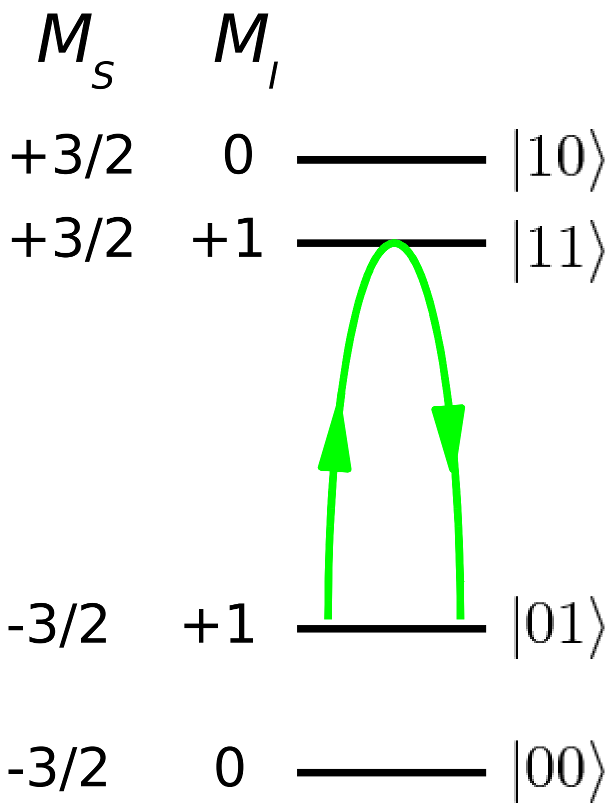
π-pulse on $S=3/2$

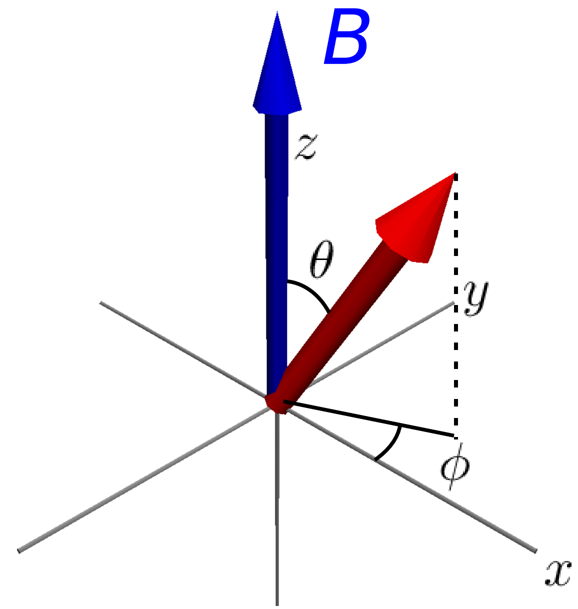


Bang-bang decoupling

What can we do with π-pulses...?

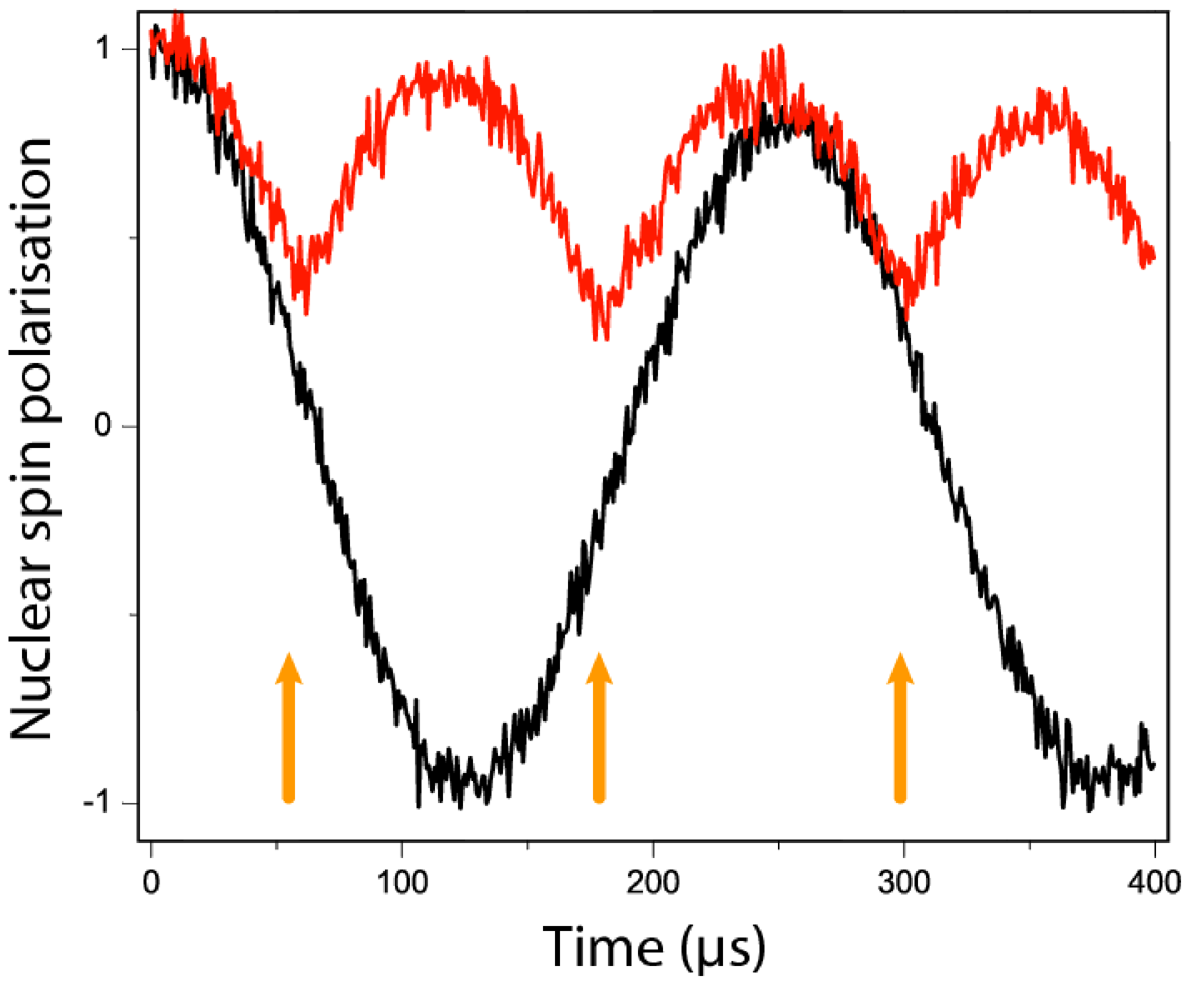
$$\alpha|00\rangle + \beta|01\rangle \longrightarrow \alpha|00\rangle - \beta|01\rangle$$



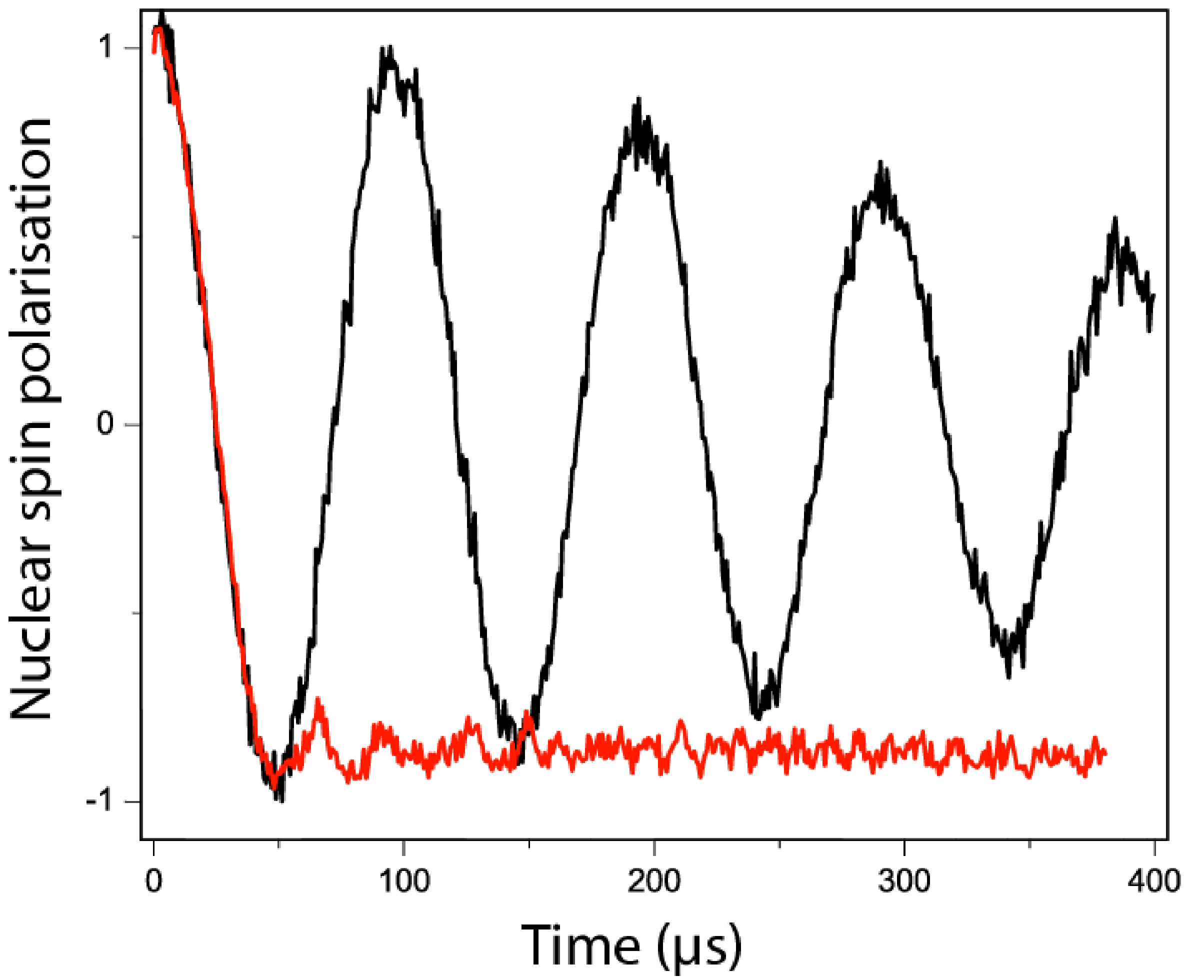


...“tunnel” across Bloch sphere *fast*.

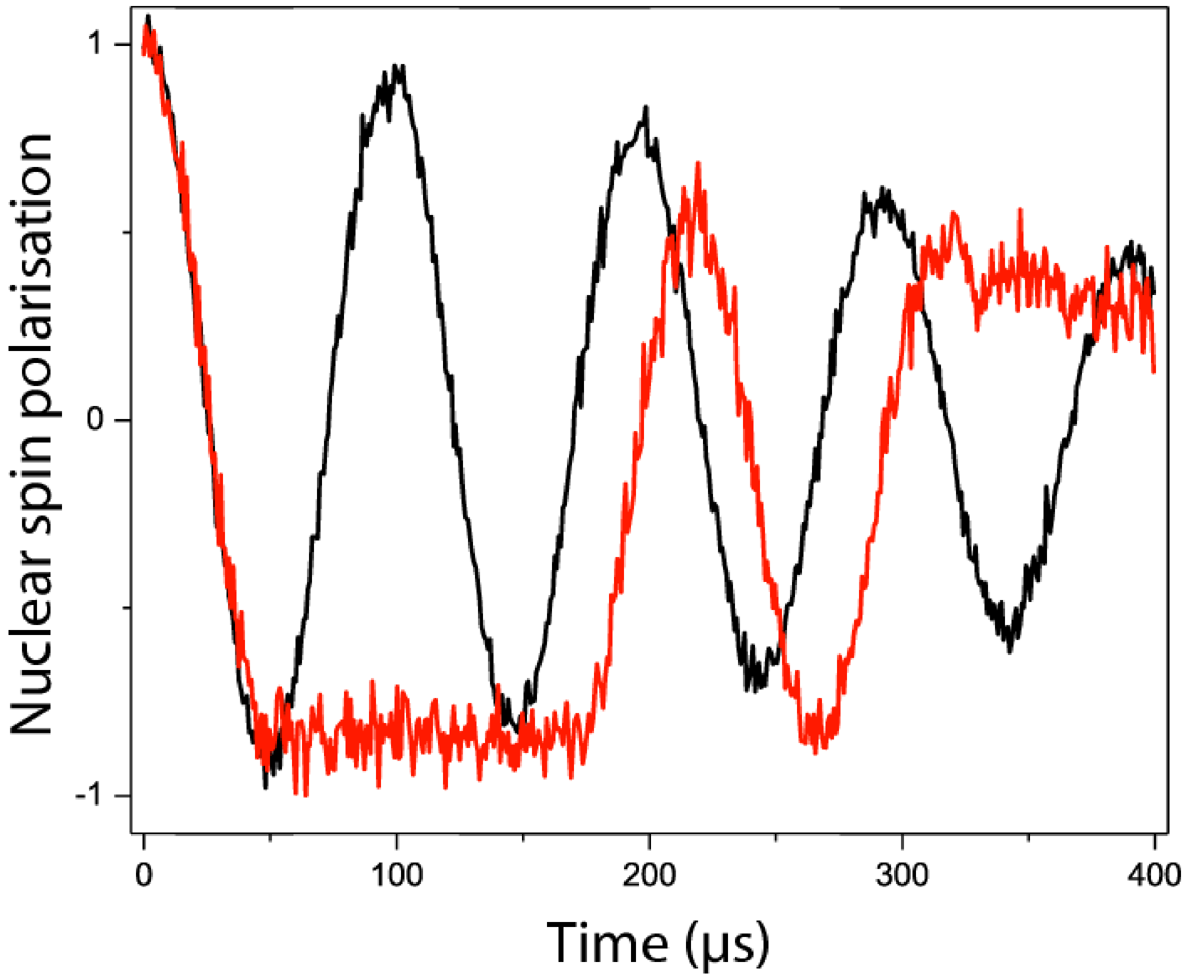
Bang-bang decoupling



Bang-bang decoupling



Bang-bang decoupling



Details of pulse sequences used

Morton *et al*., Nature Physics **2** 40 (2006)

Summary

- ESR can be used for qubit-manipulation

- N@C_{60} shows promise as a building-block for QIP devices
  - long lifetimes
  - electron and nuclear degrees of freedom

- *Technical aspects of a pulsed magnetic resonance spectrometer*
- *Application of composite sequences in other contexts*
- *Details of pulse sequences used in bang-bang experiment*