hhu.

26th International Symposium on Spin Physics

A Century of Spin

Walter Benjamin-Programm

DFG Deutsche Forschungsgemeinschaft

Spin manipulation in atomic and molecular systems for nuclear spin applications

Chrysovalantis Kannis

with

Ralf Engels, Tarek El-Kordy, Nicolas Faatz, Simon J. Pütz, Vincent Verhoeven, T. Peter Rakitzis, and Markus Büscher

JÜLICH Forschungszentrum

GSI

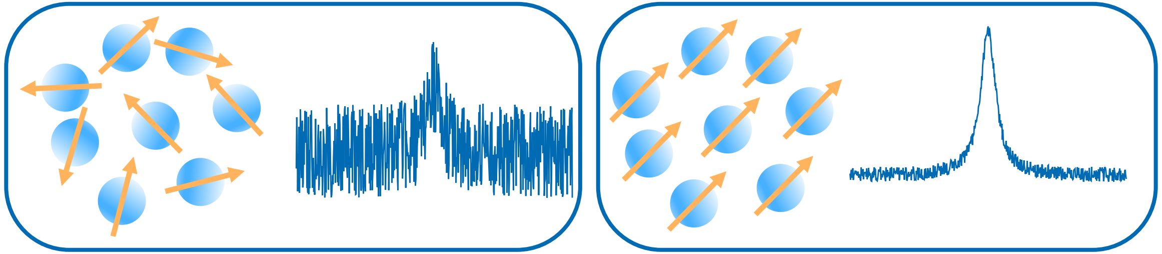
FORTH INSTITUTE OF ELECTRONIC STRUCTURE AND LASER

23.09.2025

Why nuclear spin polarization is important

Heinrich Heine Universität Düsseldorf

- Physics and medicine
  - NMR/MRI signal enhancement

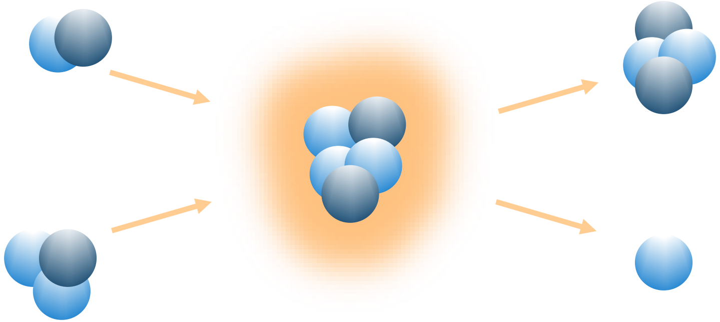


- Polarized fusion
  - Five-nucleon reactions
    - $\mathrm{d} + \mathrm{t} \longrightarrow \alpha\,(3.5\ \mathrm{MeV}) + \mathrm{n}\,(14.1\ \mathrm{MeV})$
    - $\mathrm{d} + \mathrm{h} \longrightarrow \alpha\,(3.6\ \mathrm{MeV}) + \mathrm{p}\,(14.7\ \mathrm{MeV})$
  - Four-nucleon reactions
    - $\mathrm{d} + \mathrm{d} \xrightarrow{50\%} \mathrm{t}\,(1.01\ \mathrm{MeV}) + \mathrm{p}\,(3.02\ \mathrm{MeV})$
    - $\mathrm{d} + \mathrm{d} \xrightarrow{50\%} \mathrm{h}\,(0.82\ \mathrm{MeV}) + \mathrm{n}\,(2.45\ \mathrm{MeV})$
  - ► Ralf Engels-Polarized Fusion (Plenary talk, Thu 25/09, 8:30)

Five-nucleon reactions:

i. increase in reaction rate
ii. control of the emission direction of products

Four-nucleon reactions: ?



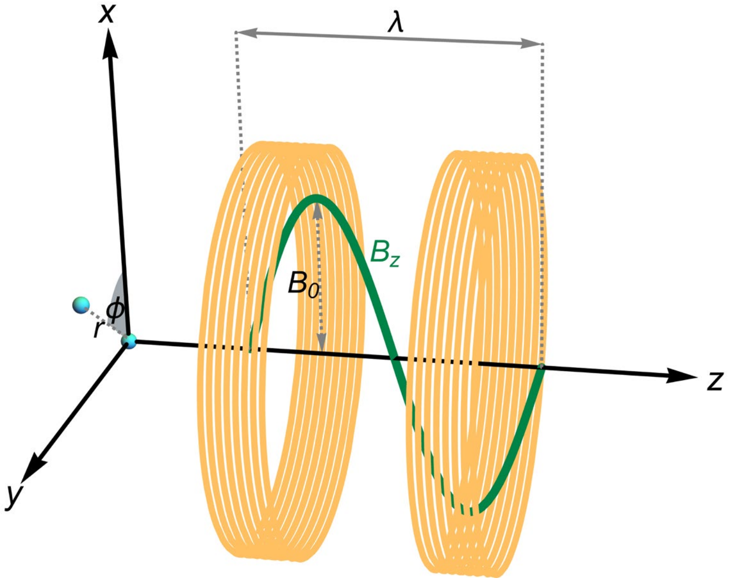
Key concepts

Heinrich Heine Universität Düsseldorf

- Spin and angular momentum in bound systems
  - Atomic states: nuclear spin I, electron spin S (with $L = 0$)
  - Molecular states: nuclear spins I, rotational angular momentum J (with $S = 0$)
- Hyperfine regime
  - Interactions between the nuclear spins and residual angular momenta: H_0
  - Spin states
    - Uncoupled basis: $|m_S, m_I\rangle$, $|m_J, m_{I_1}, m_{I_2}\rangle$
    - Coupled basis: $|F, m_F\rangle$ with $\boldsymbol{F} = \boldsymbol{S} \otimes \mathbb{1} + \mathbb{1} \otimes \boldsymbol{I}$ or $|I_t, F, m_F\rangle$ $\boldsymbol{F} = \boldsymbol{J} \otimes \mathbb{1} + \mathbb{1} \otimes \boldsymbol{I_t}$ and $\boldsymbol{I_t} = \boldsymbol{I_1} \otimes \mathbb{1} + \mathbb{1} \otimes \boldsymbol{I_2}$
- Density operator formalism
  - Ensemble of nonidentically prepared atoms or molecules→statistical mixture of states
  - Density matrix: diagonal elements→populations, off-diagonal elements→coherences
  - Liouville-von Neumann equation: $i\hbar \frac{\partial \rho}{\partial t} = [H, \rho]$
  - Average of observable O: $\langle O \rangle = Tr(\rho O)$

Key concepts

- External perturbations
  - Interaction with a magnetic field $\boldsymbol{B}$: $H_B = -\boldsymbol{\mu} \cdot \boldsymbol{B}$, with total Hamiltonian $H = H_0 + H_B$
  - Sinusoidal field, inspired by P. G. Sona [Energ. Nucl. **14**, 295 (1967)]



$$\boldsymbol{B} = B_z\, \hat{\boldsymbol{z}} + B_r\, \hat{\boldsymbol{r}}, \text{ with } B_r = -\frac{r}{2}\frac{\partial B_z}{\partial z}$$

- B_z : static longitudinal component
- Particle motion: along z with constant, nonrelativistic velocity v

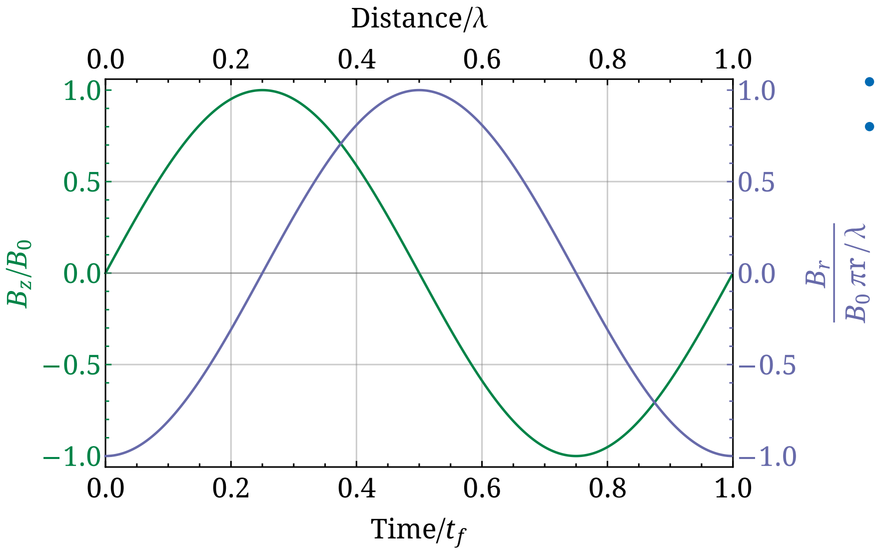
Laboratory frame: $\boldsymbol{B} = B_0 \sin\left(\frac{2\pi z}{\lambda}\right) \hat{\boldsymbol{z}} - B_0 \frac{\pi r}{\lambda} \cos\left(\frac{2\pi z}{\lambda}\right) \hat{\boldsymbol{r}}$

Particle rest frame: $\boldsymbol{B} = B_0 \sin\left(\frac{2\pi v t}{\lambda}\right) \hat{\boldsymbol{z}} - B_0 \frac{\pi r}{\lambda} \cos\left(\frac{2\pi v t}{\lambda}\right) \hat{\boldsymbol{r}}$

C. S. Kannis *et al.*, Phys. Rev. A **112**, 012801 (2025)

Key concepts

- External perturbations
  - Interaction with a magnetic field $\boldsymbol{B}$: $H_B = -\boldsymbol{\mu} \cdot \boldsymbol{B}$, with total Hamiltonian $H = H_0 + H_B$
  - Sinusoidal field, inspired by P. G. Sona [Energ. Nucl. **14**, 295 (1967)]



$$\boldsymbol{B} = B_z\,\hat{\boldsymbol{z}} + B_r\,\hat{\boldsymbol{r}}, \text{ with } B_r = -\frac{r}{2}\frac{\partial B_z}{\partial z}$$

- B_z : static longitudinal component
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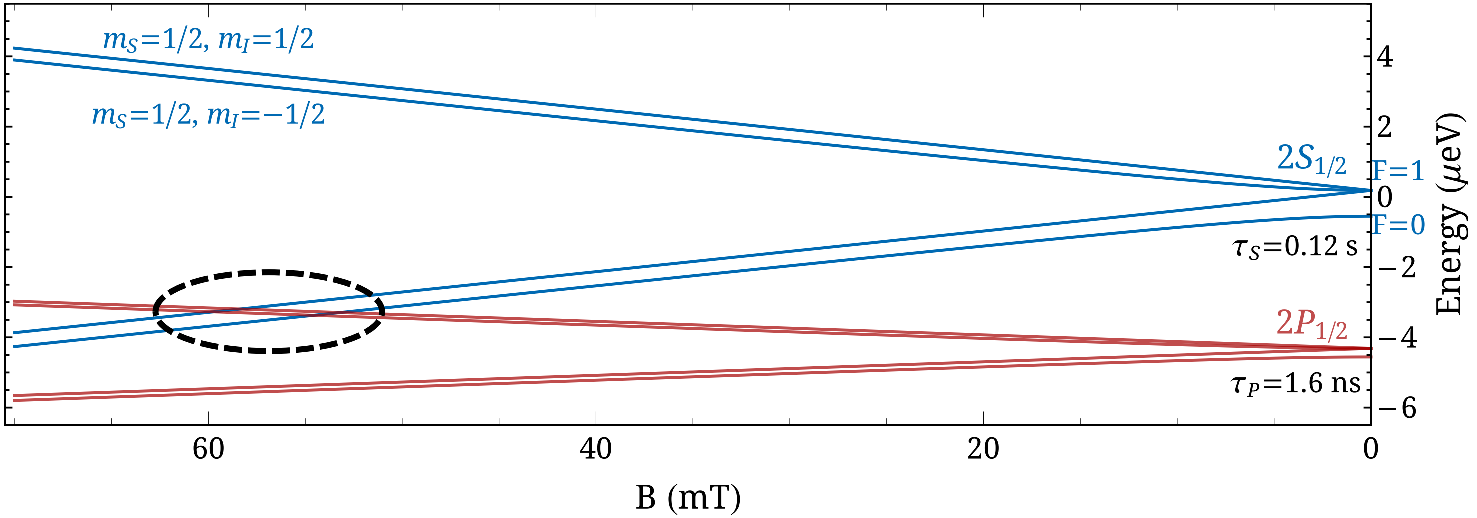
Particle rest frame: $\boldsymbol{B} = B_0 \sin\left(\frac{2\pi v t}{\lambda}\right)\hat{\boldsymbol{z}} - B_0\frac{\pi r}{\lambda}\cos\left(\frac{2\pi v t}{\lambda}\right)\hat{\boldsymbol{r}}$

$r \ll \lambda$

C. S. Kannis *et al.*, Phys. Rev. A **112**, 012801 (2025)

Incoherent initial preparation

- Hydrogen atom
  - Ground state: protons capture polarized electrons in an optically pumped, polarized Rb vapor cell (in high magnetic field)
  - Metastable state: use of the $2S_{1/2} - 2P_{1/2}$ level crossing (also at high magnetic field)



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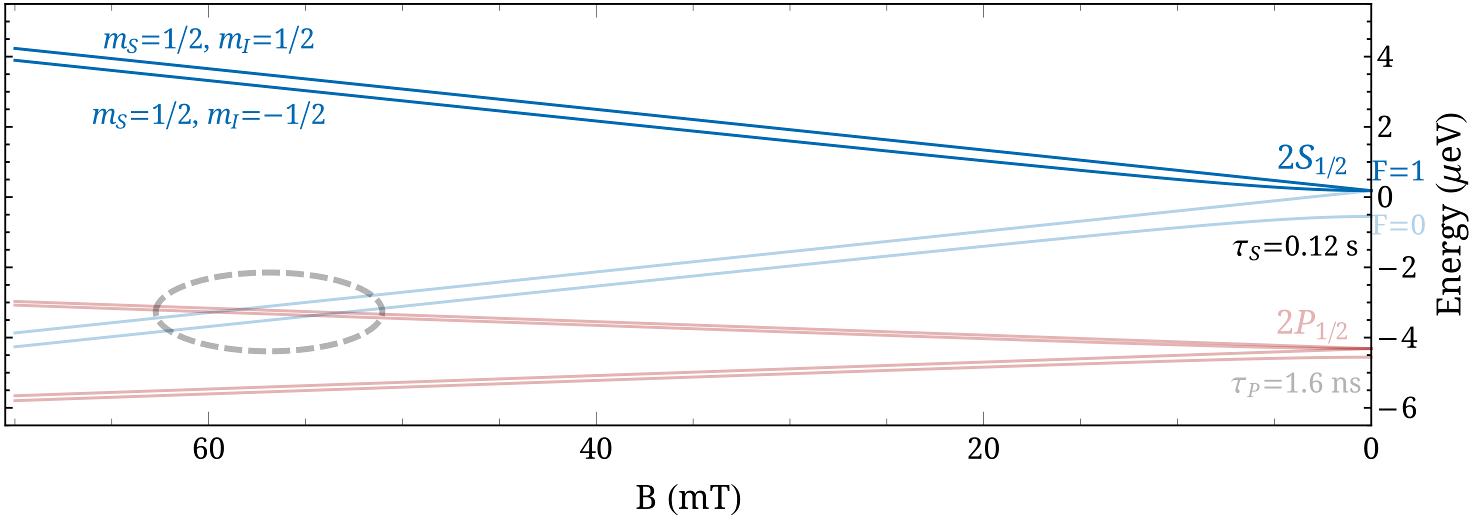


Incoherent initial preparation

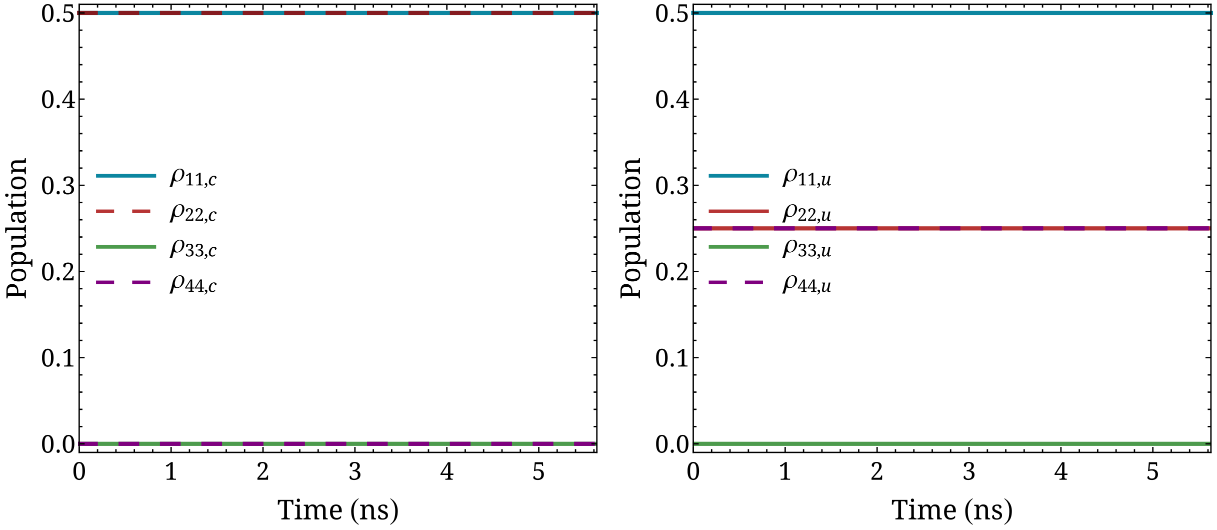
- Hydrogen atom

$$\rho_c = \begin{pmatrix} 1/2 & 0 & 0 & 0 \\ 0 & 1/2 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}$$

	coupled basis $|F, m_F\rangle$	uncoupled basis $|m_S, m_I\rangle$
1.	$|1,1\rangle$	$|1/2, 1/2\rangle$
2.	$|1,0\rangle$	$|1/2, -1/2\rangle$
3.	$|1,-1\rangle$	$|-1/2, -1/2\rangle$
4.	$|0,0\rangle$	$|-1/2, 1/2\rangle$

$H_0 = \frac{A_{2S}}{\hbar^2} \boldsymbol{I} \cdot \boldsymbol{S}$ with $\frac{A_{2S}}{h} = 177.6$ MHz

$t_f = \frac{h}{\Delta E_{hfs}} = \frac{h}{A_{2S}} \sim 5.6$ ns



Incoherent initial preparation

- Hydrogen atom

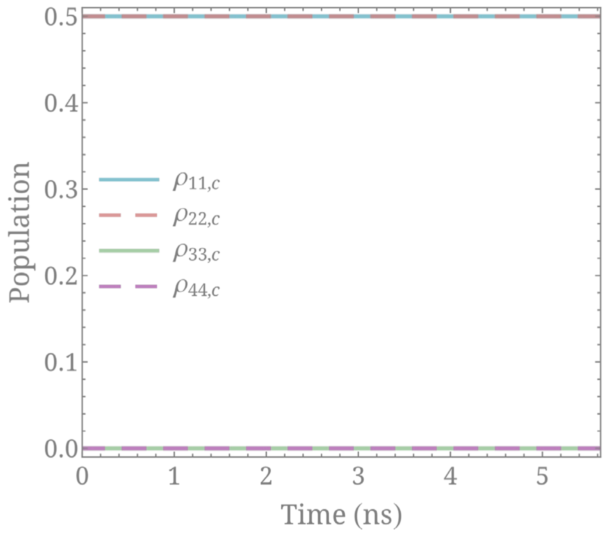
$$\rho_c = \begin{pmatrix} 1/2 & 0 & 0 & 0 \\ 0 & 1/2 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}$$

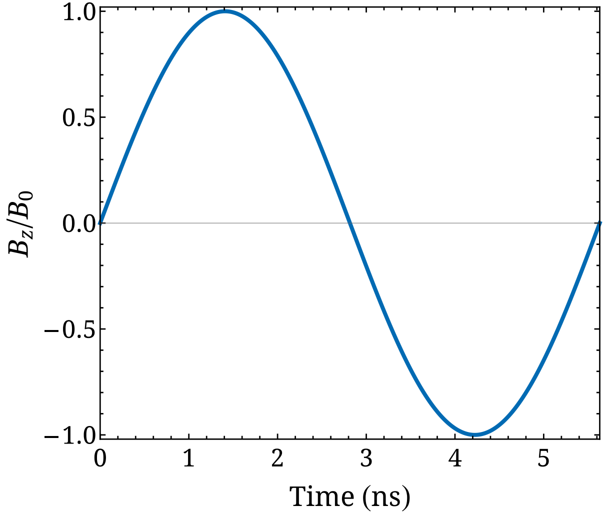
	coupled basis $\lvert F, m_F \rangle$	uncoupled basis $\lvert m_S, m_I \rangle$
1.	$\lvert 1,1 \rangle$	$\lvert 1/2, 1/2 \rangle$
2.	$\lvert 1,0 \rangle$	$\lvert 1/2, -1/2 \rangle$
3.	$\lvert 1,-1 \rangle$	$\lvert -1/2, -1/2 \rangle$
4.	$\lvert 0,0 \rangle$	$\lvert -1/2, 1/2 \rangle$

- How is population evolution affected by $B_z = B_0 \sin\left(\frac{2\pi v t}{\lambda}\right)$?

$$H_0 = \frac{A_{2S}}{\hbar^2} \boldsymbol{I} \cdot \boldsymbol{S} \quad \text{with} \quad \frac{A_{2S}}{h} = 177.6 \text{ MHz} \qquad t_f = \frac{h}{\Delta E_{hfs}} = \frac{h}{A_{2S}} \sim 5.6 \text{ ns}$$

$$\lambda = v\, t_f$$





Incoherent initial preparation

- Hydrogen atom

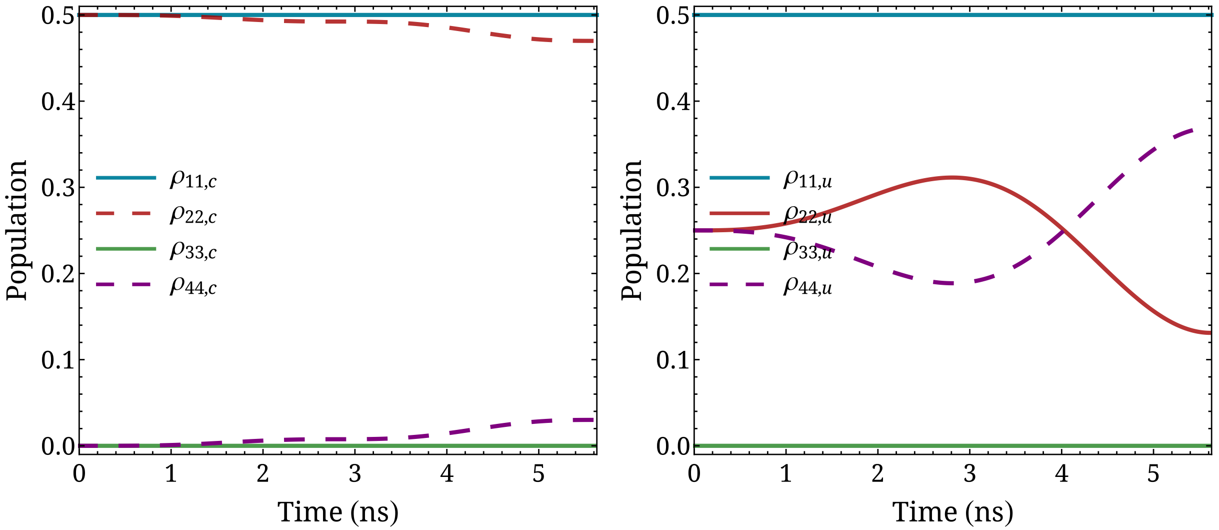
$$\rho_c = \begin{pmatrix} 1/2 & 0 & 0 & 0 \\ 0 & 1/2 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}$$

	coupled basis $\lvert F, m_F \rangle$	uncoupled basis $\lvert m_S, m_I \rangle$
1.	$\lvert 1, 1 \rangle$	$\lvert 1/2, 1/2 \rangle$
2.	$\lvert 1, 0 \rangle$	$\lvert 1/2, -1/2 \rangle$
3.	$\lvert 1, -1 \rangle$	$\lvert -1/2, -1/2 \rangle$
4.	$\lvert 0, 0 \rangle$	$\lvert -1/2, 1/2 \rangle$

➢ How is population evolution affected by $B_z = B_0 \sin\left(\frac{2\pi v t}{\lambda}\right)$?

$$H = H_0 + H_B = \frac{A_{2S}}{\hbar^2} \boldsymbol{I} \cdot \boldsymbol{S} - \left(\frac{g_S \mu_B}{\hbar} \boldsymbol{S} + \frac{g_I \mu_N}{\hbar} \boldsymbol{I}\right) \cdot \boldsymbol{B}$$

$B_0 = 1$ mT



C. Kannis *et al.*

Incoherent initial preparation

Heinrich Heine Universität Düsseldorf

- Hydrogen atom

$$\rho_c = \begin{pmatrix} 1/2 & 0 & 0 & 0 \\ 0 & 1/2 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}$$

	coupled basis $\|F, m_F\rangle$	uncoupled basis $\|m_S, m_I\rangle$
1.	$\|1,1\rangle$	$\|1/2, 1/2\rangle$
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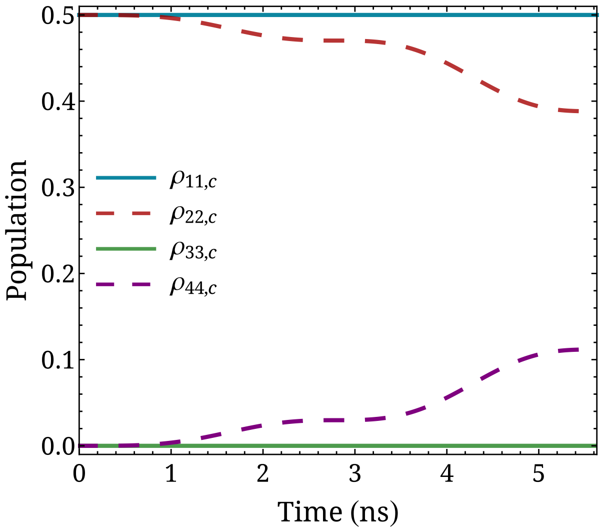
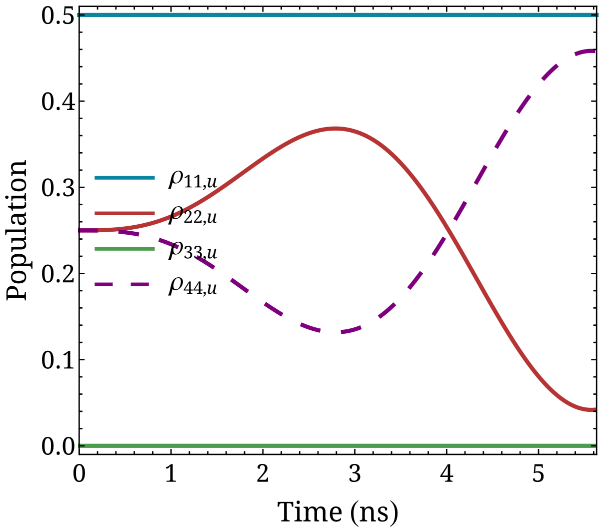
➢ How is population evolution affected by $B_z = B_0 \sin\left(\frac{2\pi v t}{\lambda}\right)$?

$$H = H_0 + H_B = \frac{A_{2S}}{\hbar^2} \boldsymbol{I} \cdot \boldsymbol{S} - \left(\frac{g_S \mu_B}{\hbar} \boldsymbol{S} + \frac{g_I \mu_N}{\hbar} \boldsymbol{I}\right) \cdot \boldsymbol{B}$$

$B_0 = 2$ mT





Incoherent initial preparation

- Hydrogen atom

$$\rho_c = \begin{pmatrix} 1/2 & 0 & 0 & 0 \\ 0 & 1/2 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}$$

	coupled basis $\lvert F, m_F\rangle$	uncoupled basis $\lvert m_S, m_I\rangle$
1.	$\lvert 1,1\rangle$	$\lvert 1/2, 1/2\rangle$
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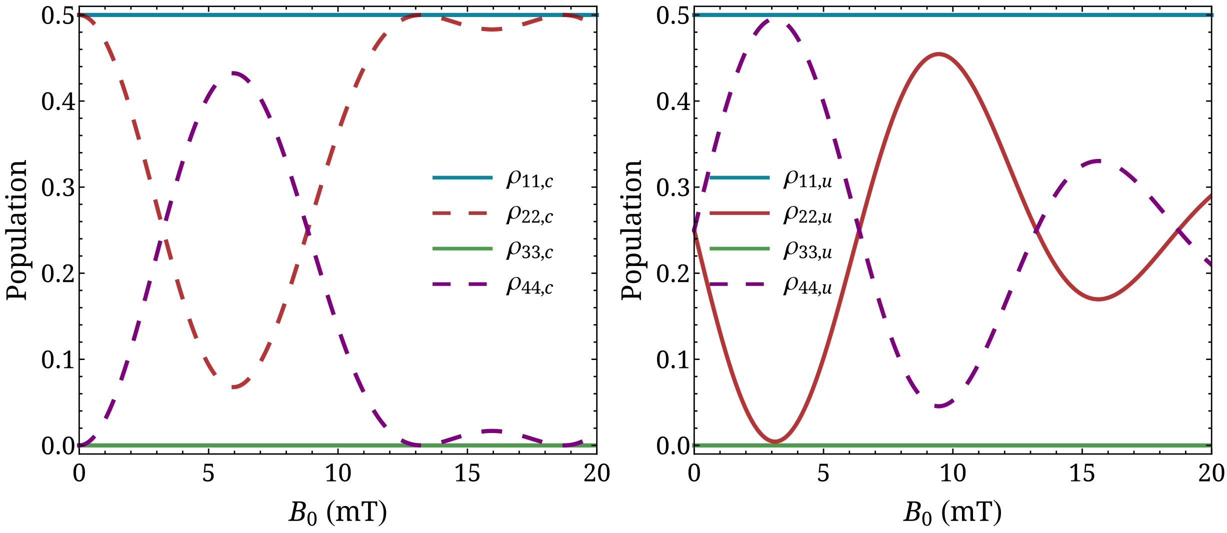
➢ How is population evolution affected by $B_z = B_0 \sin\left(\frac{2\pi v t}{\lambda}\right)$?

$$H = H_0 + H_B = \frac{A_{2S}}{\hbar^2} \boldsymbol{I} \cdot \boldsymbol{S} - \left(\frac{g_S \mu_B}{\hbar} \boldsymbol{S} + \frac{g_I \mu_N}{\hbar} \boldsymbol{I}\right) \cdot \boldsymbol{B}$$

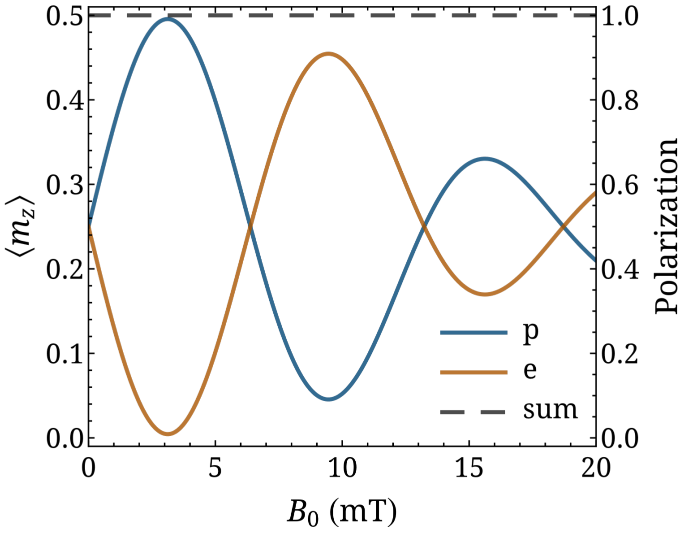


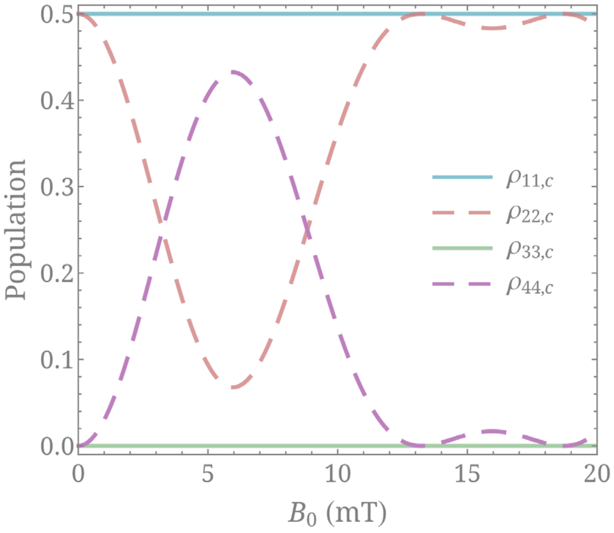
Incoherent initial preparation

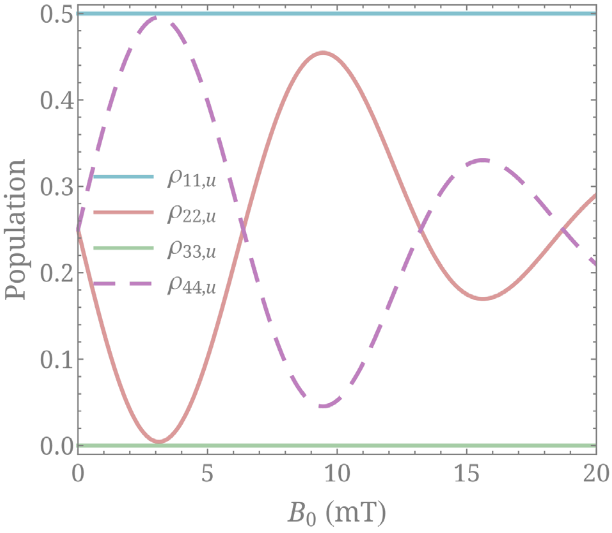
- Hydrogen atom

➢ How is population evolution affected by $B_z = B_0 \sin\left(\frac{2\pi v t}{\lambda}\right)$?

$$H = H_0 + H_B = \frac{A_{2S}}{\hbar^2}\, \boldsymbol{I}\cdot\boldsymbol{S} - \left(\frac{g_S \mu_B}{\hbar}\boldsymbol{S} + \frac{g_I \mu_N}{\hbar}\boldsymbol{I}\right)\cdot\boldsymbol{B}$$





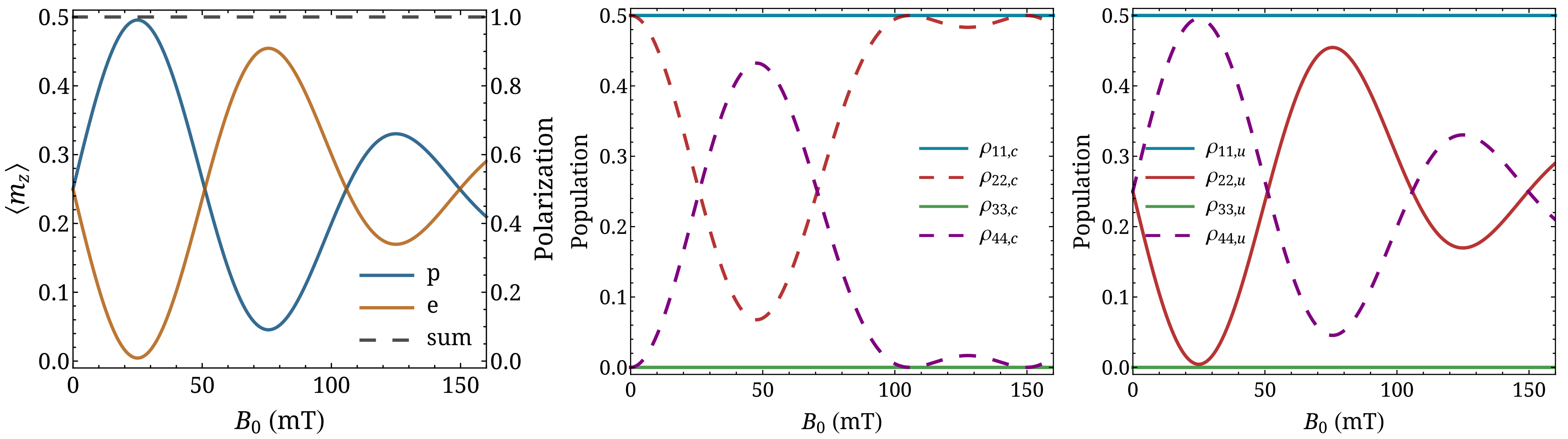


Incoherent initial preparation

- Hydrogen atom
  - Ground state ($A_{1S} = 1.42$ GHz)

➢ How is population evolution affected by $B_z = B_0 \sin\left(\frac{2\pi v t}{\lambda}\right)$?

$$H = H_0 + H_B = \frac{A_{1S}}{\hbar^2}\boldsymbol{I}\cdot\boldsymbol{S} - \left(\frac{g_S\mu_B}{\hbar}\boldsymbol{S} + \frac{g_I\mu_N}{\hbar}\boldsymbol{I}\right)\cdot\boldsymbol{B}$$



Coherent initial preparation

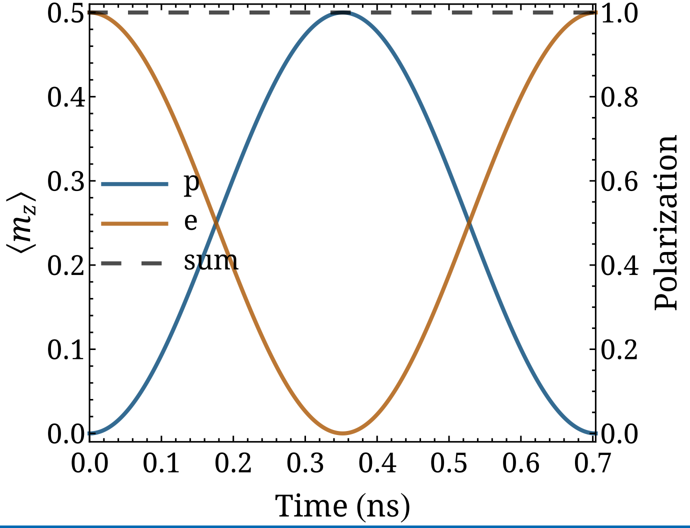
Heinrich Heine Universität Düsseldorf

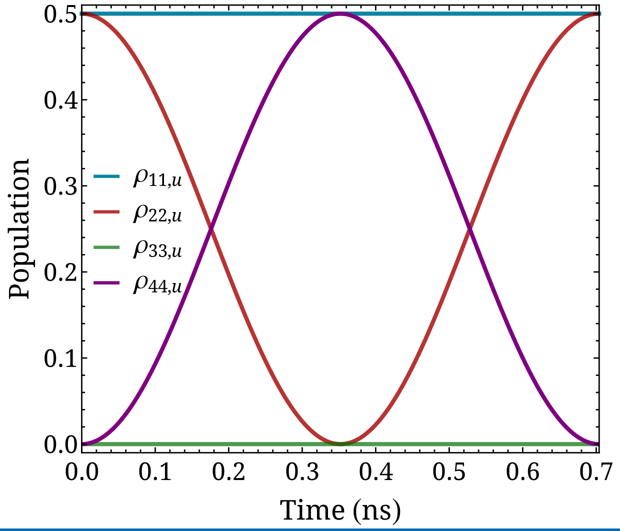
- Hydrogen atom
  - Ground state: Molecular photodissociation of a hydrogen halide
  - Example reaction: $\mathrm{HY} + hf \rightarrow \cdots \rightarrow \mathrm{H}\left(\mathrm{m_H} = \pm\frac{1}{2}\right) + \mathrm{Y}\left(\mathrm{m_Y} = \pm\frac{1}{2}\right)$

$$H_0 = \frac{A_{1S}}{\hbar^2}\,\boldsymbol{I}\cdot\boldsymbol{S}$$

$$\rho_u = \begin{pmatrix} 1/2 & 0 & 0 & 0 \\ 0 & 1/2 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}$$

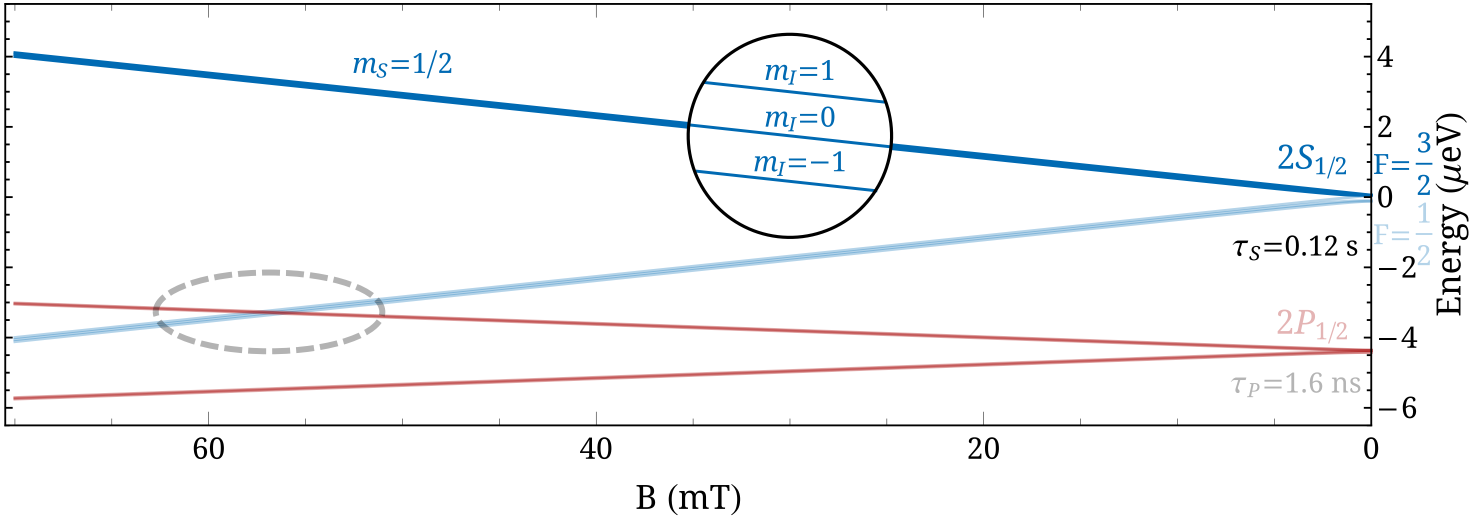
$$\rho_c = \begin{pmatrix} 1/2 & 0 & 0 & 0 \\ 0 & 1/4 & 0 & 1/4 \\ 0 & 0 & 0 & 0 \\ 0 & 1/4 & 0 & 1/4 \end{pmatrix}$$





Incoherent initial preparation

- Deuterium atom
  - Ground state: similar to hydrogen
  - Metastable state: use of the $2S_{1/2} - 2P_{1/2}$ level crossing (at high magnetic field)



Incoherent initial preparation

- Deuterium atom

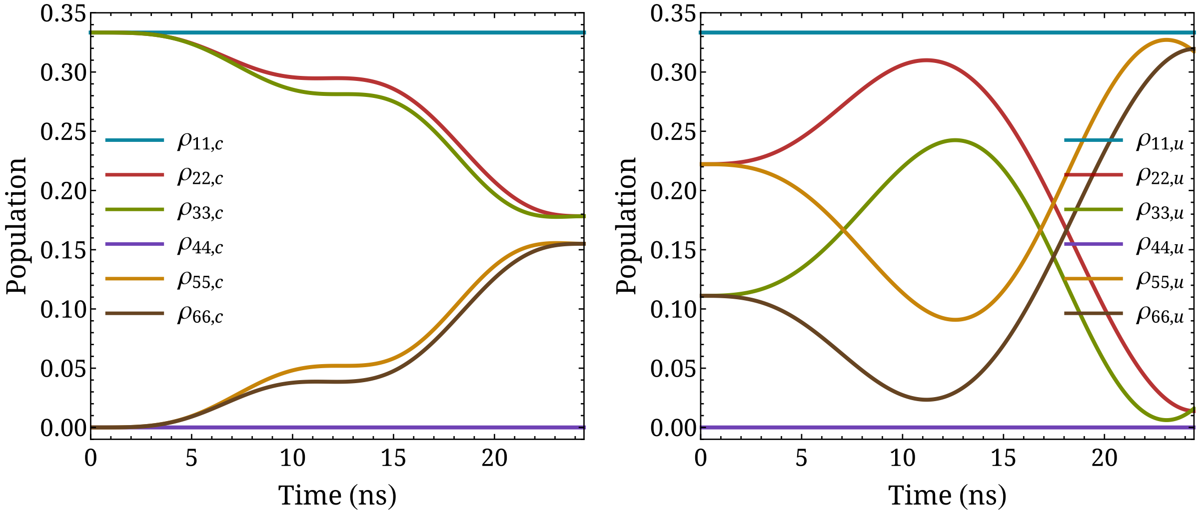
$$\rho_c = \begin{pmatrix} 1/3 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1/3 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1/3 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}$$

	coupled basis $\lvert F, m_F\rangle$	uncoupled basis $\lvert m_S, m_I\rangle$
1.	$\lvert 3/2, 3/2\rangle$	$\lvert 1/2, 1\rangle$
2.	$\lvert 3/2, 1/2\rangle$	$\lvert 1/2, 0\rangle$
3.	$\lvert 3/2, -1/2\rangle$	$\lvert 1/2, -1\rangle$
4.	$\lvert 3/2, -3/2\rangle$	$\lvert -1/2, -1\rangle$
5.	$\lvert 1/2, -1/2\rangle$	$\lvert -1/2, 0\rangle$
6.	$\lvert 1/2, 1/2\rangle$	$\lvert -1/2, 1\rangle$

$$H = H_0 + H_B = \frac{A_{2S}}{\hbar^2} \boldsymbol{I} \cdot \boldsymbol{S} - \left(\frac{g_S \mu_B}{\hbar} \boldsymbol{S} + \frac{g_I \mu_N}{\hbar} \boldsymbol{I}\right) \cdot \boldsymbol{B}$$

$$H_0 = \frac{A_{2S}}{\hbar^2} \boldsymbol{I} \cdot \boldsymbol{S} \text{ with } \frac{A_{2S}}{h} = 27.28 \text{ MHz} \qquad t_f = \frac{h}{\Delta E_{hfs}} = \frac{2h}{3A_{2S}} \sim 24.4 \text{ ns}$$

$$B_0 = 0.76 \text{ mT}$$

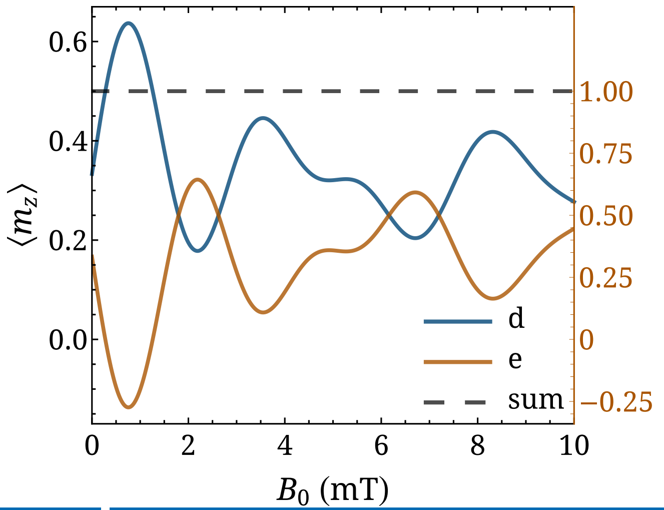


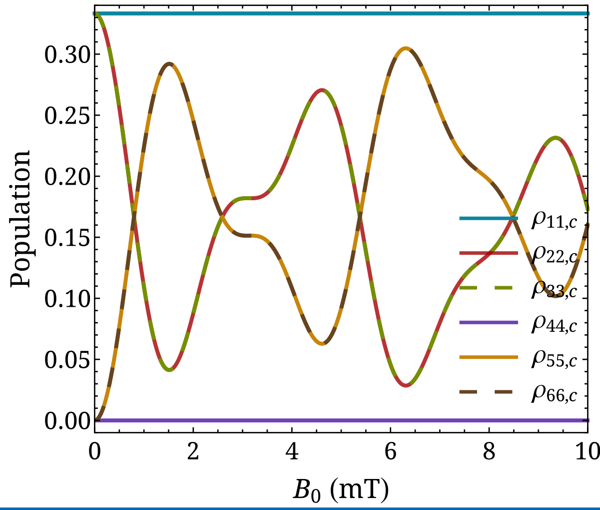
Incoherent initial preparation

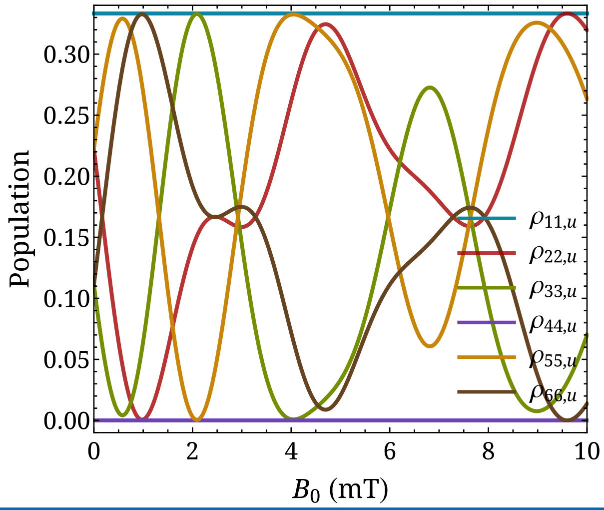
- Deuterium atom

$$H = H_0 + H_B = \frac{A_{2S}}{\hbar^2}\,\boldsymbol{I}\cdot\boldsymbol{S} - \left(\frac{g_S\mu_B}{\hbar}\boldsymbol{S} + \frac{g_I\mu_N}{\hbar}\boldsymbol{I}\right)\cdot\boldsymbol{B}$$

$$H_0 = \frac{A_{2S}}{\hbar^2}\,\boldsymbol{I}\cdot\boldsymbol{S} \ \text{ with } \ \frac{A_{2S}}{h} = 27.28\ \text{MHz} \qquad t_f = \frac{h}{\Delta E_{hfs}} = \frac{2h}{3A_{2S}} \sim 24.4\ \text{ns}$$





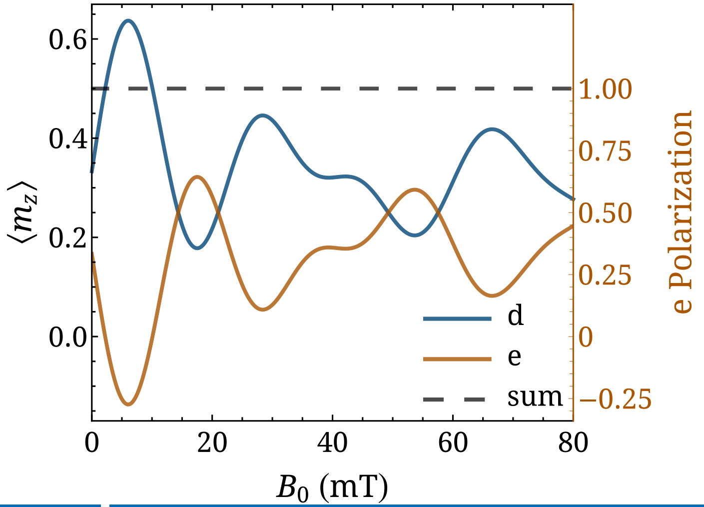


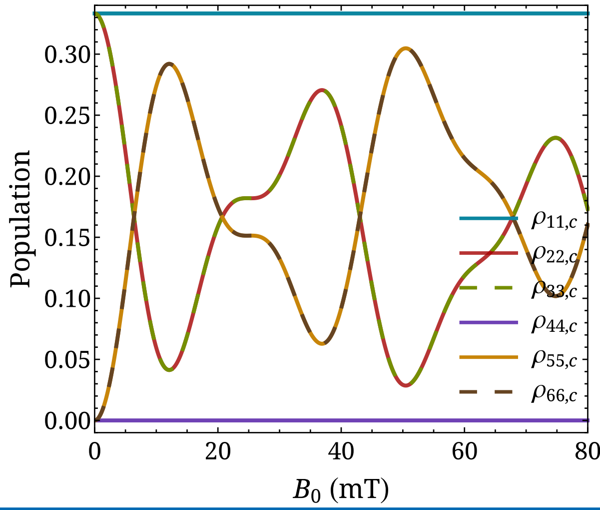
Incoherent initial preparation

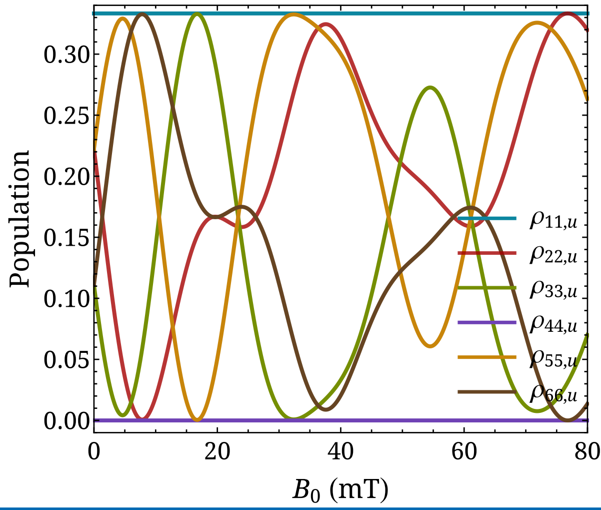
- Deuterium atom
  - Ground state

$$H = H_0 + H_B = \frac{A_{1S}}{\hbar^2}\,\boldsymbol{I}\cdot\boldsymbol{S} - \left(\frac{g_S\mu_B}{\hbar}\boldsymbol{S} + \frac{g_I\mu_N}{\hbar}\boldsymbol{I}\right)\cdot\boldsymbol{B}$$

$$H_0 = \frac{A_{1S}}{\hbar^2}\,\boldsymbol{I}\cdot\boldsymbol{S} \quad \text{with} \quad \frac{A_{1S}}{h} = 218.26 \text{ MHz} \qquad t_f = \frac{h}{\Delta E_{hfs}} = \frac{2h}{3A_{1S}} \sim 3 \text{ ns}$$







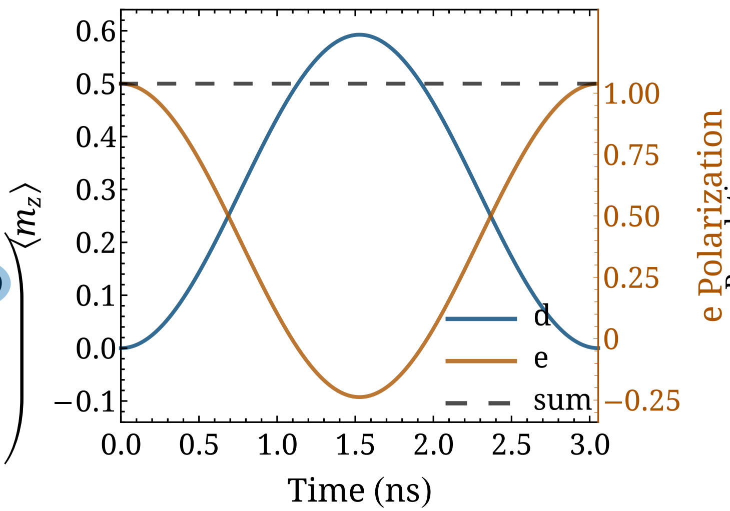
Coherent initial preparation

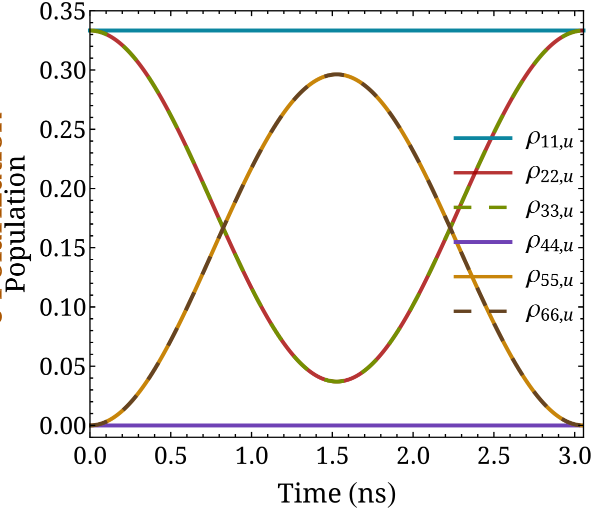
- Deuterium atom
  - Ground state: Molecular photodissociation of a deuterium halide
  - Example reaction: $\mathrm{DY} + hf \rightarrow \cdots \rightarrow \mathrm{D}\left(\mathrm{m_D} = \pm\frac{1}{2}\right) + \mathrm{Y}\left(\mathrm{m_Y} = \pm\frac{1}{2}\right)$

$$H_0 = \frac{A_{1S}}{\hbar^2}\, \boldsymbol{I} \cdot \boldsymbol{S}$$

$$\rho_u = \begin{pmatrix} 1/3 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1/3 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1/3 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}$$

$$\rho_c = \begin{pmatrix} 1/3 & 0 & 0 & 0 & 0 & 0 \\ 0 & 2/9 & 0 & 0 & 0 & \sqrt{2}/9 \\ 0 & 0 & 1/9 & 0 & \sqrt{2}/9 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & \sqrt{2}/9 & 0 & 2/9 & 0 \\ 0 & \sqrt{2}/9 & 0 & 0 & 0 & 1/9 \end{pmatrix}$$





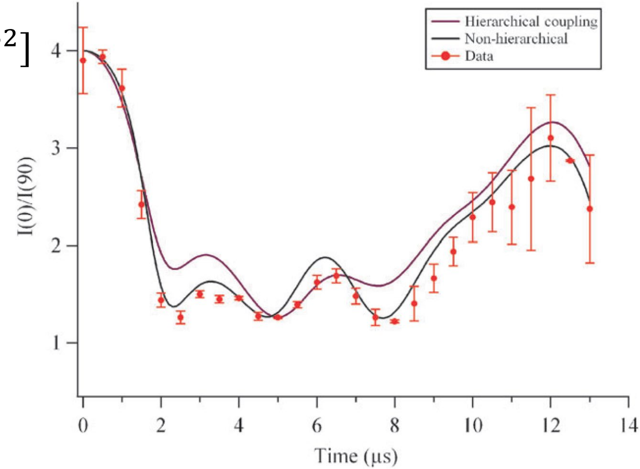
Coherently rotationally state-selected HD

hhu
Heinrich Heine
Universität Düsseldorf

- $$H_0 = -\frac{c_p}{\hbar^2}\,\boldsymbol{I_p}\cdot\boldsymbol{J} - \frac{c_d}{\hbar^2}\,\boldsymbol{I_d}\cdot\boldsymbol{J} + \frac{\delta}{\hbar^2}\,\boldsymbol{I_p}\cdot\boldsymbol{I_d} + \frac{5d_1}{(2J-1)(2J+3)\hbar^4}\left[\frac{3}{2}(\boldsymbol{I_p}\cdot\boldsymbol{J})(\boldsymbol{I_d}\cdot\boldsymbol{J}) + \frac{3}{2}(\boldsymbol{I_d}\cdot\boldsymbol{J})(\boldsymbol{I_p}\cdot\boldsymbol{J}) - \boldsymbol{I_p}\cdot\boldsymbol{I_d}\,\boldsymbol{J}^2\right] + \frac{5d_2}{(2J-1)(2J+3)\hbar^4}\left[3(\boldsymbol{I_d}\cdot\boldsymbol{J})^2 + \frac{3\hbar^2}{2}(\boldsymbol{I_d}\cdot\boldsymbol{J}) - \boldsymbol{I_d}^2\boldsymbol{J}^2\right]$$
- $$H_B = -\frac{a_p'}{\hbar}\,\boldsymbol{I_p}\cdot\boldsymbol{B} - \frac{a_d'}{\hbar}\boldsymbol{I_d}\cdot\boldsymbol{B} - \frac{b'}{\hbar}\boldsymbol{J}\cdot\boldsymbol{B} - \frac{5f'}{3(2J-1)(2J+3)\hbar^2}\left[3(\boldsymbol{J}\cdot\boldsymbol{B})^2 - \boldsymbol{J}^2\boldsymbol{B}^2\right]$$

with

$c_p/h = 85589$ Hz, $c_d/h = 13118$ Hz, $\delta/h = 43$ Hz, $d_1/h = 17764$ Hz, $d_2/h = -22452$ Hz, $a_p'/h = 4257.796 \times 10^4$ Hz/T, $f'/h = -2630$ Hz/T^2 $a_d'/h = 653.5832 \times 10^4$ Hz/T, and $b'/h = 505.5870 \times 10^4$ Hz/T

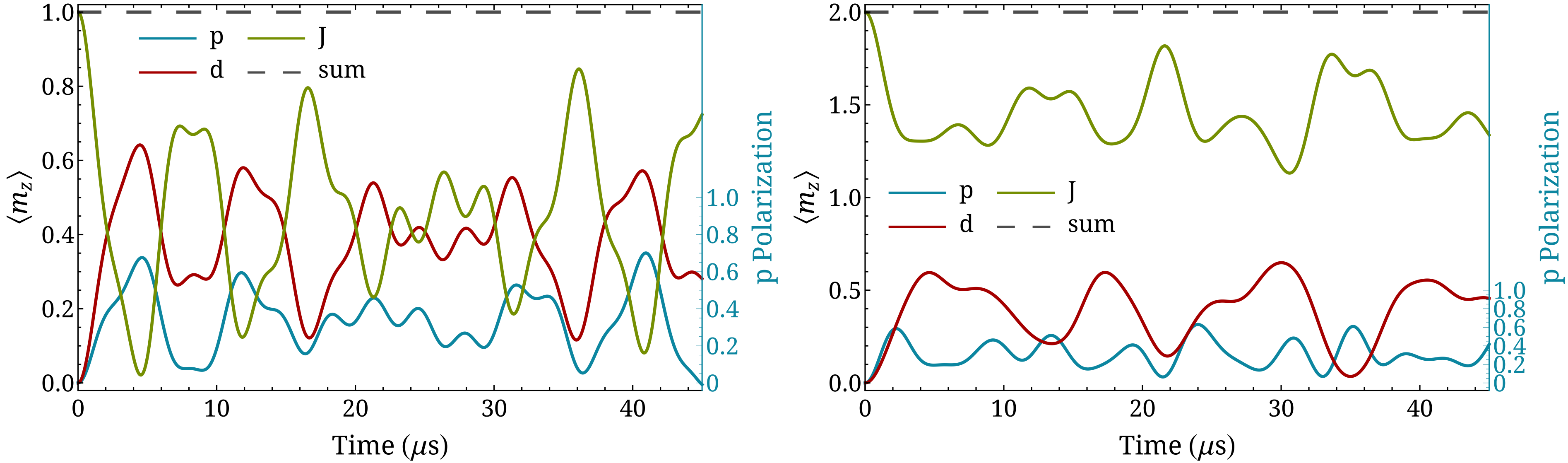


- Coherent initial preparation: $|J, m_J = +J\rangle$
- Transfer of molecular rotational polarization to nuclear spins

N. C.-M. Bartlett *et al.*, Phys. Chem. Chem. Phys. **11**, 142 (2009)

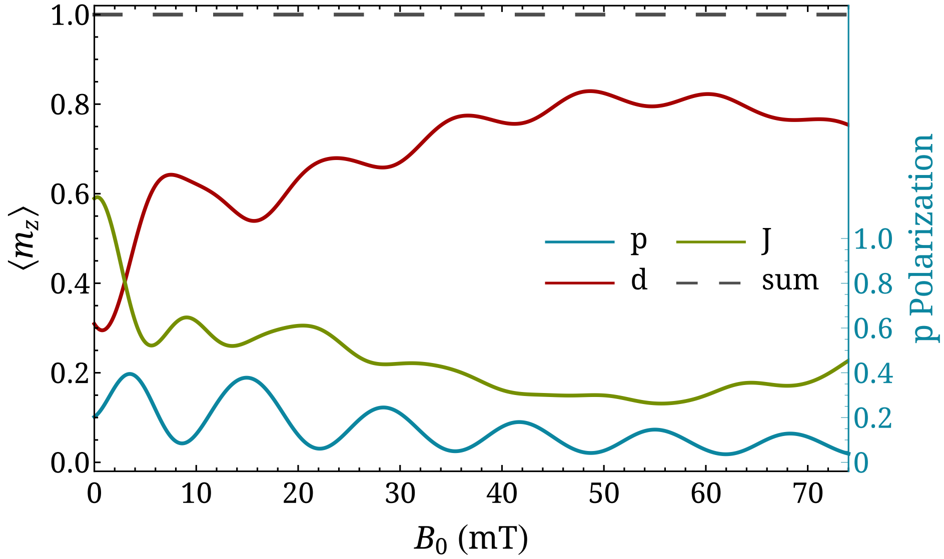
Coherently rotationally state-selected HD

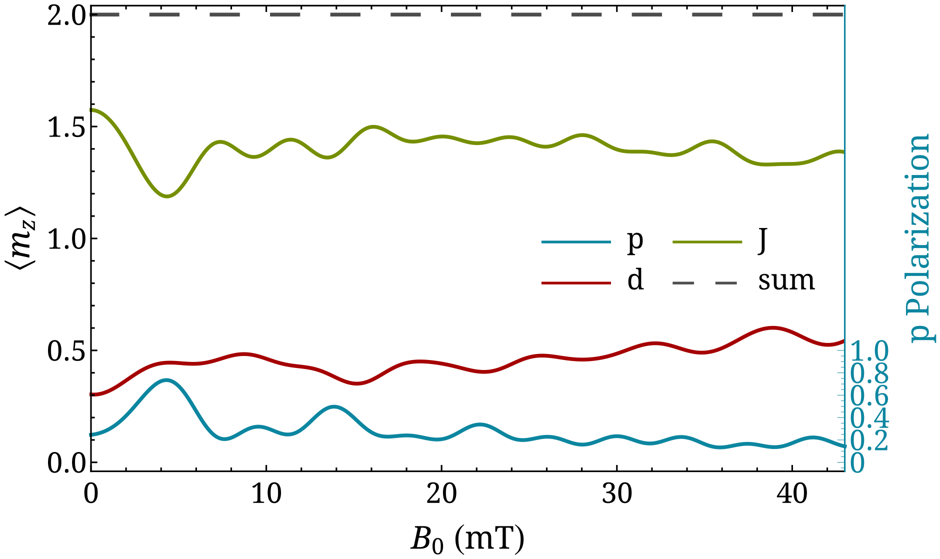
- $J = 1$: proton pol. 70% at 41 µs and deuteron pol. 64% at 4.5 µs
- $J = 2$: proton pol. 63% at 24 µs and deuteron pol. 65% at 30 µs



Coherently rotationally state-selected HD

- $J = 1$: $t_f = \frac{1}{153935\text{ Hz}} \sim 6.5$ µs, deuteron pol. 83% ($B_0 = 48.6$ mT)
- $J = 2$: $t_f = \frac{1}{87852\text{ Hz}} \sim 11.4$ µs, proton pol. 74% ($B_0 = 4.3$ mT)





Limitations

- Non-interacting particles following classical trajectories
  - External degrees of freedom (position, momentum): treated classically
  - Internal degrees of freedom (spins, rotational ang. momenta): treated quantum mechanically
  - Nonrelativistic velocities $\frac{v}{c} < 1\%$; beam kinetic energies up to: 46.9 keV (H), 93.8 keV (D), and 140.7 keV (HD)
- Radial magnetic field component B_r neglected ($r \ll \lambda$)
  - B_r induces $\Delta m_F = \pm 1$ transitions, not conserving longitudinal spin polarization
  - B_r also responsible for motional Stark effect
    - $\boldsymbol{E} = \boldsymbol{v} \times \boldsymbol{B} \Rightarrow \boldsymbol{E} = vB_r\widehat{\boldsymbol{\phi}}$
    - Electric dipole interaction does not couple states within the same hyperfine regime, only states of opposite parity
- Magnetic field need not be a perfect sinusoid
  - E.g., a sine-cubed waveform of the same frequency yields similar results with adjusted field strength

Additional information

- Theory (H)
- Theory (D)
- Incoherent preparation of metastable H (maximum nuclear polarization)
- Coherent preparation of ground-state H (signal from dissociation of HBr at 213 nm)
- Incoherent preparation of metastable D ($2S_{1/2} - 2P_{1/2}$ level crossing)
- Coherent preparation of ground-state D (signal from dissociation of DI at 266 nm)
- Coherently rotationally state-selected HD (maximum nuclear polarization)
- Experimental setup
- Applications

Theory: coupled and uncoupled bases

- Hydrogen atom: $\frac{1}{2} \otimes \frac{1}{2} = 1 \oplus 0$

	coupled basis $\lvert F, m_F\rangle$	uncoupled basis $\lvert m_S, m_I\rangle$
1.	$\lvert 1,1\rangle$	$\lvert 1/2, 1/2\rangle$
2.	$\lvert 1,0\rangle$	$\lvert 1/2, -1/2\rangle$
3.	$\lvert 1,-1\rangle$	$\lvert -1/2, -1/2\rangle$
4.	$\lvert 0,0\rangle$	$\lvert -1/2, 1/2\rangle$

$$\langle m_{z,\,p/e}\rangle = \frac{1}{\hbar}\langle S_{z,\,p/e}\rangle = \frac{1}{\hbar} Tr\left(\rho S_{z,p/e}\right)$$

$$S_{z,p,u} = \frac{\hbar}{2}\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix} \text{ and } S_{z,p,c} = Q S_{z,p,u} Q^{-1} = \frac{\hbar}{2}\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}$$

$$\lvert\chi\rangle_c = Q\lvert\chi\rangle_u \text{ with } Q = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1/\sqrt{2} & 0 & 1/\sqrt{2} \\ 0 & 0 & 1 & 0 \\ 0 & 1/\sqrt{2} & 0 & -1/\sqrt{2} \end{pmatrix}$$

$$\text{For } \rho_c = \begin{pmatrix} 1/2 & 0 & 0 & 0 \\ 0 & 1/2 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix},\ \rho_u = \begin{pmatrix} 1/2 & 0 & 0 & 0 \\ 0 & 1/4 & 0 & 1/4 \\ 0 & 0 & 0 & 0 \\ 0 & 1/4 & 0 & 1/4 \end{pmatrix}$$

Theory: coupled and uncoupled bases

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- Deuterium atom: $\frac{1}{2} \otimes 1 = \frac{3}{2} \oplus \frac{1}{2}$

	coupled basis $\vert F, m_F\rangle$	uncoupled basis $\vert m_S, m_I\rangle$
1.	$\vert 3/2, 3/2\rangle$	$\vert 1/2, 1\rangle$
2.	$\vert 3/2, 1/2\rangle$	$\vert 1/2, 0\rangle$
3.	$\vert 3/2, -1/2\rangle$	$\vert 1/2, -1\rangle$
4.	$\vert 3/2, -3/2\rangle$	$\vert -1/2, -1\rangle$
5.	$\vert 1/2, -1/2\rangle$	$\vert -1/2, 0\rangle$
6.	$\vert 1/2, 1/2\rangle$	$\vert -1/2, 1\rangle$

$$\langle m_{z,\,d/e}\rangle = \frac{1}{\hbar}\langle S_{z,\,d/e}\rangle = \frac{1}{\hbar}Tr\left(\rho S_{z,\,d/e}\right)$$

$$S_{z,d,u} = \hbar\begin{pmatrix} 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & -1 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 \end{pmatrix}$$

$$S_{z,d,c} = QS_{z,d,u}Q^{-1} = \hbar\begin{pmatrix} 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1/3 & 0 & 0 & 0 & -\sqrt{2}/3 \\ 0 & 0 & -1/3 & 0 & -\sqrt{2}/3 & 0 \\ 0 & 0 & 0 & -1 & 0 & 0 \\ 0 & 0 & -\sqrt{2}/3 & 0 & -2/3 & 0 \\ 0 & -\sqrt{2}/3 & 0 & 0 & 0 & 2/3 \end{pmatrix}$$

$$|\chi\rangle_c = Q|\chi\rangle_u \quad \text{with} \quad Q = \begin{pmatrix} 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & \sqrt{2/3} & 0 & 0 & 0 & 1/\sqrt{3} \\ 0 & 0 & 1/\sqrt{3} & 0 & \sqrt{2/3} & 0 \\ 0 & 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & \sqrt{2/3} & 0 & -1/\sqrt{3} & 0 \\ 0 & 1/\sqrt{3} & 0 & 0 & 0 & -\sqrt{2/3} \end{pmatrix}$$

For $\rho_c = \begin{pmatrix} 1/3 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1/3 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1/3 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}$, $\rho_u = \begin{pmatrix} 1/3 & 0 & 0 & 0 & 0 & 0 \\ 0 & 2/9 & 0 & 0 & 0 & \sqrt{2}/9 \\ 0 & 0 & 1/9 & 0 & \sqrt{2}/9 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & \sqrt{2}/9 & 0 & 2/9 & 0 \\ 0 & \sqrt{2}/9 & 0 & 0 & 0 & 1/9 \end{pmatrix}$

Incoherent initial preparation

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- Hydrogen atom

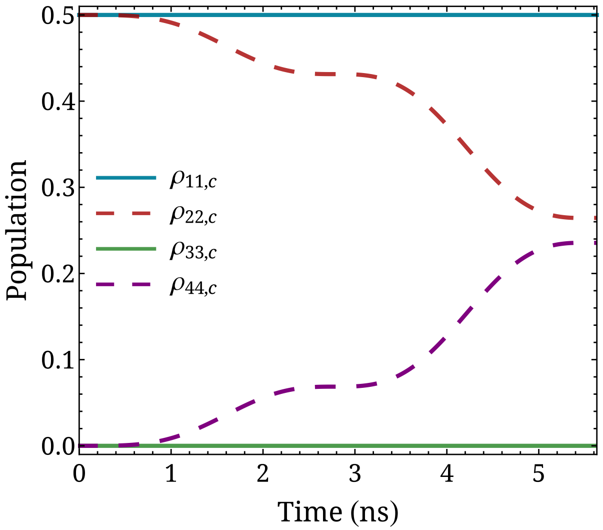
$$\rho_c = \begin{pmatrix} 1/2 & 0 & 0 & 0 \\ 0 & 1/2 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}$$

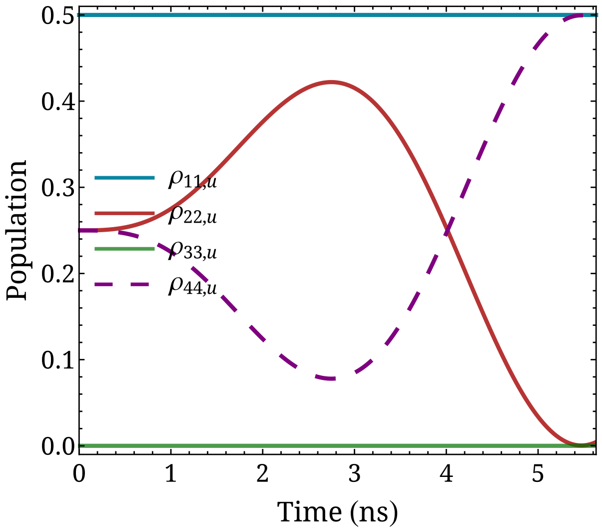
	coupled basis $\|F, m_F\rangle$	uncoupled basis $\|m_S, m_I\rangle$
1.	$\|1,1\rangle$	$\|1/2, 1/2\rangle$
2.	$\|1,0\rangle$	$\|1/2, -1/2\rangle$
3.	$\|1,-1\rangle$	$\|-1/2, -1/2\rangle$
4.	$\|0,0\rangle$	$\|-1/2, 1/2\rangle$

➢ How is population evolution affected by $B_z = B_0 \sin\left(\frac{2\pi v t}{\lambda}\right)$?

$$H = H_0 + H_B = \frac{A_{2S}}{\hbar^2} \boldsymbol{I} \cdot \boldsymbol{S} - \left(\frac{g_S \mu_B}{\hbar} \boldsymbol{S} + \frac{g_I \mu_N}{\hbar} \boldsymbol{I}\right) \cdot \boldsymbol{B}$$

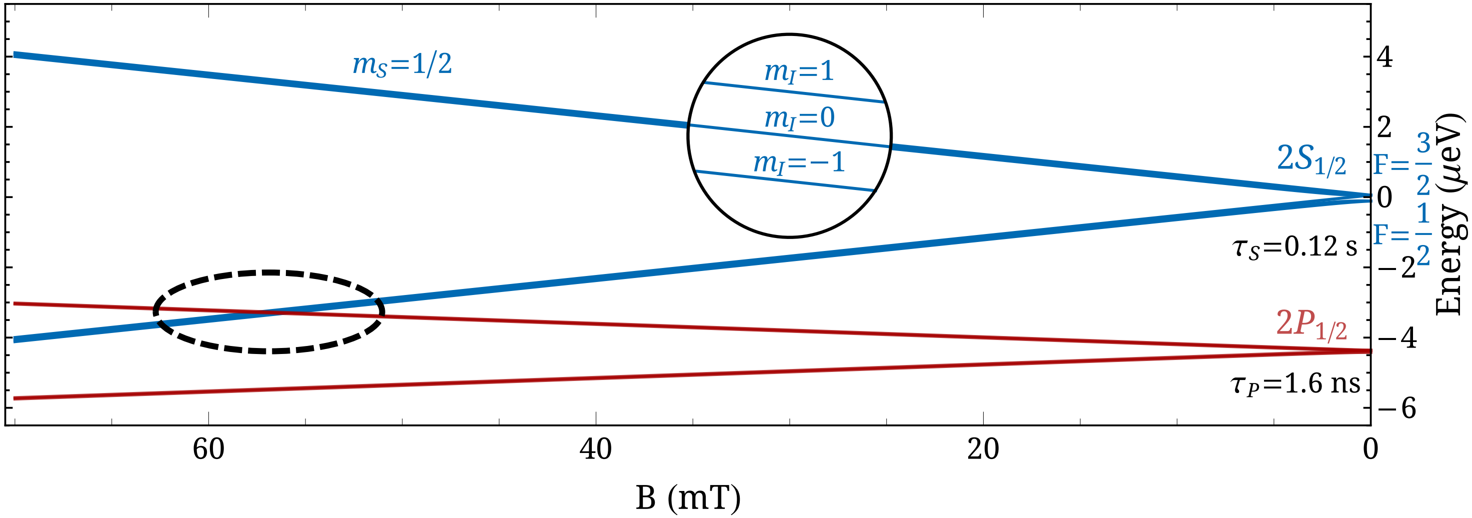
$B_0 = 3.12$ mT





Incoherent initial preparation

- Deuterium atom
  - Ground state: similar to hydrogen
  - Metastable state: use of the $2S_{1/2} - 2P_{1/2}$ level crossing (at high magnetic field)

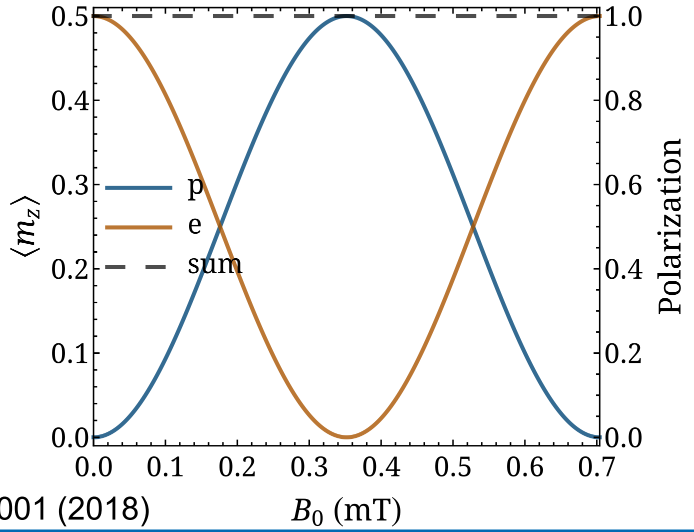


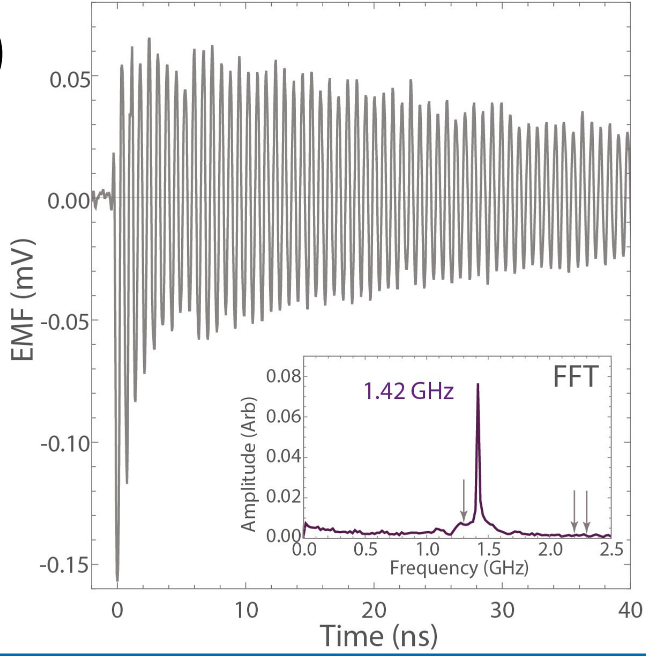
Coherent initial preparation

- Hydrogen atom
  - Ground state: Molecular photodissociation of a hydrogen halide
  - Example reaction: $\mathrm{HY} + hf \rightarrow \cdots \rightarrow \mathrm{H}\left(\mathrm{m_H} = \pm\frac{1}{2}\right) + \mathrm{Y}\left(\mathrm{m_Y} = \pm\frac{1}{2}\right)$

$$\rho_u = \begin{pmatrix} 1/2 & 0 & 0 & 0 \\ 0 & 1/2 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}$$

$$\rho_c = \begin{pmatrix} 1/2 & 0 & 0 & 0 \\ 0 & 1/4 & 0 & 1/4 \\ 0 & 0 & 0 & 0 \\ 0 & 1/4 & 0 & 1/4 \end{pmatrix}$$





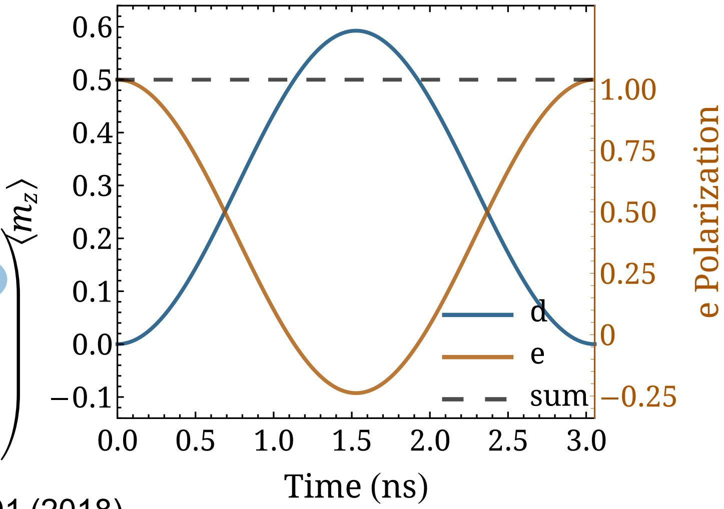
D. Sofikitis *et al.*, Phys. Rev. A **121**, 083001 (2018)

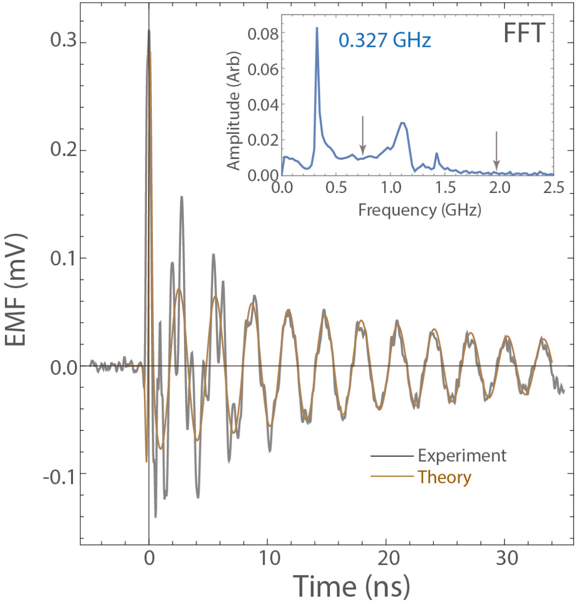
Coherent initial preparation

- Deuterium atom
  - Ground state: Molecular photodissociation of a deuterium halide
  - Example reaction: $\mathrm{DY} + hf \rightarrow \cdots \rightarrow \mathrm{D}\left(\mathrm{m_D} = \pm\frac{1}{2}\right) + \mathrm{Y}\left(\mathrm{m_Y} = \pm\frac{1}{2}\right)$

$$\rho_u = \begin{pmatrix} 1/3 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1/3 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1/3 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}$$

$$\rho_c = \begin{pmatrix} 1/3 & 0 & 0 & 0 & 0 & 0 \\ 0 & 2/9 & 0 & 0 & 0 & \sqrt{2}/9 \\ 0 & 0 & 1/9 & 0 & \sqrt{2}/9 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & \sqrt{2}/9 & 0 & 2/9 & 0 \\ 0 & \sqrt{2}/9 & 0 & 0 & 0 & 1/9 \end{pmatrix}$$

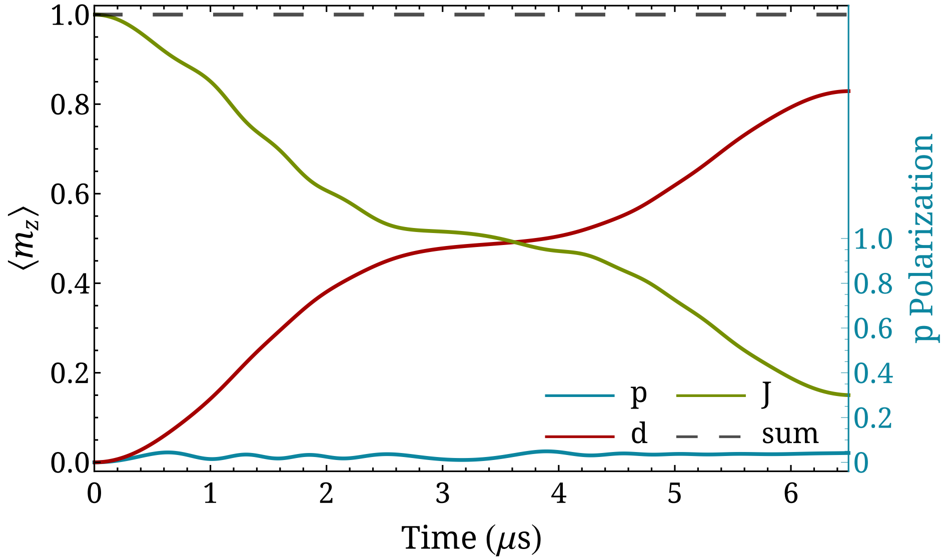


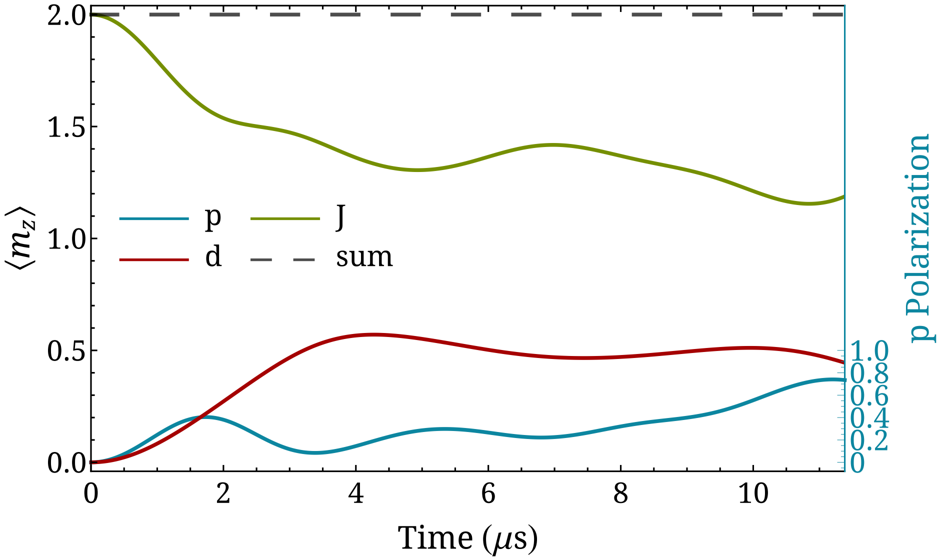


D. Sofikitis *et al.*, Phys. Rev. A **121**, 083001 (2018)

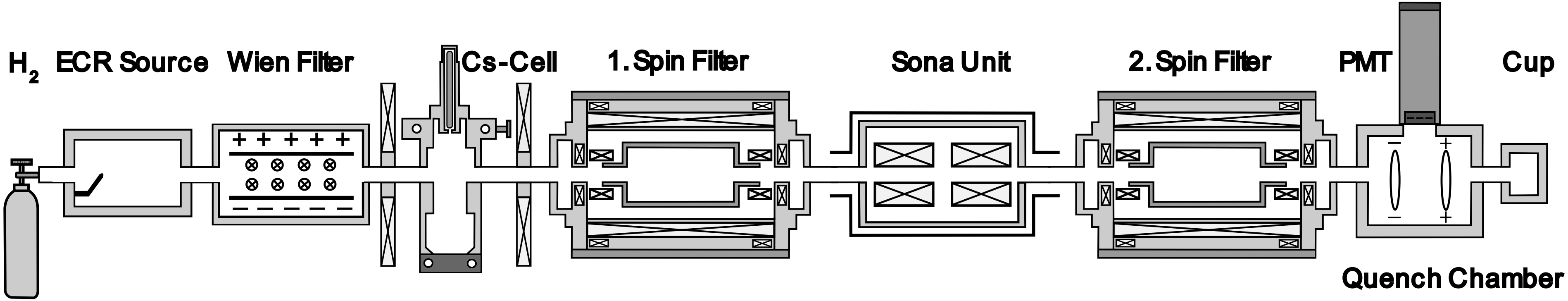
Coherently rotationally state-selected HD

- $J = 1$: $t_f = \frac{1}{153935\ \text{Hz}} \sim 6.5$ µs, deuteron pol. 83% ($B_0 = 48.6$ mT)
- $J = 2$: $t_f = \frac{1}{87852\ \text{Hz}} \sim 11.4$ µs, proton pol. 74% ($B_0 = 4.3$ mT)





Experimental setup



Applications

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- Experimental implementation
  - Enhances nuclear spin polarization
    - Atoms: $t_f \sim ns$, $\lambda \sim mm$, $B_0 \sim mT$ (nonconventional wire coils)
    - Molecules: $t_f \sim \mu s$, $\lambda \sim m$, $B_0 \sim mT$ (conventional wire coils)
  - Induces transitions within the hyperfine regime
    - B_r must be comparable to B_z [R. Engels *et al.*, Eur. Phys. J. D **75**, 257 (2021)]
- Computational modeling
  - Numerical solution of spin dynamics for systems with interacting angular momenta, assuming no entanglement with external degrees of freedom
  - Handles arbitrarily varying magnetic fields (not limited to zero-crossings or direction changes)
  - Enables Fourier analysis of time-domain observables in periodic fields