Laser preparation of intense beams & *targets* of vectorially polarized protons and tensorially polarized deuterons via molecular quantum beats.

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Dimitris Sofikitis

Spin2018

H, D, nuclear orientation

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- Hyperpolarization – our approach
  - Physical processes
  - Technological aspects

H, D, nuclear orientation

- Hyperpolarization – our approach
  - Physical processes
    - Molecular Photodissociation
    - Hyperfine polarization quantum beats
  - Technological aspects
    - Detection of spin polarization
    - Storage in B fields
    - Status of high power (table-top) laser technology
      - Why time is ripe?

H, D, nuclear orientation

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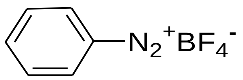
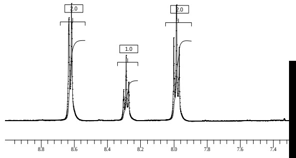
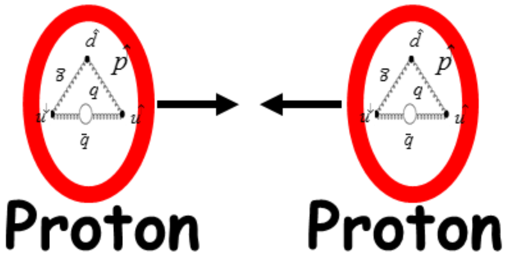
- Motivation:
  - Nuclear/particle physics
  - Reaction dynamics
  - NMR/MRI





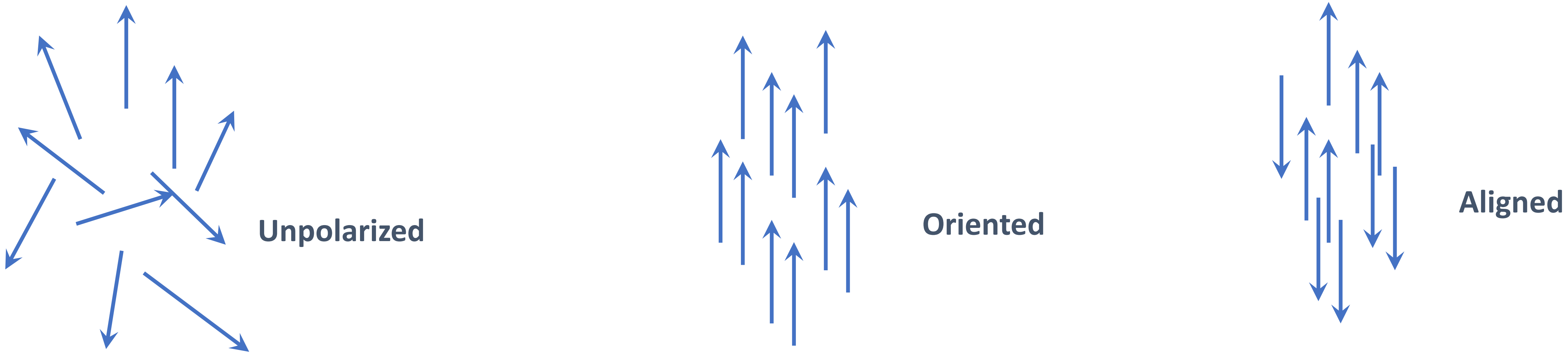


H, D, nuclear orientation

- Motivation:
  - Nuclear/particle physics
  - Reaction dynamics
  - NMR/MRI
  - Polarized Fusion
  - Wednesday 12 September A11: Prof. RAKITZIS, T. Peter

H, D, nuclear orientation

- Some vocabulary: Polarization



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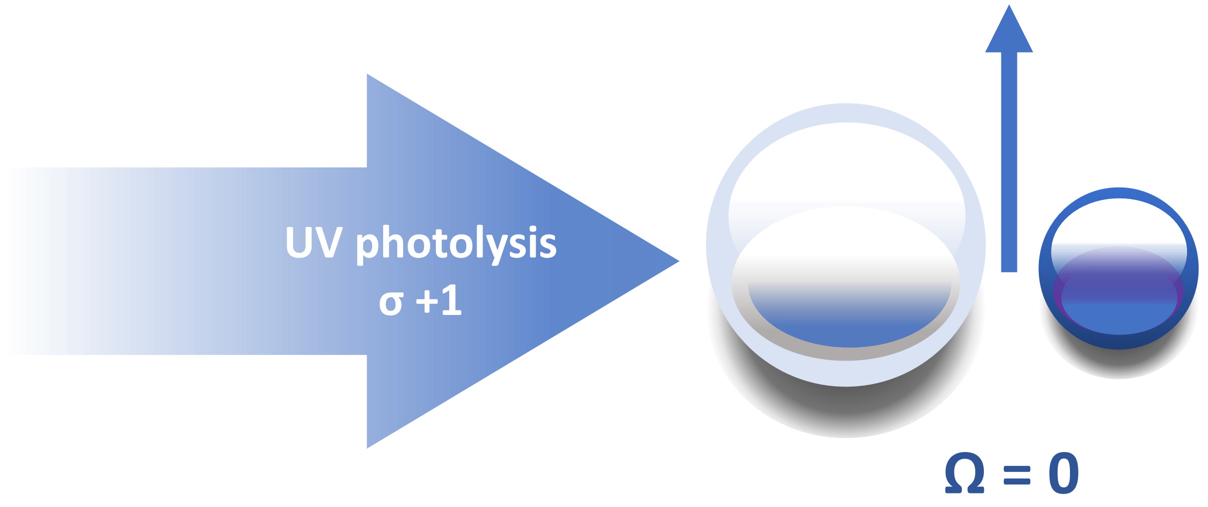
- Laser polarization (orientation/alignment) of nuclei?
  - Via the electron
  - Two-step process
    - Polarize the electron – laser dissociation
  - Polarization quantum beating
    - Transfer polarization to the nuclei

H, D, nuclear orientation

- Polarize the electron: Photodissociation
- R. J. van Brunt and R. N. Zare, J. Chem. Phys. 48, 4304 (1968)
  - Angular momentum projection → conserved during the dissociation process
  - Circularly polarized light → increase angular momentum projection (of Ω) by 1



H, D, nuclear orientation

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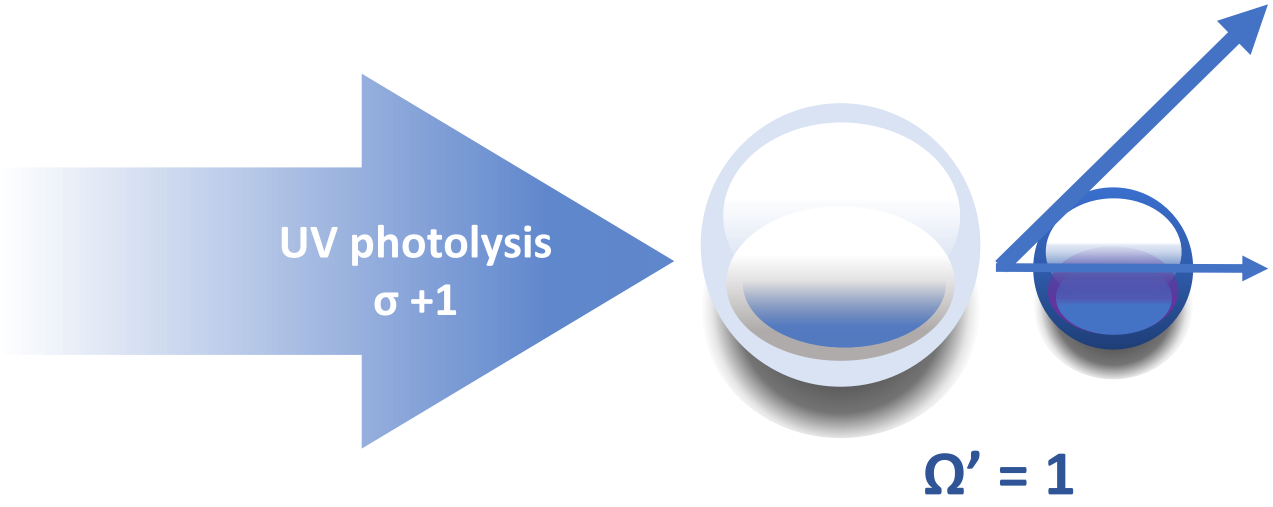

H, D, nuclear orientation

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    - Angular momentum projection → conserved during the dissociation process
    - Circularly polarized light → increase angular momentum projection (of Ω) by 1
    - In a prompt dissociation process this increase can be found in the fragments

H, D, nuclear orientation

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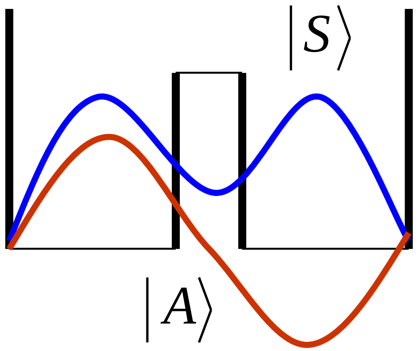
- Polarize the nucleus: Polarization beating
- Symmetric double well
  - Supports two eigenstates: S & A
  - Electron R (L) : S ± A
  - If $E_S \neq E_A$ → time dependence: Right ↔ Left

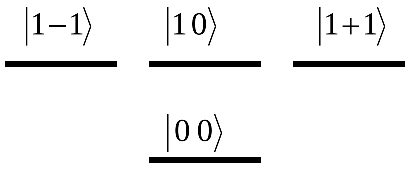


$$|R\rangle = \frac{1}{\sqrt{2}}\left(|S\rangle + |A\rangle\right) \xrightarrow{Time} |R\rangle = \frac{1}{\sqrt{2}}\left(e^{-iE_S t/\hbar}|S\rangle + e^{-iE_A t/\hbar}|A\rangle\right)$$

$$|L\rangle = \frac{1}{\sqrt{2}}\left(|S\rangle - |A\rangle\right) \xrightarrow{Time} |L\rangle = \frac{1}{\sqrt{2}}\left(e^{-iE_S t/\hbar}|S\rangle - e^{-iE_A t/\hbar}|A\rangle\right)$$

$$t = \pi\hbar / \Delta E$$

S + I → F



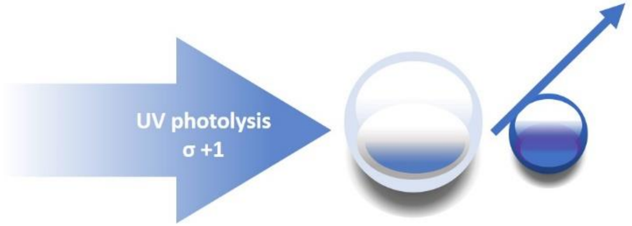
$$\uparrow\downarrow = \frac{1}{\sqrt{2}}\left(|1\,0\rangle + |0\,0\rangle\right)$$

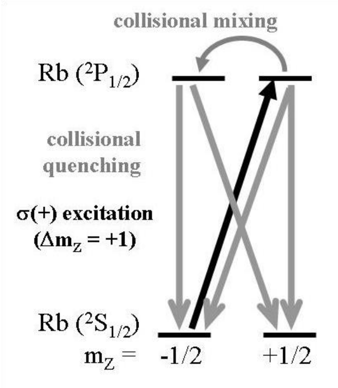
$$\downarrow\uparrow = \frac{1}{\sqrt{2}}\left(|1\,0\rangle - |0\,0\rangle\right)$$

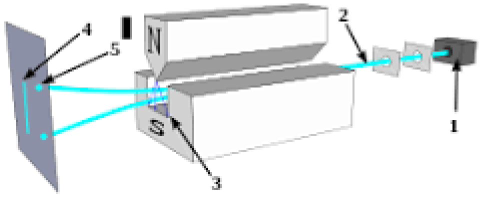
- Photodissociation does not resolve hyperfine levels
- Parallel/antiparallel = superposition F states

H, D, nuclear orientation

- Very attractive combination – timescales
- (prompt) Photodissociation ~ 100 fs
- Polarization beating (H, D) ~ ns
- State-of-the-art: SEOP and Stern Gerlach ~ μs
- Dissociation projected 10^{17} cm^{-3} with 10 ns (alkali-H depolarization cross sections)

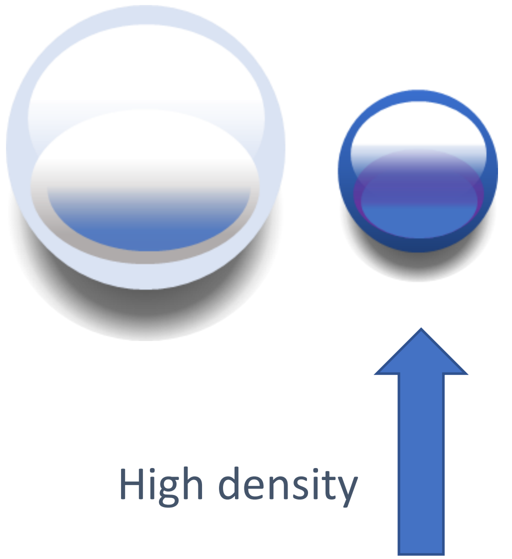






H, D, nuclear orientation

- Very attractive combination – timescales
- Needs to be proved!
  - Means detecting the polarization
  - Detect electron (nucleus) polarization in high density



H, D, nuclear orientation

- High density SPD and SPH
  - Milner et al., Phys. Rev. Lett. **118**, 243201 (2017)
  - Use an Optical centrifuge to polarize O_2 molecules
  - Detect the electron polarization via the EMF induced in a pick-up coil

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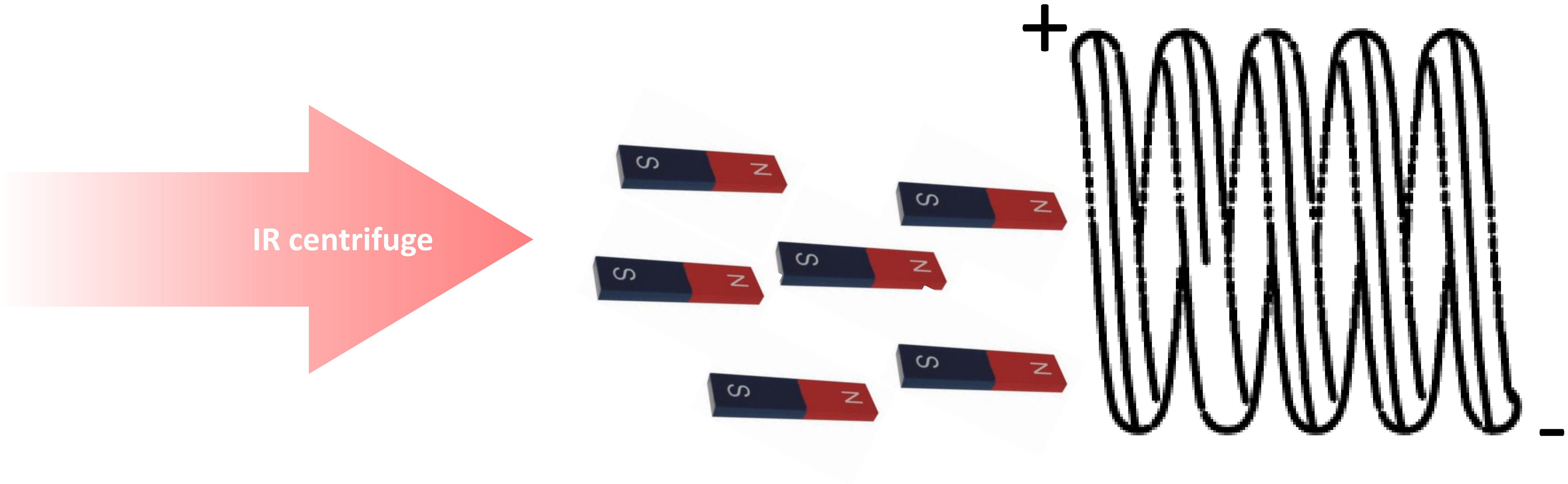

H, D, nuclear orientation

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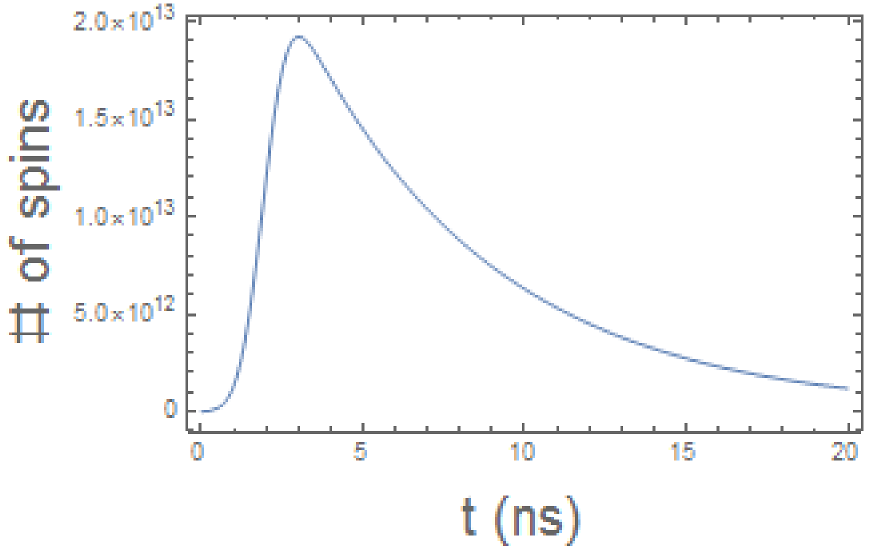
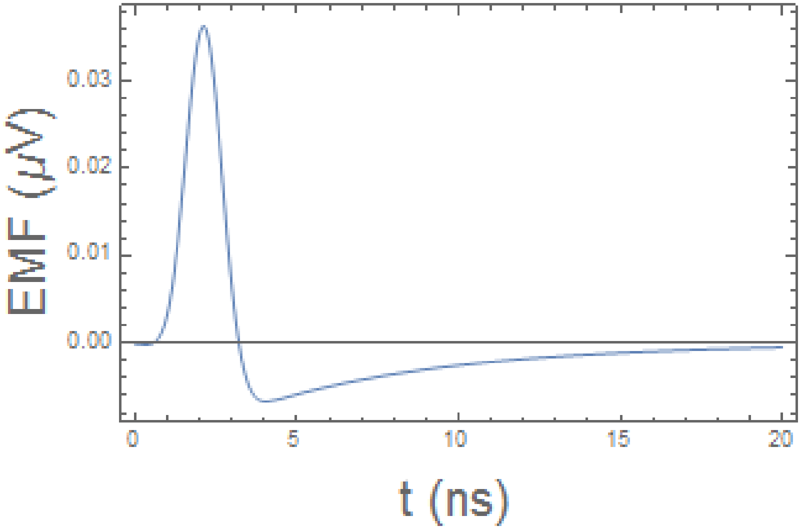
- High density SPD and SPH
  - Milner et al., Phys. Rev. Lett. **118**, 243201 (2017)
  - Detect the electron polarization via the EMF induced in a pick-up coil
  - Sudden polarization : create magnetization
  - Depolarization : exponential decay
  - EMF proportional to dP/dt





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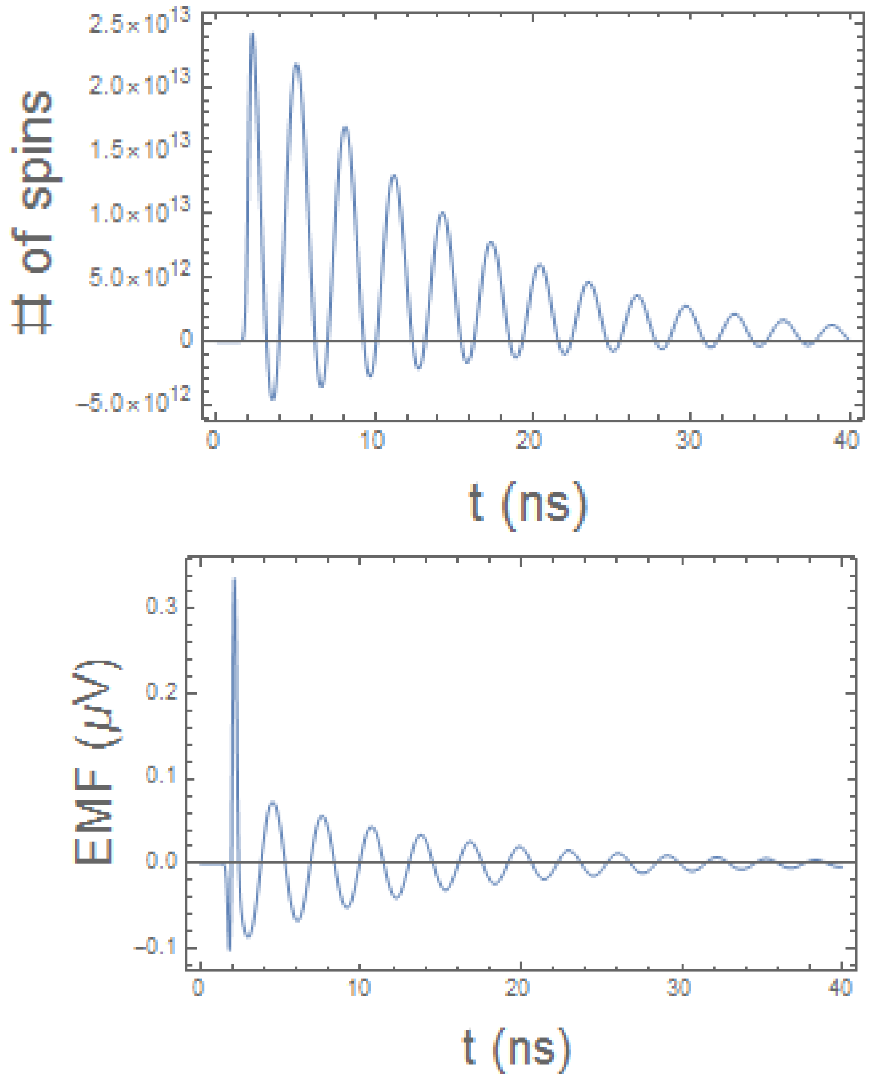
- High density SPD and SPH
  - If polarization exchange to nucleus
  - Magnetization vanishes (μ_B/μ_N~1836)



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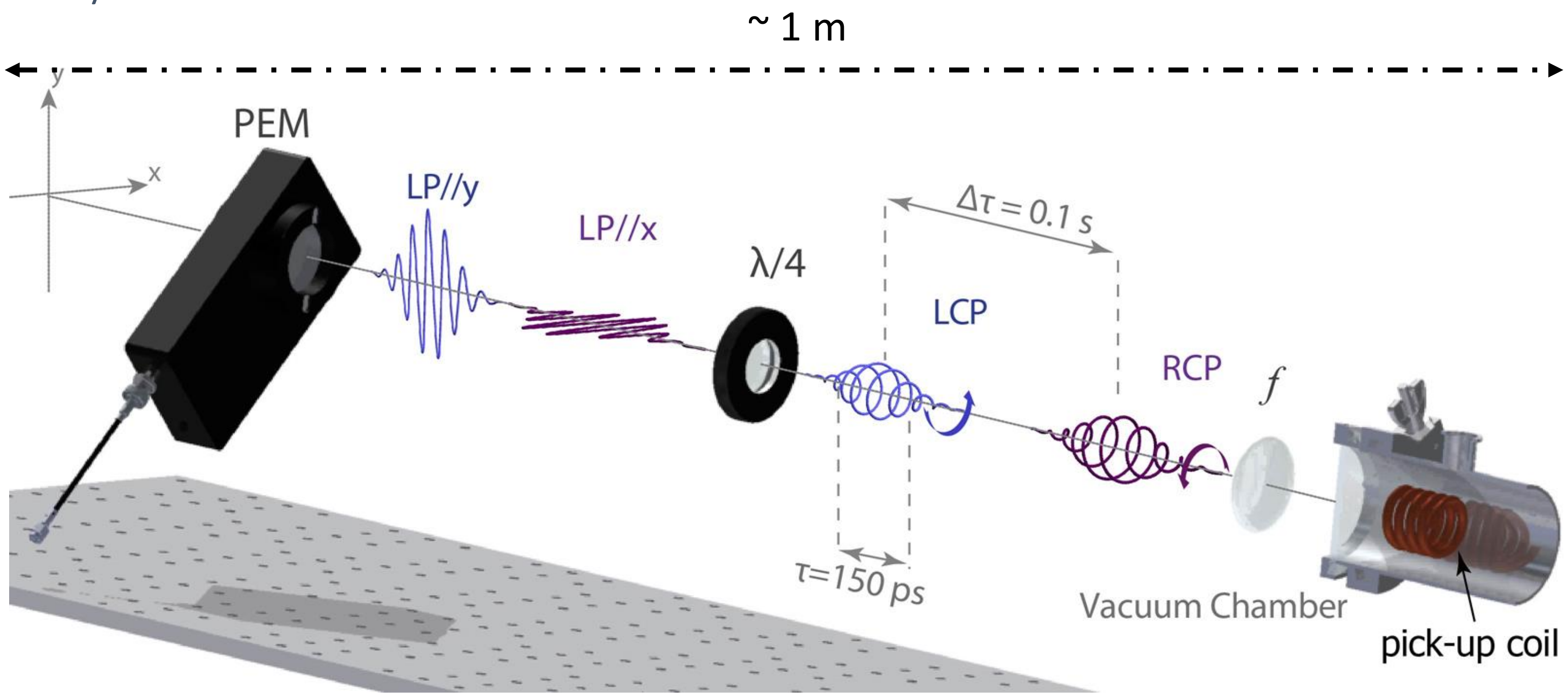
- High density SPD and SPH
  - Sofikitis et al., Phys. Rev. Lett. **121**, 083001 (2018)
  - We project signals ~ 100 µV with densities 10^{18} cm^{-3}
  - However, UV – noise ~ mV
  - PEM-Signal reversal
  - Dissociation : RCP/LCP shot-to-shot basis
  - Accumulate RCP and LCP separately



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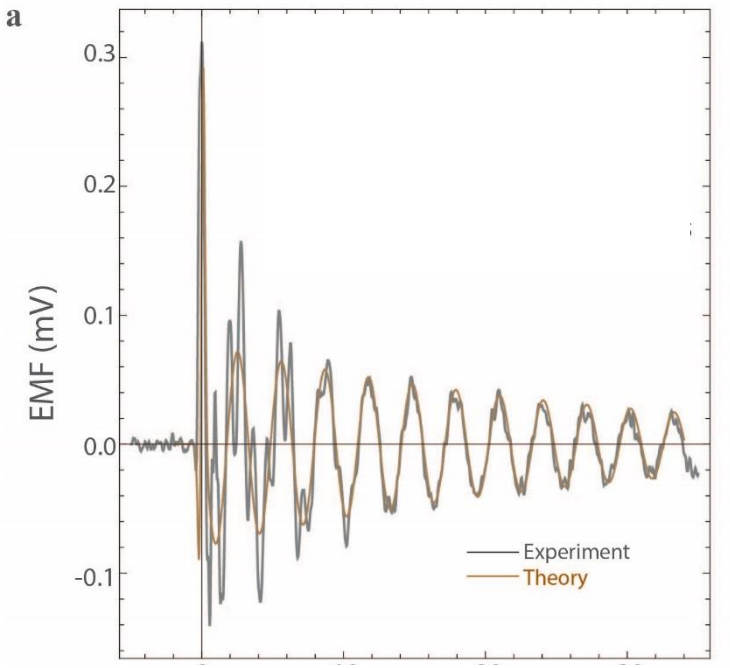
- High density SPD and SPH
  - Sofikitis et al., Phys. Rev. Lett. 121, 083001 (2018)
  - EMF signals:
    - Sharp rising edge ~ 150 ps photolysis
    - Exponential decay – depolarization
    - ns polarization exchange
    - Similar for H from HBr
    - FFT → Hyperfine frequencies



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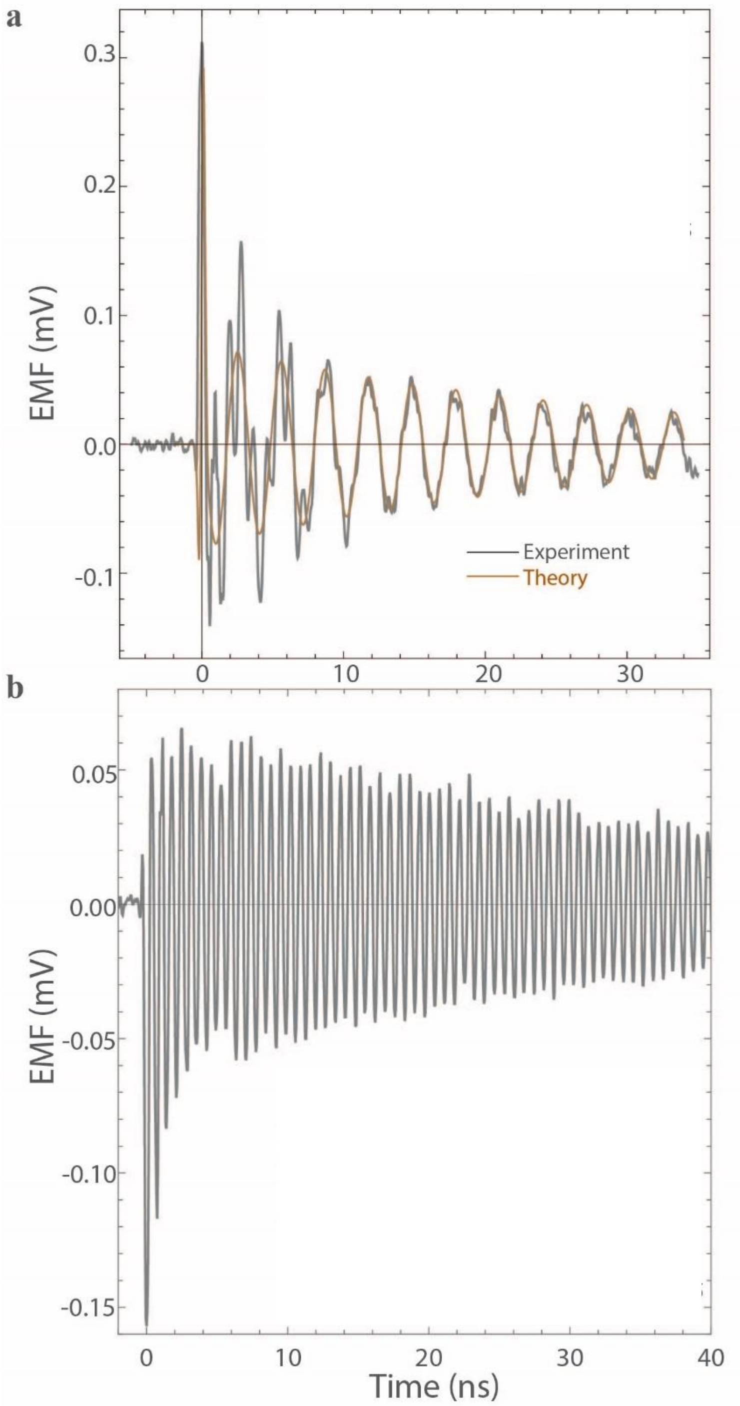

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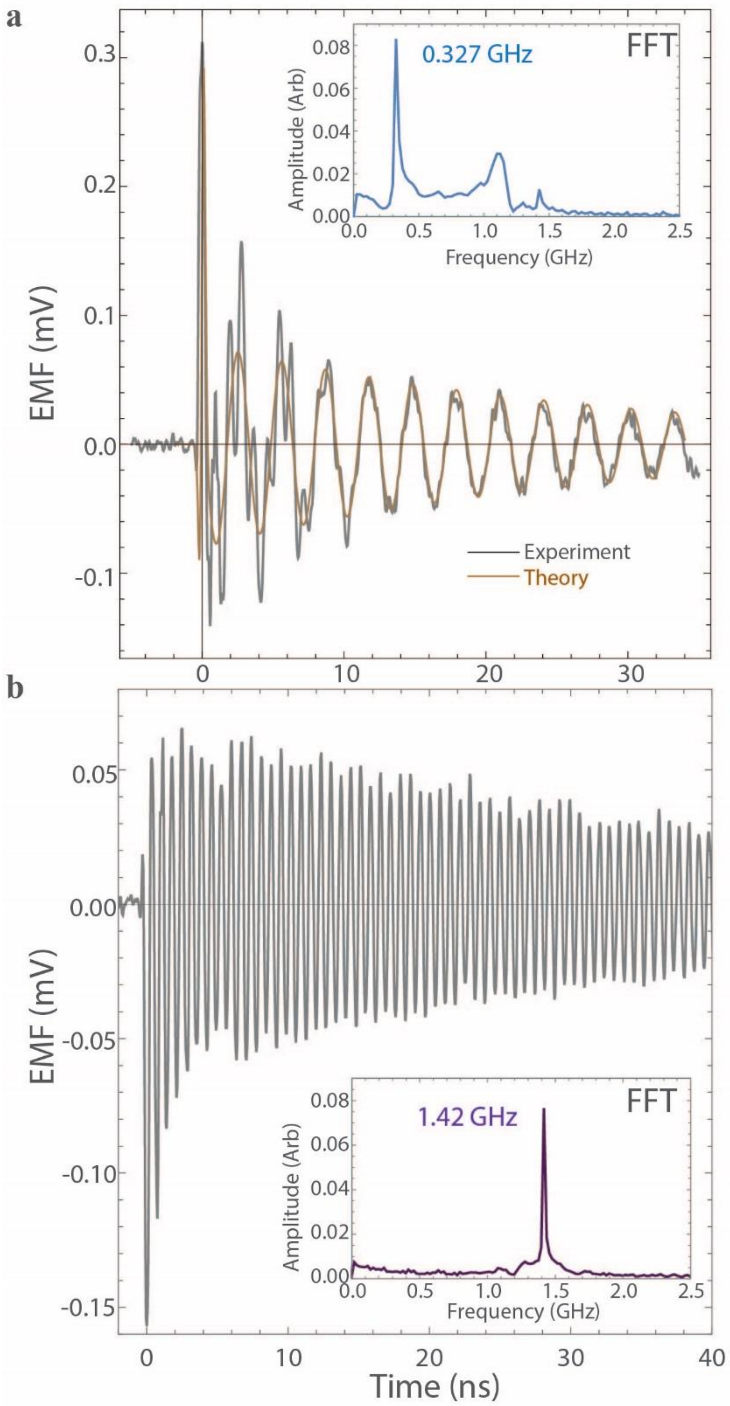

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H, D, nuclear orientation

- High density SPD and SPH
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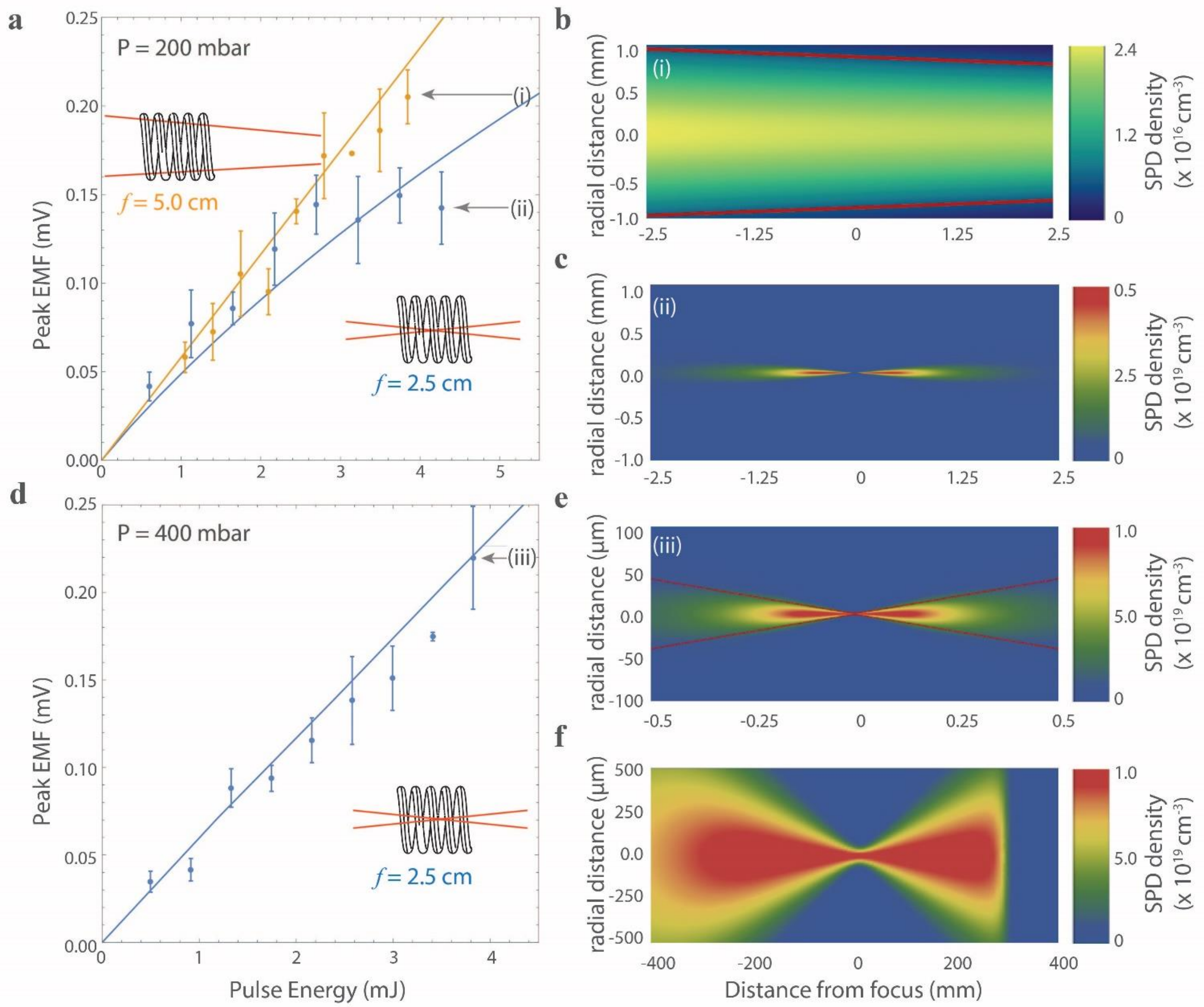
  - Vary laser intensity - multiphoton contributions limited
  - Deplete the signal – densities up to 10^{19} cm^{-3}
  - Simulate for 100 mJ
  - Laser fusion at NIF



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PULSED TARGET – INTENSE PULSE BEAMS

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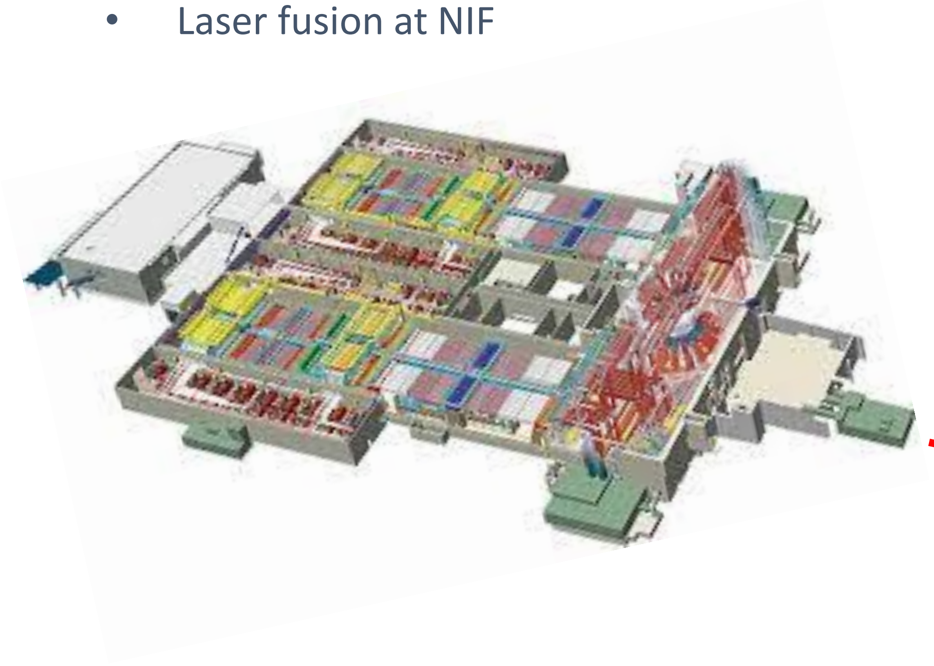

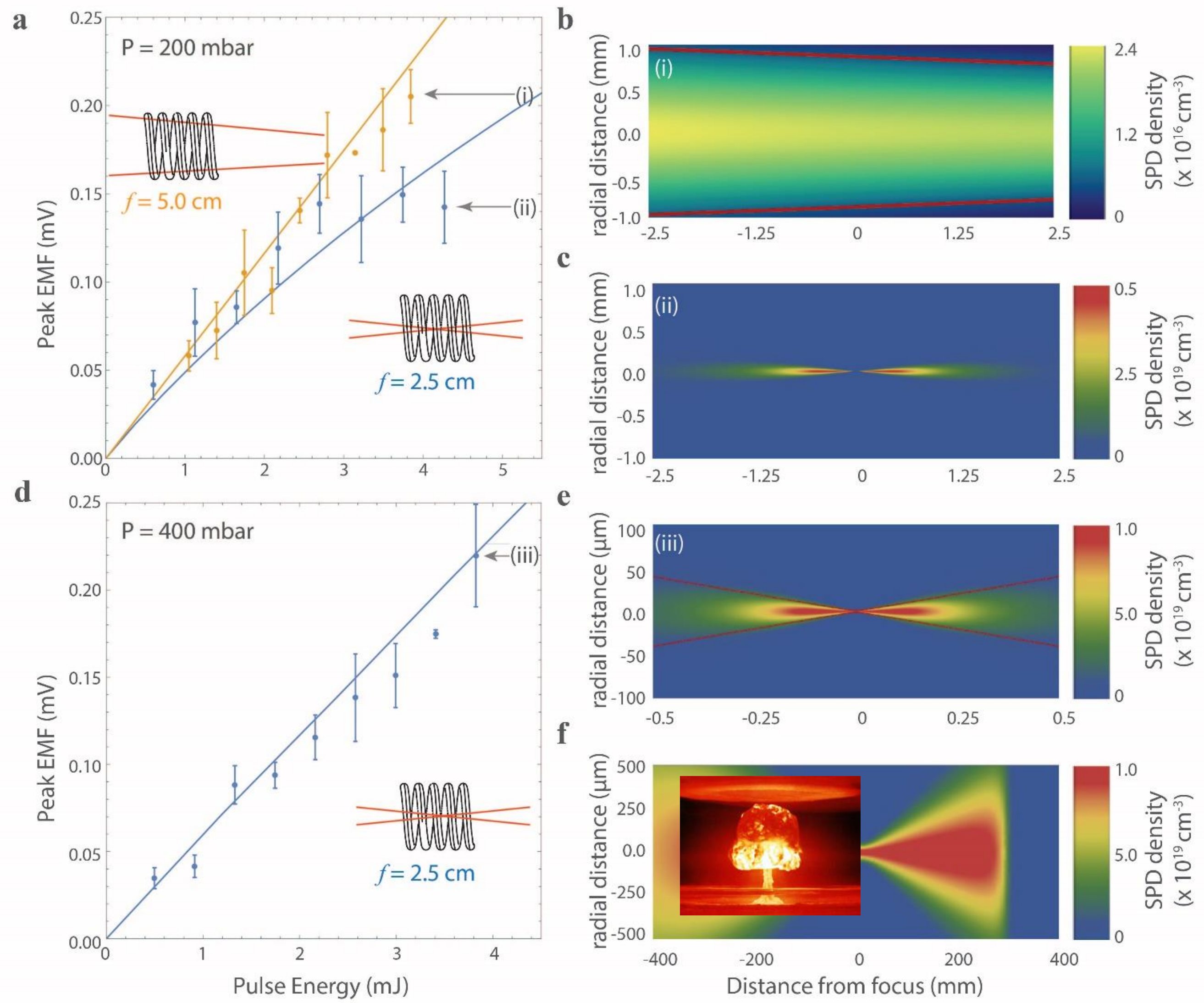

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H, D, nuclear orientation

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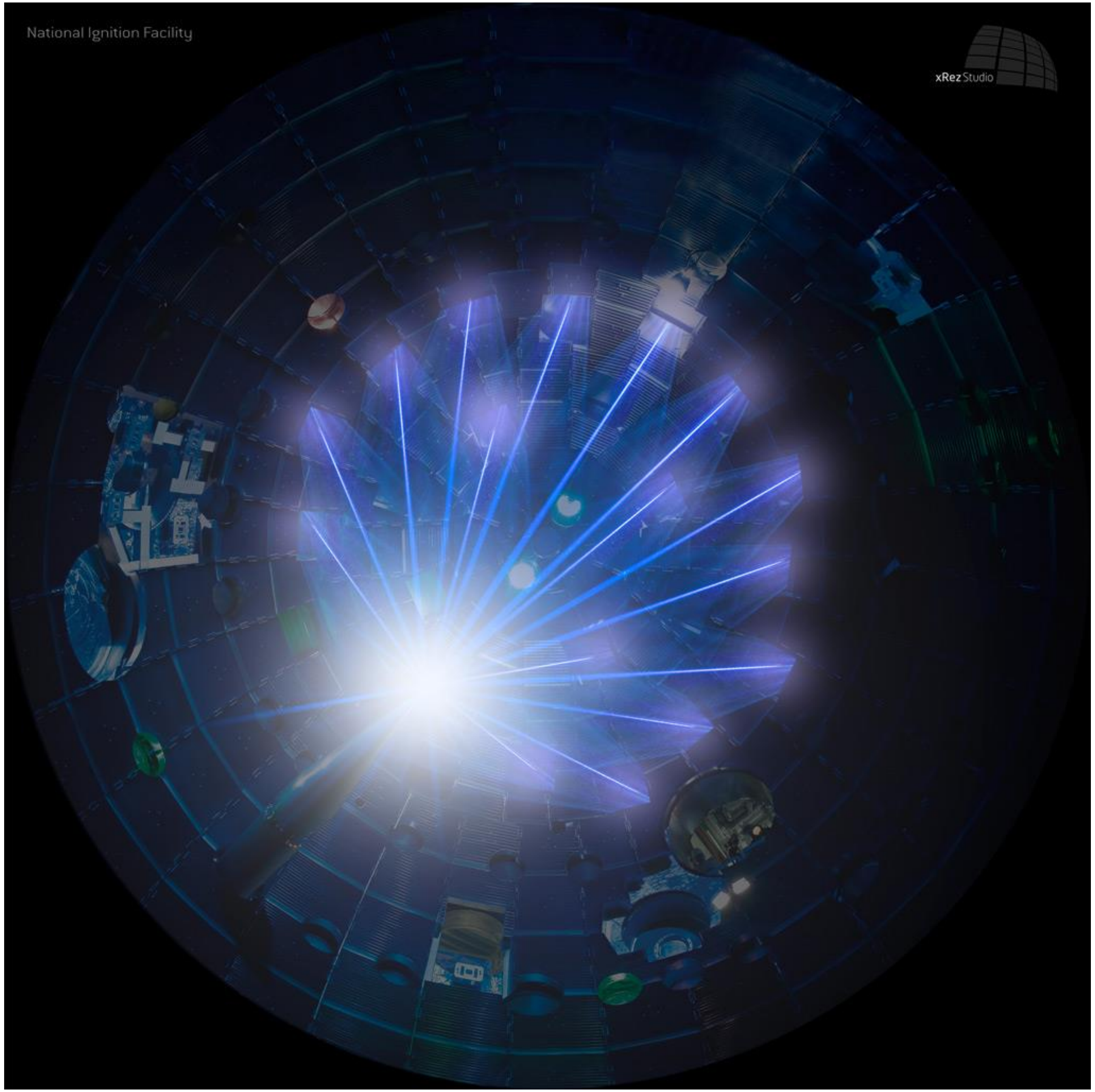
  - Deplete the signal – densities up to 10^{19} cm^{-3}
  - Simulate for 100 mJ
  - Laser fusion at NIF
    - 2 MJ
    - 2.3 ns
    - @350 nm
    - Focus at 100 μm
  - For 10^{18} cm^{-3} → 10^7 (10^5) neutrons/pulse for DT (DD)
  - Sofikitis et al., PRL **118** 233401 (2017)
  - State-of-the-art (magnetic): ~0.01 neutrons/s
  - H. Paetz gen. Schieck Eur. Phys. J. A 44, 321–354 (2010)

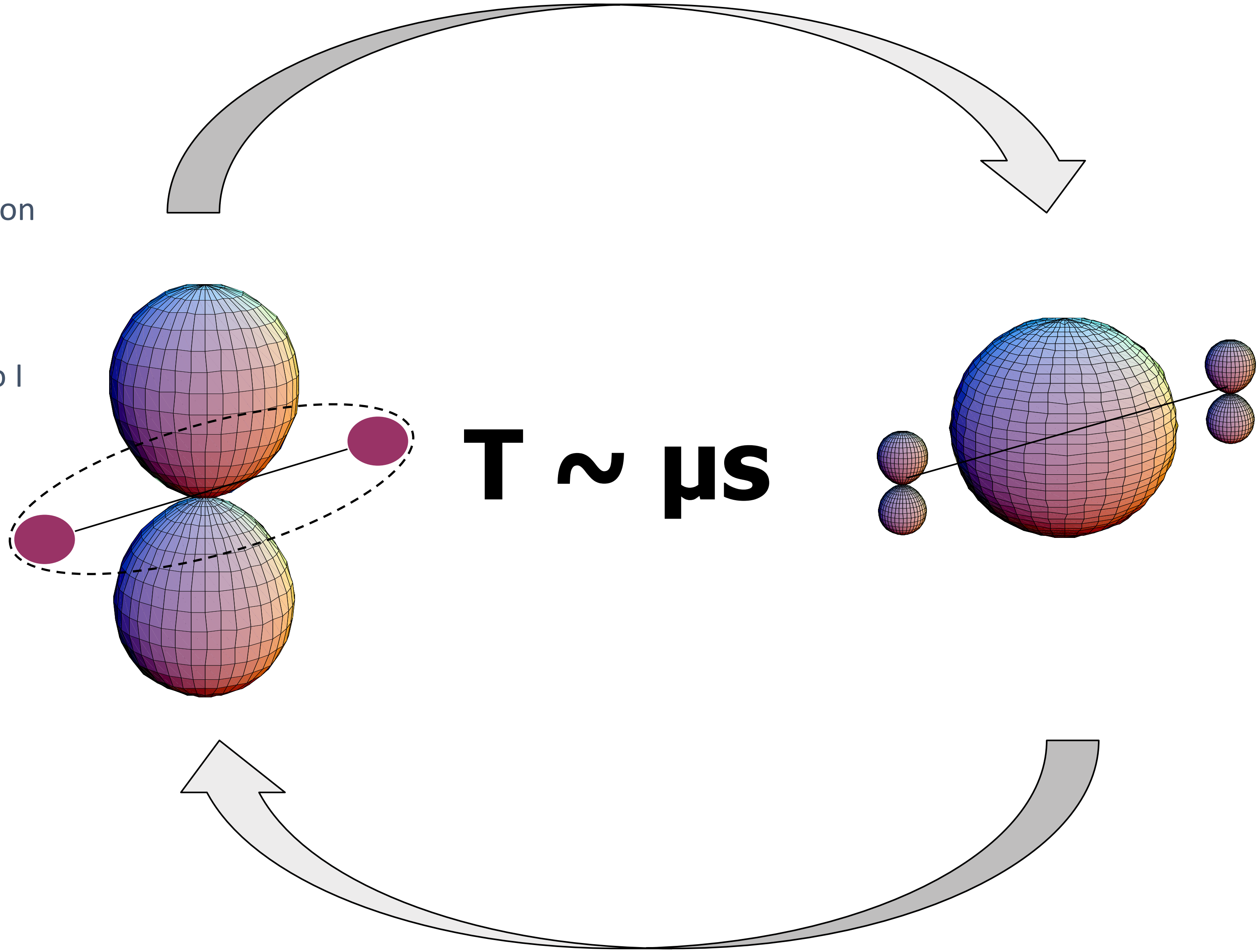


D: What about alignment?

- Obviously simple photodissociation cannot do it
  - Electron is s = ½ → no alignment
- Can we find an 'alignment reservoir' linked with nuclei?
- Molecular rotation

D: What about alignment?

- Molecular rotation
- Aligned in lab frame
  - IR (coherent) excitation
  - (IR pumping)
- HF beating
  - Transfer alignment to I
  - And back
  - microseconds

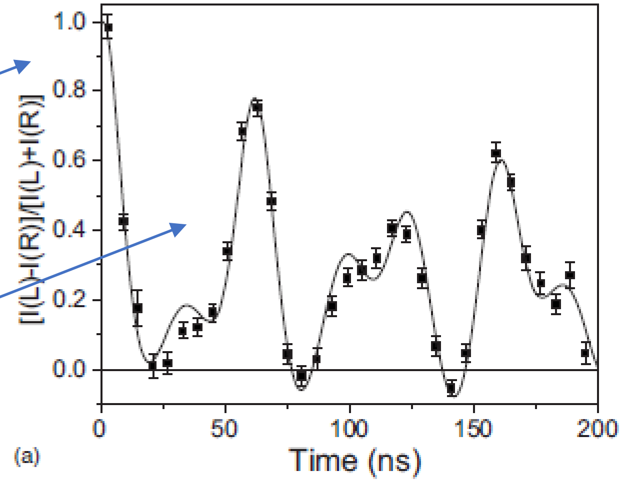


D: What about alignment?

- Polarization transfer from molecular rotation to nuclear spin
- Demonstration with HCl (orientation but still…)
- Sofikitis et al., PRA 76 012503 (2007)

IR (coherent) excitation (t_0)

HF molecular depolarization ($t>t_0$)



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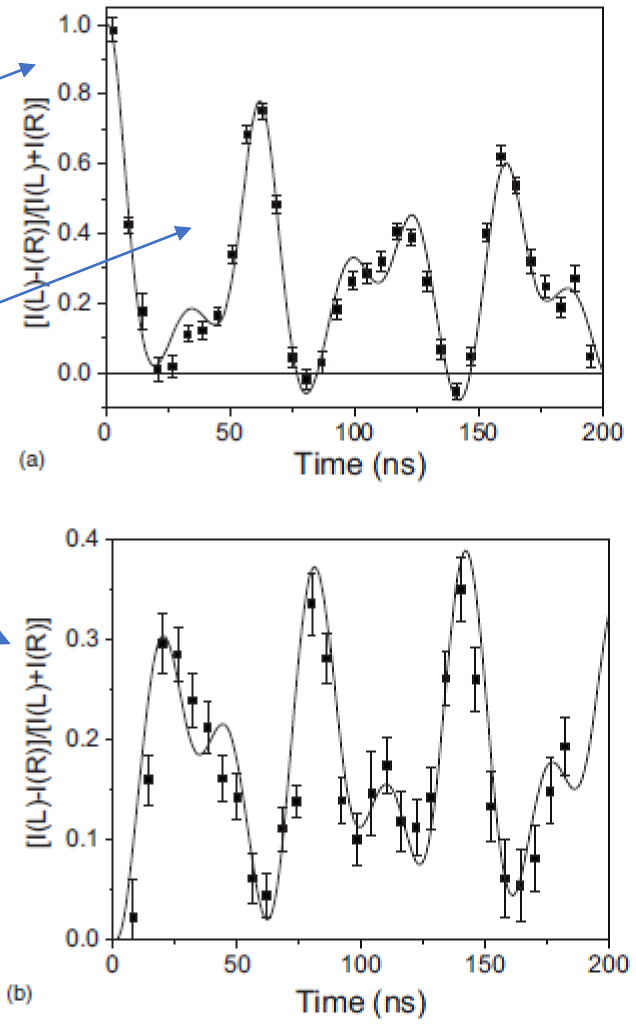
IR (coherent) excitation (t_0)

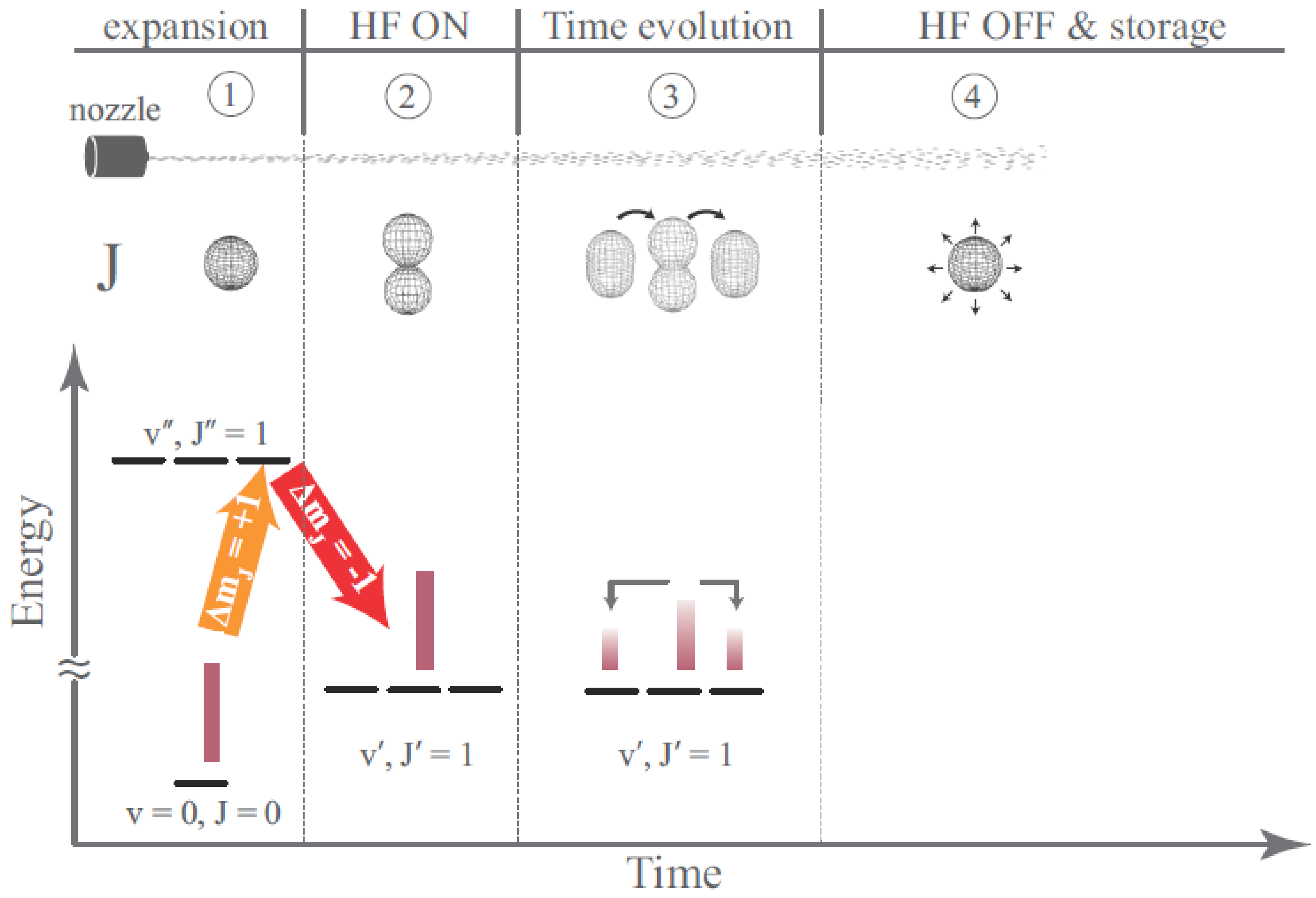
HF molecular depolarization ($t>t_0$)

HF nuclear polarization ($t>t_0$)



D: What about alignment?

- Polarization transfer from molecular rotation to nuclear spin
- A generic experiment:
- (molecular beams)
- Four stages:



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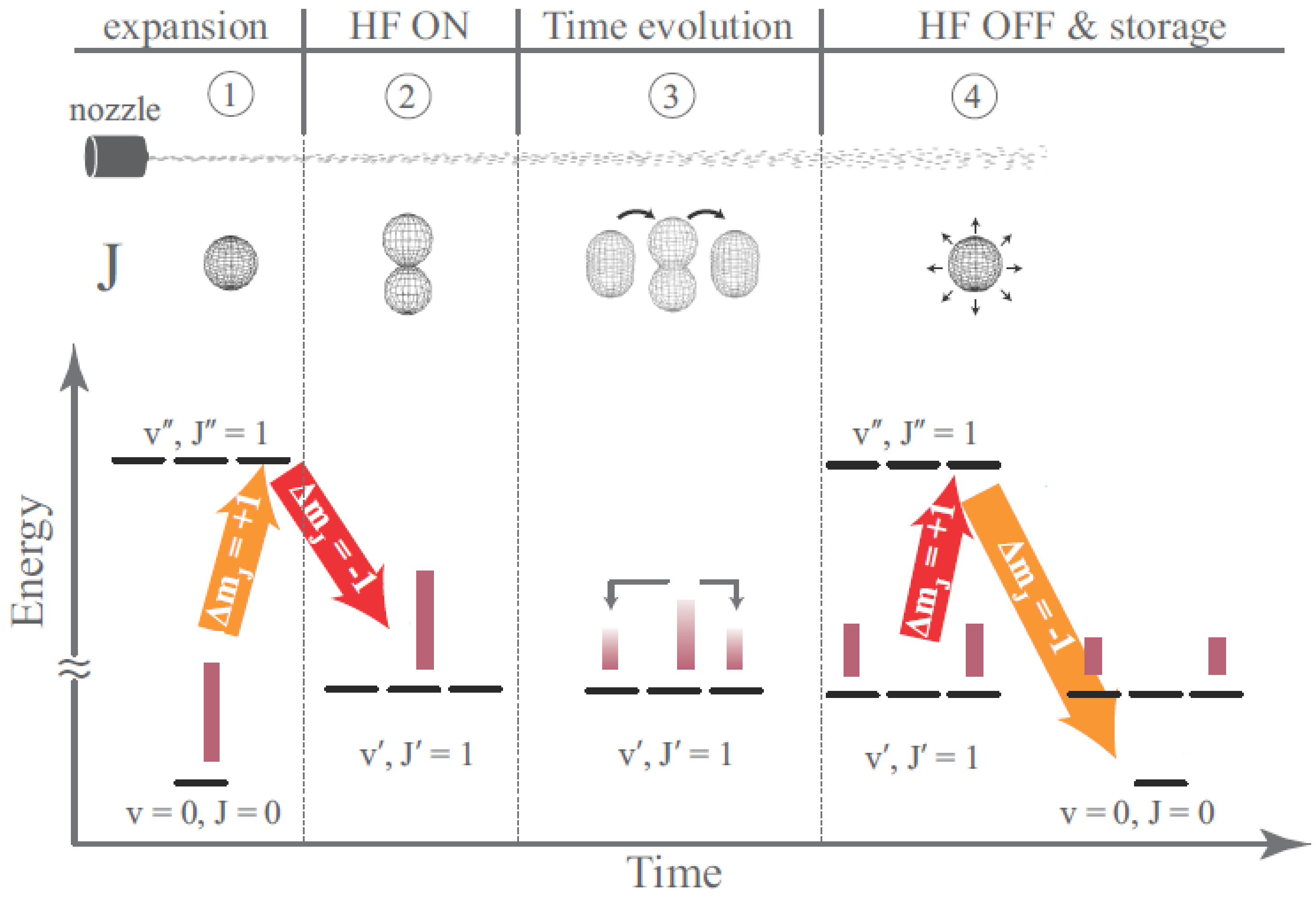
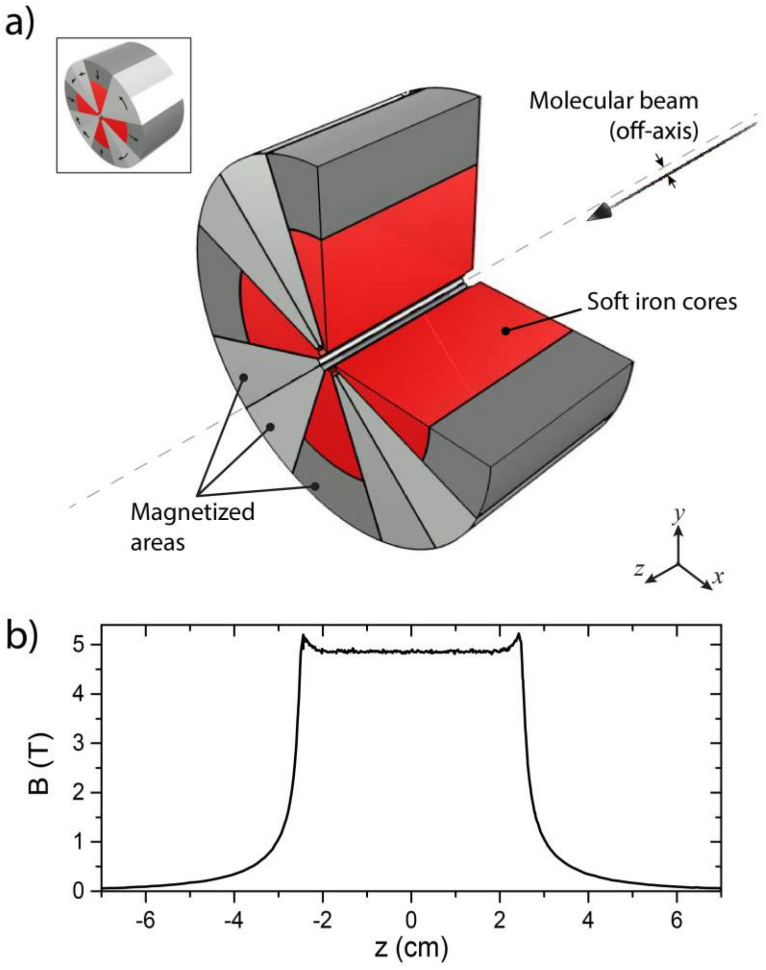


Storage?

- Polarization transfer from molecular rotation to nuclear spin
  - Storage in Hight density – molecules
  - Coupling to J – sensitive to collisions
  - Storage in B fields
  - Decoupling of hyperfine interaction
  - B(t, *l*) is not a step function
  - Make it as close as possible



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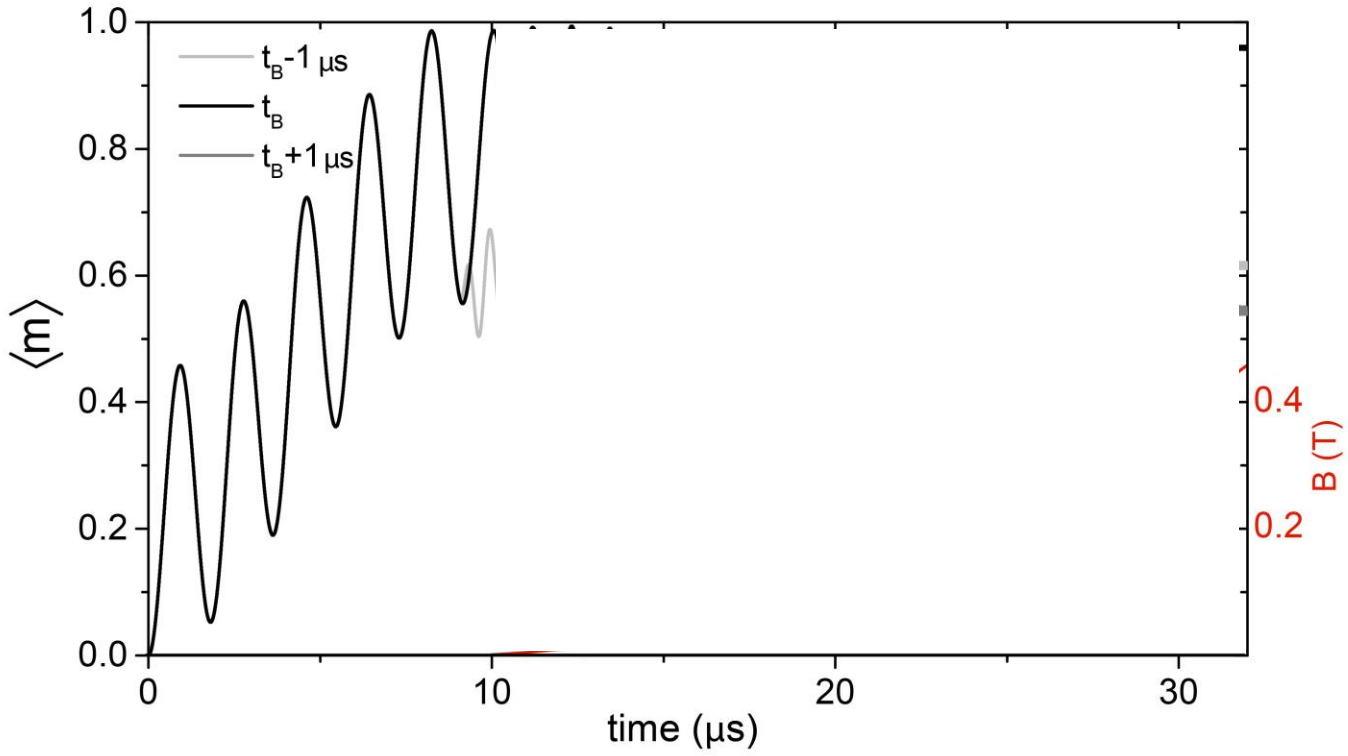
  - Example H_2
  - Timing IR – B field can be critical



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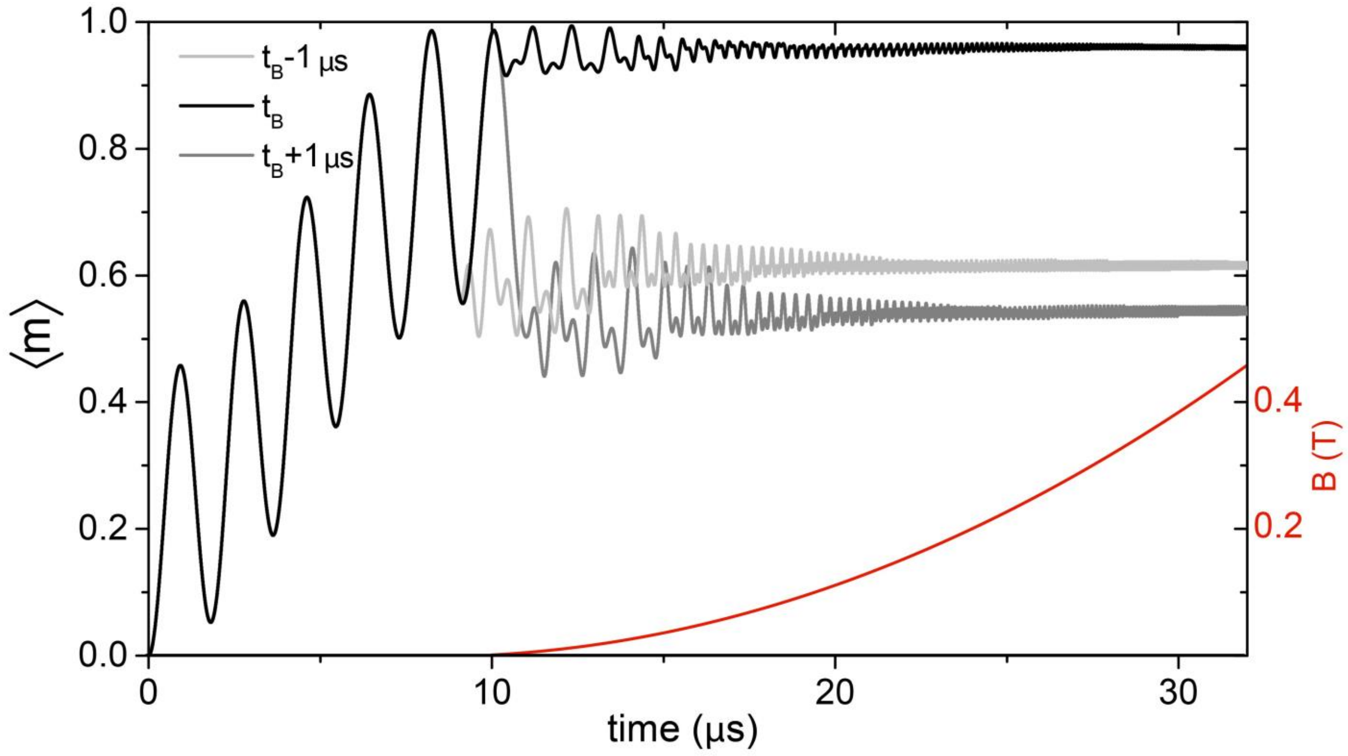

Storage?

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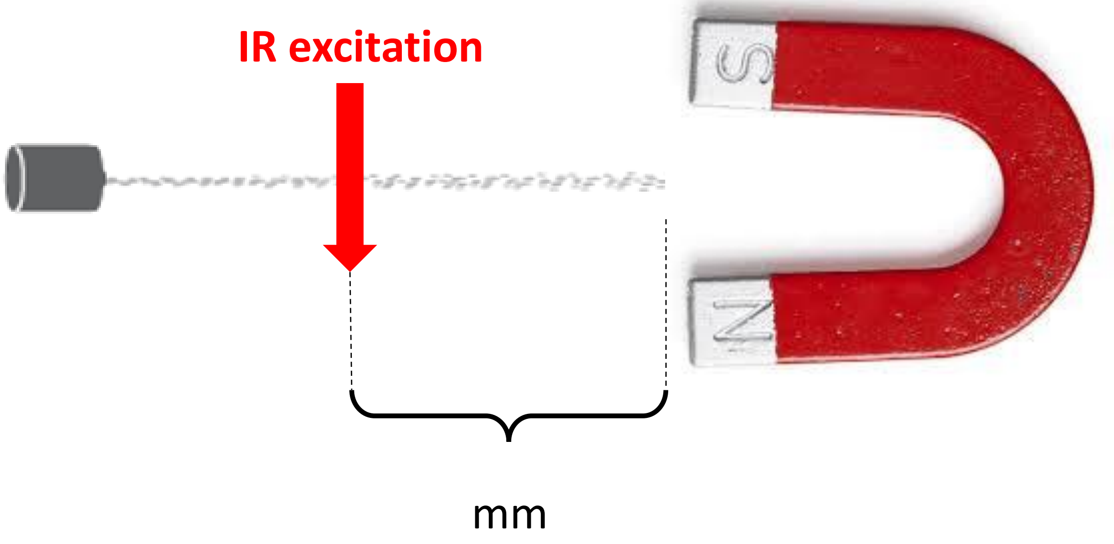
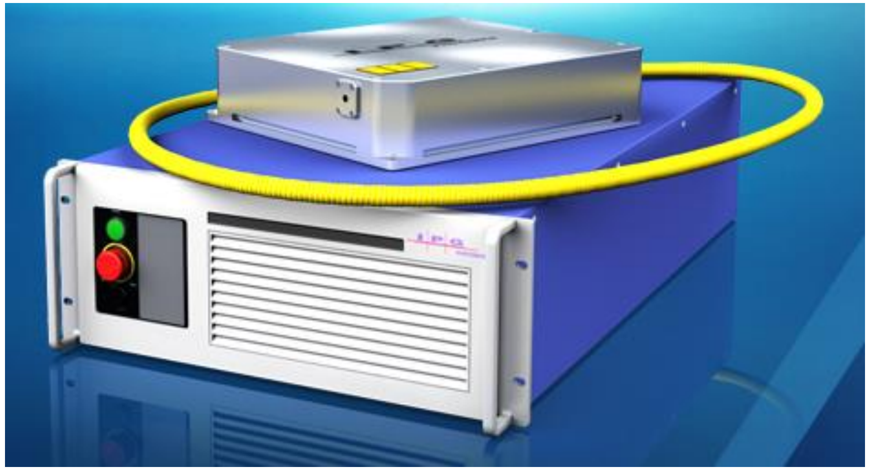
  - Storage in B fields
  - Decoupling of hyperfine interaction
  - B(t, *l*) is not a step function
  - Make it as close as possible
  - Example H_2
  - Timing IR – B field can be critical
  - Experimentally adjust timing
  - v ~ 1000 m/s → mm accuracy



D: Numbers

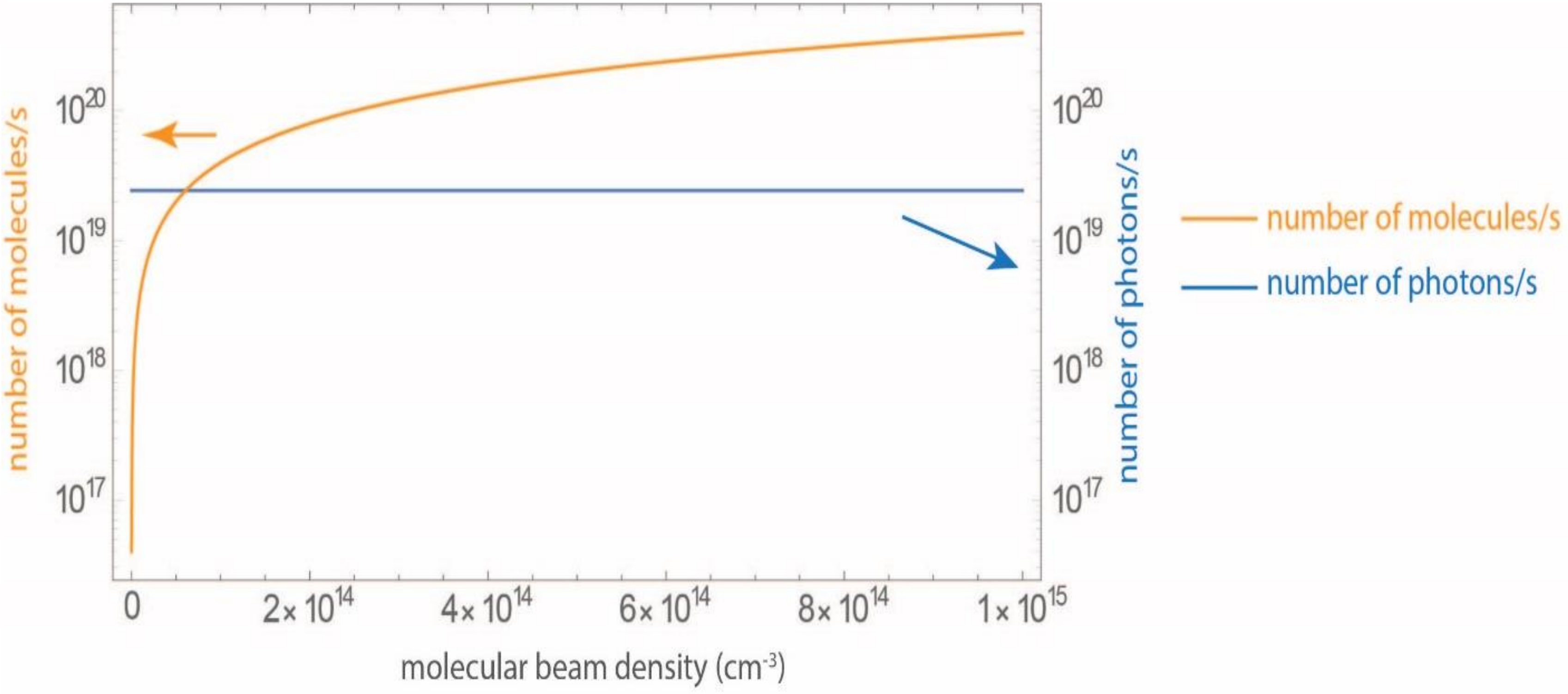
- Polarization transfer from molecular rotation to nuclear spin

  - Tunable lasers IR (pumping) P up to 5 W (λ ~ μm – 10^{19} ph/s)
  - Fixed λ lasers (dissociation) P up to 200 W (UV)
  - Fiber laser technology



D: Numbers

- Polarization transfer from molecular rotation to nuclear spin
  - For an IR power ~ 5 W N_{ph} ~ $5x10^{19}$ photons/s
  - $D_{MB} > 10^{14}$ cm^{-3} → limited by the number of photons
  - IR pumped molecules > 10^{19} mol/s
  - σ_{diss} ~ $10^{-18} – 10^{-17}$ cm^2 → P_{diss} ~ 10^{-2}
  - Flux ~ 10^{15} - 10^{20} SPD/s



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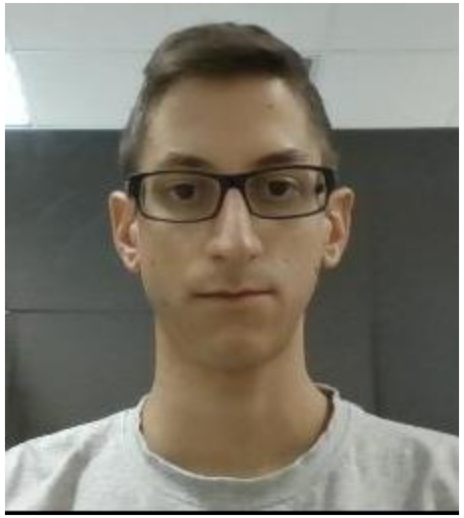
Conclusions:

- Laser-molecule interactions can provide *oriented* and *aligned* H, D
- IR pumping and/or photodissociation
  - Demonstration of 10^{19} SPD/cm^3 (orientation)
  - Alignment from IR pumped molecules
  - High power lasers – high SPD flux (up to 10^{20} SPD/s)
    - Orientation AND alignment
  - Storage in B fields

Stereodynamic effects in the dissociation of small molecules

- Collaborators
  - T. P. Rakitzis IESL FORTH, UOC Physics
  - H. Kannis
  - G. Boulogiannis
  - Luis Rubio Lago



THANK YOU!!!

Points

- Pulsed Targets 10^{19} - 10^{16} per shot- Pulsed intense beams
- Talk about particles per second
- Tune B field in cm

For the D-T or D-^{3}He nuclear fusion reactions, the angular distribution of the neutron or proton products $D(\theta,\phi)$ about the quantization axis, as a function of the nuclear vector polarizations p_z of D, T, or ^{3}He is well approximated by [25]:

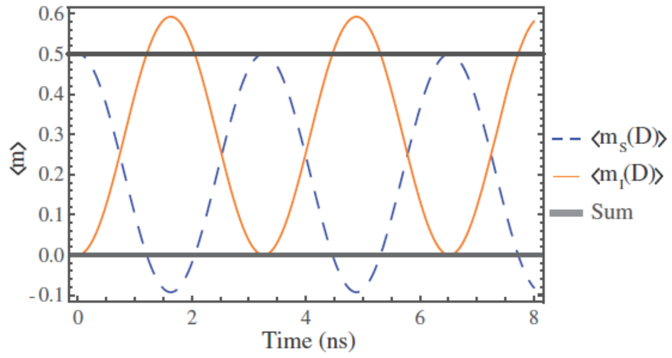
$$D(\theta,\phi) = \frac{\sigma_0}{3}[(2+p) - (2p + p_{zz})P_2(cos\theta)]/4\pi \qquad (10)$$

where $p = p_z(\mathrm{D})\mathrm{p_z}(\mathrm{Y})$, Y = T or ^{3}He, p_z is the nuclear vector polarization, p_{zz} is the tensor polarization for D nuclei, σ_0 is the fusion cross section through the intermediate ^{5}He or $^5\mathrm{Li}\frac{3}{2}^+$ state for the D-T and D-^{3}He reactions, respectively, and $P_2(x)$ is the 2$^{\mathrm{nd}}$ Legendre Polynomial. The first term in Eq. (10) is proportional to the integrated product signal, so that for maximal nuclear polarization, with $p = 1$, the product integrated intensity is increased by 50% compared to $p = 0$; also, in this case $p_{zz} = 1$, and hence $D(\theta,\phi) \sim 1 - P_2(cos\theta) \sim sin^2\theta$.

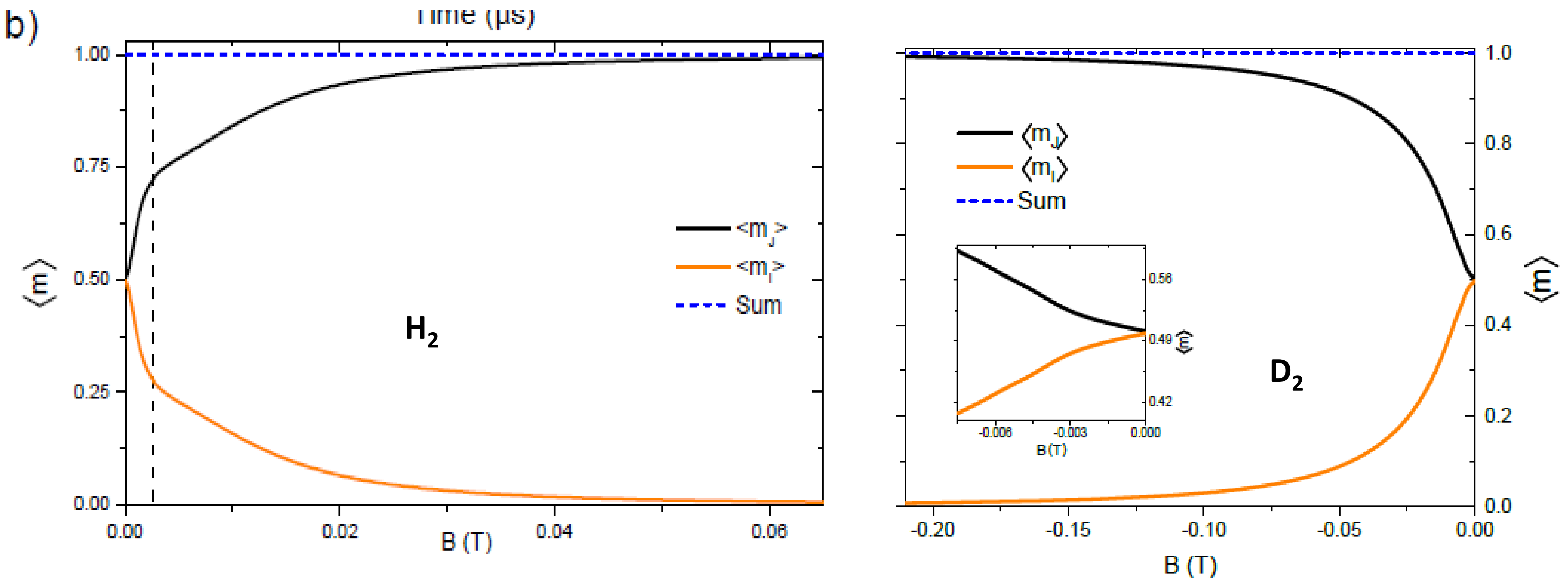
For the D-^{3}He reaction performed with $p_z(^3\mathrm{He}) = 0.76$ [26, 27], $p_z(D, t = 1.5\,ns) = 0.12$ (reported here), and $p_{zz}(D) = 0$, we predict a 14% variation in the angular distribution (between $\theta = 0°$ and $90°$) and a 4.5% increase in the integrated intensity, whereas if bond alignment is used prior to dissociation [28], $p_z(D, t = 1.5\,ns) = 0.5$ can be produced [8], leading to a 70% variation in the product angular distribution and a 19% increase in integrated intensity. In contrast, the effect of

$$\langle m_S(\mathrm{D})\rangle = \frac{16}{27}\sin^2\left(\frac{\Delta E}{2\hbar}t\right),$$

$$\langle m_I(\mathrm{D})\rangle = \frac{1}{2} - \frac{16}{27}\sin^2\left(\frac{\Delta E}{2\hbar}t\right),$$

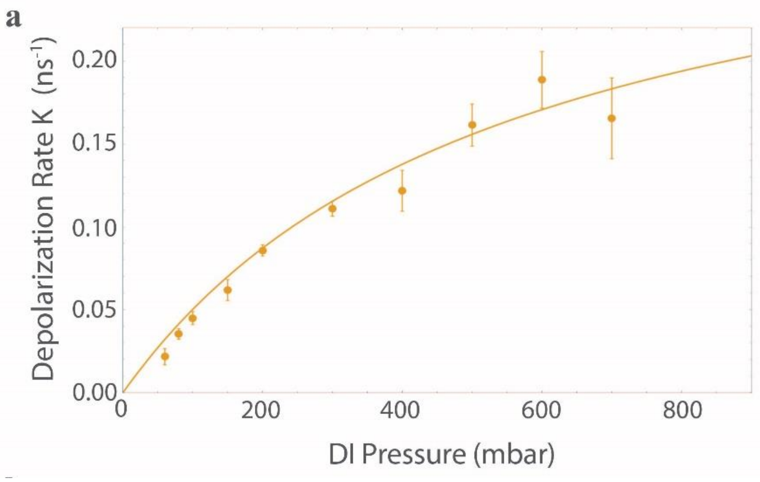


Laser-fusion neutron-yield calculations were performed using a modified [46] MEDUSA code [47], which include the following physical processes: initial laser pulse absorption, propagation of a heat wave, ionization, one-dimensional hydrodynamics of plasma by two-temperature approximation, nonlinear thermal conductivity and limitation, classical and resonant absorption of laser radiation, thermal radiation emission and its absorption by plasma, and thermonuclear reaction yield. The calculations predict that the irradiation of 10^{18} cm^{-3} deuterium and tritium atoms and 10^{19} cm^{-3} ^{3}He with a 2 MJ, 2.3 ns pulse at 350 nm focused to ~100 μm (National Ignition Facility at Lawrence Livermore National Laboratory) will heat the resulting ions to average collision (thermal) energies of ~10 keV and lead to the production of ~10^7 neutrons/pulse from the D-T reaction and ~10^5 neutrons/pulse each from the D-D and D-^{3}He reactions. Irradiation with a 6 kJ (with $\lambda \sim 1$ μm), 1 ps laser pulse [48,49], focused to 10 μm, will produce ~10^4 neutrons/pulse from D-T reactions and ~100 neutrons/pulse from D-D or D-^{3}He fusion reactions. These neutron yields compare well with the ones foreseen for ongoing polarized fusion experiments, of ~0.01 neutrons/s [50], and show that the study of polarized fusion with high signals is possible, using high-density SPD and laser-initiated fusion.



b)
Time (μs)
1.00
0.75
0.50
0.25
0.00
⟨m⟩
0.00
0.02
0.04
0.06
B (T)
<m_J>
<m_I>
Sum
H2
1.0
0.8
0.6
0.4
0.2
0.0
⟨m⟩
-0.20
-0.15
-0.10
-0.05
0.00
B (T)
⟨m_J⟩
⟨m_I⟩
Sum
0.56
0.49
0.42
-0.006
-0.003
0.000
B (T)
D2

H, D, nuclear orientation



- High density SPD and SPH
  - Sofikitis et al., Phys. Rev. Lett. 121, 083001 (2018)
  - But 10^{19} cm^{-3}???
  - 10^{17} cm^{-3} projection done using alkali-H cross sections
  - Depolarization – no two body process
  - Simplest process fitting the data
  - Inert gas breaks the complex (SF_6)
  - Minimum depolarization from I

$$\mathrm{D}^{\uparrow} + \mathrm{DI} \underset{\mathrm{k}_{-1}}{\overset{\mathrm{k}_1}{\rightleftharpoons}} \mathrm{DI{-}D}^{\uparrow}$$

Complex formation

$$\mathrm{D}^{\uparrow} + \mathrm{DI} \xrightarrow{k} \mathrm{D} + \mathrm{DI}$$

Direct depolarization DI

$$\mathrm{DI{-}D}^{\uparrow} \xrightarrow{\mathrm{k}_d} \mathrm{DI{-}D}$$

intramolecular

$$\mathrm{DI{-}D}^{\uparrow} + \mathrm{DI} \xrightarrow{\mathrm{k}_2} \mathrm{D}^{\uparrow} + 2\,\mathrm{DI}$$

Collisional dissociation DI

$$K = \frac{k_1 k_d [DI]}{k_{-1} + k_d + k_2[DI]}$$

- Dimitris Sofikitis