DEVELOPMENT OF POLARIZED SOURCES BASED ON MOLECULAR PHOTODISSOCIATION

29.09.2022 I Chrysovalantis Kannis

Workshop on Polarized Sources Targets and Polarimetry 2022 (PSTP22)

Session: Polarized Sources

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Why do we need polarized sources?

- **Accelerators**
  1. Conventional accelerators
  2. Laser-plasma accelerators
- **Polarized nuclear fusion**
  1. Four-nucleon reactions

$$\mathrm{d} + \mathrm{d} \longrightarrow \mathrm{n} + {}^{3}\mathrm{He} + 3.27\,\mathrm{MeV}$$ (neutron branch)

$$\mathrm{d} + \mathrm{d} \longrightarrow \mathrm{p} + {}^{3}\mathrm{H} + 4.03\,\mathrm{MeV}$$ (proton branch)

→
- poorly understood
- direct measurements are required

  2. Five-nucleon reactions

$$\mathrm{d} + {}^{3}\mathrm{H} \longrightarrow \mathrm{n} + {}^{4}\mathrm{He} + 17.58\,\mathrm{MeV}$$

$$\mathrm{d} + {}^{3}\mathrm{He} \longrightarrow \mathrm{p} + {}^{4}\mathrm{He} + 18.34\,\mathrm{MeV}$$

→
- control the angular distribution of products
- increase the reaction rate

Why do we need polarized sources?

- **Polarized nuclear fusion**

  ii. Five-nucleon reactions

  - Angular distribution of fusion products: $W(\theta) = 1 - \frac{1}{2} P_z^d P_z^y + \frac{3}{2} P_z^d P_z^y \sin^2\theta + \frac{1}{4} P_{zz}\left(1 - 3\cos^2\theta\right)$,

    where $y = {}^3H$ or ${}^3He$ and the polarization factors:

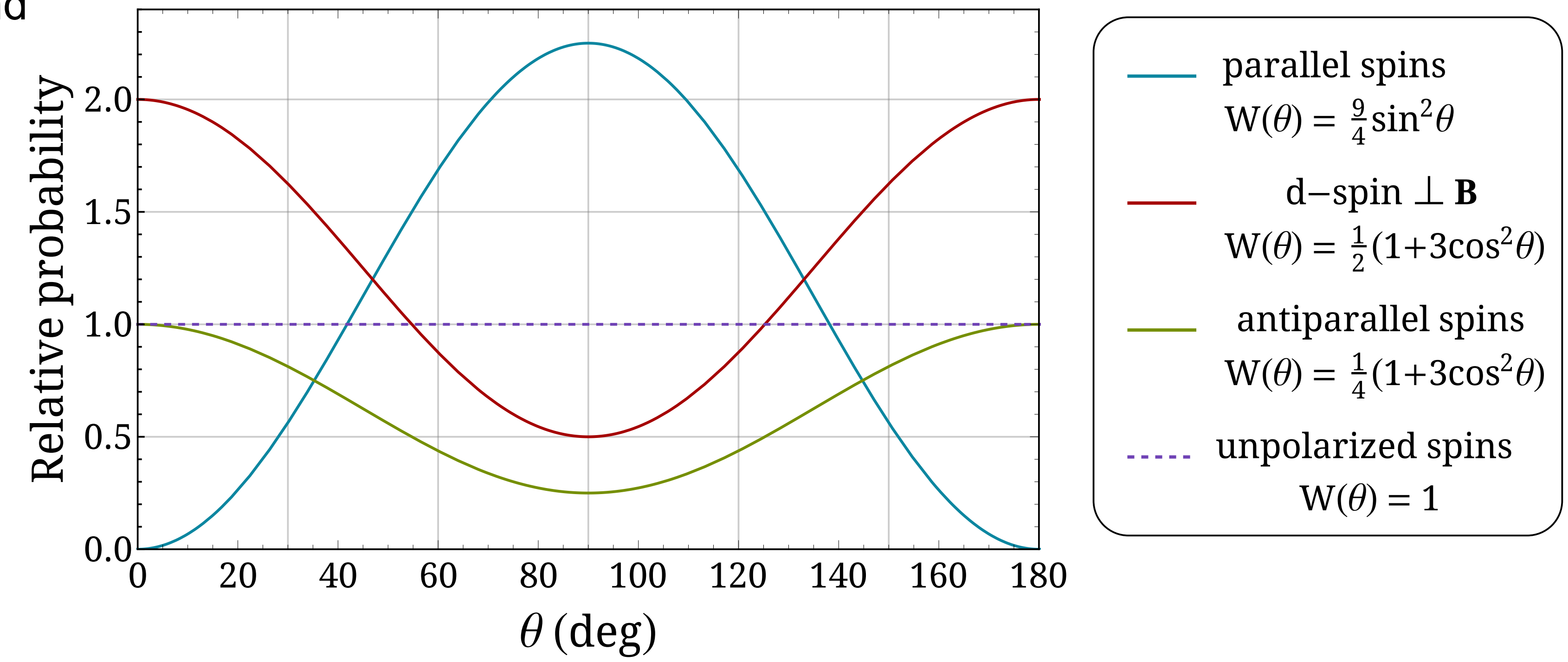
    $$P_z^d = \frac{N_{+1} - N_{-1}}{N_{+1} + N_{-1} + N_0}$$

    $$P_z^y = \frac{N_{+1/2} - N_{-1/2}}{N_{+1/2} + N_{-1/2}}$$

    $$P_{zz} = \frac{N_{+1} + N_{-1} - 2N_0}{N_{+1} + N_{-1} + N_0}$$

    with $P_z \epsilon [-1, 1]$

    and $P_{zz} \epsilon [-2, 1]$.



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Why do we need polarized sources?

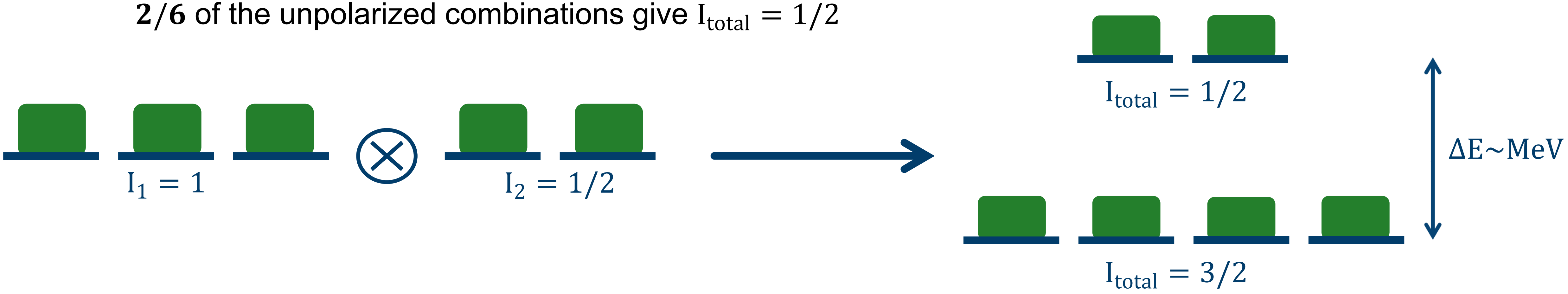
- **Polarized nuclear fusion**

  ii. Five-nucleon reactions

  - Fusion rate for unpolarized reactants:

    $\mathbf{4/6}$ of the unpolarized combinations give $\mathrm{I_{total}} = 3/2$

    $\mathbf{2/6}$ of the unpolarized combinations give $\mathrm{I_{total}} = 1/2$



Why do we need polarized sources?

- **Polarized nuclear fusion**

  ii. Five-nucleon reactions

  - Fusion rate for unpolarized reactants: $\mathbf{2/3}$

    $\mathbf{4/6}$ of the unpolarized combinations give $I_{total} = 3/2$

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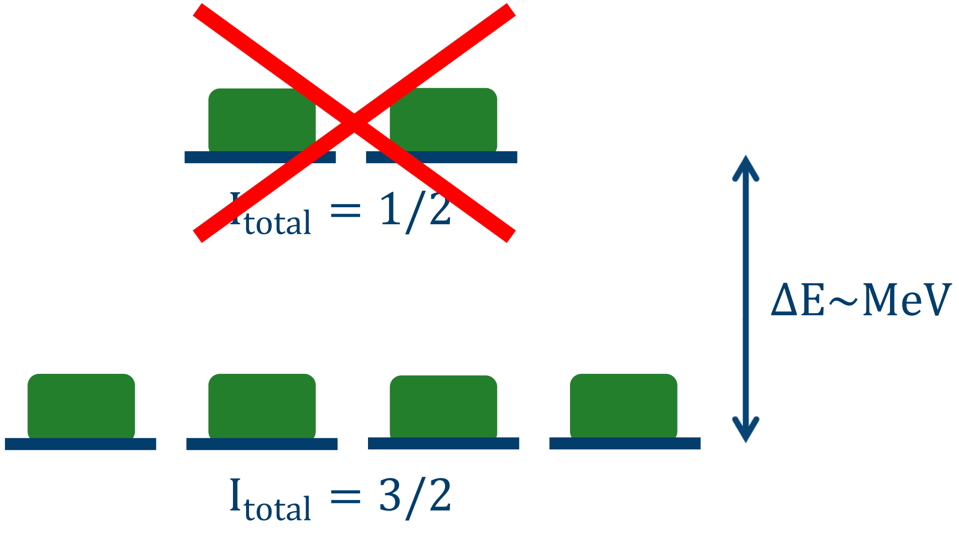


Only $\mathbf{4/6}$ of the unpolarized substates contribute to the fusion reaction!

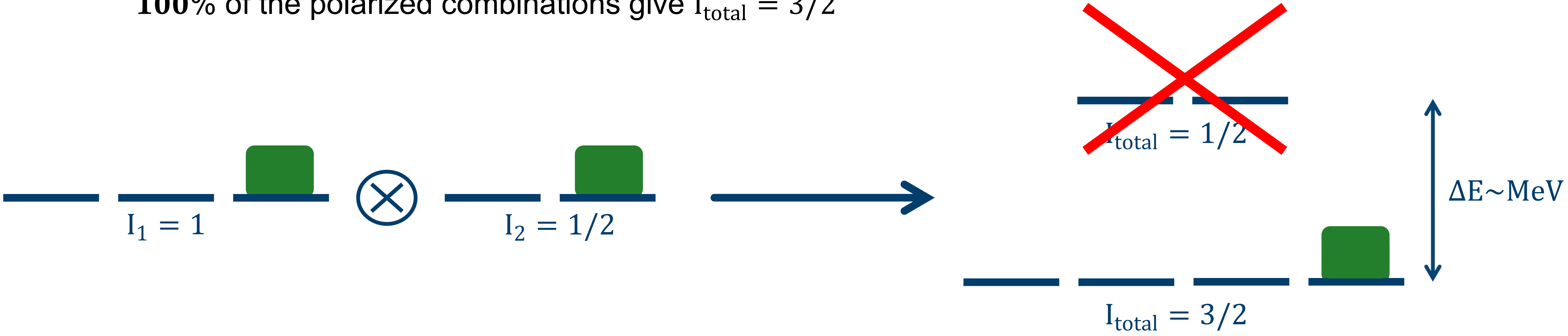
Why do we need polarized sources?

- **Polarized nuclear fusion**

  ii. Five-nucleon reactions

  - Fusion rate for unpolarized reactants: $\mathbf{1}$

  $\mathbf{100\%}$ of the polarized combinations give $I_{total} = 3/2$



Increased efficiency by $\mathbf{50\%}$!

Conventional polarization methods

- **Stern-Gerlach spin-separation**

  required time for spin-separation: ms

  limited by the beam divergence

  density limit: $\sim 10^{12}$ cm^{-3}

  highest flux: $\sim 10^{17}$ H/s

- **Spin-exchange optical pumping**

  two-step process: (i) optical pumping of a mediating species

  (ii) polarization transfer through spin-exchange collisions

  characteristic time in a spin-exchange cell: ms

  highest density: $\sim 10^{14}$ cm^{-3} (low polarization)

  highest flux: $\sim 10^{18}$ H/s (low polarization)

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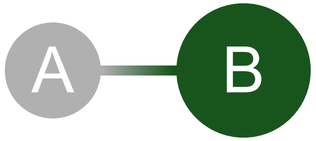
- **Molecular photodissociation**

  in situ production

  production time: ns

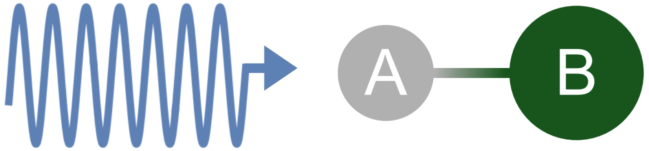
  density $> 10^{19}$ cm^{-3}

Mechanism of molecular photodissociation



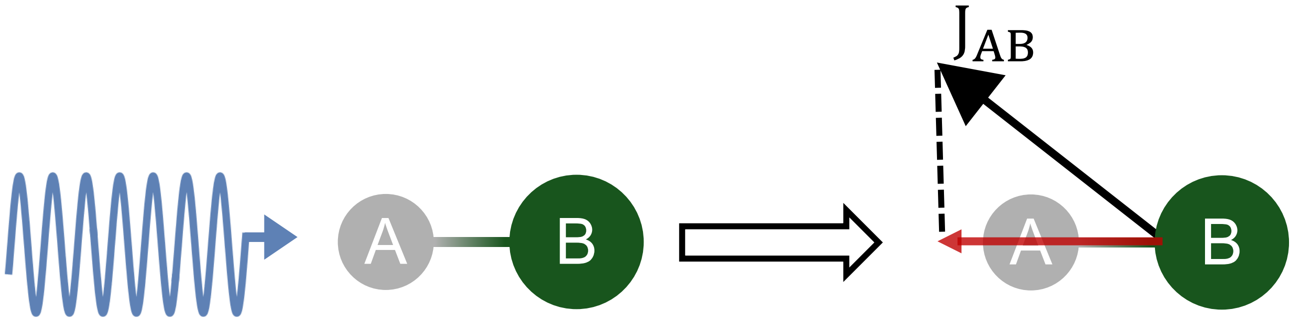
Ω_g

Mechanism of molecular photodissociation



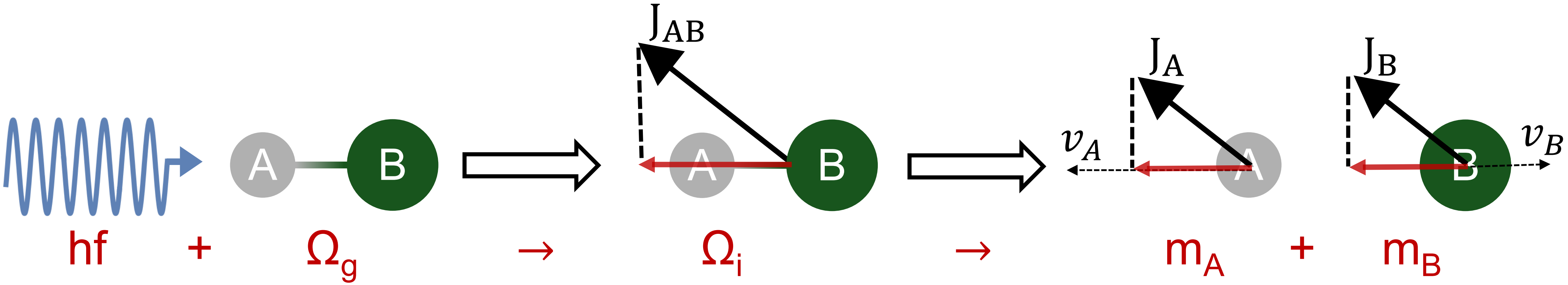
hf + Ω_g

Mechanism of molecular photodissociation

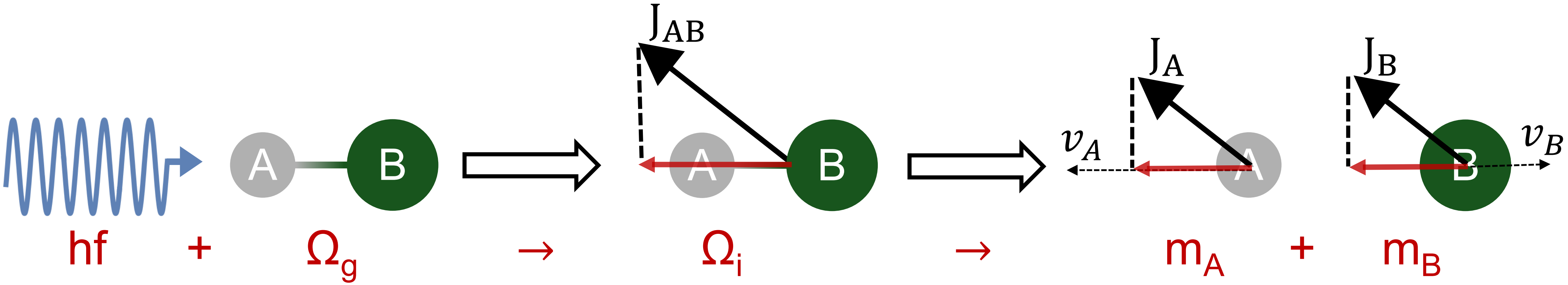


$hf + \Omega_g \rightarrow \Omega_i$

Mechanism of molecular photodissociation



Mechanism of molecular photodissociation



$$hf + AB\,(\Omega_g) \rightarrow AB\,(\Omega_i) \rightarrow A\,(J_A, m_A) + B\,(J_B, m_B)$$

Ω_i is the **projection** of the total electronic angular momentum of electronic state i along the AB bond axis.
m is the **projection** of atomic angular momentum J along the bond axis.
Conservation of angular momentum **projection** along the recoil direction yields the constraint:

$$\Omega_i = m_A + m_B$$

Mechanism of molecular photodissociation

Hydrogen halides (HY)

- 3 excited electronic states play a role in photodissociation:

$$\mathrm{HY}\left(\mathrm{X}\ {}^1\Sigma_{0^+};\ \Omega_g = 0\right) + hf \rightarrow \mathrm{HY}\left(\alpha\ {}^3\Pi_1;\ \Omega_i = \pm 1\right) \rightarrow \mathrm{H}\left(m_H = \pm\tfrac{1}{2}\right) + \mathrm{Y}\left(m_Y = \pm\tfrac{1}{2}\right)$$

$$\mathrm{HY}\left(\mathrm{A}\ {}^1\Pi_1;\ \Omega_i = \pm 1\right) \rightarrow \mathrm{H}\left(m_H = \mp\tfrac{1}{2}\right) + \mathrm{Y}\left(m_Y = \pm\tfrac{3}{2}\right)$$

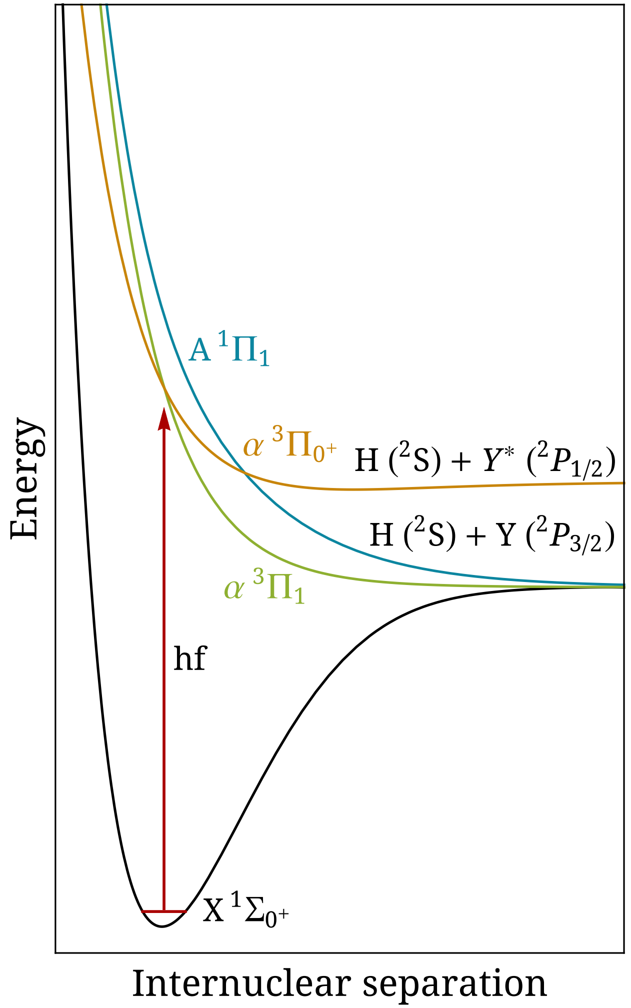
$$\mathrm{HY}\left(\alpha\ {}^3\Pi_{0^+};\ \Omega_i = 0\right) \rightarrow \mathrm{H}\left(m_H = \pm\tfrac{1}{2}\right) + \mathrm{Y}^*\left(m_{Y^*} = \mp\tfrac{1}{2}\right)$$

- Photodissociation of HY can occur exclusively through one of these states:

$$\mathrm{HI}\left(\mathrm{X}\ {}^1\Sigma_{0^+};\ \Omega_g = 0\right) + hf\,(213\ \mathrm{nm}) \xrightarrow{\mathrm{A}\ {}^1\Pi_1} \mathrm{H}\left(m_H = \mp\tfrac{1}{2}\right) + \mathrm{I}\left(m_I = \pm\tfrac{3}{2}\right)$$

$$\mathrm{HI}\left(\mathrm{X}\ {}^1\Sigma_{0^+};\ \Omega_g = 0\right) + hf\,(266\ \mathrm{nm}) \xrightarrow{\alpha\ {}^3\Pi_1} \mathrm{H}\left(m_H = \pm\tfrac{1}{2}\right) + \mathrm{I}\left(m_I = \pm\tfrac{1}{2}\right)$$

$$\mathrm{HCl}\left(\mathrm{X}\ {}^1\Sigma_{0^+};\ \Omega_g = 0\right) + hf\,(213\ \mathrm{nm}) \xrightarrow{\alpha\ {}^3\Pi_1} \mathrm{H}\left(m_H = \pm\tfrac{1}{2}\right) + \mathrm{Cl}\left(m_I = \pm\tfrac{1}{2}\right)$$



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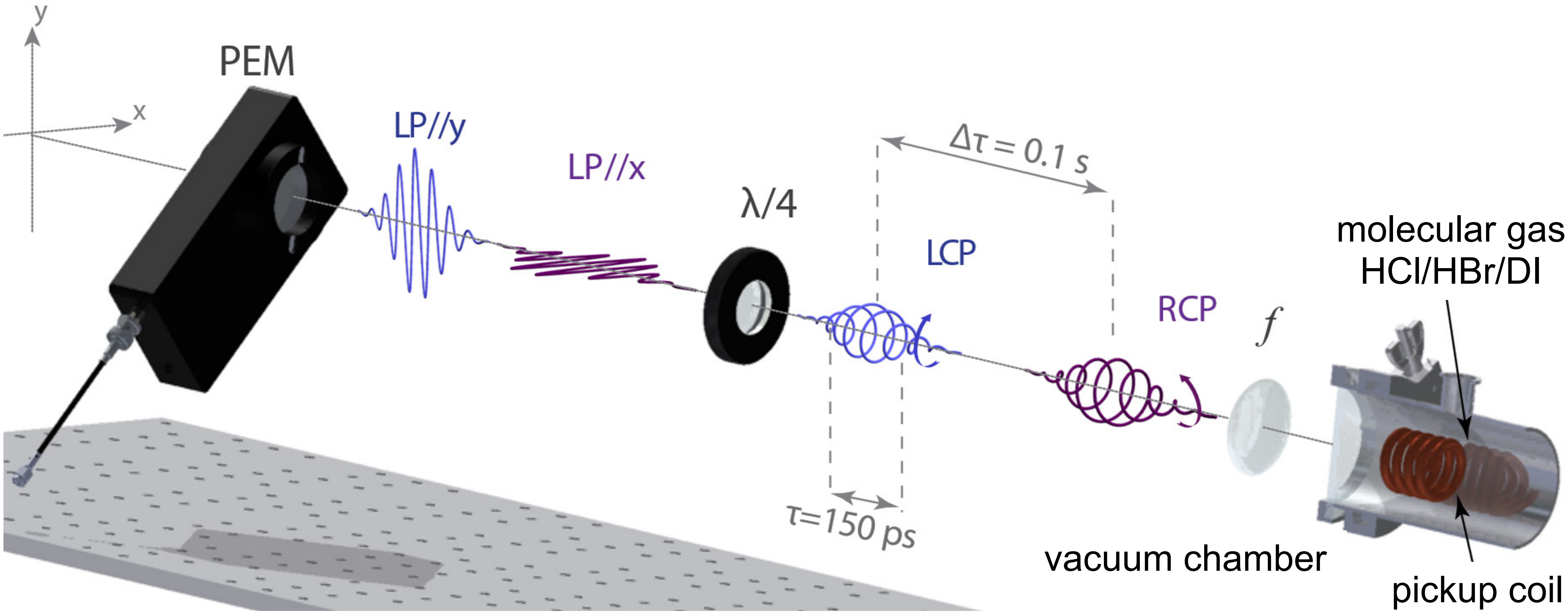
High-density spin-polarized H and D atoms

Produced from photodissociation and detected with a pickup coil



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

High-density spin-polarized H and D atoms

Detection of magnetization quantum beats with a pickup coil

- Magnetic moment of hydrogen halides (HY) in the ground state: $\boldsymbol{\mu}_{\mathbf{mol}} = \boldsymbol{\mu}_{\mathbf{I_H}} + \boldsymbol{\mu}_{\mathbf{I_Y}} = g_{I_H}\mu_N \mathbf{I_H} + g_{I_Y}\mu_N \mathbf{I_Y}$
- Magnetic moment of hydrogen in the ground state: $\boldsymbol{\mu}_{\mathbf{H}} = \boldsymbol{\mu}_{\mathbf{S}} + \boldsymbol{\mu}_{\mathbf{I_H}} = g_S\mu_B \mathbf{S} + g_{I_H}\mu_N \mathbf{I_H}$

$\mu_{B,N}$ are the Bohr and nuclear magnetons $\begin{cases} \mu_B = 9.274 \times 10^{-24}\ \text{J/T} \\ \mu_N = 5.051 \times 10^{-27}\ \text{J/T} \end{cases}$

g_{S,I_H} are the electron and nuclear g-factors $\begin{cases} g_S = -2.002 \\ g_{I_H} = 5.586\ \left(g_{I_D} = 0.857\right) \end{cases}$

- The absorbed photons (N_a) produce electron-spin-polarized atoms with magnetization:

$$M(t) = N_a g_S \mu_B m_S\, e^{-\frac{t}{\tau_p}} \cos^2\left(\frac{\omega\, t}{2}\right)$$

with τ_p: polarization lifetime and ω: angular hyperfine frequency.

High-density spin-polarized H and D atoms

Detection of magnetization quantum beats with a pickup coil

- A time-dependent magnetic flux $\Phi_B(t)$ is created through the coil: $\Phi_B(t) = M(t)\, A\, \mu_0$,

where A: coil area and $\mu_0 = 4\pi \times 10^{-7}$ H/m (vacuum permeability).

- According to Faraday's law of induction, an electromotive force $\mathcal{E}(t)$ is induced: $\mathcal{E}(t) = -N_t \frac{d\Phi_B(t)}{dt}$,

where $N_t = 4.5$ (number of turns).

- Expected signal: $\mathcal{E}(t) = V_R(t) + V_L(t) \Rightarrow -N_t\, A\, \mu_0 \frac{dM(t)}{dt} = V_R(t) - \frac{L}{R}\frac{dV_R(t)}{dt}$

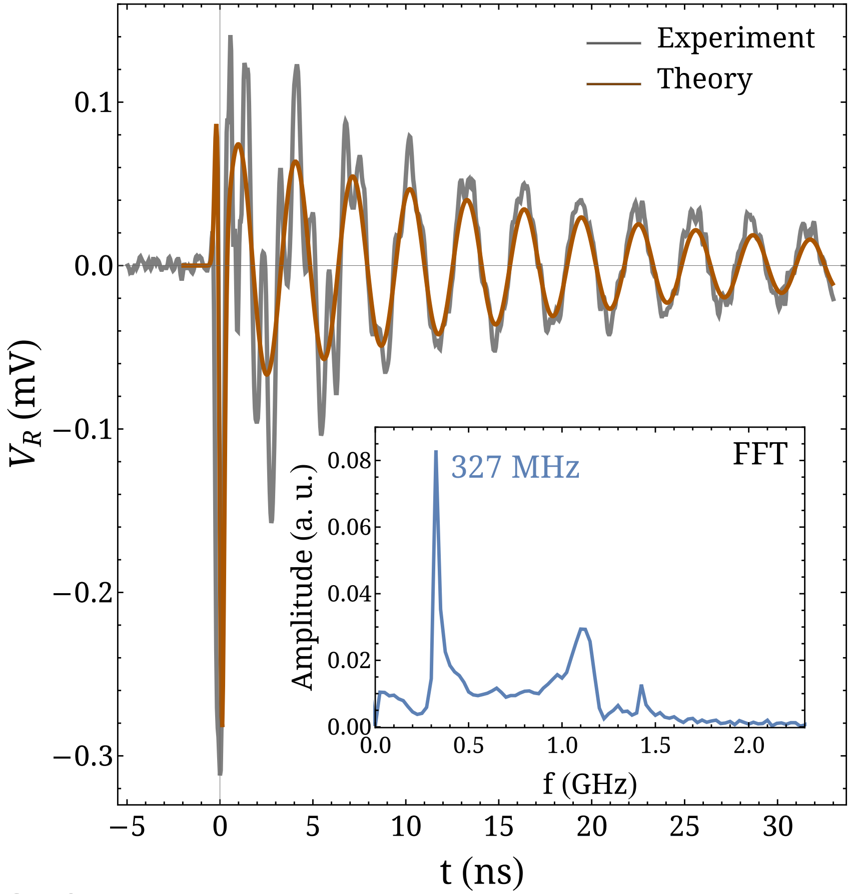
$$\Rightarrow V_R(t) = N_t\, A\, \mu_0 \frac{dM(t)}{dt} - \frac{L}{R}\frac{dV_R(t)}{dt},$$

where $R = 50\ \Omega$ (load resistor) and L: inductance.

- For UV beams (213 nm or 266 nm) with an energy of a few mJ and a pulse duration of 150 ps, interacting with $\sim 10^2$ mbar of HY, a signal of the order of 10^{-1} mV can be detected with a 5-mm-long and 2-mm-diameter coil.

High-density spin-polarized H and D atoms

Detection of magnetization quantum beats with a pickup coil



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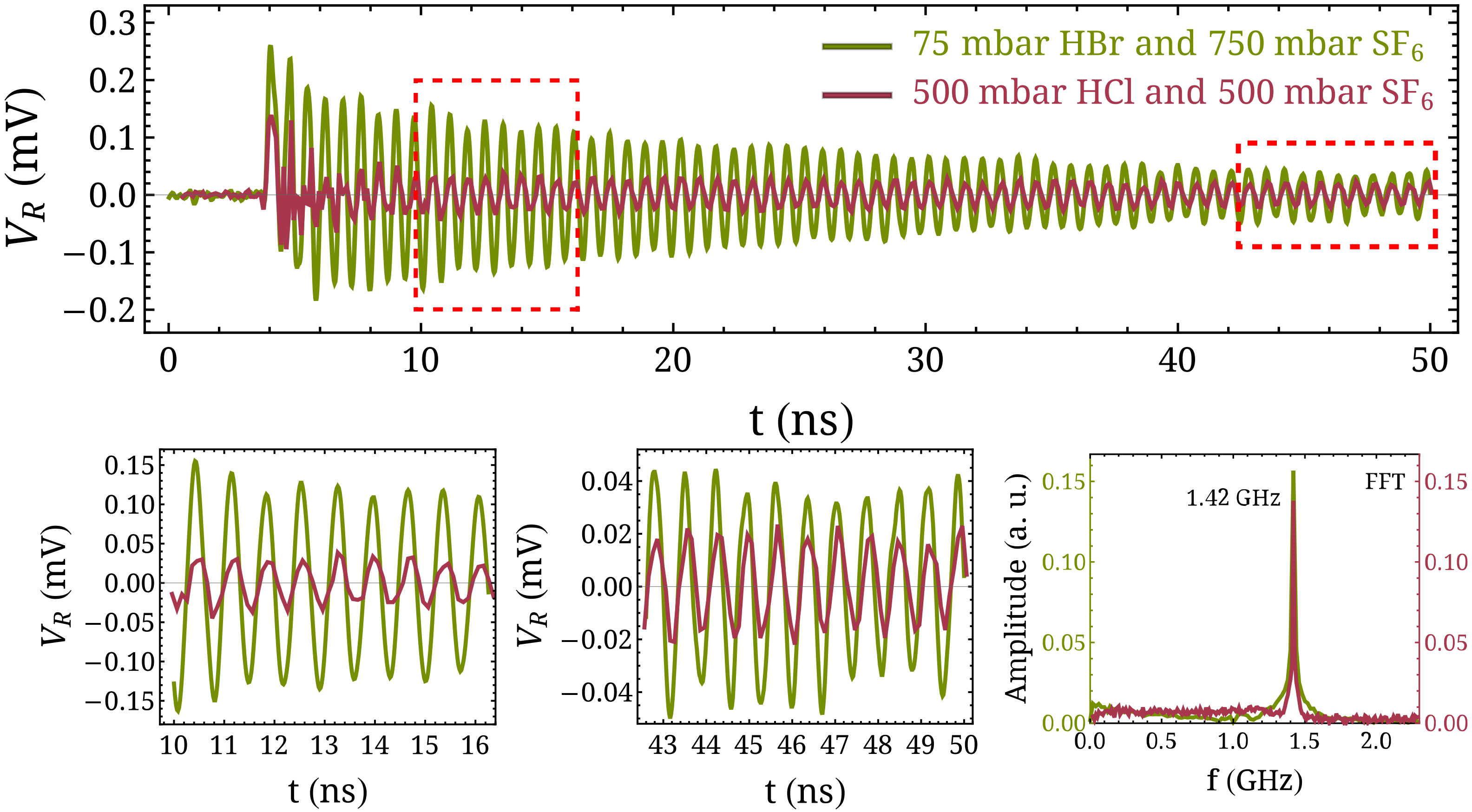

Sofikitis et al., Phys. Chem. Chem. Phys. **21**, 14000 (2019)

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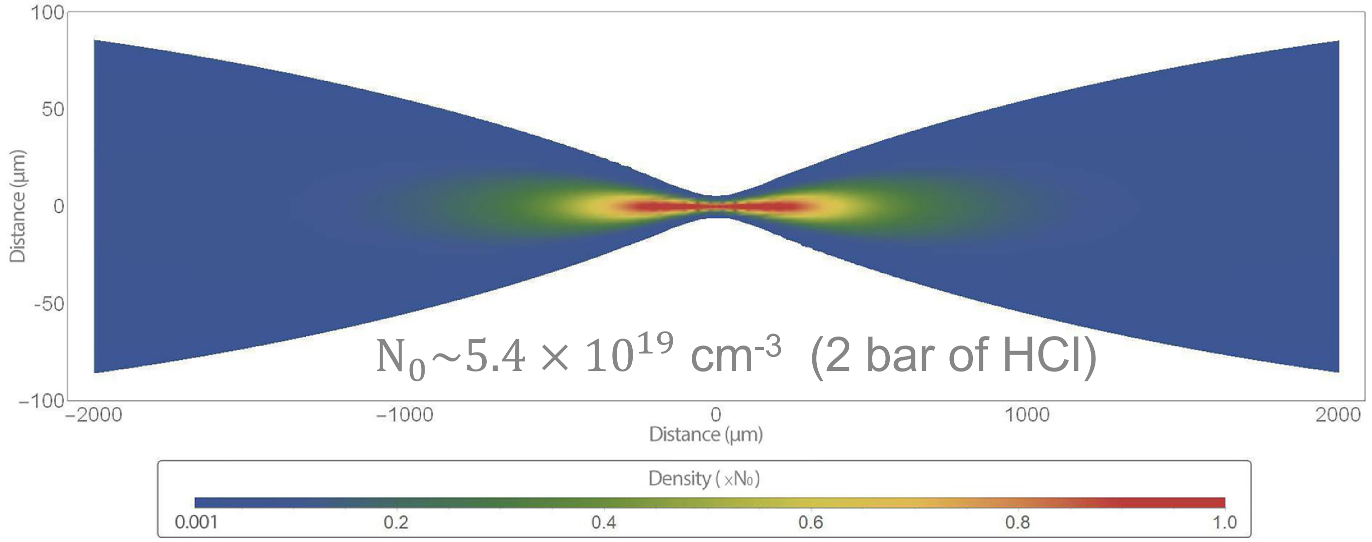
High-density spin-polarized H and D atoms

Spin-polarized H (SPH) density and focusing geometries

Photodissociation regimes

i. "low"-density regime ([Y]≪[HY]):
- 0.1% of HY molecules are dissociated
- SPH density $\sim 10^{16}$ cm^{-3}
- depolarization via an SPH-HY complex

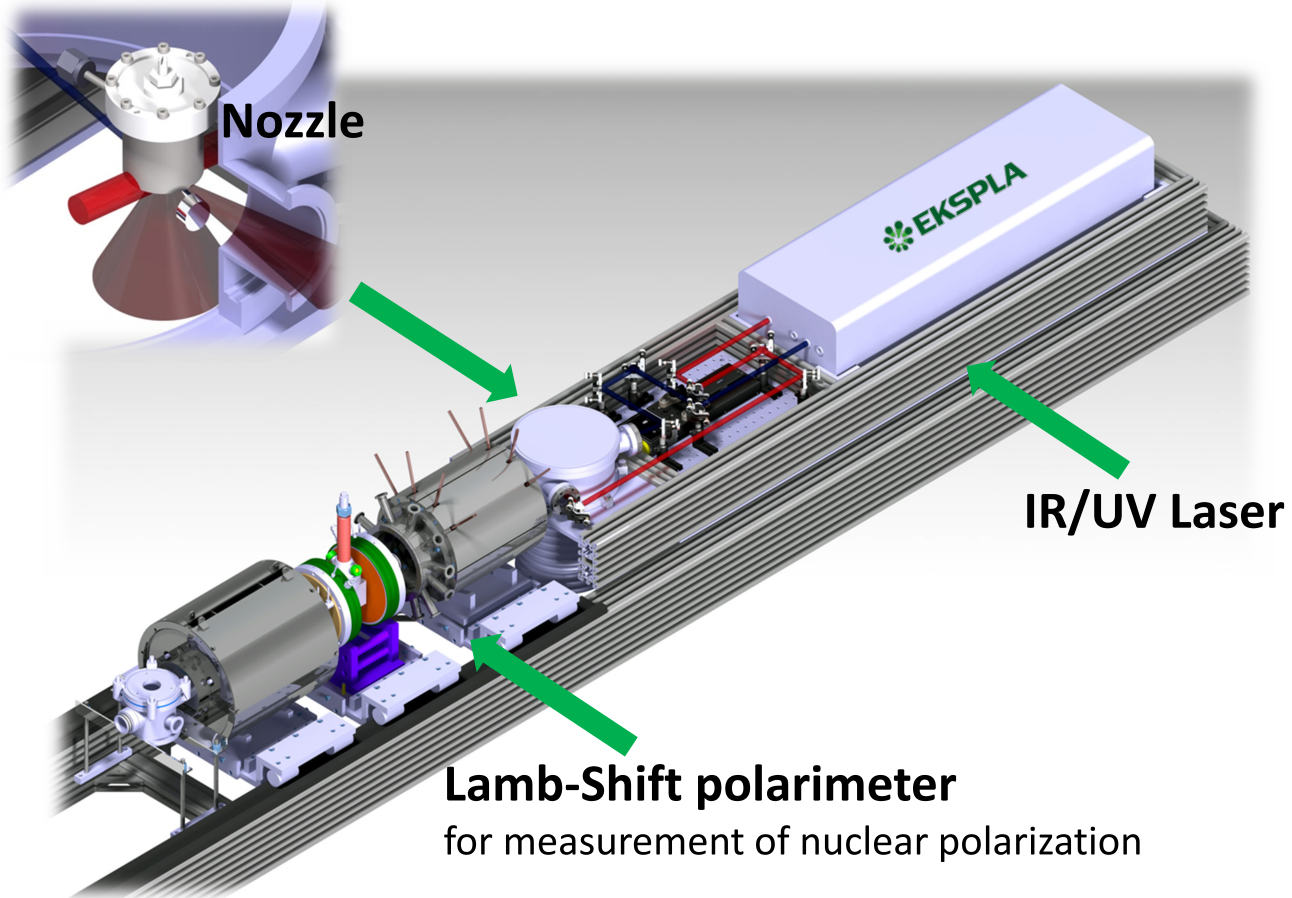
ii. "high"-density regime ([Y]≫[HY]):
- virtually all HY molecules are dissociated
- SPH density $\sim 10^{19}$ cm^{-3}
- depolarization via collisions between SPH and Y (depolarized within less than 1 ns)
- inert gas with a high heat capacity can cool down the SPH and lower the collision rate

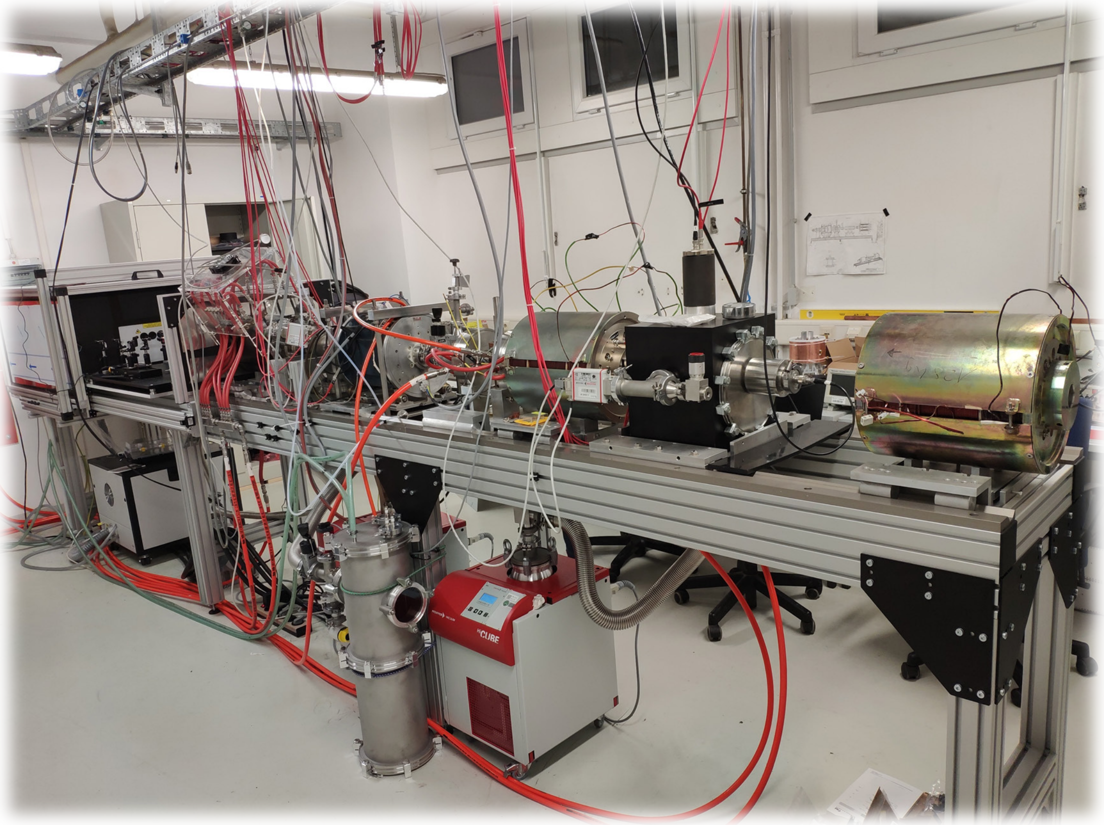


Spiliotis et al., Chem. Phys. Impact **2**, 100022 (2021)

Future developments

Production and detection of polarized proton beams from photodissociation (FZJ)





A. Hützen, PhD thesis (Heinrich-Heine-University Düsseldorf, 2021)

Future developments

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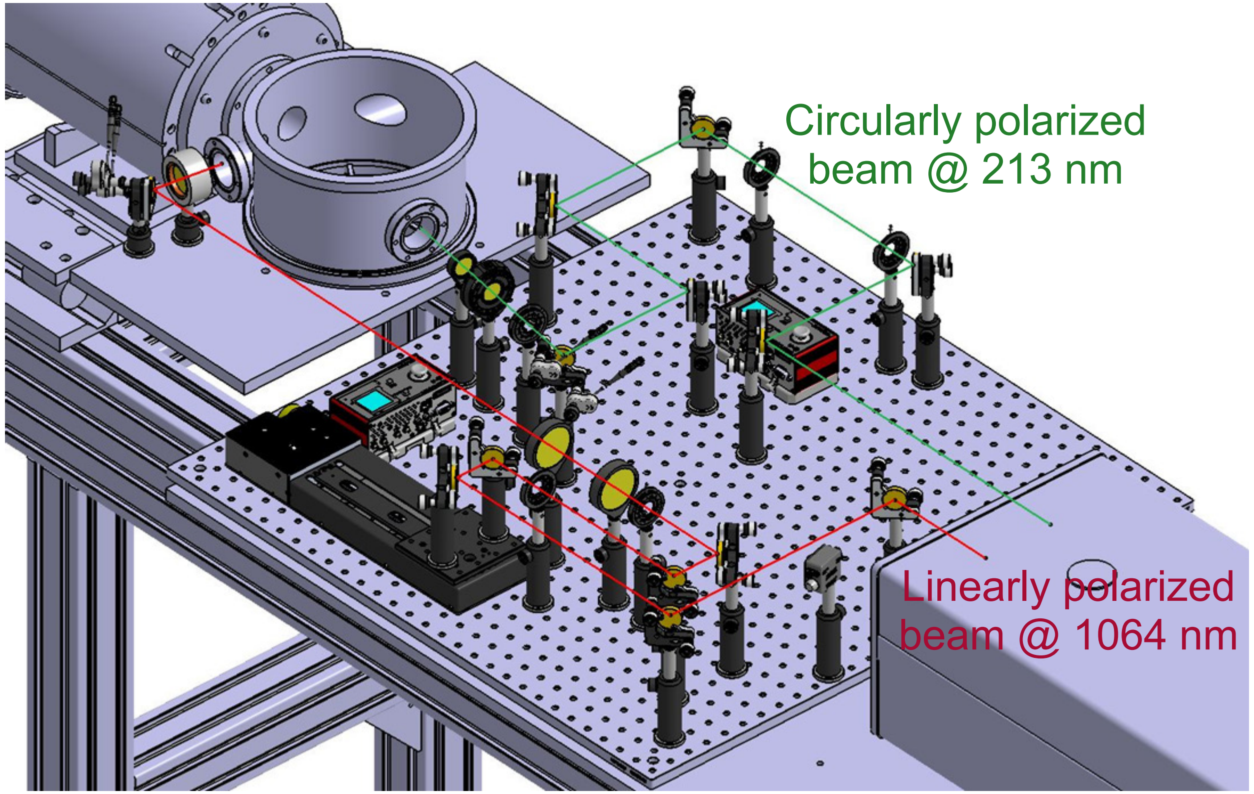

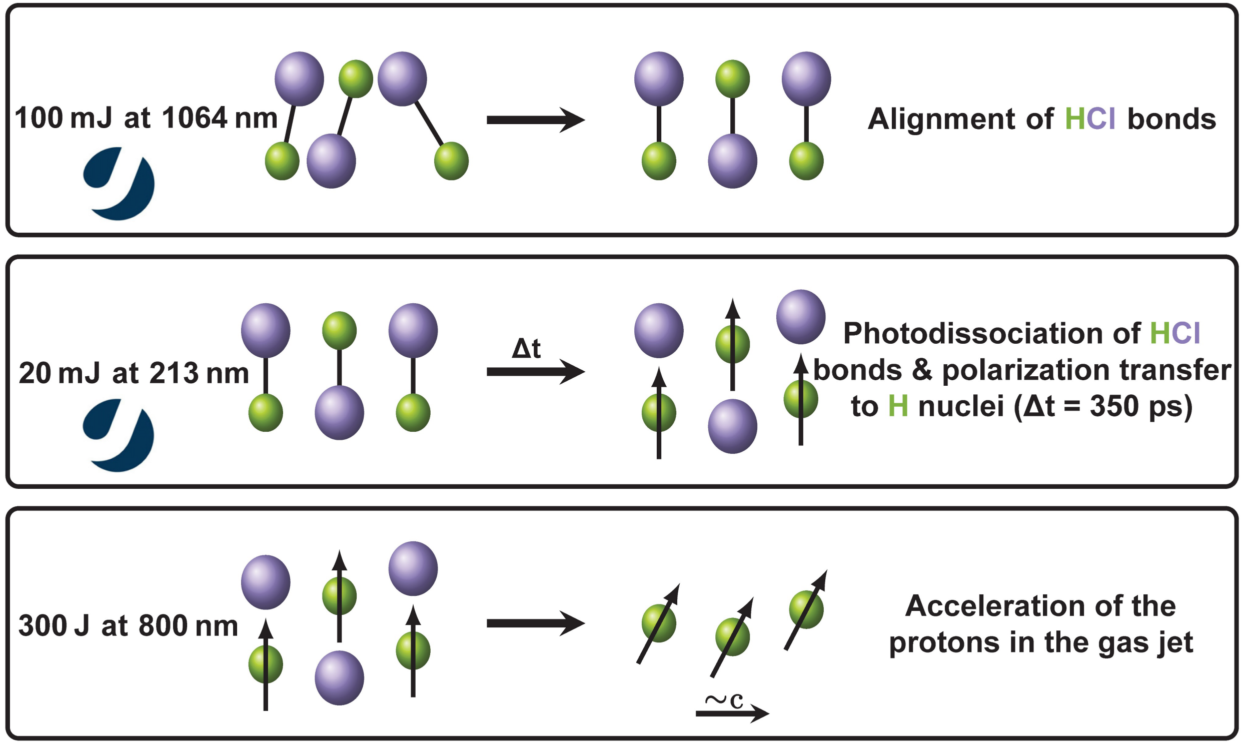

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