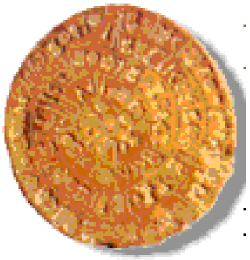
Optical excitation of molecules for Spin-Polarized Nuclear Fusion



T. Peter Rakitzis

Department of Physics, University of Crete and

IESL-FORTH

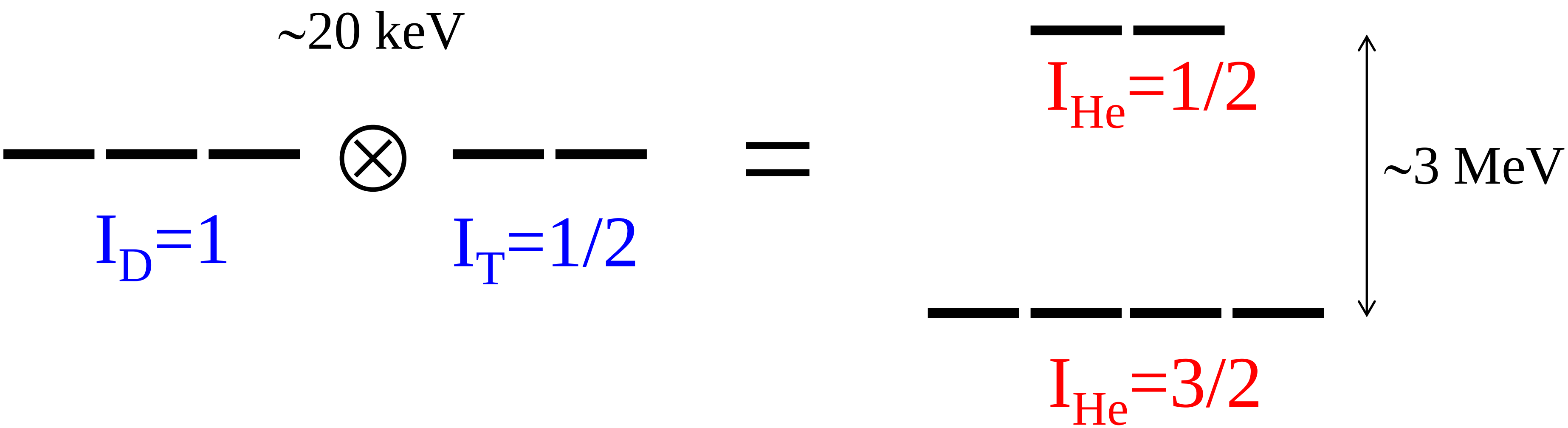
Institute of Electronic Structure and Laser

Foundation for Research and Technology - Hellas

Fusion polarization dependence for (1) and (2)

(1) $^{2}D + ^{3}T \rightarrow ^{5}He\ (I=3/2) \rightarrow ^{4}He + n$

(2) $^{2}D + ^{3}He \rightarrow ^{5}Li\ (I=3/2) \rightarrow ^{4}He + p^{+}$



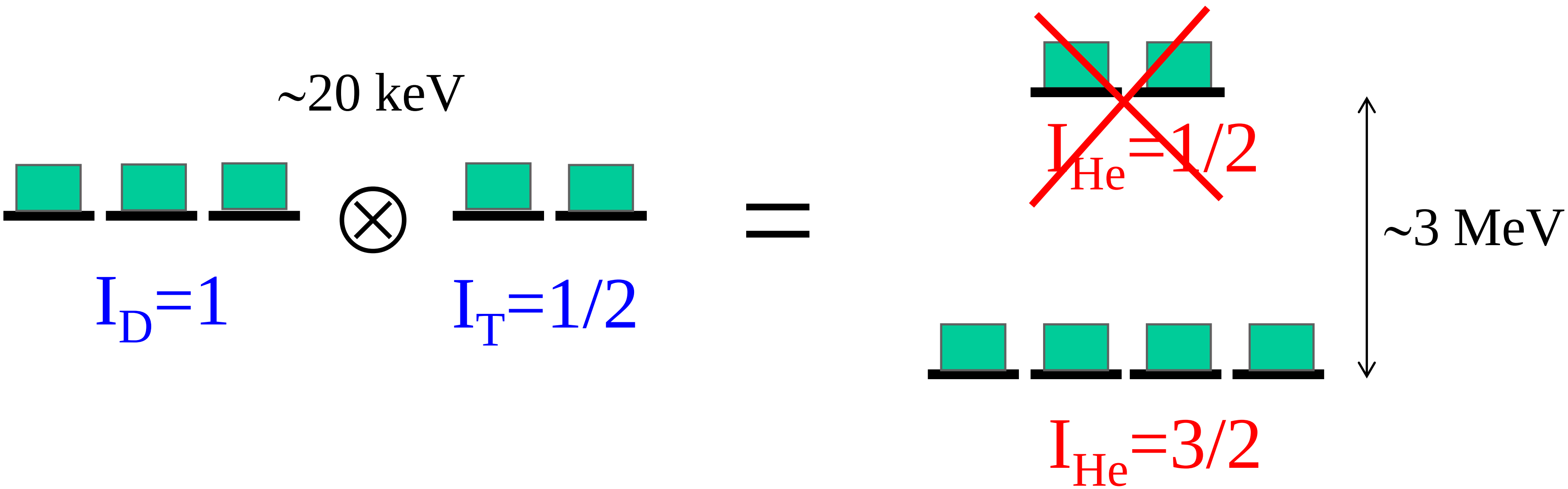
4/6 of unpolarized m-state combinations give I=3/2

2/6 of unpolarized m-state combinations give I=1/2

Fusion polarization dependence for (1) and (2)

(1) $^{2}D + ^{3}T \rightarrow ^{5}He\ (I=3/2) \rightarrow ^{4}He + n$

(2) $^{2}D + ^{3}He \rightarrow ^{5}Li\ (I=3/2) \rightarrow ^{4}He + p^{+}$

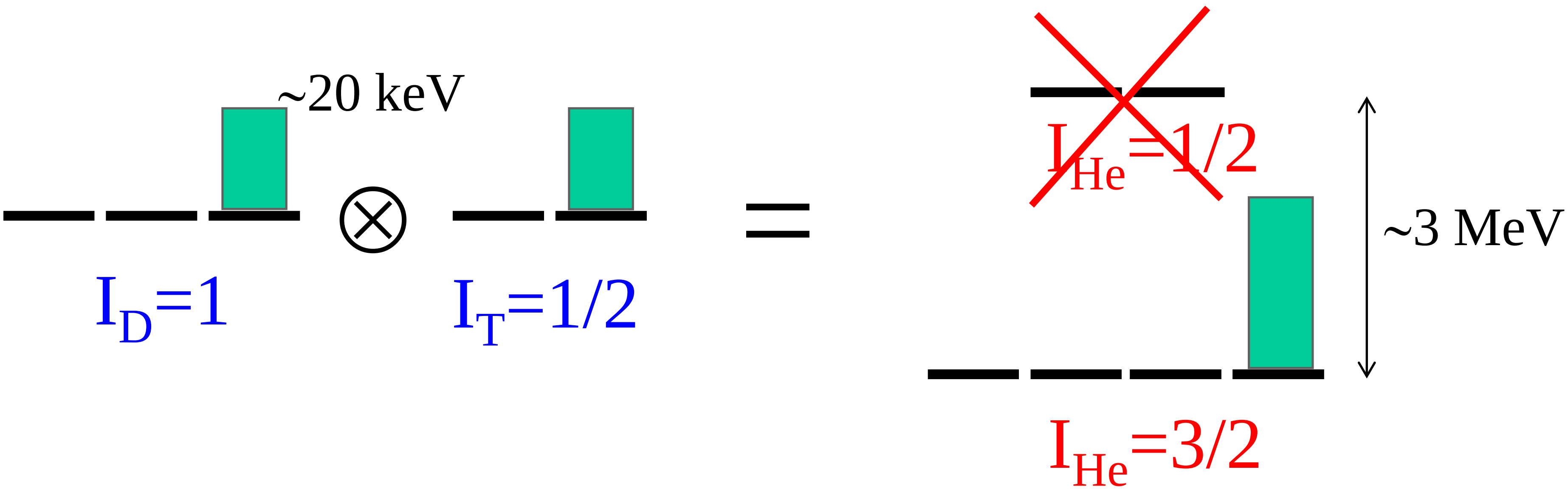


Only 4/6 of unpolarized m-state combinations can react

Fusion polarization dependence for (1) and (2)

(1) $^{2}D + {}^{3}T \rightarrow {}^{5}He\ (I=3/2) \rightarrow {}^{4}He + n$

(2) $^{2}D + {}^{3}He \rightarrow {}^{5}Li\ (I=3/2) \rightarrow {}^{4}He + p^{+}$



100% of **polarized** reactants give I=3/2 $\Rightarrow$

(1) Polarization increases rate by 50%

(2) Directional neutron distribution ~$\sin^{2}\theta$

(3) Together, reactor efficiency may improve by up to a factor of 2.

Polarized Fusion in Plasma has never been observed (!)
due to a lack of sufficient spin-polarized D

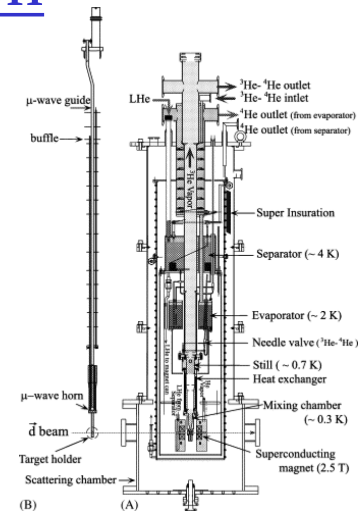
Open questions in Polarized Fusion

(1) Does the polarization survive the ~20 keV plasma?

(2) What is the polarization dependence of DD fusion?

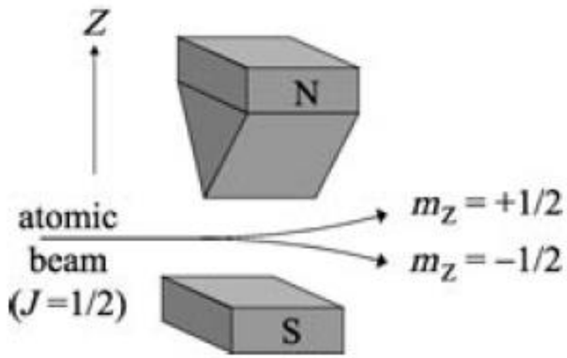
(3) Can 10^{22} pol-D/s be produced, which is needed for fusion reactors (ITER)?

Standard Methods of SPH production

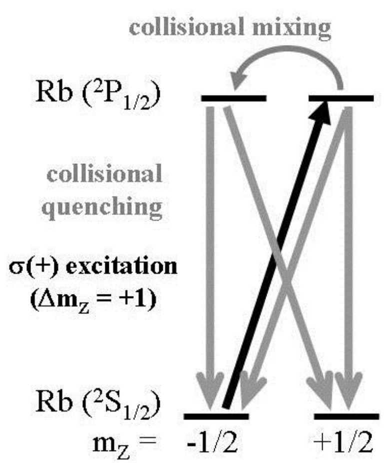


- Strong B-field/cryogenic cooling, DNP

- Stern-Gerlach separation



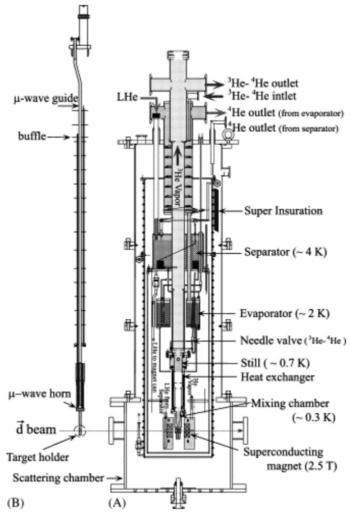
- Optical-pumping



Strong B-field/cryogenic cooling

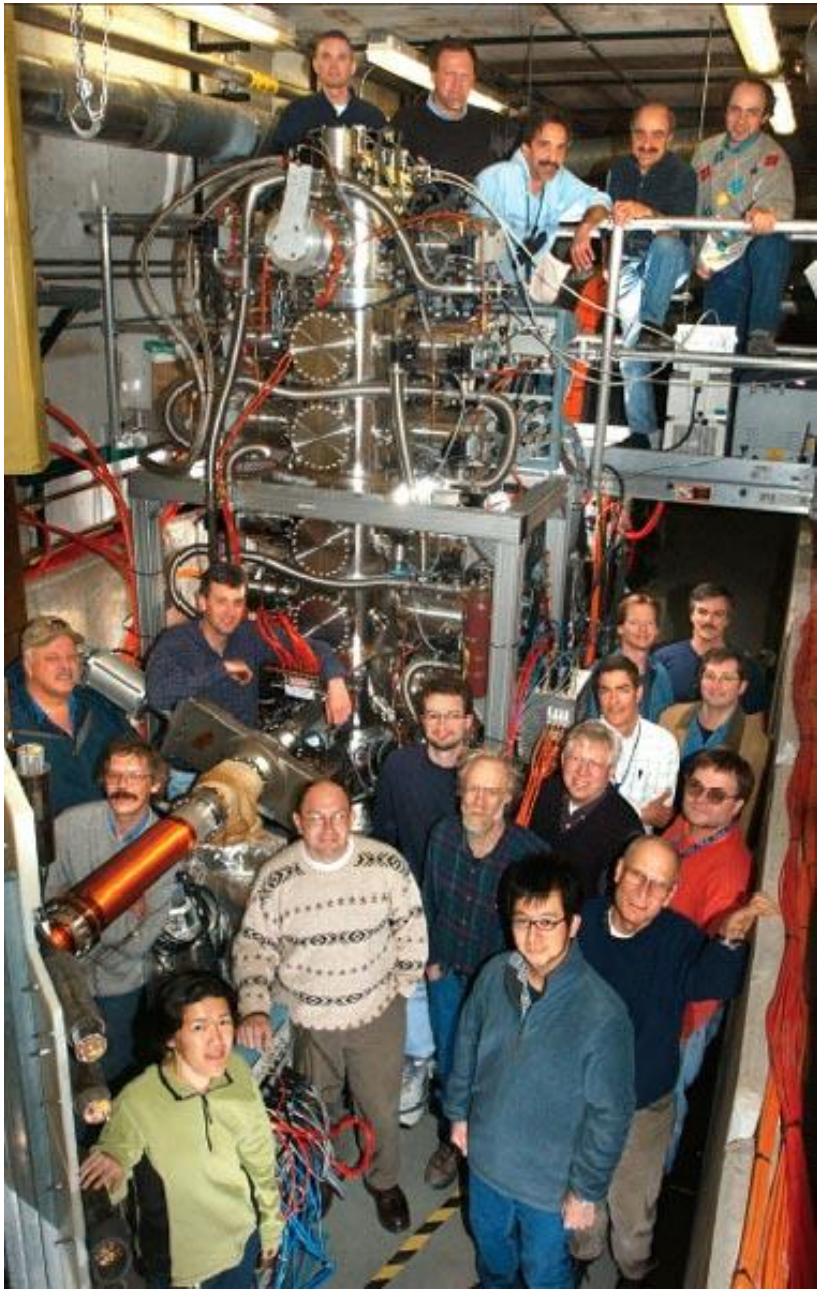
D_2 cooled to 25 mK (2 weeks) with 15T B field:

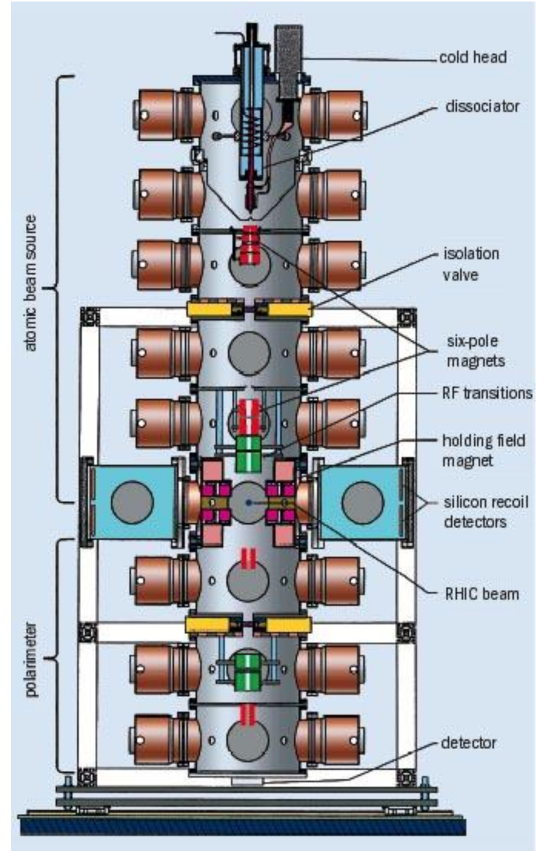
- Polarization only ~20%
- Takes too long
- Makes too little



Unclear how the D_2 "ice" can be introduced into a reactor, without depolarization

SPH production at BNL (Stern-Gerlach technique)



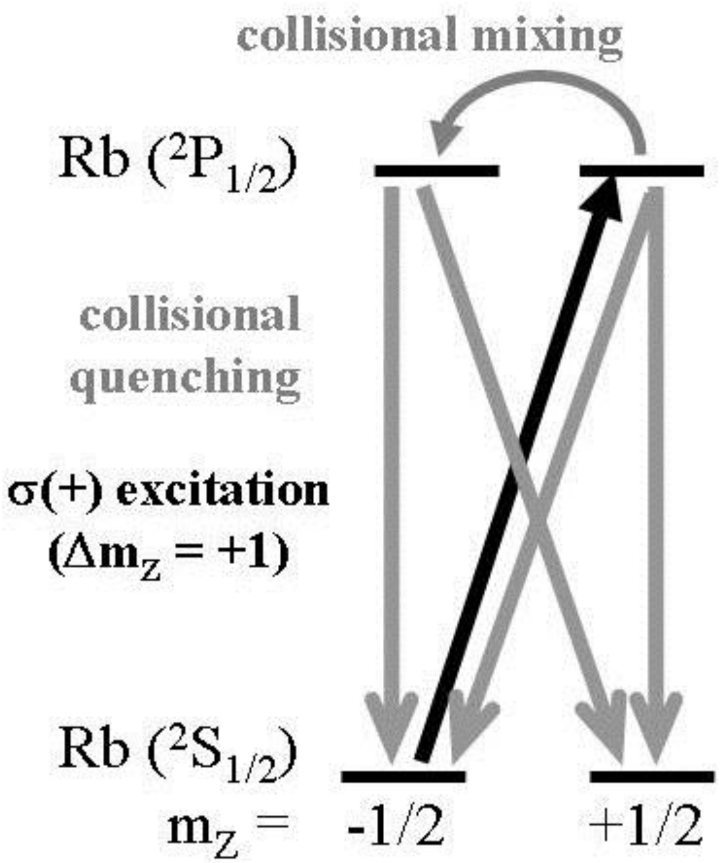


10^{17}/s << 10^{22}/s

needed for
a fusion reactor

Current Methods of SPH production

- Optical-pumping Limited to alkali atoms (and noble gases and SPH via Spin-exchange optical pumping)



10^{17}/s << 10^{22}/s

needed for
a fusion reactor

Each technique has significant drawbacks.

Laser excitation of molecules
offers advantages
compared to traditional methods

Two new methods:

(1) UV photodissociation of Hydrogen Halides

(2) IR rovribration excitation of molecules

(1) Molecular Photodissociation

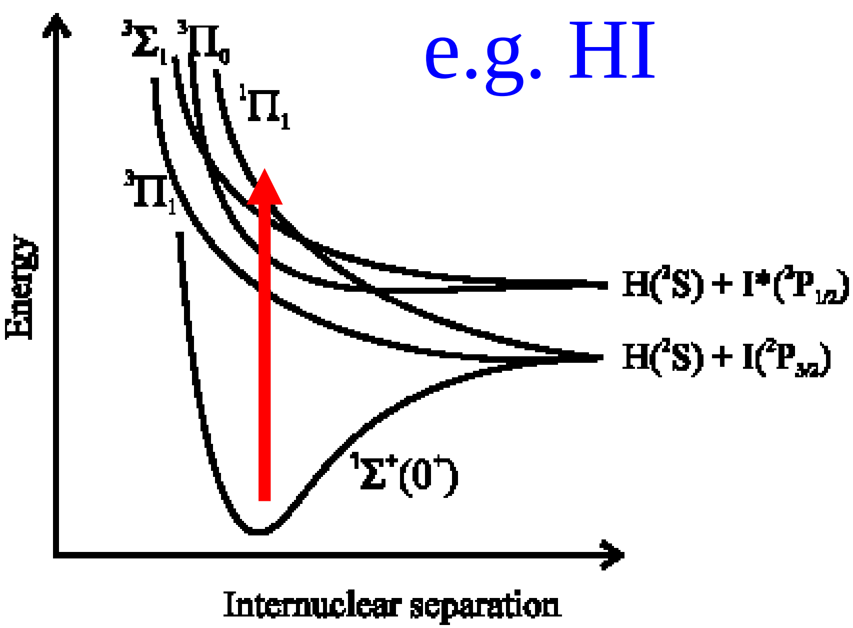
Adiabatic correlation of molecular electronic states to specific atomic m states

van Brunt and Zare, *J. Chem. Phys.* **48**, 4304 (1968).

$$\mathrm{AB}(\Omega_g=0) + h\nu \rightarrow \mathrm{AB}(\Omega_i=1) \rightarrow \mathrm{A}(J_A,m_A) + \mathrm{B}(J_B,m_B)$$

Circularly Polarized

e.g. HI



$$\Omega_i \rightarrow \left| m_A, m_B \right\rangle$$

$$\left|+1\right\rangle_{A^1\Pi} \xrightarrow{R\to\infty} \left| -\frac{1}{2}, +\frac{3}{2} \right\rangle$$

$$\left|+1\right\rangle_{a^3\Pi} \xrightarrow{R\to\infty} \left| +\frac{1}{2}, +\frac{1}{2} \right\rangle$$

$$\left|0\right\rangle_{a^3\Pi} \xrightarrow{R\to\infty} \left| \mp\frac{1}{2}, \pm\frac{1}{2} \right\rangle$$

(1) Molecular Photodissociation

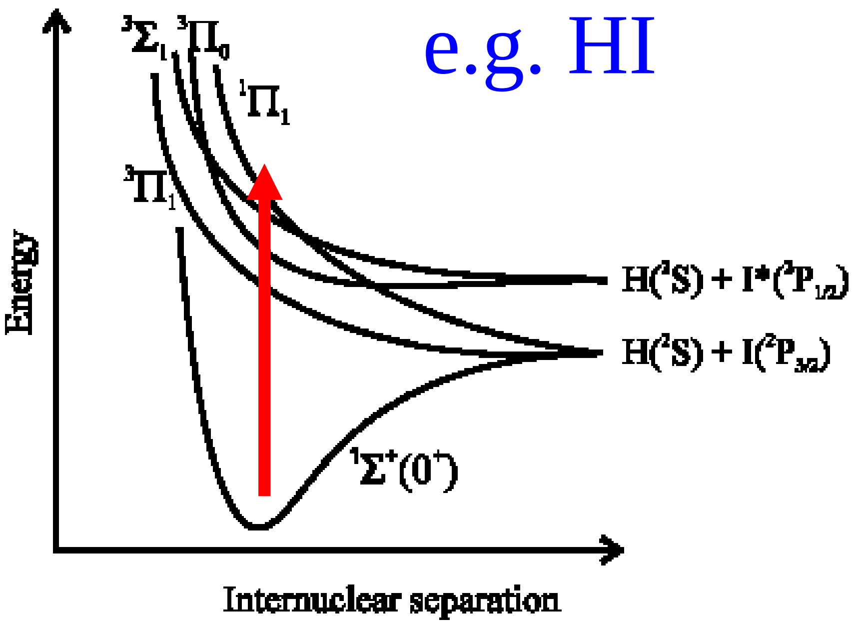
Adiabatic correlation of molecular electronic states to specific atomic m states

van Brunt and Zare, *J. Chem. Phys.* **48**, 4304 (1968).

$$AB(\Omega_g{=}0) + hf \rightarrow AB(\Omega_i{=}1) \rightarrow A(J_A,m_A) + B(J_B,m_B)$$

Circularly Polarized

e.g. HI



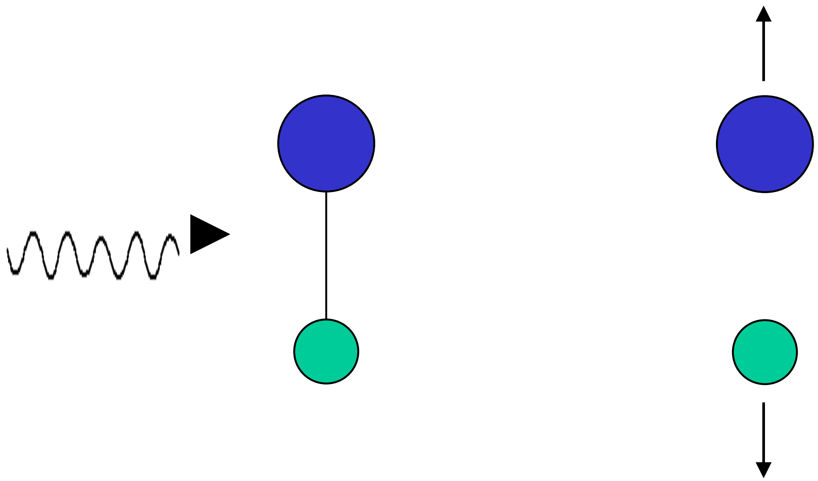
$$\Omega_i \rightarrow \left| m_A, m_B \right\rangle$$

$$\left|+1\right\rangle_{A^1\Pi} \xrightarrow{R\to\infty} \left|-\frac{1}{2},+\frac{3}{2}\right\rangle$$

$$\left|+1\right\rangle_{a^3\Pi} \xrightarrow{R\to\infty} \left|+\frac{1}{2},+\frac{1}{2}\right\rangle$$

$$\left|0\right\rangle_{a^3\Pi} \xrightarrow{R\to\infty} \left|\mp\frac{1}{2},\pm\frac{1}{2}\right\rangle$$

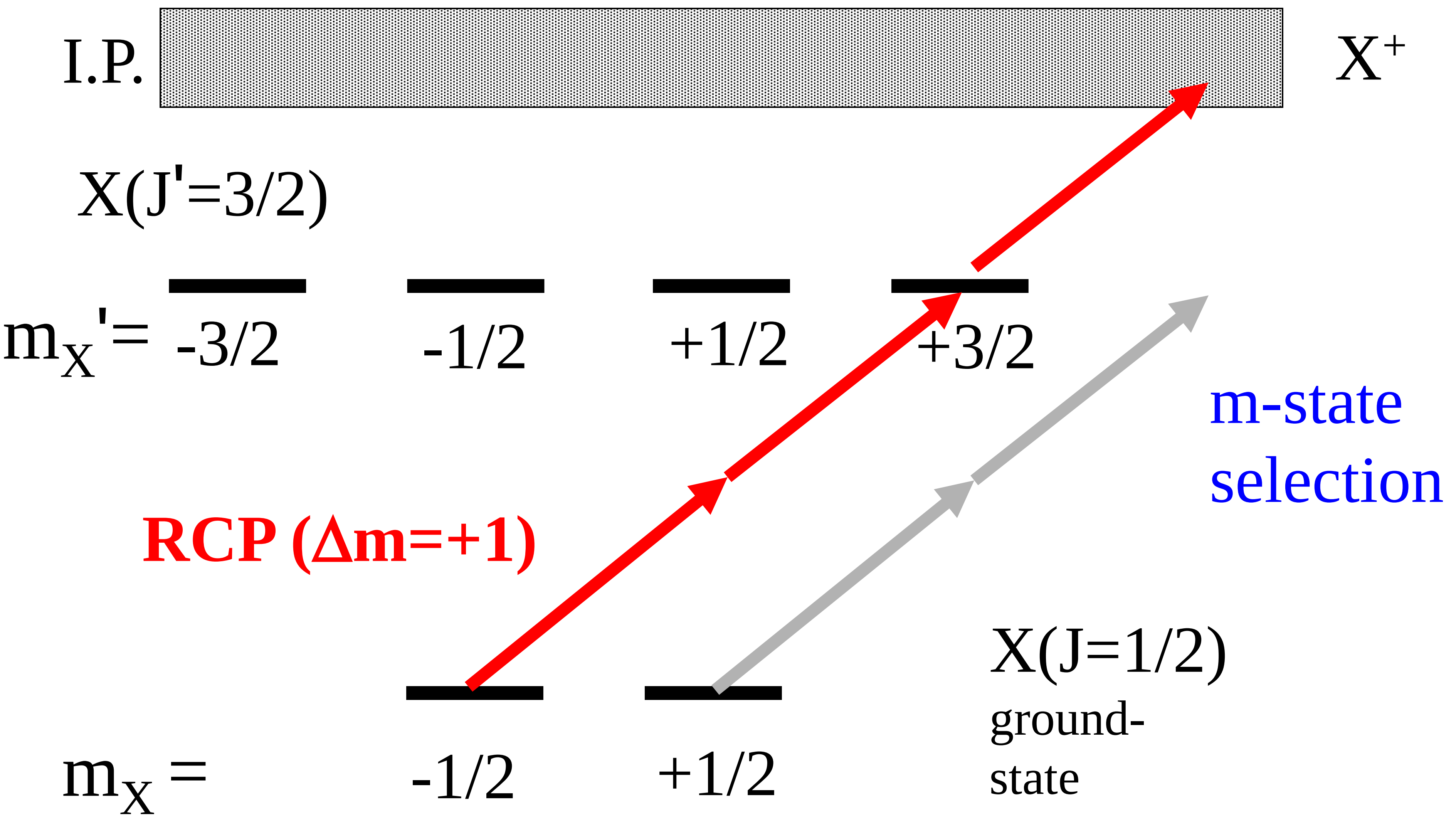
Production of highly-polarized atoms from molecular photodissociation



Advantages

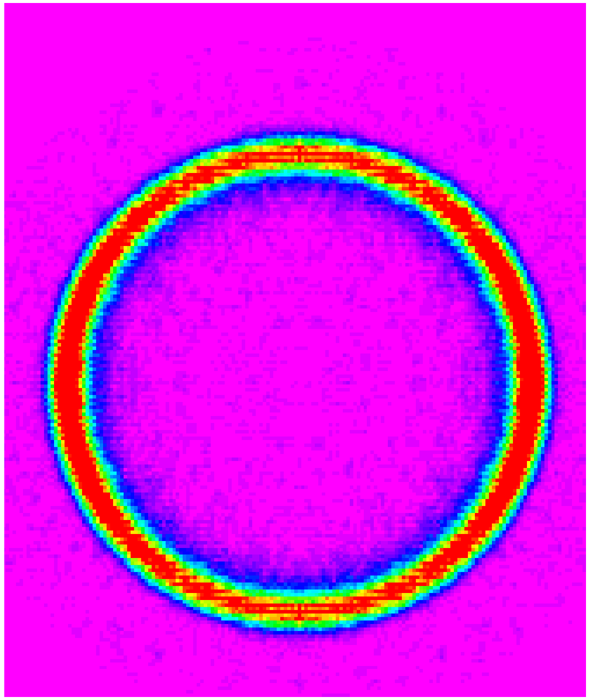
- short timescales of production (100 fs dissociation)
- Transitions in UV, where powerful lasers exist ($\sim 10^{22}$ phot/s)
- Efficient: 1 photon $\rightarrow$ 1 polarized H/D
- high-density (from stable molecules, e.g. DI, HBr, at high pressure)

Halogen atoms Polarization measured with Resonance Enhanced Multi-photon Ionization (REMPI)

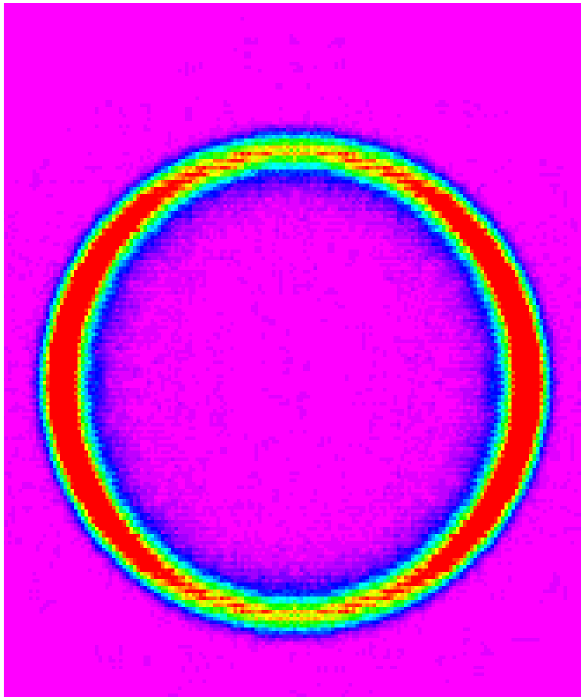


$HCl \rightarrow H + Cl\ (J=1/2)$

RR



RL

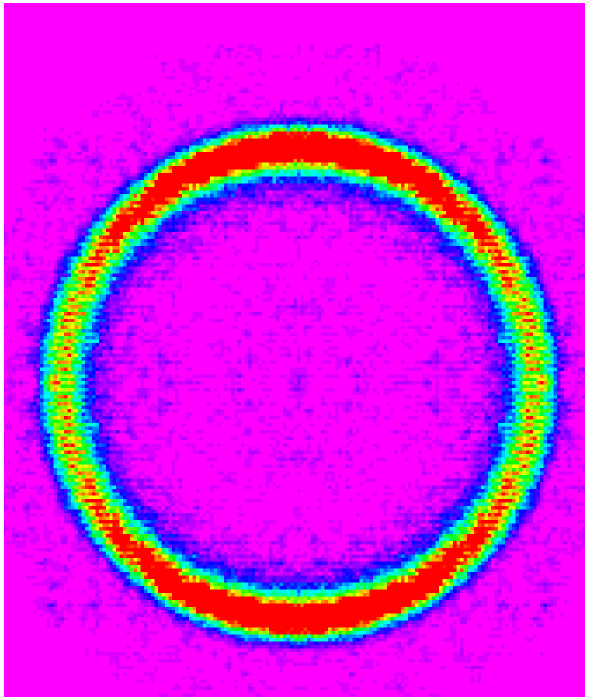


$$\frac{I_{RCP}}{I_{LCP}} = 1.8 \pm 0.2$$

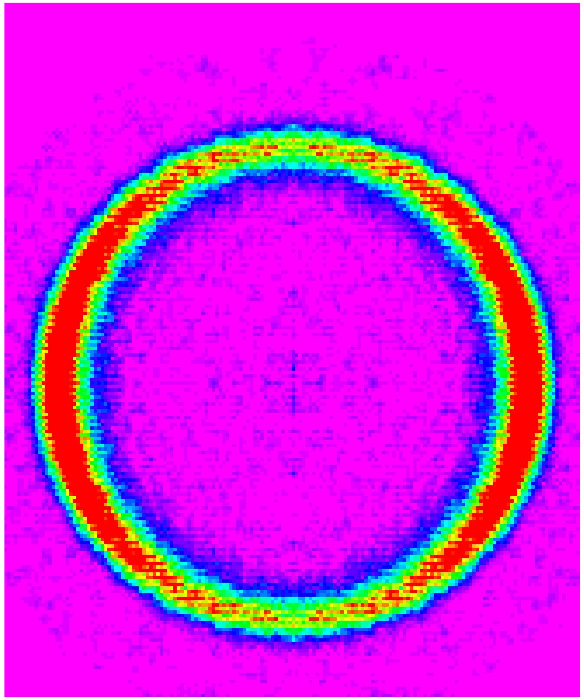
$$\Delta\beta_2 = -0.3 \pm 0.1$$

HCl $\rightarrow$ H + **Cl** (J=3/2)

RR



RL

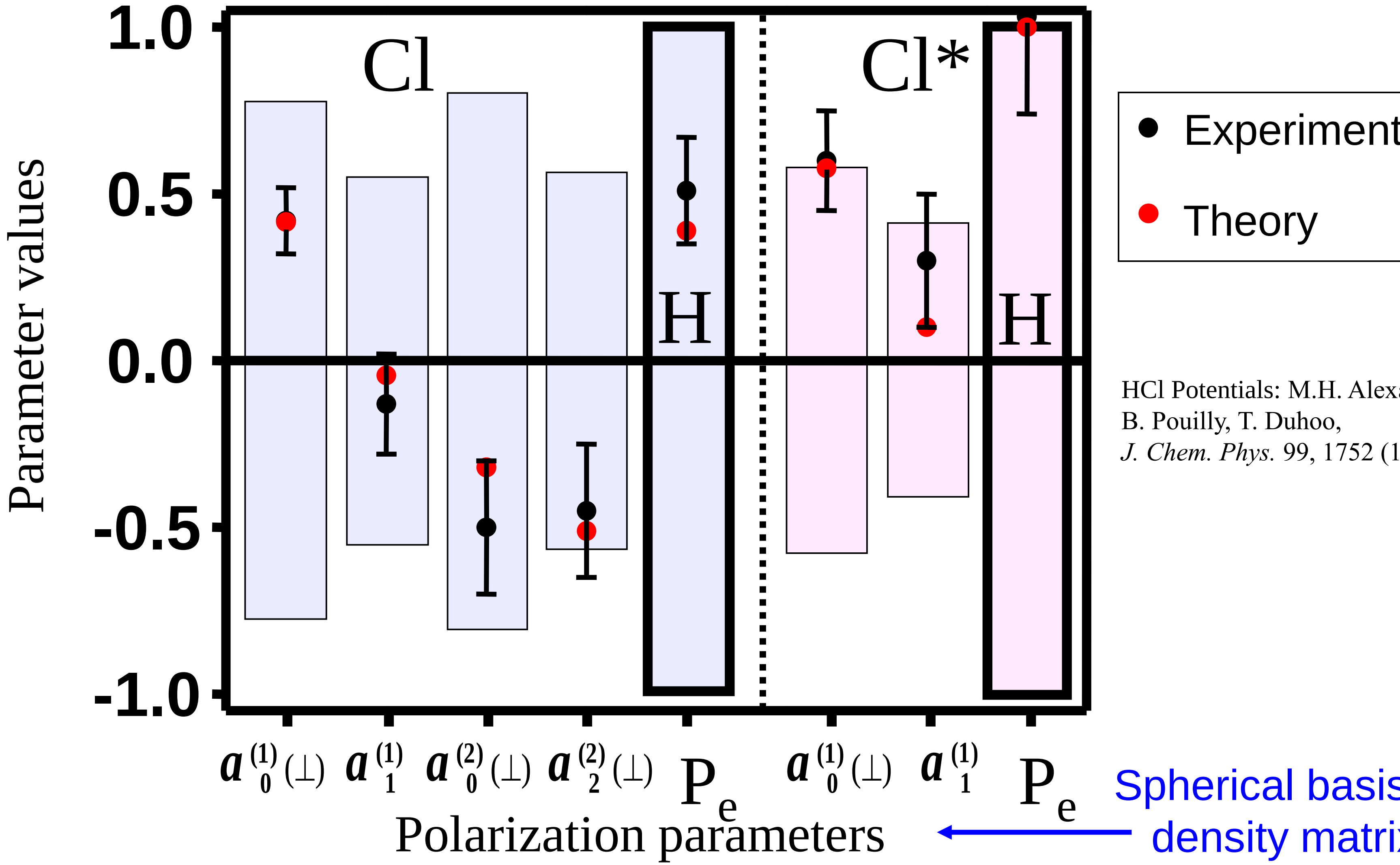


$$\frac{I_{RCP}}{I_{LCP}} = 1.4 \pm 0.2$$

$$\Delta\beta_2 = -1.0 \pm 0.1$$

Cl polarization from HCl photodissociation at 193 nm

(Diatomics understood)



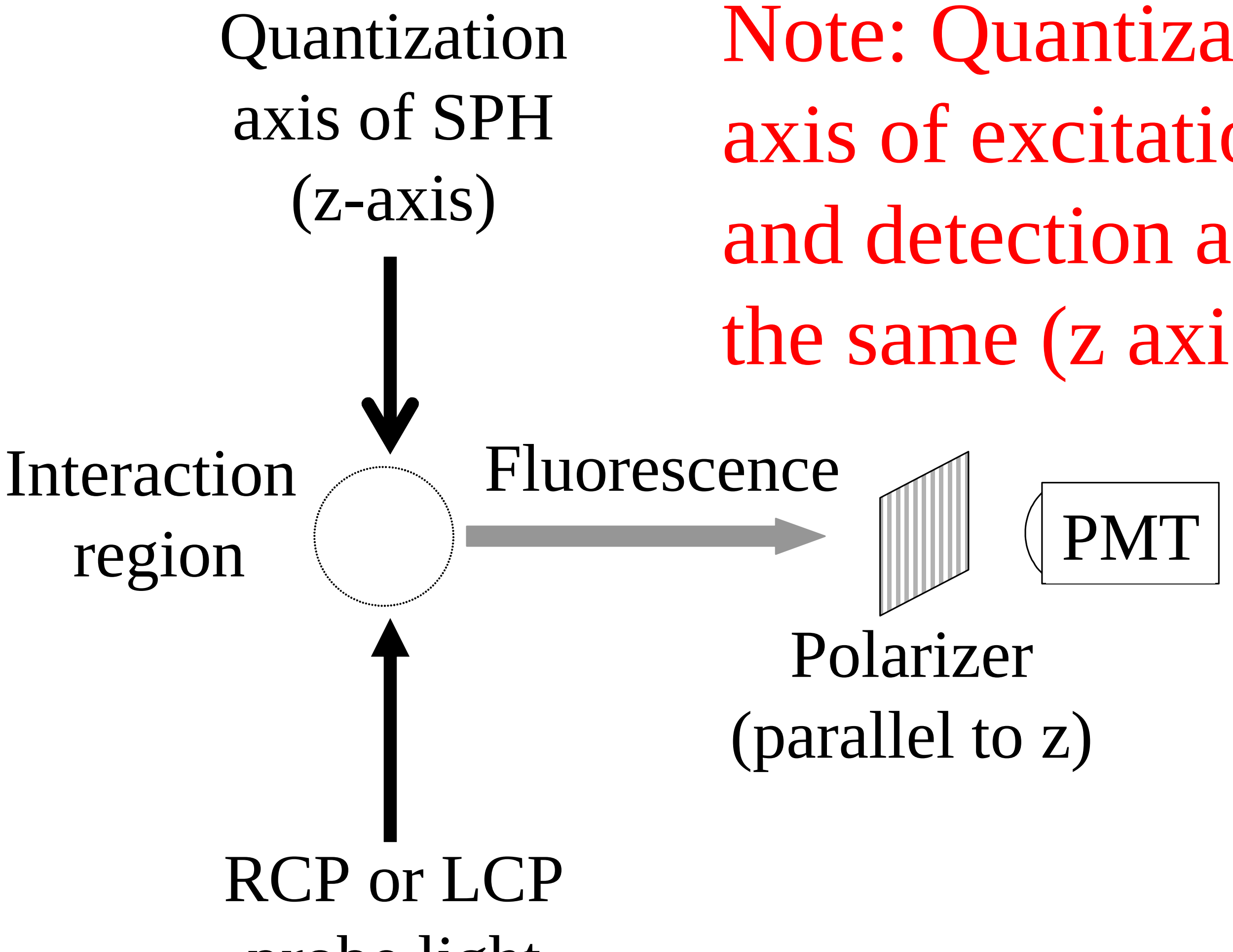
HCl Potentials: M.H. Alexander, B. Pouilly, T. Duhoo, *J. Chem. Phys.* 99, 1752 (1993).

Spherical basis of density matrix

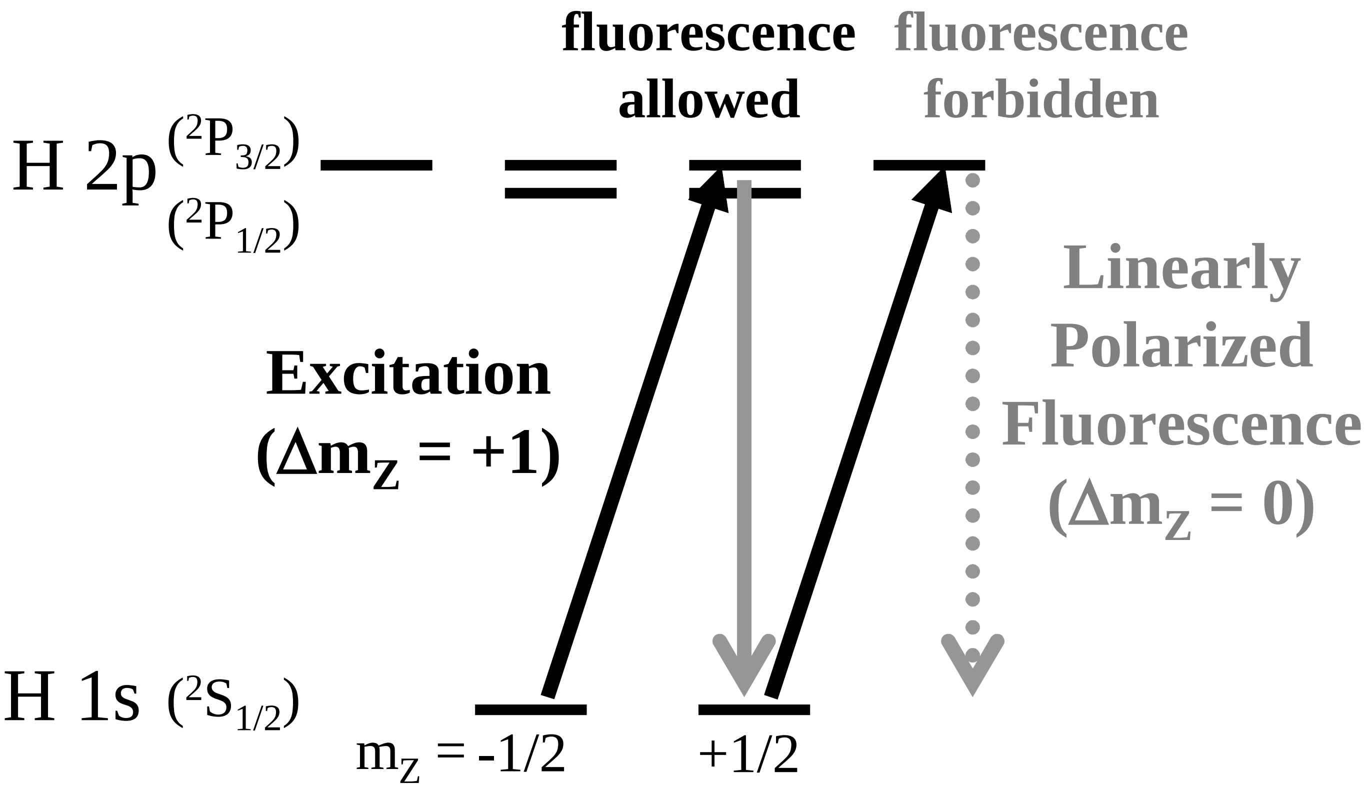
- T.P. Rakitzis et al., *Science* **300**, 1936 (2003), "SPH Atoms from Molecular Photodissociation"

Measure the SPH be seen **directly!**

Note: Quantization axis of excitation and detection are the same (z axis)

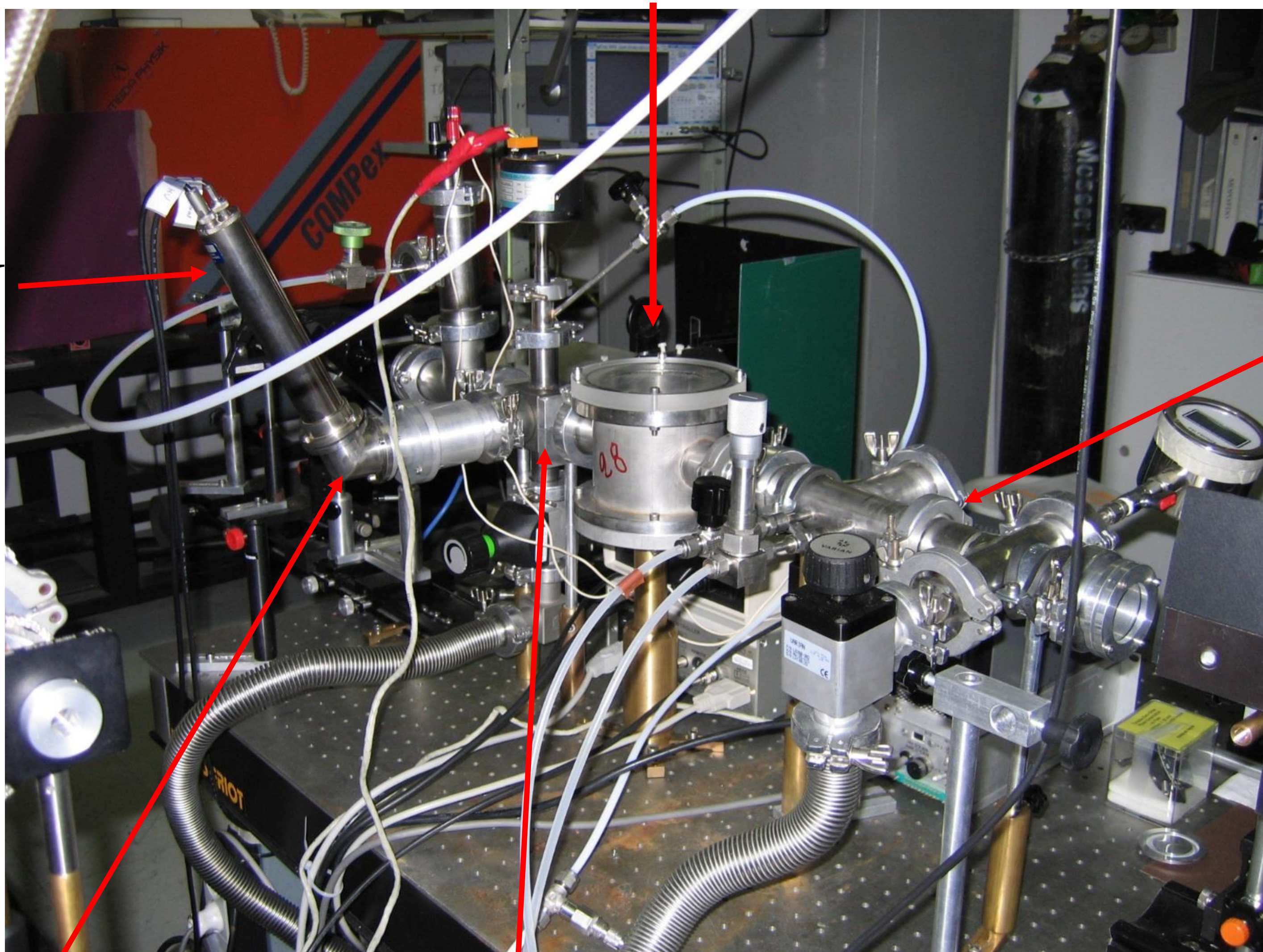


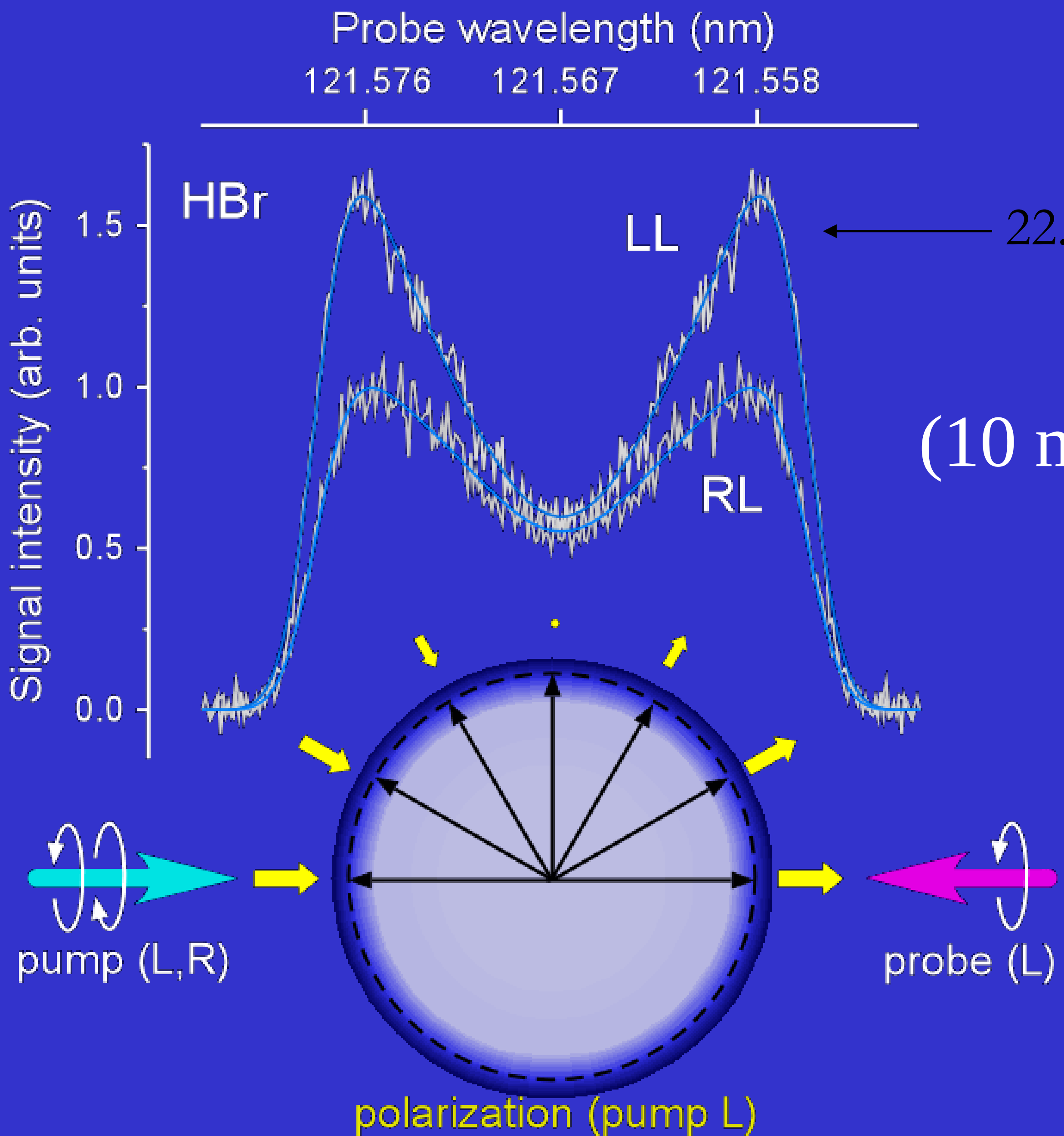
SPH fluorescence detection



T.P. Rakitzis, *ChemPhysChem*, **5,** 1489 (2004).

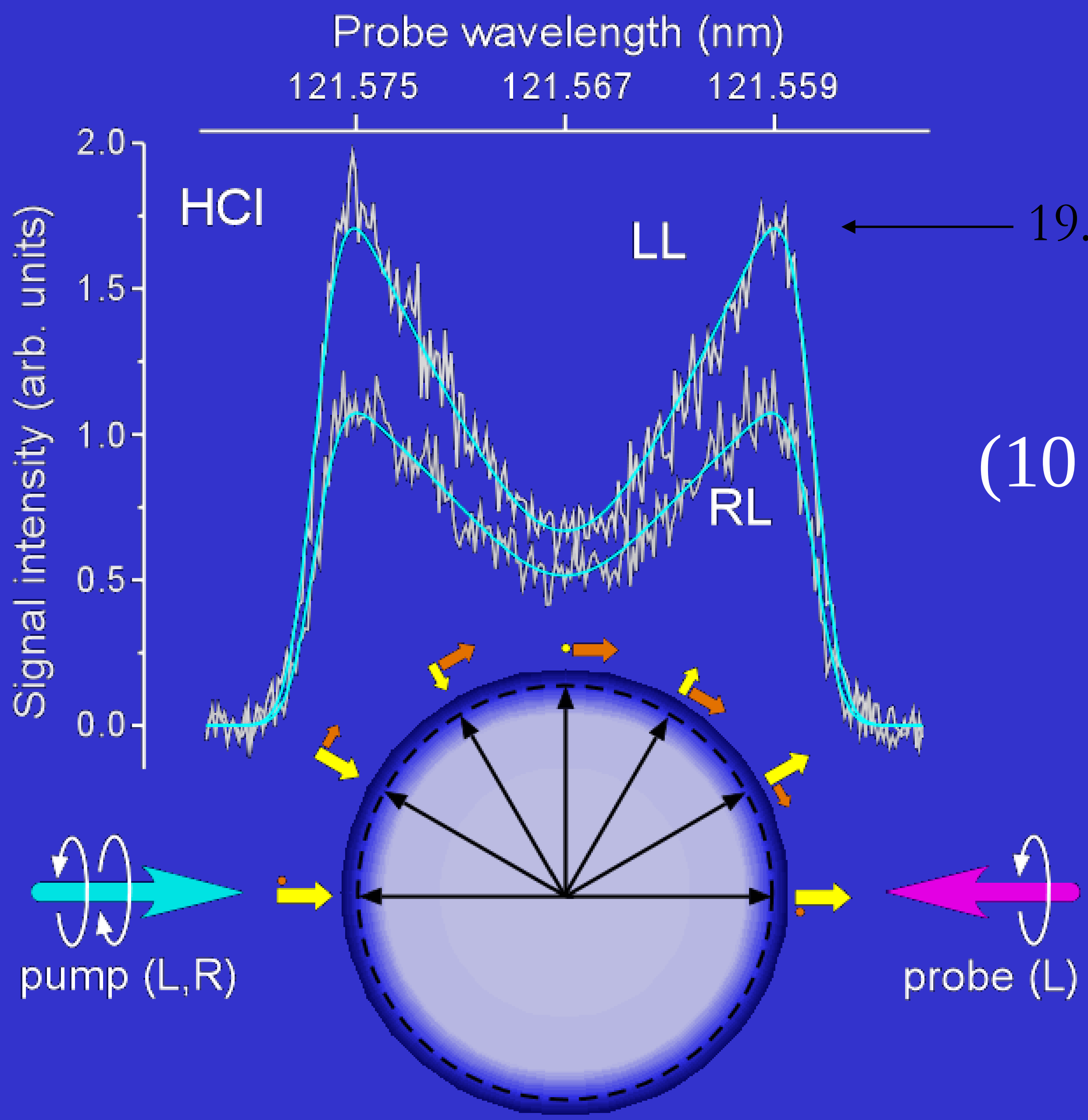
Advantages:
1) Hyperfine resolution not necessary
2) Sensitive to SPH velocity (Doppler shift)
3) Detect on nanosecond timescale
4) Very sensitive


VUV λ/4 plate
VUV PMT
VUV generation
VUV polarizer
Interaction region
COMPex



22.6 km / s

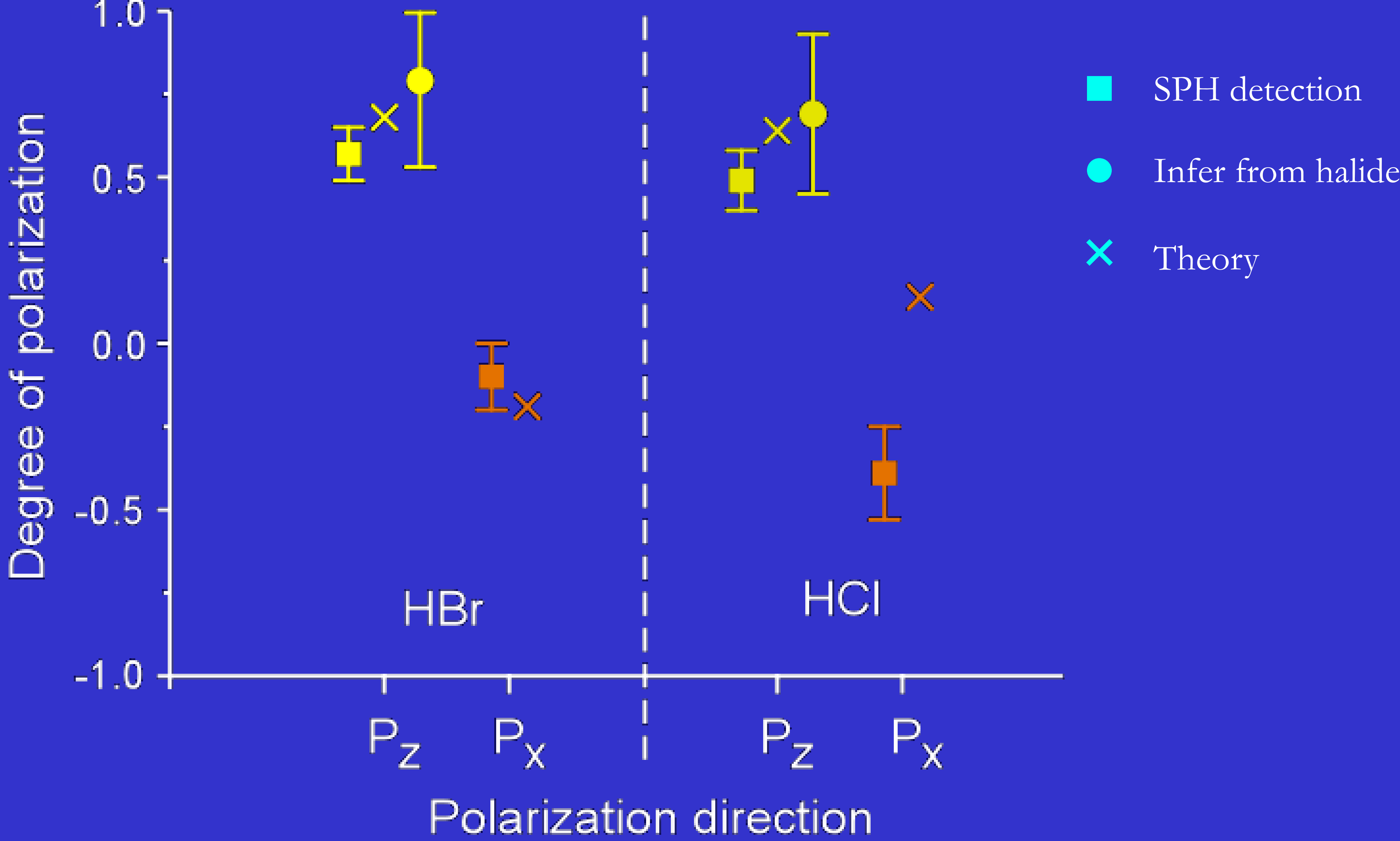
(10 mTorr HBr)



19.3 km / s

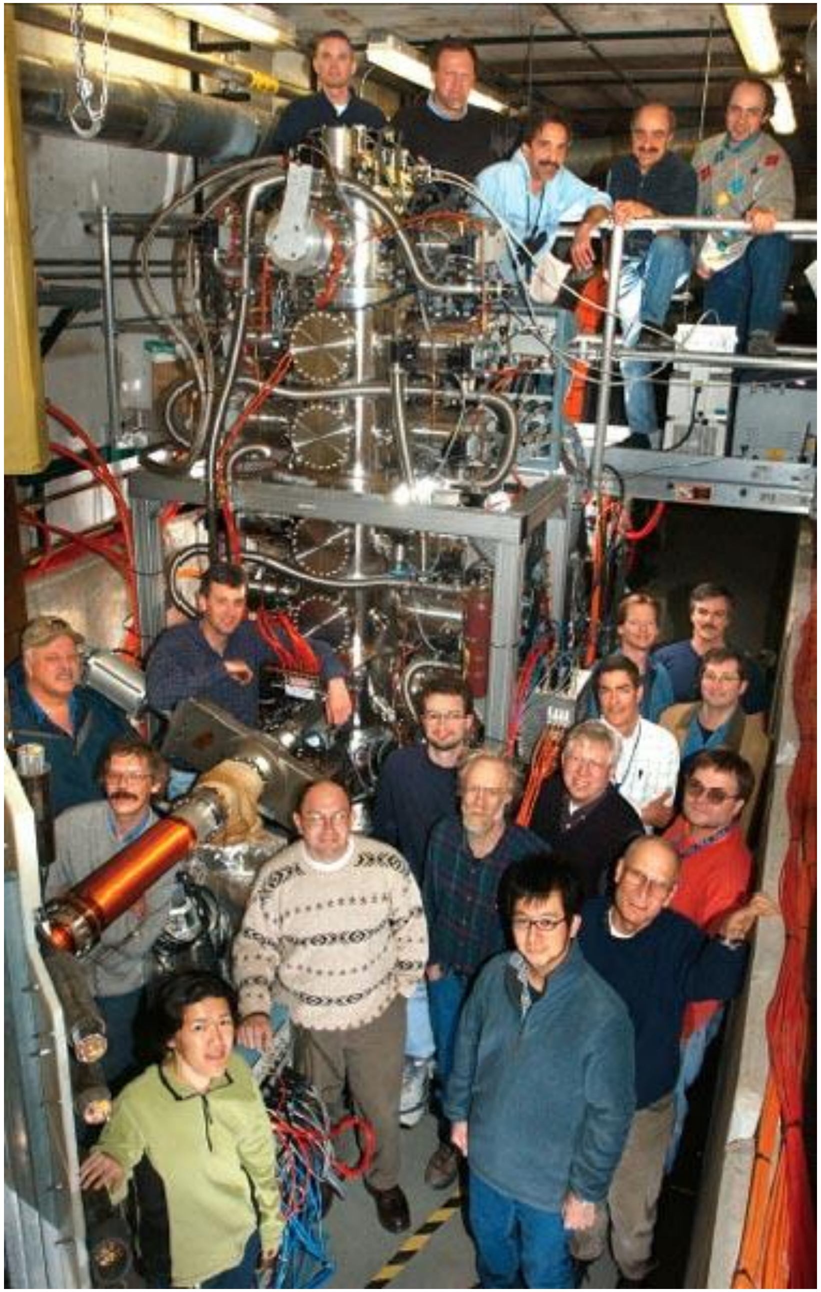
(10 mTorr HCl)

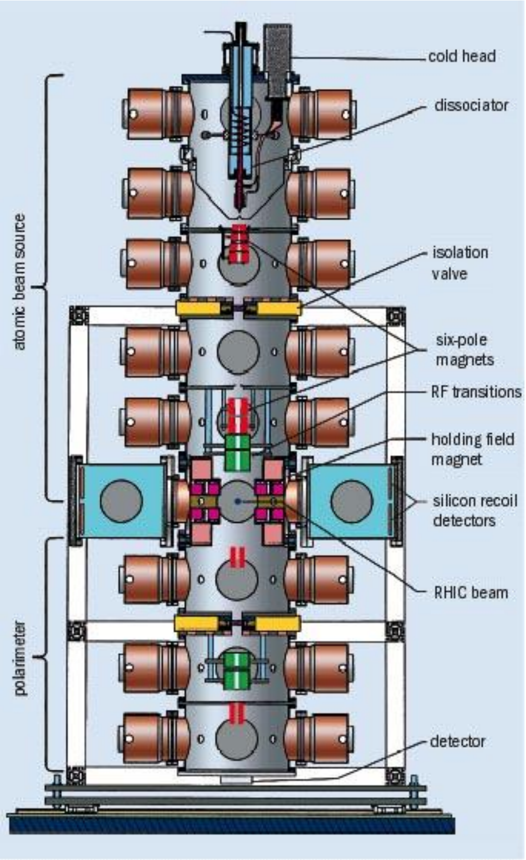
Results

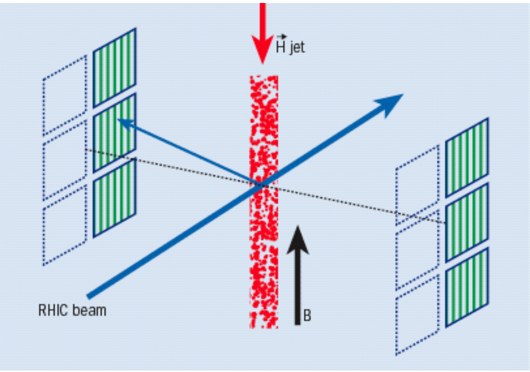


Theory: Brown & co., *JPC.A.*, 108 (2004) 7790; 110 (2006) 5371.

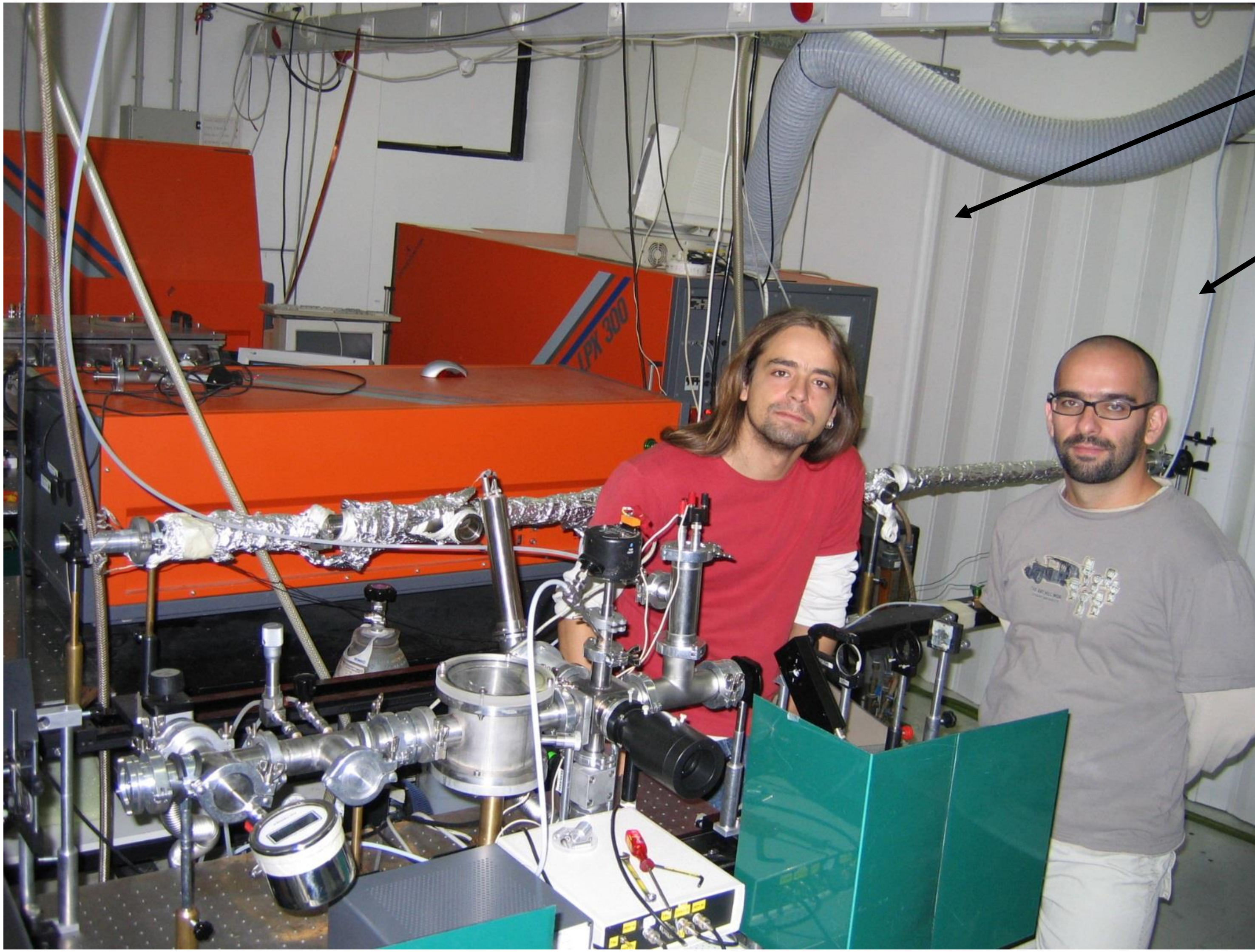
SPH production at BNL (current density record)







SPH production and detection on Crete



Dimitris

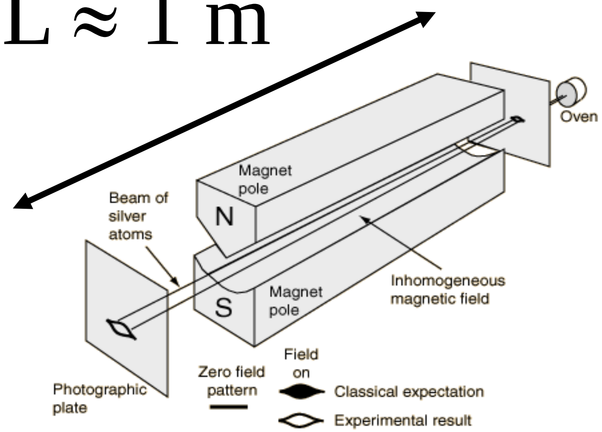
Luis

Comparison

	S-G(BNL)	Crete (lasers)
1) Density (SPH/cm^3)	10^{12}	$>10^{18}$
	Continuous	**Pulsed**
2) Production rate s^{-1}	10^{17}	10^{19}-10^{23}
3) Detection timescale	ms	ns
4) Velocity resolution	None	100 m/s
5) Polarization	0.92	0.45 (0.90)

"Single-molecule Stern-Gerlach Spin-Separator"

$L \approx 1$ m



$T \approx 1$ ms

X H

Explains why density of polarized atoms can be many orders of magnitude higher than traditional S-G experiment

$L \approx 1$ nm $T \approx 100$ fs

10^{18} cm^{-3} (1 ns) density estimated from H-Rb depolarization rate

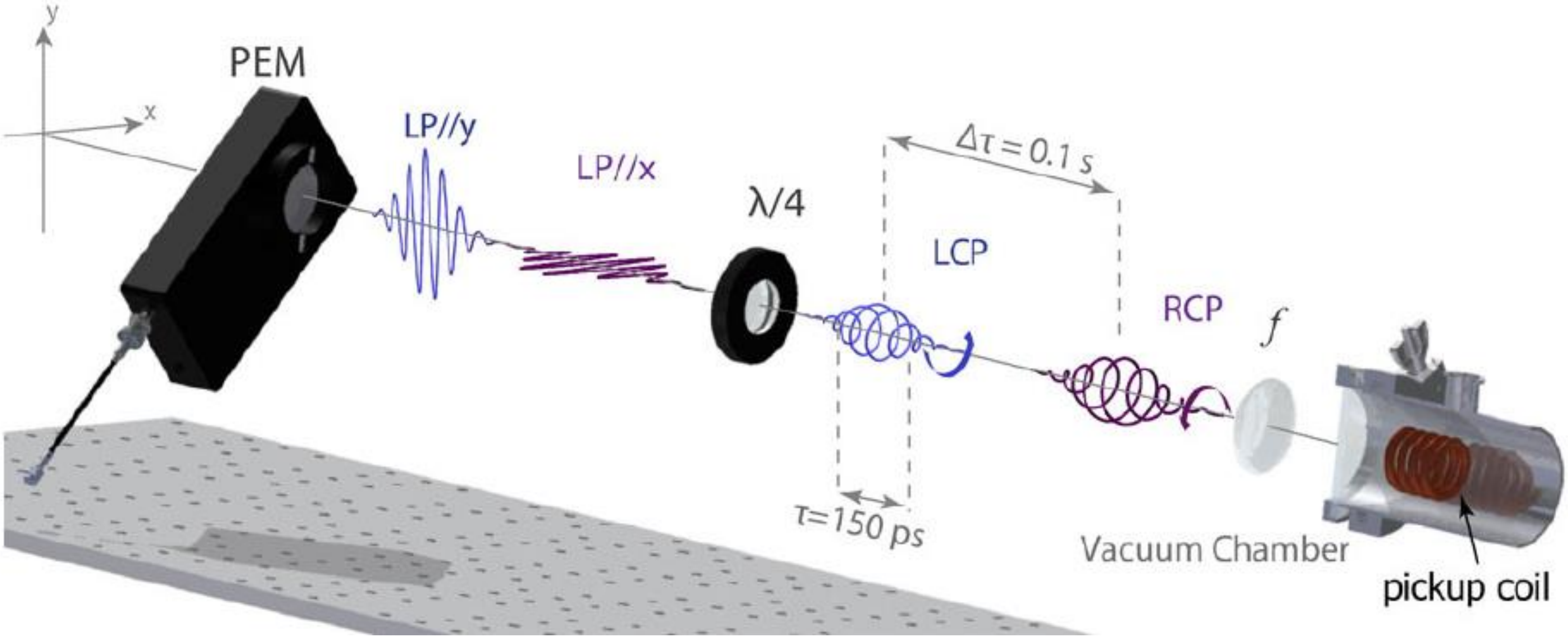
H-halogen-atom depolarization unknown

How high can the SPH density reach?

A direct measurement is necessary, before applications can be considered.

Recently inspired by development of kJ-MJ laser pulses for laser fusion

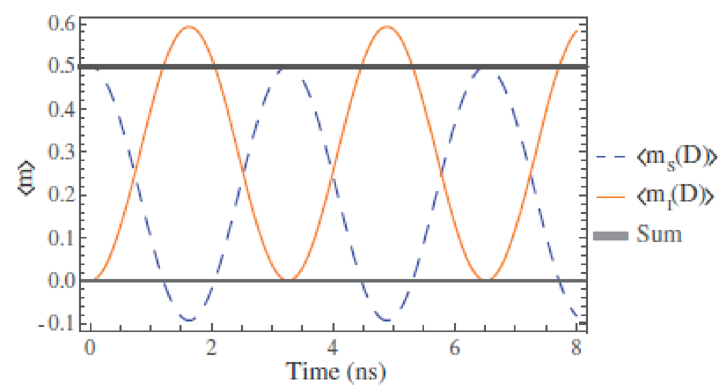
Time-dependent magnetization measurements with pickup coil



Hyperfine beating in electron polarization and magnetization:

$$\langle m_s(t)\rangle = \frac{1}{2}e^{-t/\tau_p}[1-\alpha\sin^2(\omega t/2)]$$

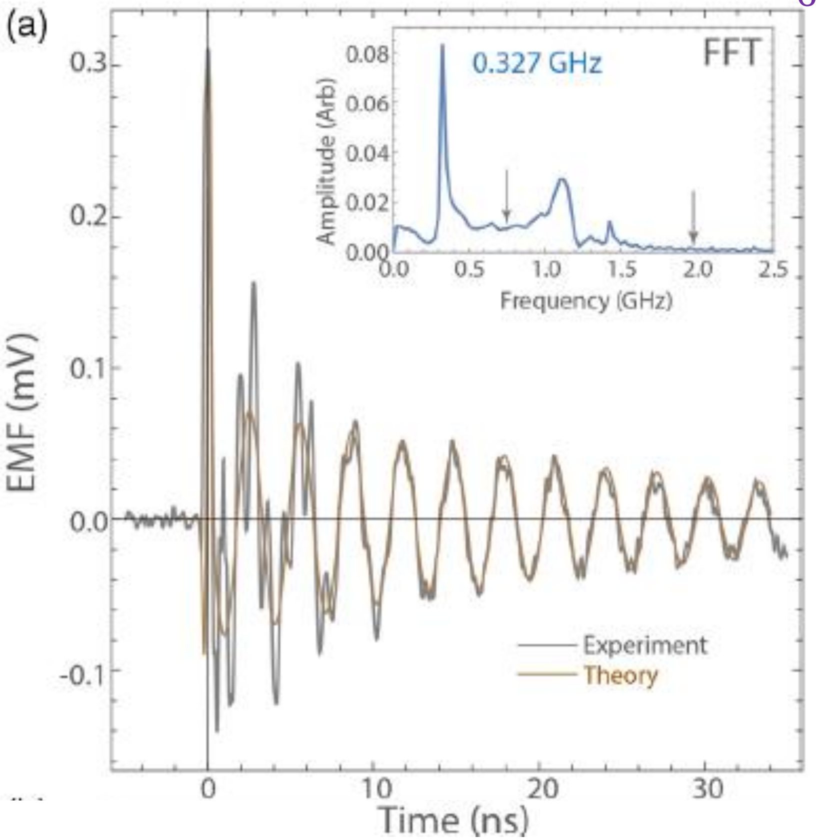
$$\langle m_I(t)\rangle = \frac{1}{2}e^{-t/\tau_p}[\alpha\sin^2(\omega t/2)]$$

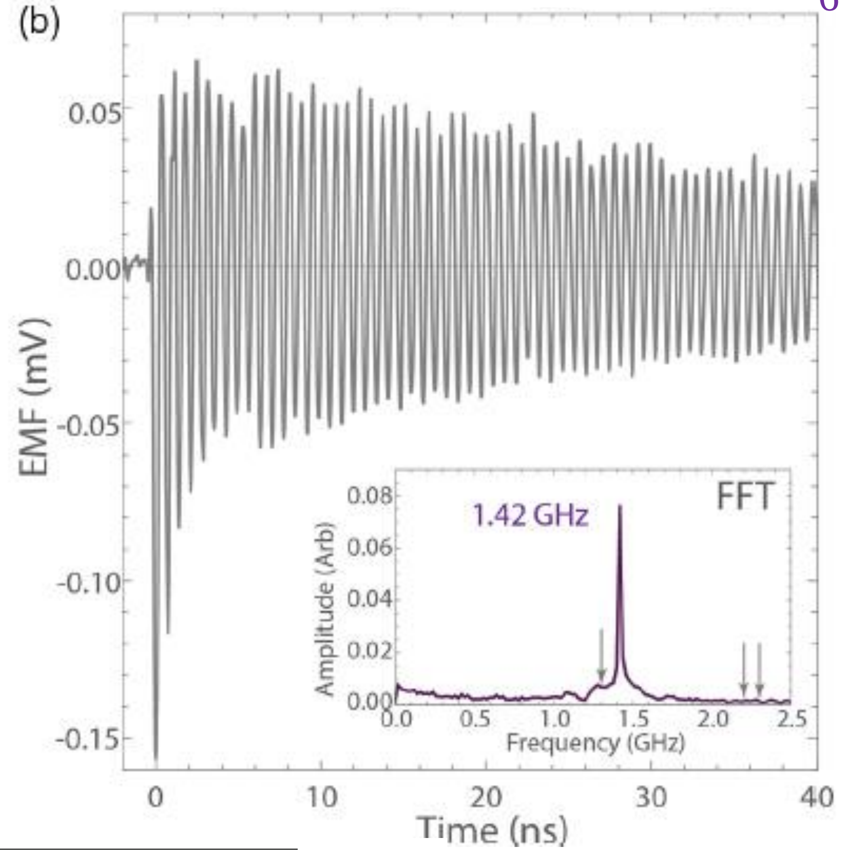


Magnetization oscillation at hyperfine beat frequency

125 mbar DI / 875 mbar SF_6

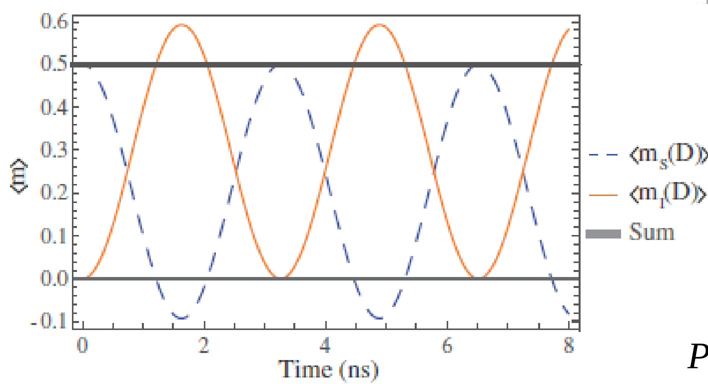
70 mbar HBr / 930 mbar SF_6





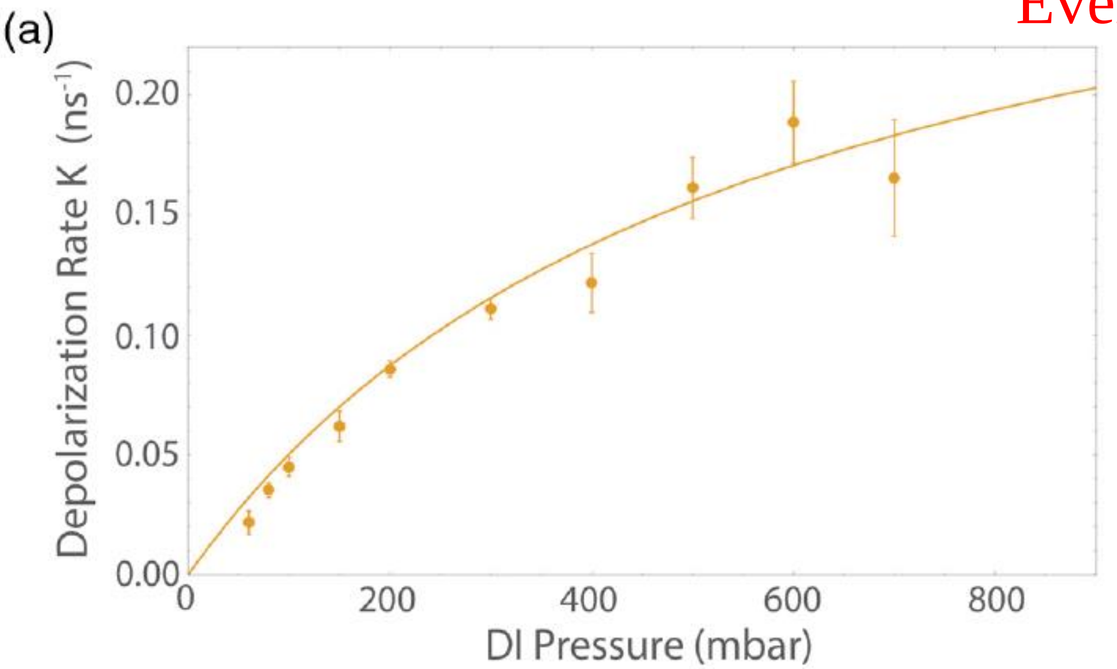
$$\langle m_s(t)\rangle = \frac{1}{2}e^{-t/\tau_p}[1-\alpha\sin^2(\omega t/2)]$$

$$\langle m_I(t)\rangle = \frac{1}{2}e^{-t/\tau_p}[\alpha\sin^2(\omega t/2)]$$



Phys. Rev. Lett. **121**, 083001 (2018).

SPD depolarization rate vs. DI pressure



Depolarization saturating.
Even higher densities possible?

$$D^{\uparrow} + DI \underset{k_{-1}}{\overset{k_1}{\rightleftharpoons}} DI{-}D^{\uparrow}$$

Complex formation

$$DI{-}D^{\uparrow} \xrightarrow{k_d} DI{-}D$$

Intramolecular depolarization

$$DI{-}D^{\uparrow} + DI \xrightarrow{k_2} D^{\uparrow} + 2\,DI$$

Collisional dissociation DI

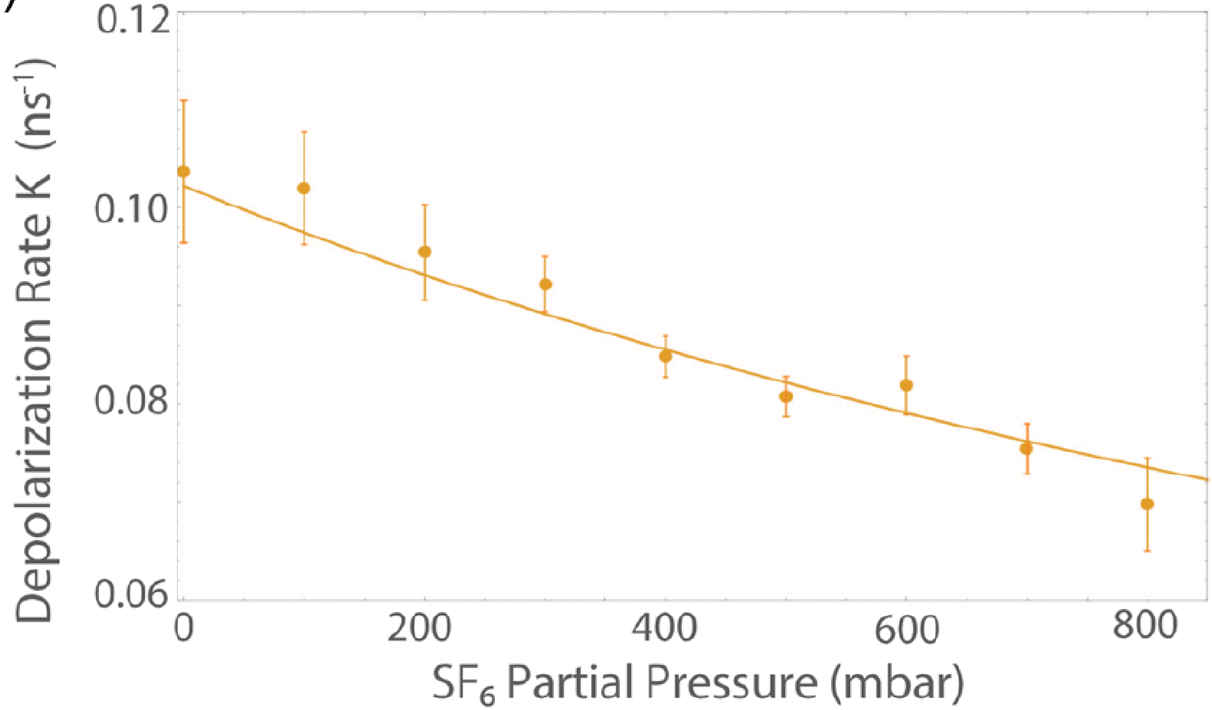
>10^{19} cm^{-3} density (~10 ns lifetime)
>100 time higher than expected

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

SPD depolarization rate vs. SF_6 pressure:

Evidence of DI-D complex destruction

(b)

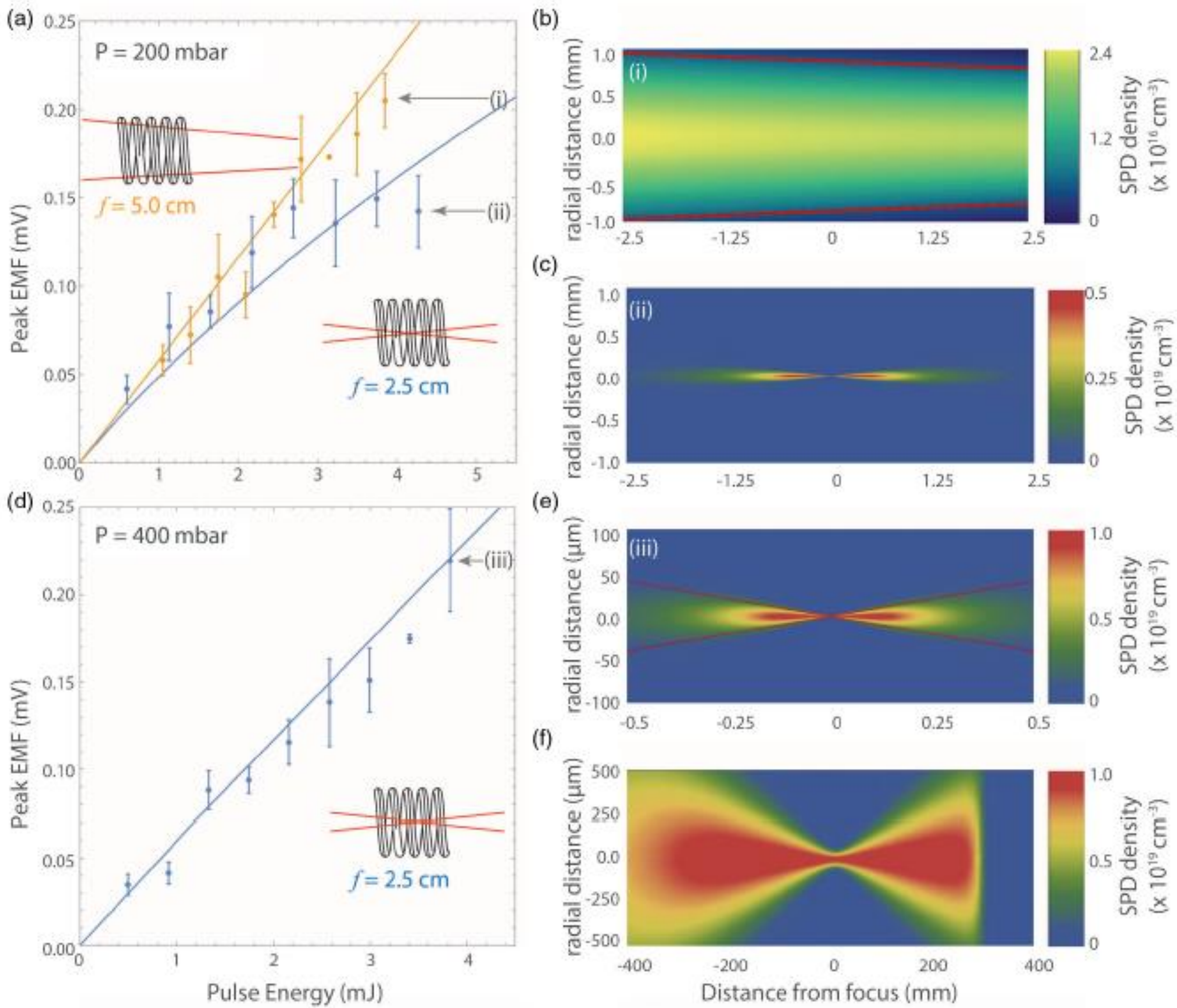


$$\text{DI-D}^{\uparrow} + X \xrightarrow{k_2^X} \text{D}^{\uparrow} + \text{DI} + X$$

Collisional dissociation by inert gas X

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

Signal vs. laser intensity and focusing



Summary I

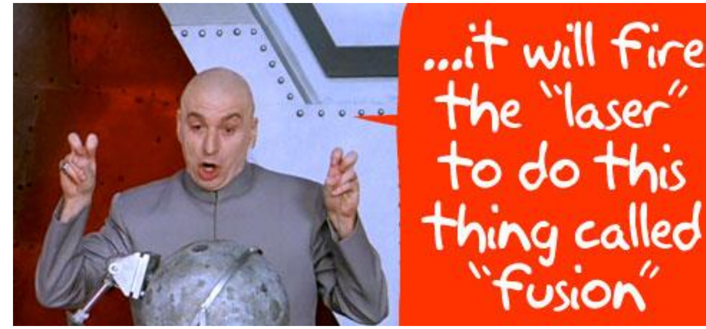
- SPH densities of 10^{19} cm^{-3} (~1 atm)

depolarization rate flattens at high pressure.

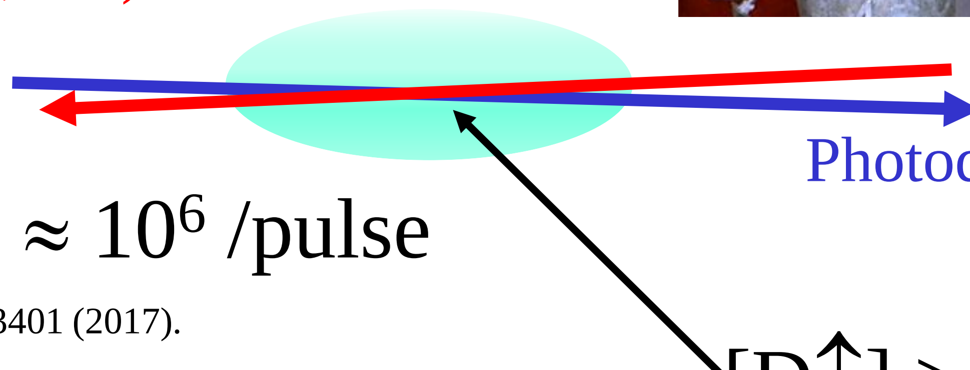
- Future work: Go to higher pressures (5-10 atm),

as even higher densities are likely possible.

Pump-probe polarized Fusion
(D-^3He or D-T)



Fusion laser
2 MJ/shot
at NIF (Livermore, CA)



Photodissociation
laser

Fusion counts ≈ 10^6 /pulse

Phys. Rev. Lett. **118**, 233401 (2017).

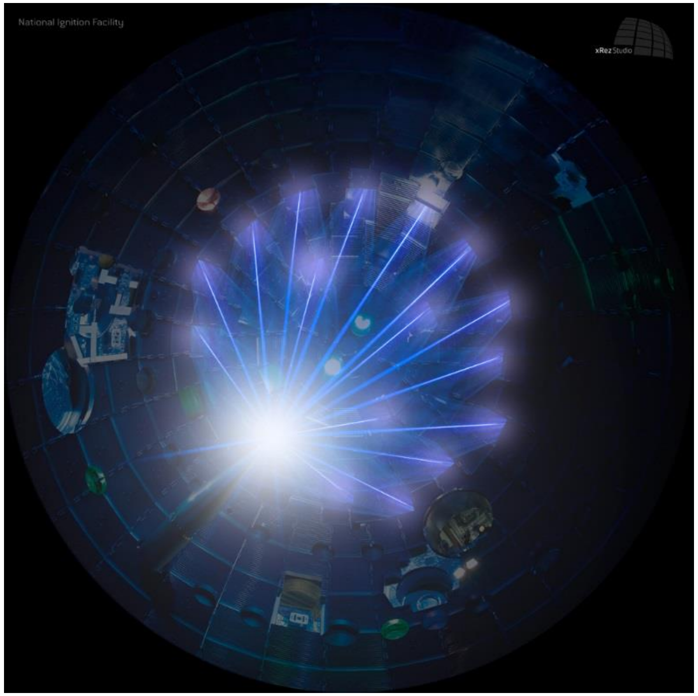
$[D\uparrow] > 10^{19}$ cm^{-3}

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

Neutron angular distribution becomes anisotropic ONLY if polarization survives, $p > 0$.

R. M. More, Phys. Rev. Lett. 51, 396 (1983).

National Ignition Facility (NIF, Livermore CA



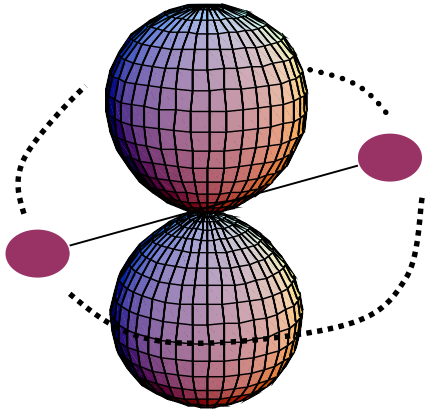
2 MJ / (ns pulse)

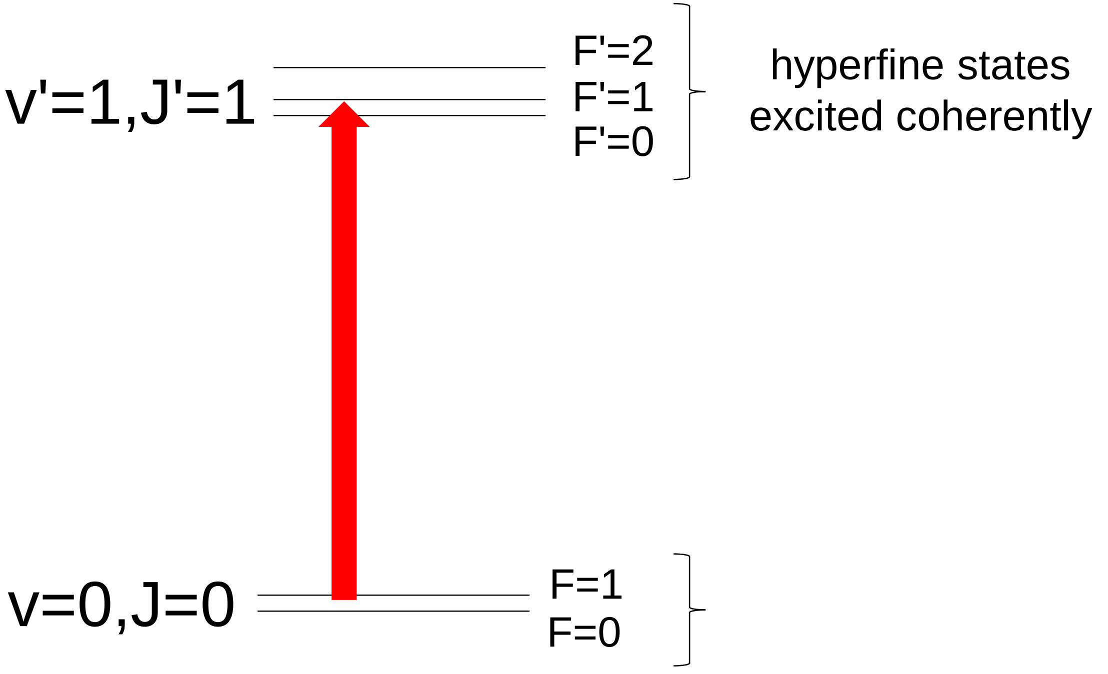
Summary II

- UV photodissociation method good for generating high SPD densities for **demonstration** of polarized laser fusion (**not** for generating pellets for laser fusion).

Method 2: Pulsed IR rovibrational excitation of molecular beams

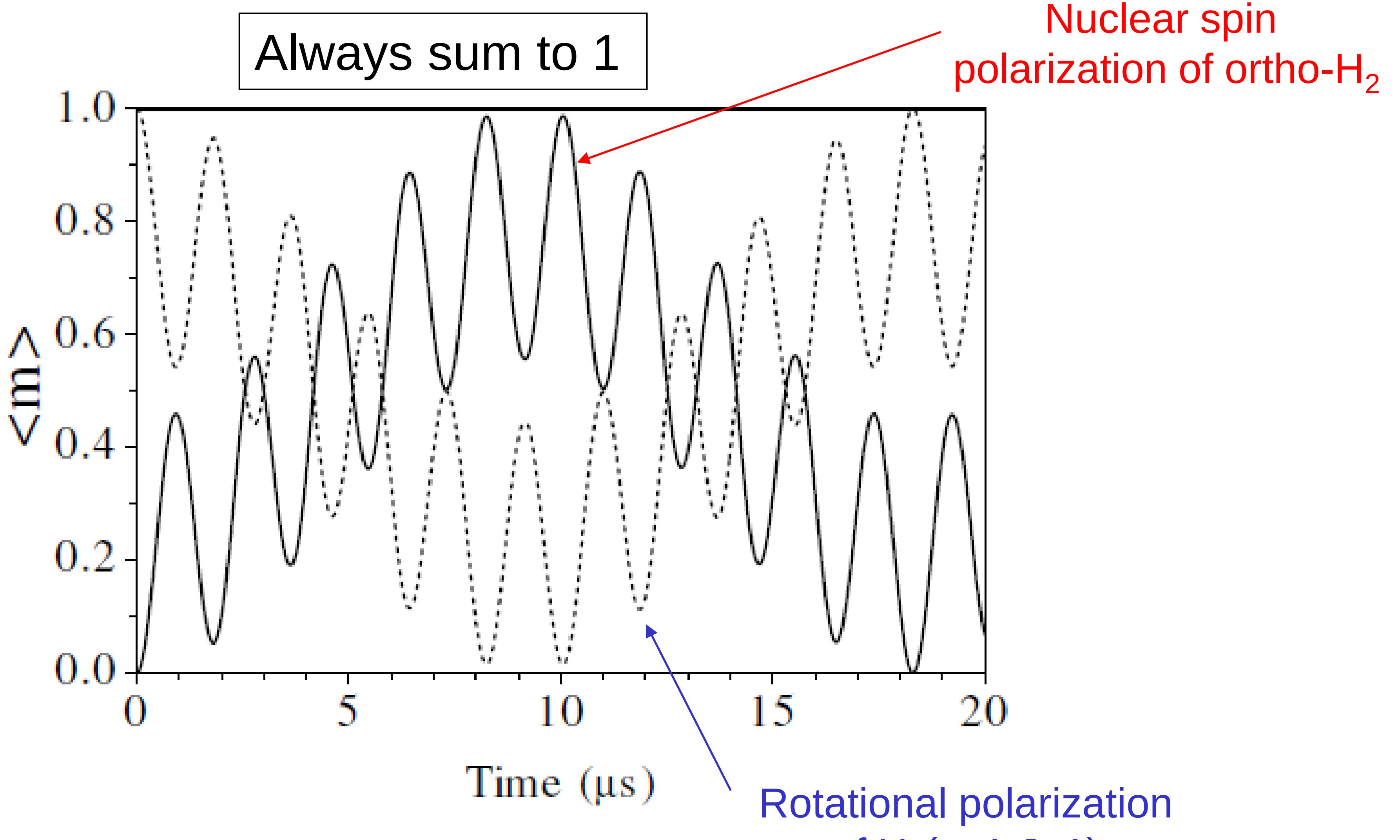
Polarizing molecular rotation (instead of electronic spin):



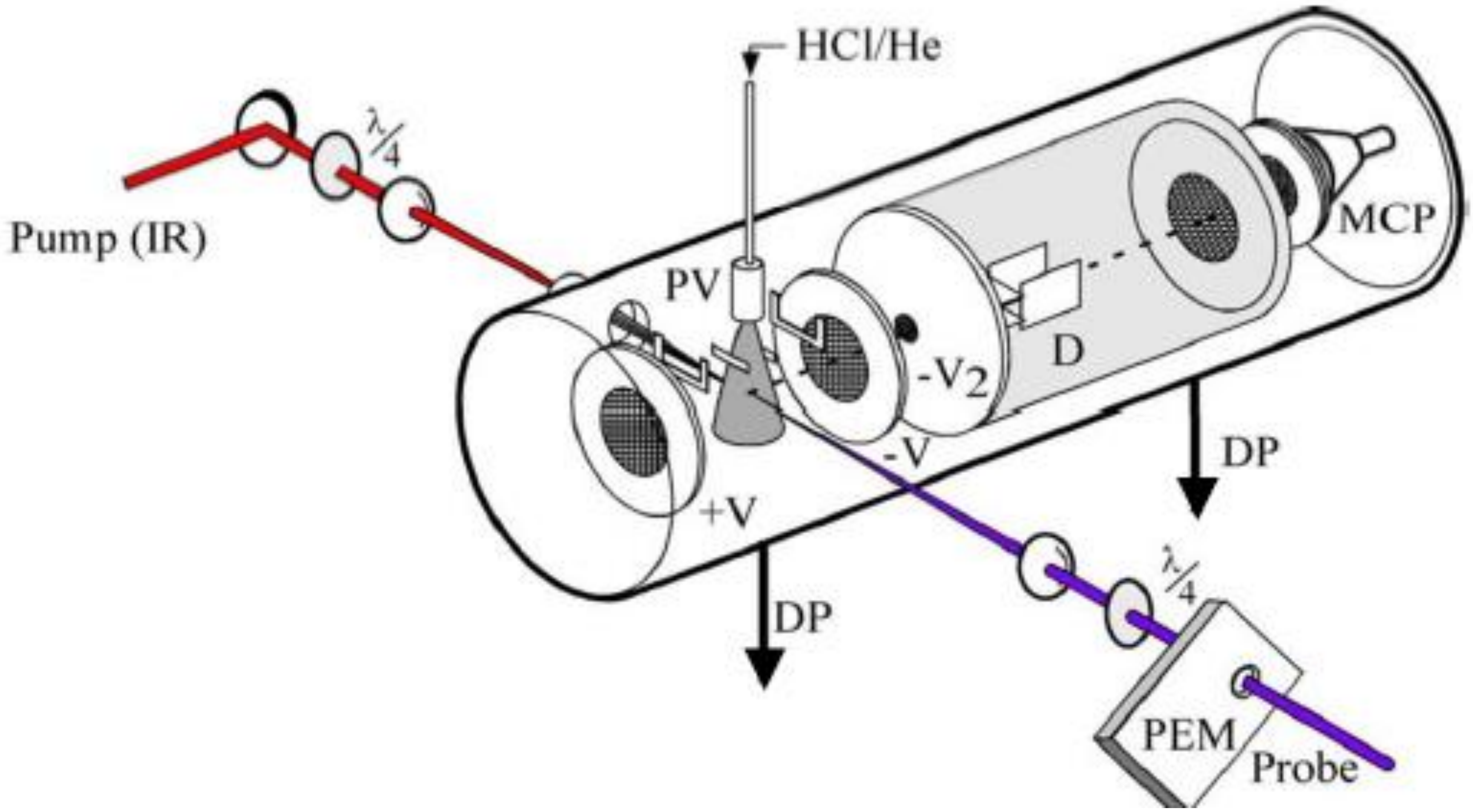


T. Peter Rakitzis, *Phys. Rev. Lett.* **94**, 83005 (2005).

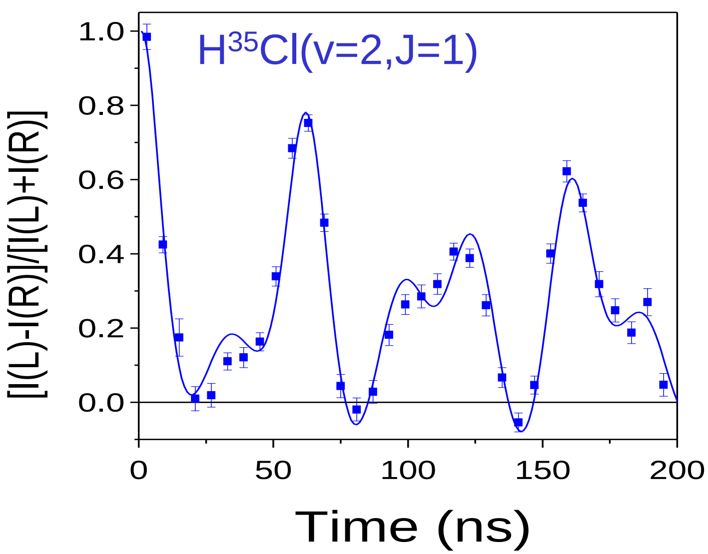
Pulsed Excitation of H_2

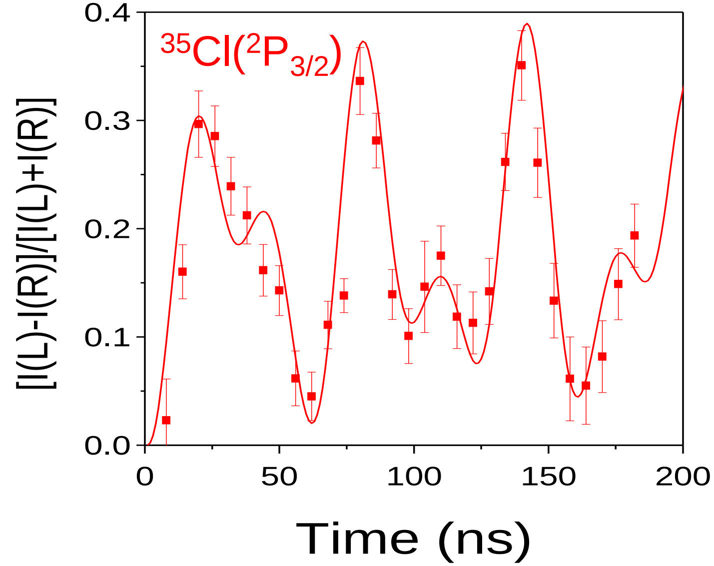


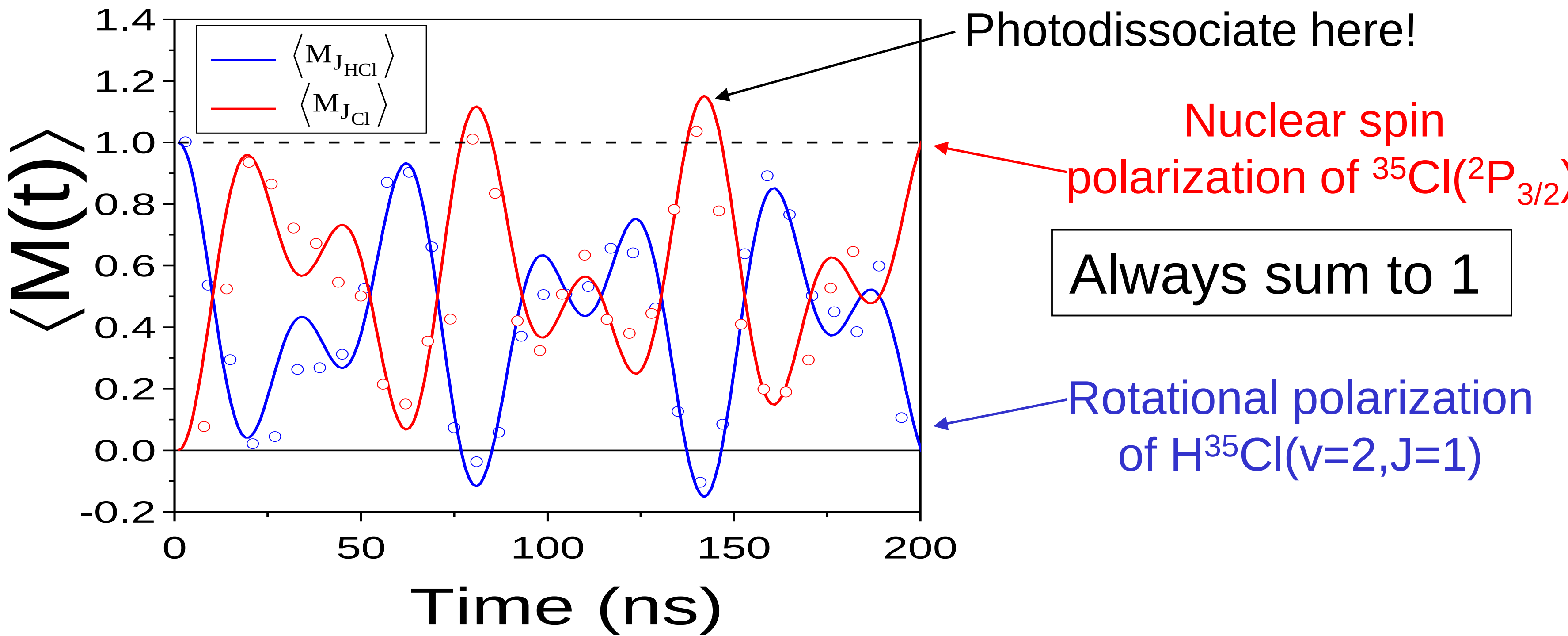
Pump-Probe (IR-UV) setup for measurement of hyperfine beating



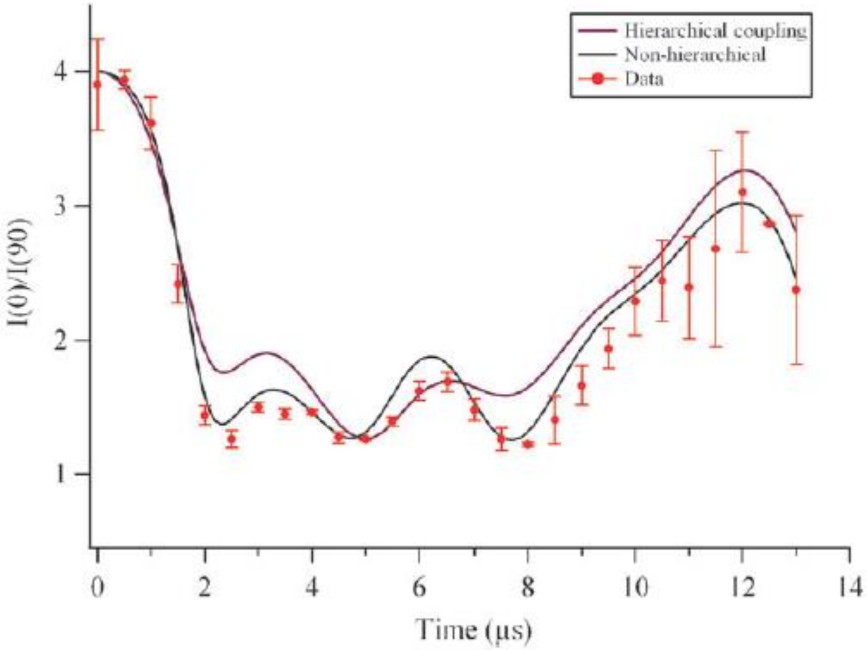
Pulsed Excitation of HCl







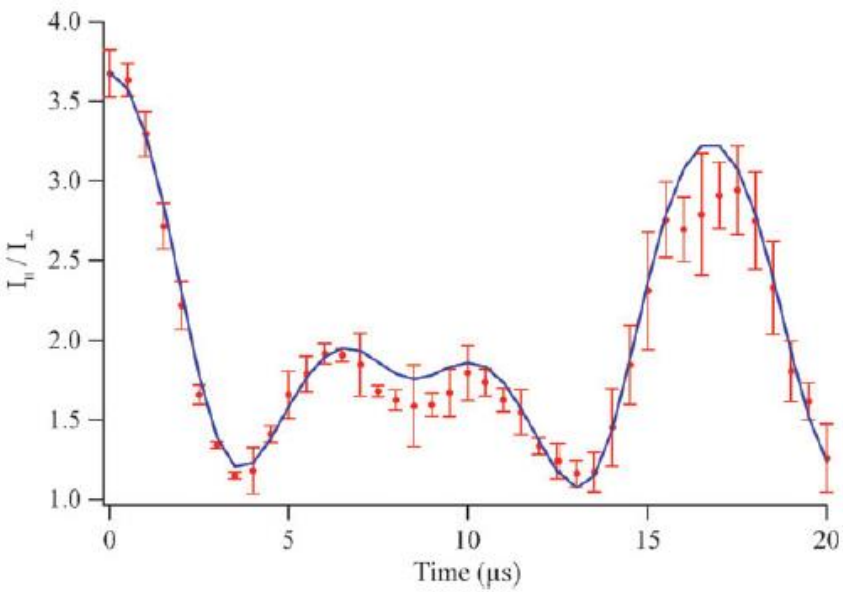
Hyperfine beating in HD and D_2



HD(v=1, j=2, m=0)

Phys. Chem. Chem. Phys., 2009, **11**, 142

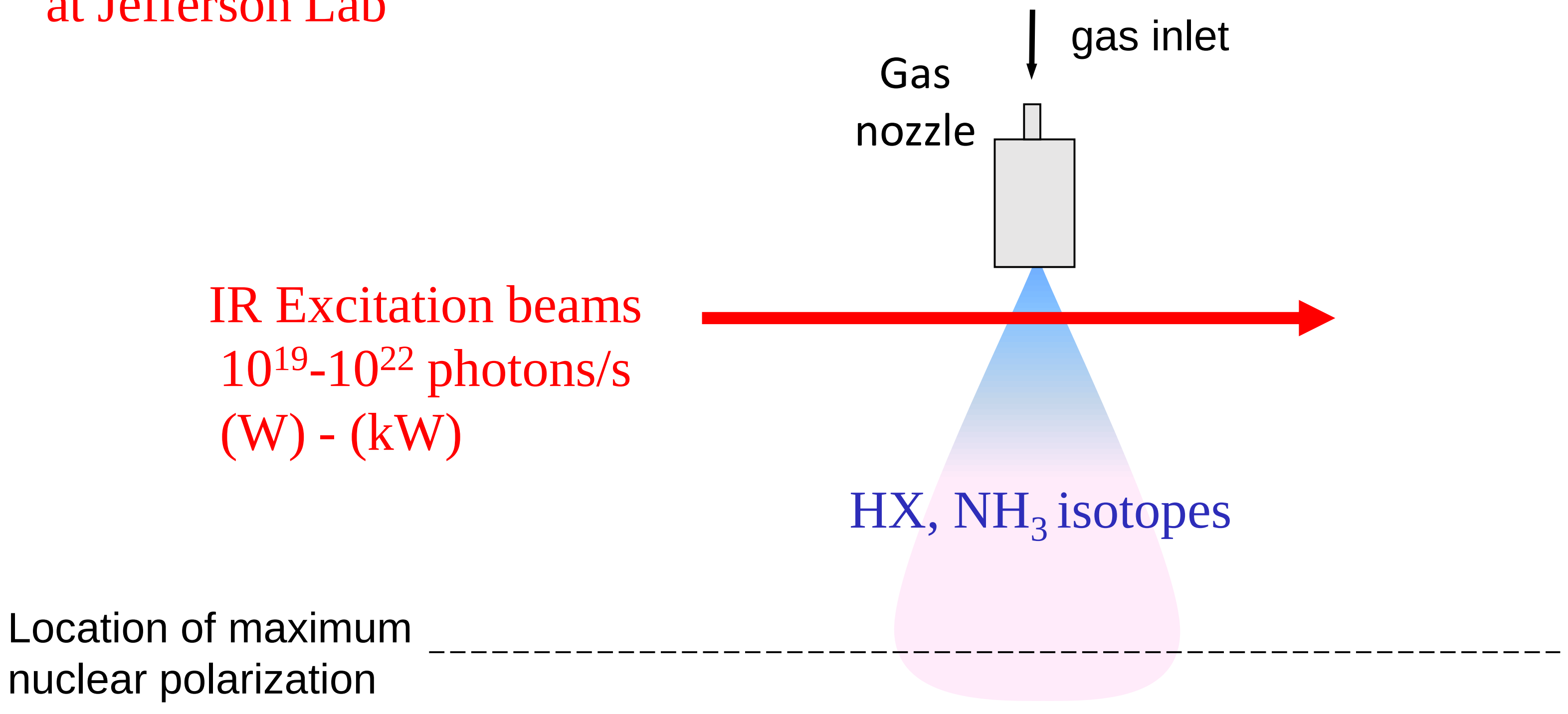
Note: IR excitation not possible here, rovibrational excitation performed with 2-photon stimulated Raman pumping, of low efficiency.



D_2(v=1, j=2, m=0)

Phys. Chem. Chem. Phys., 2010, **12**, 15689

Planning scaled-up experiments at Jefferson Lab

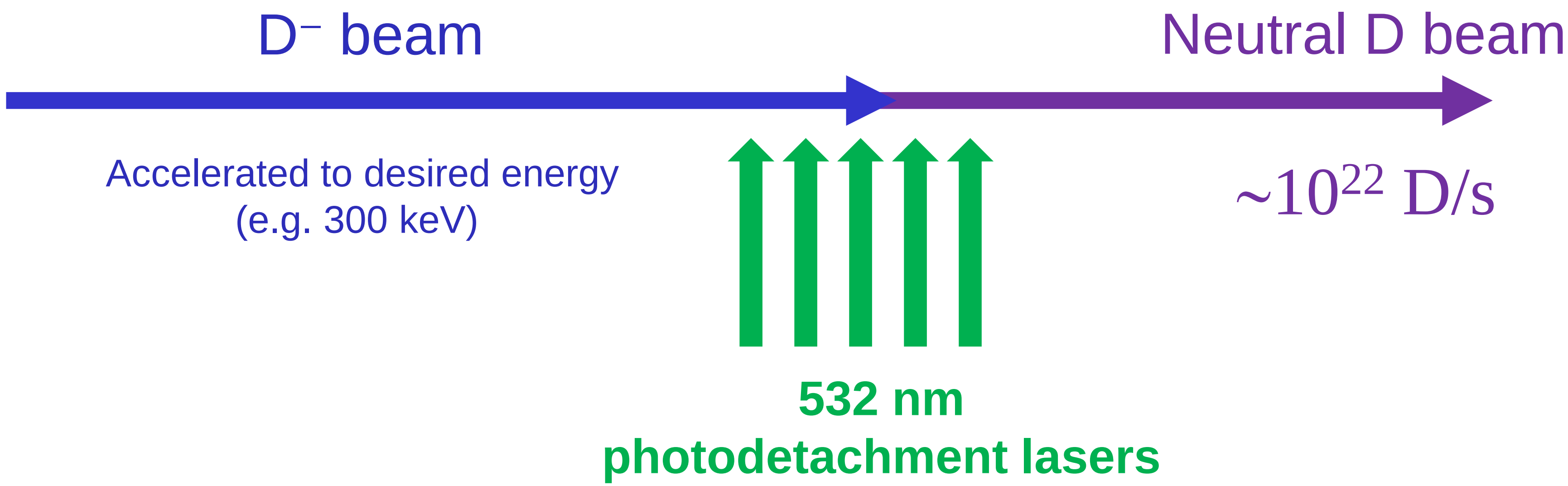


Freeze polarization in nuclei by:

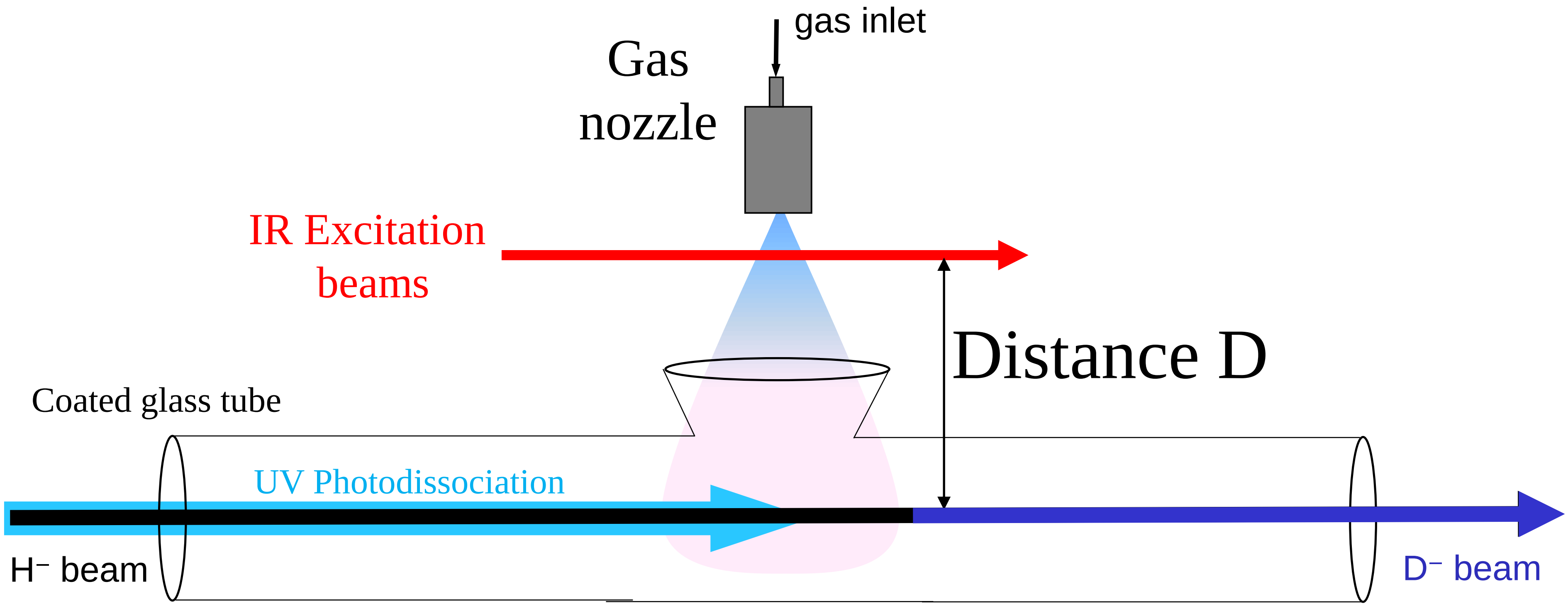
(1) Freezing molecules at surface, OR
(2) Introduction a B-field, with $B>B_c$, OR
(3) Photodissociate molecule

Planned spin-polarized production rate of 10^{19}-10^{22} molecules/s

Fast neutral D beam for fusion reactor



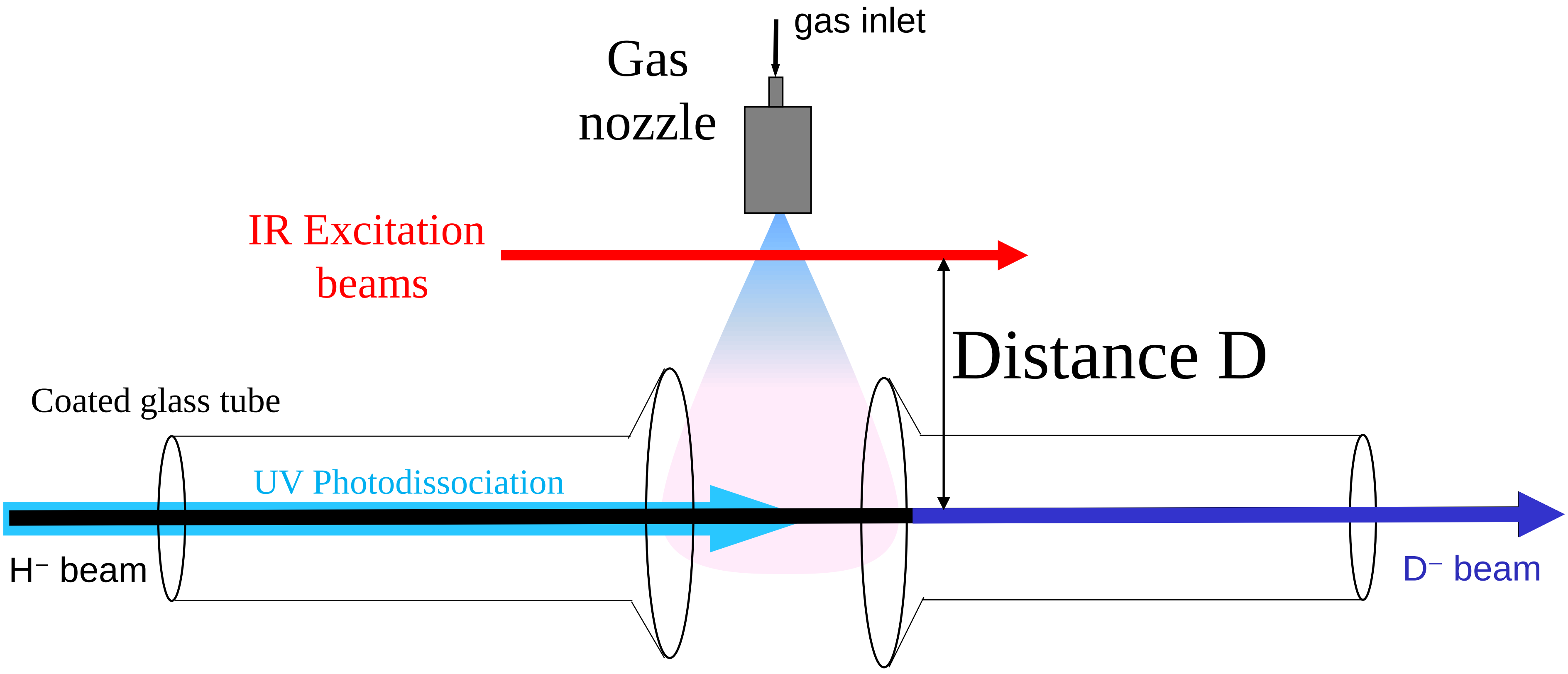
Proposal for D⁻ beam production



$$H^- + D \rightarrow H + D^-$$

Charge exchange efficiency demonstrated at 15%

Proposal for D^- beam production



$$H^- + D \rightarrow H + D^-$$

Charge exchange efficiency demonstrated at 15%

High-power lasers and production rates

• Commercial appropriate IR and UV lasers exist with

10^{19}-10^{21} photon/s

• Assuming 1-10% total efficiency, 10^{18}-10^{20} D/s beams possible.

• Crucial to understand the efficiency of each step, in proof-of-principle experiment.

• Clearly (even if very efficient) 10^{22} D/s beams for ITER will require custom, industrial-scale lasers (100s of kW, which exist).

Conclusions

• Two new methods for production of spin-polarized molecules:

(1) UV photodissociation ⇒ Demonstration of Polarized laser Fusion

(2) IR rovibrational excitation ⇒

(a) Large production rates of spin-polarized molecules

(b) Likely large production rates of spin-polarized D^- and T^- possible

All ideas are welcome

Acknowledgements

FORTH
Dimitris Sofikitis
Chrysovalandis Kannis

IESL-FORTH and
University of Crete

Alexander Andreev (Szeged, Berlin)

Juelich
Markus Büscher
Ralf Engels
Anna Hützen

← Method 1

European Research Council

Jefferson Lab
Andy Sandorfi
Matt Poelker
Amy Sy

← Method 2

MARIE CURIE ACTIONS