



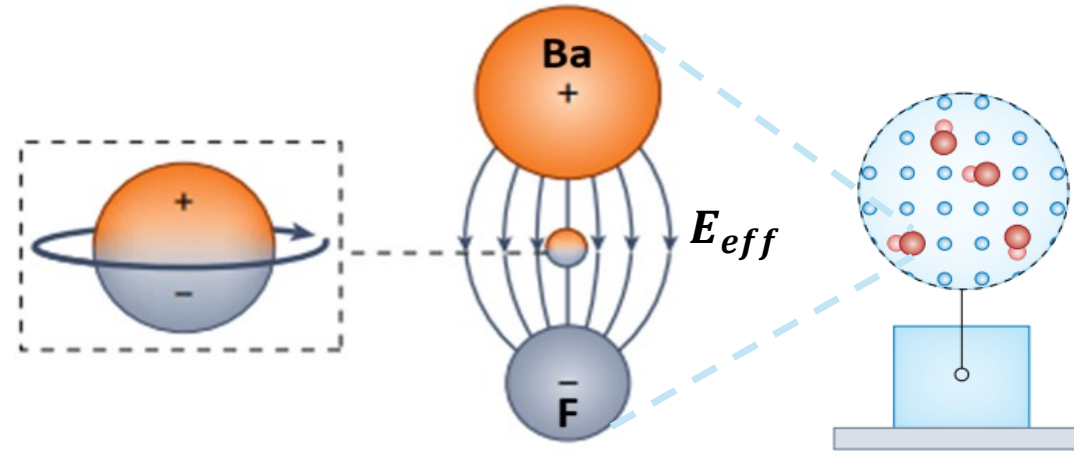
PNRR MUR project PE000023-NQSTI



Istituto Nazionale di Fisica Nucleare

CSN gruppo V

Measuring the electric dipole moment of the electron using polar molecules in a parahydrogen matrix



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on behalf of the PHYDES Collaboration

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Outline

- Motivation for an eEDM measurement
- Phydes proposal
- $p\text{H}_2$ production and characterization
- BaF molecular source
- Conclusions and future work

BSM physics with eEDM

A nonzero eEDM implies an aspherical charge distribution along the electron's spin axis

→ violation of T-symmetry

→ violation of CP.

STANDARD MODEL PREDICTION

$$d_e^{SM} \leq 10^{-38} e \text{ cm}$$

TOO SMALL

SM extensions allow a much larger eEDM that is within reach of near-term experiments.

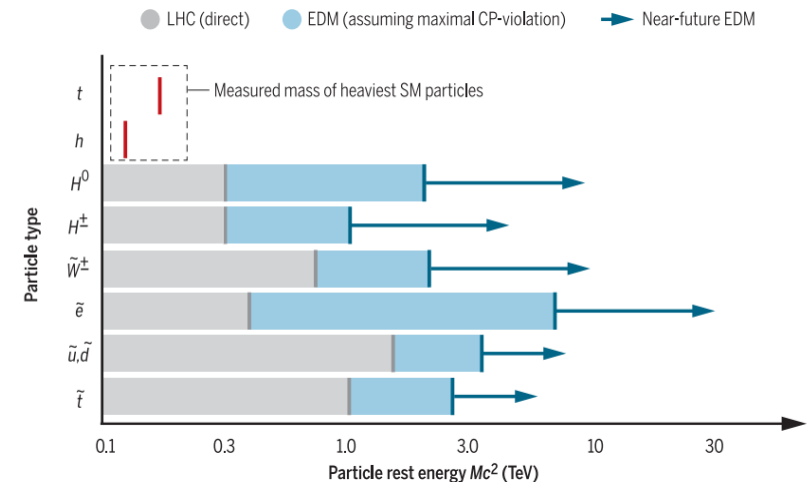
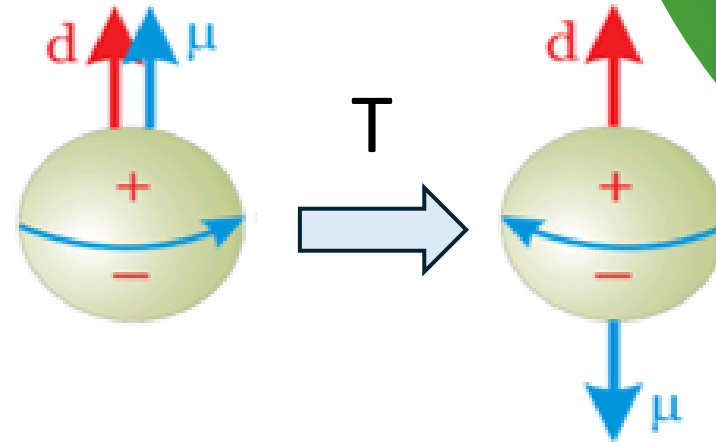
CURRENT EXPERIMENTAL LIMIT

$$d_e < 4.1 \times 10^{-30} e \text{ cm}$$

@ 90% confidence level

[T. S. Roussy *et al.*, *Science* **381**,46-50 (2023)]

(obtained with trapped HfF+)

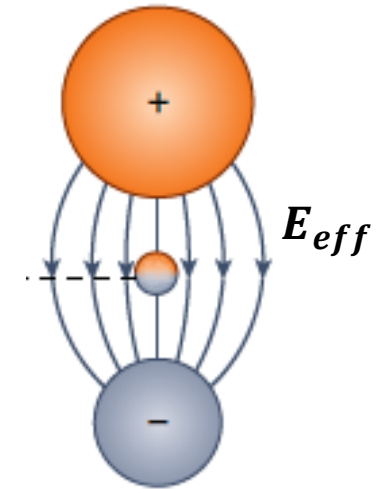


[DeMille *et al.*, *Science* **357**, 990–994 (2017)]

Powerful window on energy scales much larger than those probed directly at LHC!

Ingredients for an eEDM measurement

Diatomic polar molecules (BaF, YbF, ThO) have a single valence electron exposed to a huge effective molecular electric field ($E_{\text{eff}} \sim 10 \text{ GV/cm}$)



Measure electron spin precession frequency in a magnetic and electric field and detect changes in precession rate when the electric field direction is reversed.

FIGURE OF MERIT

$$\delta d_e = \frac{\hbar}{2 E_{\text{eff}} \sqrt{N} t_p}$$

E_{eff} = effective electric field inside the molecule ($> \sim 10 \text{ GV/cm}$)

t_p = spin precession measurement time $\leq T_{\text{coherence}}$

N = integrated n° of electrons whose precession is detected

Assuming:

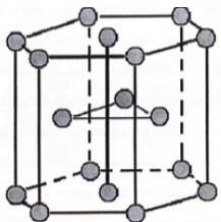
$N \approx 10^{16}$ number of molecules (electrons) interrogated

$t_p \approx 3 \text{ ms}$

$$\delta d_e = \sim 10^{-31} e \text{ cm}$$

Matrix isolation technique

pH₂ : anti-parallel nuclear spins, lower-energy state of molecular H₂

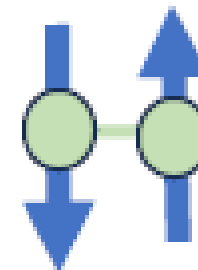


Stable hexagonal closed packed (hcp) structure.

Lattice parameter $\approx 3.78 \text{ \AA}$

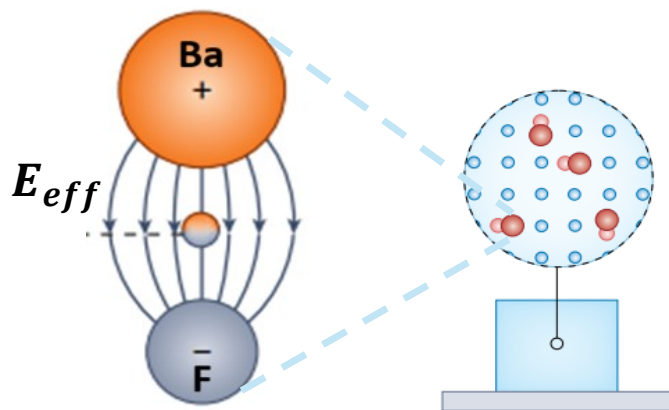
$\rho = 0.086 \text{ g / cm}^3$

optically transparent media



Guest molecules in a matrix of pH₂ gas solidified at cryogenic temperature

- Big intermolecular distance \rightarrow no significant deformations of the crystalline structure due to doping
- Minimal interactions \rightarrow long coherence time



modified from:

[DeMille, et al., *Nat. Phys.* **20**, 741–749 (2024)]

$T_{\text{coherence}}$ measured in alkali atoms in pH₂ \approx hundreds of ms
[J. Weinstein et al., PRL 125, 043601(2020)]

Pros: high number number of molecules within the measurement volume (BaF is very reactive!)

Cons: solid environment, need to control systematics to preserve coherence times

PHYDES

(Parahydrogen and diatomic molecules for EDM studies)

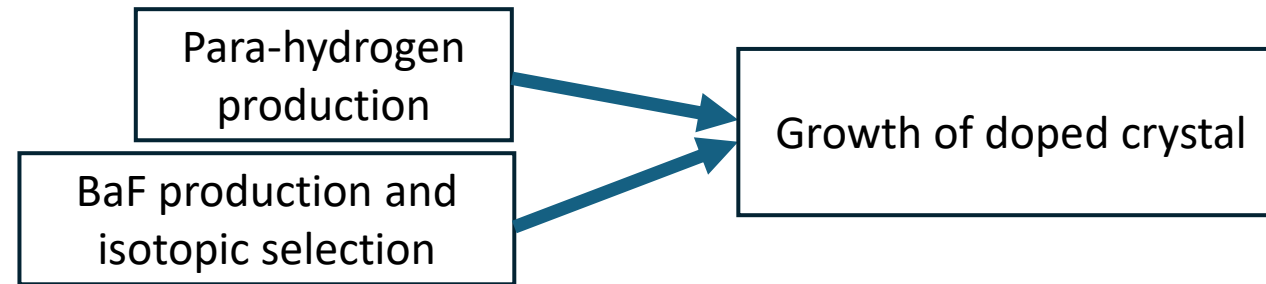
R&D project (CSN5 INFN) to study the production of BaF-doped pH₂ crystal matrices ($n \approx 10^{15}$ BaF molecules / cm³, approx. 0.1 ppm)

Detection methods under study:

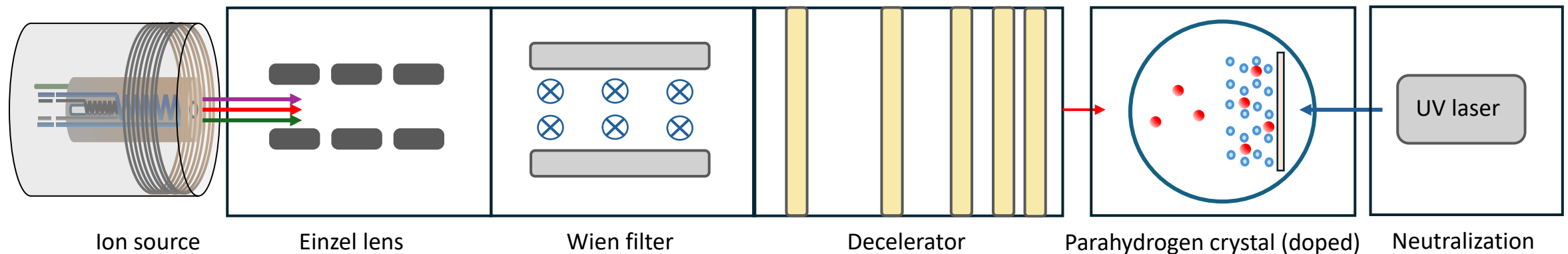
- **Optical detection** with Laser Induced Fluorescence spectroscopy:
Measure the population of a prepared coherent superposition state and detect any change in population (precession angle) when E is reversed.
- **Electron Paramagnetic Resonance (EPR)** detection:
Microwave induced precession frequency shift in a high external field.

BaF-doped crystal production

2 different sub-systems



- BaF⁺ is produced in a glow discharge chamber from BaF₂ powder
- molecules are accelerated to 1 keV
- isotopically selected with a Wien velocity filter ($\vec{E} \times \vec{B}$)
- decelerated to ≈ 5 eV and mixed with a pH₂ gas flow on a sapphire substrate
- neutralized with photo-extracted electrons from a gold layer on the substrate

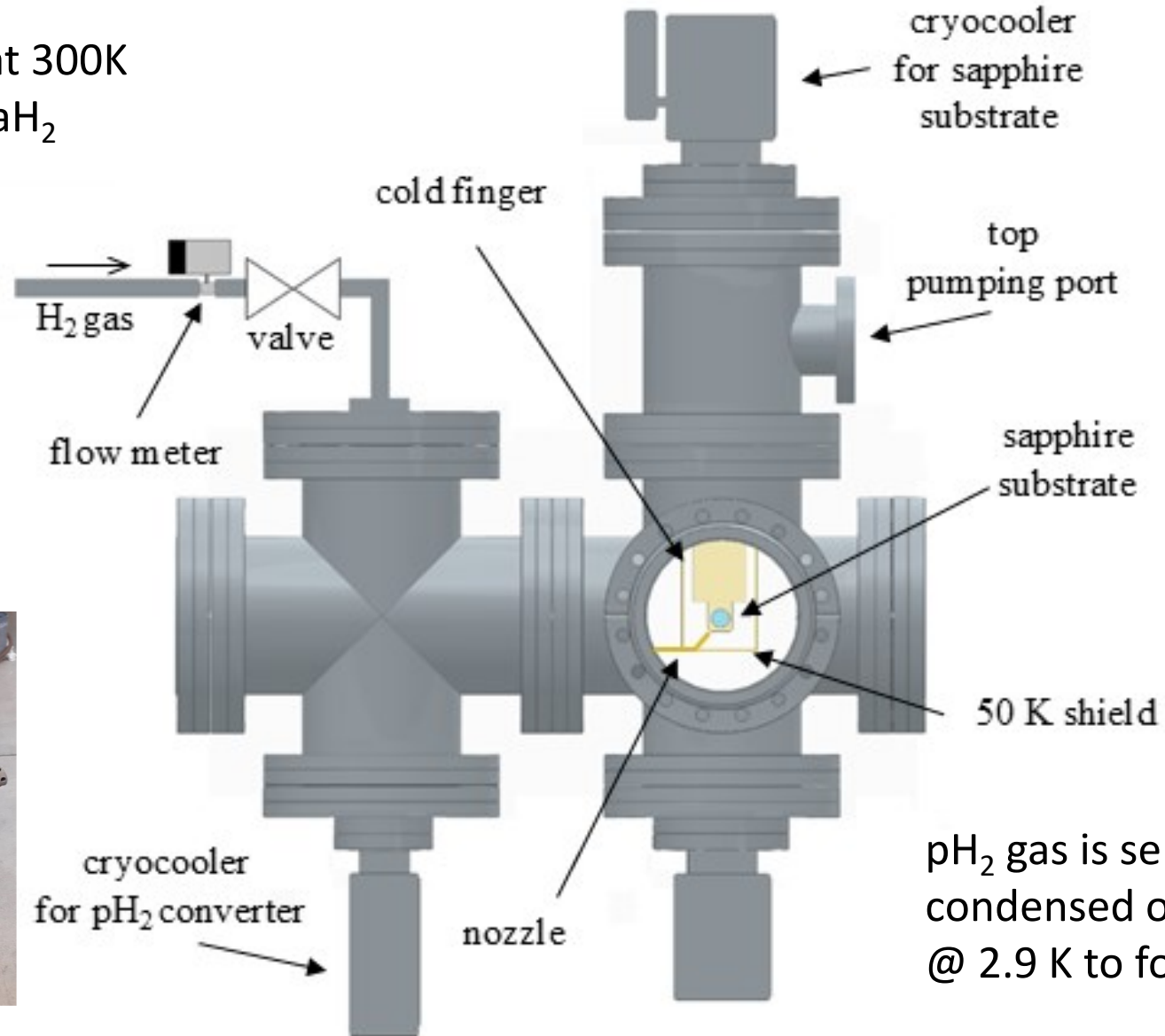


Parahydrogen production and crystal growth

High purity hydrogen gas at 300K
75% orthoH₂, 25% paraH₂

CONVERTER

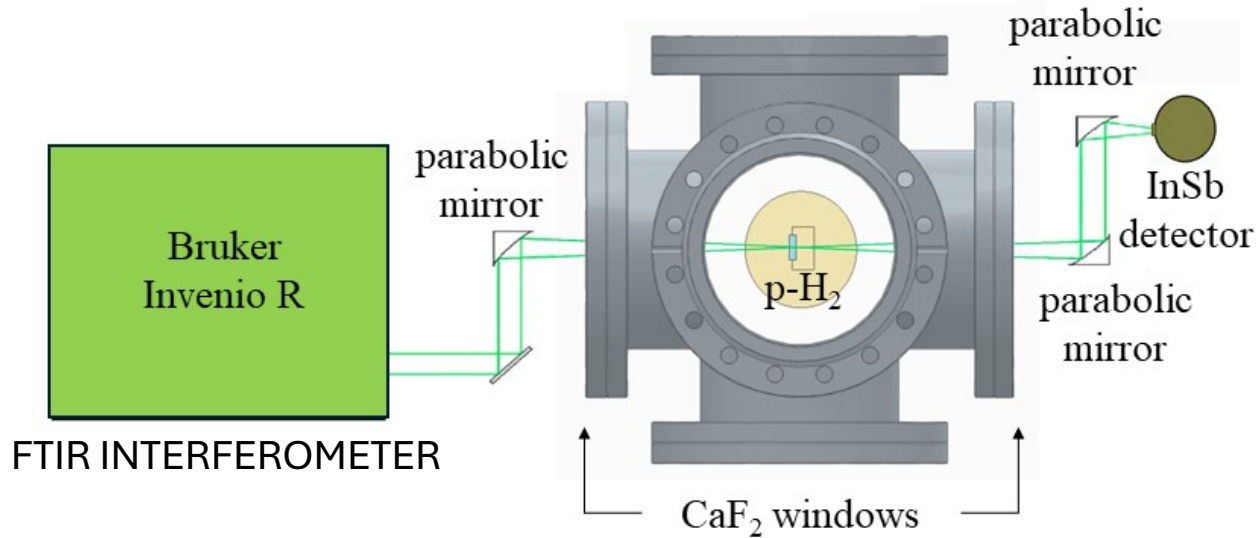
Copper line filled with
hydrous ferric oxide catalyst
 $T_{\text{converter}} \approx 20 \text{ K}$



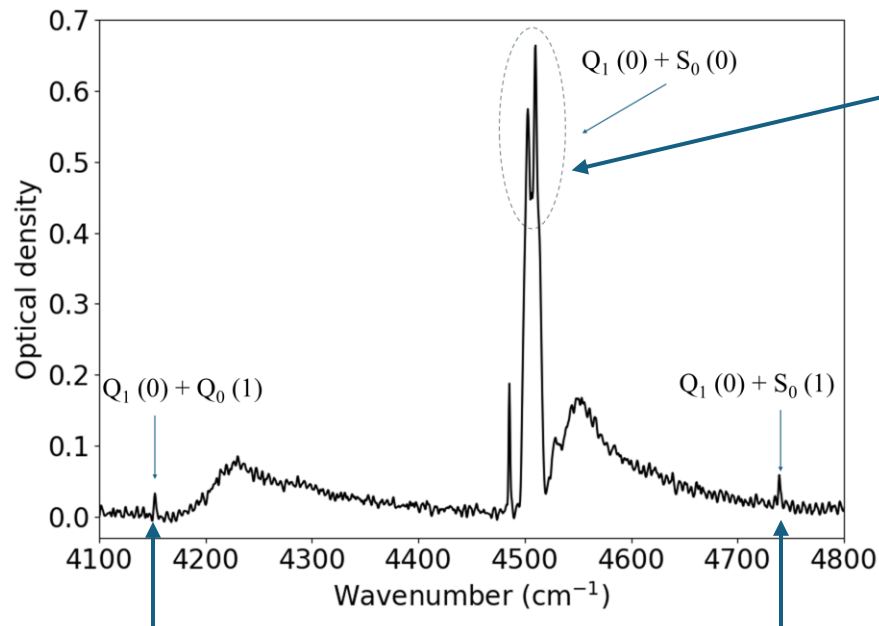
Sapphire substrate
 $\phi = 1''$
 $T_{\text{sapphire}} = 2.9 \text{ K}$

pH₂ gas is sent through a nozzle and
condensed on a sapphire substrate
@ 2.9 K to form a solid crystal

Characterization of the cryogenic matrix (I)



A Fourier transform interferometer (FTIR) with a near-infrared light source is used to measure the absorption spectrum of the p-H₂ matrix.



Thickness of deposited layer

Ortho/para-H₂ ratio

$$h(\text{mm}) = 0.048 \int_{4485 \text{ cm}^{-1}}^{4520 \text{ cm}^{-1}} OD(\nu) d\nu$$

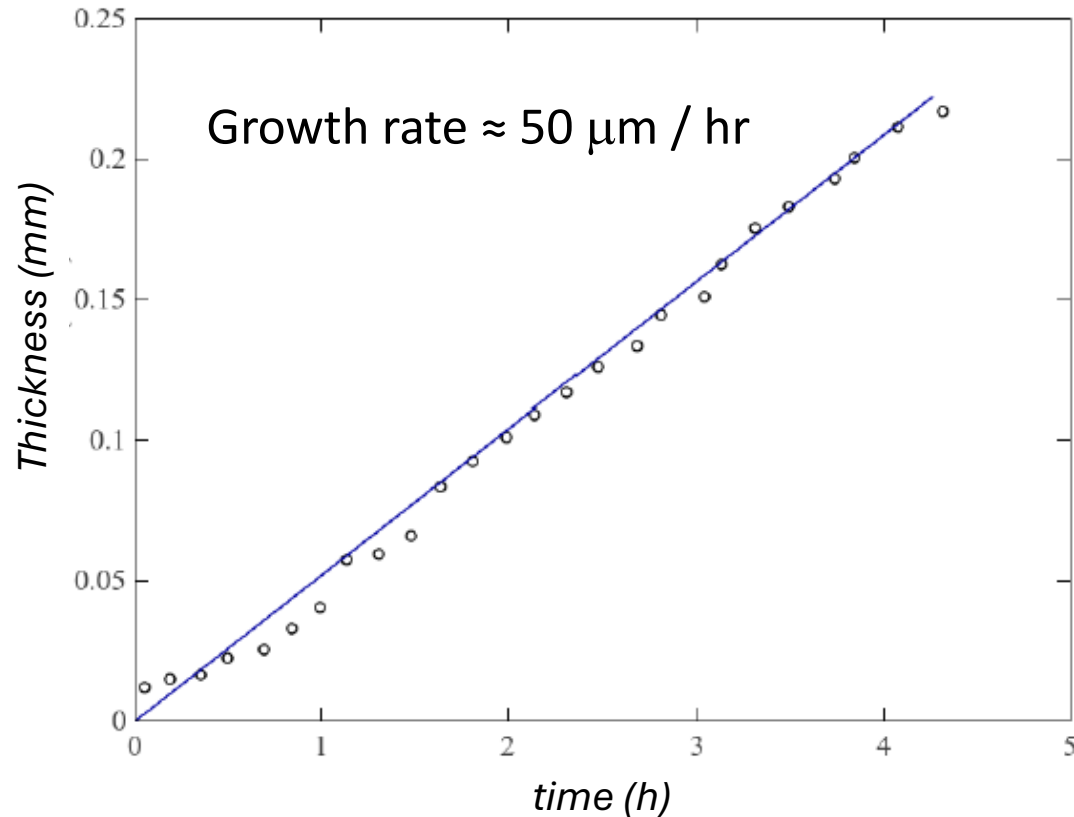
$$f_{\text{ortho}} = \frac{0.124 \text{ mm}}{h(\text{mm})} \int_{4151 \text{ cm}^{-1}}^{4154 \text{ cm}^{-1}} OD(\nu) d\nu$$

$$f_{\text{ortho}} = \frac{0.0787 \text{ mm}}{h(\text{mm})} \int_{4732 \text{ cm}^{-1}}^{4742 \text{ cm}^{-1}} OD(\nu) d\nu$$

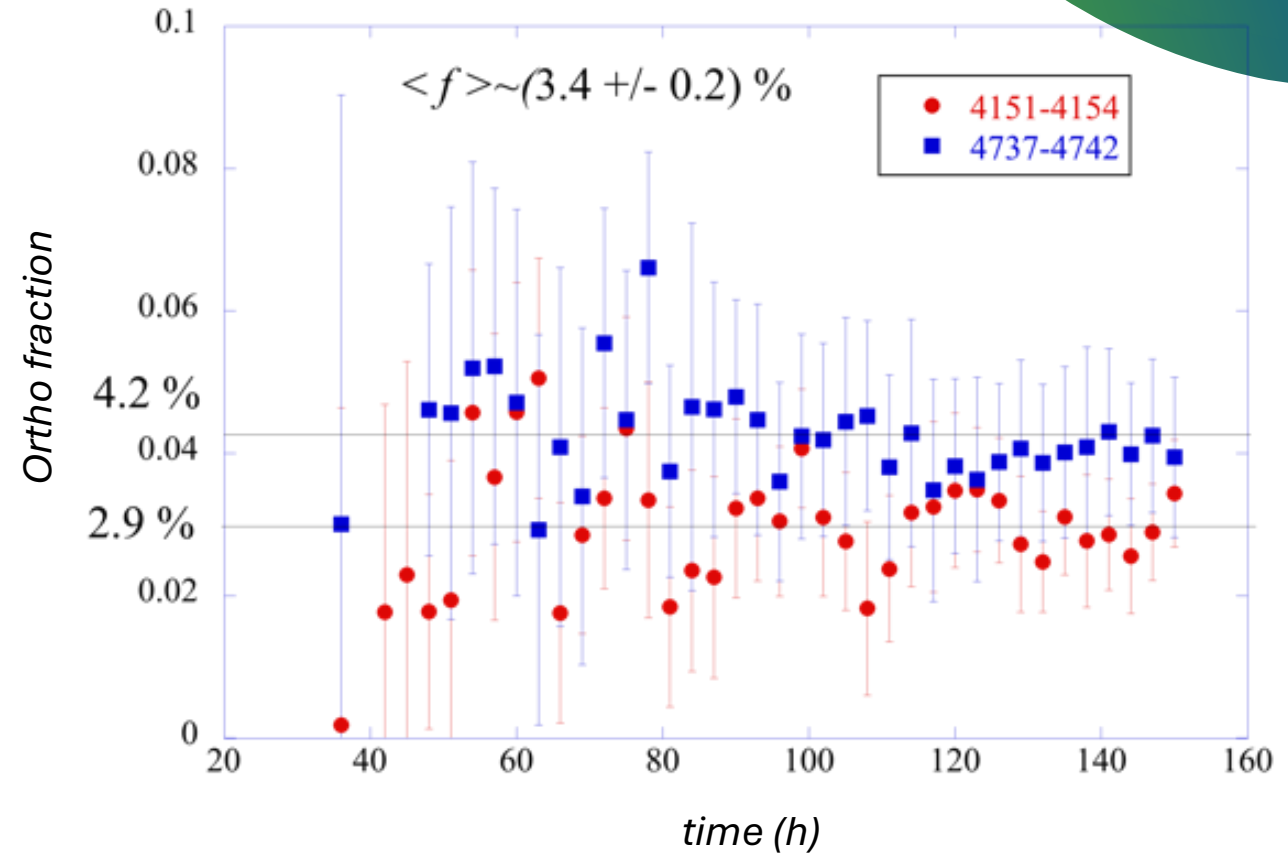
[Fajardo, Applied Spectroscopy 73.12, 1403–1408 (2019)]

Characterization of the cryogenic matrix (II)

Parahydrogen Thickness

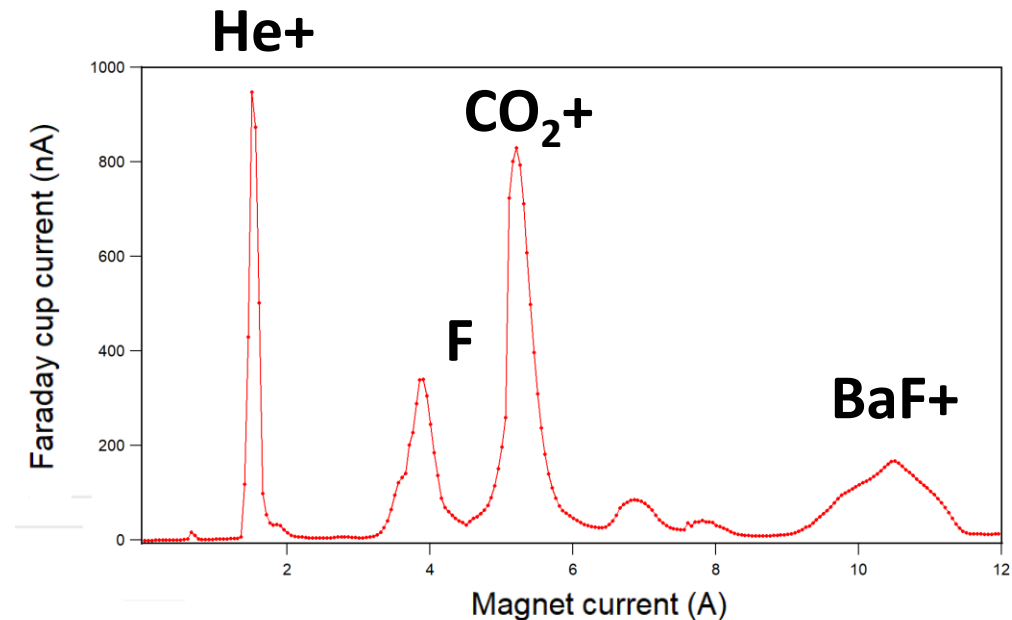
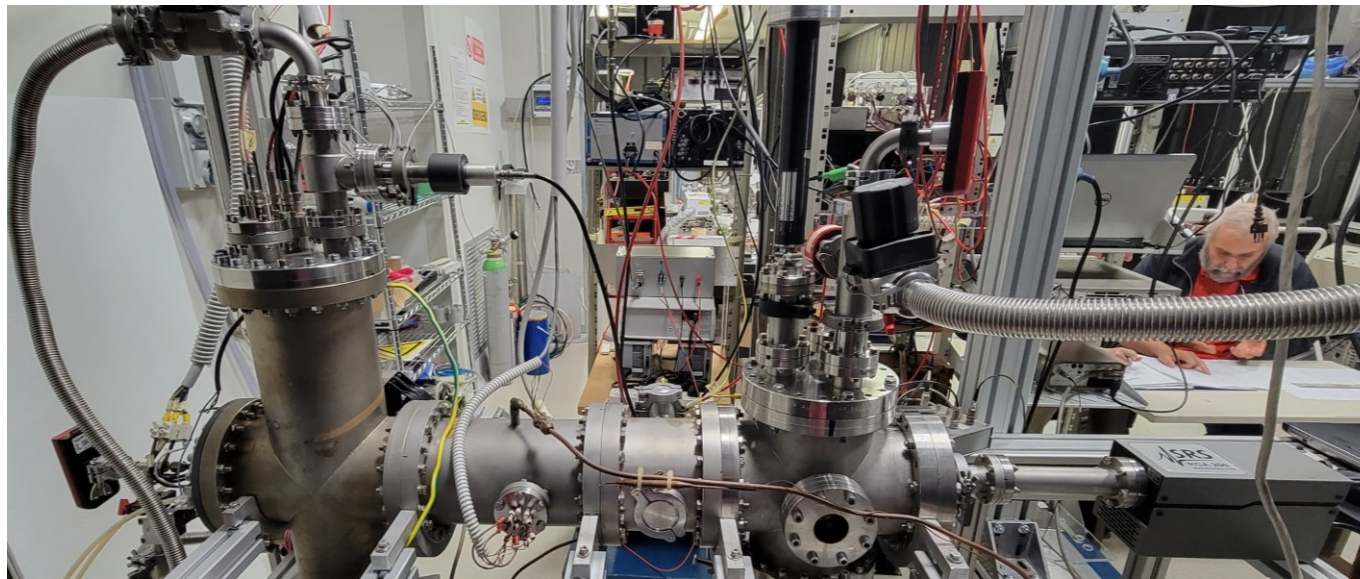
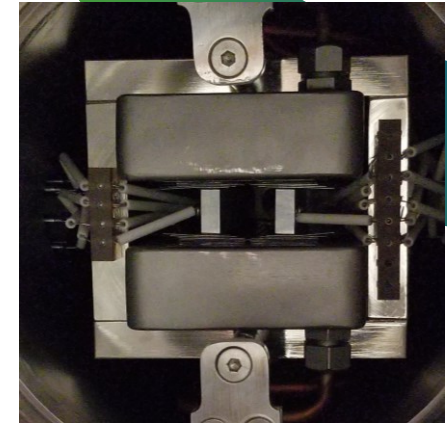
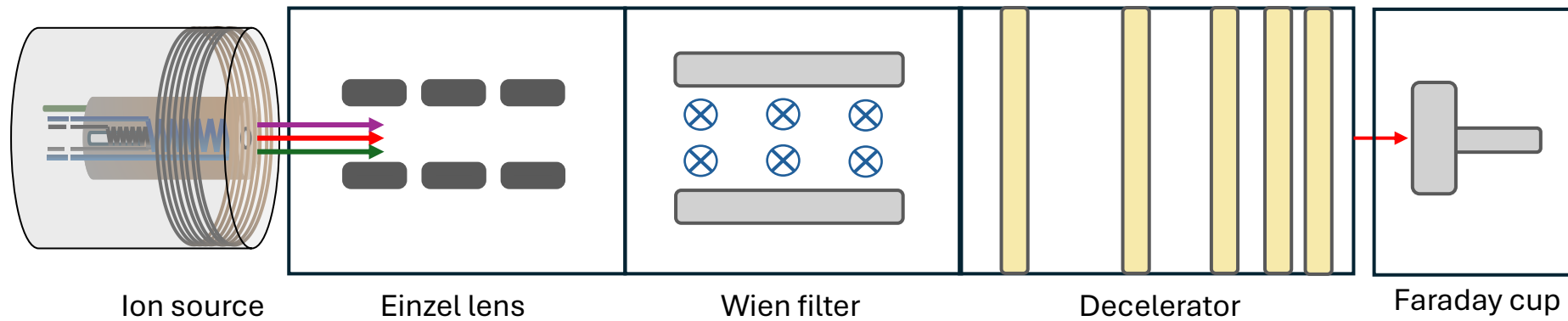


o-H₂ @ 3.5% level



can be reduced to 100 ppm level
if $T_{\text{converter}}$ is lowered to 15 K

BaF production: molecular source

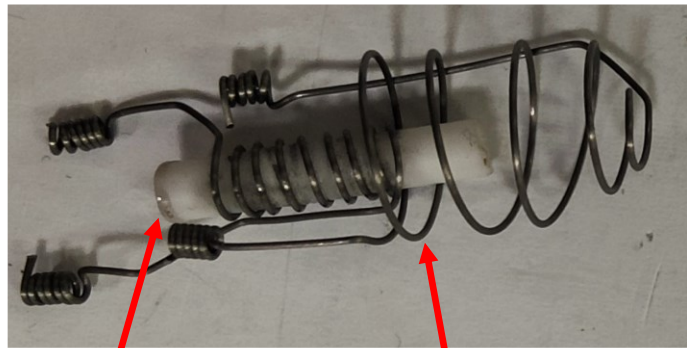


Current status:

- extracted a BaF⁺ beam @ 1 KeV with few μA
- after decelerator 20nA @ 5eV, 10 cm from output (10^{14} molecules / hr)

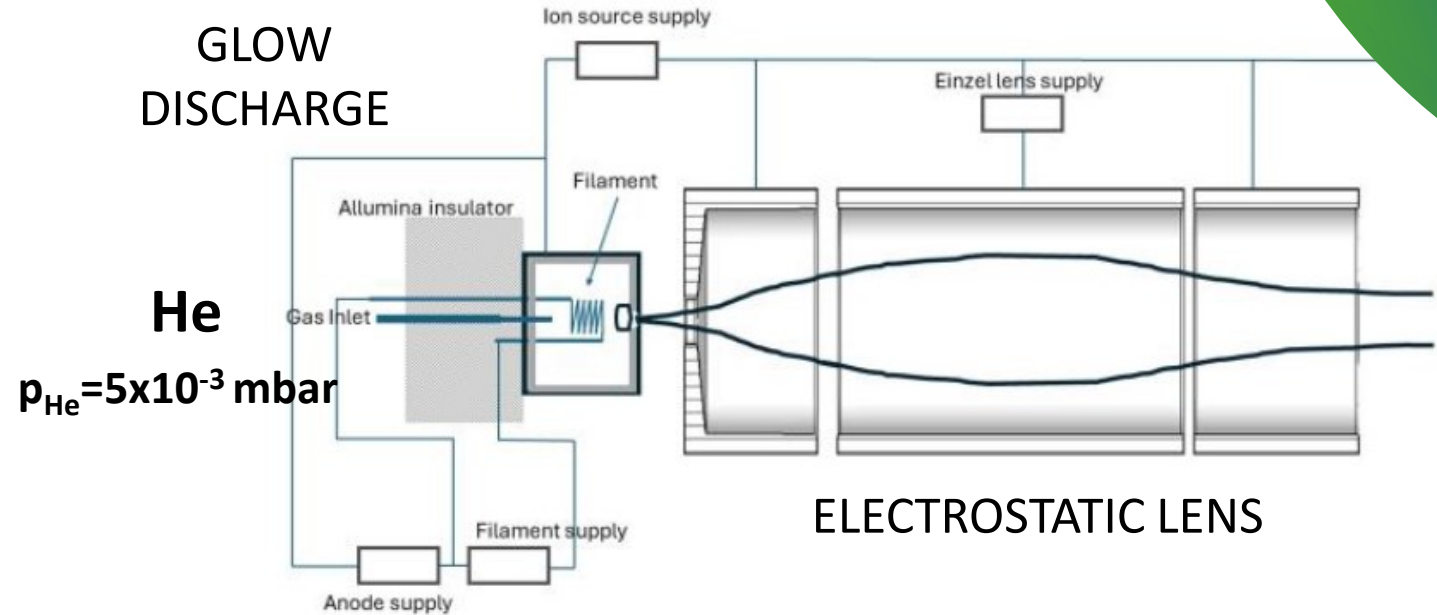
NOT STABLE OVER TIME!

BaF molecular source

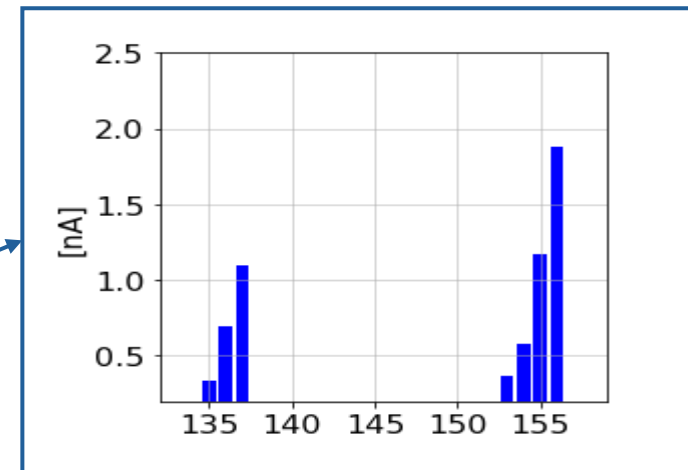
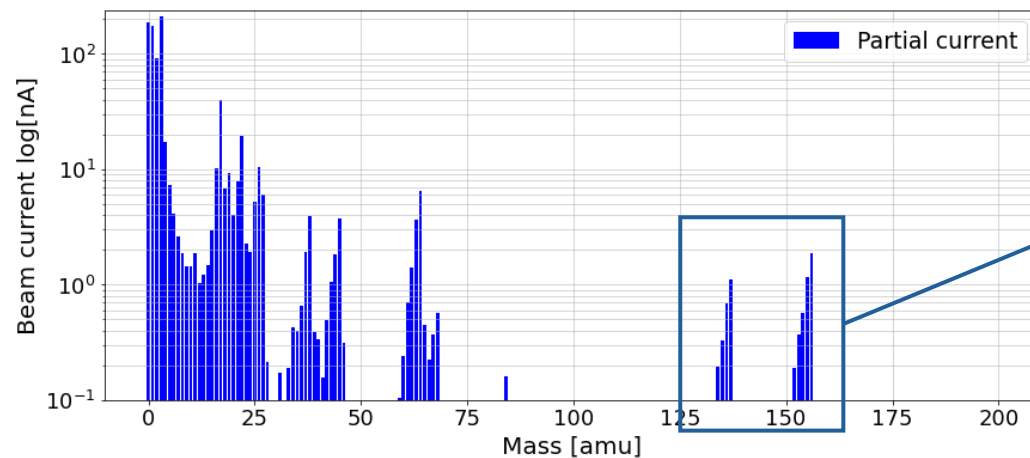


BaF₂ oven

e⁻ filament

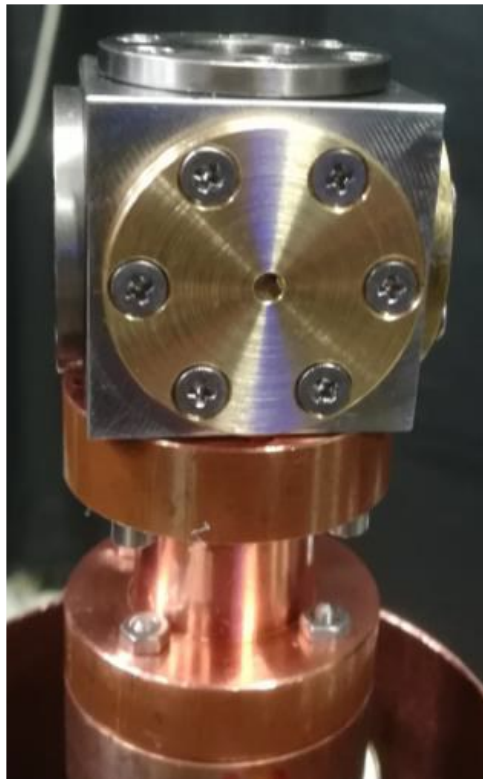


Ba and BaF isotopes beam current (with RGA)



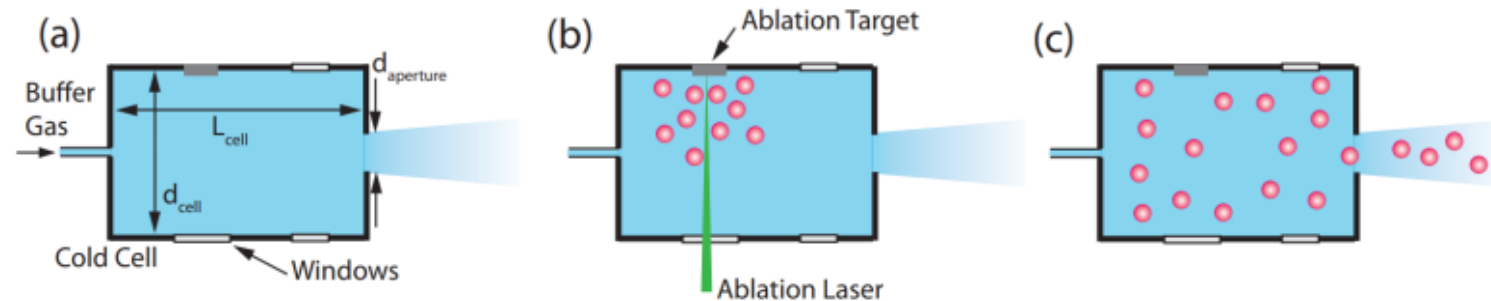
BaF production: laser ablation

Producing BaF molecules with laser ablation and pH_2 as a buffer gas



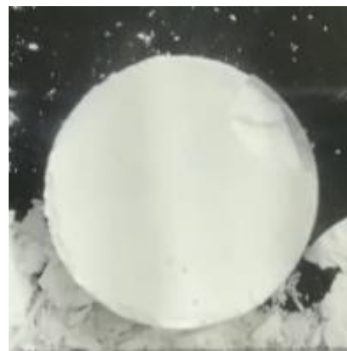
Cell: 25x25x20mm
Output aperture
 $\phi = 2\text{mm}$

Attached to pH_2 converter



[The NL-eEDM collaboration., *Eur. Phys. J. D* 72, 197 (2018)]

Laser for ablation: Q-switch @ 1064nm, mJ energy pulses



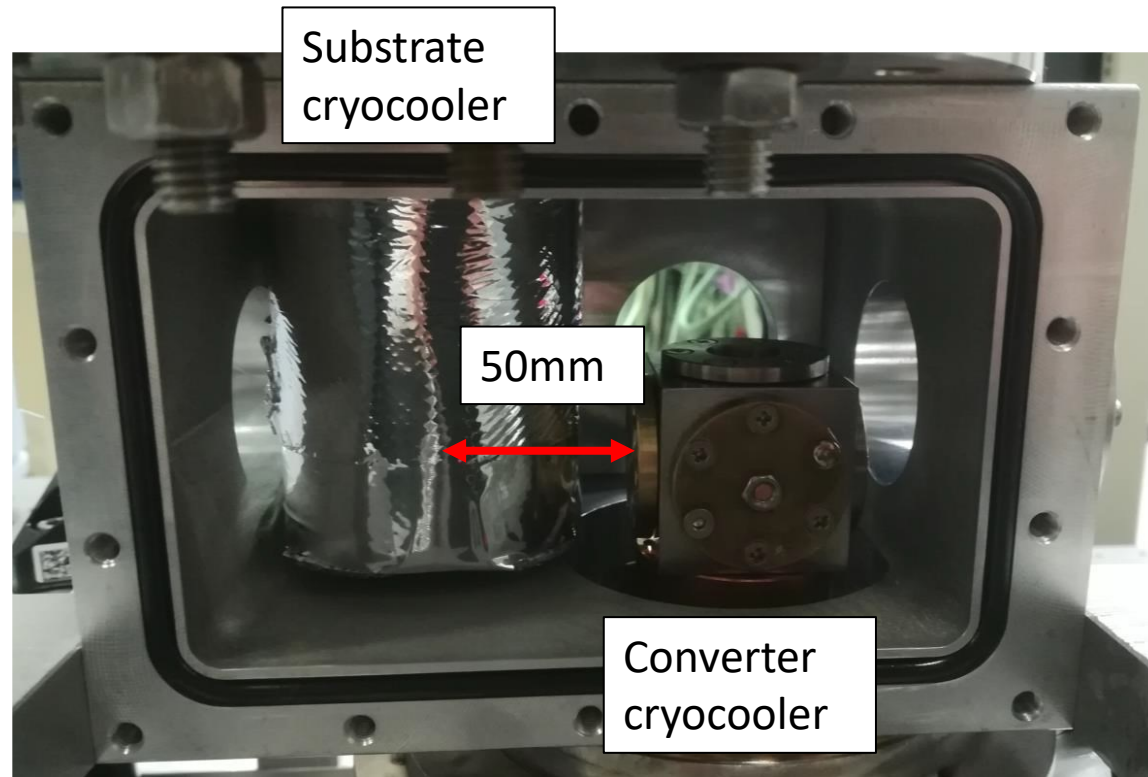
BaF₂ powder pad



optical windows on 2 sides:

- Ablation
- BaF absorption probe (laser light tuned to $X^2\Sigma^+ \rightarrow A^2\Pi^{1/2}$ $\lambda = 859.8\text{ nm}$)

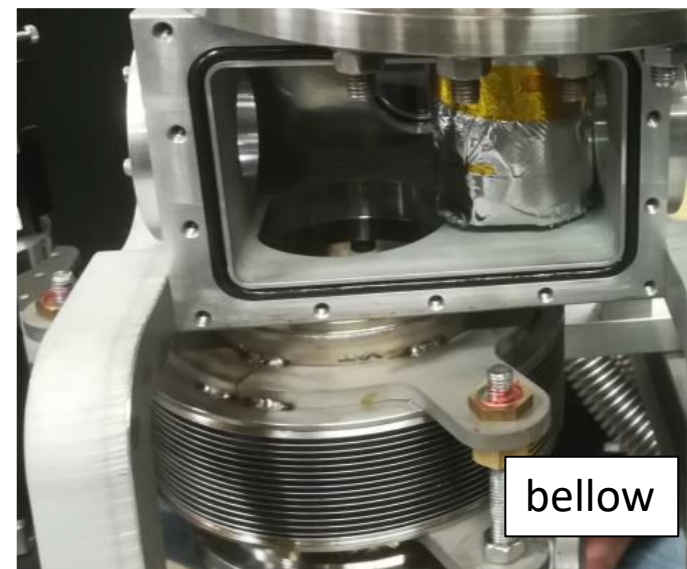
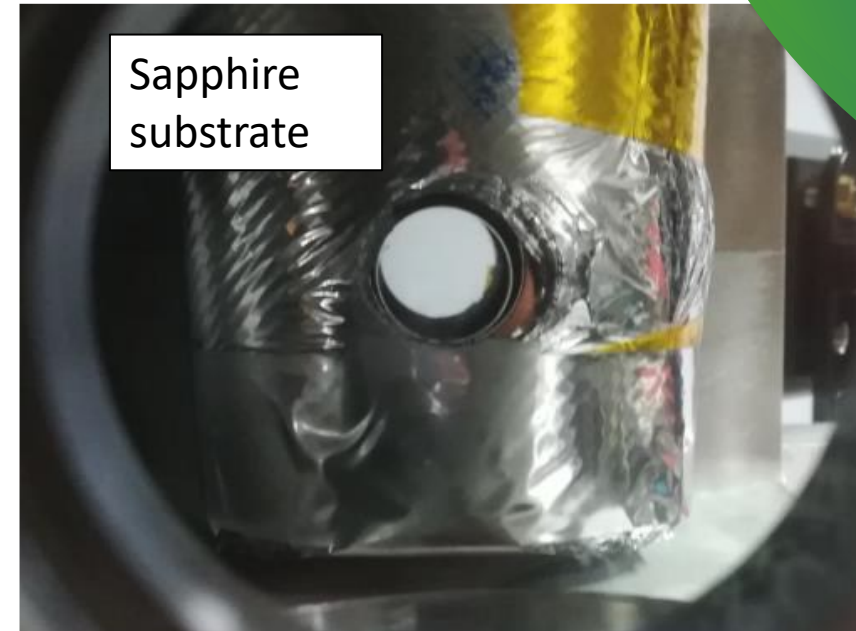
BaF-doped pH2 crystal



Distance cell to sapphire substrate (pH2): 50mm

Grew a doped crystal, but...

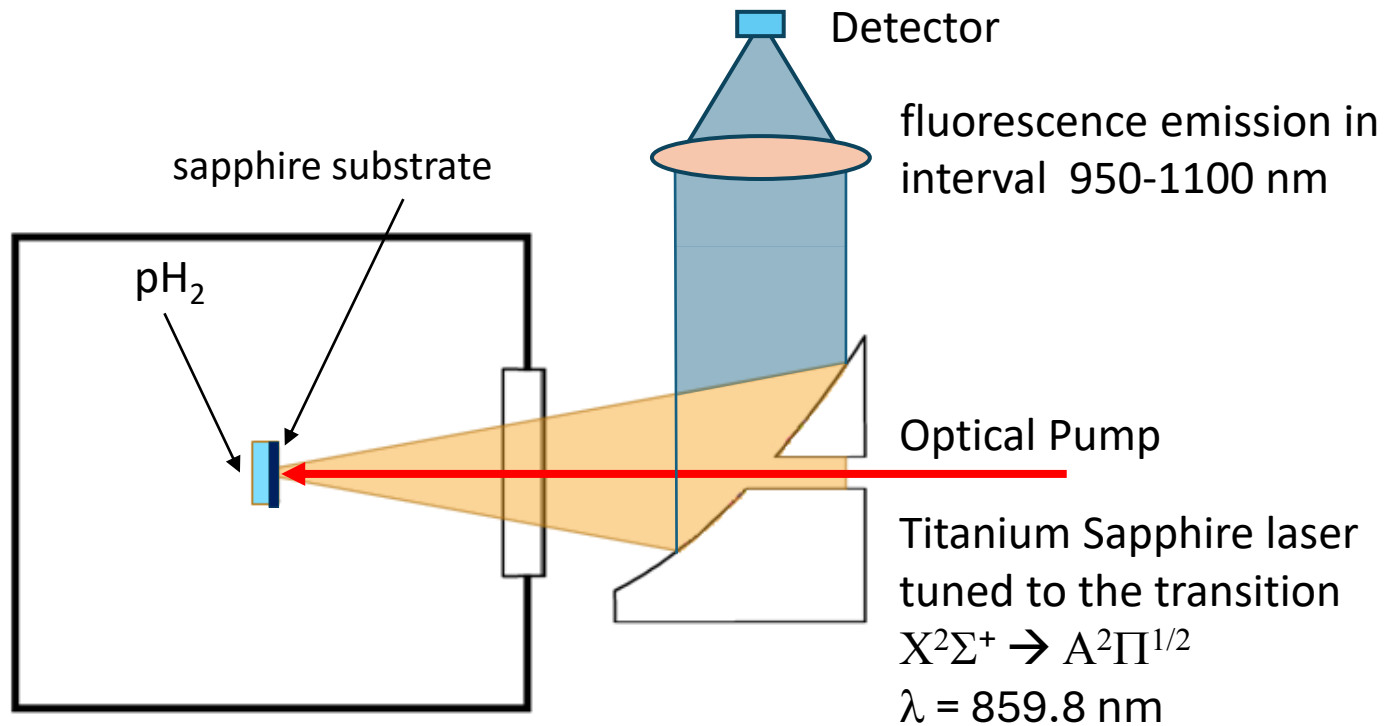
Bad optical quality → strong absorption in all NIR range



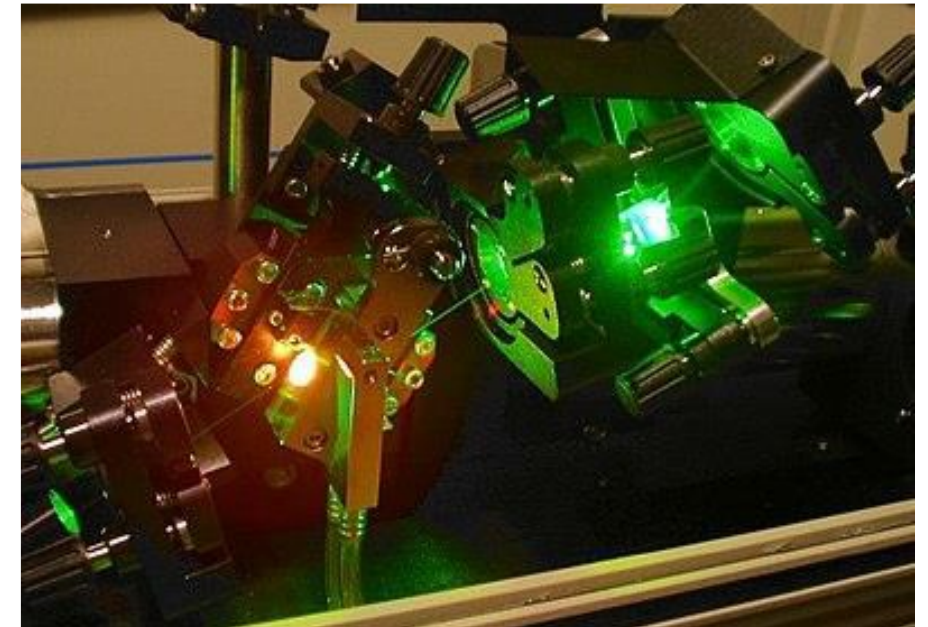
Cell can be moved out of the optical axis with a bellow for spectroscopy and absorption measurements.

Laser Induced Fluorescence (LIF) spectroscopy

- Measure shifts and broadening of lines in solid from gas phase
- Infer quality of BaF doping procedure and quantity of molecules.

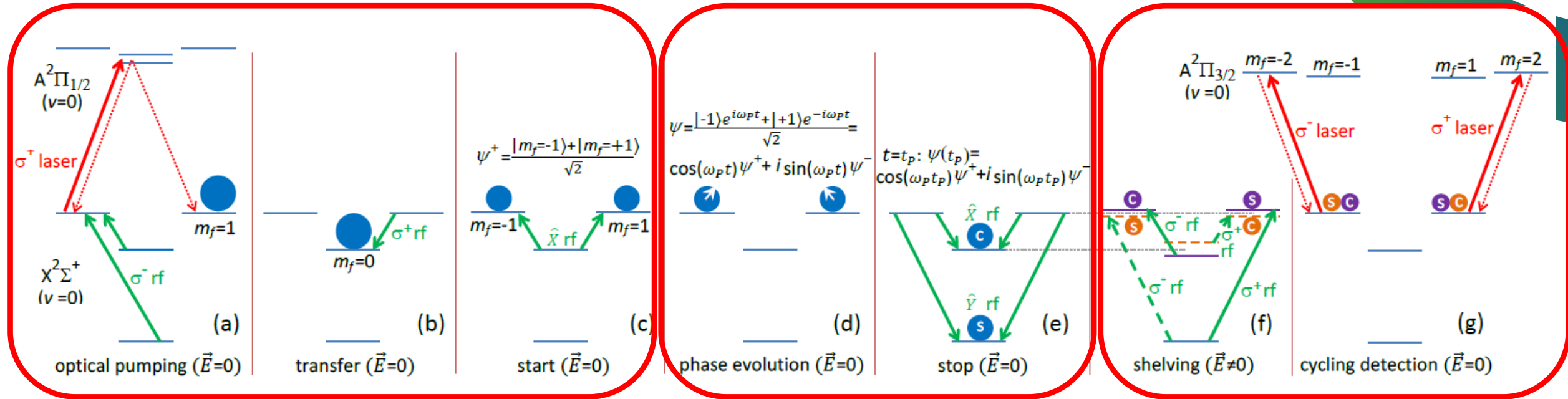


Ti:S laser (720-850 nm, linewidth 10kHz)



Optical detection scheme

[A. Vutha et al., PRA 98, 032513 (2018)]



STATE PREPARATION

EVOLUTION

MEASUREMENT

EDM measurement using laser-induced fluorescence spectroscopy

The ratio of the fluorescence for the σ^- and σ^+ determines:

$$\omega_P = (g\mu_B B_Z \pm d_e \mathcal{E}_{\text{eff}}) / \hbar$$

EPR detection scheme

$$H = -\mu_B \frac{\mathbf{S}}{S} \cdot \mathbf{B}_0 - d_e \frac{\mathbf{S}}{S} \cdot \mathbf{E}_0$$

$$\omega_S = \gamma_e B_0 + \frac{d_e}{\hbar} E_0 = \omega_L + \omega_d$$

$$\Delta\omega_{1/2} = 2\pi \left(\frac{g\mu_B}{h} \right) \Delta H_{1/2} = \gamma \Delta H_{1/2} = \frac{2}{T_2},$$

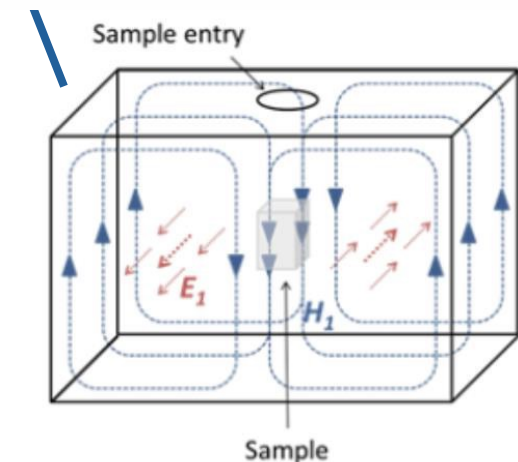
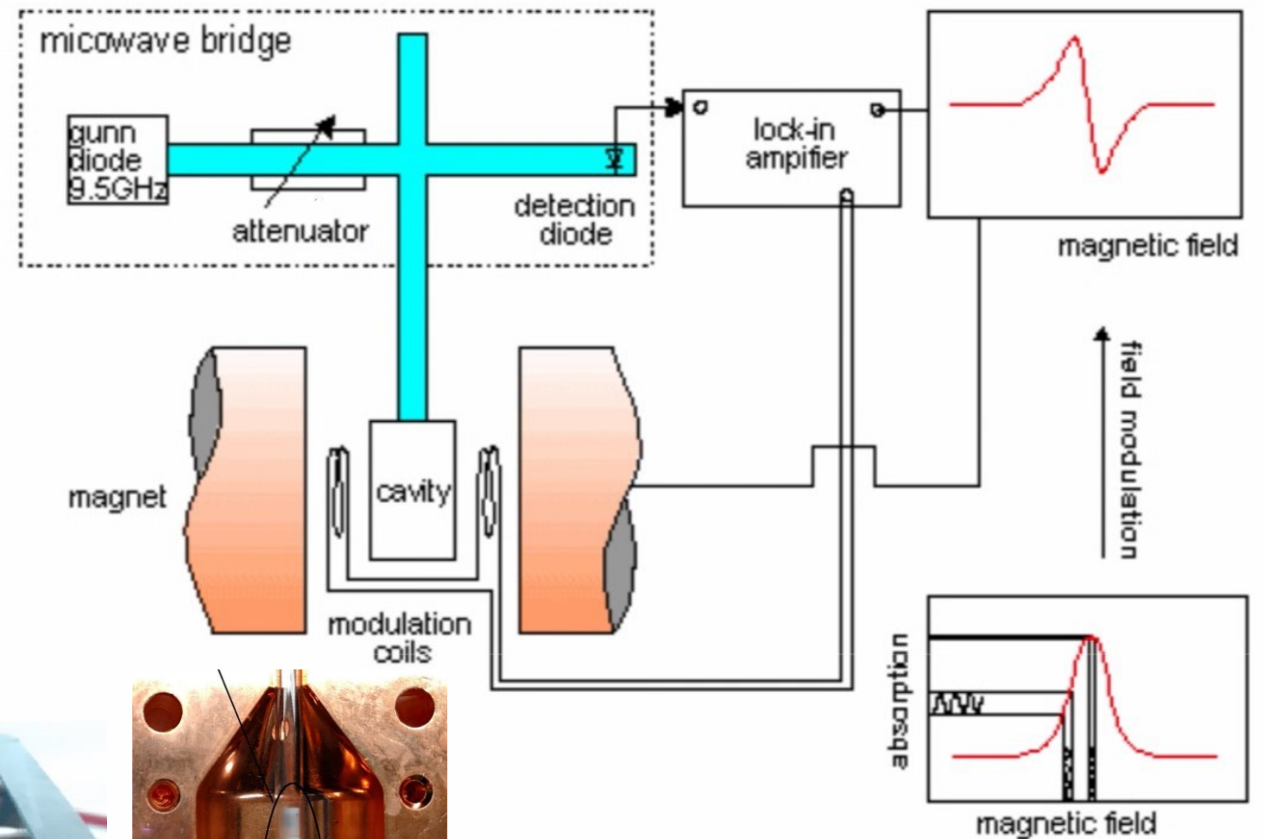
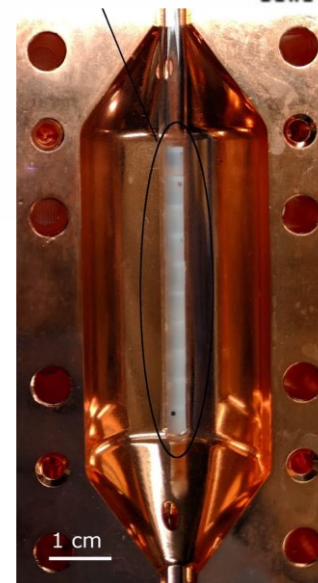
Electron Gyro-
Magnetic Ratio
28 GHz/T

Biassing a microwave cavity
with a high DC Electric Field

$$\nu \approx 10 \text{ GHz}$$

$$E \approx 10^6 \text{ V/m}$$

$$\nu_{\text{mod}} \approx 100 \text{ kHz}$$



Conclusions and future work

1) We have demonstrated:

- Ability to grow $p\text{H}_2$ cryogenic crystal with $t \approx 500$ mm and characterize them with IR spectroscopy
- a BaF molecular source producing 20nA @ 5eV (10^{14} molecules / hr) with isotopic selection

2) We started integrating in $p\text{H}_2$ BaF molecules from a laser ablation source

FUTURE WORK:

- New laser ablation cell in setup to grow doped crystals
- Laser Induced Fluorescence spectroscopy studies
- BaF molecular source: improve stability and output current to ≈ 100 nA
- Increase purity of $p\text{H}_2$ crystals f_{ortho} to 100 ppm with $T_{converter} \approx 15$ K
- EPR setup and measurements of paramagnetic samples