The PHYDES activity: BaF in para-hydrogen for EDM studies

The PHYDES R&D project

Marco Guarise*
on behalf of PHYDES collaboration

*University of Ferrara and INFN

EDMs: complementary experiments and theory connections

Outlook

Contents:

- **PHYDES R&D project**
  - Approach
  - Main features
  - Milestones
- **Para-hydrogen**
  - Properties
  - Growing
  - Recent results
- **BaF molecules**
  - Properties
  - Production
  - Recent results
- **Future steps**
- **Conclusions**

PHYDES para-hydrogen and diatomic molecules for EDM studies

Who: National PI of the project G. Carugno

Where: 4 INFN groups in Italy:

- Padova
- Laboratori Nazionali Legnaro (main setup)
- Ferrara
- Laboratori Nazionali del Sud

When: 2022-2024 + new R&D 2025-2028

Idea: PHYDES is **R&D project** (CSN5 INFN) whose main goal is to study the feasibility of the approach based on para-hydrogen crystal doped with BaF molecules in different configuration of detection (magnetization or optical scheme) in the electron EDM study.
Our aim is to study in few years the possible showstoppers related to the frontiers experimental technologies needed in these schemes.
We benefits from the **expertise gained in previous R&D for axion detection in cold crystals (cryogenic crystals of Rare gas)** [AXIOMA & DEMIURGOS R&D projects]

Wrt other approaches:

- Possible pro: high number
- Possible cons: short coherence time

→ **To be measured**

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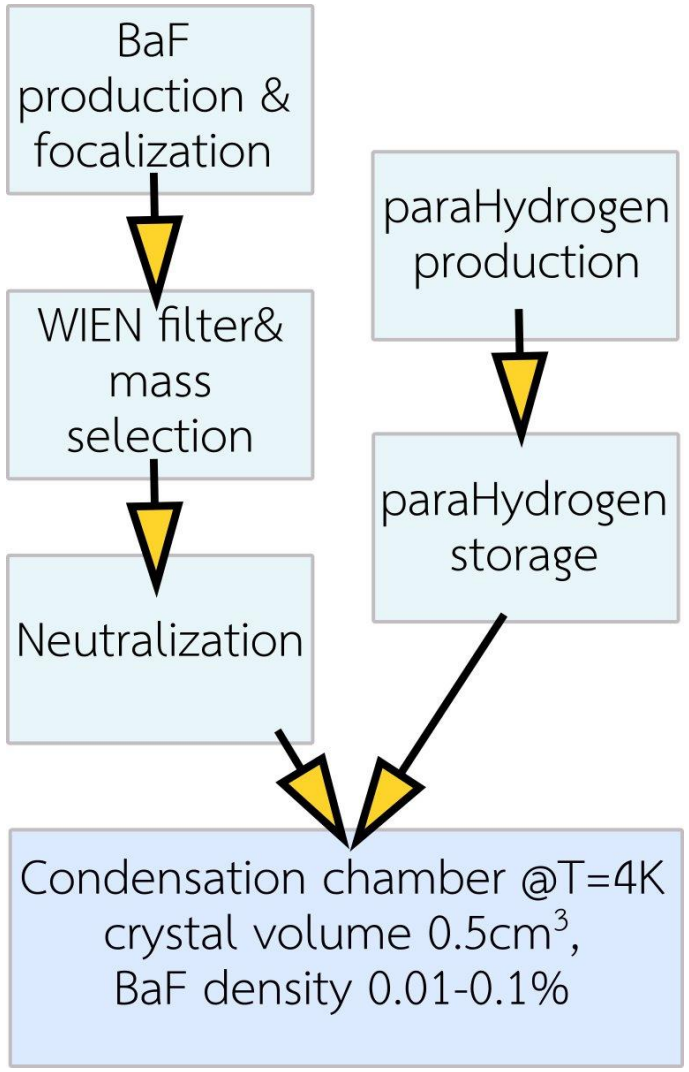
PHYDES Experimental scheme



Implemantation in **5 different subchambers**:

- BaF **production, acceleration and focusing**;
  This is needed to obtain a high numbers of molecules
- **Isotopical selection**;
  Isolate only the good isotope
- **Neutralization and cooling**
  Create neutral BaF ready for doping
- **Para-hydrogen production** and storage
  Production in parallel of the host atoms for the matrix
- **Condensation** chamber
  Final grow of the doped crystal

We decided to proceed doing each step separately in order to understand the best way to move forward, improvements of the receipt and possible drawbacks of each part

PHYDES Detection protocols

We are applying an R&D approach also to detection protocols in order to understand the pros/cons of different schemes in terms of S/N, and unveil possible showstoppers from the experimental point of view.

Different detection approaches:

- Magnetization detection
- EPR detection
- Optical detection

Magnetization approach:

d_e aligns with E, spin also align with E, so net electronic spin polarization that generates a bulk magnetization (very feeble but lots of electrons)

Using superconducting quantum interference device (SQUID), one can detect this EDM-induced magnetic field

$$B^{\mathrm{CP}} = \mu_0 \gamma\, n\, \langle \mu_{\mathrm{mol}}^{\mathrm{CP}} \rangle \qquad \langle \mu_{\mathrm{mol}}^{\mathrm{CP}} \rangle \approx \mu_B \frac{d_e E_{\mathrm{eff}}}{k_B T} \times \langle n_z \rangle$$

n= molecular number density

$< n_z >$ = average projection of the molecular axis onto the E-field

Optical detection of the phase shift when E is reversed

Probe the relative population of the states after B, E application and detect phase differences between the measurements with E fields in opposite orientation.

This needs state preparation which is similar to the approaches seen in other talks (M. Tarbutt,...)

EPR approach:

Using EPR/ESR technique to probe frequency shifts when E is applied

PHYDES Details

In the next slides we will see experimental details of each part of the PHYDES R&D program:

- Cryogenic crystals: growing techniques & main properties;
- Para Hydrogen;
- BaF;
- Neutralization chamber;
- BaF line;

Cryogenic crystals

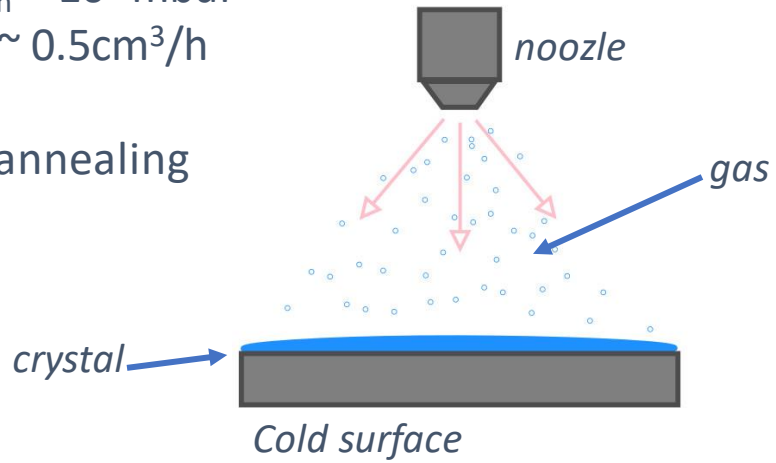
Overview

Solid crystals made of inert and unreactive material can be grown at cryogenic temperatures

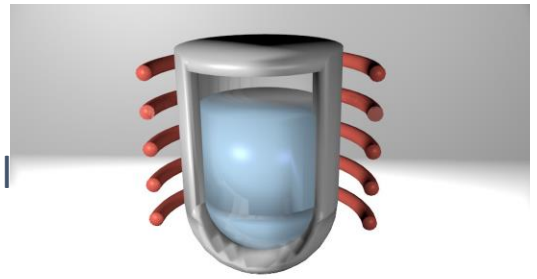
Application in Fundamental physics researches (light components of DM candidates, neutrino coherent scattering, EDM study), medical (MRI/NMR) with hyperpolarized Xe, chemical spectroscopy

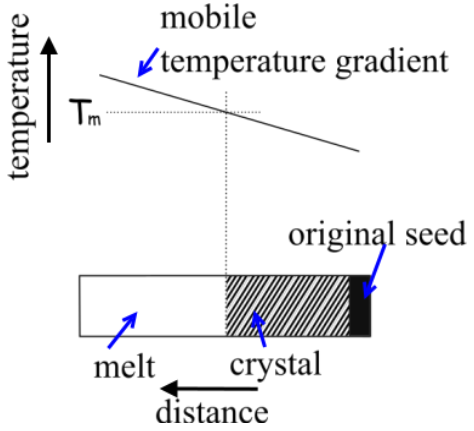
Growing techniques:

- **Vapour deposition technique**
  - Spray deposition through noozle
  - Cold surface
  - P_{growth} ~ 10^{-5}mbar
  - Rate ~ 0.5cm^3/h
  - Films
  - Easy annealing



- **Modified Bridgeman Stockbargher technique**
  - Growth from liquid
  - Low temperature container
  - Fine tuning temperature control
  - Rate ~ 2cm^3/h
  - Large solids





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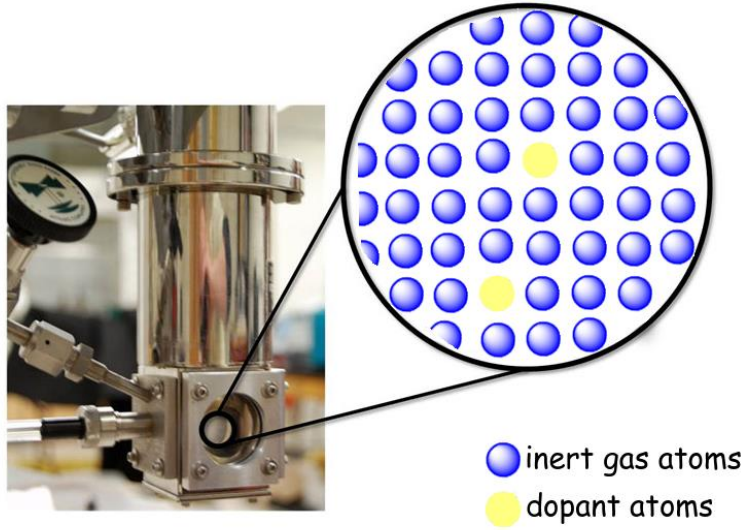
Doped cryogenic crystals

Matrix Isolation Spectroscopy (MIS): G. Pimentel in the 50's → study free radicals, unstable and transient species embedded into inert gas matrices

- **low interacting environment** → low line broadening;
  - feeble interaction host host;
  - feeble interaction host guest;
- the species can be **accumulated** in the matrix over many minutes → high density;
- **large interstitial space** → good doping;
- possible issues: clustering; line broadening; impurity; interstitial/substitutional sites;



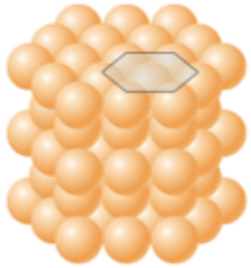
NB: Rare gases crystal generally can tolerate an impurity concentration in the order of 0.1% without generating much problems to the lattice. If too many impurities are present, they can coalesce to form clusters and destroy the crystal structure.

I. Gerhardt, et al. J. Chem. Phys. 137, 014507 (2012);
S. Upadhyay, et al. Phys. Rev. Lett. 117, 175301 (2016)

p-H_2

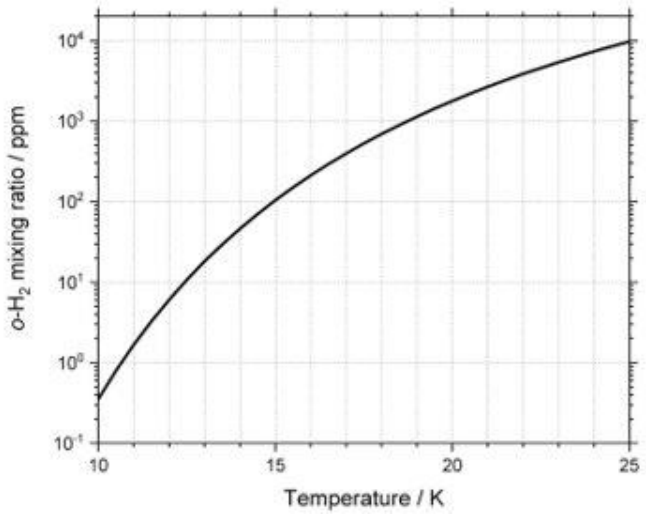
Properties

Molecular H_2 is classified into two nuclear-spin isomers, ortho (parallel) and para (antiparallel), according to the total nuclear spin. Para-hydrogen is the lower-energy state.



When solid p-H_2 is prepared by direct deposition, the solid might have mixed hcp-ccp structures, but annealing at ~5 K converts the ccp structure to the more stable hcp structure.

- Lattice phonon energy for p-H_2 ~100 cm^{-1}
- p-H_2 has no quadrupole moment



The amplitude of zero-point motion relative to the distance to the nearest neighbor for solid p-H_2 is ~18% of the lattice constant (for solid Ar is only 5%). This is a measure of the localization of particles in their equilibrium positions in a crystal: the softness.

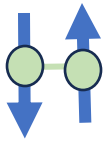
p-H_2 is a quantum solid!

- Softness: molecules are almost free from the cage effect
- Softness: imperfection of the crystal structure around the dopant molecules are very small.
- Relaxation of excited molecules in solid p-H_2 is extraordinarily slow: small line broadening

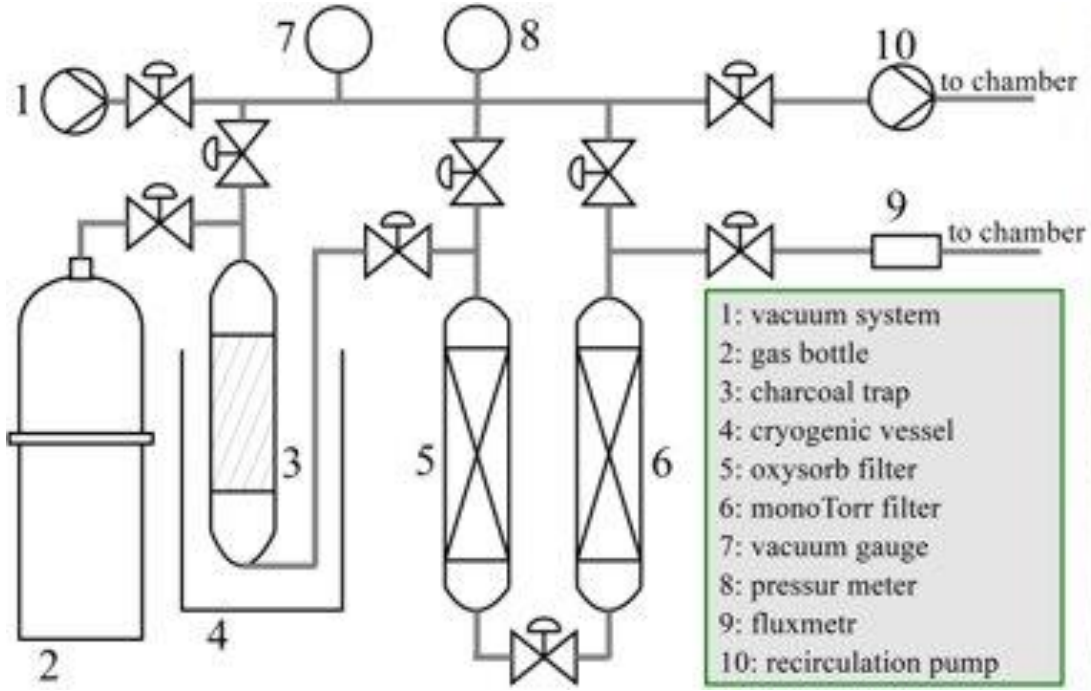
NB Weinstein group: spin coherence times as long as 0.1 s for an ensemble of rubidium atoms trapped in a solid parahydrogen matrix! [PRL 125, 043601 (2020)]

p-H_2

Purification and enrichment chamber

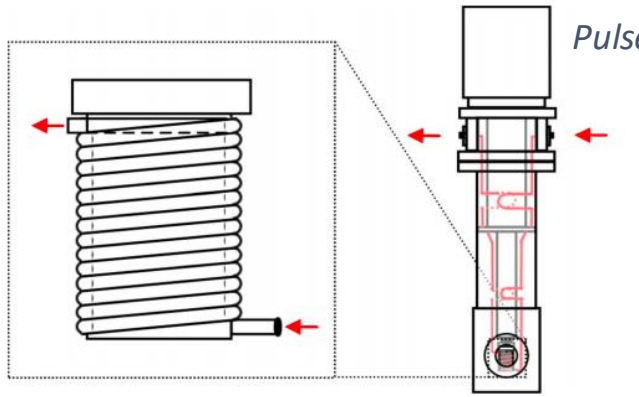


Para-hydrogen: anti-parallel proton spin



- **Hydrogen purification system:**
  - Cryo charcoal trap;
  - MonoTorr selective filter;
- **Para-hydrogen enrichment system:**
  - Pulse-tube refrigerator
  - Coil tube of oxygen free, high-conductivity copper
  - Hydrous ferric oxide catalyst

Generation of highly enriched p-H_2 at rates of up to 0.5 liters per minute (SLM)

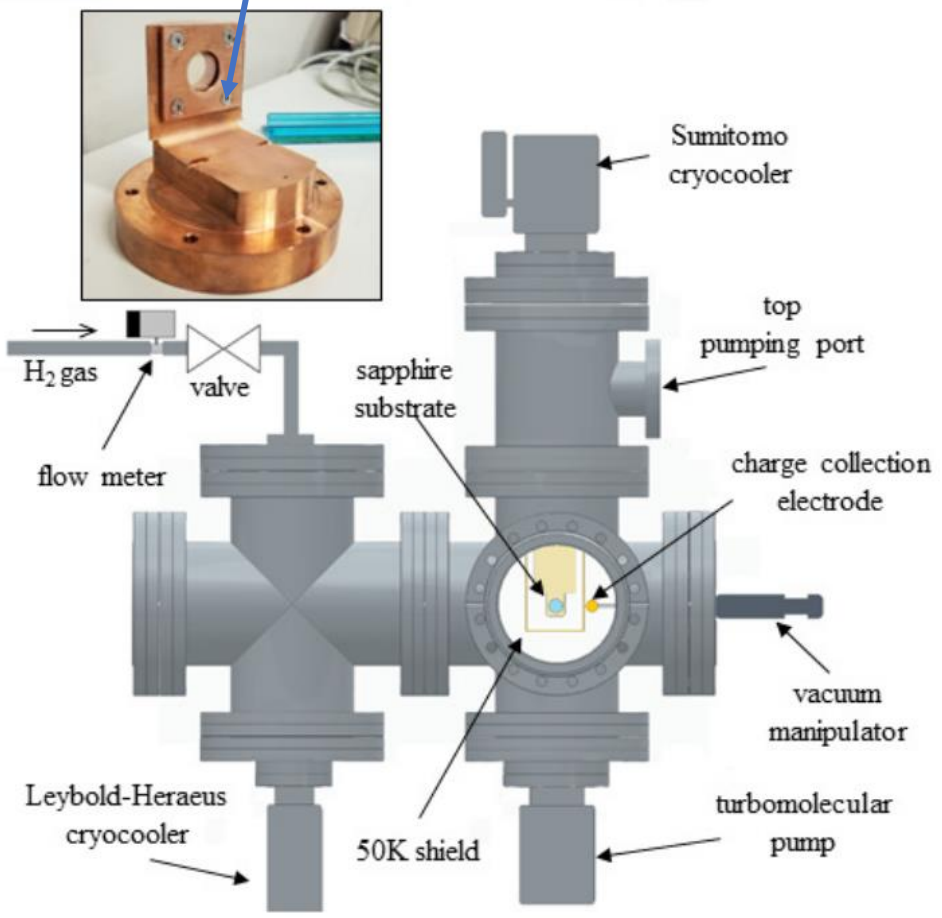


Pulse-tube

Coil tube filled with catalyst

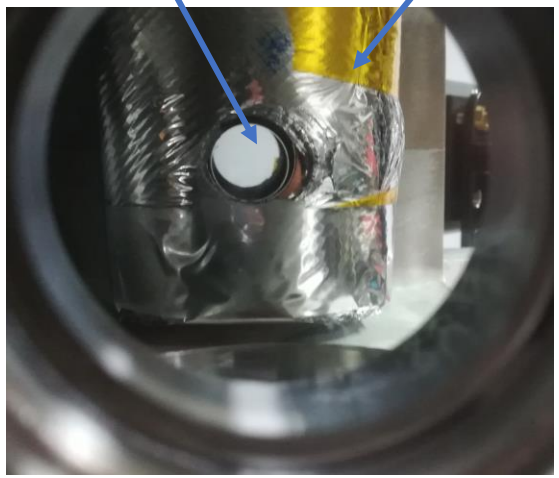
P-H_2 Crystal growth

Cold finger unmounted from the pulse tube



Mylar foils covering the cold finger

Sapphire window



Front view

Later view

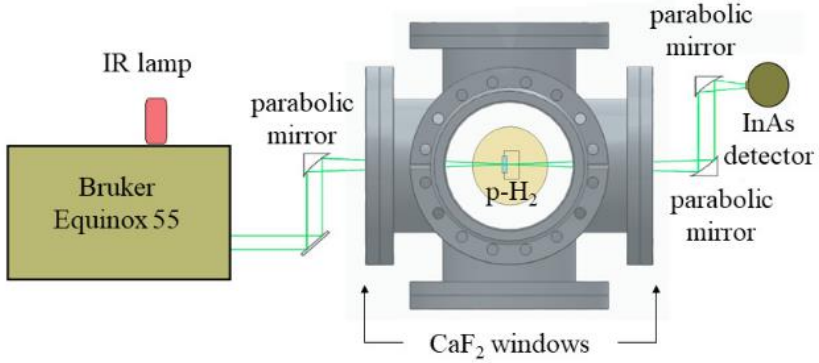
Features:

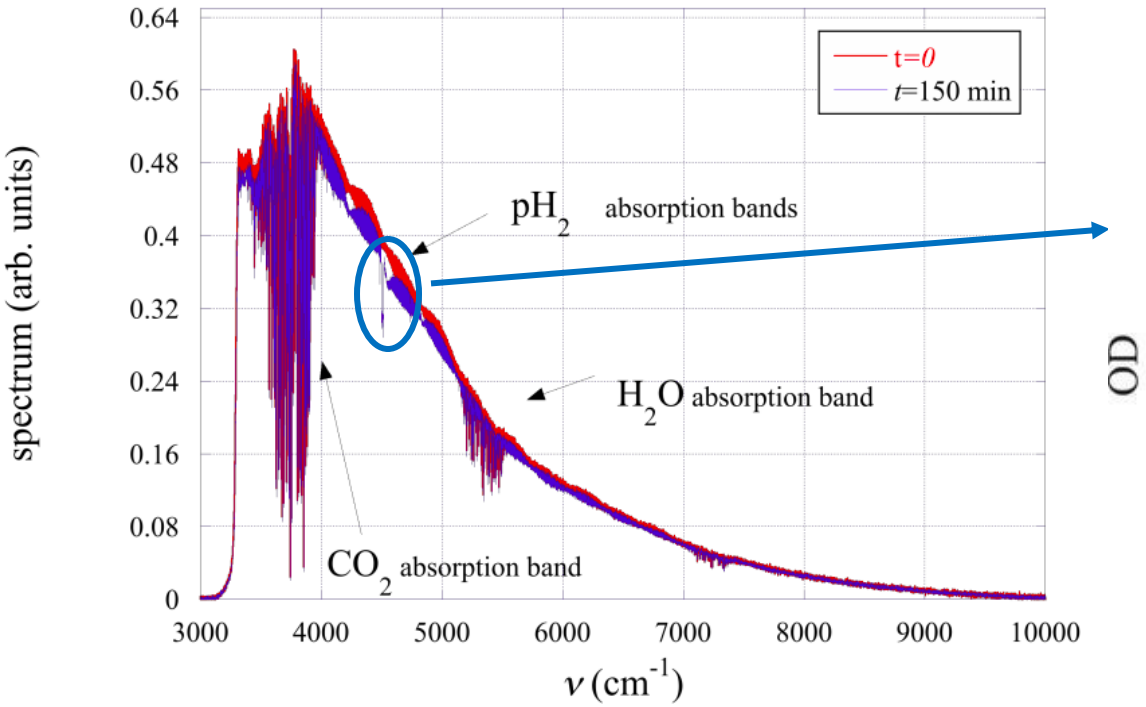
- OFHC copper cold finger
- Sapphire substrate 1"
- Precise thermal control <0.2°
- Flux 0.5 cm^3/min
- 3 mm diameter gas nozzle
- Spray from 30 mm
- Clear aperture ≈ 15 mm

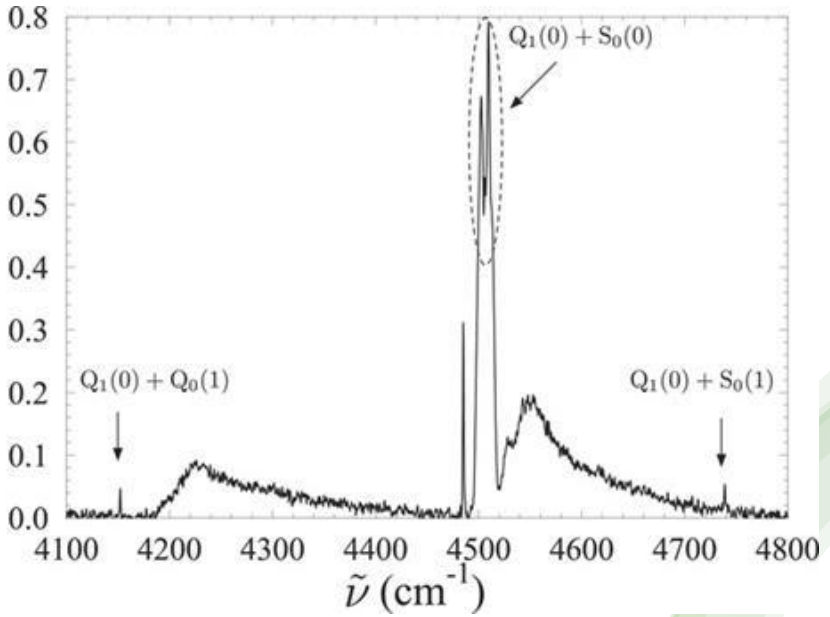
p-H_2 Characterization

Quantities:

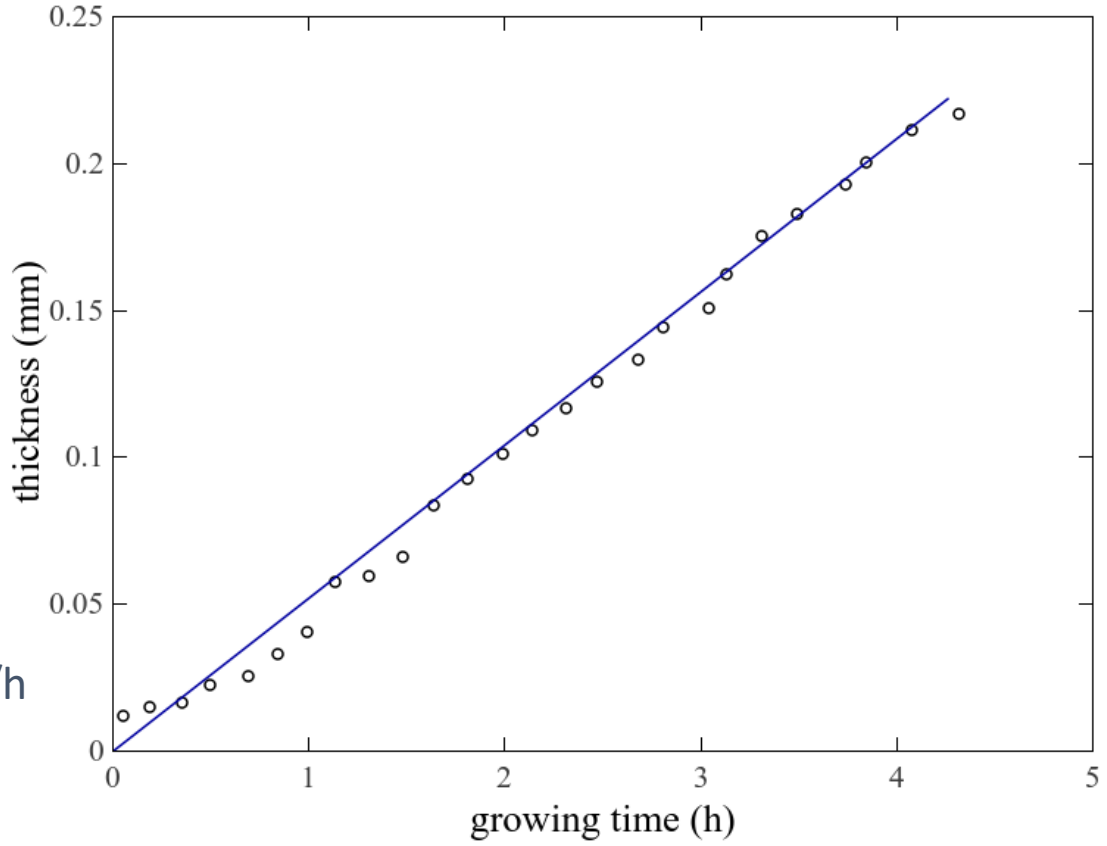
- Spectrum: I(v)
- Reference spectrum: $I_0(v)$
- Transmission spectrum: T→ $I(v)/I_0(v)$
- Optical density: OD=-log T

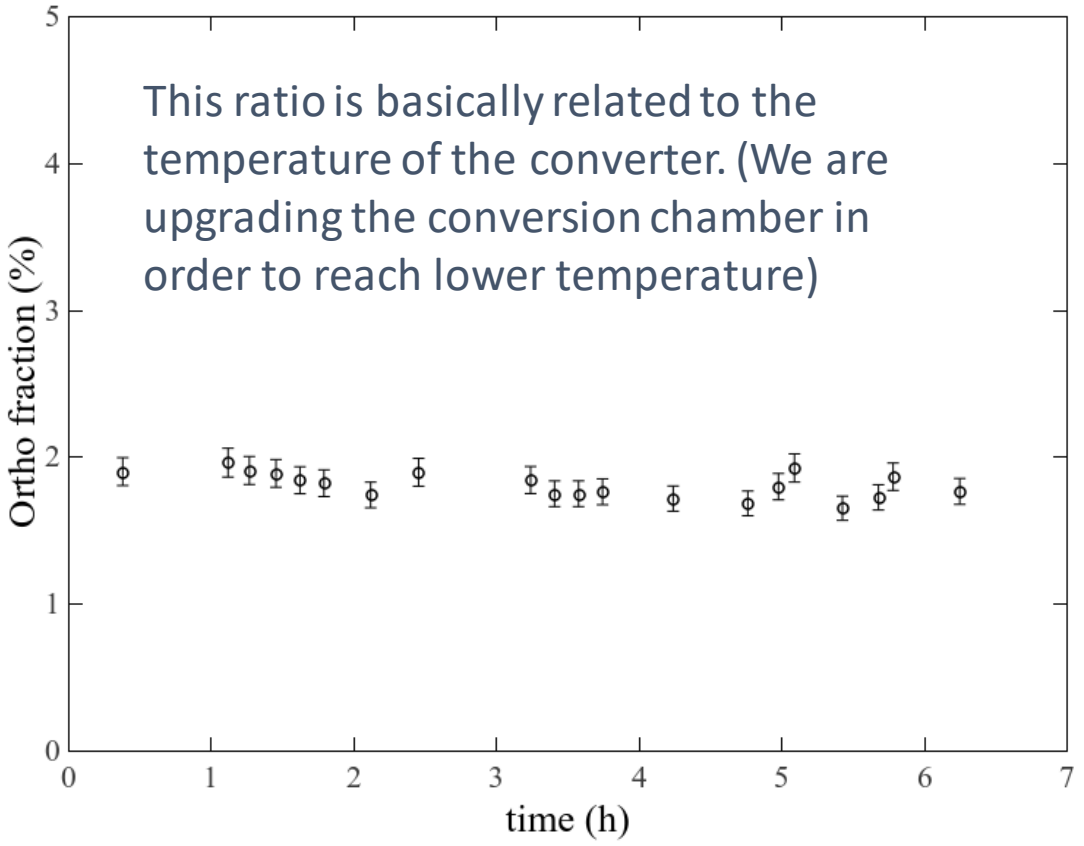






p-H_2 Characterization





This ratio is basically related to the temperature of the converter. (We are upgrading the conversion chamber in order to reach lower temperature)

Slope = 52um/h

Film thickness: $t = 4.8 \times 10^{-2} \int_{4495}^{4525} OD \, d\tilde{\nu}$

In 2.5h of growing → thickness of 0.2mm

The ortho ratio is computed used lines at 4150cm^{-1} and 4750cm^{-1}

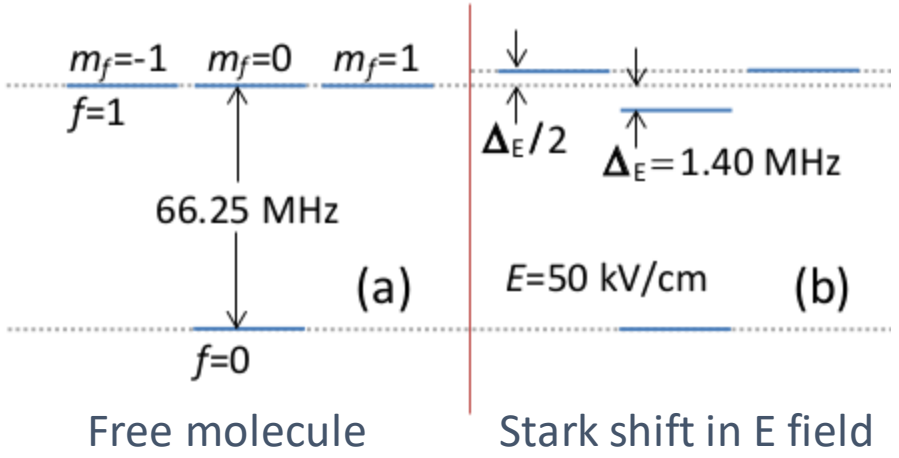
$$f = \frac{0.124}{h} \times \int_{4151}^{4154} \mathrm{OD}(\tilde{\nu}) \, d\tilde{\nu}$$

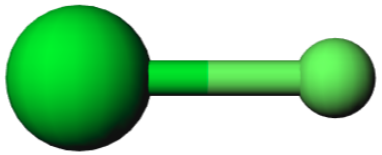
$$f = \frac{0.0787}{h} \times \int_{4732}^{4742} \mathrm{OD}(\tilde{\nu}) \, d\tilde{\nu}$$

BaF

Properties

Ground rotational state $X^2\Sigma^+$ (n = 0)





	BaF
E_{eff}(GV/cm)	8

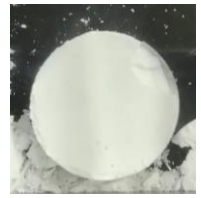
Transition $X^2\Sigma^+ \rightarrow A^2\pi_{1/2}$ in the NIR @ 859nm

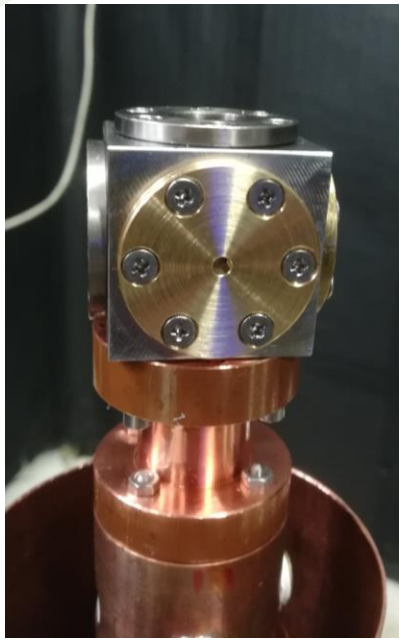
We foresee to perform lot of spectroscopic measurements both in "vacuum" environment and in solid to understand the line shifts and broadening that can help us to infer how much interacting is the environment.
These measurements are also important to understand the quality of BaF during doping, the quality of the doping procedure and the quantity of molecules involved in the process.

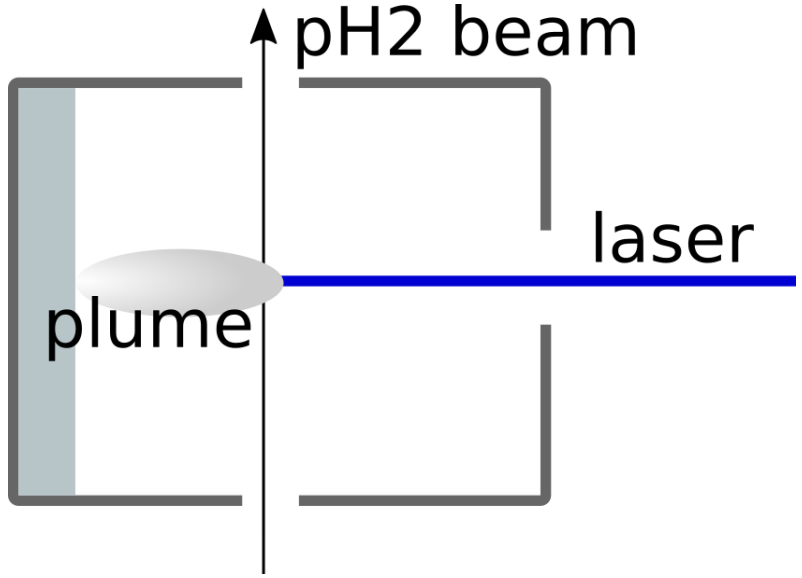
BaF

Production

In order to try to embed BaF molecules in pH2, we initially would like to use an easier approach: we produce BaF close to the condensation chamber via laser ablation and we mixed with pH2 during the growth.







Features:

- Laser source: Q-switch 1064nm
- BaF powder pad (sintering process from high purity powders)
- Chamber: 25x25x20mm
- Exit hole: 2mm diameter
- Distance to pH2 chamber: 50mm
- Possibility to perform optical transmission during ablation: optical windows
- Test BaF quality
- Test of stability during growth

Furthermore laser ablation can be pointed on different places in order to guarantee homogeneity and plume formation

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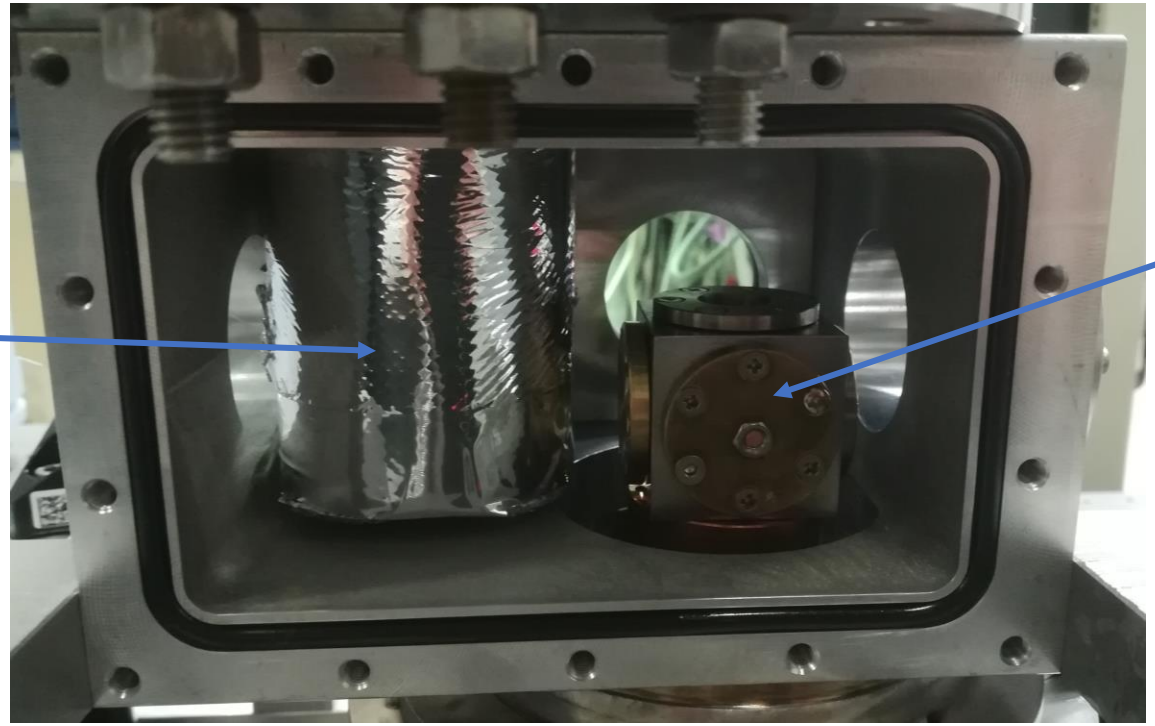
P-H_2 doped BaF

Crystal growth

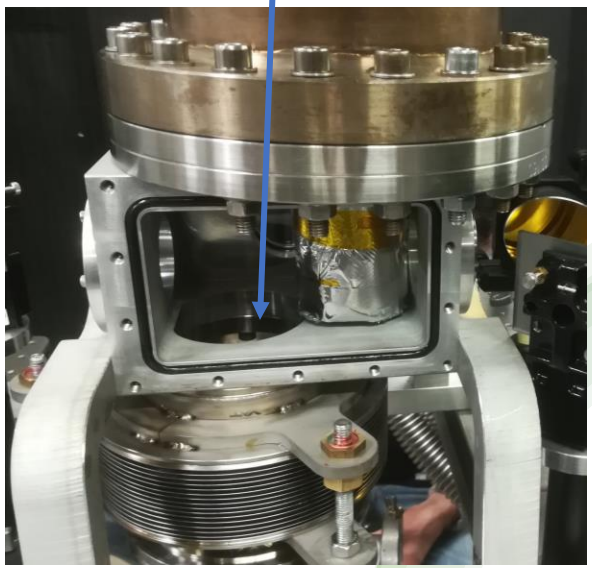
We are using a unique complex vacuum chamber for pH2 mixing with BaF. The two parts (mixing and deposition) are attached to 2 different cryocoolers. Mixing chamber can be moved up or down in the optical axis in order to perform spectroscopy

Deposition Sapphire window



BaF & p-H2 mixing chamber

- On axis
- Off axis (mounted on bellows seals)



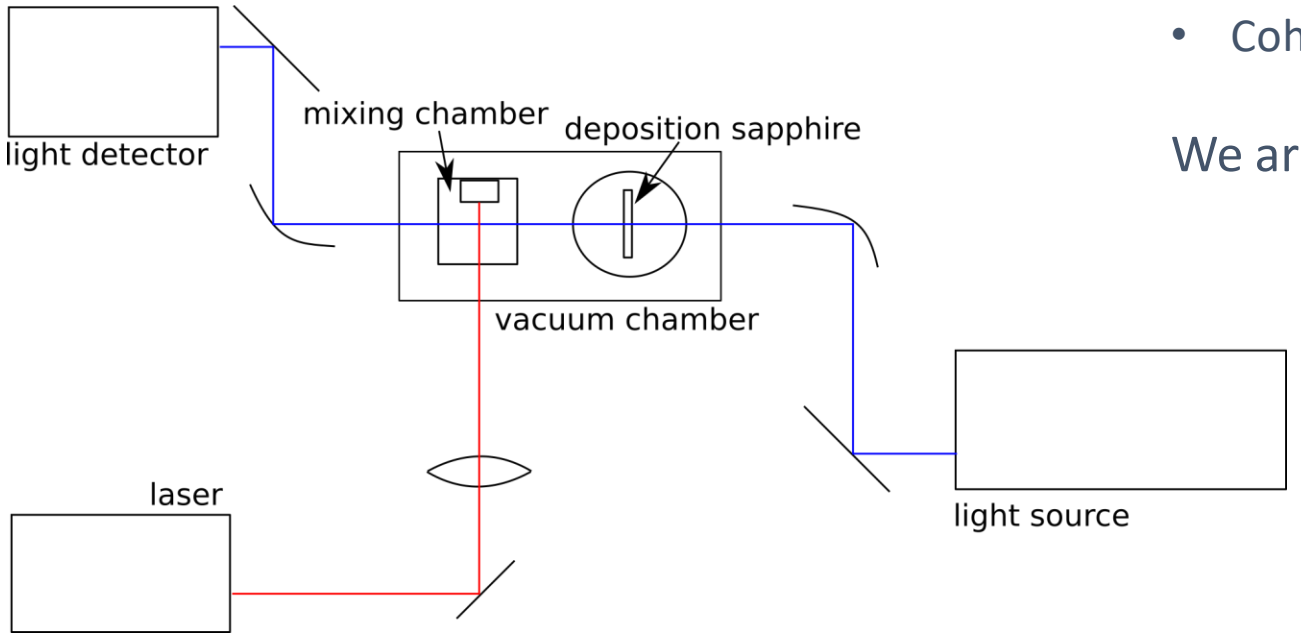
NB We will add a Helmotz and RF coils around the chamber

P-H_2 doped BaF

Procedure & tests

Receipt:

- Start pH_2 production
- Pure pH_2 condensation @ 4K (few um)
- Start laser ablation on BaF
- Spray mixed pH_2+BaF



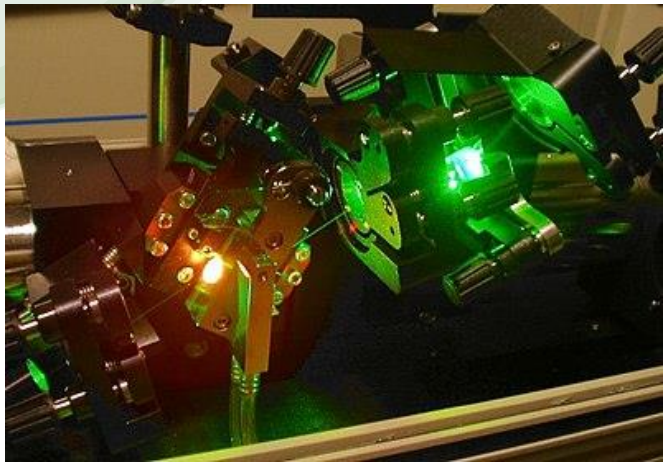
Ongoing steps:

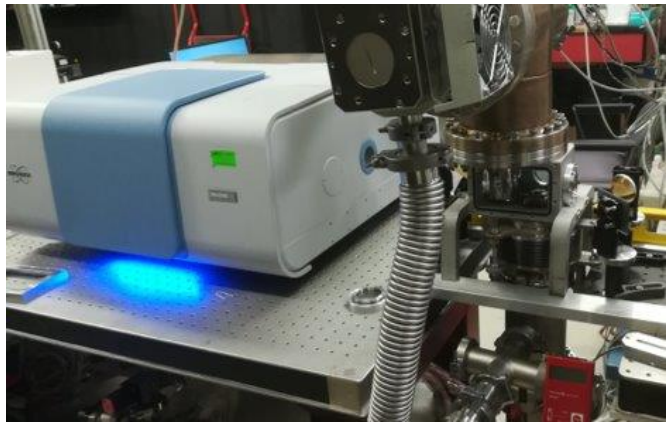
- Understand ratio pH_2/BaF during growth (as function of laser intensity, pH_2 flux, chamber temperature,…)
- Crystal quality and levels (using UV laser induced fluorescence)
- Lines shifts in BaF (through optical spectroscopy)
- Coherence time (through optical spectroscopy)

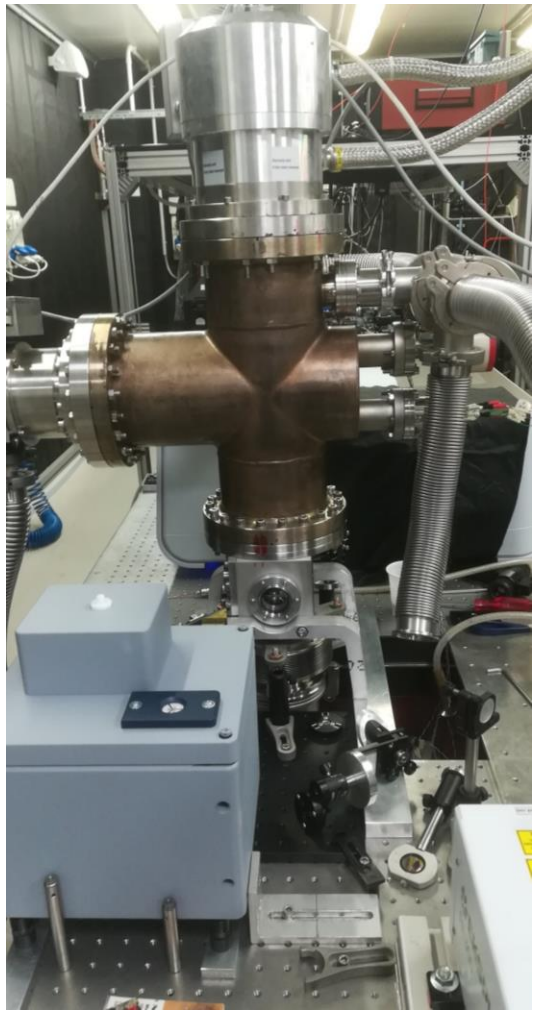
We are working hard on this!!

P-H_2 doped BaF

Future measurements







Main instrumentation:

Vacuum:

- Turbo and rotative pumps (10^{-10}mbar);

Cryogenics:

2 pulse tube refrigerators:

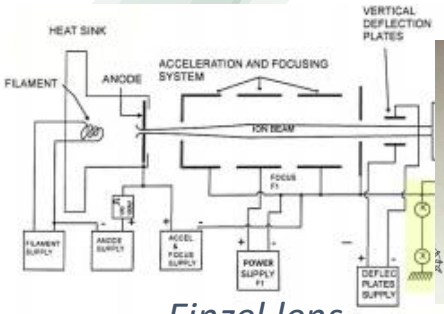
- pH2 production;
- crystal condensation;

Optics:

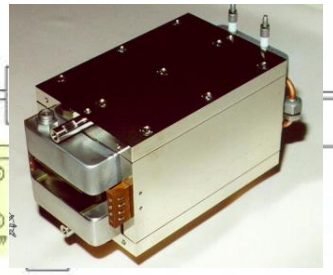
- Ti:Sa laser (tunable range [720-850]nm, linewidth 10kHz);
- Ti:Sa laser (tunable range [695-1005]nm, linewidth 10MHz);
- Dye laser (tunable range [565,595]nm, linewidth 10kHz);
- Q-swithc pulsed NdYAG 266nm;
- Q-swithc pulsed NdYAG 1064nm;
- CW 447nm laser;
- FTIR interferometer Bruker Invenio R882 with 3 detectors:
  - Si Diode [9500-25000]cm^{-1}
  - InSb [1850-10000]cm^{-1}
  - DLaTGS [20-12000]cm^{-1}
- PMTs;

BaF beam Setup

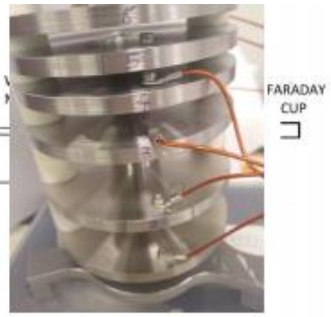
Plasma production + acceleration + focusing + isotopic selection + deceleration



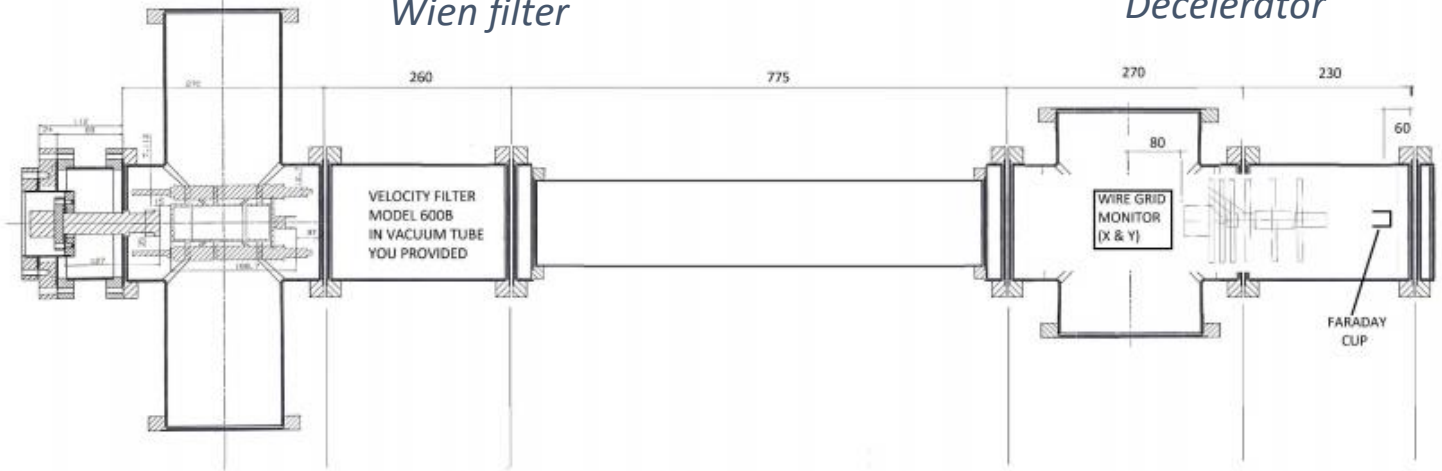
Einzel lens

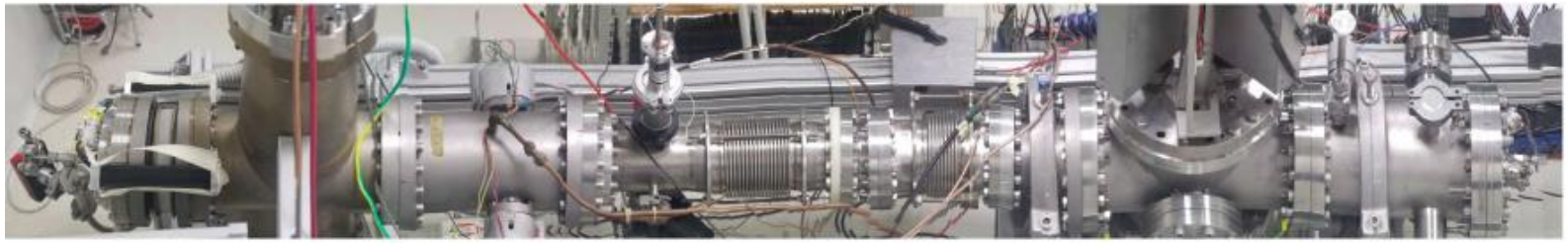


Wien filter



Decelerator





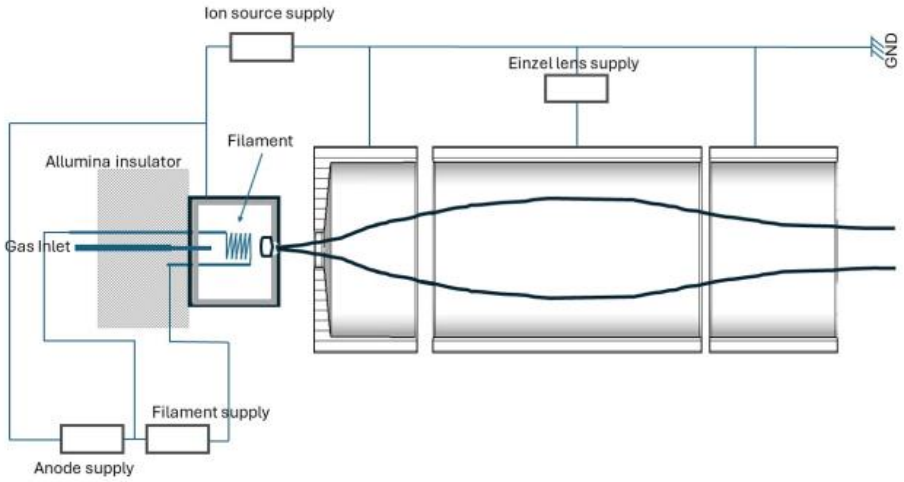
A glow plasma discharge, ignited by a background Argon gas at 10^{-4}mbar, is kept at an overall current of 150 mA so it ionizes the BaF molecules present within the filament and the Tantalum extraction electrode.

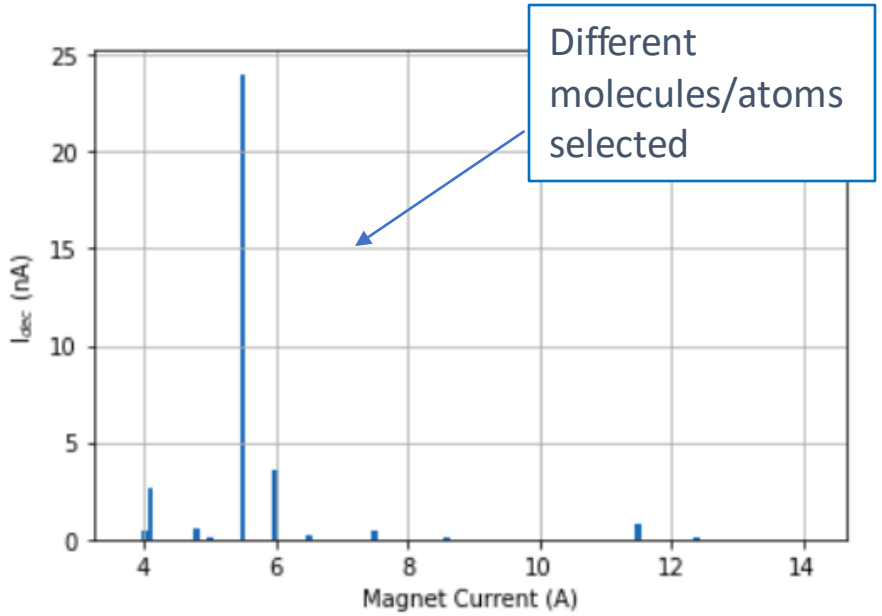
BaF+ molecules are accelerated, focused in an electrostatic lens and selected in a Wien filter.

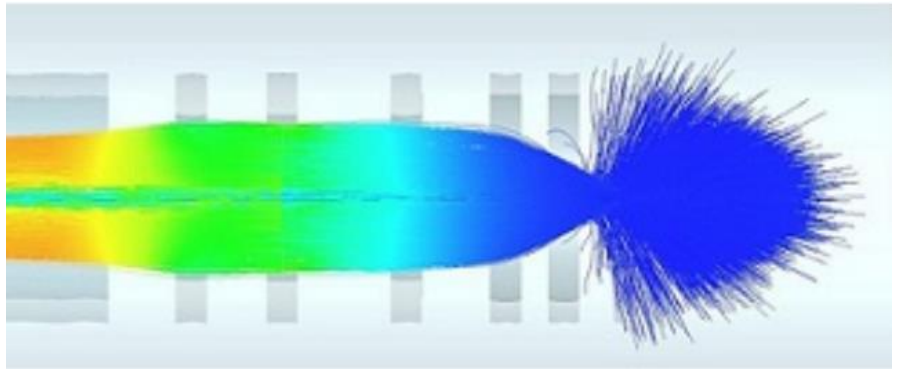
At the end they are decelerated in a "cooled BaF+ beam". Decelerator is configured as a double Einzel lens, to keep the beam focused against space charge.

During test phase we placed a wire grid (250um spaced) and a Faraday cup to monitor the beam.

BaF beam Tests







Unfortunately in the 1st setup built, the current is not to much stable due to the particular mechanical configuration of the crucible so we are upgrading this part

Initial test with Xe^+:

- Similar mass wr to BaF
- 100nA current
- Final energy 5eV

Test with BaF:

- Isotopic separation with Wien filter
- extracted BaF+ beam @ 1 KeV with few uA
- 20nA current @ 5eV, 10 cm far from decelerator

Neutralization chamber

Approach

BaF^+ molecules must be neutralized:

- Not modify matrix structure
- Non disturb EDM measure

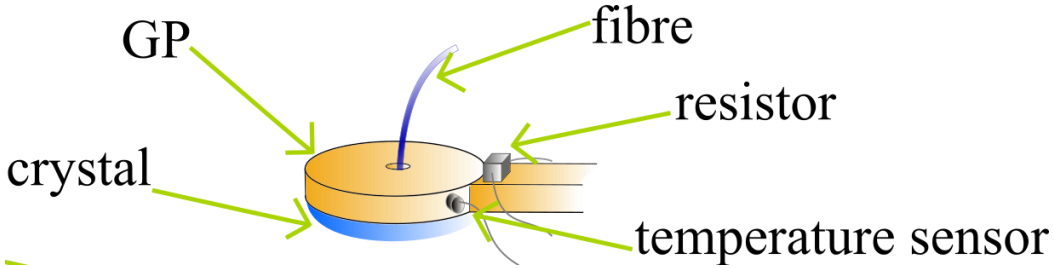
Possible approaches:

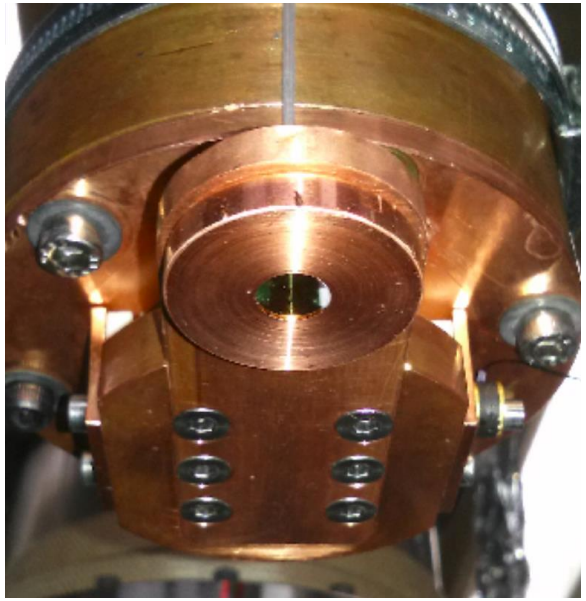
- Cs charge exchange → alkali vapour may contaminate purity
- Graphene → high velocity can destroy graphene structure
- **Direct charge exchange in pH_2 matrix** → Fill the crystal with free electrons

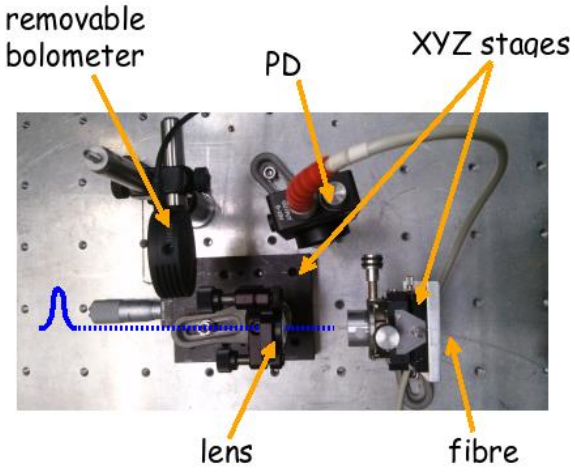
NB We are still probing different approaches

We are developing a system that uses photoelectric effect in AU and inject electrons in the crystal to check the thermalization length in solid para-hydrogen

- UV laser (ns pulses @455nm @ 10Hz)
- Quartz window + Au
- Pulse tube cryocooler + optical fibre







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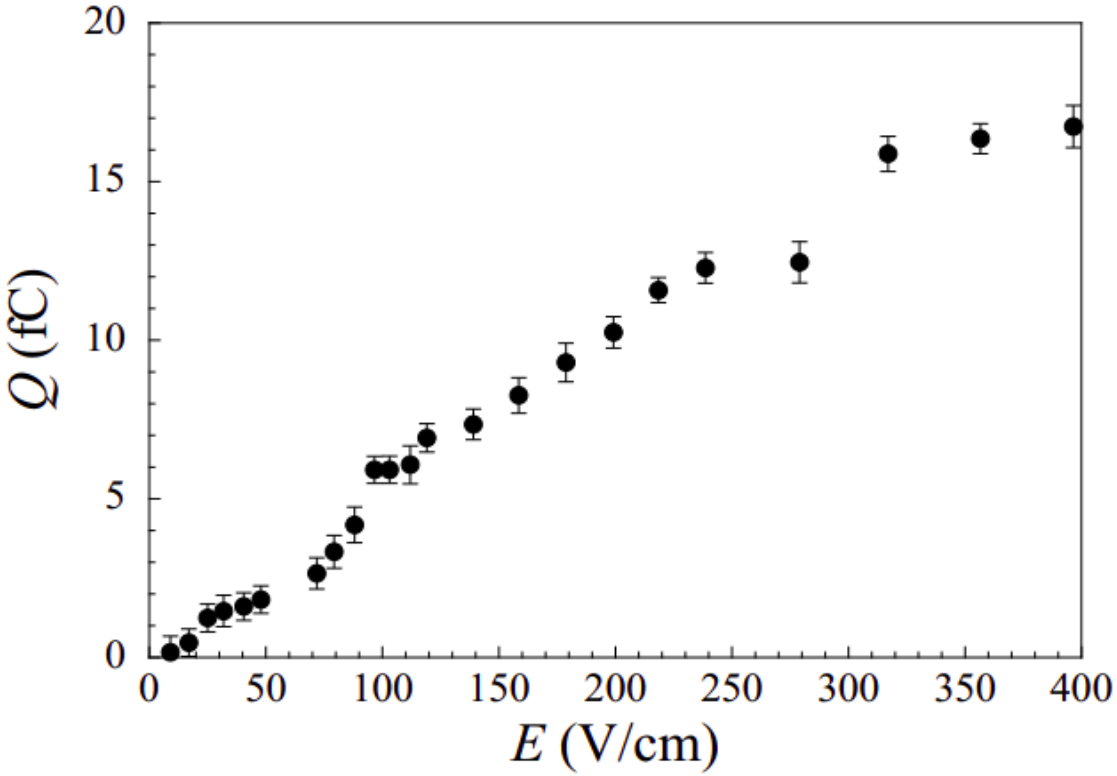
Neutralization chamber

Tests

We performed a measurement of the electrons injected in a pH_2 solid crystal



Charge collected at the anode plotted as a function of the electric field E in the p-H2 film

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Electrons photoinjected into the solid p-H2 film are epithermal.
Upon injection, electrons will lose their excess energy by collisions in the solid until they get thermalized and then propagate in the conduction band of the crystal.
They can reach the crystal-vacuum interphase and can be extracted.

$$\frac{Q}{Q_0} = \frac{B}{D} e^{-z_m/z_0}$$

Q_0 charge collected in vacuum
B describes the effect of the barrier
D describes the backdiffusion process
Z_m is the max of the potential energy of the electrons
Z_0 thermalization length

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Conclusions

- eEDM is a promising probe for physics beyond standard model;
- PHYDES: R&D program to study the feasibility of a possible apparatus that uses BaF molecules embedded into solid $p\text{-}H_2$. Our effort is to measure important quantities such as the number of molecules that can be embedded in the crystal (without disturbing), and the coherence time. Furthermore we would like to understand if there are showstoppers of this approach;
- Based at Legnaro National laboratory of INFN (Italy);
- Different setup for test different approaches:
  - Baf production and selection chamber;
  - Baf neutralization chamber;
  - $P\text{-}H_2$ production and growth;
  - Doped matrix;
- Very early stage but promising results;
- Lots of work to do!

The end

Thank you

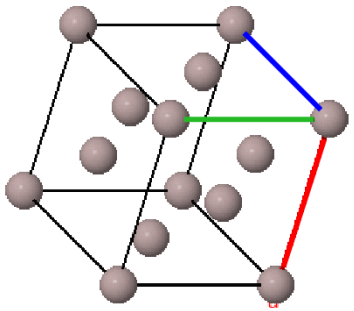
Group: P. Antonini, M. Benettoni, F. A. Borghesani, C. Braggio, R. Calabrese, G. Carugno, F. Chiossi, U. Gasparini, F. Gonella, M. Guarise, A. Khanbekyan, A. Lombardi, E. Mariotti, G. Messineo, J. Pazzini, G. Ruoso, L. Tomassetti, M. Zanetti
Thanks to the technical support of: E. Berto, F. Calaon, M. Tessaro

Backup slides

Cryogenic crystals

Solid crystals made of inert and unreactive material can be grown at cryogenic temperatures

- Examples:



Material	A	T_m(K)	T_b(K)	ρ(g/cm^3)	E_{gap}(eV)	stucture	a(pm)
H_2	2	13.8	20.3	0.08	11.4	hcp	379
Ne	20	24.5	27.1	1.44	21.4	ccp	443
Ar	40	83.8	87.3	1.62	14.2	ccp	525
Kr	84	115.8	119.9	2.83	11.6	ccp	570
Xe	131	161.4	165	3.54	9.3	ccp	620
CH_4	16	91	112	0.51	24.5	ccp	588

- Stable electronic configuration
- Low melting temperature → difficulty of grow and manipulate
- Lennard-Jones potential: $\phi(r) = 4\epsilon\left[\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6}\right]$

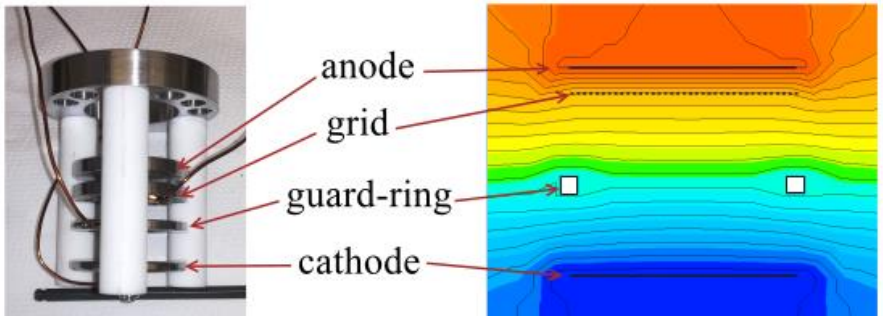
σ = distance among 2 neighboring
ε = energy of interaction

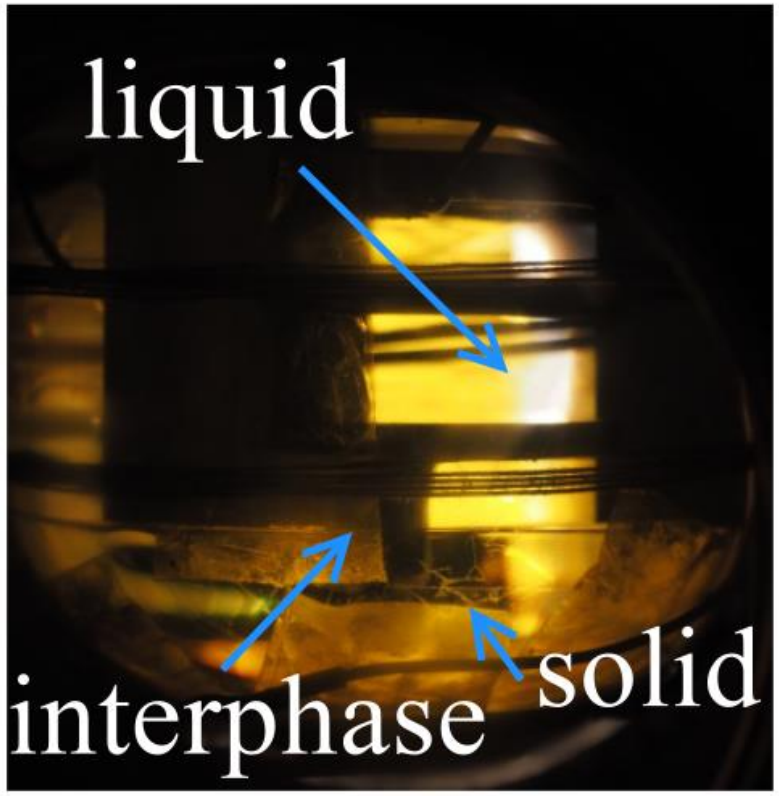
- Weak inter-molecular forces
- Dispersion forces increase with the radius (neon has the lower boiling temperature)

Large Xe crystal

Features:

- Bridgman-Stockbarger modified technique;
- 24 hours of growing;
- T_{melt} ~160 K;
- ∇T=1.5K/cm;
- 0.5 K/h;
- total volume ~150 cm^3;
- total mass~ 0.5 kg;
- 4 electrodes embedded in;
- electric field ~ 0.5 kV/cm;





Large Xenon crystal with
high optical quality