







# Doped cryogenic crystals for fudamental physics researches

The PHYDES apparatus for e-EDM study

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# Outlook

Contents:

- Cryogenic crystals
  - Main features
  - Doping
  - Growing techniques
  - Para hydrogen
- Electron EDM
  - Phenomenon
  - Measure the eEDM
- **PHYDES project** 
  - Approach
  - BaF chamber
  - P-H<sub>2</sub> chamber
  - Results
- Conclusions



# Cryogenic crystals

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

- **Examples: Material**  $\rho(g/cm^3)$ Α T<sub>m</sub>(K) T<sub>b</sub>(K) E<sub>gap</sub>(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 21.4 443 сср Ar 40 83.8 87.3 1.62 14.2 525 сср Kr 84 115.8 119.9 2.83 11.6 570 сср Xe 131 161.4 165 3.54 9.3 620 сср  $CH_4$ 16 91 112 0.51 24.5 588 сср
- Stable electronic configuration
- Low melting temperature  $\rightarrow$  difficulty of grow and manipulate

• Lennard-Jones potential: 
$$\phi(r) = 4\epsilon \left[ \left( \frac{\sigma}{r} \right)^{12} - \left( \frac{\sigma}{r} \right)^{6} \right]$$

 $\sigma = \underline{distance} \ among \ 2 \ neighboring$  $\varepsilon = energy \ of \ interaction$ 

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



# Doped cryogenic crystals

Matrix Isolation Spectroscopy (MIS): G. Pimentel in the 50's  $\rightarrow$  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)



# Cryogenic crystals

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 ۲
  - Cold surface
  - $P_{growth} \sim 10^{-5} mbar$
  - Rate ~ 0.5 cm<sup>3</sup>/h
  - Films
  - Easy annealing



- Modified Bridgeman Stockbargher technique
  - Growth from liquid
  - Low temperature container ۲
  - Fine tuning temperature control
  - Rate ~  $2 \text{ cm}^3/\text{h}$
  - Large solids







## Para-Hydrogen (p-H<sub>2</sub>)

Molecular H<sub>2</sub> 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<sub>2</sub> ~100 cm<sup>-1</sup>
- p-H2 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<sub>2</sub> 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-H2 is extraordinarily slow: small line broadening



# The electron electric dipole moment (e-EDM)

- Electron electric dipole moment (EDM) d<sub>e</sub> is an asymmetric charge distribution along the particle spin: "displacement of centre of charge from centre of mass"
- In the standard model, EDM arises from the CP-violating components of the CKM matrix
- CP violation necessary to explain matter-antimatter asymmetry
- In the SM d<sub>e</sub> <10 <sup>-38</sup> e·cm, but many extensions predict new contribution to EDM at a higer level

THIS MAKES EDM's search an ideal probe for detecting NEW PHYSICS associated with CP violation and a powerful window on energy scales much larger than those that can be probed directly at LHC!





#### How to measure the e-EDM

If we place an electron in a magnetic  $B_0$  and electric field  $E_0$ , we have:

$$H = -\mu_B \frac{\mathbf{S}}{\mathrm{S}} \cdot \mathbf{B_0} - \mathrm{d_e} \frac{\mathbf{S}}{\mathrm{S}} \cdot \mathbf{E_0}$$

If d<sub>e</sub> is non-zero, the interaction with the applied fields make the spin precess:

$$\omega_S = \gamma_e B_0 + rac{\mathrm{d}_\mathrm{e}}{\hbar} \mathrm{E}_0 = \omega_L + \omega_d$$

*Current limit (Cornell group): 4.1x10<sup>-30</sup> ecm* 

Where the magnitude related to the EDM is:  $\omega_d = 2\pi \times 2.4 \times 10^{-5} \text{Hz} \left(\frac{E}{10^{10} \text{V/cm}}\right) \left(\frac{d_e}{10^{-29} \text{e cm}}\right)$ 

To make largest the electric field on the electron

**Molecules:** molecular effective electric field  $E_{\mbox{\tiny eff}}$ 

	BaF	YbF	HgCl
E <sub>eff</sub> (GV/cm)	8	23	115



#### The PHYDES apparatus

Idea: study e-EDM in BaF molecules embedded into p-H<sub>2</sub> matrices —

- R&D program @ INFN Legnaro national laboratory (Italy)
- Implementation in 5 different subchambers:
  - molecular beam production (acceleration, focusing, isotopical selection, neutralization and cooling) paraHydrogen production (storage and control)
  - cryogenic condensation chamber
- Test the feasibility of each step and the limits
- Verify different approaches and other possibilities



Increase number of molecules under test (ppm in a solid matrix)

- BaF: good molecule for EDM
- ParaHydrogen: good matrix

We are currently setting up the system (each chambers) trying to optimize each part: details in next slides



#### BaF beam

Plasma production + acceleration + focusing + isotopic selection + deceleration



INFŃ

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#### Neutralization chamber

BaF<sup>+</sup> molecules must be neutralized:

- Not modify matrix structure
- Non disturb EDM measure

Possible approaches:

- Cs charge excange  $\rightarrow$  alkali vapour may contaminate purity
- Graphene → high velocity can destruct graphene structure
- Direct charge excange in  $pH_2$  matrix  $\rightarrow$  Fill the crystal with free electrons

NB We are still probin 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)
- Quartz window + Au
- Pulse tube cryocooler + optical fibre





### P-H<sub>2</sub> production 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 catayst

Generation of highly <u>enriched</u>  $p-H_2$  at rates of up to 0.5 liters per minute (SLM)





12

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#### Condensation chamber

- Vapour deposition technique
- Ultra high vacuum chamber
- Nozzle on a moving plate
- Pulse tube refrigerator (1W @ 4K)
- Resistors for fine regulation of temperature
- Quartz window for UV-VIS-NIR spectroscopy
- Differet laser systems







- Copper oxygen free high thermal conductivity
- Annealing possibility
- In future will be instrumented with coils
- for RF detection



### p-H<sub>2</sub> characterization

0.5

0.45

0.4

0.35

0.3

0.25

#### **Quantities:**

- Spectrum: I(v)
- Reference spectrum: I<sub>0</sub>(v)
- Transmission spectrum:  $T \rightarrow I(v)/I_0(v)$
- Optical density: OD=-log T





#### Conclusions

- eEDM is a promising probe fo physics beyond standard model
- PHYDES: R&D program to study the feasibility in BaF molecules embedded into solid p-H<sub>2</sub>
- Based at Legnaro National laboratory of INFN (Italy)
- Setup for test different approaches
  - Baf production and selection chamber
  - Baf neutralization chamber
  - P-H<sub>2</sub>roduction and growth
- Promising results

Some references:

Appl. Sci. 2022, 12, 6492. <u>https://doi.org/10.3390/app12136492</u> J. Instrum.,2018 15(3) <u>https://doi.org/10.1088/1748-0221/15/03/C03004</u>. Eur. Phys. J. Plus (2022) 137:673 <u>https://doi.org/10.1140/epjp/s13360-022-02876-4</u>





# Thank you



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