

Beyond standard model using atom in cryogenic matrix: the electron electric dipole moment

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Numerous problem to be solved ☾ New physics

Questions in Fundamental Physics:

- Nature of dark matter and dark energy.
 - Unification of quantum mechanics (probabilistic, discrete) and general relativity (deterministic, continuous).
 - Neutrino mass existence: Standard Model massless, but neutrino oscillations ☾ they have mass
- Charge (C): $e^+ \leftrightarrow e^-$
Parité (P): $r \leftrightarrow -r$
Temps (T): $t \leftrightarrow -t$
- **Tensions:**
 - Matter-antimatter asymmetry: CP violation predicted is insufficient to explain the
 - Muon g-2 anomaly: measured magnetic moment disagree with theory -> new particles or forces?
 - B meson decay anomalies: decays deviate from Standard Model ☾ new particles leptoquarks?
 - Proton radius puzzle: Value obtained via electron scattering or by muonic hydrogen differs
 - **Not happy with:**
 - Strong CP Problem: In quantum chromodynamics no observed CP violation even if possible ($\theta < 1e-10$) ☾ Axions
 - Hierarchy problem.
 - Vast difference in the strengths of the fundamental forces.
 - Higgs boson's mass contributions due to its interactions with other particle should increase its Higgs mass dramatically but to avoid this require coupling constants values tuned to an extreme precision

AMO Physics can play a (big) role

Gravitation and quantum mechanics

Search for exotic force (spin-gravity)

Search for violation of quantum statistics

Precision test of Quantum electrodynamics

Atomic parity violation

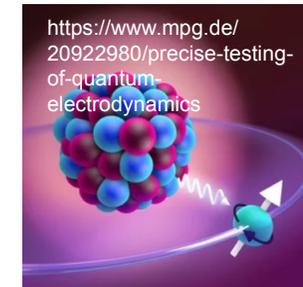
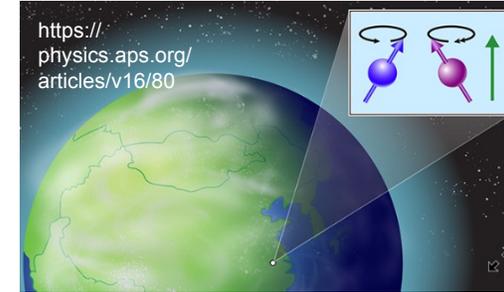
Time reversal violation: electric dipole moments

Test of CPT theorem: matter-antimatter comparisons

Lorentz symmetry tests

Search for (light) dark matter

Search for time variation of fundamental constants



Charge (C): $e^+ \leftrightarrow e^-$
 Parité (P): $r \leftrightarrow -r$
 Temps (T): $t \leftrightarrow -t$



Axion as dark matter
 → osc. eEDM

$$(\partial^\mu \partial_\mu - m^2) a(x) = 0$$

$$\Rightarrow d_e \propto E a_0 \sin(m_a t) \quad \text{Axion mass}$$

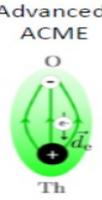
$\frac{m_a t}{\hbar = 6.582 \times 10^{-16} \text{ eV}}$

Mariana Safranova

Fundamental symmetries with quantum science techniques

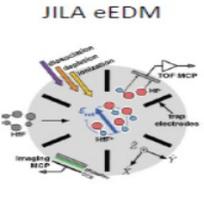
Searches for electron electric-dipole moment (eEDM)

Advanced ACME



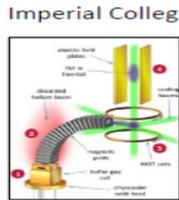
ThO

JILA eEDM



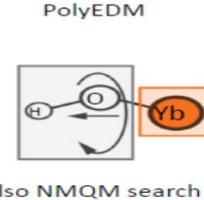
HfF⁺, ThF⁺

Imperial College



YbF

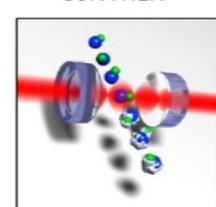
PolyEDM



Also NMQM search
YbOH, ...

Searches for hadronic EDMs

CeNTREX

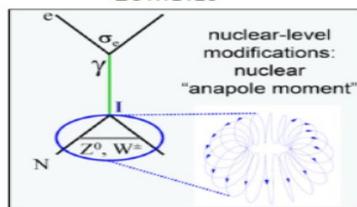


TIF (proton EDM)

Hg
Xe
Ra
EDMs

Enhanced parity violation

ZOMBIES



nuclear-level modifications: nuclear "anapole moment"

Also Yb (Mainz), Fr (FRIUMF & Japan)

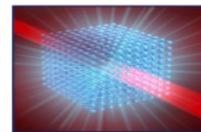
Rapid advances in ultracold molecule cooling and trapping; polyatomic molecules; future: molecules with Ra & "spin squeezed" entangled states

Atomic and Nuclear Clocks & Cavities

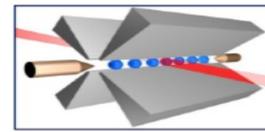
Major clock & cavities R&D efforts below, also molecular clocks, portable clocks and optical links

BSM searches with clocks

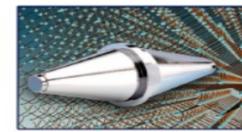
- Searches for variations of fundamental constants
- Ultralight scalar dark matter & relaxation searches
- Tests of general relativity
- Searches for violation of the equivalence principle
- Searches for the Lorentz violation



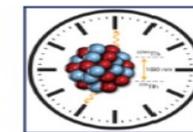
3D lattice clocks



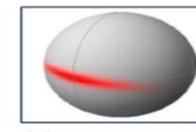
Multi-ion & entangled clocks



Ultrastable optical cavities



Nuclear & highly charge ion clocks



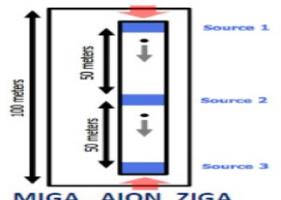
Measurements beyond the quantum limit

Atom interferometry

BSM searches:
Variation of fundamental constants
Ultralight scalar DM & relaxation searches
Violation of the equivalence principle

Prototype gravitational wave detectors

MAGIS-100  Fermilab

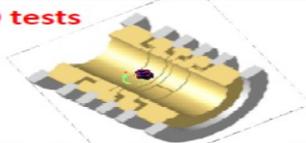


100 meters
50 meters
50 meters

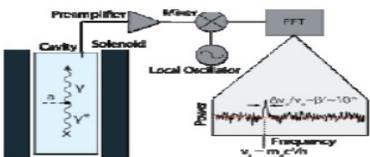
Source 1
Source 2
Source 3

MIGA, AION, ZIGA

QED tests



Highly charged ions and simple systems (H, D, ³He⁺, He, Li, HD, ...)

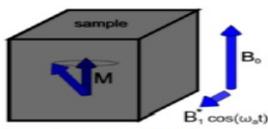


Pre-amplifier, Solenoid, Local Oscillator, FFT

Microwave cavities: HAYSTAC
AMO: measurements beyond quantum limits

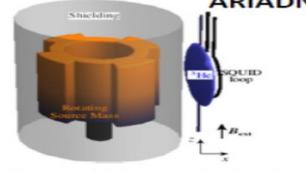
Axion and ALPs searches

CASPER-electric, solids (coupling to gluons)



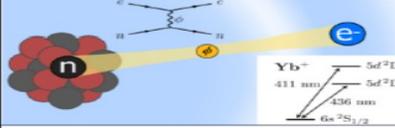
sample, B₀, B₁ cos(ω₀t)

CASPER-wind, Xe (coupling to fermions)



Shielding, SQUID loop, ARIADNE

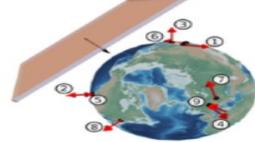
Resonantly detecting axion-mediated forces with NMR



Yb⁺ 5d²D_{5/2}, 5d²D_{3/2}, 411 nm, 436 nm, 6s²S_{1/2}

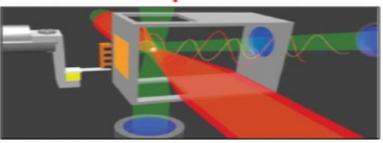
Fifth force searches with precision spectroscopy with atoms and ions

Other dark matter & new force searches



GNOME: network of optical magnetometers for exotic physics

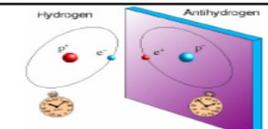
Levitated optomechanics



Also: GW detection and testing the Newtonian inverse square law

CPT tests

\bar{p}, \bar{H}



Hydrogen, Antihydrogen

Many other current & future experiments: tests of the gravity-quantum interface, and HUNTER, SHAFT, ORGAN & UPLOAD (axions), solid-state directional detection with NV centers (WIMPs), doped cryocrystals for EDMs, Rydberg atoms, ...

Fundamental Physics in Small Experiments

T. Blum, P. Winter

T. Bhattacharya, T.Y. Chen, V. Cirigliano, D. DeMille, A. Geraci, N.R. Hutzler, T.M. Ito, D. Kaplan, O. Kim, R. Lehnert, W.M. Morse, Y.K. Semertzidis

nature physics

Review article

<https://doi.org/10.1038/s41567-024-02499-9>

Quantum sensing and metrology for fundamental physics with molecules

Received: 10 August 2023

David DeMille^{1,2}✉, Nicholas R. Hutzler³, Ana Maria Rey^{4,5} & Tanya Zelevinsky⁶

Accepted: 2 April 2024

PHYSICAL REVIEW LETTERS **132**, 190001 (2024)

Essay: Quantum Sensing with Atomic, Molecular, and Optical Platforms for Fundamental Physics

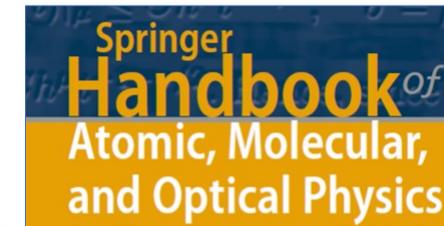
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Searches for New Physics

Marianna S. Safronova

REVIEWS OF MODERN PHYSICS, VOLUME 97, APRIL–JUNE 2025

Spin-dependent exotic interactions

P and T violation —> Good observable for new physics

$$H = H_0\left(\frac{q_i q_j}{4\pi\epsilon_0 \|\mathbf{r}\|}, p^2\right)$$

Conserved parity

Time reversal

Charge conjugation

$P (r \leftrightarrow -r)$

$T (t \leftrightarrow -t)$

$C (q \leftrightarrow -q)$

size of known violating observable zero

•Physicists have been wrong before...

•P violation---Wolfgang Pauli:¹

"I do not believe that the Lord is a weak left-hander, and I am ready to bet a very high sum that the experiments will give symmetric results."

•CP violation---Lev Landau:²

"If CP is violated, I will hang myself."

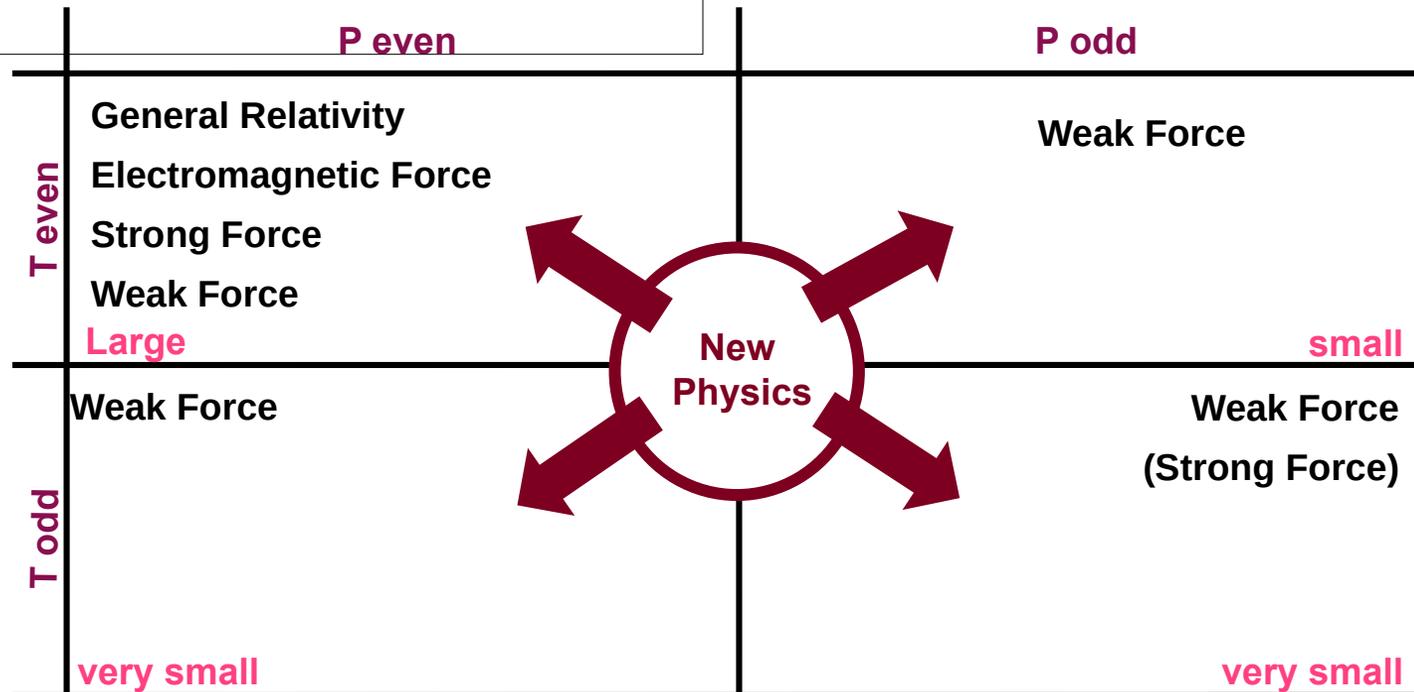
¹Pauli in a letter to Victor Weisskopf, quoted in the Ambidextrous Universe, by Martin Gardner.

²Oral history, as related by Dima Budker.

T violation via CPT \leftrightarrow CP violation (can explain matter/antimatter asymmetry: Sakharov)

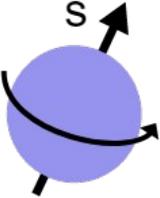
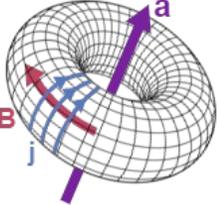
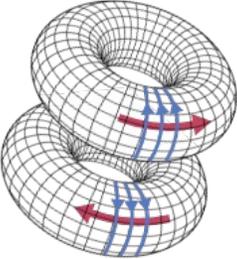
$$H = H^0 + V^{EM} + V^N$$

$$V^N = \underbrace{-V^P}_{\substack{P\text{-odd} \\ T\text{-even}}} - \underbrace{V^{PT}}_{\substack{P\text{-odd} \\ T\text{-odd}}} - \underbrace{V^T}_{\substack{P\text{-even} \\ T\text{-odd}}}$$



small

Parity **P** and Time reversal **T**

	P even	P odd
T even	Magnetic Dipole moment  Electric Quadrupole, Magnetic Octupole, ...	Anapole moment 
P&T violating moments		
T odd	Tetrapole moment 	Electric Dipole moment  Magnetic Quadrupole, Electric Octupole, ...

8

Atomic Hamiltonians

T-odd, P-odd

$$H_{EDM} = S\mathbf{I} \cdot [\nabla\delta(\mathbf{r})]$$

T-even, P-odd

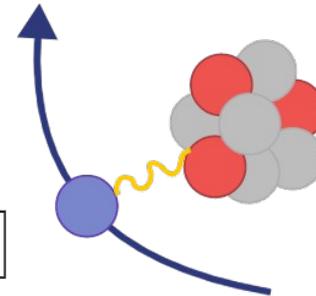
$$H_{\text{anapole}} = \frac{a}{I} \frac{1}{2m} \left[\mathbf{I} \cdot \{ \mathbf{p}, n(\mathbf{r}) \} + i(\mathbf{I} \times \boldsymbol{\sigma}) \cdot \{ \mathbf{p}, n(\mathbf{r}) \} \right]$$

T-odd, P-even

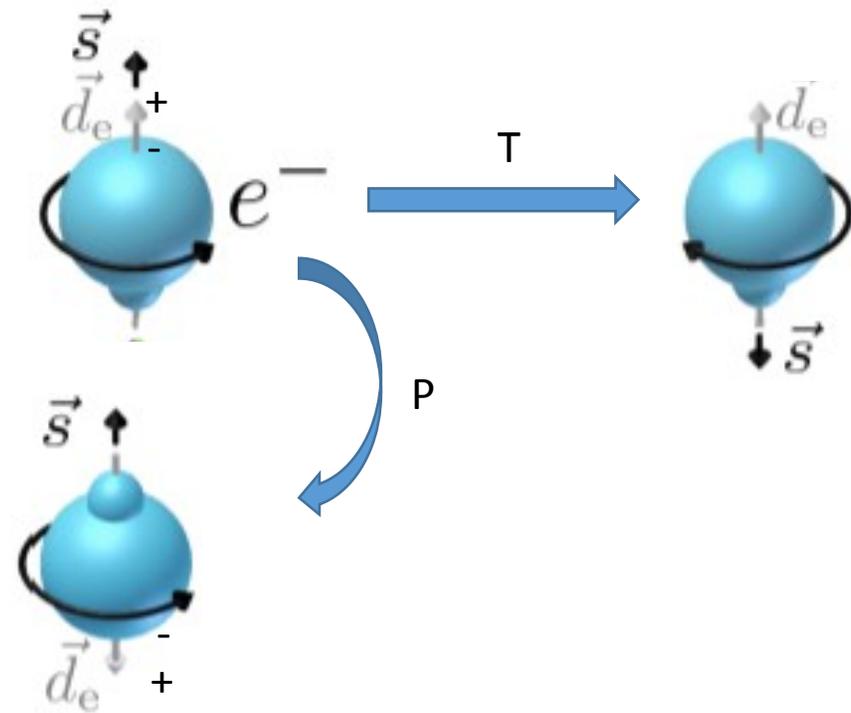
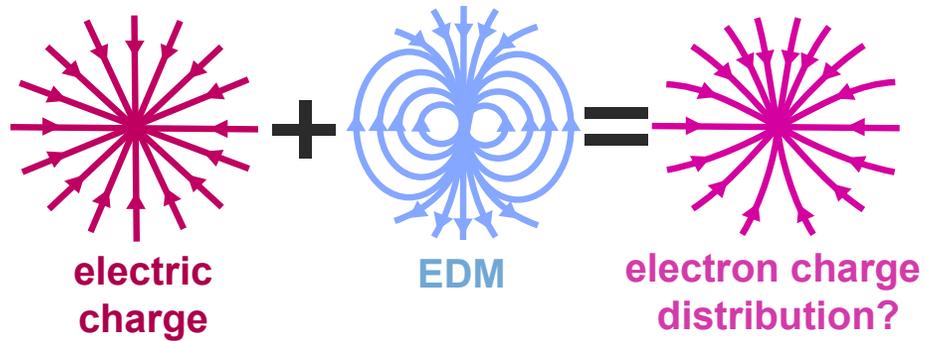
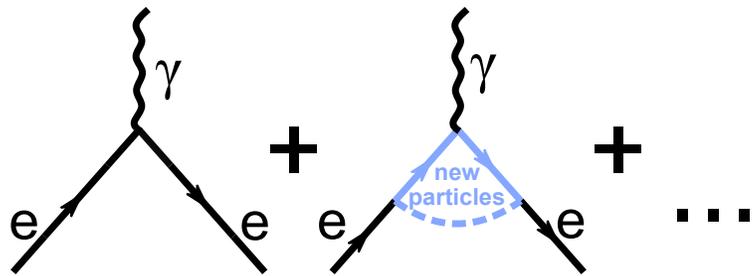
$$H_{\text{tetrapole}} = \frac{t}{2m} \left[iQ_{ij}^{I^2} [\partial_i \partial_j, \delta(\mathbf{r})] + \frac{3 \left\{ (\mathbf{I} \times \boldsymbol{\sigma})_i, I_j \right\} \left[[\partial_i \partial_j \delta(\mathbf{r})] - \delta(\mathbf{r}) \partial_i \partial_j \right]}{2I(2I-1)} \right]$$

nuclear spin

electron spin



P and T violation: Electric Dipole Moment (EDM)



T violation via CPT \Leftrightarrow CP violation (can explain matter/antimatter asymmetry: Sakharov)

EDM – worldwide

<https://www.psi.ch/en/nedm/edms-world-wide> In 2020

Neutrons: (~ 200 ppl.)

- Beam EDM @ Bern
- LANL nEDM @ LANL
- nEDM @ PSI
- nEDM @ SNS
- PanEDM @ ILL
- PNPI/FTI/ILL @ ILL
- TUCAN @ TRIUMF

Storage rings: (~ 400 ppl.)

- CPEDM/JEDI
- muEDM @ PSI
- g-2 @ FNAL
- g-2 @ JPARC

Atoms: (~ 60 ppl.)

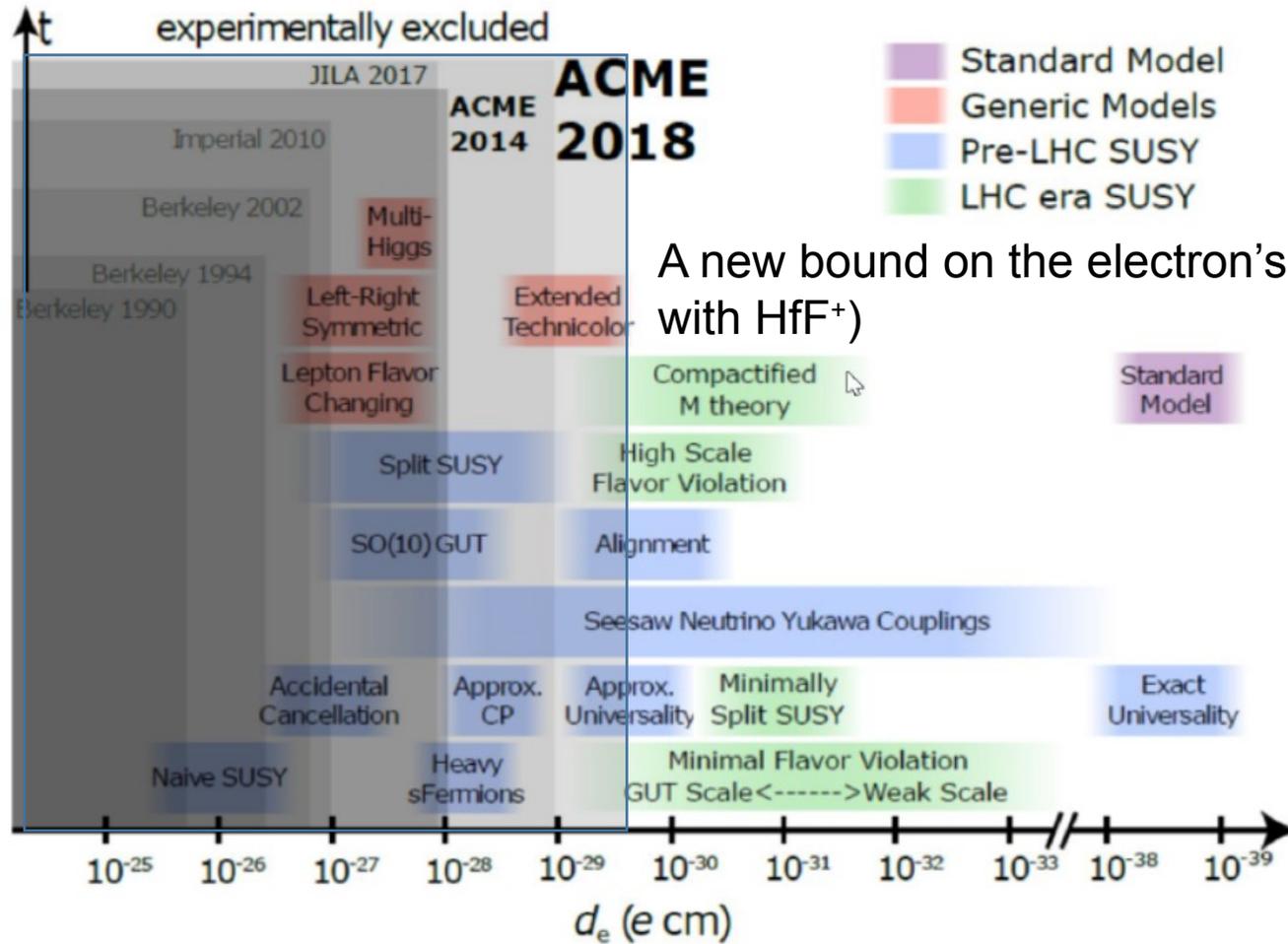
- Cs @ Penn State
- Fr @ Riken
- Hg @ Bonn
- Hg @ Seattle
- Ra @ Argonne
- Xe @ Heidelberg
- Xe @ PTB
- Xe @ Riken



Molecules: (~ 55 ppl.)

- BaF (EDM³) @ Toronto
- BaF (NLeEDM) @ Groningen/Nikhef
- HfF⁺ @ JILA
- ThO (ACME) @ Yale
- YBF @ Imperial

Predicted electron electric dipole moments (compiled by D. DeMille)



A new bound on the electron's electric dipole moment (JILA with HfF+)

Super high energy physics with table top experiments

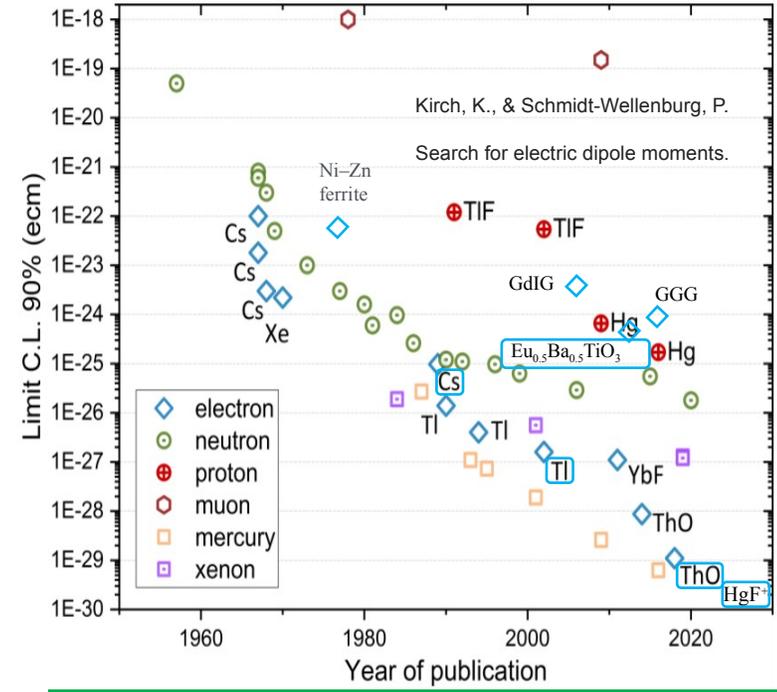
$$m \approx \sqrt{\frac{10^{-26} e \cdot cm}{d_e}} TeV$$

0.05 0.5 5 50 500 5000 One loop (TeV)

0.005 0.05 0.5 5 50 500 Two loops (TeV)

EDM studies and project (for electron)

Some EDM history



Beyond Standard Models

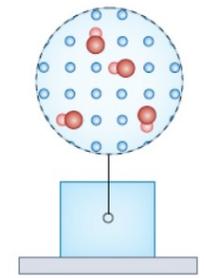
- Supersymmetry (SUSY),
- Multi-Higgs,
- Symmetrical Left-Right
- Leptoquarks,
- Additional dimensions,
- Technicolor and Composite Models,
- Axion Models,
- Neutrino Mass Models,
- Grand Unification Theories (GUT),
- CP-Violant QED Sector Extensions,
- Mirror Matter Models

Standard Model electron EDM $\sim 10^{-38}$ e.cm

$$\Delta H = \langle \psi | \mathbf{d} \cdot \mathbf{E} | \psi \rangle = d_{\text{eff}} \cdot E_{\text{eff}} \cos(\hat{d}\hat{E}) \approx \hbar / \tau$$

$$d \cdot E_{\text{eff}} = \frac{\hbar}{\epsilon_{\text{polar}} \tau \sqrt{N}}$$

Particle	N_{τ} time integrated	Time [s]	Polarization Epsilon,	Eff. Field E_{eff} (V/cm)	EDM e.cm
Cs	10^{14}	0.015	0.7	$5 \cdot 10^5$	$1 \cdot 10^{-26}$
ThO	10^{13}	0.002	0.1	10^{11}	10^{-29}
HgF ⁺	10^7	3	0.8	$2 \cdot 10^{10}$	$4 \cdot 10^{-30}$



Ideal

**N big
 τ long**

Atoms (Cs) in matrix	10^{16}	0.01	0.1	10^6	$6 \cdot 10^{-27}$
	10^{20}	1	1	10^9	$6 \cdot 10^{-35}$
Molecule in matrix	10^{18}	0.1	0.1	10^{10}	$6 \cdot 10^{-33}$
	10^{21}	1	1	10^{11}	10^{-37}

Which atom/molecule ?

$$H = -d \cdot E_{\text{eff}}$$

(laboratory field $E_{\text{lab}} = 10 \text{ kV/cm}$)

$$\mathcal{L}_d = -i \frac{d}{2} \bar{\psi}(\mathbf{x}) \sigma^{\mu\nu} \gamma_5 \psi(\mathbf{x}) F_{\mu\nu}(\mathbf{x}) \quad \rightarrow \quad H_d = -d [2 \gamma_0 \mathbf{S} \cdot \mathbf{E} + i \gamma_0 \boldsymbol{\alpha} \cdot \mathbf{B}]$$

$$H_d = H_d^{\text{clas}} + H_d^{\text{rel}} = \begin{pmatrix} -2d \mathbf{S} \cdot \mathbf{E} & 0 \\ 0 & -2d \mathbf{S} \cdot \mathbf{E} \end{pmatrix} + \begin{pmatrix} 0 & 0 \\ 0 & 4d_e \mathbf{S} \cdot \mathbf{E} \end{pmatrix}$$

Schiff's theorem

$$\Delta \mathcal{E} = \langle \psi | H_d | \psi \rangle = \langle \psi | H_d^{\text{rel}} | \psi \rangle = \langle \psi | \begin{pmatrix} 0 & 0 \\ 0 & 4d_e \mathbf{S} \cdot \mathbf{E} \end{pmatrix} | \psi \rangle = \boxed{4d_e \langle \psi_L | \mathbf{S} \cdot \mathbf{E} | \psi_L \rangle}$$

Need mixed state $|\psi\rangle = \epsilon_s |s\rangle + \epsilon_p |p\rangle \quad \rightarrow \quad \boxed{\langle \psi | H_e | \psi \rangle \sim d_e (100 \text{ GV/cm}) (Z/80)^3 \epsilon_s \epsilon_p}$

with $\epsilon_s \approx 1$ and $\epsilon_p \approx \frac{\langle s | ez | p \rangle}{\mathcal{E}(s) - \mathcal{E}(p)} E_{\text{lab}}$

Ex: Atoms 1st order (Perturbation Stark effect)

$$\langle \psi | H_e | \psi \rangle \sim 125 d_e E_{\text{lab}} \quad \text{Cs}$$

Ex: Molecule aligned on field $\epsilon_s \approx \epsilon_p \approx 1/\sqrt{2}$

$$\langle \psi | H_e | \psi \rangle \sim d_e (100 \text{ GV/cm}) \quad \text{ThO, HfF}^+, \dots$$

How to measure → Ramsey spectroscopy

PHYSICAL REVIEW

VOLUME 108, NUMBER 1

OCTOBER 1, 1957

Experimental Limit to the Electric Dipole Moment of the Neutron

J. H. SMITH,* E. M. PURCELL, AND N. F. RAMSEY

Oak Ridge National Laboratory, Oak Ridge, Tennessee, and Harvard University, Cambridge, Massachusetts

(Received May 17, 1957)

An experimental measurement of the electric dipole moment of the neutron by a neutron-beam magnetic resonance method is described. The result of the experiment is that the electric dipole moment of the neutron equals the charge of the electron multiplied by a distance $D = (-0.1 \pm 2.4) \times 10^{-20}$ cm. Consequently, if an electric dipole moment of the neutron exists and is associated with the spin angular momentum, its magnitude almost certainly corresponds to a value of D less than 5×10^{-20} cm.

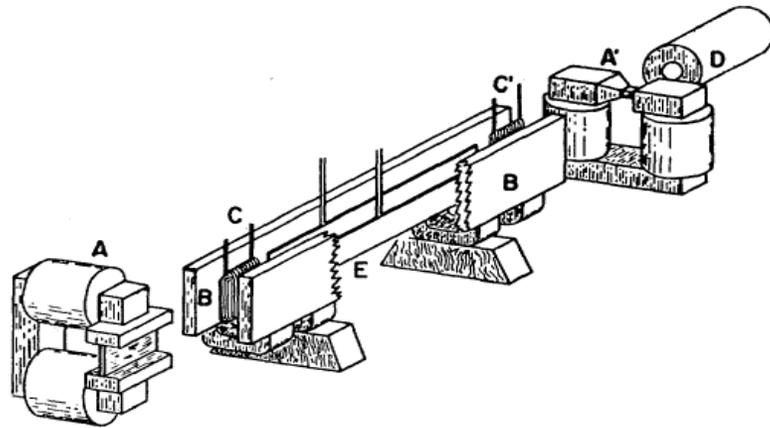
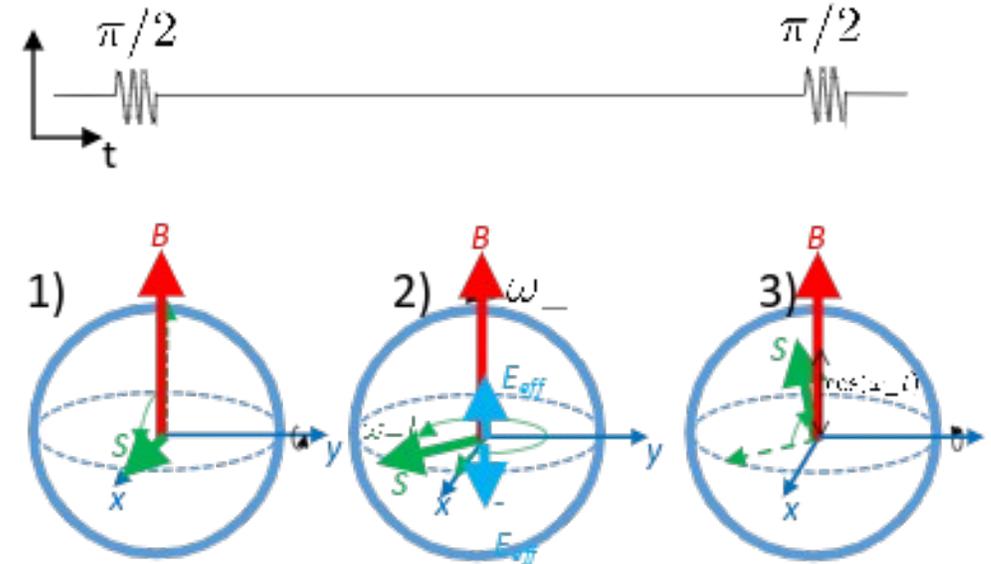
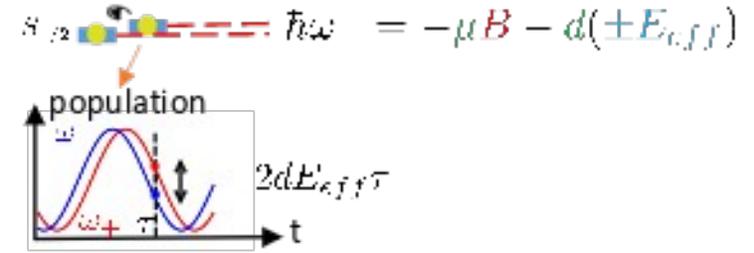


FIG. 1. Schematic diagram of the apparatus. *A*, the magnetized iron mirror polarizer. *A'*, the magnetized iron transmission analyzer. *B*, the pole faces of the homogeneous field magnet. Note the horseshoe-like magnets bolted along the bottom. *C*, *C'*, the coils for the radio-frequency magnetic field. *D*, the BF_3 neutron counter. The magnetic fields in the polarizing magnet and the homogeneous field magnet are at right angles, and two twisted iron strips were used between them to rotate the neutron spins adiabatically.

- 1) Spin preparation $S_{1/2}$
- 2) Precession $\hbar\omega = -\mu B - d(\pm E_{eff})$
- 3) Readout



EDM in matrix



Wutharath Chin, **Claudine Crépin-Gilbert**

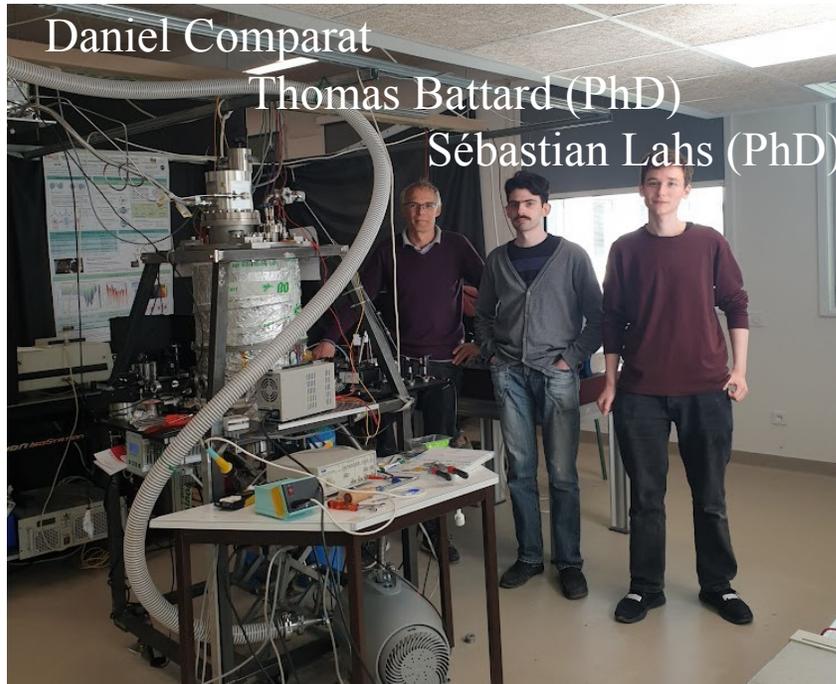
Julie Douady, **Benoit Gervais**, Erwan Hochard, Emmanuelle Jacquet, Raphael Photopoulos

Chloé Malbrunot

Benoit Darquié, Mathieu Manceau,

Hemanth Dinesan (Post-Doc)

Subham Mahapatra (M2)

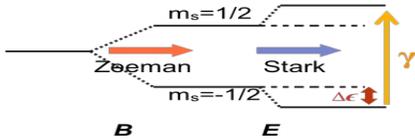
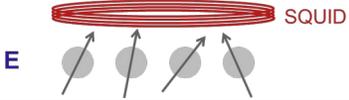


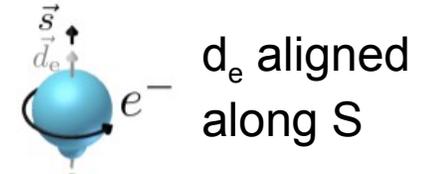
PHYSICAL REVIEW A **108**, 042820 (2023)

Cesium atoms in a cryogenic argon matrix

T. Battard ¹, S. Lahs ¹, C. Crépin ² and D. Comparat ^{1,*}

History of eEDM in solid state systems

year	sample	eEDM limit in e-cm	method	$H = d_e E + \mu B$ $d_e // \mu$
1961	KCr- & NH ₄ -(SO ₄) ₂ 12H ₂ O	$d_e < 10^{-13}$	EPR spectroscopy	$\Delta \text{Energy} = -d \cdot E_{\text{eff}}$ 
1963	Al ₂ O ₃ :Cr & MgO:Cr	$d_e = (1 \pm 4.6) \times 10^{-16}$	EPR spectroscopy	
1979	Nickel-Zink-Ferrite	$d_e = (8.1 \pm 11.6) \times 10^{-23}$	Magnetometry	
2004	GdIG	$d_e = (2 \pm 3) \times 10^{-24}$	Voltage measurement	B field, align spin \leftrightarrow dipole \leftrightarrow Create +/- charges
2011	GGG	$d_e = (-5.57 \pm 7.98_{\text{stat}} \pm 0.12_{\text{syst}}) \times 10^{-25}$	Magnetometry	E field, align dipole \leftrightarrow Spin \leftrightarrow Create B field
2012	Eu _{0.5} Ba _{0.5} TiO ₃	$d_e = (-1.07 \pm 3.06_{\text{stat}} \pm 1.74_{\text{syst}}) \times 10^{-25}$	Magnetometry	



Broken symmetry
->
magnetoelectric

$$E_i^{\text{int}} = E_i^* + \frac{1}{\epsilon_0} P_i^{\text{stat}} + \chi_{ij} E_j^* + \alpha_{ij} B_j^* + \frac{1}{2} \beta_{ijk} B_j^* B_k^* + \frac{1}{2} \gamma_{ijk} B_j^* E_k^* + \frac{1}{2} \chi_{ijk}^{(2)} E_j^* E_k^*$$

$$B_i^{\text{int}} = B_i^* + \frac{1}{\mu_0} M_i^{\text{stat}} + \bar{\chi}_{ij} B_j^* + \alpha_{ij} E_j^* + \frac{1}{2} \beta_{ijk} E_j^* B_k^* + \frac{1}{2} \gamma_{ijk} E_j^* E_k^* + \frac{1}{2} \bar{\chi}_{ijk}^{(2)} B_j^* B_k^*$$

Using cryogenic matrix: An old idea

“ARTIFICIAL VACUUM” FOR *T*-VIOLATION EXPERIMENT 1987

Craig PRYOR
Department of Physics, University of California, Santa Barbara, CA 93106, USA

and

Frank WILCZEK
Institute for Theoretical Physics, University of California, Santa Barbara, CA 93106, USA

PHYSICS LETTERS B

Can paramagnetic atoms in superfluid helium be used to search for permanent electric dipole moments?

M. Arndt, S.I. Kanorsky, A. Weis and T.W. Hänsch

Physics Letters A 174 (1993) 298–303

Proposal for a Sensitive Search for the Electric Dipole Moment of the Electron with Matrix-Isolated Radicals

PRL 97, 063001 (2006)

M. G. Kozlov
Petersburg Nuclear Physics Institute, Gatchina 188300, Russia

Andrei Derevianko

 **atoms** 

Article

Oriented Polar Molecules in a Solid Inert-Gas Matrix: A Proposed Method for Measuring the Electric Dipole Moment of the Electron

A. C. Vutha^{1,*}, M. Horbatsch² and E. A. Hessels²

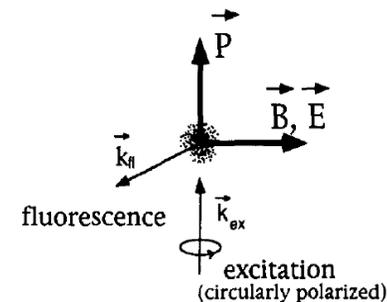
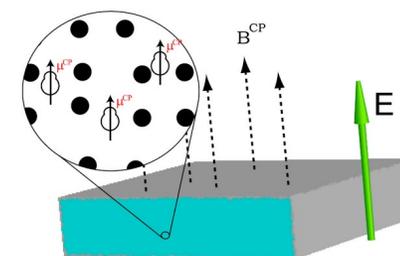
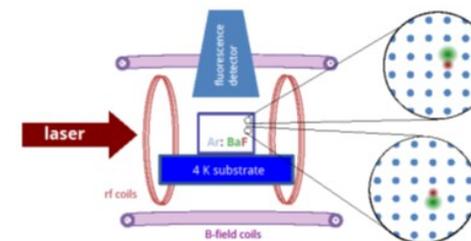


Fig. 4. Geometry of the proposed EDM experiment.

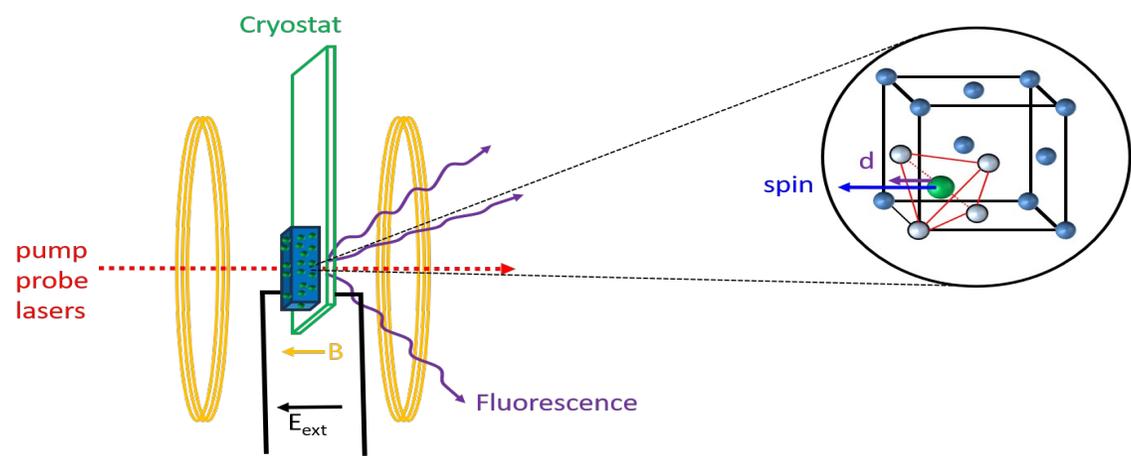


Experiment schematic



BaF
...

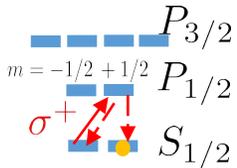
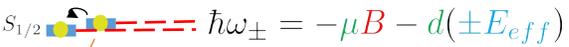
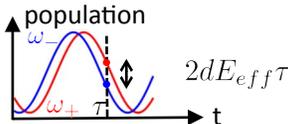
EDM in Matrix

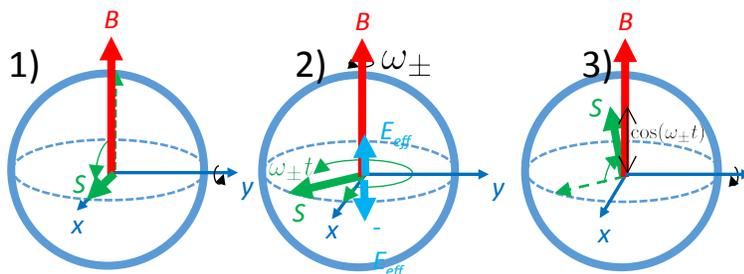


- atoms (Rb, Cs, Tm, Yb, ...)
- molecules (BaF, RaF, ..)
- in rare-gas (Ne, Ar mainly) or parahydrogen
- Lambo R, Xu CY, Pratt S, Xu H, Zappala J, Bailey K, **Lu ZT**, Mueller P, O'Connor T, Kamorzin B et al.. 2021 High-resolution spectroscopy of neutral **Yb atoms** in a solid Ne matrix. Physical Review A 104, 062809.
- Braggio C, Calabrese R, **Carugno G**, Fiscelli G, Guarise M, Khanbekyan A, Noto A, Passante R, Rizzuto L, Ruoso G et al.. 2022 Spectroscopy of **Alkali Atoms** in Solid Matrices of Rare Gases: Experimental Results and Theoretical Analysis. Applied Sciences 12, 6492.
- Gaire V, **Parker C**, Raman C, Li J, Pei Y. 2022 Excitation of magnetic dipole transition of **Thulium** atoms trapped in rare gas crystals. In APS Division of Atomic, Molecular and Optical Physics Meeting Abstracts vol. 2022 pp. H04–009.
- Li S, Ramachandran H, Anderson R, **Vutha A**. 2023 Optical control of **BaF molecules** trapped in **neon ice**. New Journal of Physics.
- Lambo R, Koyanagi G, Horbatsch M, Fournier R, **Hessels E**. 2023 Calculation of the local environment of a **barium monofluoride** molecule in a **neon** matrix. arXiv preprint arXiv:2305.10667.
- Ballof J, Nusgart N, Lalain P, Au M, Heinke R, Leimbach D, Stegemann S, Schutt M, Rothe S, **Singh JT**. 2023 Progress towards the FRIB-EDM3-Frontend: A tool to provide **radioactive molecules** from isotope harvesting for fundamental symmetry studies. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms 541, 224–227.
- Azevedo LO, Costa RJ, Wolff W, Oliveira AN, Sacramento RL, Silveira DM, **Cesar CL**. 2023 **A platform** for trapped cryogenic electrons, anions and cations **for fundamental physics** and chemical studies. arXiv preprint arXiv:2301.13248.
- Lambo RL, Koyanagi GK, Ragyanszki A, Horbatsch M, Fournier R, **Hessels EA**. 2023 Calculation of the local environment of a barium monofluoride molecule in an argon matrix: a step towards using **matrix-isolated BaF for determining the electron electric dipole moment**. Molecular Physics 121, e2198044.

Spin precession align by optical pumping + coherence

USE ATOMS (circular)

- 1) Optical pumping (alignement) 
- 2) Precession 
- 3) Readout 



PHYSICAL REVIEW A VOLUME 54, NUMBER 2 AUGUST 1996
Millihertz magnetic resonance spectroscopy of Cs atoms in body-centered-cubic ⁴He
 S. I. Kanorsky,* S. Lang, S. Lücke, S. B. Ross,† T. W. Hänsch, and **A. Weis**
 Max-Planck-Institut für Quantenoptik, Hans Kopfermann Strasse 1, D-85748 Garching, Germany

PHYSICAL REVIEW A **88**, 063404 (2013)
Optical pumping of rubidium atoms frozen in solid argon

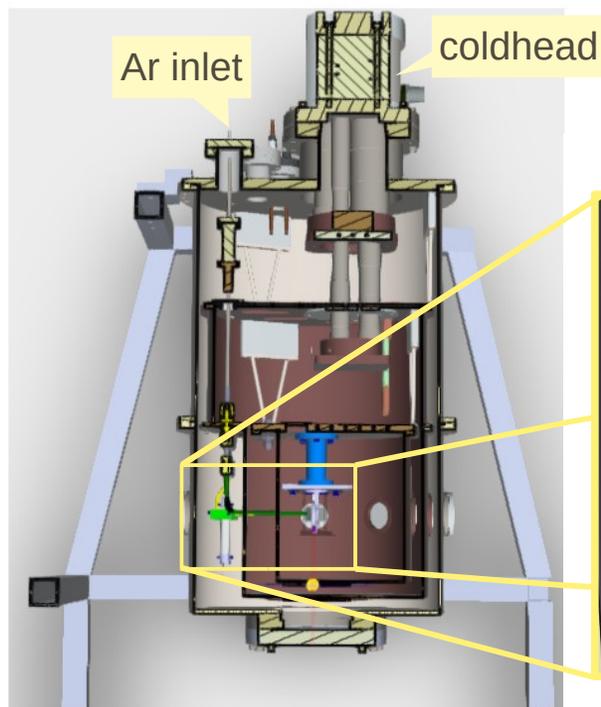
T_1 (population) ~ 1 s

T_2 (coherence, phase) ~ 0.1 s (for Cs)

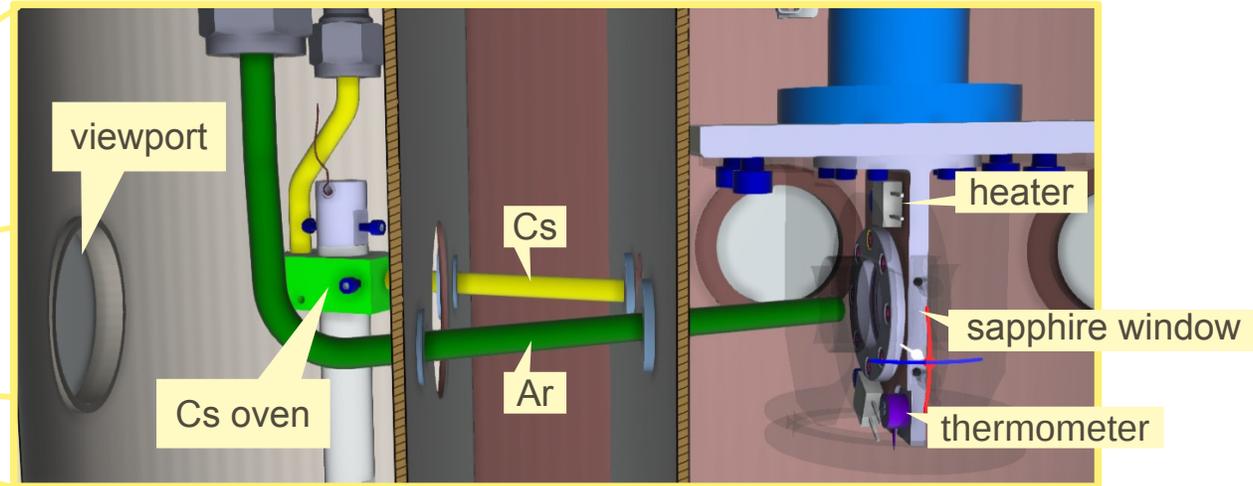
Spin coherence and optical properties of alkali-metal atoms in solid parahydrogen

Sunil Upadhyay,¹ Ugne Dargyte,¹ Vsevolod D. Dergachev,² Robert P. Prater,¹ Sergey A. Varganov,² Timur V. Tscherbul,¹ David Patterson,³ and **Jonathan D. Weinstein^{1,*}**
¹Department of Physics, University of Nevada, Reno NV 89557, USA

Phys. Rev. A 100, 063419 (2019)



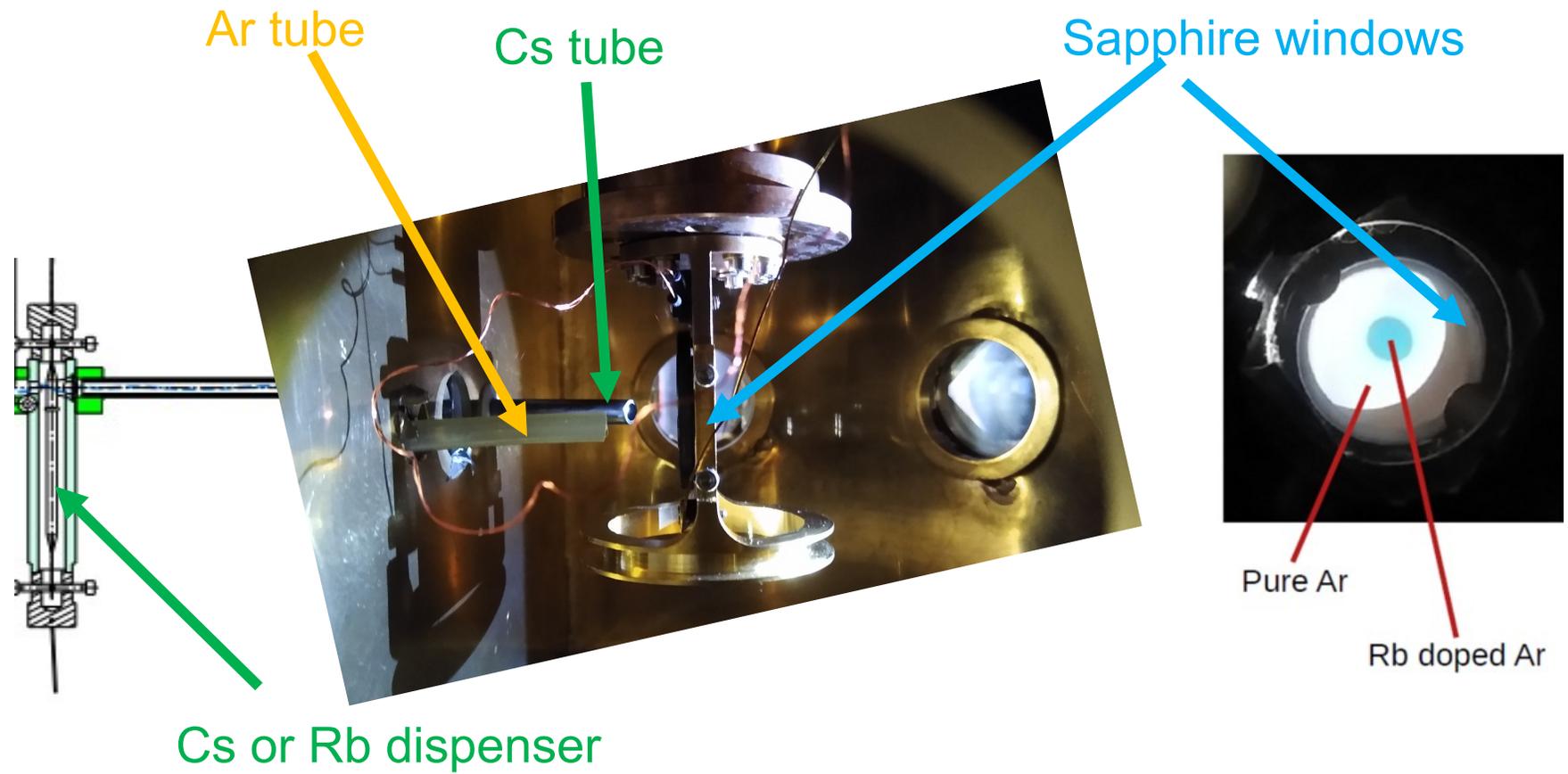
Experimental setup

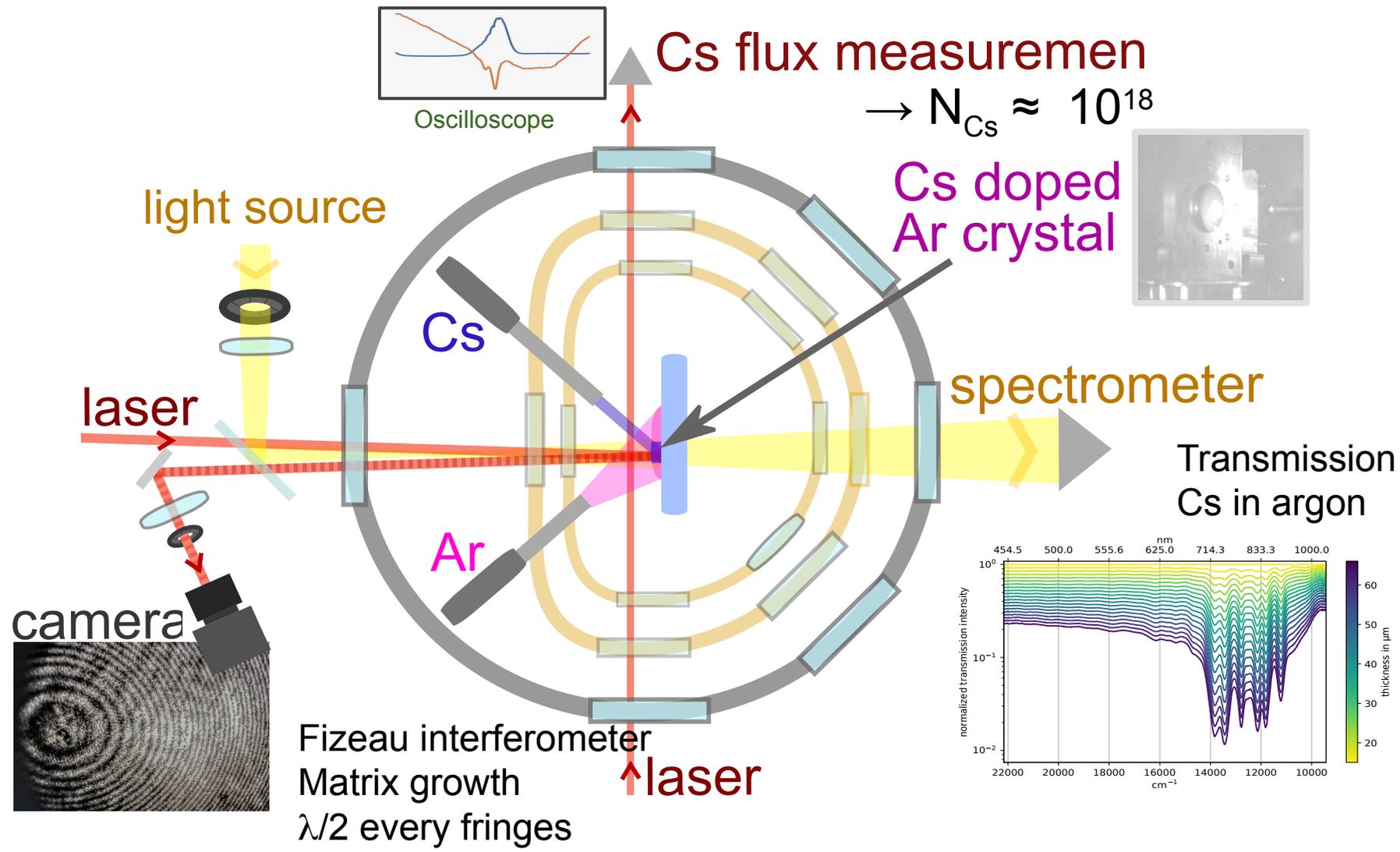


PHYSICAL REVIEW A **108**, 042820 (2023)

Cesium atoms in a cryogenic argon matrix

T. Battard ¹, S. Lahs ¹, C. Crépin ², and D. Comparat ^{1,*}

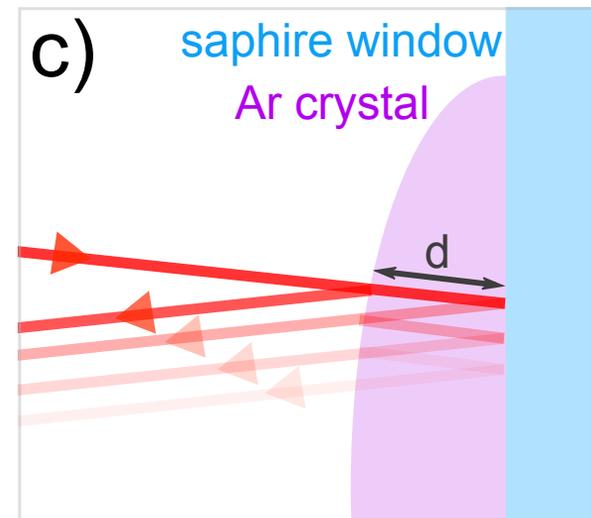
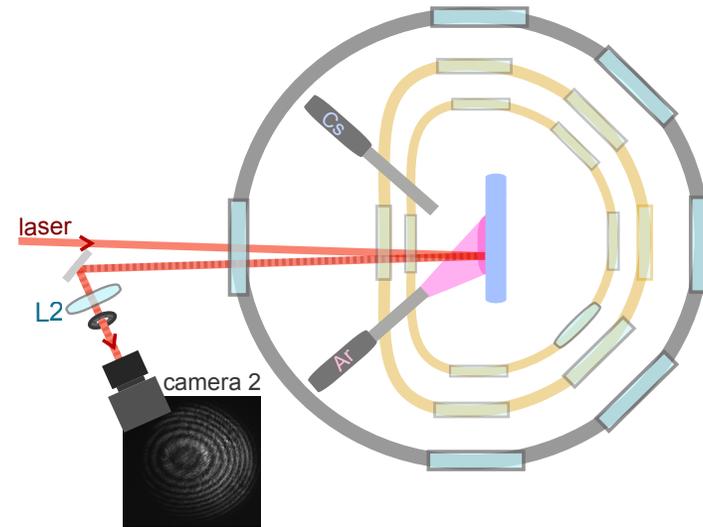




Crystal thickness and quality



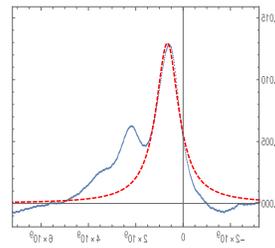
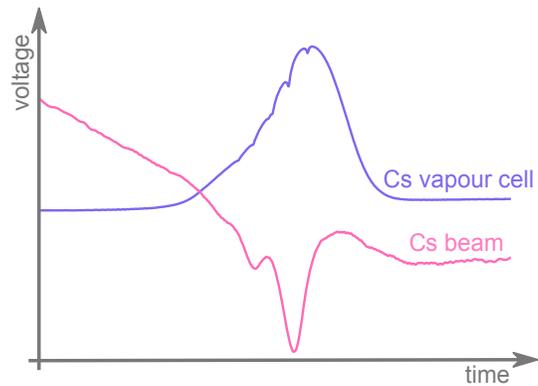
Growth rate $\approx 1\mu\text{m}/\text{min}$



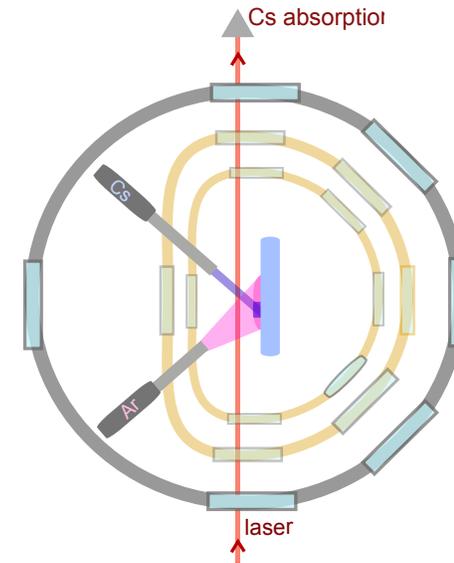
Cs flux measurement

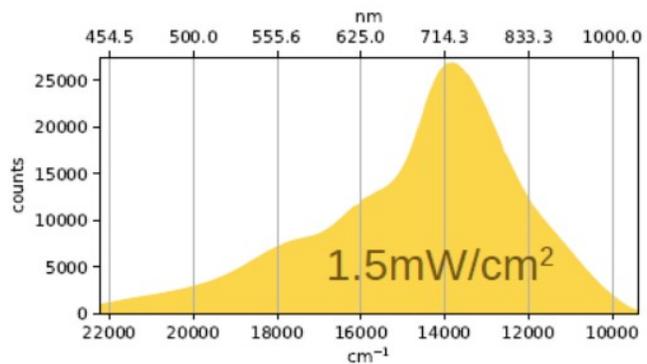
→ In Ar crystal $\sim 10^{18}$ per cm^3

Cs in Ar at Similar distance (10nm) than dislocations!

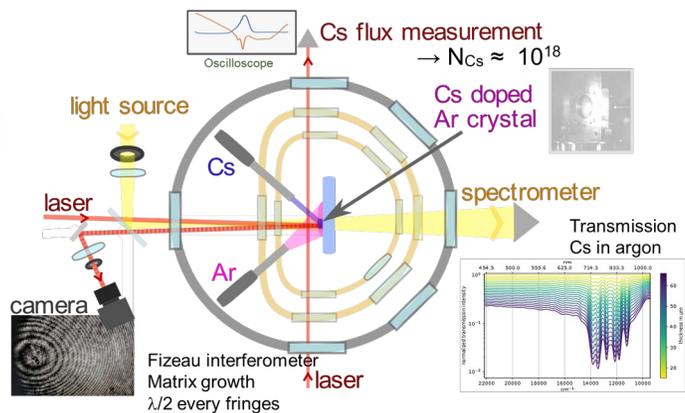


Effusive beam model, broadening by Doppler effect

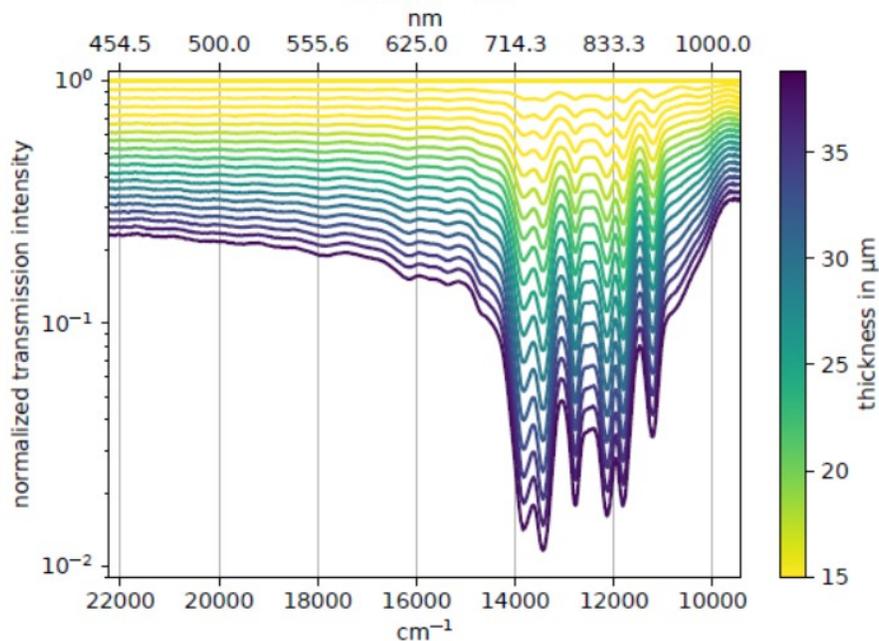




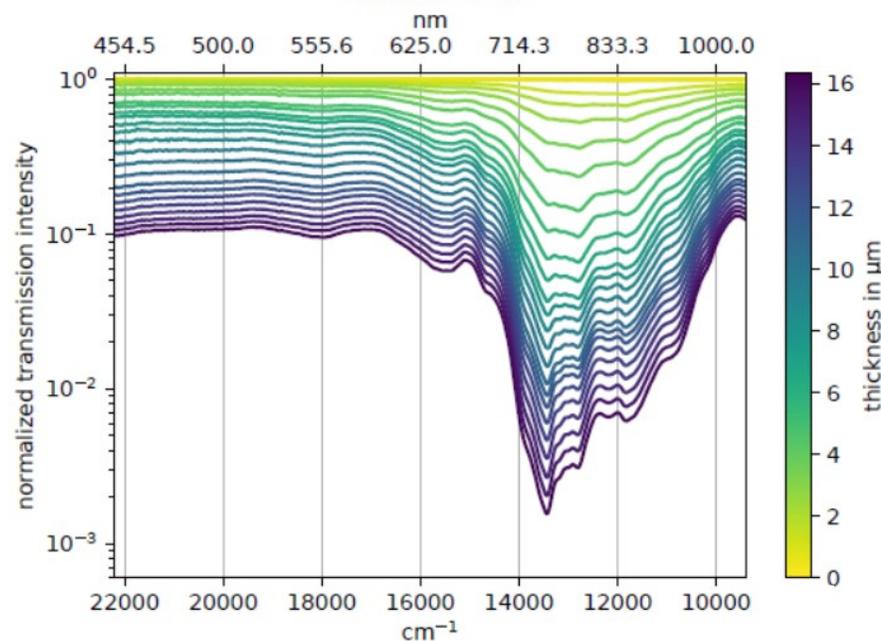
Transmission Spectroscopy



Cs in Ar - 8K



Cs in Ar - 14K



Interpretation of Cs spectrum

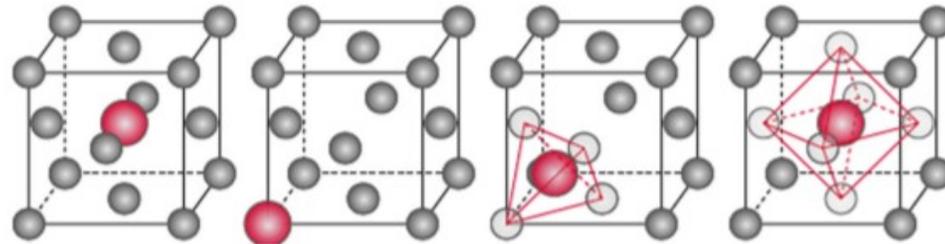
Optical Absorption Spectra of Alkali Atoms in Rare-Gas Matrices

W. WEYHMANN† AND F. M. PIPKIN

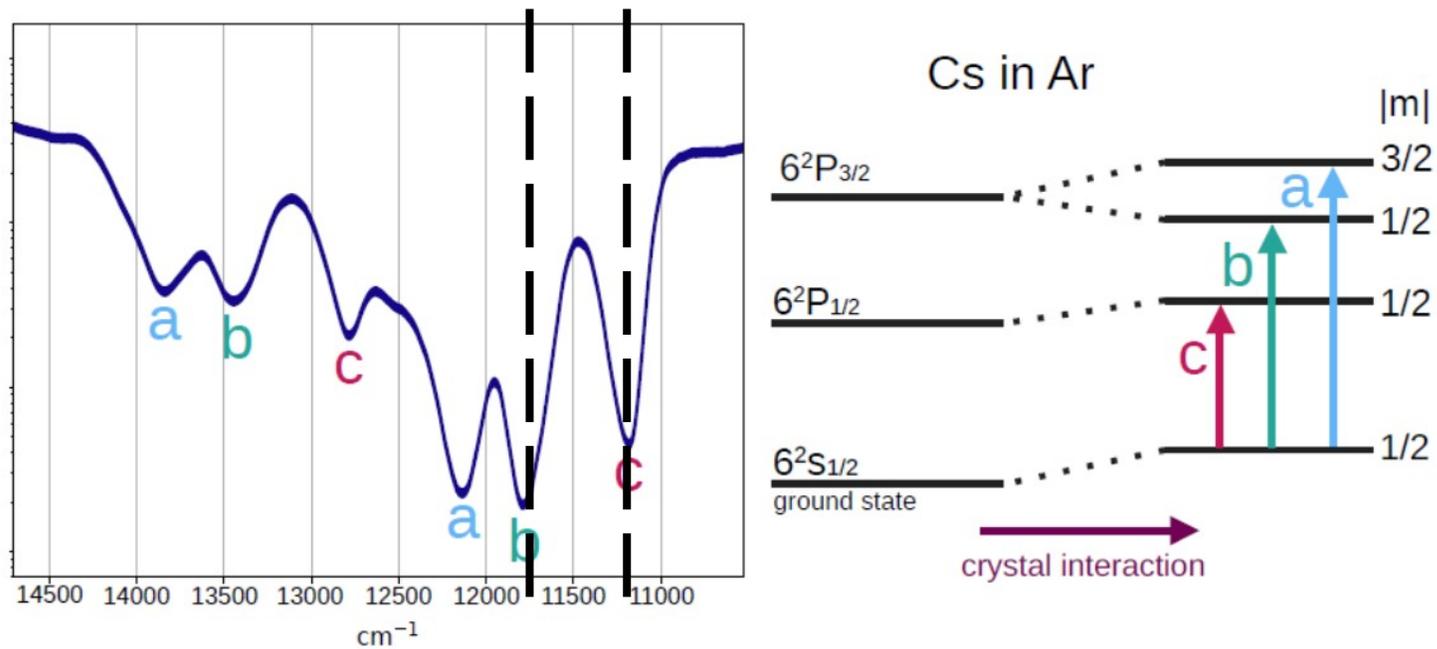
Lyman Laboratory of Physics, Harvard University, Cambridge, Massachusetts

(Received 19 August 1964)

Probably 2 different Trapping sites

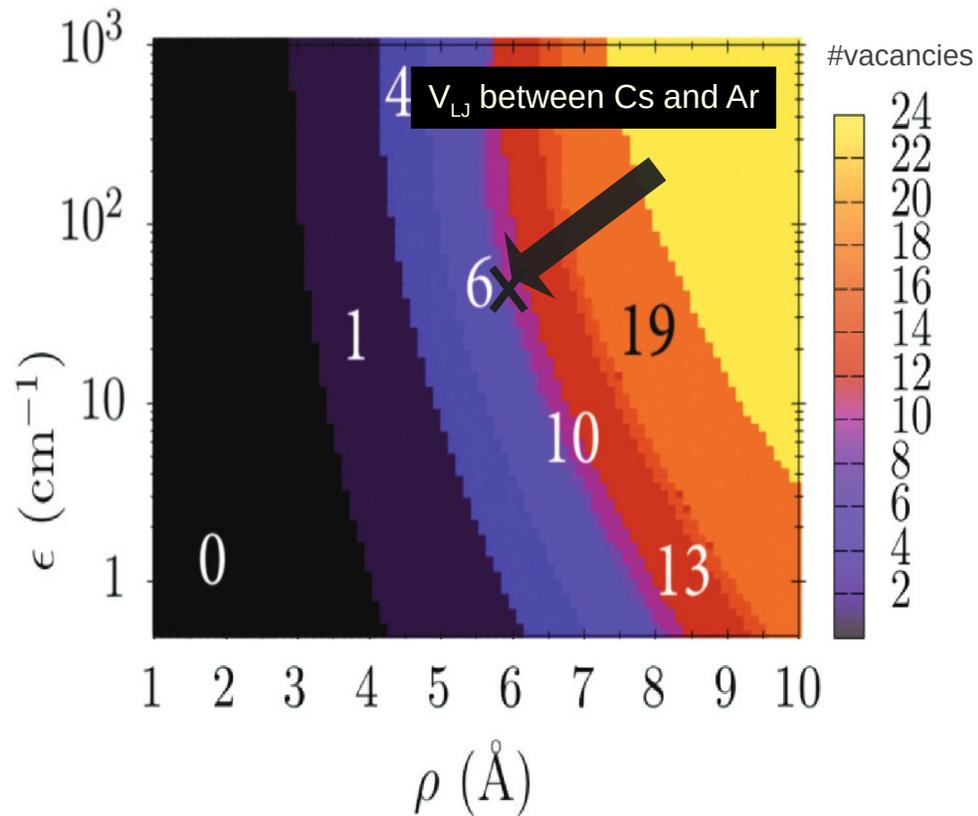


Gas phase Cs(6s)—Cs(6p_J)
852nm 894nm

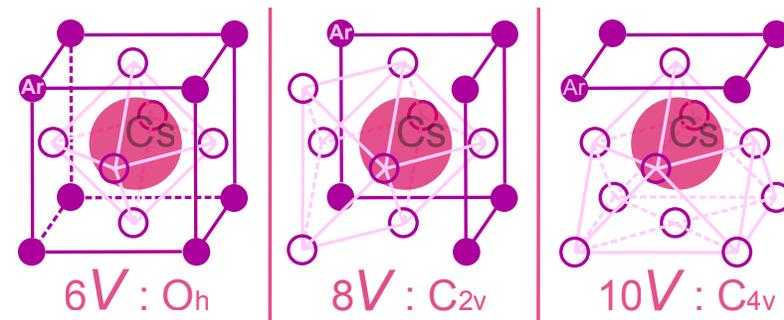


Most probable trapping sites (n=6,8,10 vacancies)?

Free energy of trapping sites in fcc argon for different Lennard-Jones-potentials $V_{LJ}(r) = \epsilon \left[\left(\frac{\rho}{r} \right)^{12} - 2 \left(\frac{\rho}{r} \right)^6 \right]$



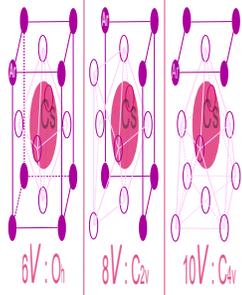
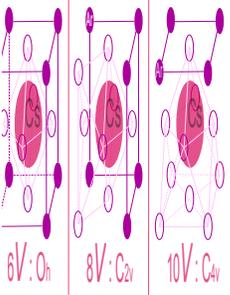
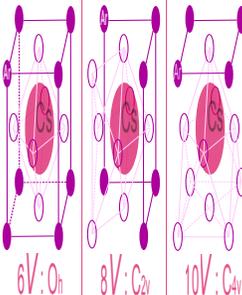
G. K. Ozerov, D. S. Bezrukov, and
A. A. Buchachenko, Phys. Rev. B 103, 184110 (2021)



Different magneto-electric couplings

$$E_i^{\text{int}} = E_i^* + \frac{1}{\epsilon_0} P_i^{\text{stat}} + \chi_{ij} E_j^* + \alpha_{ij} B_j^* + \frac{1}{2} \beta_{ijk} B_j^* B_k^* + \frac{1}{2} \gamma_{ijk} B_j^* E_k^* + \frac{1}{2} \chi_{ijk}^{(2)} E_j^* E_k^*$$

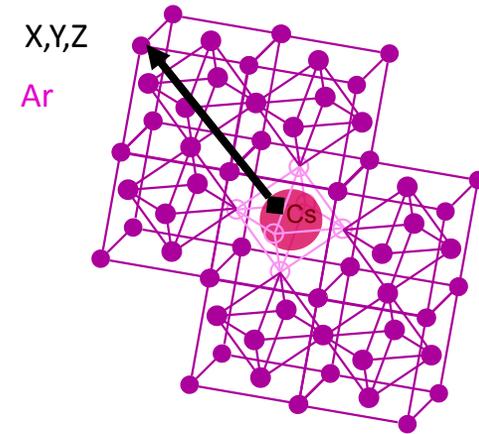
$$B_i^{\text{int}} = B_i^* + \frac{1}{\mu_0} M_i^{\text{stat}} + \bar{\chi}_{ij} B_j^* + \alpha_{ij} E_j^* + \frac{1}{2} \beta_{ijk} E_j^* B_k^* + \frac{1}{2} \gamma_{ijk} E_j^* E_k^* + \frac{1}{2} \bar{\chi}_{ijk}^{(2)} B_j^* B_k^*$$

	 <p>6V: O_h 8V: C_{2v} 10V: C_{4v}</p>	 <p>6V: O_h 8V: C_{2v} 10V: C_{4v}</p>	 <p>6V: O_h 8V: C_{2v} 10V: C_{4v}</p>
α :	$\alpha^{O_h} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$	$\alpha^{C_{2v}} = \begin{pmatrix} 0 & \alpha_{12} & 0 \\ \alpha_{12} & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$	$\alpha^{C_{4v}} = \begin{pmatrix} 0 & \alpha_{12} & 0 \\ -\alpha_{12} & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$
β & γ :	zero	Orthogonal \mathbf{E} - \mathbf{B} -mixing	Orthogonal \mathbf{E} - \mathbf{B} -mixing

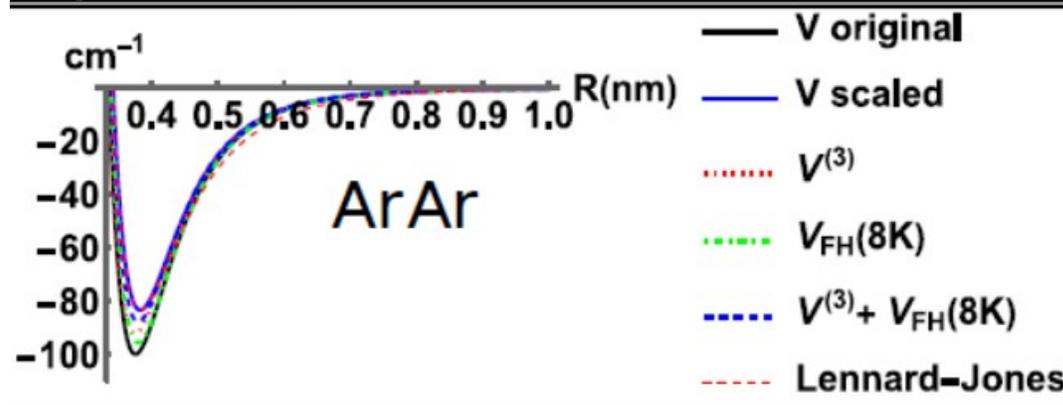
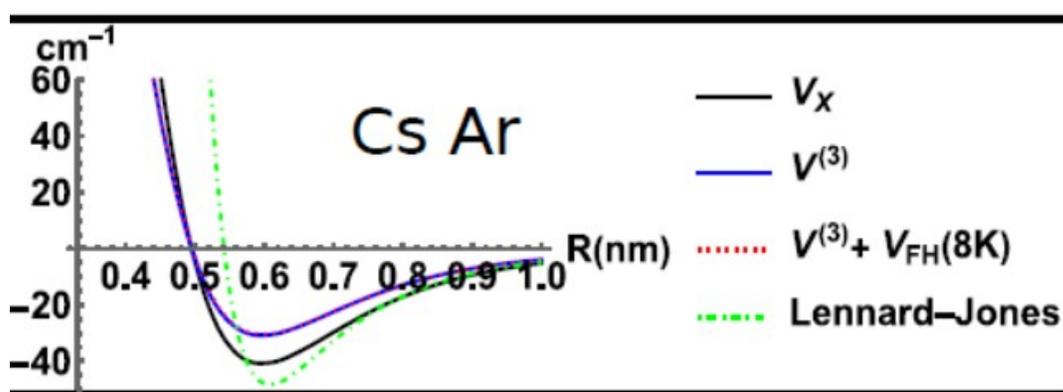
Ar-Ar and Cs-Ar Pair wise potential approximation

$$V = E_{Cs}(n, N)$$

$$V = \sum_{i=1}^{N-n} V_{Ar-Cs}(r_{Cs-Ari}) + \sum_{1 \leq i < j \leq N-n} V_{Ar}(r_{ij})$$



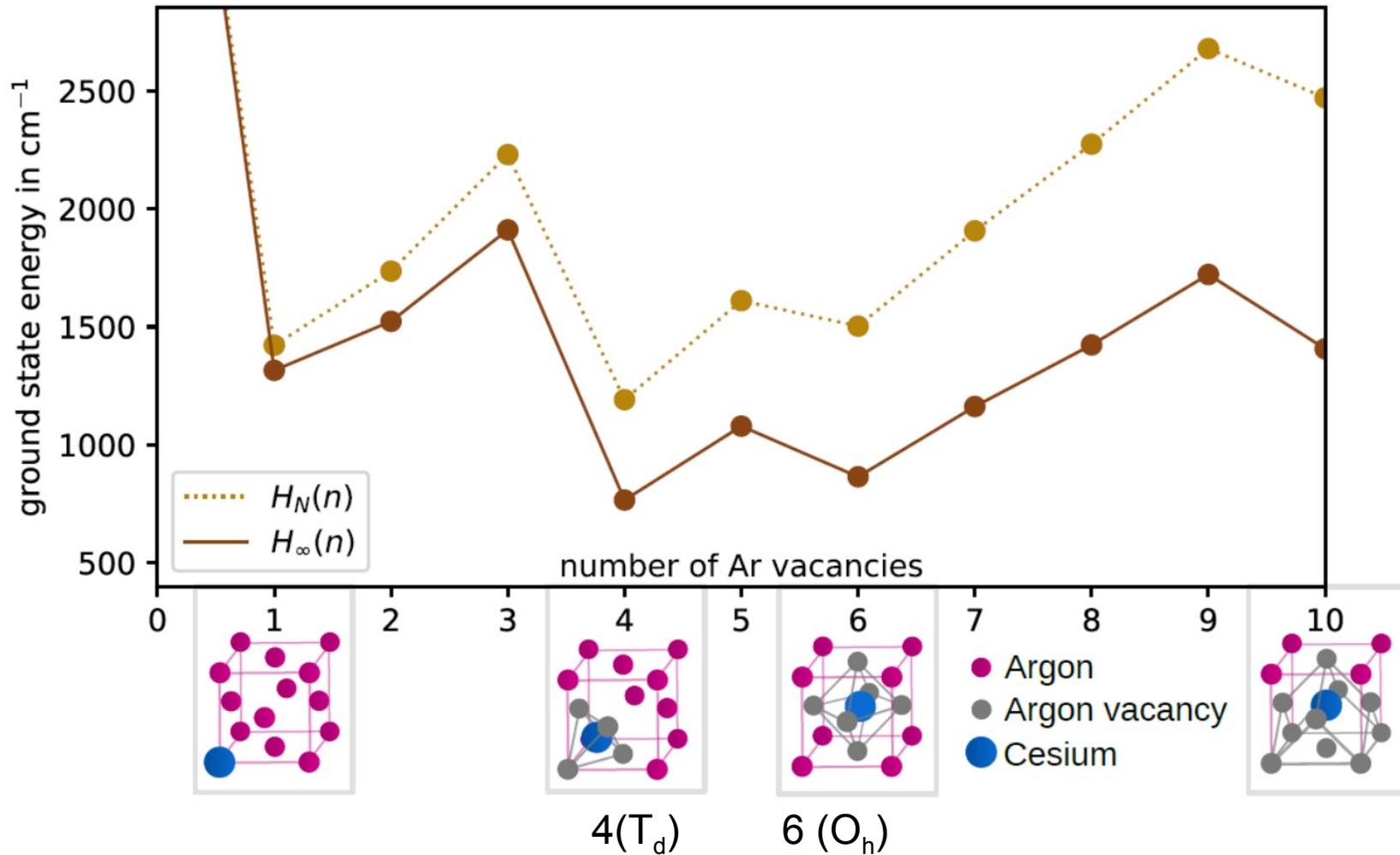
$N \sim 1000$ atoms
 n vacancies



Stability diagram

Accommodation energy: system (Cs+Ar) energy – energy of the pure Ar crystal

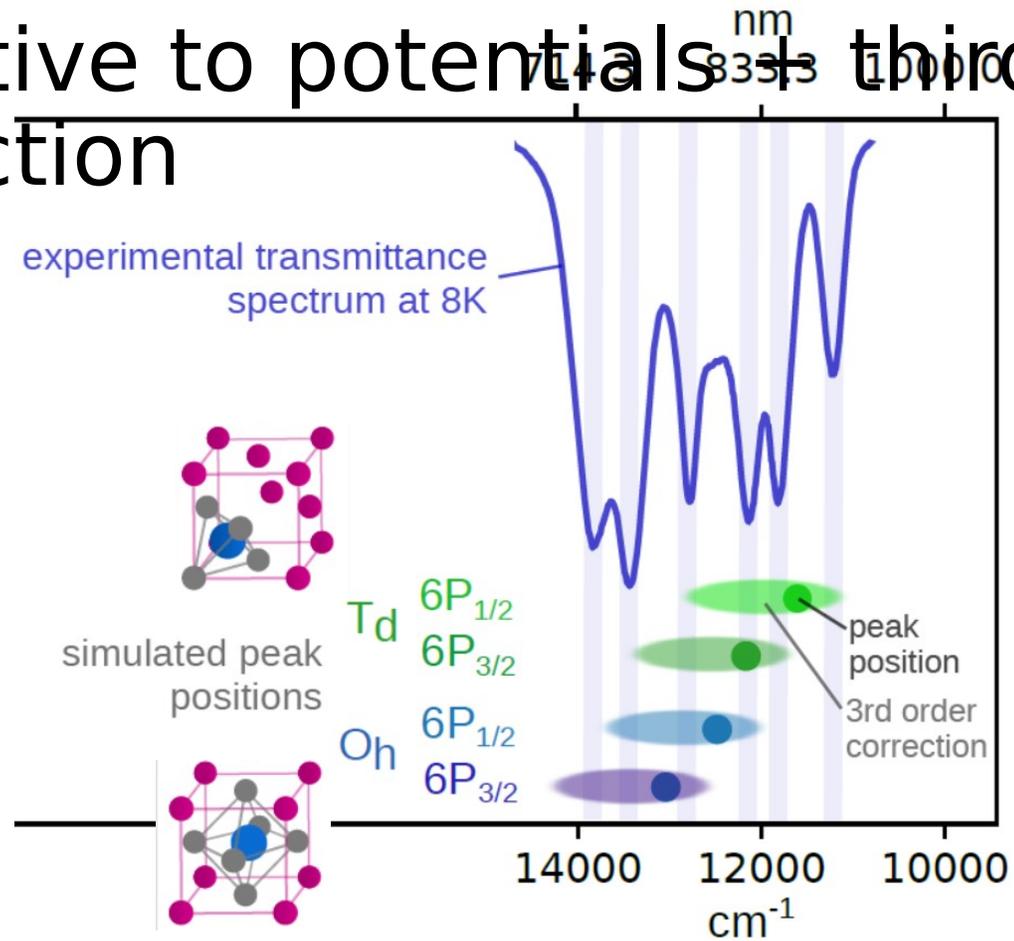
$$\Delta E_N(n) = E_{\text{Cs}}(n, N) - E_{\text{Ar}}(N)(N - n)/N$$



Convex stability (ex: 5+5 \in 4 +6) \in **4 vacancies or 6 vacancies**

Reasonable line positions for $4(T_d) - 6(O_h)$

Sensitive to potentials + third order correction

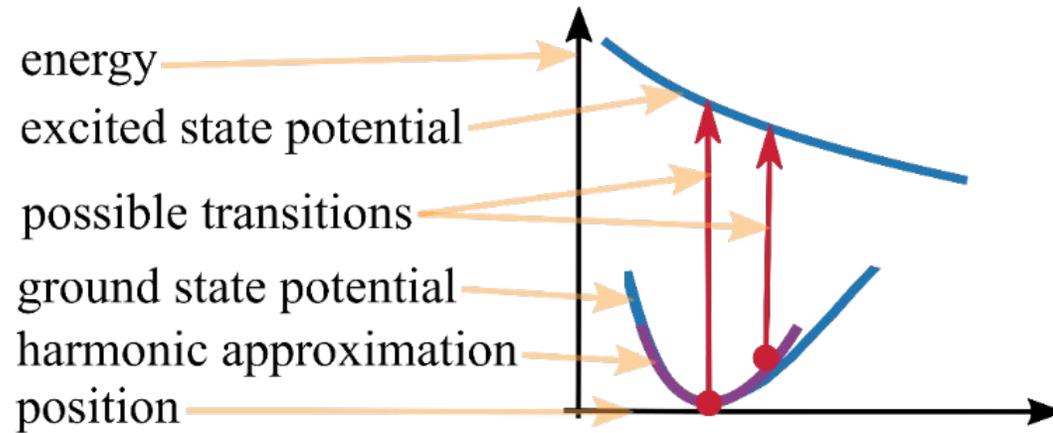


Oh(6) blue shift

$T_d(4) \sim 0$ shift

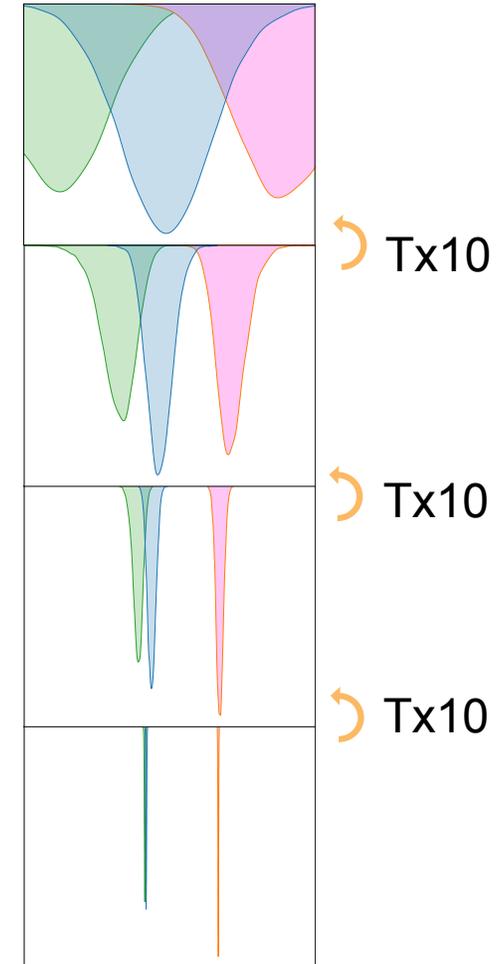
Jahn-Teller Dynamical (thermal) effect

☾ Broadening and splitting



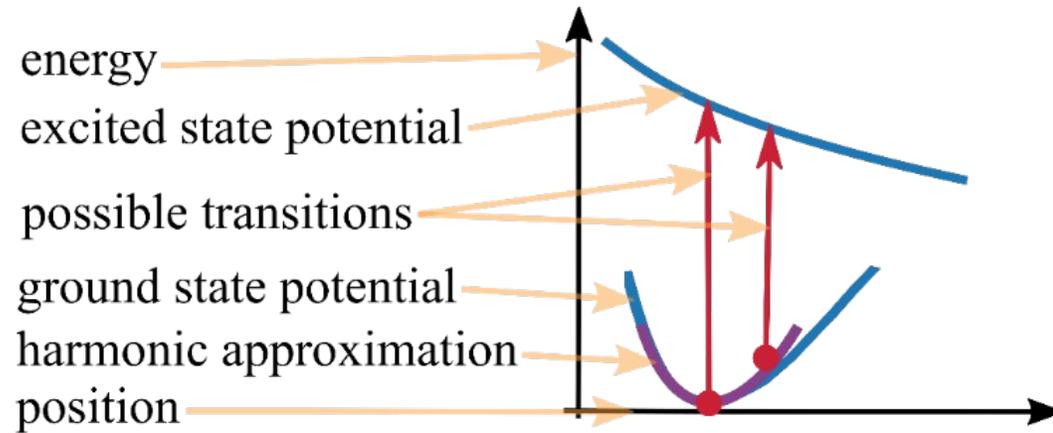
Semi-classical Franck-Condon approximation

Crystal field
1st order in excited state



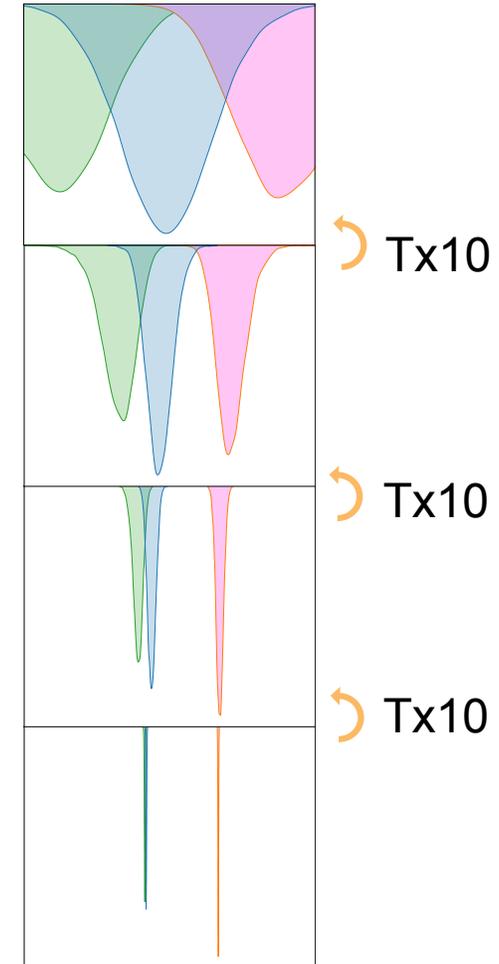
Jahn-Teller Dynamical (thermal) effect

☾ Broadening and splitting

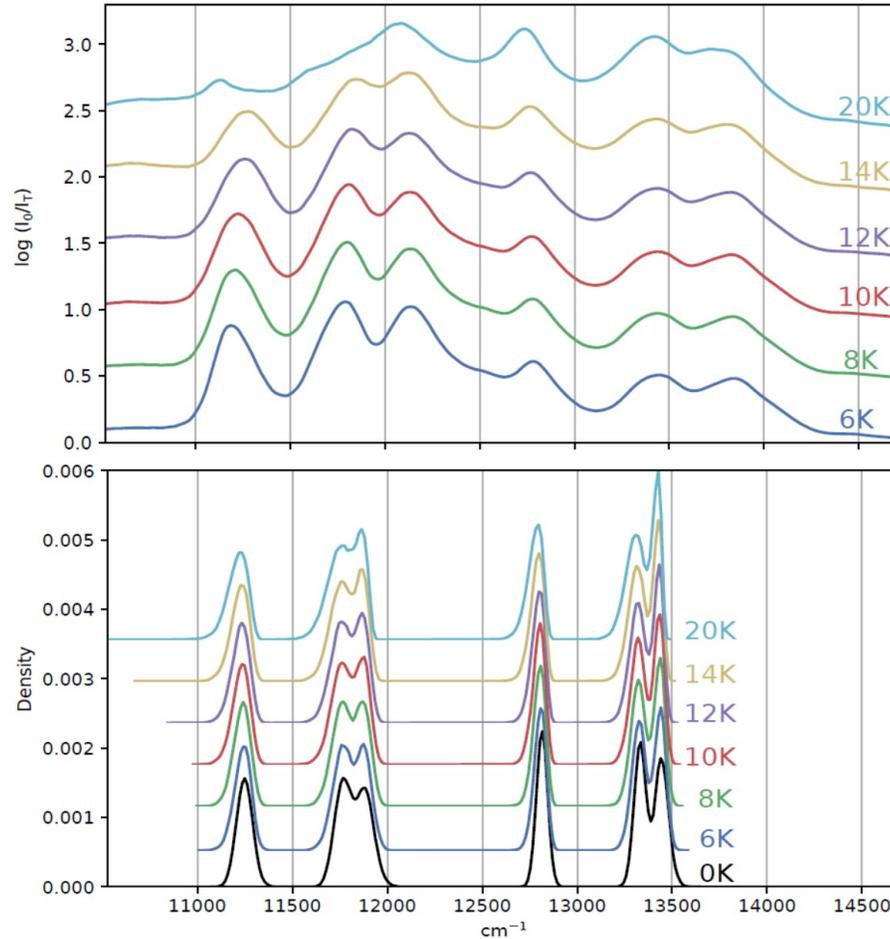


Semi-classical Franck-Condon approximation

Crystal field
1st order in excited state

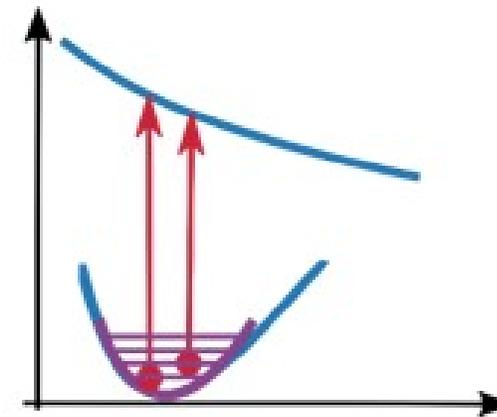


But incorrect shape (Reflexion approximation + Crystal Field theory)



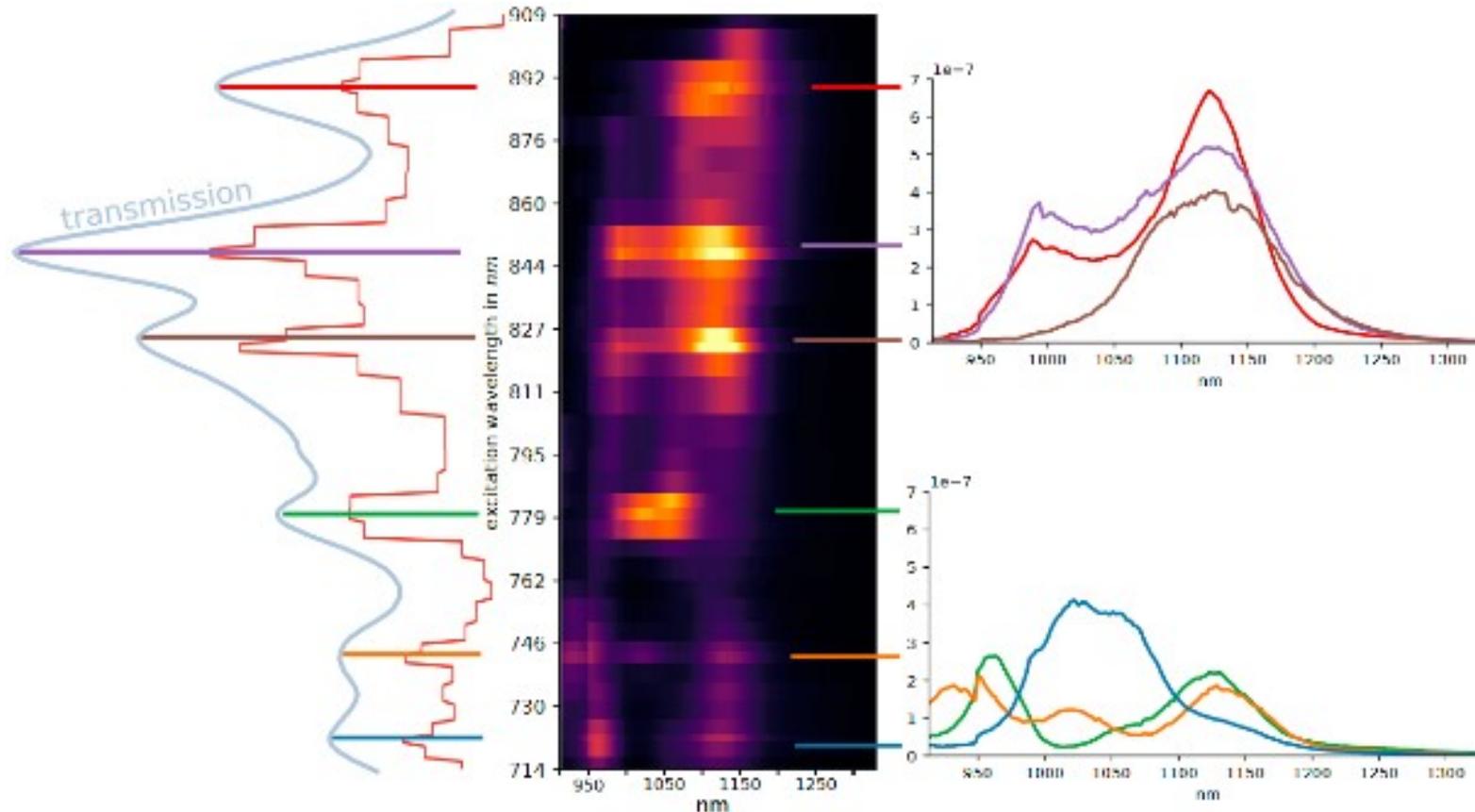
Experiment

Theory



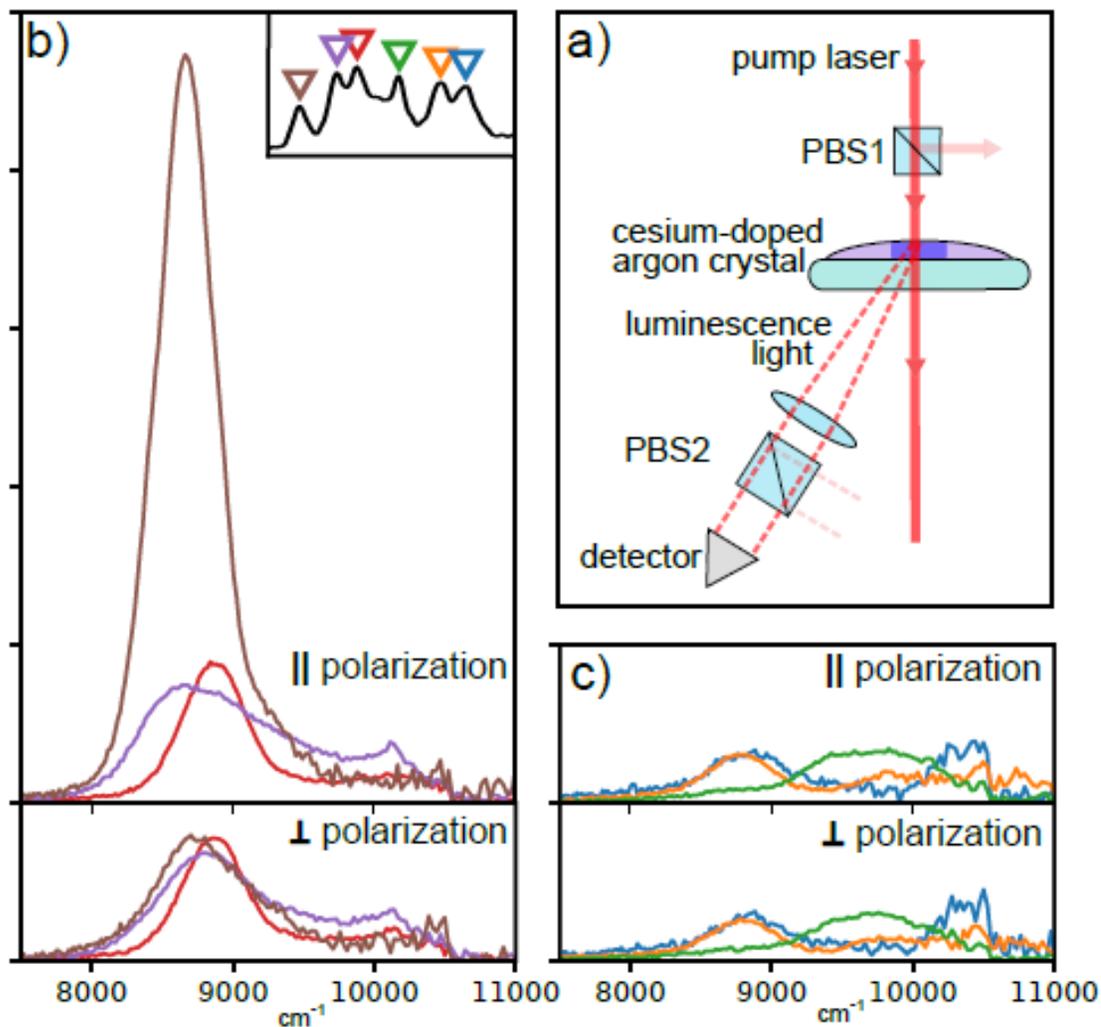
$$A(E) \propto \sum_i P_i \int |\Psi_i(\mathbf{Q})|^2 \delta[E - (V_e(\mathbf{Q}) - E_i)] d\mathbf{Q}$$

Next steps: Preliminary results on Fluorescence

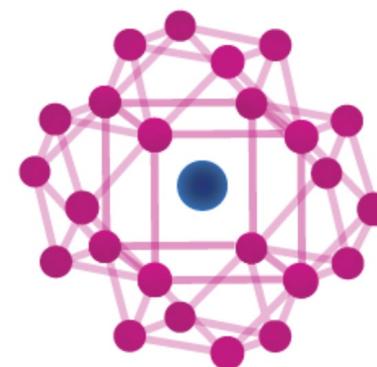
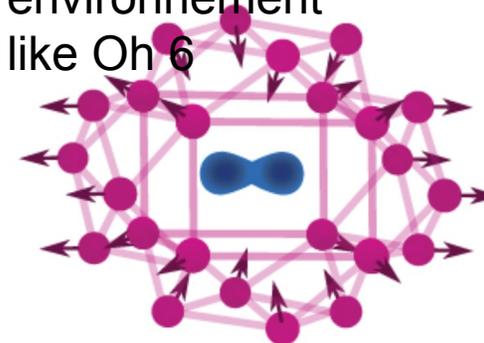


Unexpected (non) relaxation $p_{3/2}$ ☾
 $p_{1/2}$

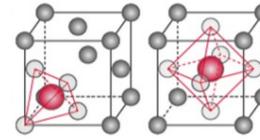
Preliminary results with polarized light



The reddest line might be $6s \rightarrow 6p_{1/2}$ excitation in a very symmetric environment like Oh 6



Conclusion



- Cs in Ar seems promising for EDM measurement below 10^{-30} e.cm ☾ new physics
- In solid state systems, there can be dangerous mixing terms.
- Absorption spectra shows that multiple trapping sites are present at the same time. **Is it Td (4 vacancies) + Oh (6 vacancies) ? What about hcp phase ?**

Next steps

More absorption (temperature dependence)

Fluorescence measurement

Temperature dependence

Magnetic circular dichroism

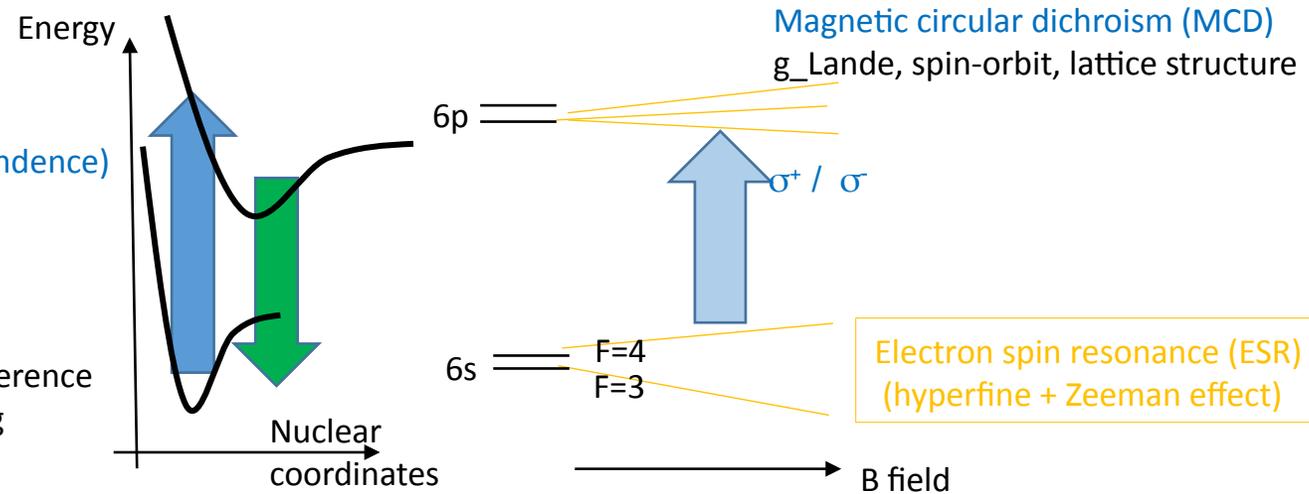
Optical pumping

Electron Spin Resonance ☾ Spin coherence

Magneto electric effects E/B coupling

Other atoms (Li, Na, K, Rb, Cs)

Other gaz (Kr, Xe, para-H₂)



P and *T* violating moments of Cs in cryogenic matrices.

• <https://cnrs.zoom.us/j/96208775835?pwd=GfpE5GcMiTpYfyACZKb2yB1vNXaddNov 3rd 3pm>

- Polarizabilities as probes for P, T, and PT violation : S. Lahs and D. Comparat 2024 *New J. Phys.* **26** 125001
- Atomic Observables Induced by Cosmic Fields : S. Lahs, D. Comparat, F. Kirk, B. Roberts arXiv:2510.08007

New observables in cristal vs gaz phase

Sébastien Lahs



$$V^{EM} = -\underset{\substack{\text{electric dipole}}}{\mathbf{d}} \cdot \boldsymbol{\mathcal{E}} - \underset{\substack{\text{magnetic dipole}}}{\boldsymbol{\mu}} \cdot \boldsymbol{\mathcal{H}} - \underset{\substack{\text{electric quadrupole}}}{Q_{ij}} \partial_i \mathcal{E}_j$$

$$d_{PT}^{\mu} \alpha_{ij}^{\ell} = \sum_{k,l} \frac{\langle n | V^{PT} | l \rangle \langle l | d_i | k \rangle \langle k | \mu_j | n \rangle}{(\omega_{kn}^2 - (\omega^{\ell})^2) \omega_{ln}} \omega_{kn} + \text{permutations}$$

P odd $\Delta E \propto \frac{dQ}{P} \alpha_{t_m} (\boldsymbol{\mathcal{H}} \cdot \hat{\mathbf{n}}) (\boldsymbol{\mathcal{E}} \times \boldsymbol{\mathcal{H}}) \cdot \hat{\mathbf{n}}$

T odd $\Delta E \propto \frac{\mu Q}{T} \alpha_{t_m} (\boldsymbol{\mathcal{E}} \cdot \hat{\mathbf{n}}) (\boldsymbol{\mathcal{H}} \times \boldsymbol{\mathcal{E}}) \cdot \hat{\mathbf{n}}$

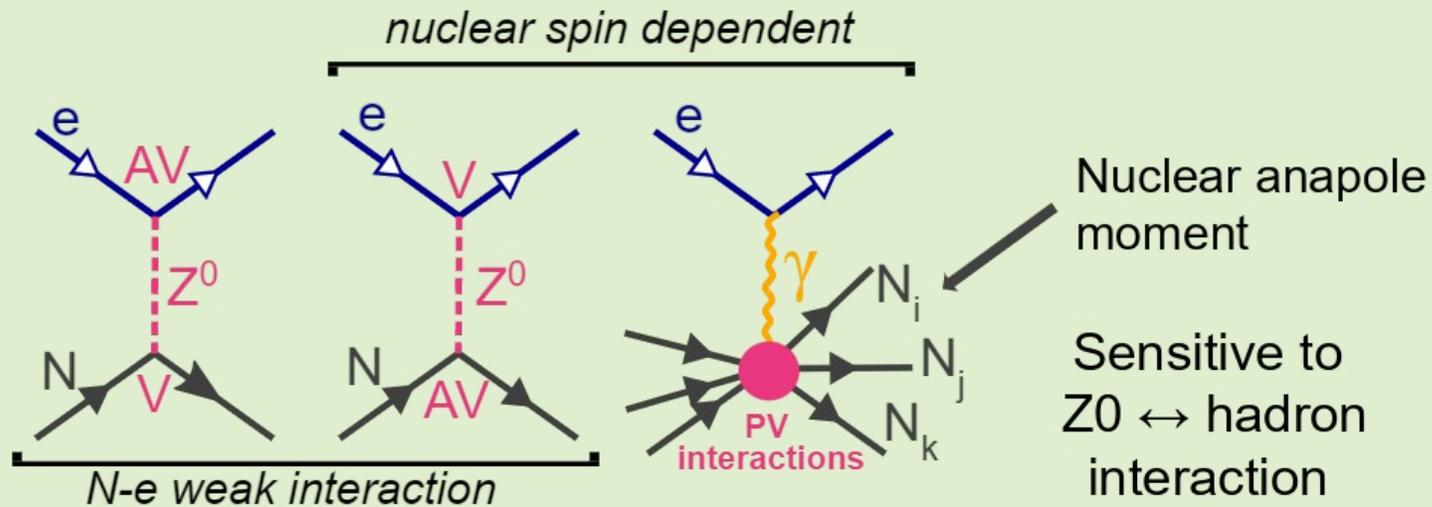
P&T odd $\Delta E \propto \frac{dQ}{PT} \alpha_{v_m} (\boldsymbol{\mathcal{E}} \cdot \boldsymbol{\mathcal{H}}) - (\boldsymbol{\mathcal{E}} \cdot \hat{\mathbf{n}}) (\boldsymbol{\mathcal{H}} \cdot \hat{\mathbf{n}})$



.Bouchiat, M & Bouchiat C.
Eur. Phys. J. D, 15.1 (2001): 5-18.

Nuclear Anapole Moment

1997 only successful measurement in Cs beam [2]



Bouchiat & Bouchiat:

Observable as a linear stark shift in cryogenic matrix [3]

$$\Delta E_{stark} \propto \frac{(\hat{n} \cdot \vec{B}) \hat{n} \cdot (\vec{E} \times \vec{B})}{B^2}$$

with symmetry
breaking crystal axis $\frac{\partial \epsilon}{\partial x_j}$