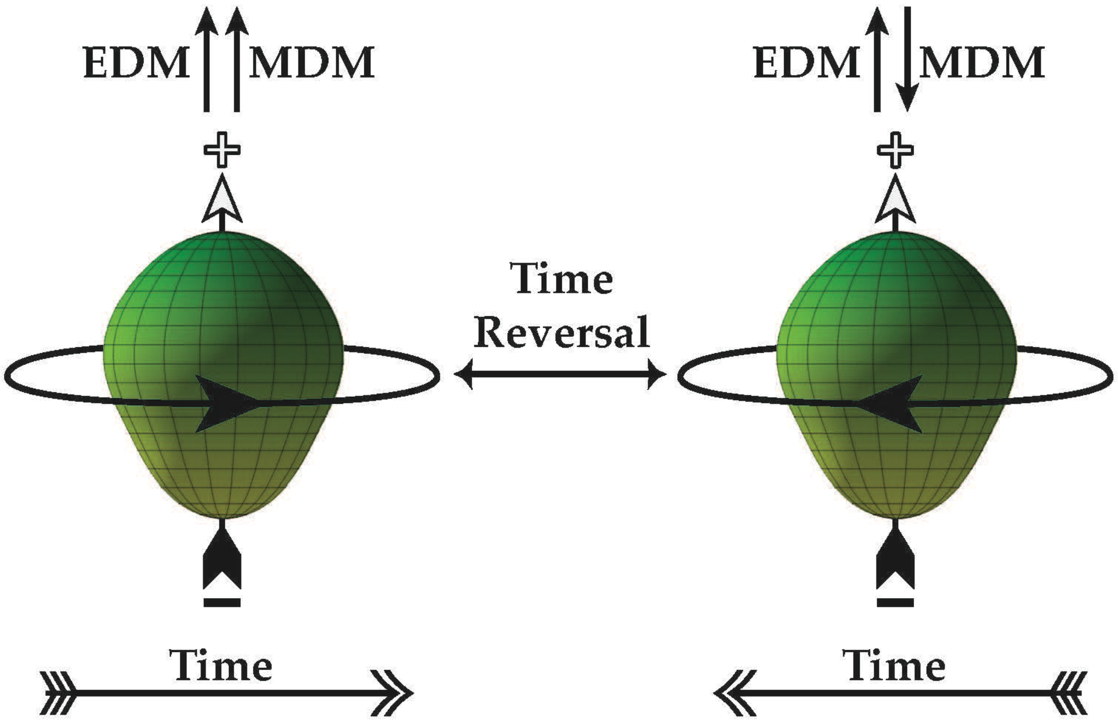
FRIB Updates on Center-Level Funding, Pa-229, and Radioactive Molecules in Solids



Lise Meitner

Jaideep Taggart Singh (he/him/his)

Facility for Rare Isotope Beams / MSU

EDM Workshop 2025 (Caltech & TDLI)

Resnick Sustainability Center rooms 110 and 120

https://caltech.zoom.us/j/88942455905?pwd=Wfebb93TbVyUMLcinAbczJbxt80hcl.1

Monday May 12, 2025 @ 15:45 to 16:30 (PST)

FRIB

SCAN ME

Main Take Home Message: Let's Work Together!

Exhortation: The appearance of inefficiently duplicating efforts at multiple facilities runs some risk under the present funding context.

FRIB is eager to proactively support the needs of the FS community in places where it may be able to play a unique role:

1. Offline access to some interesting isotopes (^{225}Ra, ^{229}Pa, others?) by direct production via the Isotope Harvesting program.
2. General collaborative support for radiochemistry for anyone anywhere
3. People (searching for an FS staff scientist now- see **QR Code**), space, and capital equipment for FS experiments physically hosted at FRIB
4. Plans for a general-purpose beamline that utilizes harvested isotopes (US NSF MRI)
5. Administrative/logistical/organizational support for Center-level effort to help provide a "home-base" within the US context
6. DOE Office of Science has requested a FRIB Director's Review.

FRIB PIs: Jaideep Taggart Singh (Spinlab), Xing Wu (QUEST)
Shane Wilkins (starting July 1)

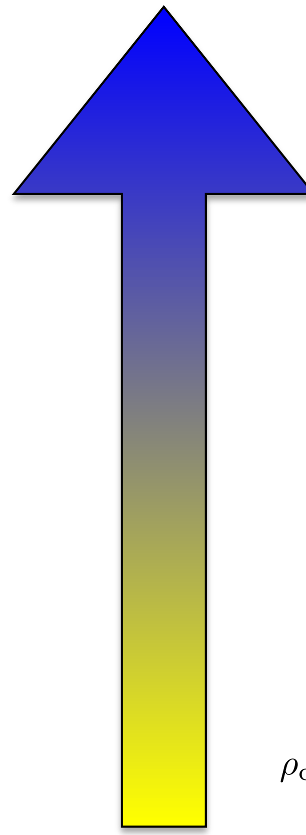
- Atomic ^{225}Ra EDM in a laser trap
- Polar molecules trapped in noble gas solids (EDM-cubed)
- ^{229}Pa nuclear structure measurements
- ^{229}Pa ions implanted in optical crystals/diamonds
- Centrifuge slowing of cold atoms and molecules
- Optical cycling properties of interesting molecules
- Cold sources for radioactive molecules

FS Scientist 959065

Electric Dipole Moment (EDM): Measures the Separation of Charges

$$\vec{d} = \int \rho_{\text{charge}} \left(\vec{r} - \vec{R}_{\text{CM}}\right) \, d^3r = \langle \rho_{\text{charge}} \vec{r} \rangle - Q\vec{R}_{\text{CM}}$$

"Thunder Cloud as Generator #2" (1971) by Paterson Ewen [Art Gallery of Ontario]



$$\vec{d}$$

ρ_{charge} = charge distribution

$\vec{r}$ = position vector

$\vec{R}_{\text{CM}}$ = center of mass

Q = net charge

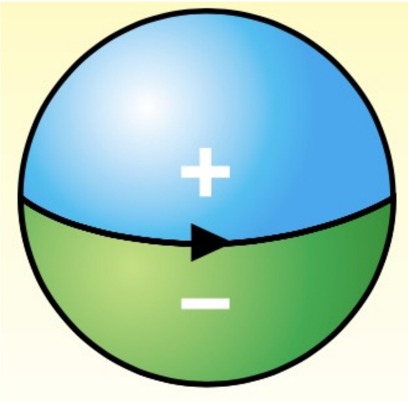
2023 EDM Limits:
“Free” of Standard Model (SM) “Backgrounds”

Chupp, Fierlinger, Ramsey-Musolf, JTS RMP 91:015001 (2019) & Nature 562:355 (2018)
& PRL 124:081803 (2020) & PRL 129:231801 (2022) & Science 381:46 (2023)

System	Best Limit (95%) 1E-28 *e* cm	SM estimate 1E-28 *e* cm	Method (Location)
Neutron	220	$\sim 10^{-4}$	ultracold neutrons in a bottle (PSI)
“Electron”	0.11 0.05	$\sim 10^{-7}$	cold ThO beam (JHU/UC/Harvard/Northwestern) trapped HfF^{+} (JILA/Boulder)
^{199}Hg	0.074	$\sim 10^{-6}$	atoms in vapor cell (UW-Seattle)

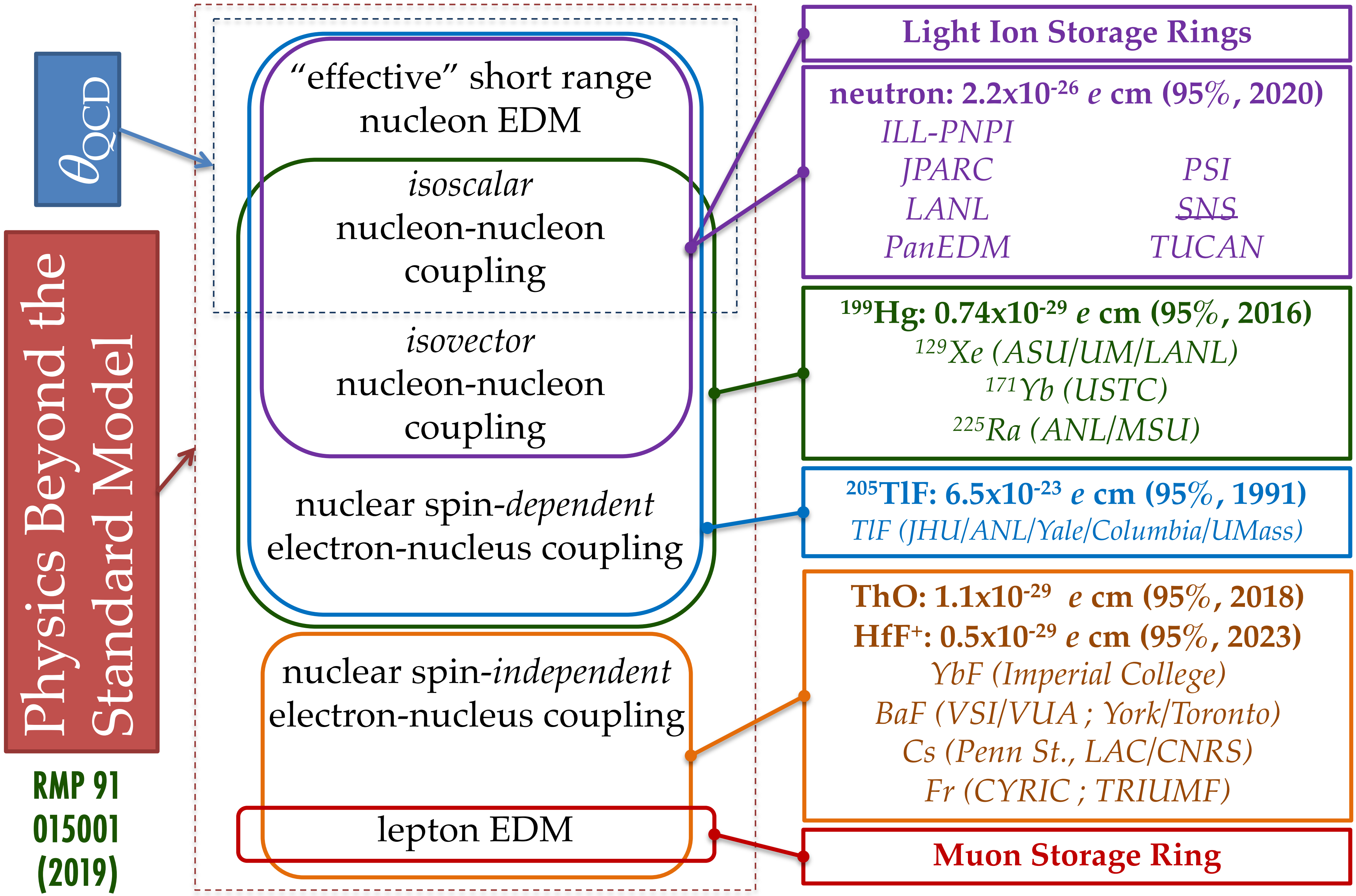
Imagine a ^{199}Hg atom that is composed of two oppositely charged hemispherical shells each with charge magnitude *e*…

…if the ^{199}Hg atom was the size of the Earth, then the maximum thickness of these shells would be less than the diameter of a strand of human hair.



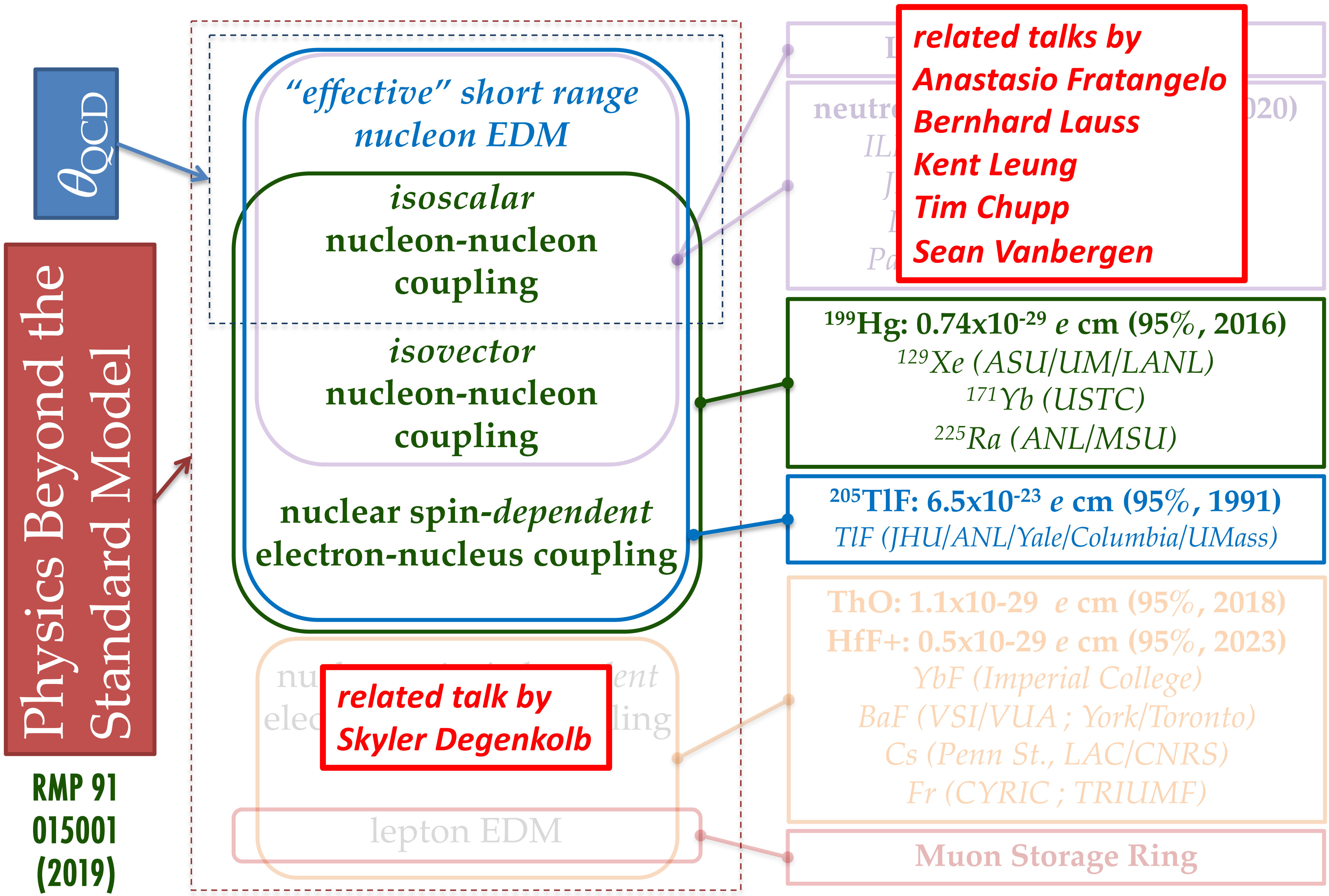
Physics Today, June 2003

Different Sources of $\not{T}$ in EDMs of Different Systems

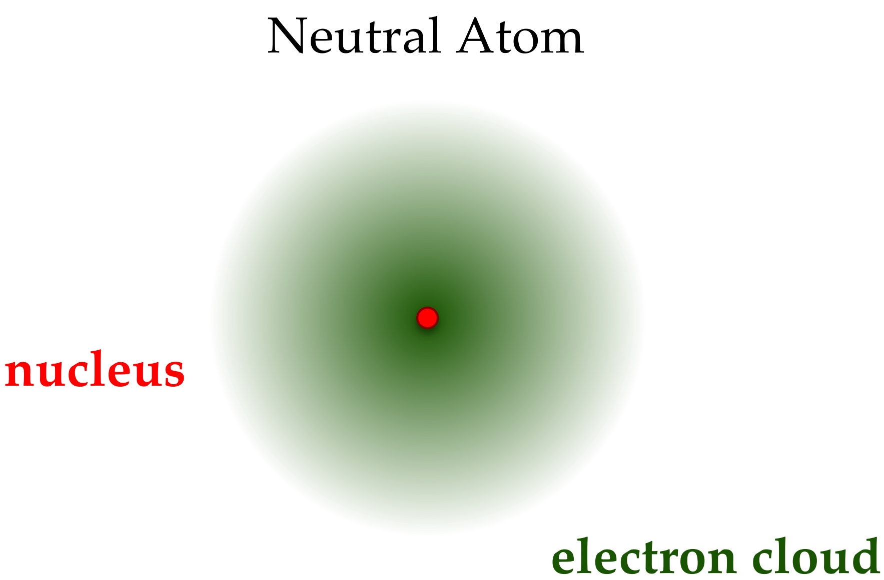


RMP 91 015001 (2019)

Different Sources of $\not{T}$ in EDMs of Different Systems



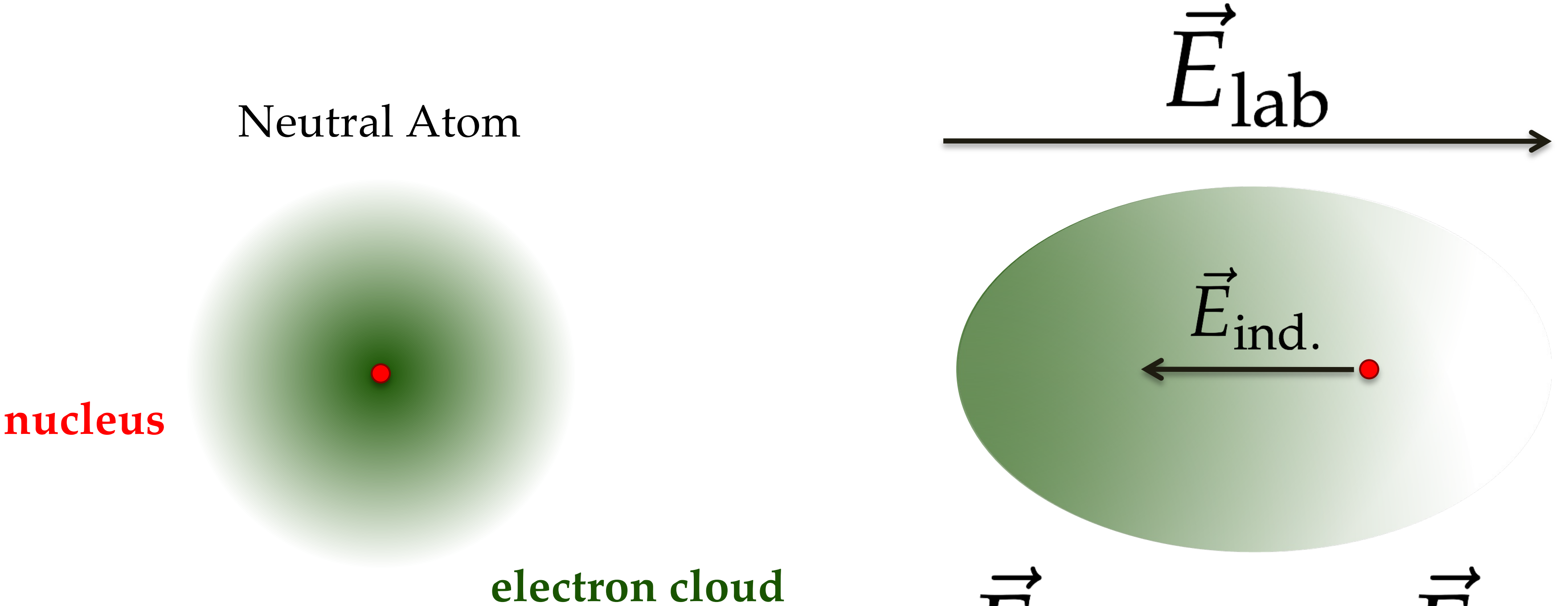
Diamagnetic Atoms: All Electrons Are Paired



Schiff Shielding in Diamagnetic Atoms

- **Shielding in Diamagnetic Atoms**

  Schiff PR 132:2194 (1963)



$$\vec{E}_{\mathrm{ind.}} \approx -\vec{E}_{\mathrm{lab}}$$

Shielding Imperfect in Relativistic Atoms With Nonzero Nuclear Size

- **Shielding in Diamagnetic Atoms**
  Schiff PR 132:2194 (1963)

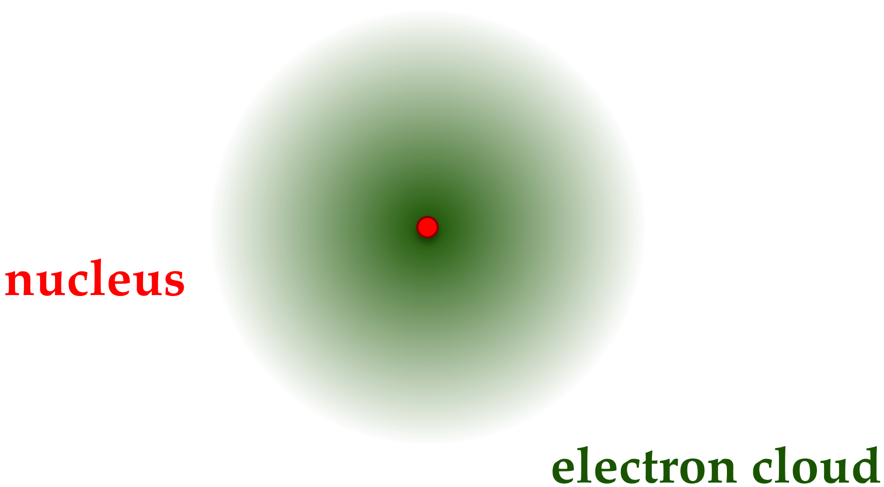
- **Relativistic atoms: The Sandars-Bouchiat Z^3 "Law"**
  Physics Letters 22:290 (1966) & Physics Letters 48B:111 (1974)

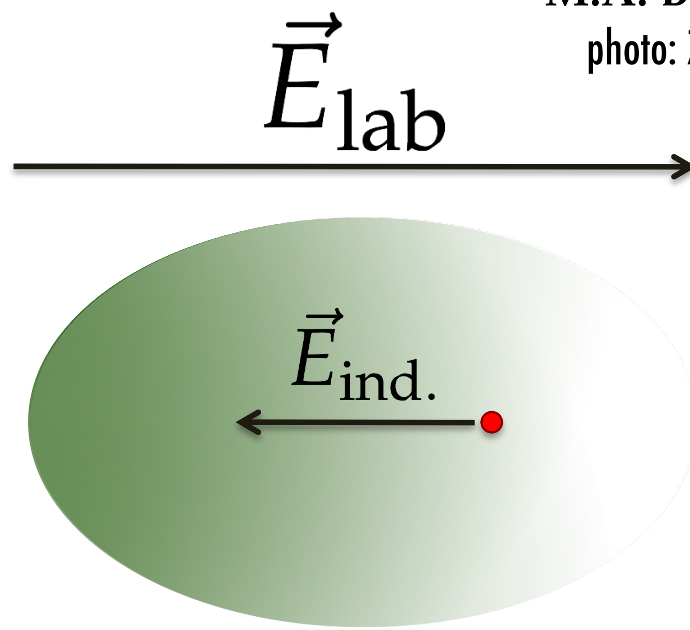
- **^{225}Ra vs ^{199}Hg vs. ^{3}He: 2.8 to 1 to 10^{-5}**
  JPB:AMOP 53:195004 (2020) & Phys. Rev. A 106, 022817 (2022)

Madame Professor Marie-Anne Bouchiat

M.A. Bouchiat
photo: Zolotrev

Neutral Atom





$$\vec{E}_{\text{ind.}} \approx -\vec{E}_{\text{lab}}$$

Residual Ᵽ & Ŧ Observable: Nuclear Schiff Moment

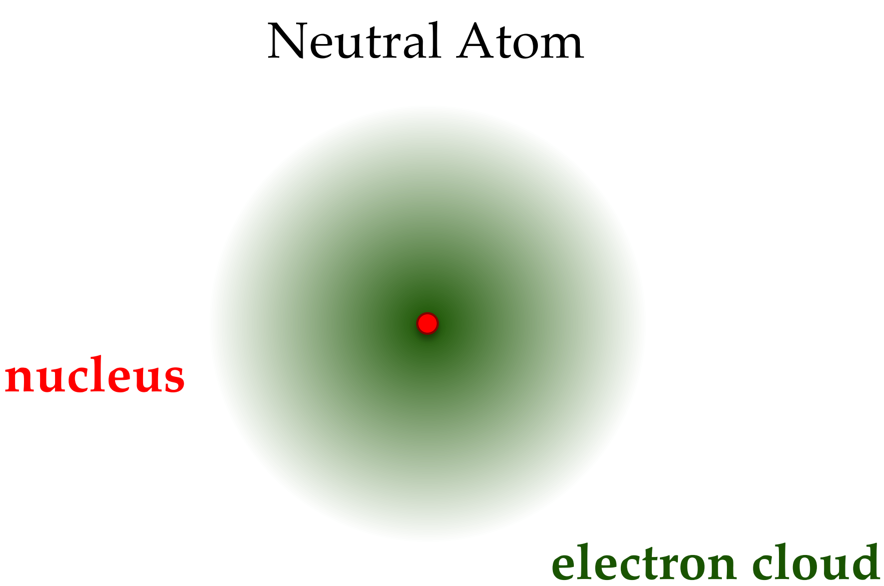
- **Shielding in Diamagnetic Atoms**
  Schiff PR 132:2194 (1963)
- **Relativistic atoms: The Sandars-Bouchiat Z^3 "Law"**
  Physics Letters 22:290 (1966) & Physics Letters 48B:111 (1974)
- **^{225}Ra vs ^{199}Hg vs. ^{3}He: 2.8 to 1 to 10^{-5}**
  JPB:AMOP 53:195004 (2020) & Phys. Rev. A 106, 022817 (2022)

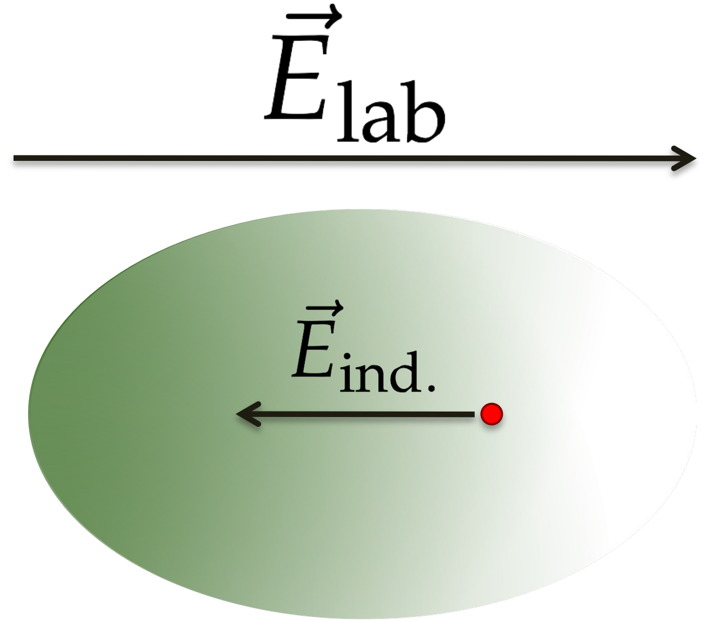
$$\vec{d}_{\mathrm{atom}} = \kappa_{\mathrm{atom}} Z^3 \vec{S}$$

Schiff Moment

$$\vec{S} = \frac{\langle e r^2 \vec{r} \rangle}{10} - \frac{\langle r^2 \rangle \langle e \vec{r} \rangle}{6}$$

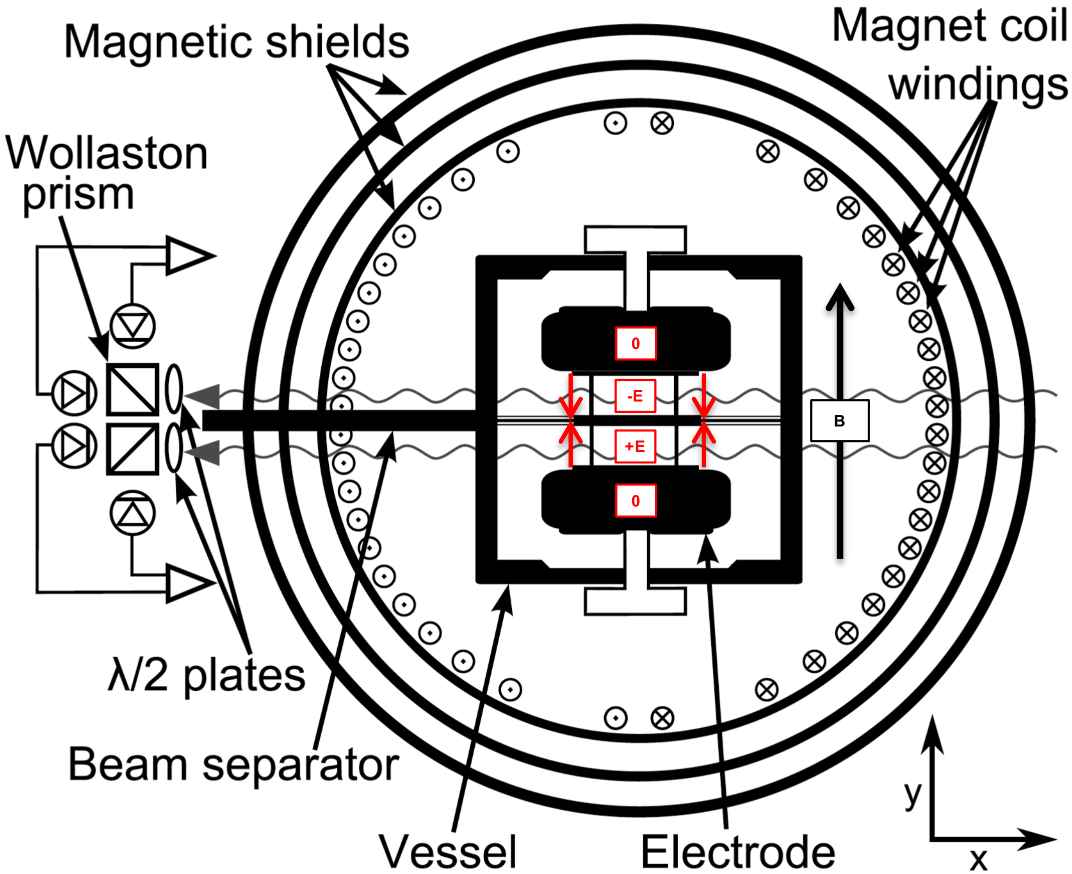
Zh. Eksp. Teor. Fiz. 87, 1521-1540 (1984)





$$\vec{E}_{\mathrm{ind.}} \approx -\vec{E}_{\mathrm{lab}}$$

2016: Atomic EDM of ^{199}Hg (Stable) In A Vapor Cell
The Gold Standard For Over 40 Years!



- **diamagnetic**, $^1S_0$ ground state
- ***I* = ½**, no elect. quad. moment
- **high Z**, (80) rel. atomic struct.
- **stable**, (17% n.a.) 92% enriched
- **high vapor pressure**, (10^{13}/cm^3)
- **modest electric field,** 10 kV/cm
- **40+ year old experiment!**

Limiting systematic appears to be ~10 nm scale motion of vapor cells when HV is switched in the presence of 2nd order *B*-field gradients.

$$\nu = 8.3 \text{ Hz}$$

$$\Delta\nu \leq 0.1 \text{ nHz}$$

The best limit on atomic EDM:

EDM(^{199}Hg) < 0.74×10^{-29} *e*-cm (95% C.L.)

Graner et al., PRL 116:161601 (2016)

Ᵽ & Ŧ Physics: First Order Perturbation Theory

$$S_z = \frac{\langle er^2 z \rangle}{10} - \frac{\langle r^2 \rangle \langle ez \rangle}{6}$$

$$S \equiv \langle \Psi_0 | S_z | \Psi_0 \rangle = \sum_{k \neq 0} \frac{\langle \Psi_0 | S_z | \Psi_k \rangle \langle \Psi_k | V_{\text{ⱣŦ}} | \Psi_0 \rangle}{E_0 - E_k} + \text{c.c.}$$

- The Ᵽ and Ŧ physics that we seek (unknown & common to all isotopes)

Isotopes With Nearly Degenerate Nuclear States

$$S_z = \frac{\langle e r^2 z \rangle}{10} - \frac{\langle r^2 \rangle \langle e z \rangle}{6}$$

$$S \equiv \langle \Psi_0 | S_z | \Psi_0 \rangle = \sum_{k \neq 0} \frac{\langle \Psi_0 | S_z | \Psi_k \rangle \langle \Psi_k | V_{\not{P}\not{T}} | \Psi_0 \rangle}{E_0 - E_k} + \text{c.c.}$$

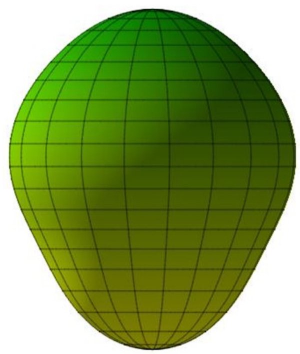
- Difference in lab-frame nuclear energy levels
- The $\not{P}$ and $\not{T}$ physics that we seek (unknown & common to all isotopes)

Nuclear Schiff Moment in the Lab Frame

$$S_z = \frac{\langle er^2 z\rangle}{10} - \frac{\langle r^2\rangle \langle ez\rangle}{6}$$

$$S \equiv \langle \Psi_0 | S_z | \Psi_0 \rangle = \sum_{k\neq 0} \frac{\langle \Psi_0 | S_z | \Psi_k \rangle \langle \Psi_k | V_{\not{P}\not{T}} | \Psi_0 \rangle}{E_0 - E_k} + \text{c.c.}$$

- Body-frame Schiff moment – large when there are intrinsic nuclear deformations
- Difference in lab-frame nuclear energy levels
- The $\not{P}$ and $\not{T}$ physics that we seek (unknown & common to all isotopes)



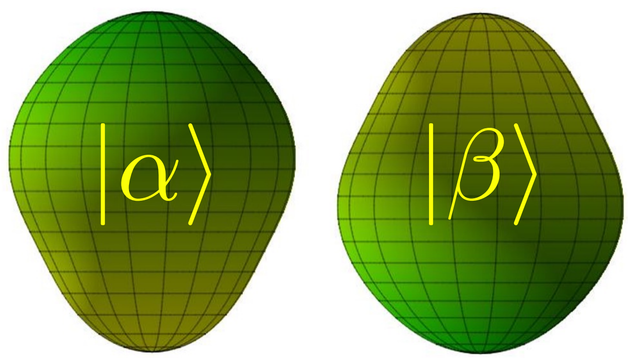
Example of large intrinsic Schiff moment: Pear-shaped nucleus in the "body-frame"

Pear-Shaped Nuclei = Nearly Degenerate Parity Doublets

$$S_z = \frac{\langle e r^2 z \rangle}{10} - \frac{\langle r^2 \rangle \langle e z \rangle}{6}$$

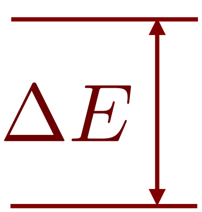
$$S \equiv \langle \Psi_0 | \, S_z \, | \Psi_0 \rangle = \sum_{k \neq 0} \frac{\langle \Psi_0 | \, S_z \, | \Psi_k \rangle \langle \Psi_k | \, V_{\not{PT}} \, | \Psi_0 \rangle}{E_0 - E_k} + \text{c.c.}$$

Parity Doublet



- **Nearly degenerate parity doublet**

Haxton & Henley PRL 51:1937 (1983)



$$|\Psi_1\rangle = \frac{|\alpha\rangle \mp |\beta\rangle}{\sqrt{2}}$$

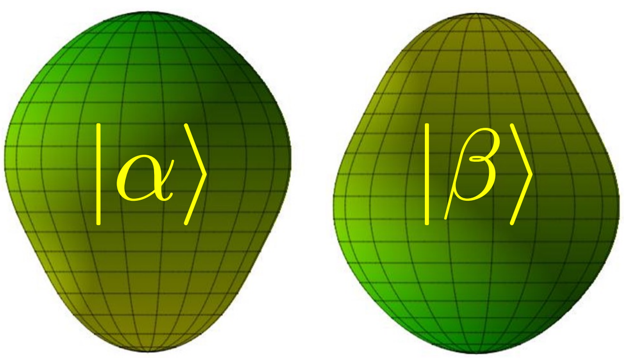
$$|\Psi_0\rangle = \frac{|\alpha\rangle \pm |\beta\rangle}{\sqrt{2}}$$

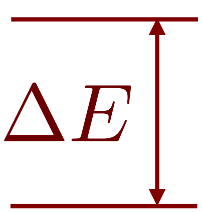
Pear-Shaped Nuclei = Enhanced Intrinsic Schiff Moments

$$S_z = \frac{\langle er^2 z\rangle}{10} - \frac{\langle r^2\rangle\langle ez\rangle}{6}$$

$$S \equiv \langle\Psi_0|\, S_z\, |\Psi_0\rangle = \sum_{k\neq 0} \frac{\langle\Psi_0|\, S_z\, |\Psi_k\rangle \langle\Psi_k|\, V_{\not{P}\not{T}}\, |\Psi_0\rangle}{E_0 - E_k} + \text{c.c.}$$

Parity Doublet





$$|\Psi_1\rangle = \frac{|\alpha\rangle \mp |\beta\rangle}{\sqrt{2}}$$

$$|\Psi_0\rangle = \frac{|\alpha\rangle \pm |\beta\rangle}{\sqrt{2}}$$

- **Nearly degenerate parity doublet**

  Haxton & Henley PRL 51:1937 (1983)

- **Large intrinsic Schiff moment due to octupole deformation**

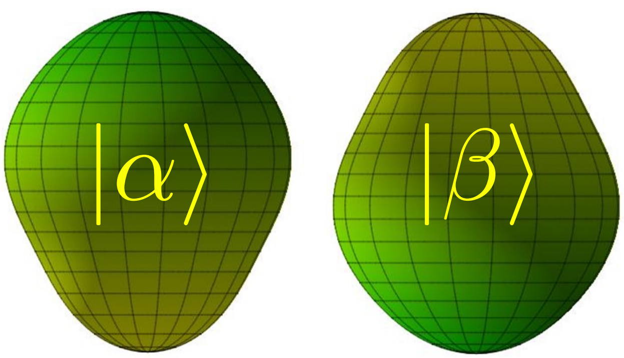
  Auerbach, Flambaum, & Spevak PRL 76:4316 (1996)

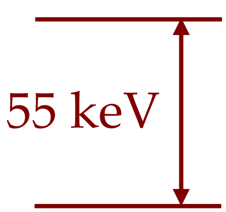
Example: Enhanced Sensitivity in Radium-225

$$S_z = \frac{\langle er^2 z \rangle}{10} - \frac{\langle r^2 \rangle \langle ez \rangle}{6}$$

$$S \equiv \langle \Psi_0 | S_z | \Psi_0 \rangle = \sum_{k \neq 0} \frac{\langle \Psi_0 | S_z | \Psi_k \rangle \langle \Psi_k | V_{\not{P}\not{T}} | \Psi_0 \rangle}{E_0 - E_k} + \text{c.c.}$$

Parity Doublet





$$|\Psi_1\rangle = \frac{|\alpha\rangle \mp |\beta\rangle}{\sqrt{2}}$$

$$|\Psi_0\rangle = \frac{|\alpha\rangle \pm |\beta\rangle}{\sqrt{2}}$$

- **Nearly degenerate parity doublet**

  Haxton & Henley PRL 51:1937 (1983)

- **Large intrinsic Schiff moment due to octupole deformation**

  Auerbach, Flambaum, & Spevak PRL 76:4316 (1996)

Total Enhancement Factor: EDM (^{225}Ra) / EDM (^{199}Hg)

Skyrme Model	Isoscalar	Isovector
SIII	300	4000
SkM*	300	2000
SLy4	700	9000

^{225}Ra: Dobaczewski & Engel PRL 94:232502 (2005)
^{199}Hg: Ban et al. PRC 82:015501 (2010)

Nuclear Structure Calculations Are Challenging!

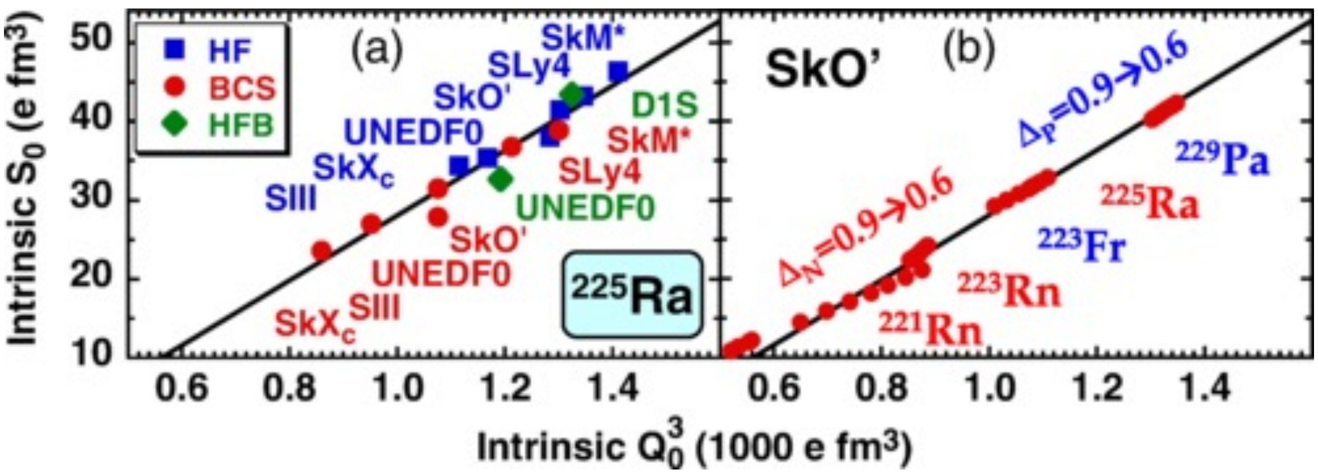
type	^{199}Hg	^{225}Ra	ratio*2.8	references
SIII	0.005	7.0	4300	PRC 82 015501 (2010)
SkM*	-0.027	21.5	-2400	PRC 82 015501 (2010)
SLy4	-0.006	16.9	-8600	PRC 82 015501 (2010)
SkO′		6.0		
DE05	0.071			PRC 72 045503 (2005)
DS03	0.055			PAN 66 1940 (2003)
"Best"	+/-(0.02)	6.0	+/-(900)?	Prog. PNP 71 21 (2013)

related talk by Jon Engel

- Isovector coupling is given by "chromo"-EDMs
- Nuclei are the most sensitive to this source of new physics
- Opportunity for ^{225}Ra or other octupole deformed species

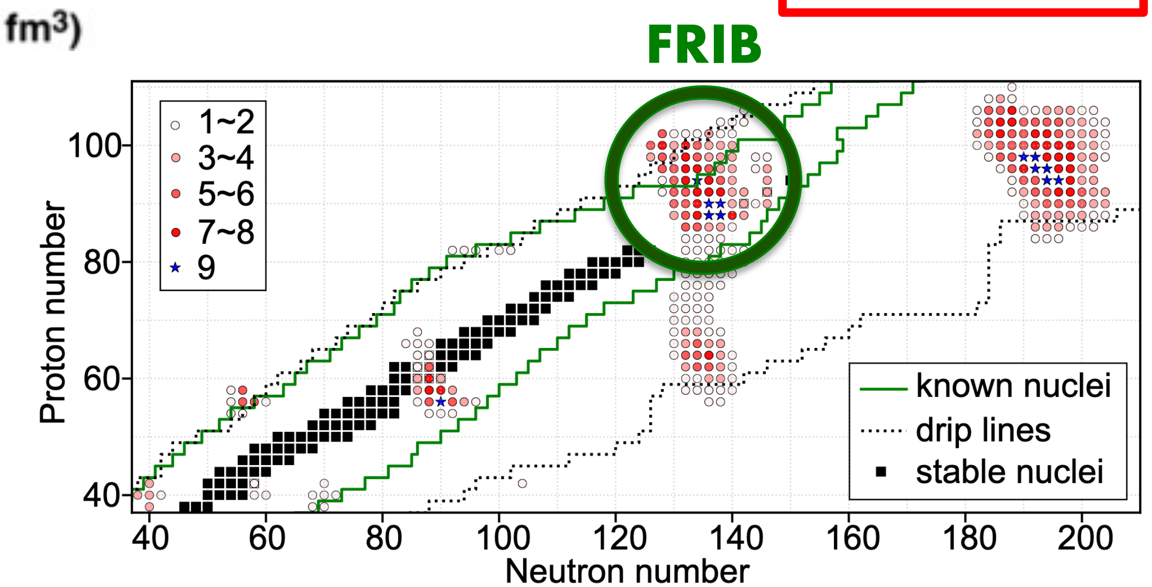
Calibrating the Intrinsic Schiff Moment

$$S \equiv \langle \Psi_0 | S_z | \Psi_0 \rangle = \sum_{k \neq 0} \frac{\langle \Psi_0 | S_z | \Psi_k \rangle \langle \Psi_k | V_{PT} | \Psi_0 \rangle}{E_0 - E_k} + \text{c.c.}$$



PRL 121, 232501 (2018)
Phys. Rev. C, 102:024311 (2020)

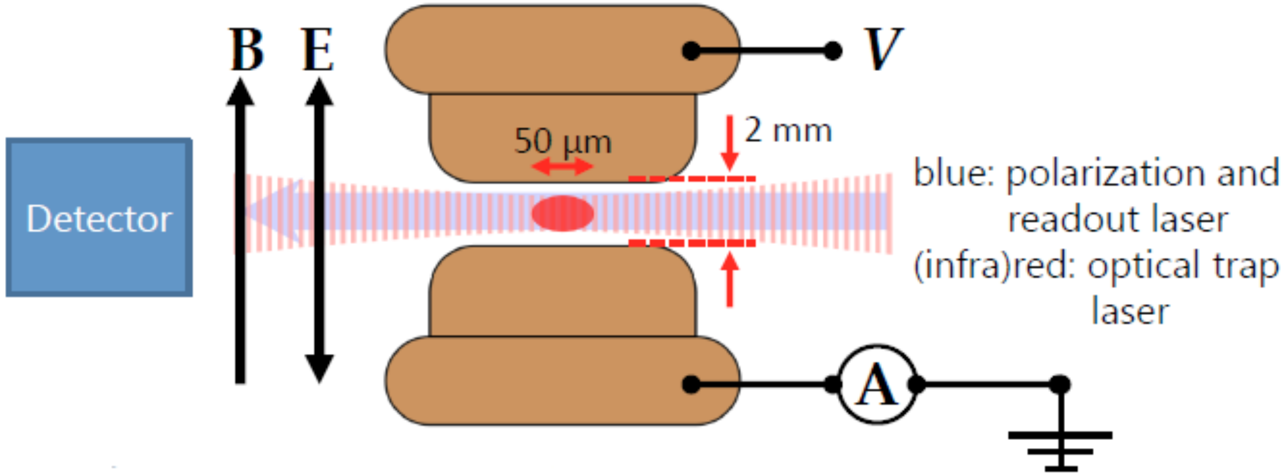
related talk by Jon Engel



Nuclear structure measurements combined with nuclear theory can calibrate the new physics sensitivity of "enhancer" isotopes with uncertainty quantification!

Ongoing: The Atomic EDM of ^{225}Ra at Argonne

$|d(^{225}Ra)| < 50 \times 10^{-23}$ *e*-cm (95%)
PRL 114:233002 (2015)
$|d(^{225}Ra)| < 1.4 \times 10^{-23}$ *e*-cm (95%)
equivalent to ~1000xEDM(^{199}Hg)
PRC 94:025501 (2016)
Upgrades underway to improve sensitivity by x1000
Spectrochimica Acta Part B 172 105967 (2020)



EDM search using atoms held in Optical Lattice

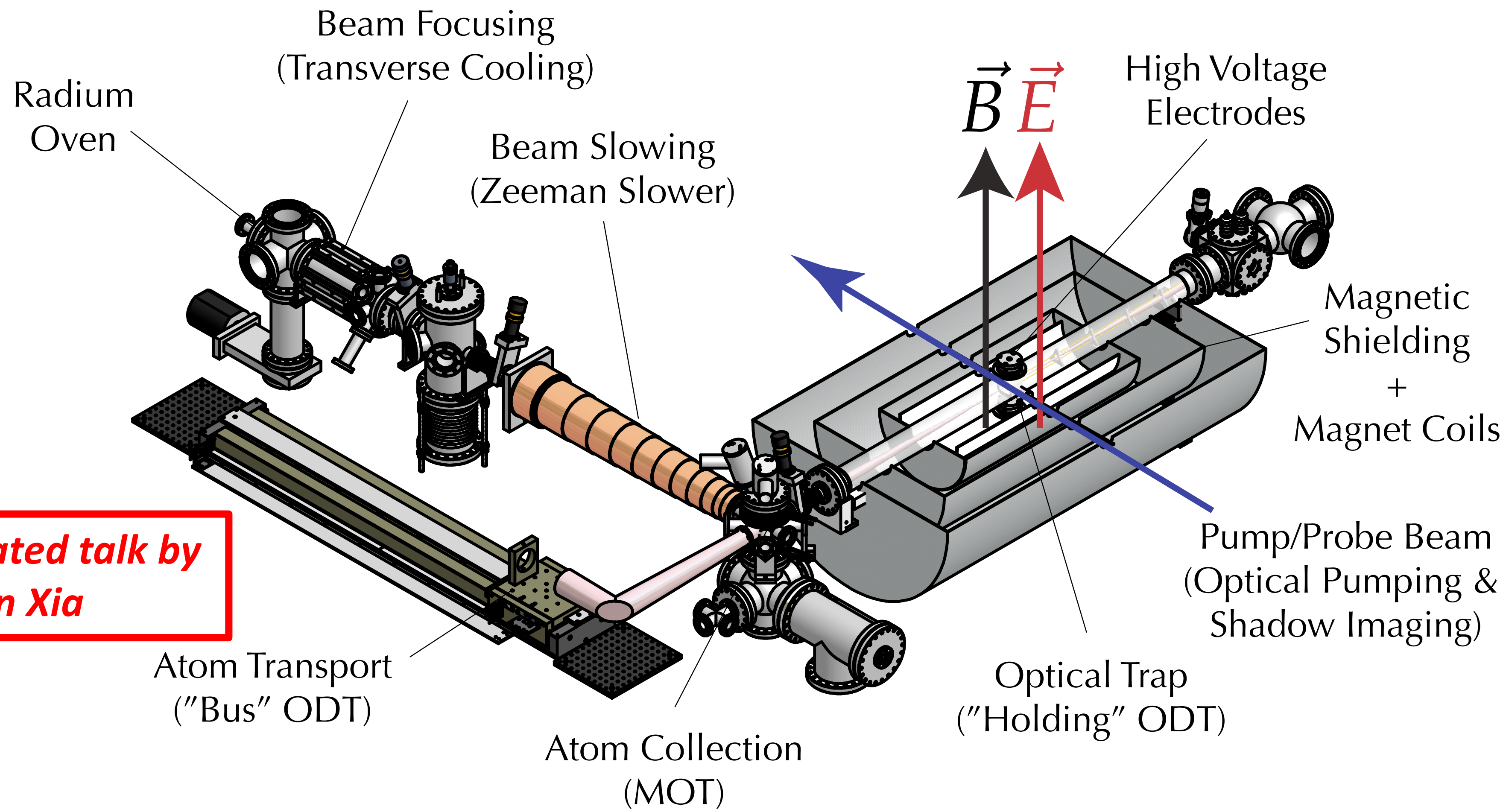
Romalis & Fortson PRA 59:4547 (1999)
Chin et al. PRA 63:033401 (2001)
Bishof et al. PRC 94:025501 (2016)

^{226}Ra
nuclear spin = 0
$t_{1/2}$ = 1600 years
Low vapor pressure

^{225}Ra
Nuclear Spin = ½
$t_{1/2}$ = 15 days
Low vapor pressure

- Atoms concentrated in a very small region
- Long coherence time (100 s) PRL 129, 083001 (2022)
- negligible "v x E" systematics
- High electric field (>300 kV/cm) in vacuum NIMA 1014 165738 (2021)
- Light-induced systematic effects can be controlled!

Status: Three Key Upgrades Being Developed Now



related talk by Tian Xia

PRC 94:025501

|d(Ra-225)| < 1.4×10^{-23} *e* cm (95%)

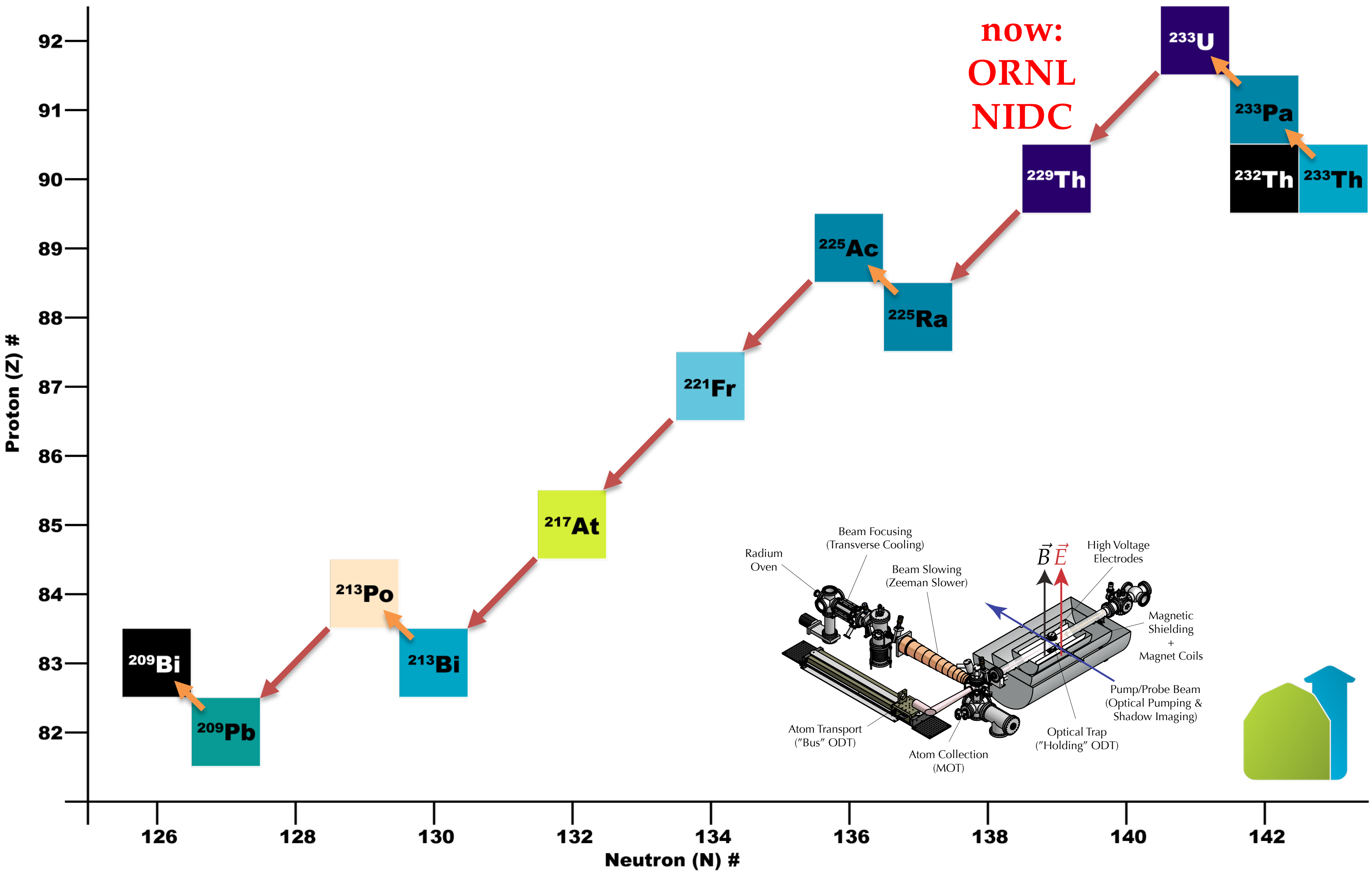
completely statistics limited

several upgrades underway

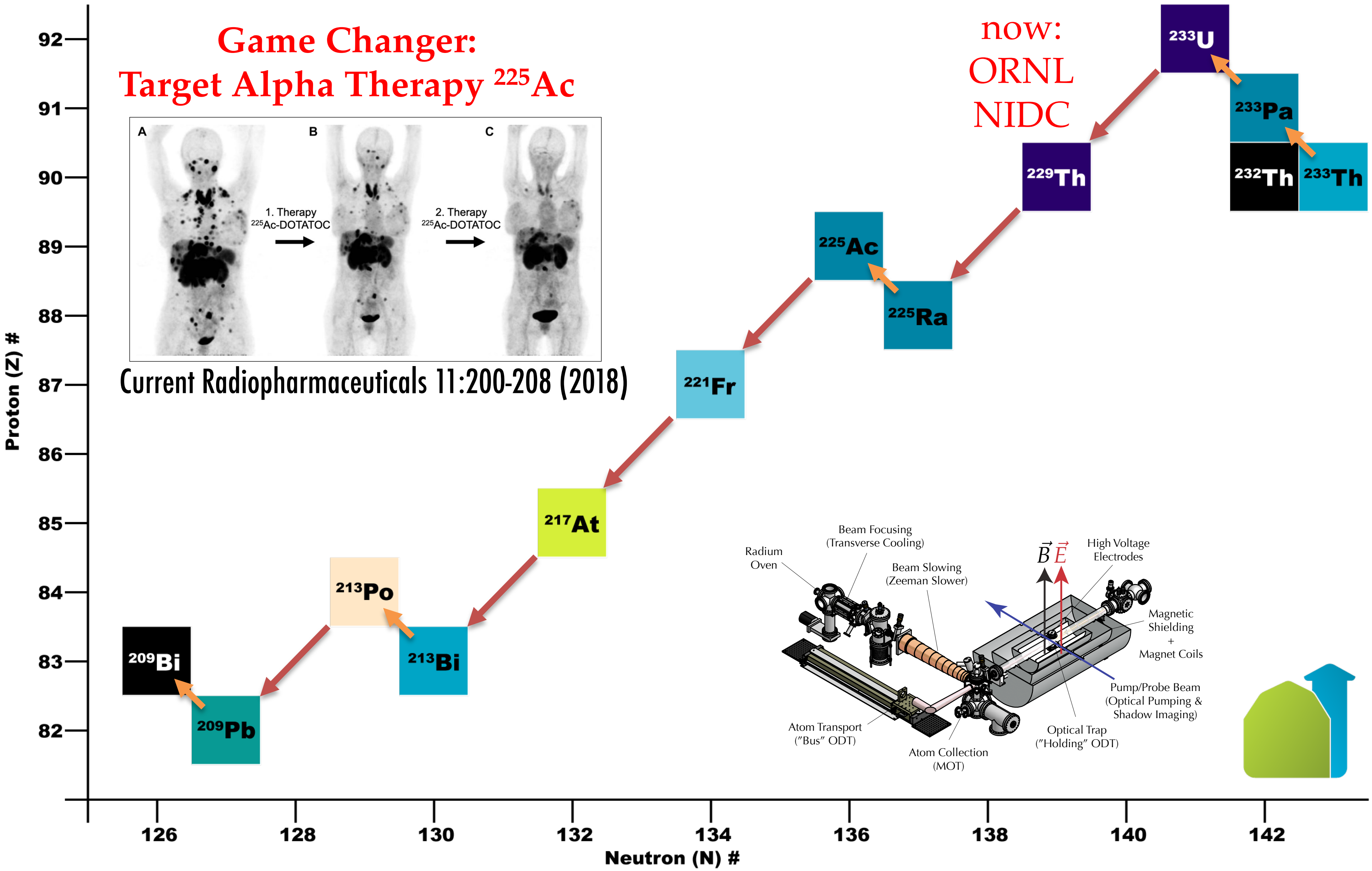
Gordon Arrowsmith-Kron (PhD defended):

1. **atom slowing/trapping efficiency**
2. **more exact reversal of higher *E*-field**
3. **more efficient detection scheme**

Source of Radium-225 Atoms ($\tau_{1/2}$ = 15 days)



~~Source of Radium-225 Atoms ($\tau_{1/2}$ = 15 days)~~



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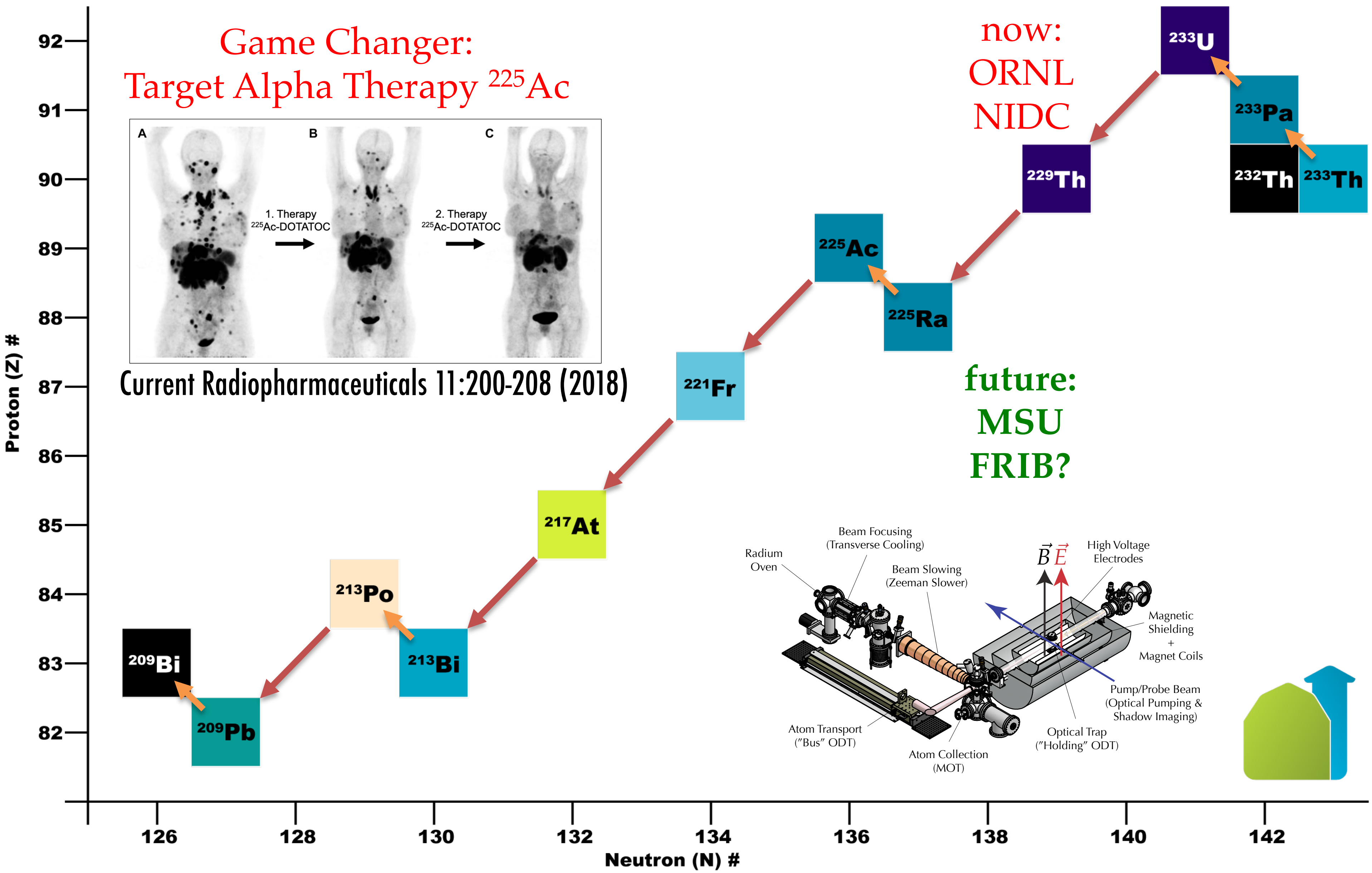


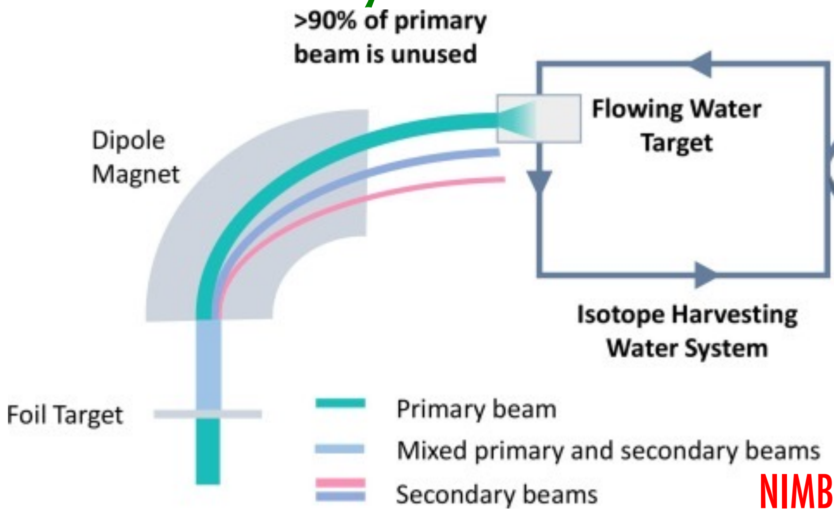
Facility for Rare Isotope Beams @ MSU

Michigan State University
East Lansing, MI
Very Bad at American Football
Home of FRIB

University of Michigan
Ann Arbor, MI
Very Good at American Football
no FRIB

Google Maps & Wikipedia Commons

"Isotope Harvesting" at
The Facility for Rare Isotope Beams (MSU/East Lansing)



NIMB 478 34 (2020)

This is funded and on its way!

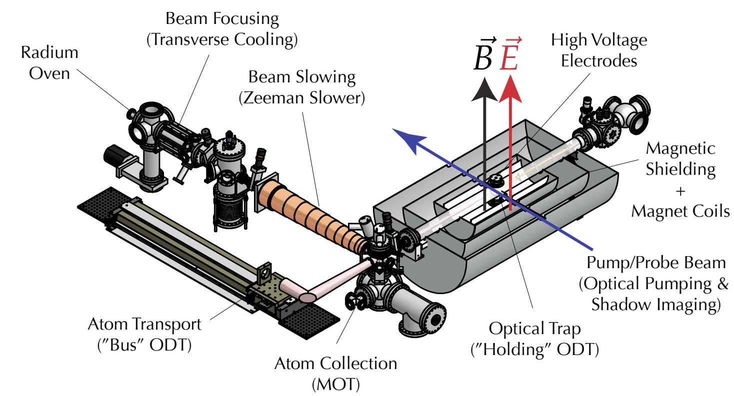
Adobe Stock

Isotope harvesting at FRIB: additional opportunities for scientific discovery [J. Phys. G: Nucl. Part. Phys. 46 100501 (2019)]

Adobe Stock

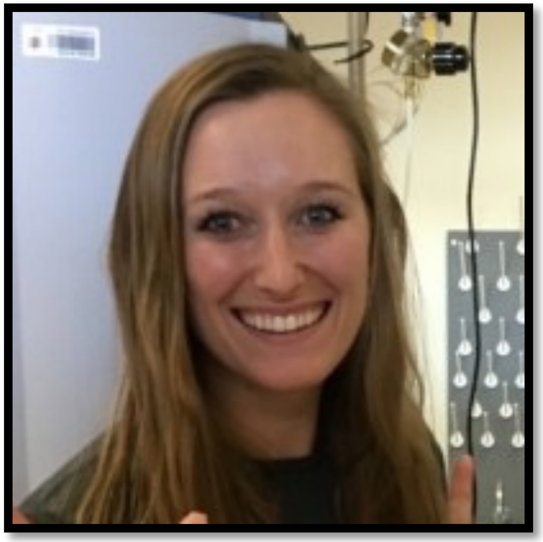
This needs support and we should work together:

^{225}RaF/^{225}RaO
^{227}ThO



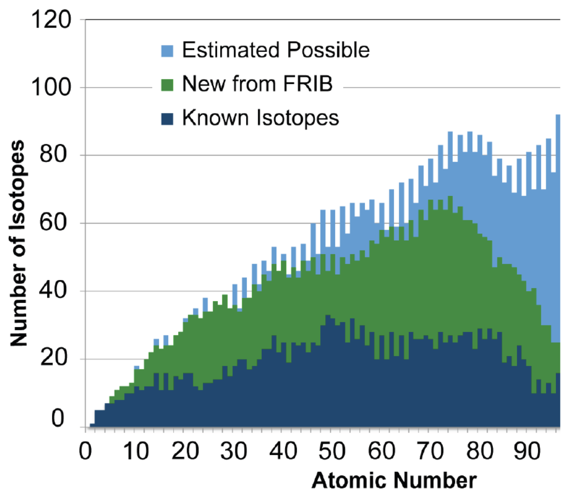
The Radiochemistry Team at FRIB/MSU

Prof. Greg Severin

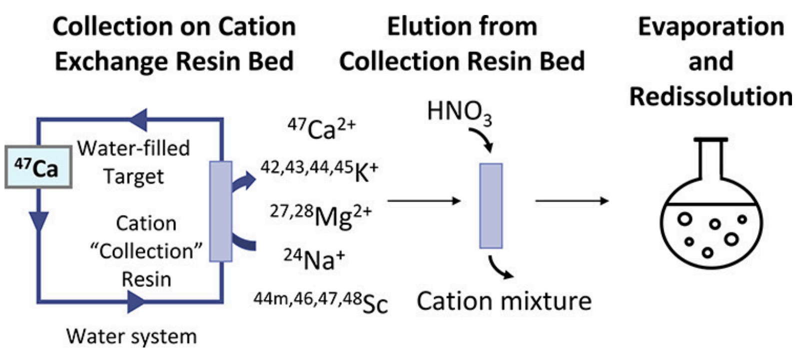


Prof. Alyssa Gaiser

Prof. Katharina Domnanich



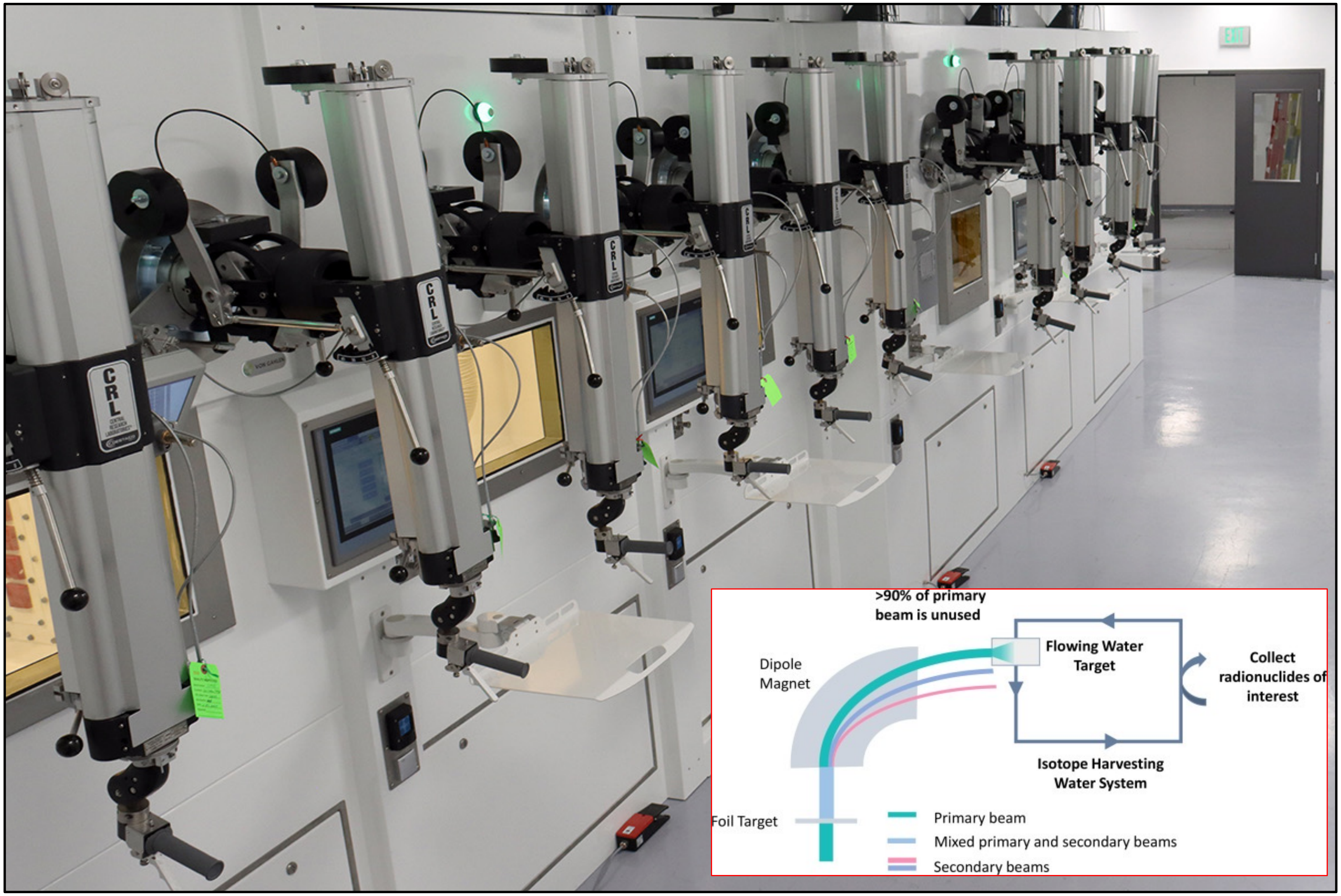
Nature 486, 509–512 (2012)



Recovery of 92% to 99% of ^{47}Ca (surrogate for Radium)

Abel et al., ACS Omega 5(43) 27864 (2020)

Isotope Harvesting Vault has been installed!

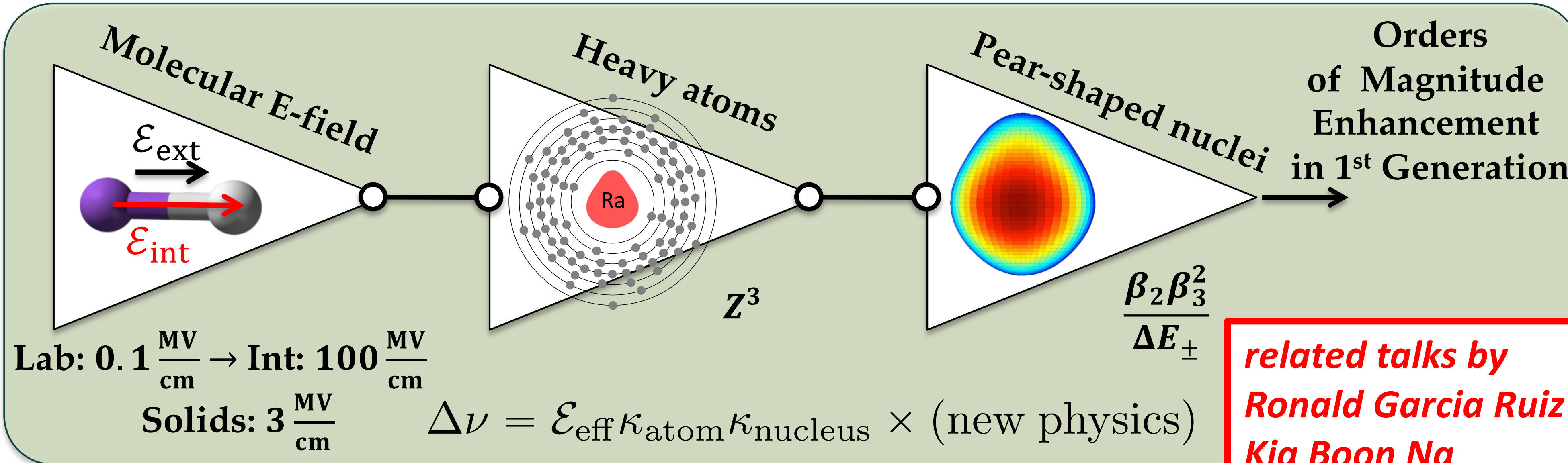


C. Vyas

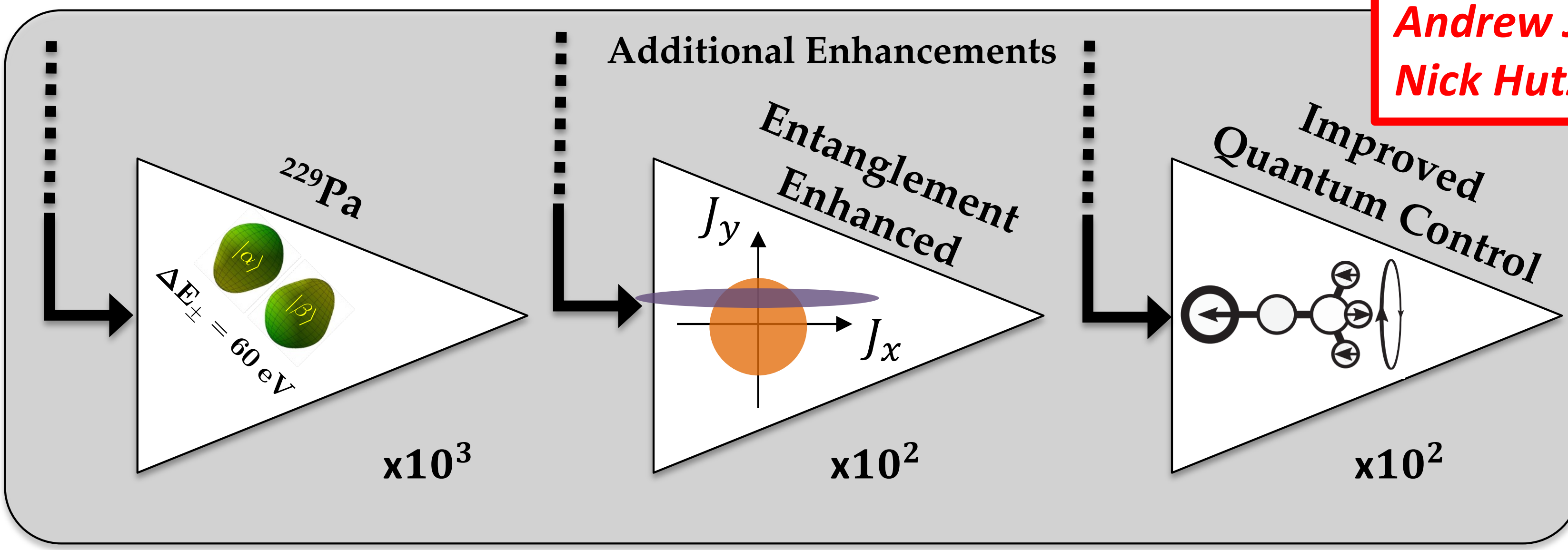
NIMB 478 34 (2020)

30 feet

New Laboratory: Trapped Radioactive Molecules Containing Heavy Pear-Shaped Nuclei



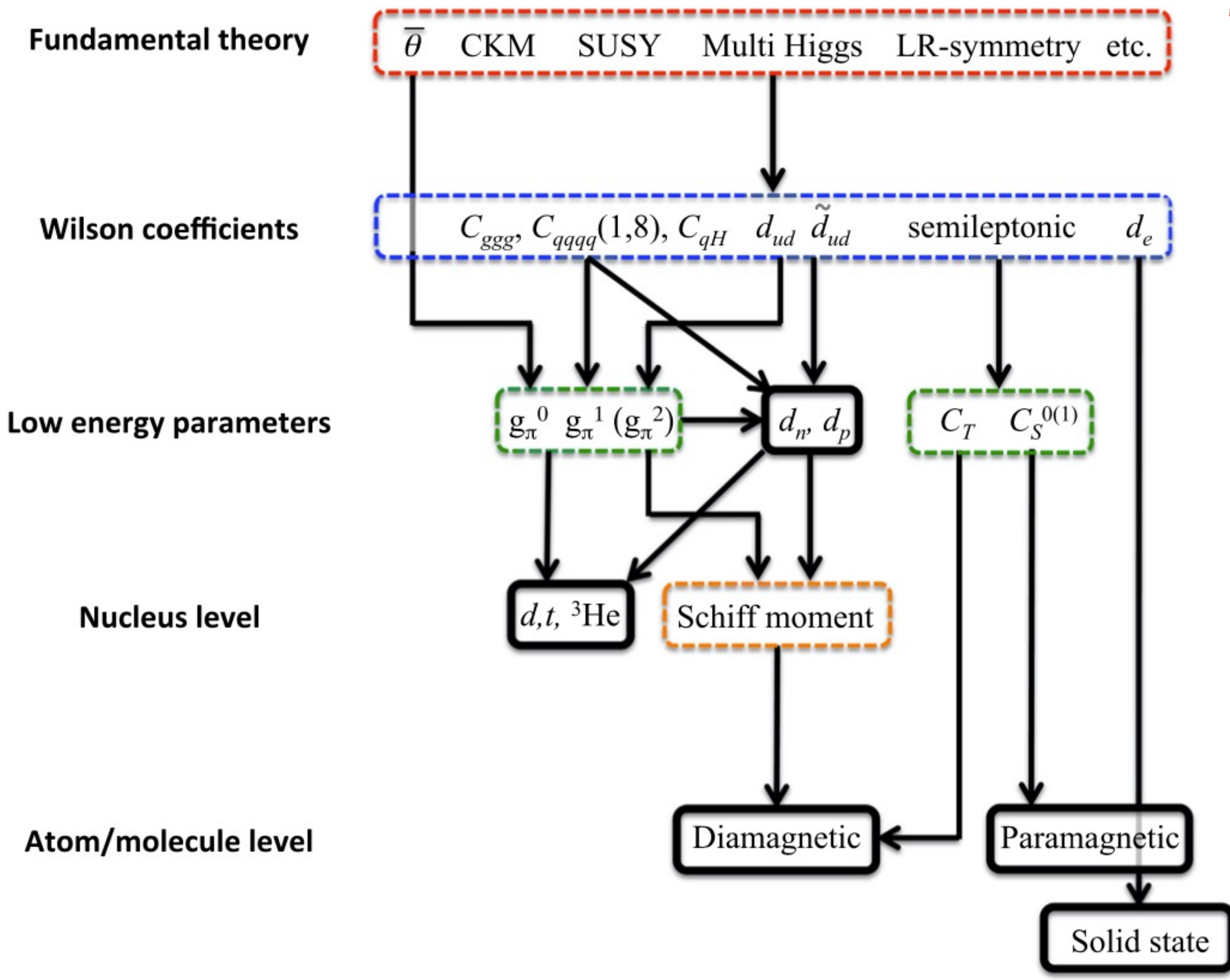
related talks by
Ronald Garcia Ruiz
Kia Boon Ng
Andrew Jayich
Nick Hutzler



Xing Wu
MSU 2023

Connecting New Physics to EDMs

T.E. Chupp, P. Fierlinger, M. Ramsey-Musolf, JTS, RMP 91:015001



Sources of *CP*-violation

Particle Physics Theory

Effective Field Theory

Lattice QCD Theory

Nuclear Theory
+
Nuclear Experiment

Atomic Theory
+
Atomic Experiment

Molecular Theory
+
Molecular Experiment

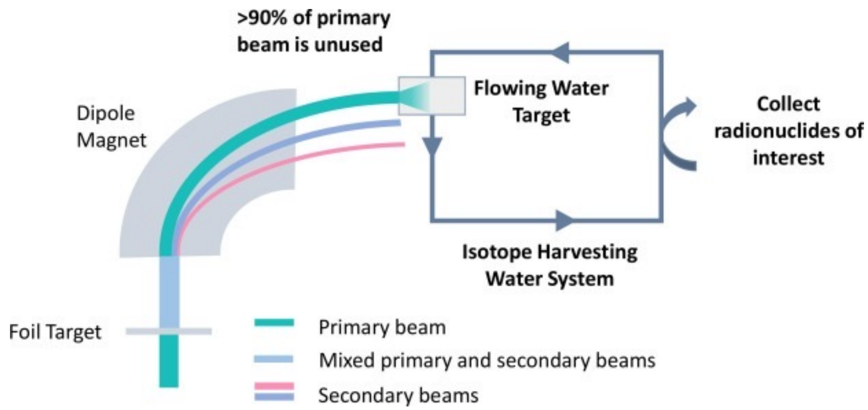
Radiochemistry

...EDMs

The Nuclear Pear Factory: A Proposed Center

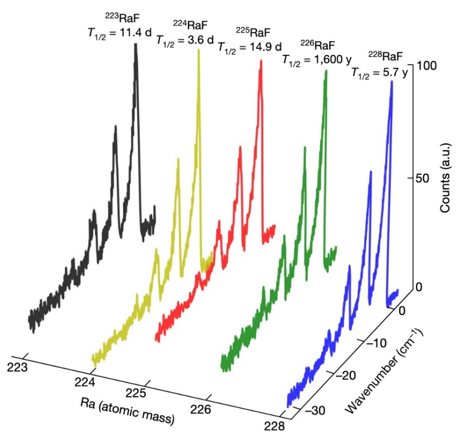
Nature 497:199 (2013)

A joint Experiment/Theory & AMO/Nuclear/Radiochemistry effort to calibrate the new physics sensitivity of pear-shaped nuclei and to carry out the requisite precursory work leading to ultrasensitive EDM searches.

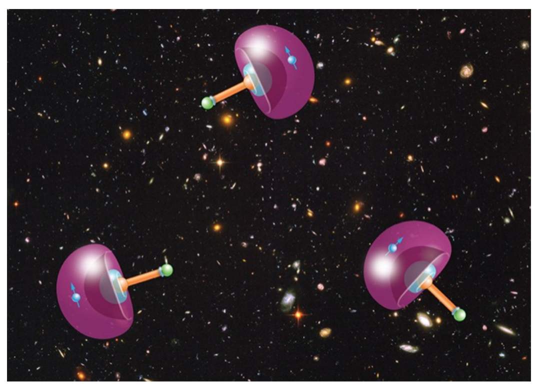


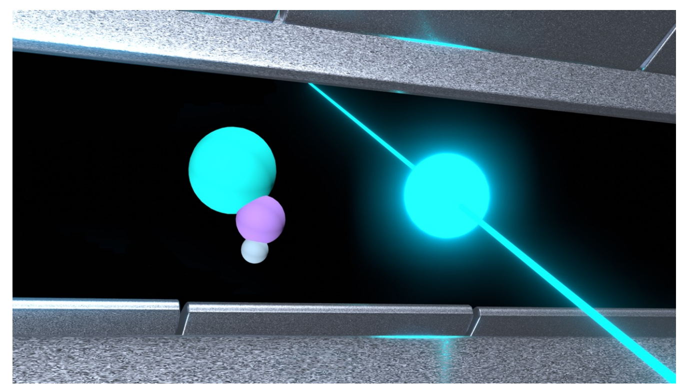
NIMB 478 34 (2020)

N P F



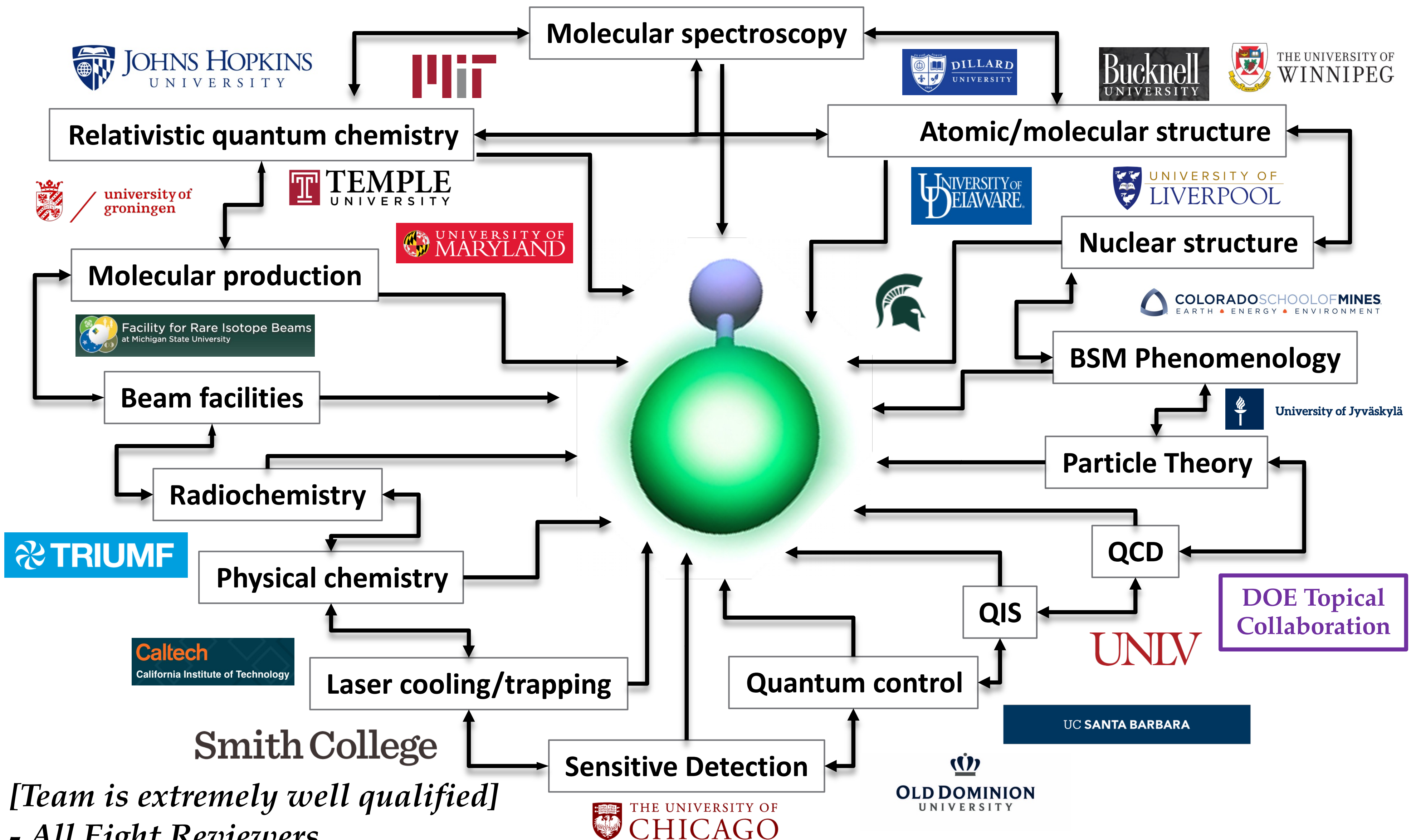
Nature 581:396 (2020)





https://physics.aps.org/articles/v14/103 & A.M. Jayich

A Center (US NSF Physics Frontiers Center) Is An Ideal Mechanism For Carrying Out This Highly Interdisciplinary Long-Term Effort



[Team is extremely well qualified]
- All Eight Reviewers

The Nuclear Pear Factory: Some History & Plans

The 2023 US NSF PFC Competition:
2022-05-02: US NSF PFC Solicitation Posted
2022-08-01: Preproposal submitted
2022-11-10: Invitation to submit Full Proposal
2023-01-27: Full Proposal submitted
2023-03-31: Invitation to present at Reverse Site Visit @ NSF (Final Round)
2023-05-03 & 2023-05-04: NPF Team in DC for Reverse Site Visit @ NSF
2023-05-22: Unofficial notice that NPF was not selected
(4 of ~10^1 were funded: 3 renewals and 1 new PFC)
2023-08-18: Official notice that full NPF proposal was declined

Email me if you want to join us... especially if you are a Theorist... especially if you do Nuclear/Lattice!
singhj@frib.msu.edu

The 2026 US NSF PFC Competition?
2024-12-20: Very positive conversation with NSF PFC program manager: solicitation written, requires internal approval, intended to be posted in May 2025, strongly encouraged to submit a stronger NPF proposal...

2025-05-12: No solicitation on NSF website...

2025-06-05: Presentation to MSU Research Foundation Board for Internal Center Development Funds ($240k total for two years – lots of funds available for workshops...)

2025-07-01: Internal MSU Center Development Funds become available if funded...

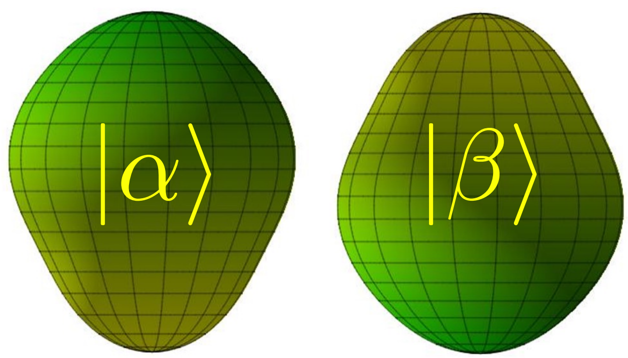
^{229}Pa *May* Be Unusually Sensitive!

Choose an isotope with large deformations

$$S \equiv \langle \Psi_0 | S_z | \Psi_0 \rangle = \sum_{k \neq 0} \frac{\langle \Psi_0 | S_z | \Psi_k \rangle \langle \Psi_k | V_{\not{P}\not{T}} | \Psi_0 \rangle}{E_0 - E_k} + \text{c.c.}$$

Unknown

Parity Doublet



ΔE

$$|\Psi_1\rangle = \frac{|\alpha\rangle \mp |\beta\rangle}{\sqrt{2}}$$

$$|\Psi_0\rangle = \frac{|\alpha\rangle \pm |\beta\rangle}{\sqrt{2}}$$

Pa-229: Haxton & Henley PRL 51:1937 (1983)
I. Ahmad et al Phys. Rev. C 92:024313 (2015)
Dobaczewski et al PRL 121, 232501 (2018)

Isotope	ΔE (keV)	$\tau_{1/2}$ (sec)	sensitivity
Hg-199	1800	stable	1
Rn-223	~10^2?	10^3	10^2
Ra-225	55	10^6	10^3
Pa-229	**(0.06 +/- 0.05)?**	**10^5**	**10^6**

FRIB will make lots of Pa-229!

Ground State Parity Doublet Energy Splitting
The Muddled Picture for ^{229}Pa (1980-2015)

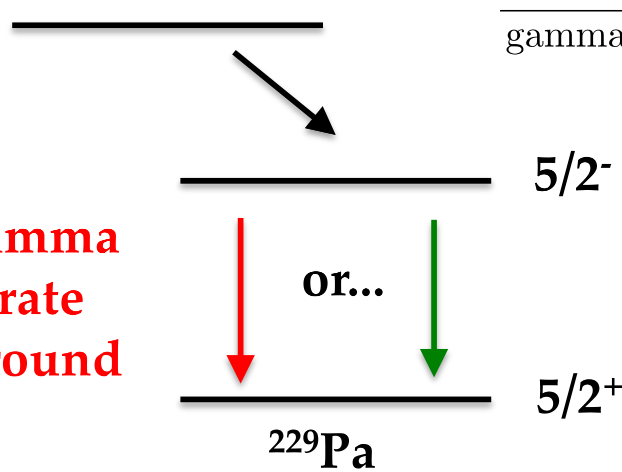
1980: Theoretical prediction: 100 eV
PLB 96(1-2):7-10
1982: Experimental claim from ANL: 220(50) eV
PRL 49:1758-61
1988: Theoretical calculation: 400 eV
PRC 37:2744-78

1991: No ground state parity doublet found in German experiment!
PRC 44:R1728-31
1993: No evidence for octupole deformation from German experiment!
PPNP 28:429-34
1994: Weak octupole collectivity now claimed by German experiments!
NPA 576:267-307

2015: Full reanalysis of 1982 ANL experiment + incorporation of data from German experiments: Original claim of 220 eV conversion electrons is gone.
If there is a ground state parity doublet, then it must be 60(50) eV.
PRC 92:024313 (2015)

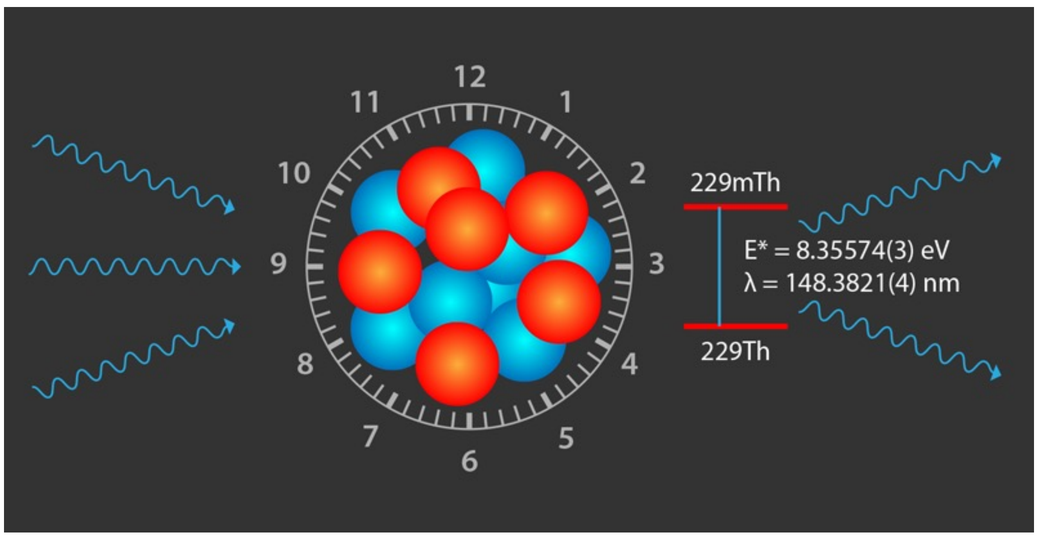
Two Complementary Paths
To Search For a Low-Lying Nuclear State in ^{229}Pa

$$\frac{\text{electrons}}{\text{gammas}} \approx \kappa_{\text{Schiff}}\,\alpha^4 Z^3 \left(\frac{L}{L+1}\right)\left(\frac{2m_e c^2}{\Delta E}\right)^{L+5/2} \approx 10^6 \text{ to } 10^{18}$$



low energy gamma
- **low signal rate**
- **low background**

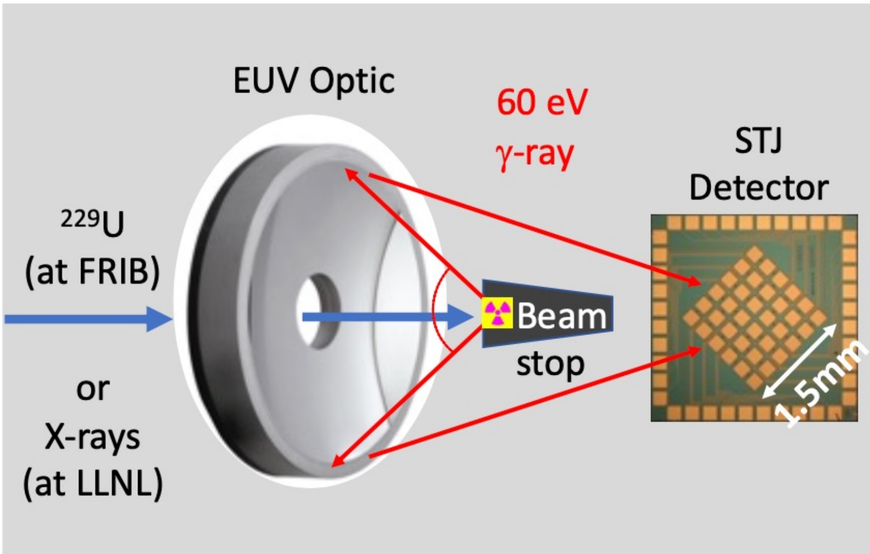
conversion electron
- **high signal rate**
- **high background**

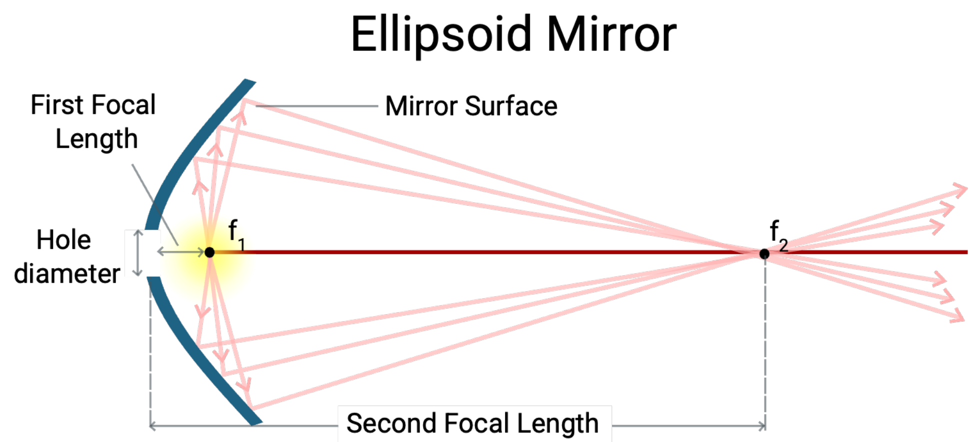


https://physics.aps.org/articles/v17/71

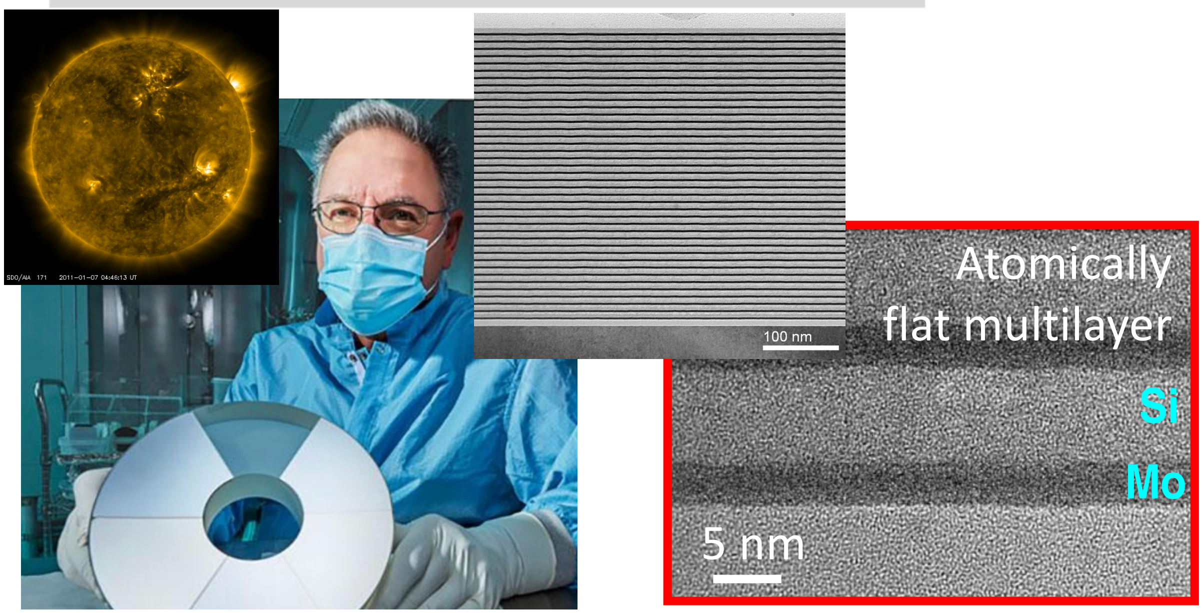
	^{229}Th	^{229}Pa
A	229	229
Z	90	91
N	139	138
ΔE	8.36 eV	(60 +/- 50) eV?
PD?	5/2$^+$ 3/2$^+$	5/2$^+$ 5/2$^-$

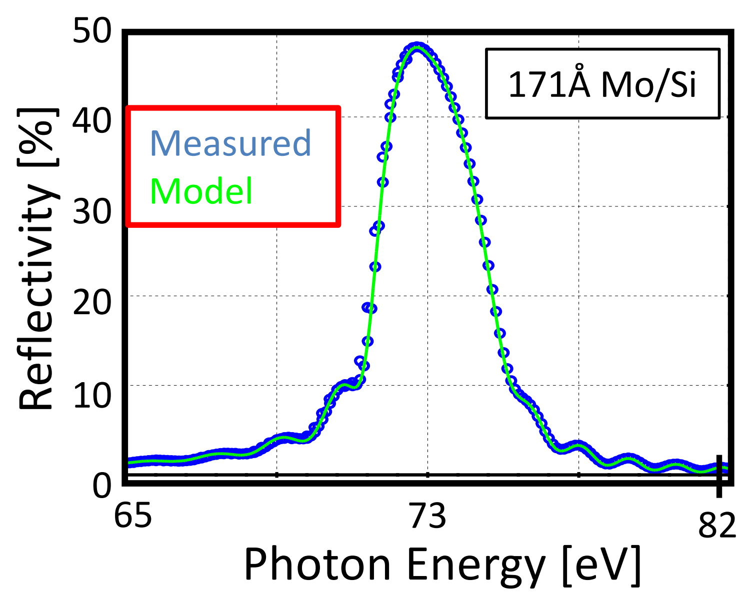
Awarded Today @ 09:56! Direct Gamma Detection (40-80 eV) w/ LLNL: Stephan Friedrich & Marie-Anne Descalle





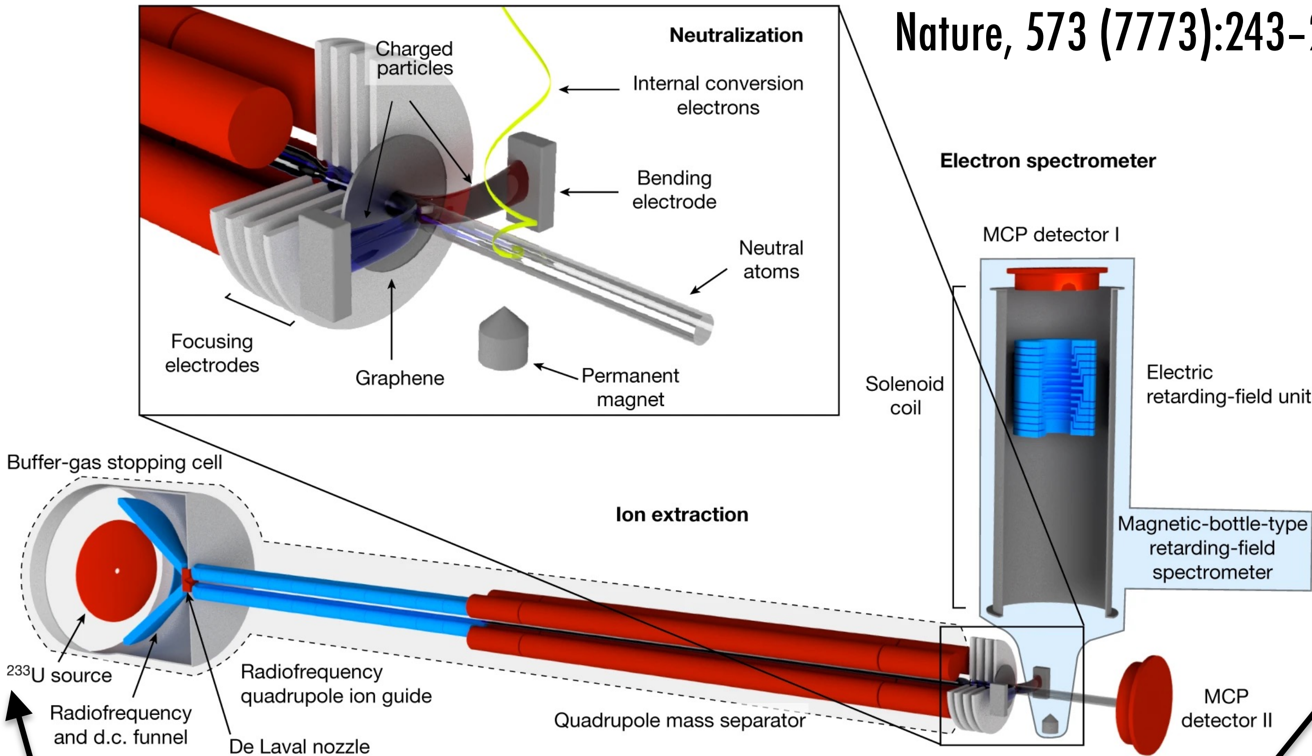
https://www.meetoptics.com/academy/ellipsoidal-mirrors#what-are-ellipsoidal-mirrors



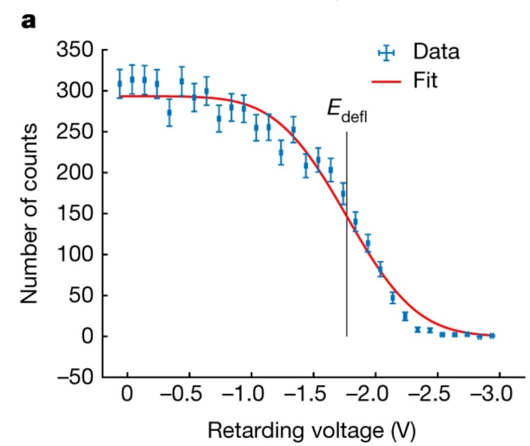


Spinlab Pending: The ^{229}Th Measurement Technique: Integrated Conversion Electron Spectroscopy

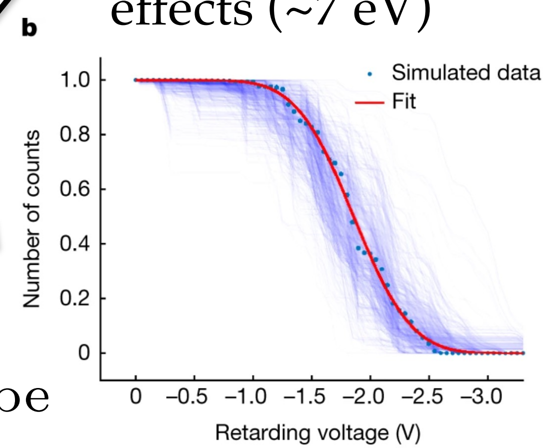
Nature, 573 (7773):243-246 (2019)



measurement



atomic binding effects (~7 eV)



^{229}U implanted from FRIB

We are just looking for nonzero counts and then the order of magnitude of ΔE if a signal is observed. Unlike for ^{229}Th, knowledge of atomic binding effects is not crucial.
Main background: 8.36 eV from ^{229}Th!

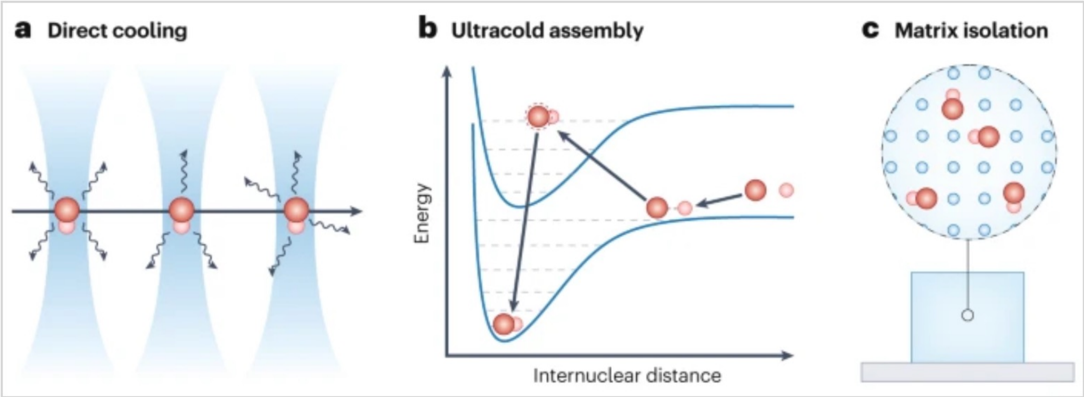
$$\Delta E = E_{\text{defl}} + E_{\text{abe}}$$

Heavy Pear-Shaped Nuclei Inside Molecules/Solids

Enhancements: nuclear Schiff moment enhancement of x1000 (^{225}Ra)
to maybe(!?!) x1000000 (^{229}Pa)
and ~100 MV / cm effective internal *E*-field (lab < 1 MV / cm)
[N.B. the nucleus feels a different E_{eff} than the electrons!]

Potential: x10^5 to x10^{10} more new physics sensitivity than the ^{199}Hg experiment <u>on a per atom basis.</u>

Opportunity:
Isotope harvesting @ FRIB:
from "Beam to Beaker"
(^{225}Ra, ^{229}Pa, ...)



Nature Physics 20, p741-749 (2024)

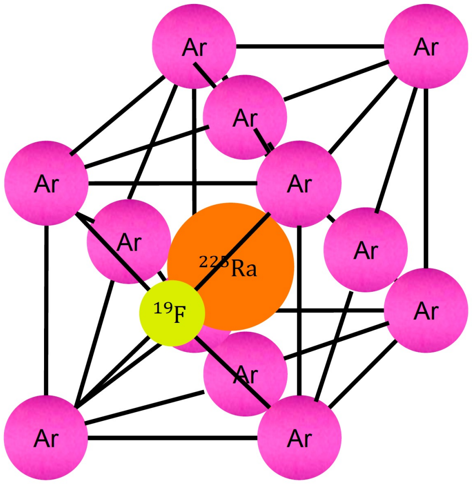
Challenges:

- How do we get the harvested isotopes from "Beaker" into an experiment?
- How do we calibrate the new physics sensitivity of these "enhancer isotopes" inside of molecules?
- How do we efficiently form & probe short-lived radioactive molecules?

Spinlab: Pear-Shaped Nuclei Implanted In Cryogenic Solids: ^{225}RaF/ ^{225}RaO ($\tau_{1/2}$ = 15 days) & ^{229}Pa ($\tau_{1/2}$ = 1.5 days)

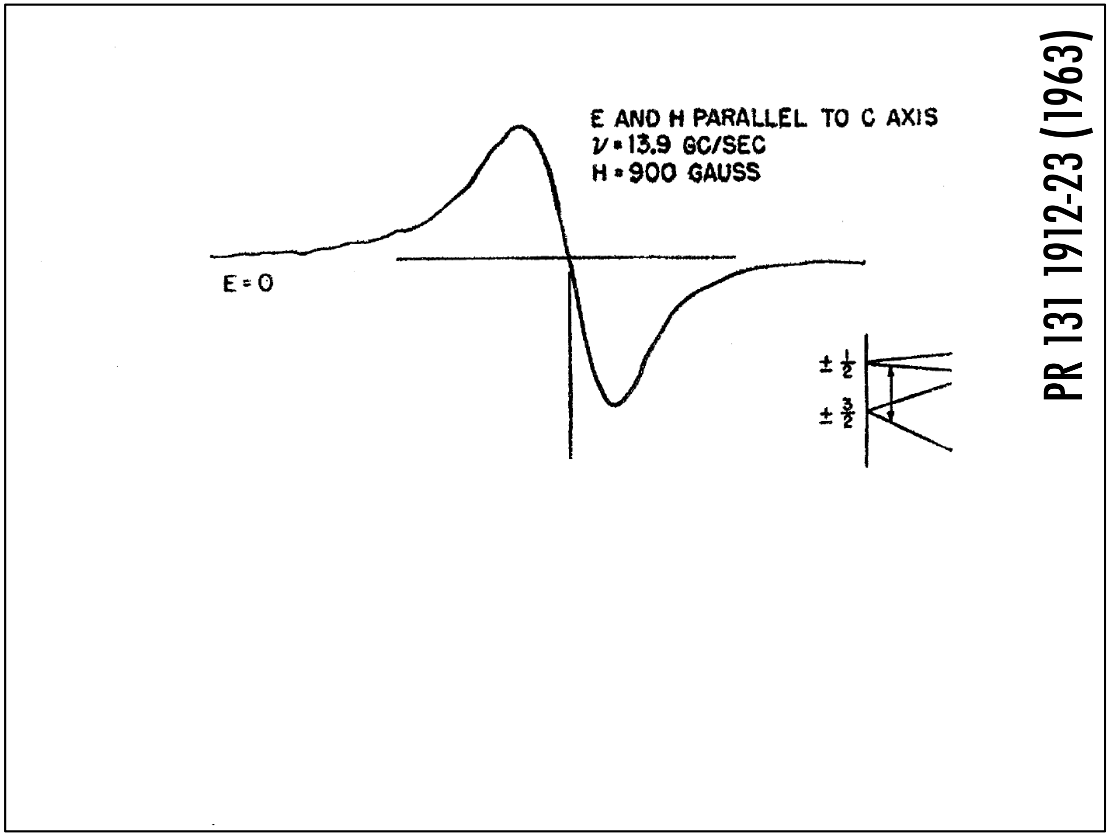
- **Efficient trapping of a wide variety of species**
- **Very high number densities**
- Stable and chemically inert confinement
- Transparent in the optical regime for optical probing
- Under certain conditions, polar molecules orient themselves along the crystal axes which allows for control of systematics: PRA 98:032513 (2018)
- **Challenge: quantum control in rare gas solids**
- Ions implanted in optical crystals or diamonds allowing for optically-addressable nuclear spins

  Hyp. Int. 240:29 (2019), Phil. Trans. R. Soc. A.382: 20230169 (2024), PRA 108, 012819 (2023)
- Implanted ions can sit at two distinct sites with opposite pointing internal E-fields which allows for control of systematics PR 131 1912 (1963)



Controlling Systematics in Solids: The RB-Experiment

Cr^{3+} in Al_2O_3: Royce & Bloembergen Phys. Rev. 131 1912 (1963)



external B-field

Key Concept: In the absence of external fields, the non-degeneracy of a Kramers Doublet is an indication of time-reversal symmetry violation.

"Stark Splitting:" There Are Two Ensembles of Ions!

Cr^{3+} in Al_2O_3: Royce & Bloembergen Phys. Rev. 131 1912 (1963)

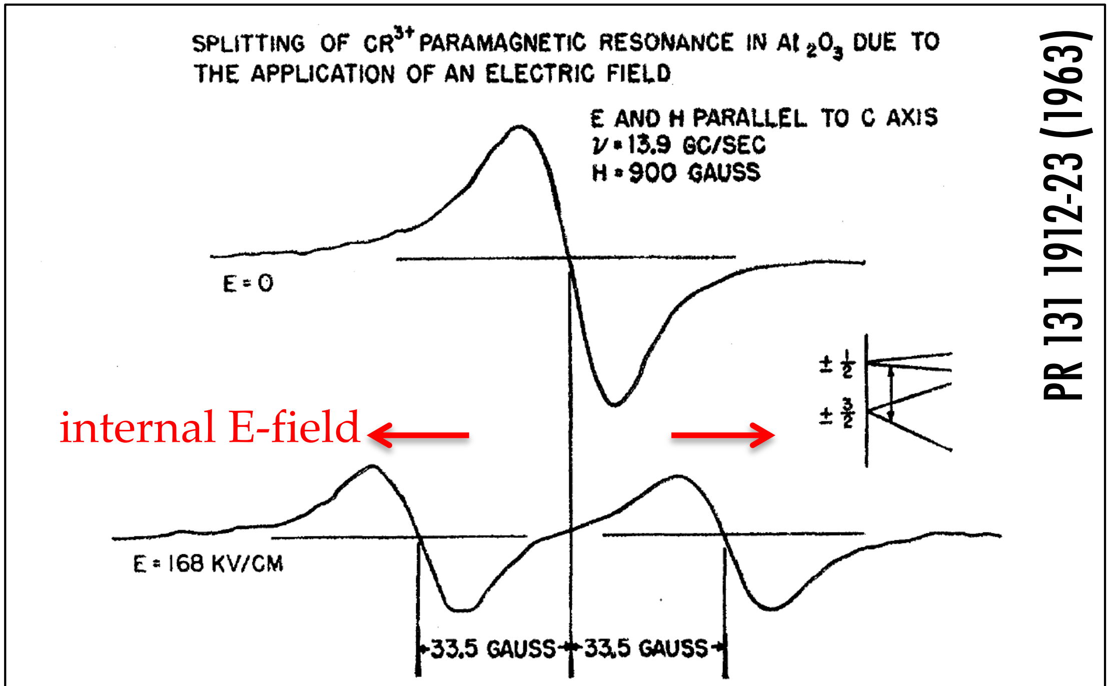


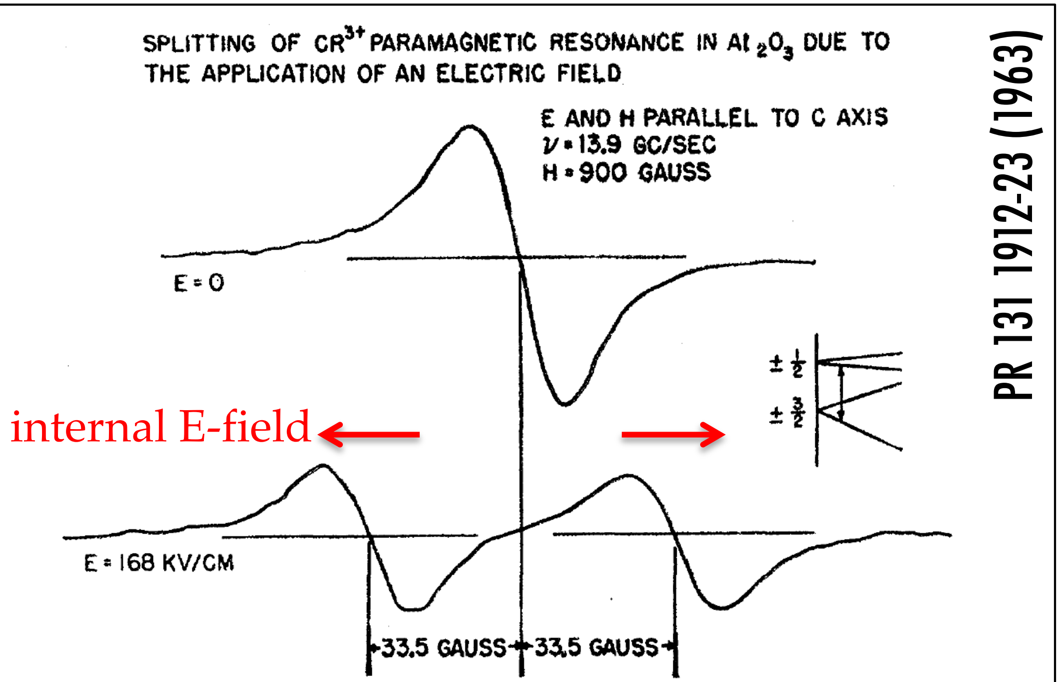
FIG. 1. Experimental absorption derivative curves showing the electric-field-induced splitting of the $\frac{1}{2} \rightarrow \frac{3}{2}$ resonance line of Cr^{3+} in ruby.

→ external B-field

Key Concept: Under the right conditions, ions (& molecules...) in solids are oriented along the crystal axes.

Apply E-Field Only During State Detection!

Cr^{3+} in Al_2O_3: Royce & Bloembergen Phys. Rev. 131 1912 (1963)



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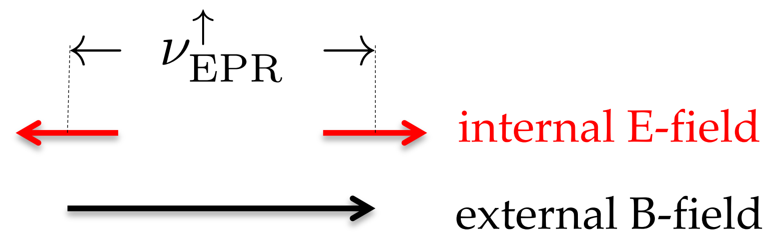
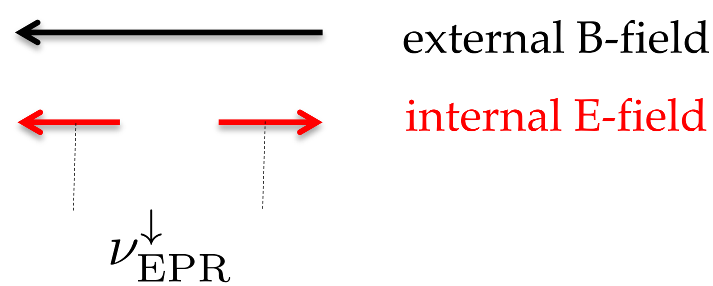
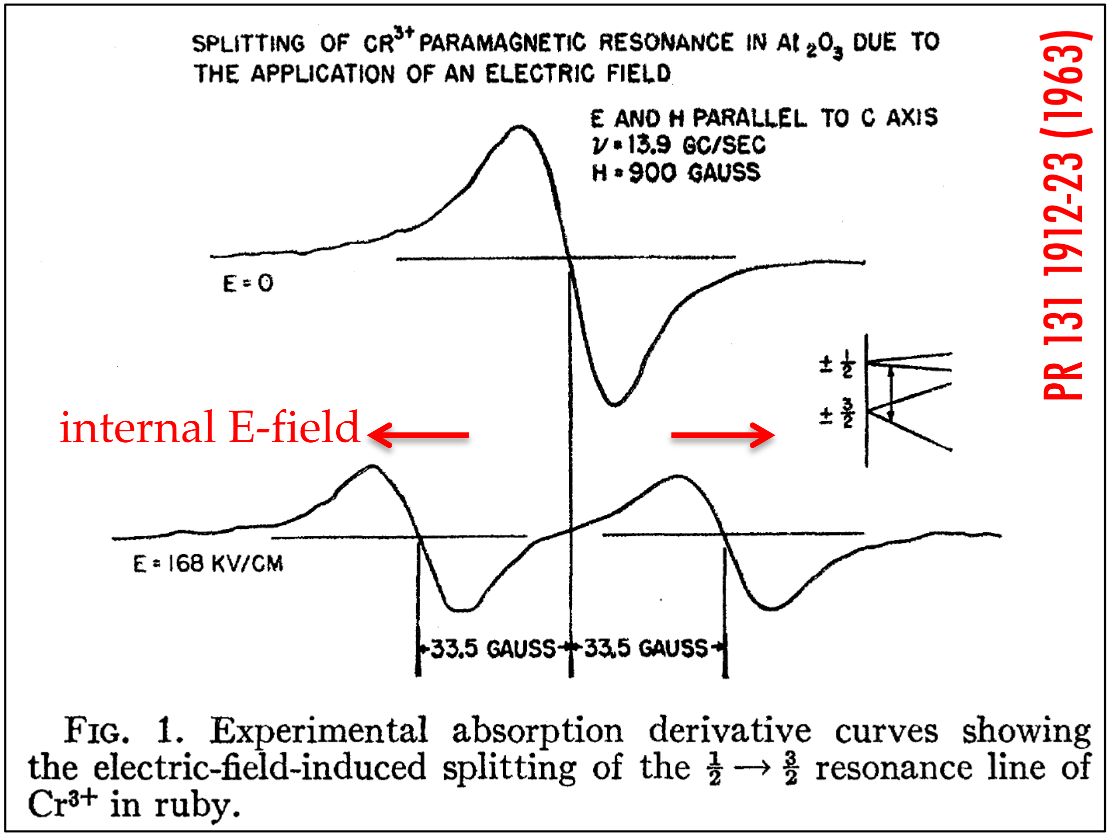


$$\Delta\nu_{\mathrm{EPR}} = \frac{4\Delta m d E}{J}$$



Key Concept: Apply a lab *E*-field only during the detection/readout step just to spectroscopically distinguish the two ensembles!

Key Idea to Control Systematics in Solids: Two Nearby Sites Where Effective E-fields Are Opposed



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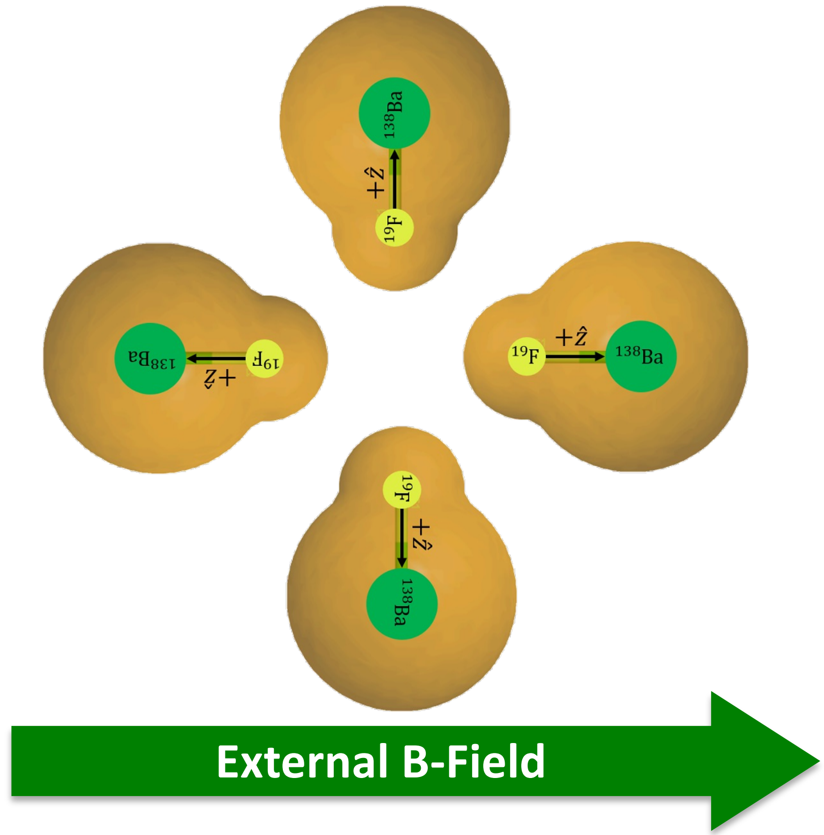
→ external B-field

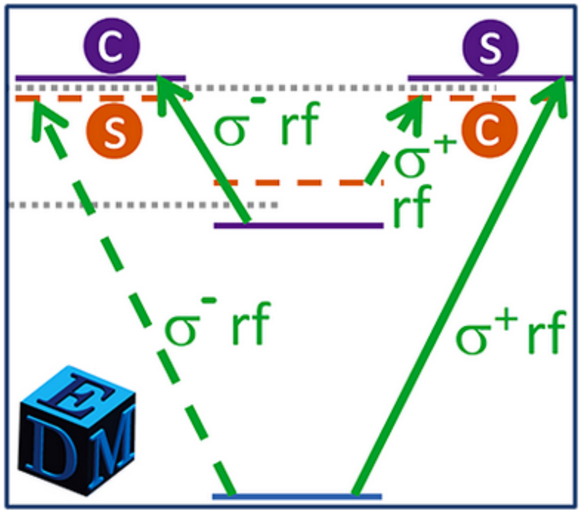


PRA 98:032513 (2018)

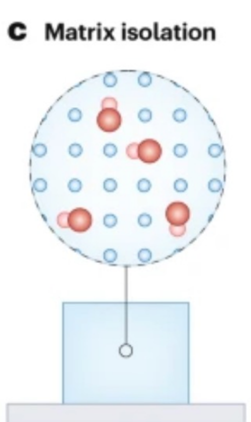
BaF/RaF orientations
(in/out not depicted here)

Key Concept: No lab *E*-field during the spin precession step minimizes the sensitivity of the frequency measurement to external *E*-fields.

One Example of a Modern "RB-Experiment"



EDM³
E. A. Hessels (York, Canada)
A. Vutha (Toronto)
JTS (MSU)



Nature Physics 20, p741–749 (2024)

EDITORS' SUGGESTION

Orientation-dependent hyperfine structure of polar molecules in a rare-gas matrix: A scheme for measuring the electron electric dipole moment

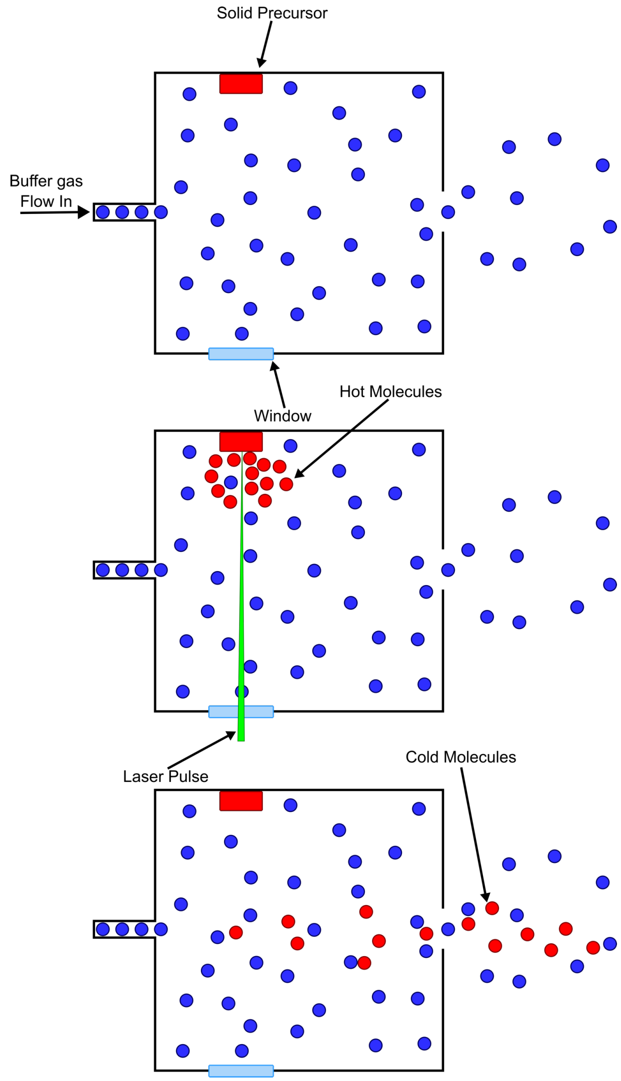
The Stark shift of the hyperfine states of polar molecules embedded in a solid rare-gas matrix is found to depend on the molecular orientation. This finding may significantly improve the measurements of the electron electric dipole moment by using large ensembles of polar molecules trapped in rare-gas matrices with orientation-dependent detections.

A. C. Vutha, M. Horbatsch, and E. A. Hessels

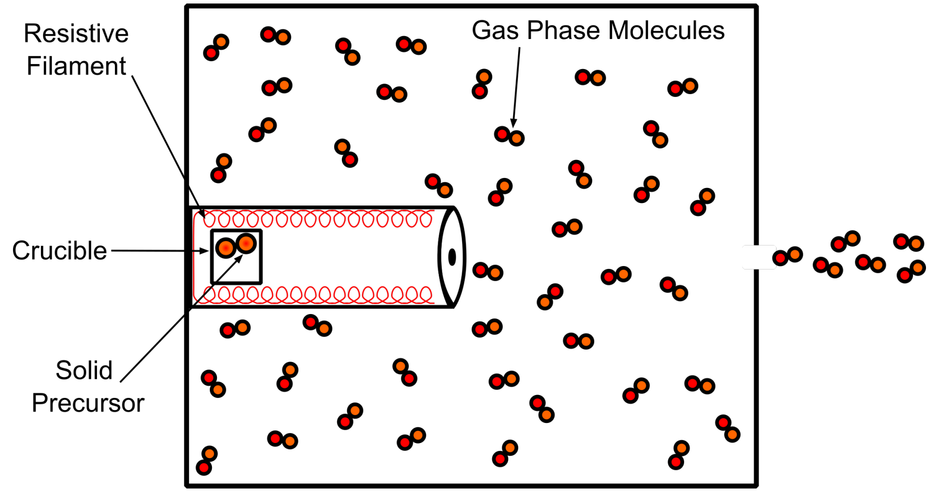
Phys. Rev. A **98**, 032513 (2018)

Some Molecule Production Techniques

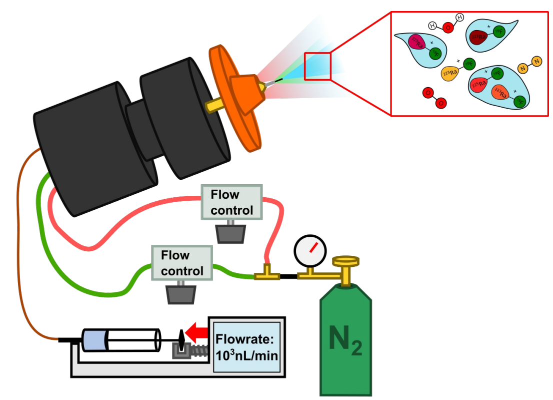
Less Brute Force:
Laser Ablation + Cold Buffer Gas



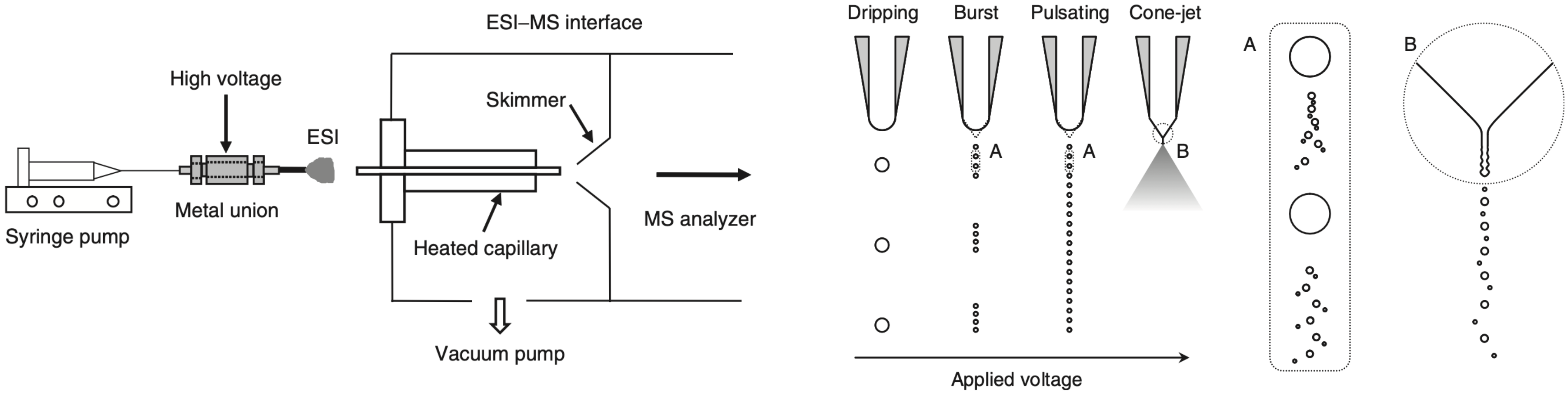
Most Brute Force:
High Temperature Oven

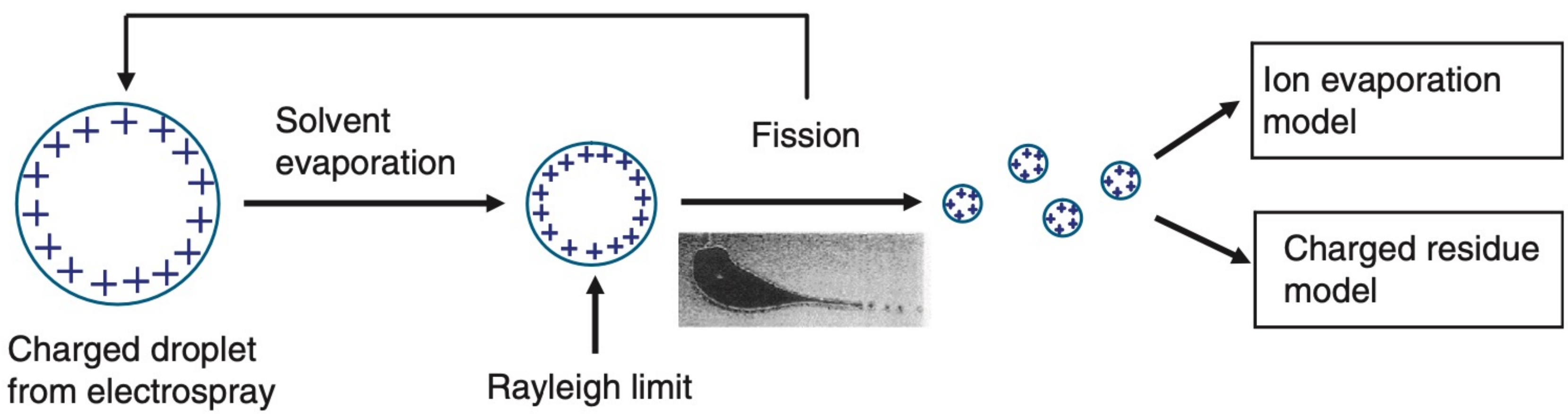


Crazy Enough to Work:
Electrospray Ionization



Electrospray Ionization Basics





Encyclopedia of Spectroscopy and Spectrometry, Third Edition http://dx.doi.org/10.1016/B978-0-12-803224-4.00319-8

Nobel Prize Chemistry 2002: John B. Fenn

Electrospray Ionization Could Be Very Efficient!

Anal. Chem. **2010,** *82,* 9344–9349

Achieving 50% Ionization Efficiency in Subambient Pressure Ionization with Nanoelectrospray

Ioan Marginean, Jason S. Page, Aleksey V. Tolmachev, Keqi Tang, and Richard D. Smith*

Biological Sciences Division, Pacific Northwest National Laboratory, P.O. Box 999, Richland, Washington 99352, United States

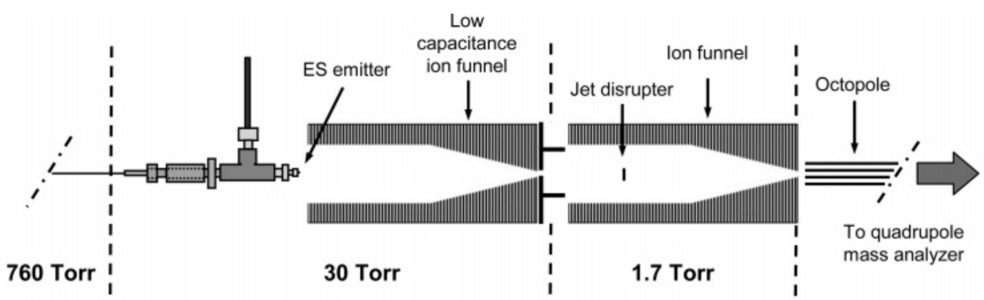
Inefficient ionization and poor transmission of the charged species produced by an electrospray from the ambient pressure mass spectrometer source into the high vacuum region required for mass analysis significantly limits achievable sensitivity. Here, we present evidence that, when operated at flow rates of 50 nL/min, a new electrospray-based ion source operated at ∼20 Torr can deliver ∼50% of the analyte ions initially in the solution as charged desolvated species into the rough vacuum region of mass spectrometers. The ion source can be tuned to optimize the analyte signal for readily ionized species while reducing the background contribution.

Anal. Chem. **2008,** *80,* 1800−1805

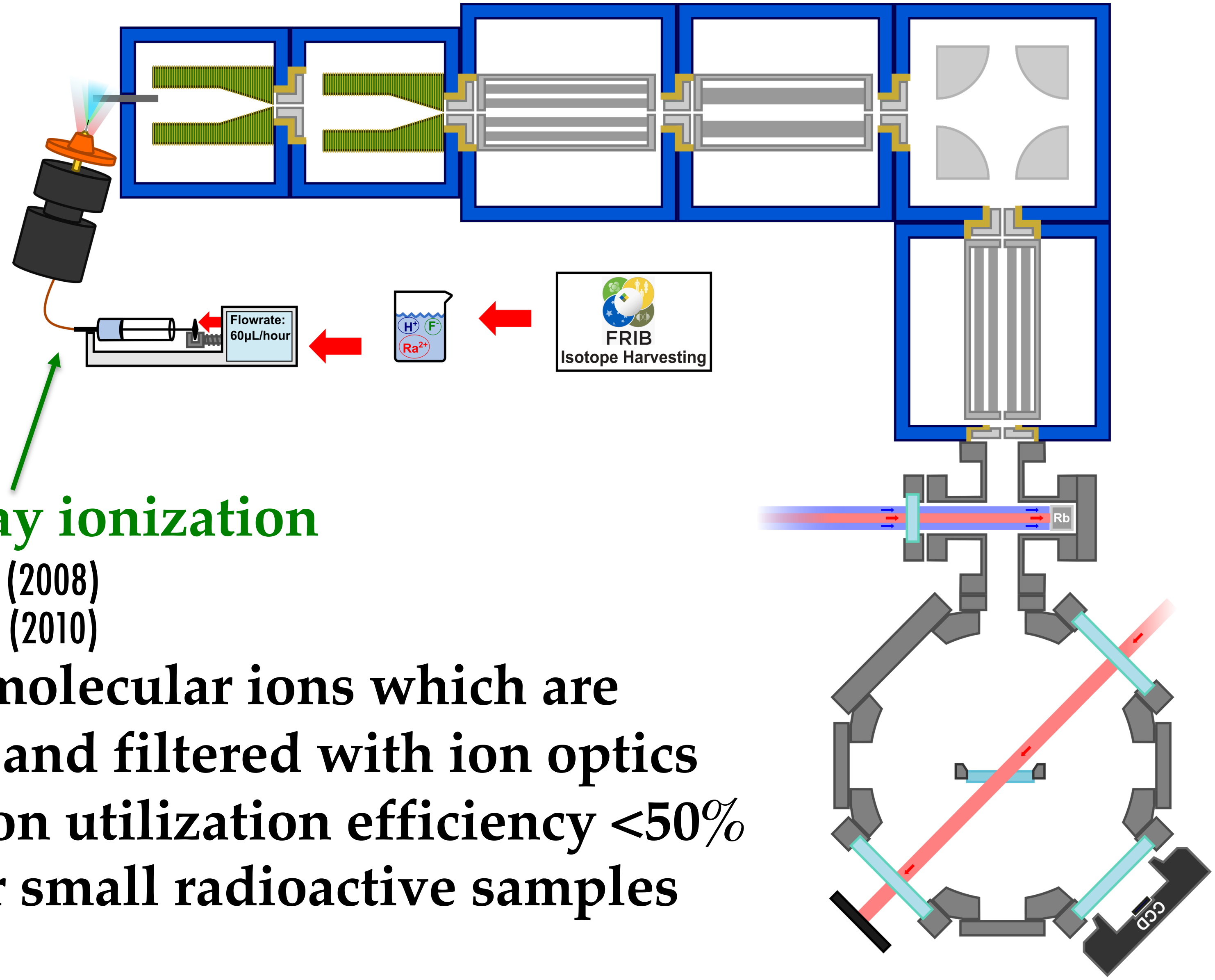
Subambient Pressure Ionization with Nanoelectrospray Source and Interface for Improved Sensitivity in Mass Spectrometry

Jason S. Page, Keqi Tang, Ryan T. Kelly, and Richard D. Smith*

Biological Sciences Division, Pacific Northwest National Laboratory, P.O. Box 999, Richland, Washington 99352



Spinlab: ^{225}RaF/ ^{225}RaO in Noble Gas Solids

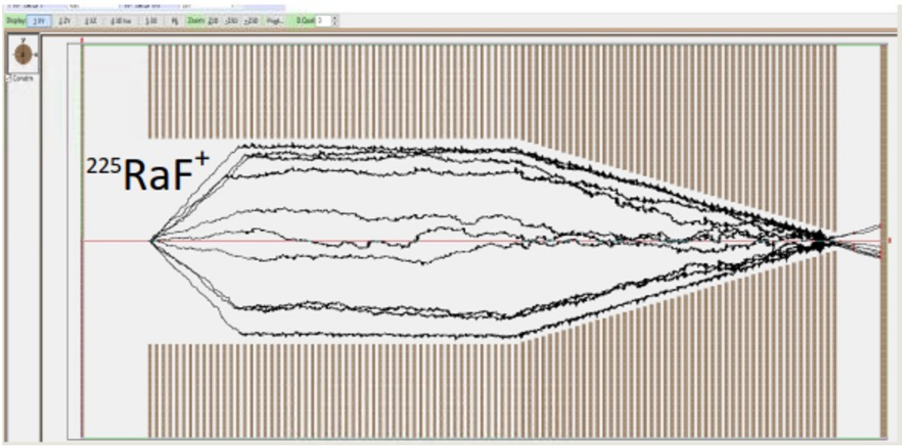


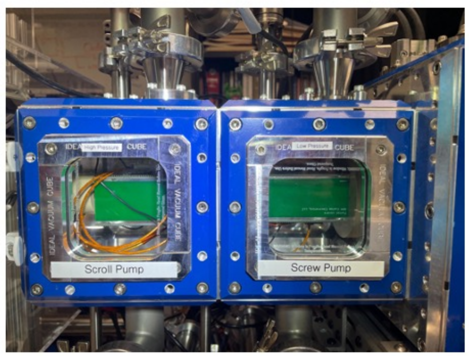
Electrospray ionization

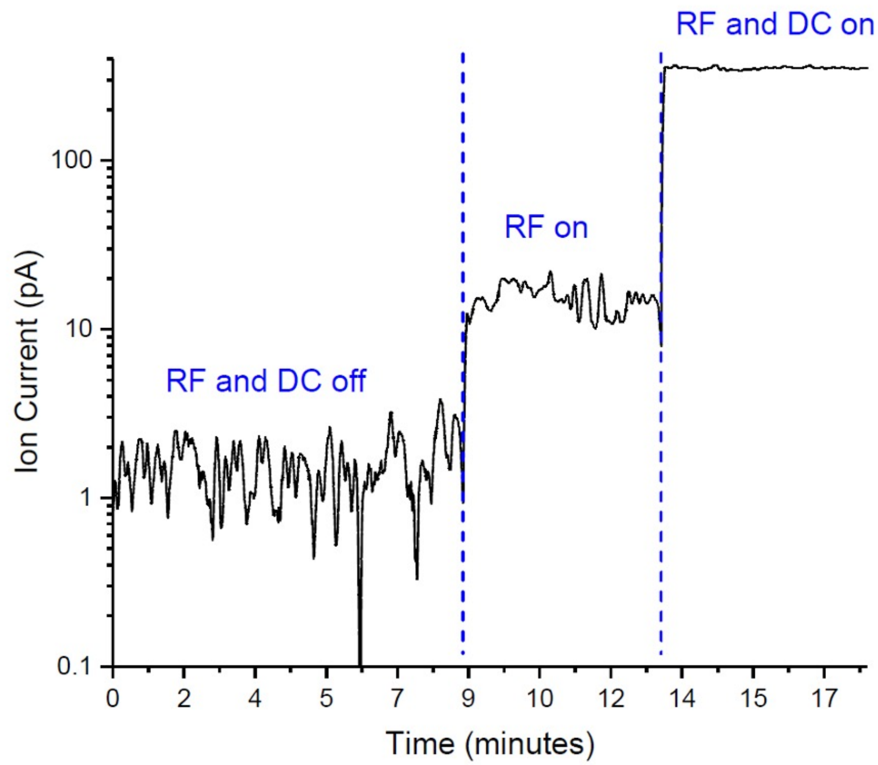
Anal. Chem.80:1800 (2008)
Anal. Chem.82:9344 (2010)

- creates molecular ions which are focused and filtered with ion optics
- ionization utilization efficiency <50%
- ideal for small radioactive samples

Spinlab: Testing With Isopropanol:
Ion Funnels Work Well! (~$5k off the shelf)



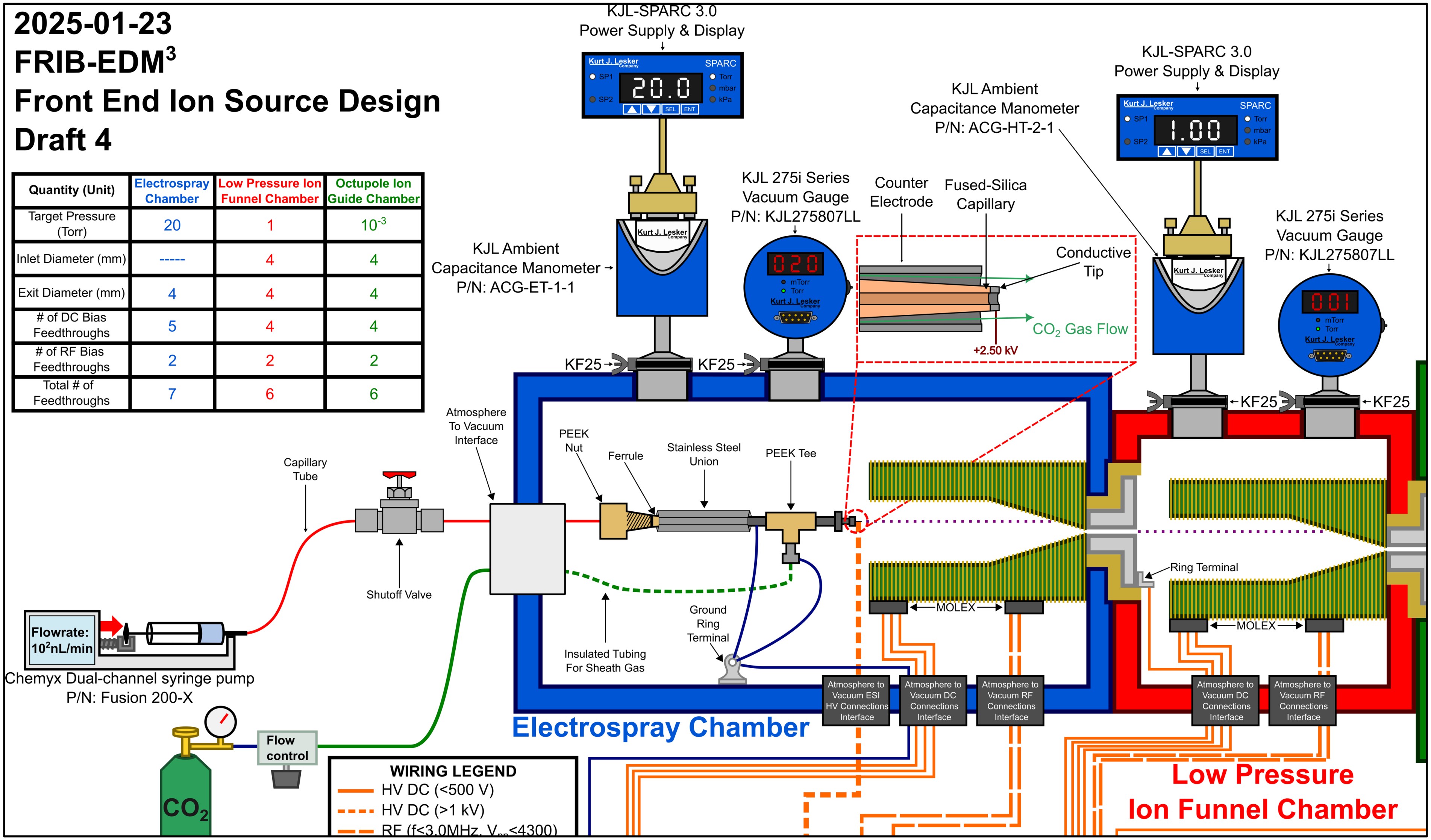




Stable ion beams are possible:
- Can be used during installation to test ion optics
- Optimizing applied bias

Challenges:
- Low reproducibility
- Lack of control over positioning and probe alignment

Spinlab: Current Status of Source Interface Design



Quantity (Unit)	Electrospray Chamber	Low Pressure Ion Funnel Chamber	Octupole Ion Guide Chamber
Target Pressure (Torr)	20	1	10^{-3}
Inlet Diameter (mm)	-----	4	4
Exit Diameter (mm)	4	4	4
# of DC Bias Feedthroughs	5	4	4
# of RF Bias Feedthroughs	2	2	2
Total # of Feedthroughs	7	6	6

FRIB is eager to invest in unique off-line opportunities!

Exhortation: The appearance of inefficiently duplicating efforts at multiple facilities runs some risk under the present funding context.

FRIB is eager to proactively support the needs of the FS community in places where it may be able to play a unique role:

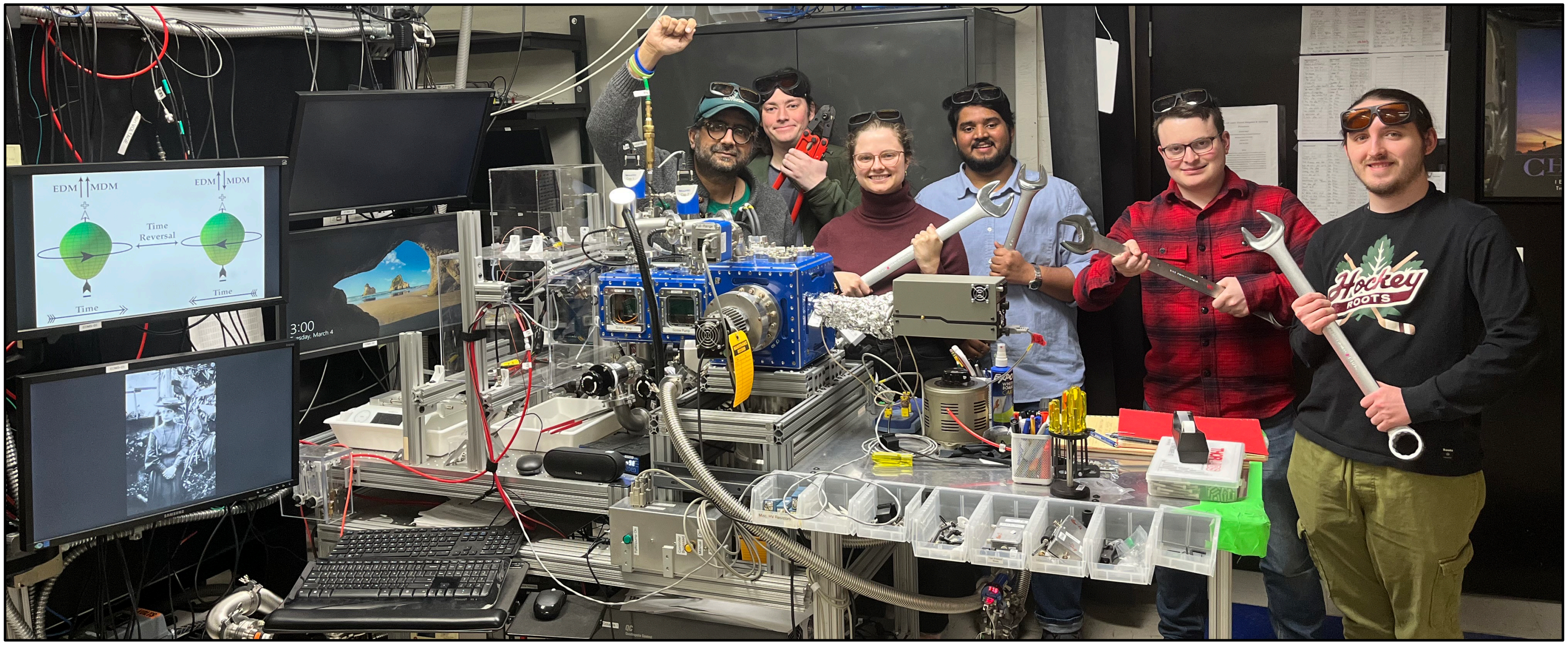
1. Offline access to some interesting isotopes (^{225}Ra, ^{229}Pa, others?) by direct production via the Isotope Harvesting program.
2. General collaborative support for radiochemistry for anyone anywhere
3. People (searching for an FS staff scientist now- see **QR Code**), space, and capital equipment for FS experiments physically hosted at FRIB
4. Plans for a general-purpose beamline that utilizes harvested isotopes (US NSF MRI)
5. Administrative/logistical/organizational support for Center-level effort to help provide a "home-base" within the US context
6. DOE Office of Science has requested a FRIB Director's Review.

FRIB PIs: Jaideep Taggart Singh (Spinlab), Xing Wu (QUEST)
Shane Wilkins (starting July 1)

- Atomic ^{225}Ra EDM in a laser trap
- Polar molecules trapped in noble gas solids (EDM-cubed)
- ^{229}Pa nuclear structure measurements
- ^{229}Pa ions implanted in optical crystals/diamonds
- Centrifuge slowing of cold atoms and molecules
- Optical cycling properties of interesting molecules
- Cold sources for radioactive molecules

FS Scientist 959065

Thanks For Your Attention!



U.S. DEPARTMENT of ENERGY

DE-SC0019015 (ECA-EDM3)
DE-SC0019455 (Ra EDM)
DE-NA0003996 (Pa-229/SAM)
DE-SC0025679 (Ra EDM+EDM3)

NSF

#1654610
(CAREER-SAM)
#2412951 (SAM)

GORDON AND BETTY MOORE FOUNDATION

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GBMF8863
G-2019-12503