Experiments using molecules (stable or radioactive) to detect P,T-violating new physics and measure hadronic P-violation

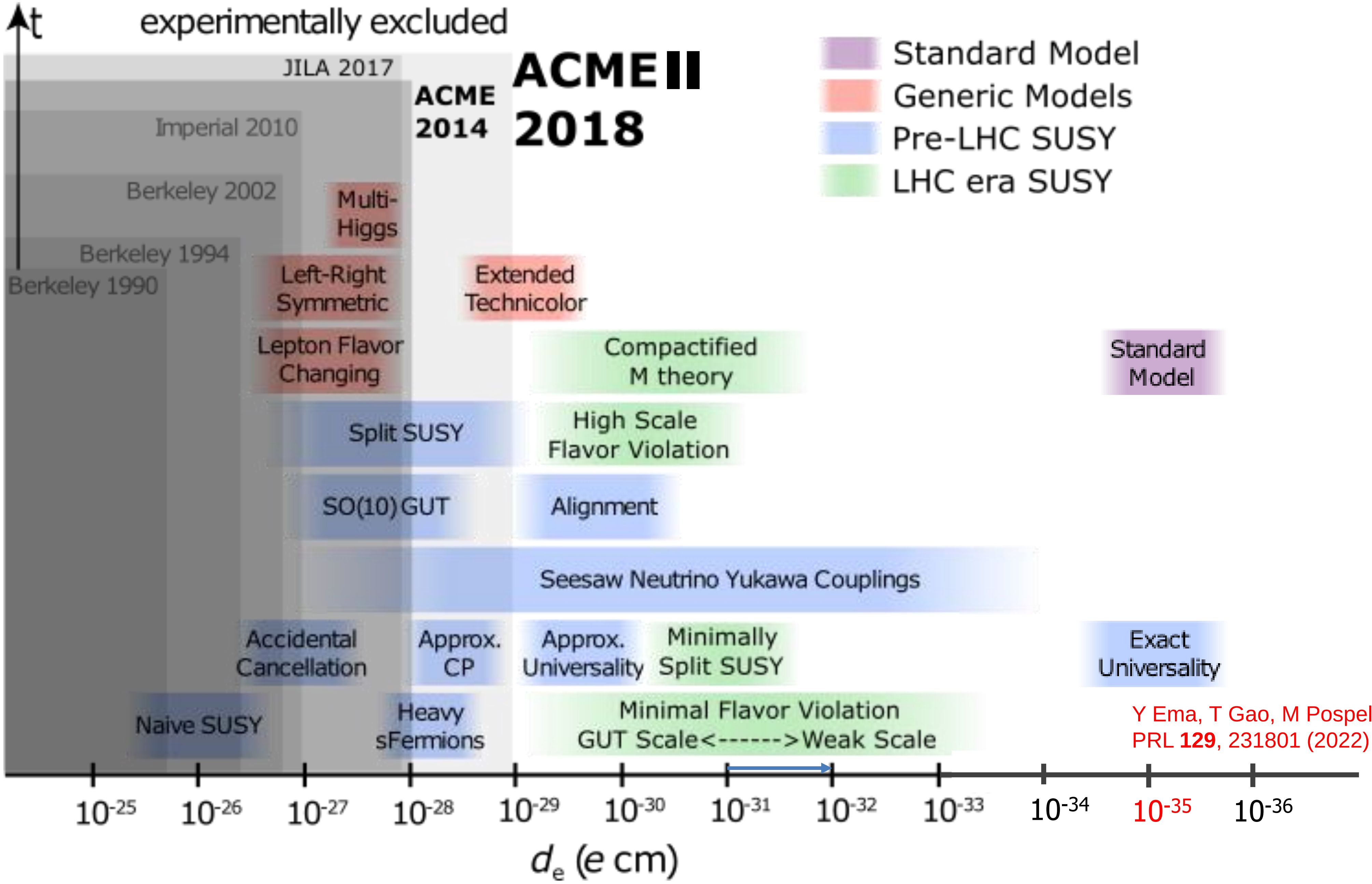
- Overview: particle electric dipole moments (EDMs) as probes of high-scale physics
- Polar molecules as amplifiers of EDM signals
- Case study: ACME search for the electron EDM
- Assembled ultracold molecules for EDMs`
- New initiative: nuclear “EDM” search with 223FrAg molecules
- ZOMBIES: measuring nuclear anapole moments

Dave DeMille
Department of Physics, University of Chicago
Physics Division, Argonne National Lab

Funding
NSF, Moore Foundation, Sloan Foundation, DOE

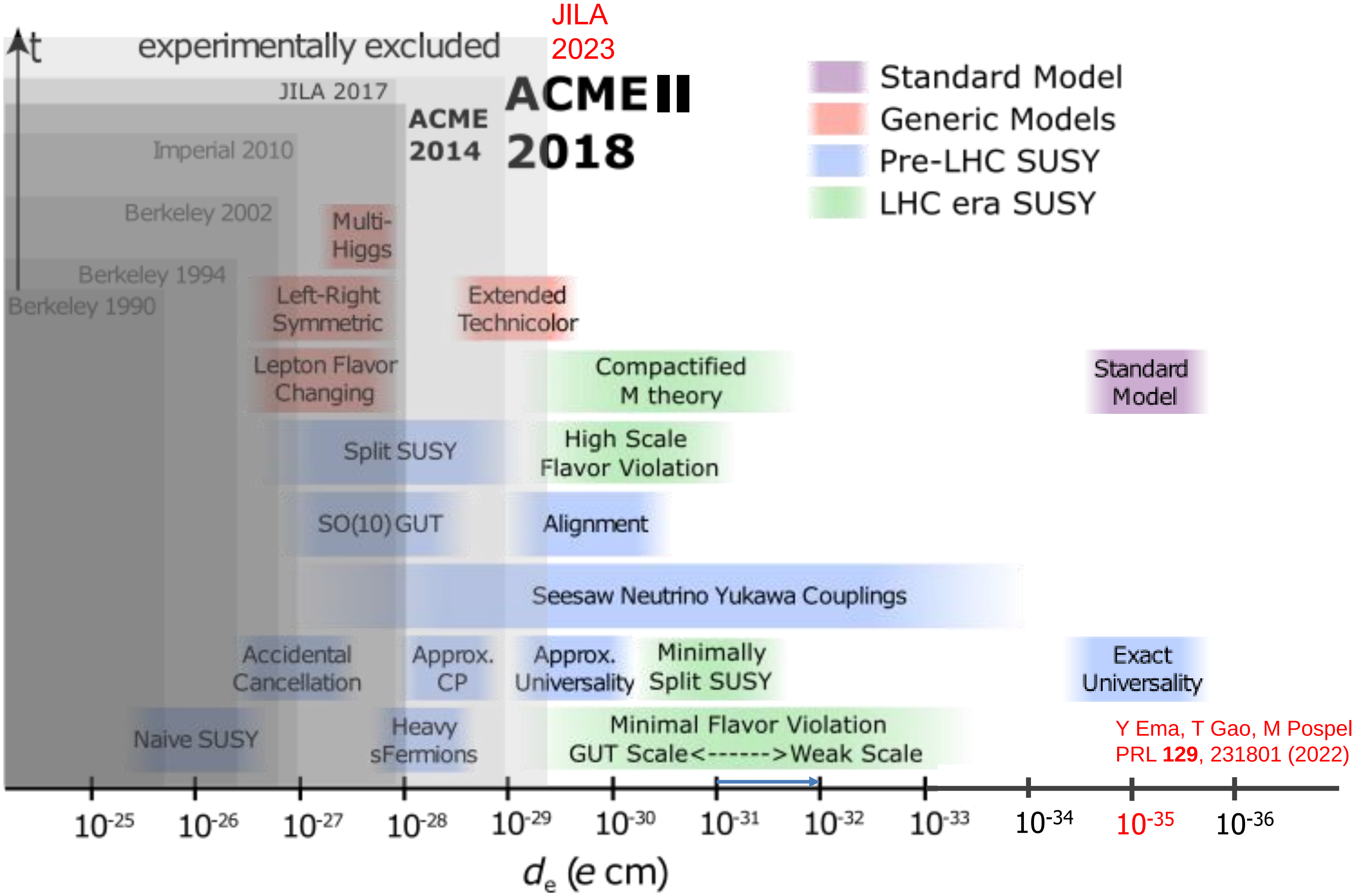
DeMille
Group

Search for new physics via electron EDM



Y Ema, T Gao, M Pospelov
PRL **129**, 231801 (2022)

Search for new physics via electron EDM



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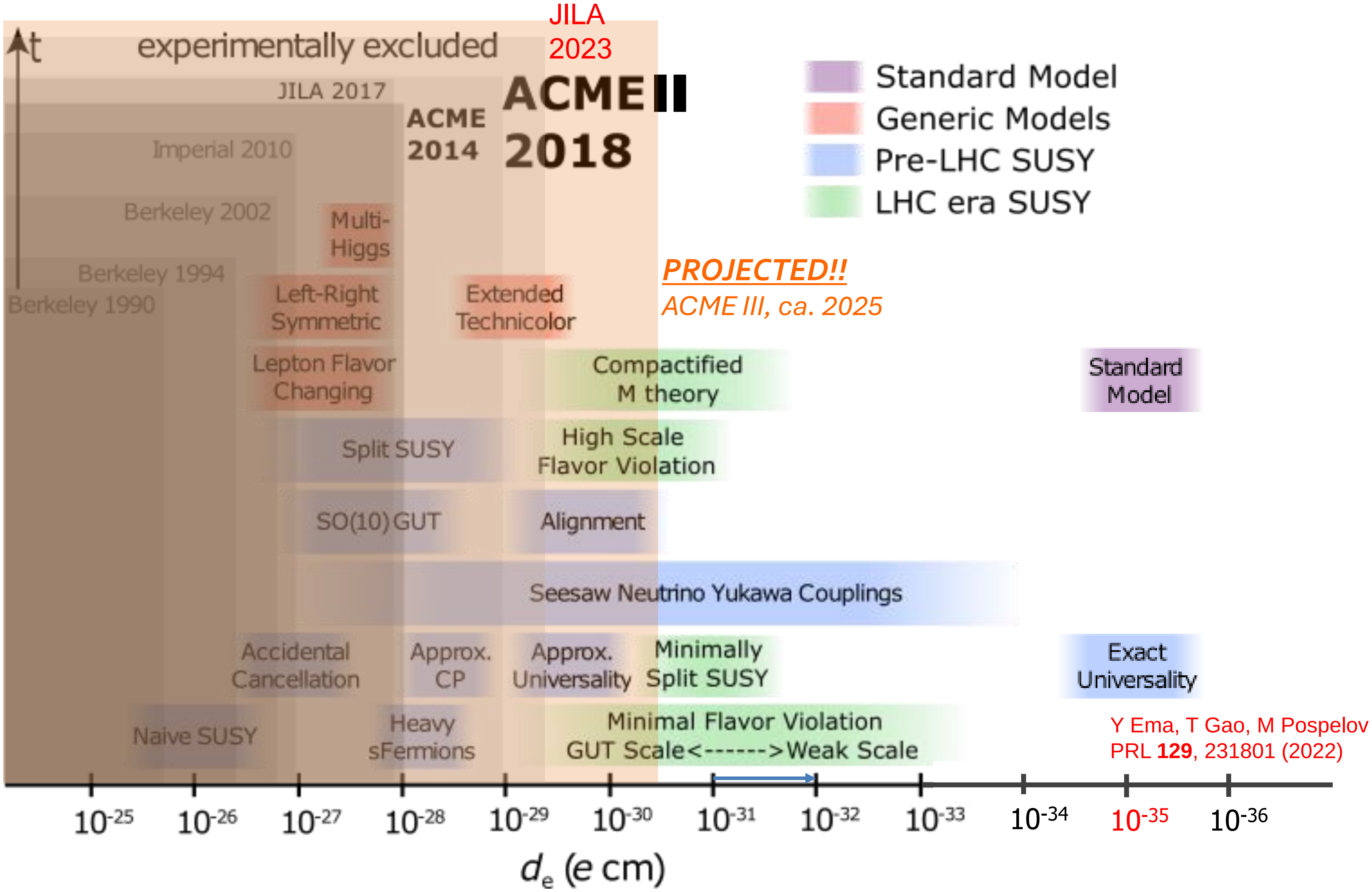
Search for new physics via electron EDM



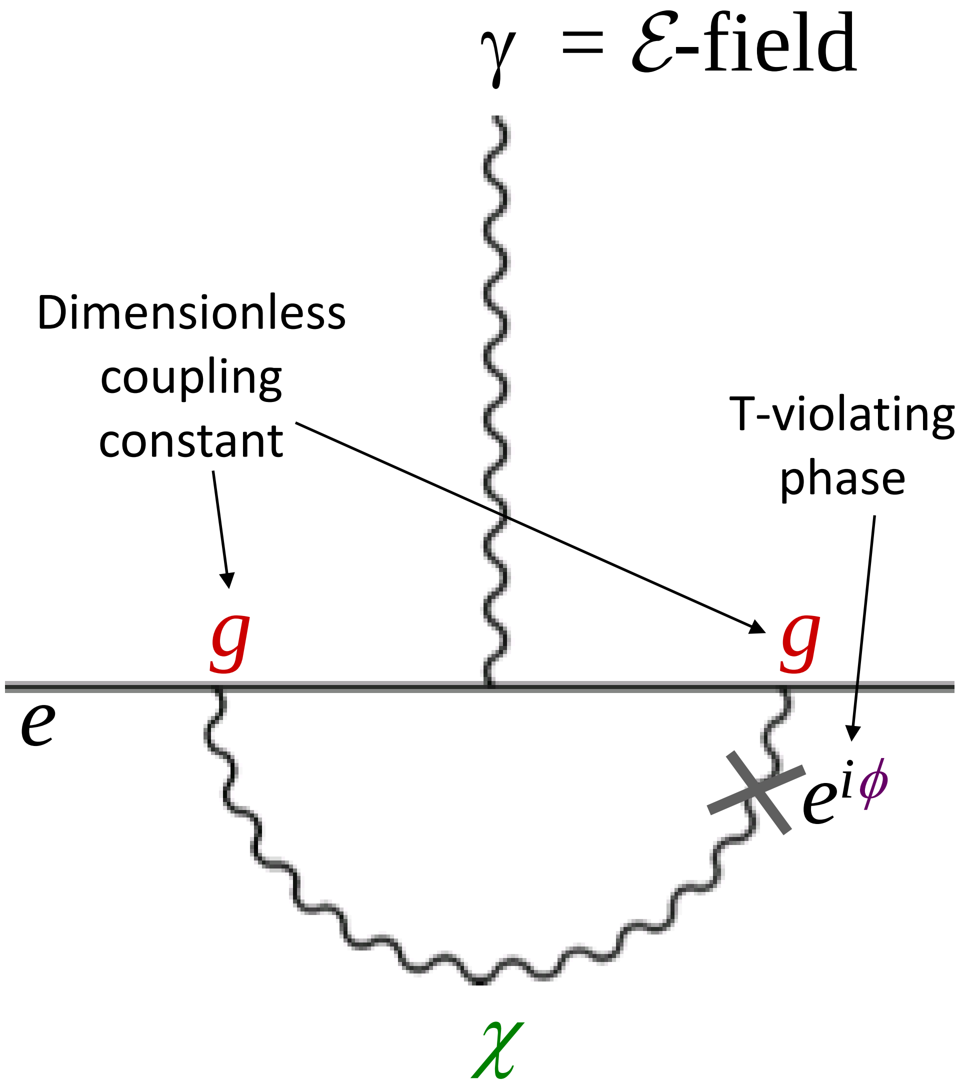
JILA
2023

PROJECTED!!

ACME III, ca. 2025

Y Ema, T Gao, M Pospelov
PRL **129**, 231801 (2022)

Crude dimensional estimates for eEDM



(new heavy particle)

typical e-*EDM*

$$d_e \sim e\left(\frac{g^2}{2\pi}\right)^N \frac{m_e}{m_\chi^2}\sin\phi$$

N = # loops

“natural” assumptions

$g^2/\hbar c \approx \alpha$

$\sin(\phi) \sim 1$

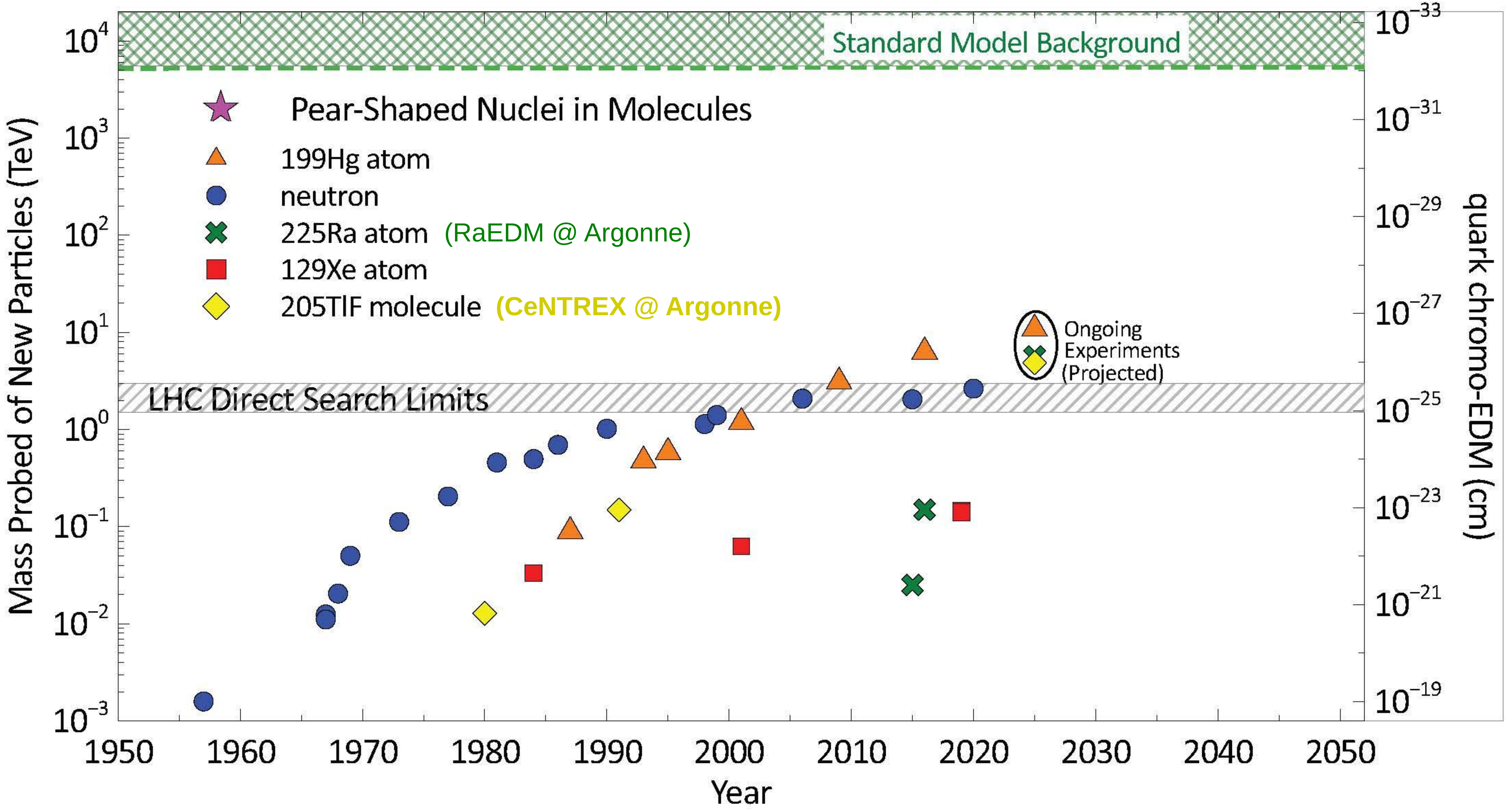
$m_\chi \sim 50$ TeV

$\Downarrow$

d_e ~ current limit

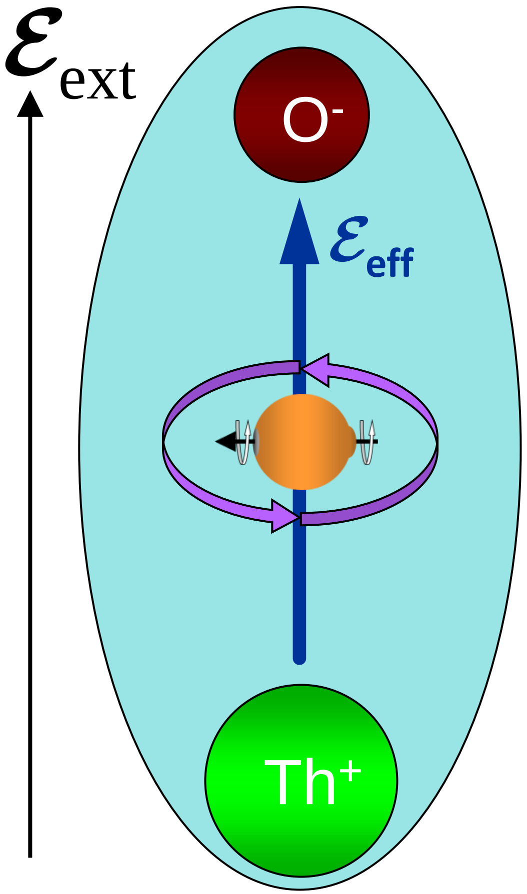
(*N* = 1 loop)

Limits on quark chromo-EDMs from hadronic "EDM" experiments



Polar molecules amplify observable effect of EDMs

Easily polarized → **10^3-10^4x enhanced sensitivity** vs. atoms



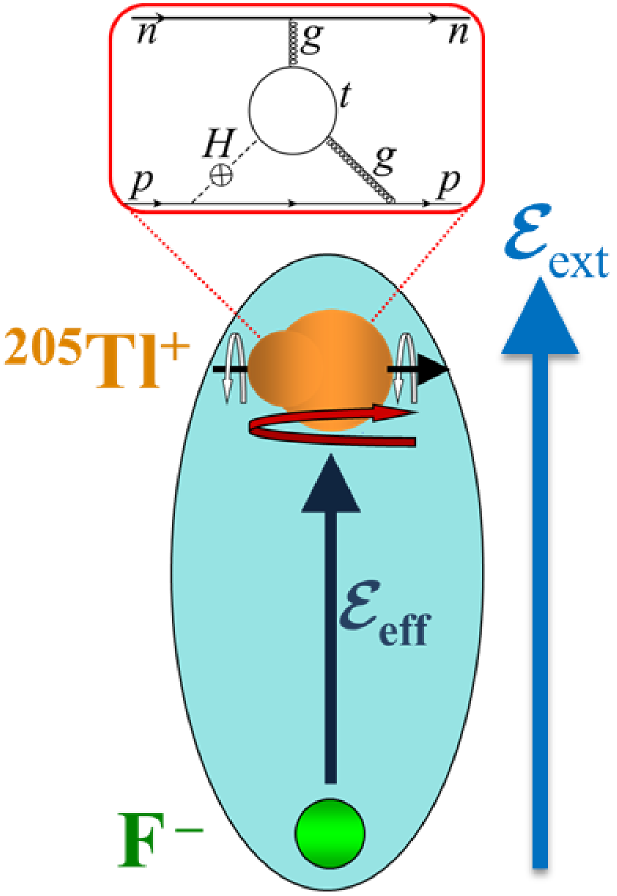
Observable

energy shift

$$\Delta E_{P,T} \propto Z^3$$

in both cases

$$\mathcal{E}_{\text{eff}} \equiv \Delta E_{P,T} / d$$



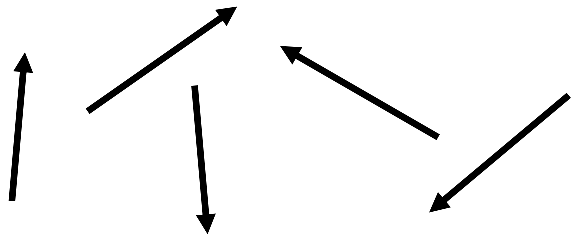
Electron EDM

in ThO*, HgF^{+*}, YbF, RaF, YbOH…

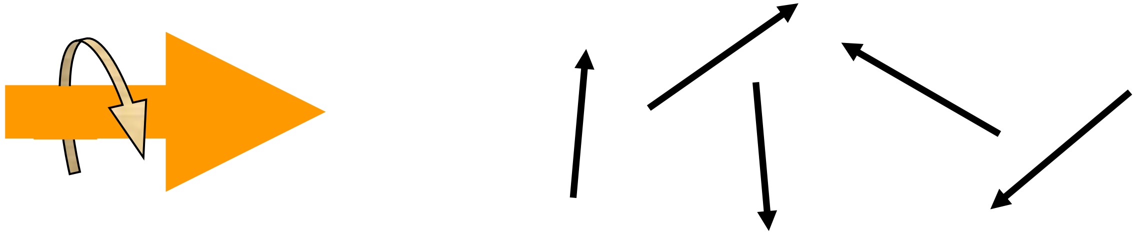
Nuclear Schiff moment ≈ "EDM" in TlF, …

Magnetic Quad. Moment in YbOH, …

General method to detect an "EDM"



General method to detect an "EDM"



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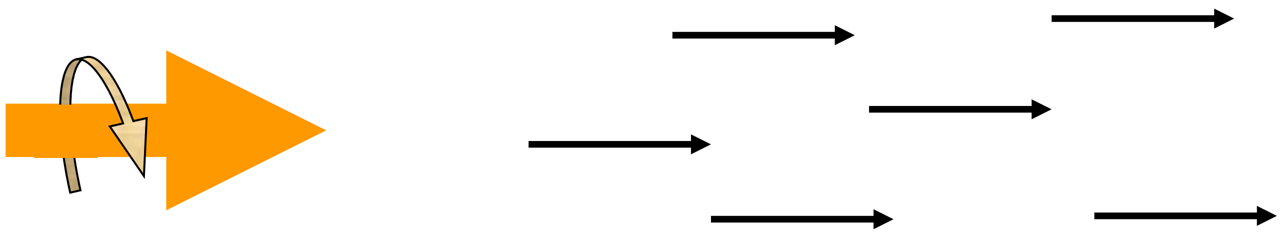

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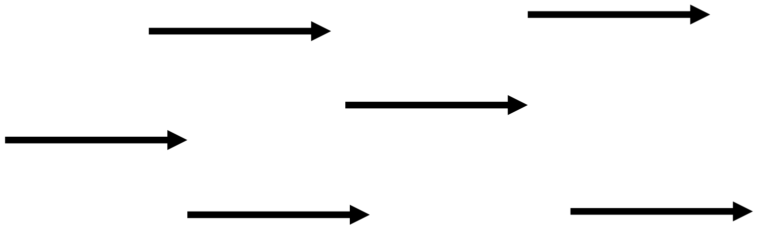

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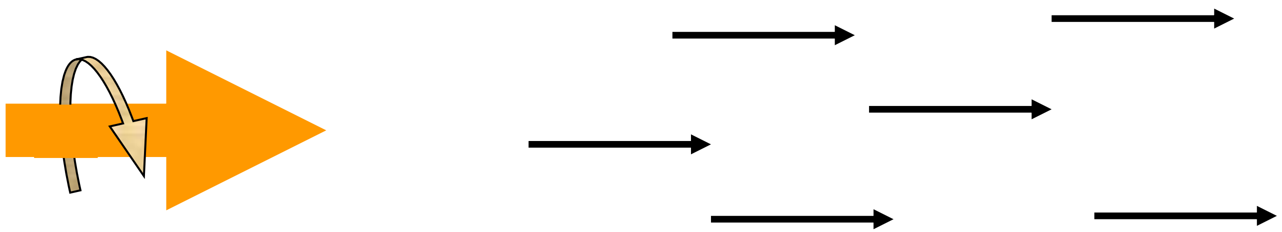
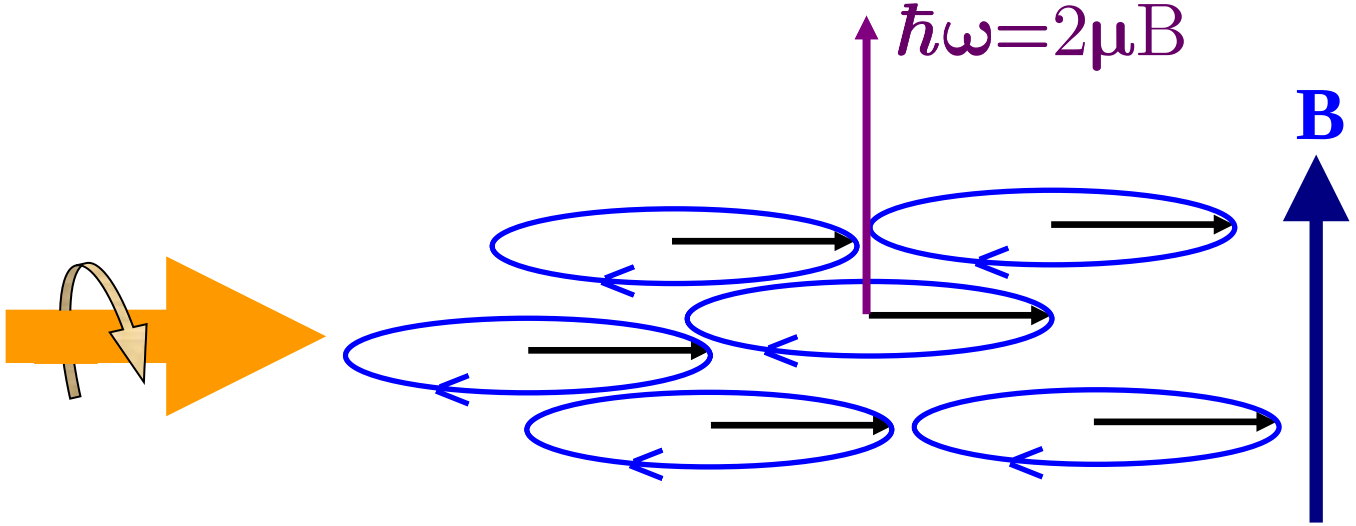
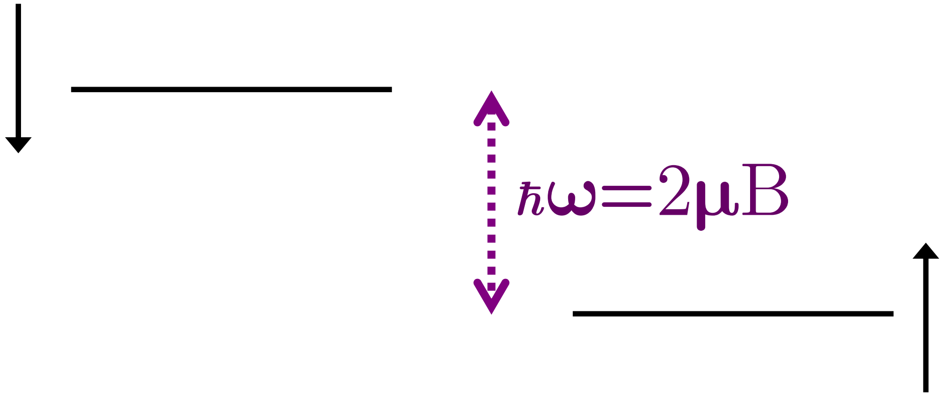


Energy level picture:

General method to detect an "EDM"



Energy level picture:



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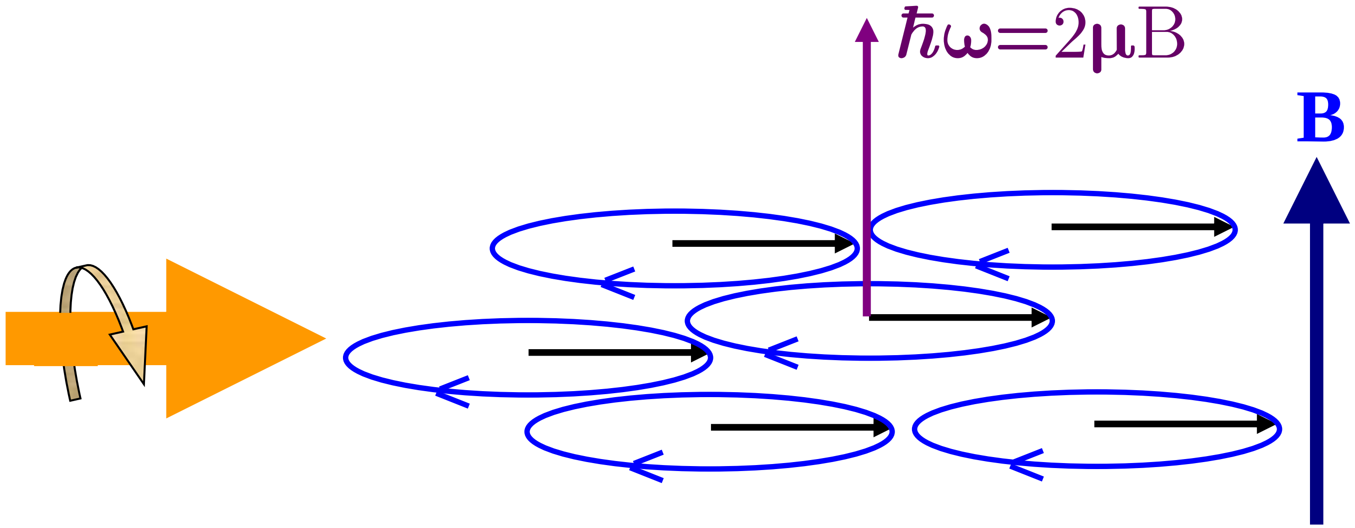
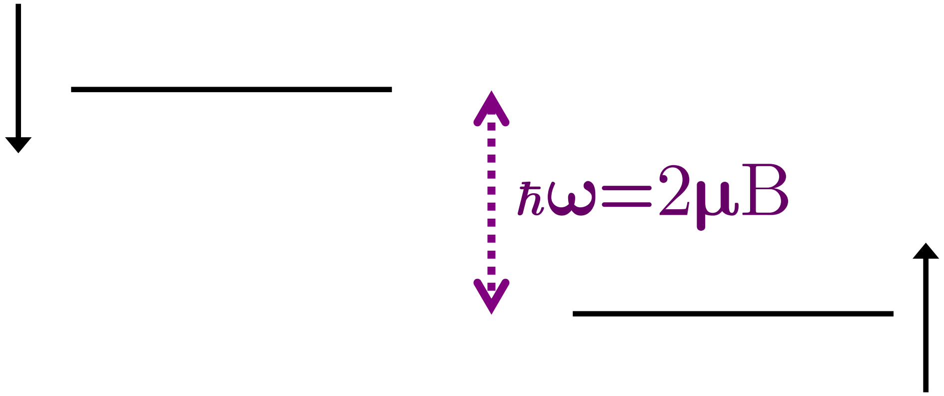
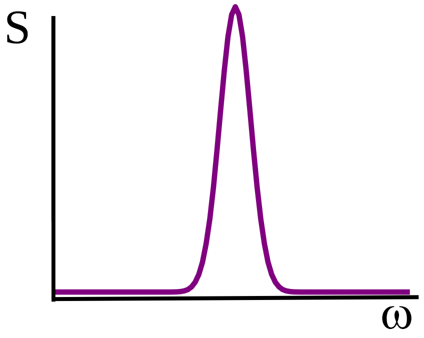


Energy level picture:





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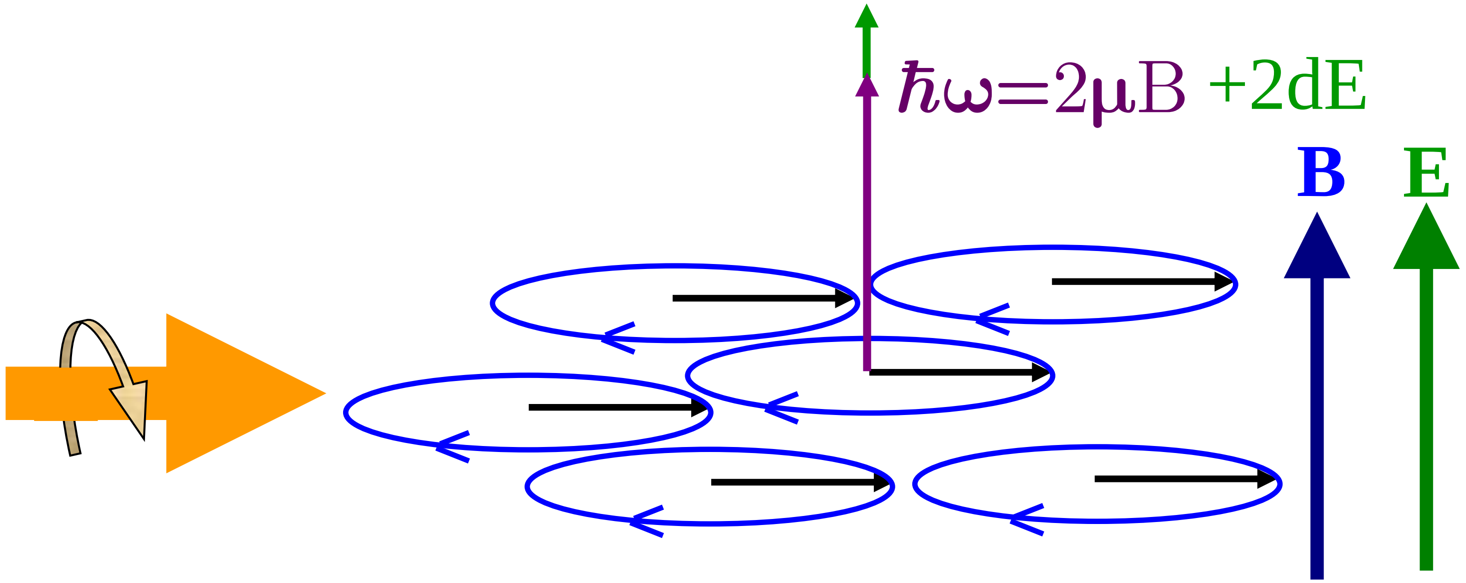

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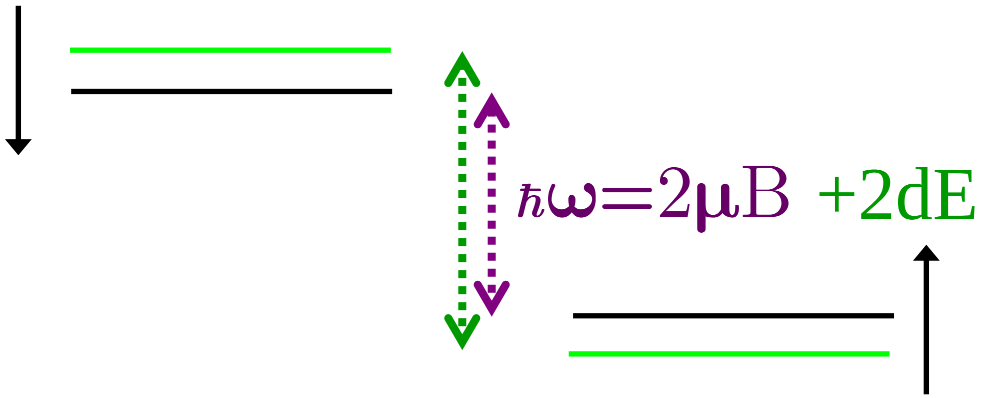

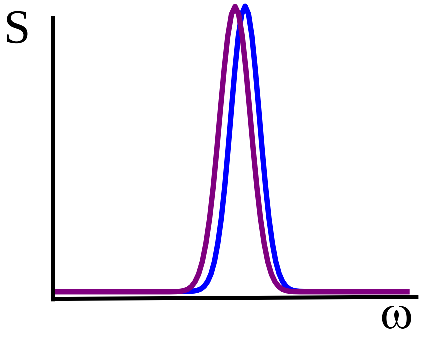

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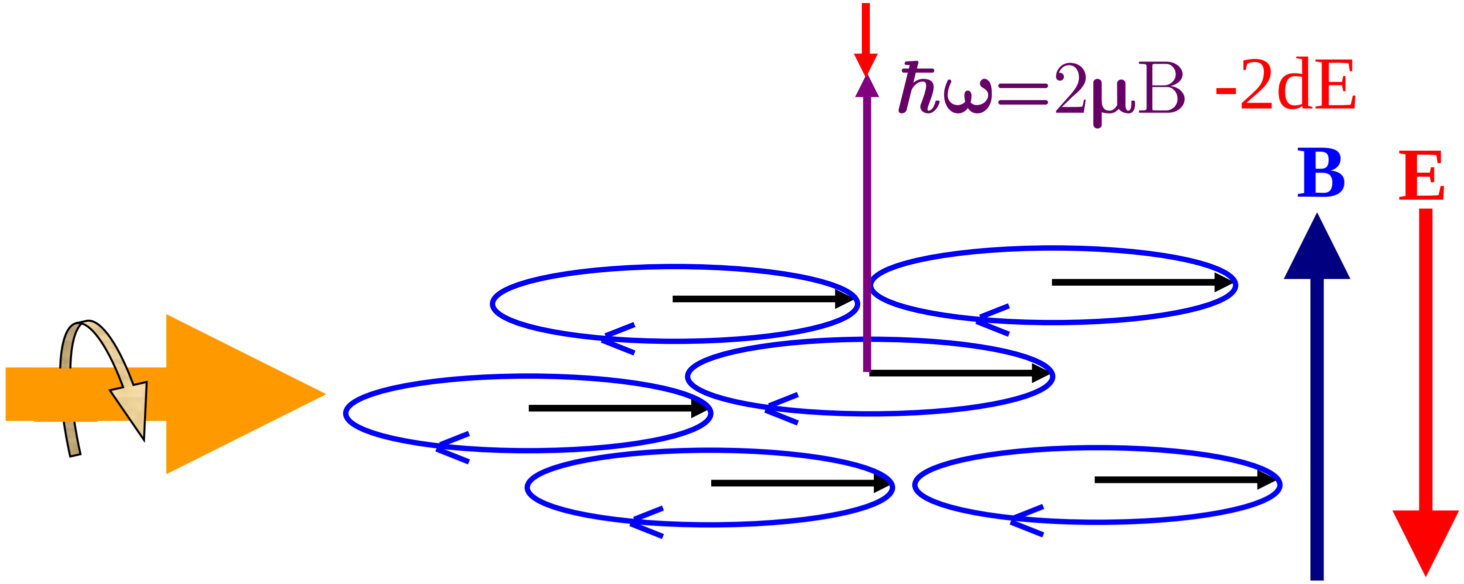
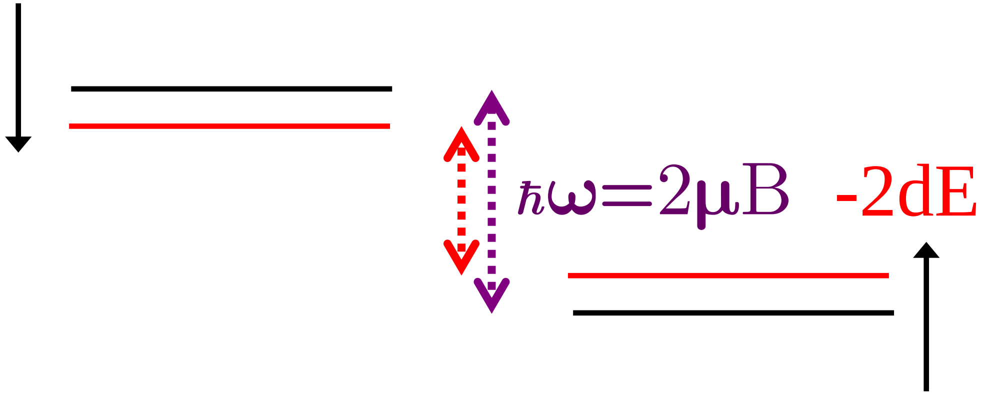
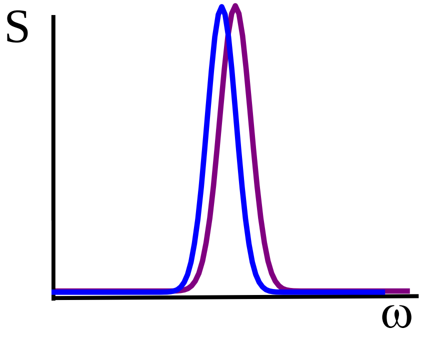


Energy level picture:





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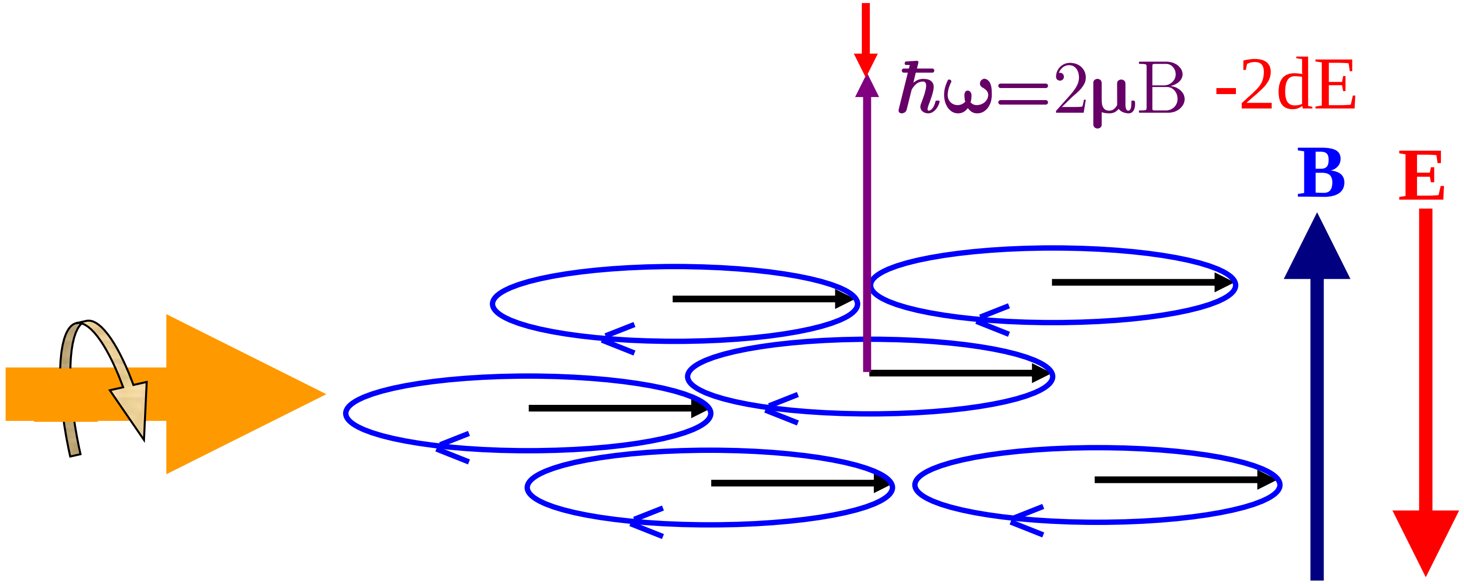
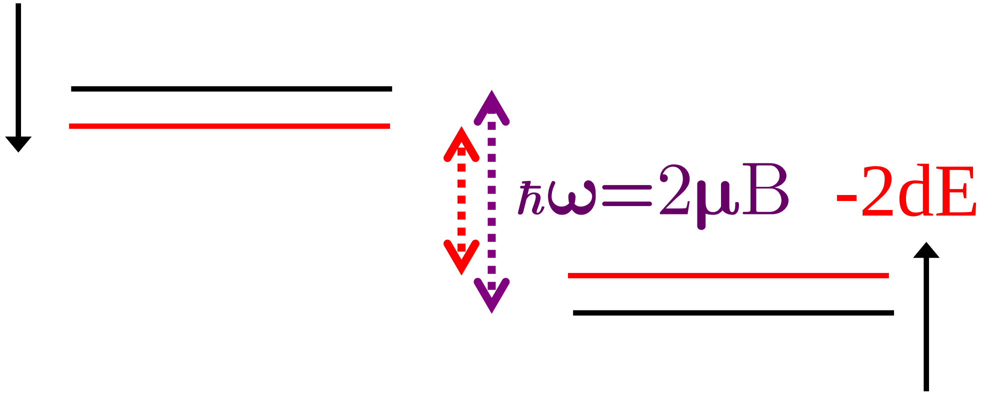


Energy level picture:



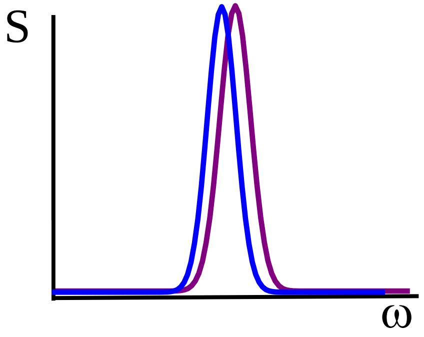


Figure of merit for
Statistical sensitivity:

$$\frac{shift}{resolution} = \frac{d\mathcal{E}}{\left(\tau_{coh}\right)^{-1}\left(S/N\right)^{-1}} \propto \mathcal{E} \cdot \tau_{coh} \cdot \sqrt{\dot{N} \cdot T_{\text{int}}}$$

ACME III collaboration

THE UNIVERSITY OF CHICAGO

Northwestern University

HARVARD UNIVERSITY

OKAYAMA UNIVERSITY

University of Chicago
David DeMille (PI)
Zhen Han (grad student)
Peiran Hu (grad student)

Northwestern University
Gerald Gabrielse (PI)
Xing Fan (postdoc)
Siyuan Liu (grad student)
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Harvard University
John Doyle (PI)

Okayama University
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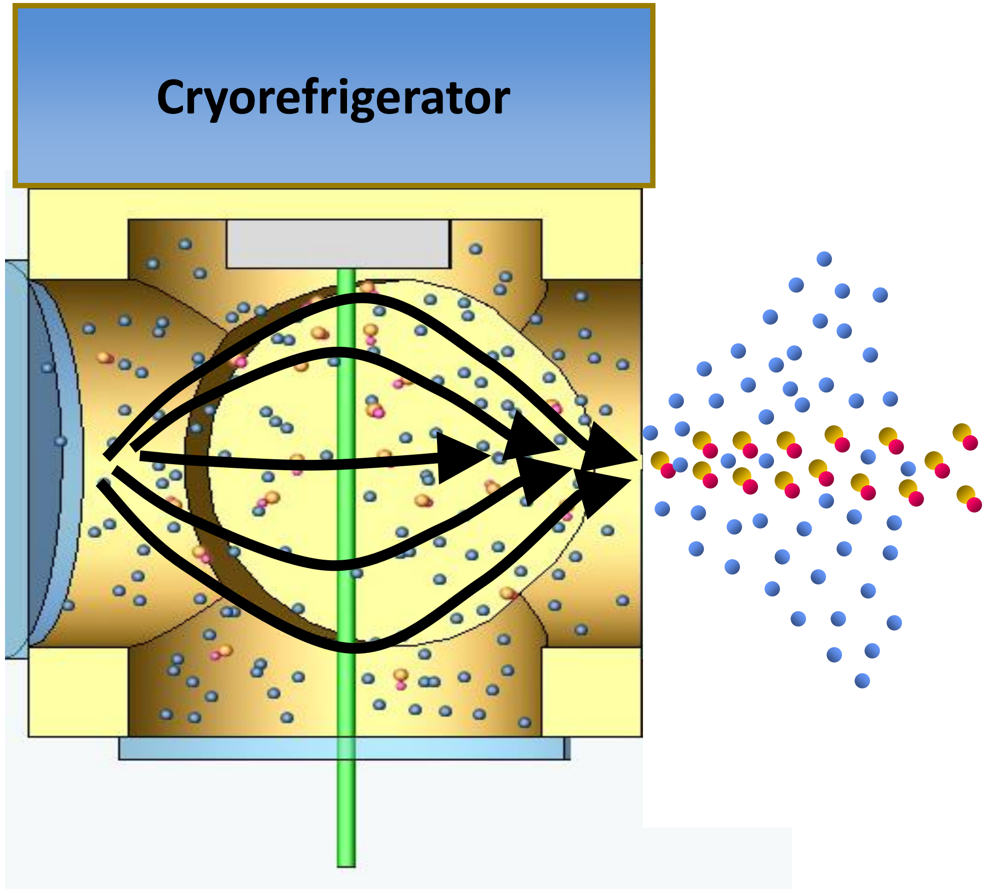
Koji Yoshimura
Noboru Sasao
Satoshi Uetake

Other collaborators
Cris Panda (Berkeley)
Nick Hutzler (Caltech)
Xing Wu (Michigan St.)

New molecular beam technology: hydrodynamically enhanced cryogenic buffer gas beam

[Maxwell *et al.* PRL 2005; Patterson & Doyle JCP 2007;
Barry *et al.* PCCP 2011; Hutzler *et al.* PCCP 2011]

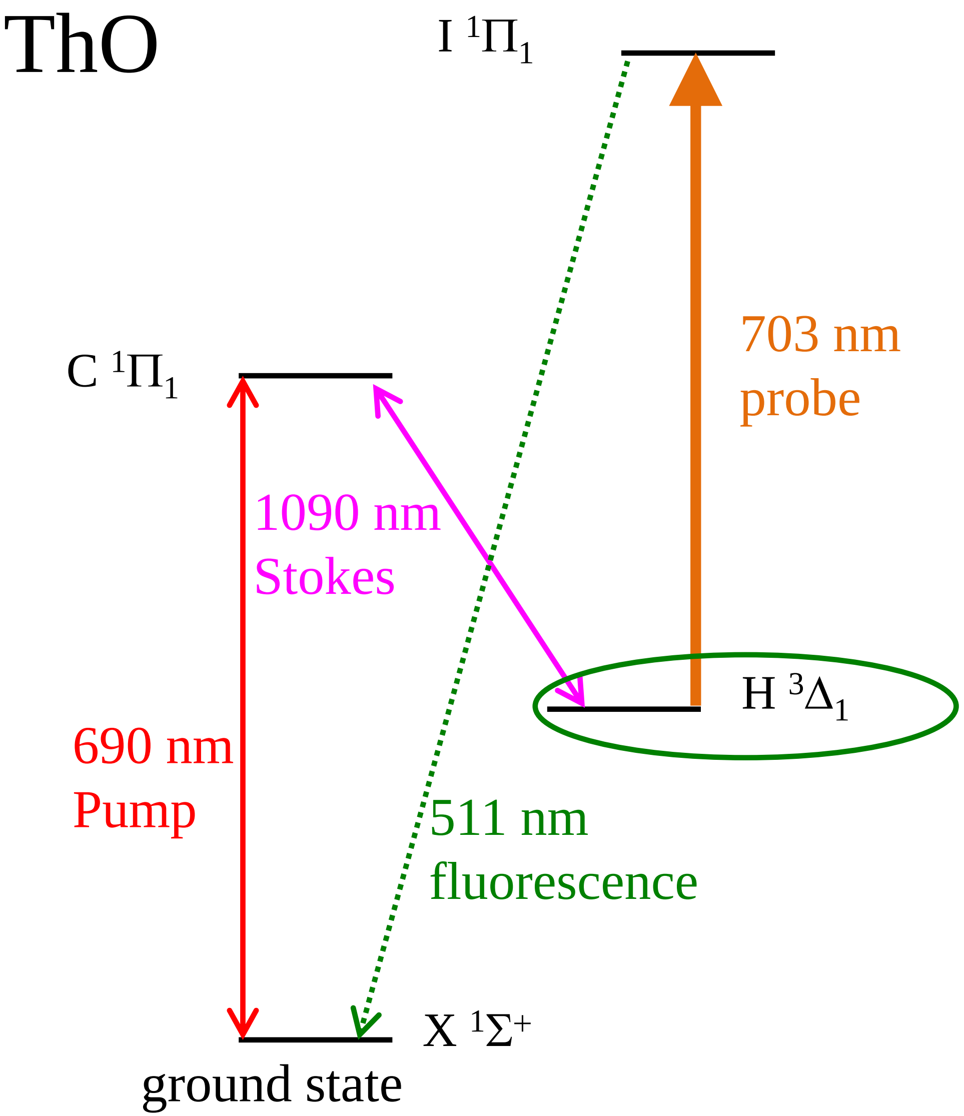


- Inject hot molecules (e.g. via laser ablation)
- Cool w/cryogenic buffer gas *@high density*
- Efficient extraction to beam via "wind" in cell: $10^{-3} \rightarrow >10\%$
- "Self-collimated" by extraction dynamics
- Rotationally cooled by supersonic expansion
- **Cold** (~1-4 K) & **moderately slow** (v ~ 150-200 m/s)

Beam brightness ***~10^3 × larger*** than prior sources for refractory/free radical species

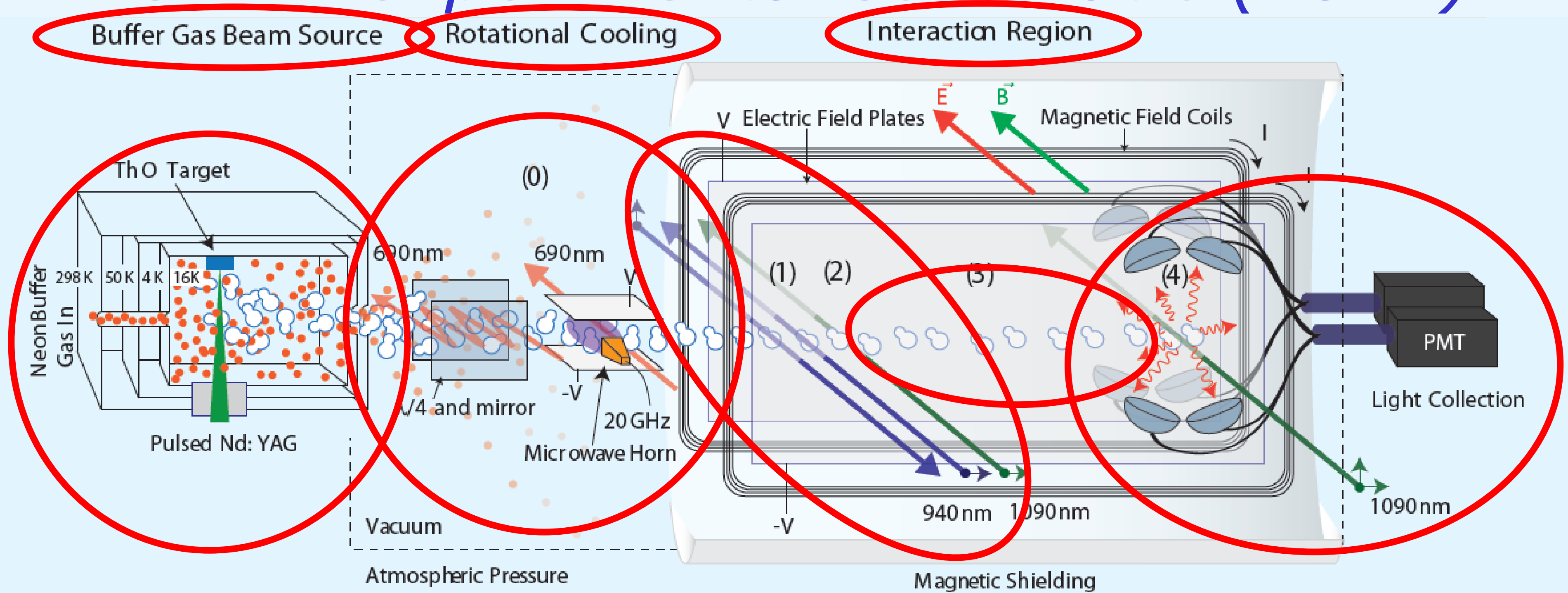
*“New” molecular species: ThO**

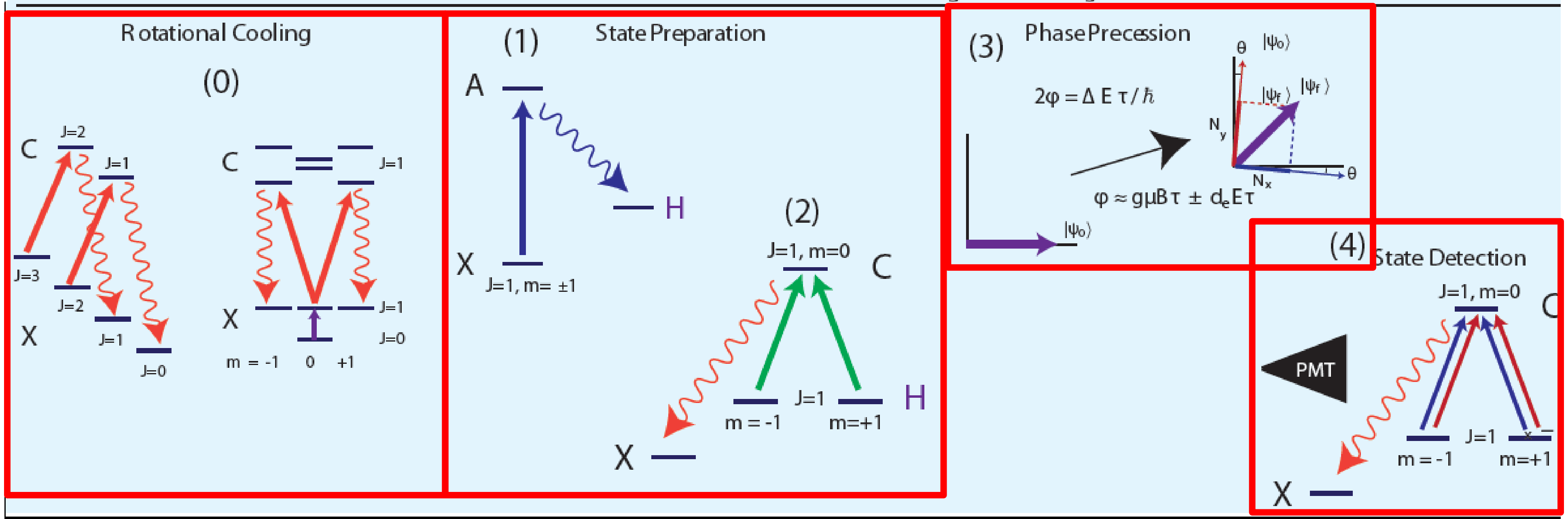
[A.C. Vutha *et al.* J. Phys B 2010]



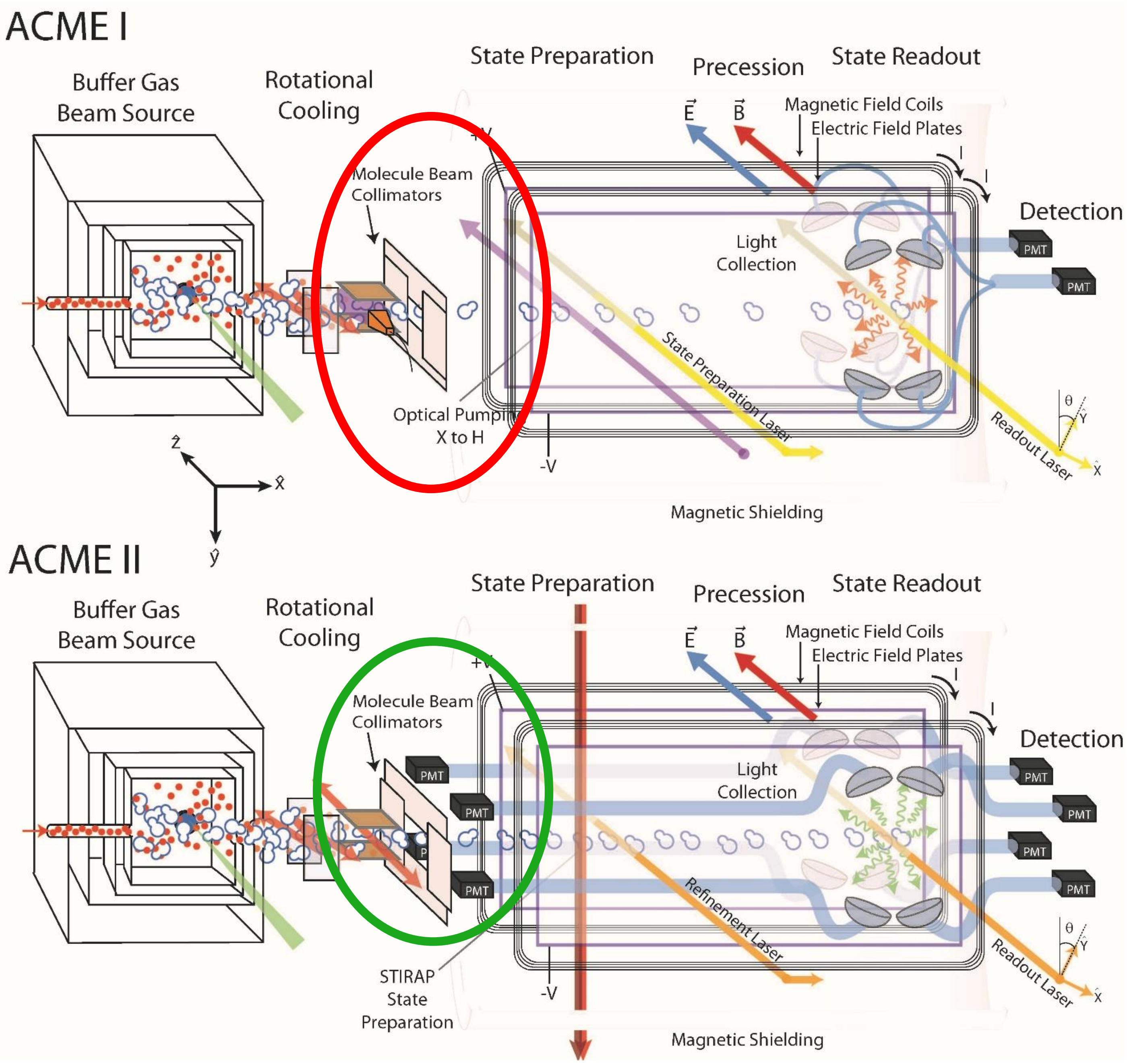
- **Sufficient coherence time for measurement**
  in *metastable* state $H\ ^3\Delta_1$
- **Largest effective internal $\mathcal{E}$:** ~80 GV/cm
  [Skripnikov et al. (2016), Fleig & Nayak (2016)]
- **Suppressed magnetic moment**
  <0.01 μ_B in $H\ ^3\Delta_1$ reduces *B*-field systematics
  [Idea: Meyer, Bohn, Cornell *et al.* (JILA);
  Measured: A.C. Vutha *et al.*, PRA 2011]
- **Omega-doublet co-magnetometer**
  suppresses many possible systematics
  & requires only very modest polarizing $\mathcal{E}$-field
- All spectroscopic data previously known
- State preparation and readout
  w/standard, robust diode & fiber lasers
- Blue-shifted fluorescence from probe laser
  ⇒no problem with backgrounds
- **High beam source yield**

ACME I experimental schematic (2014)





ACME I→ II upgrades to increase signal

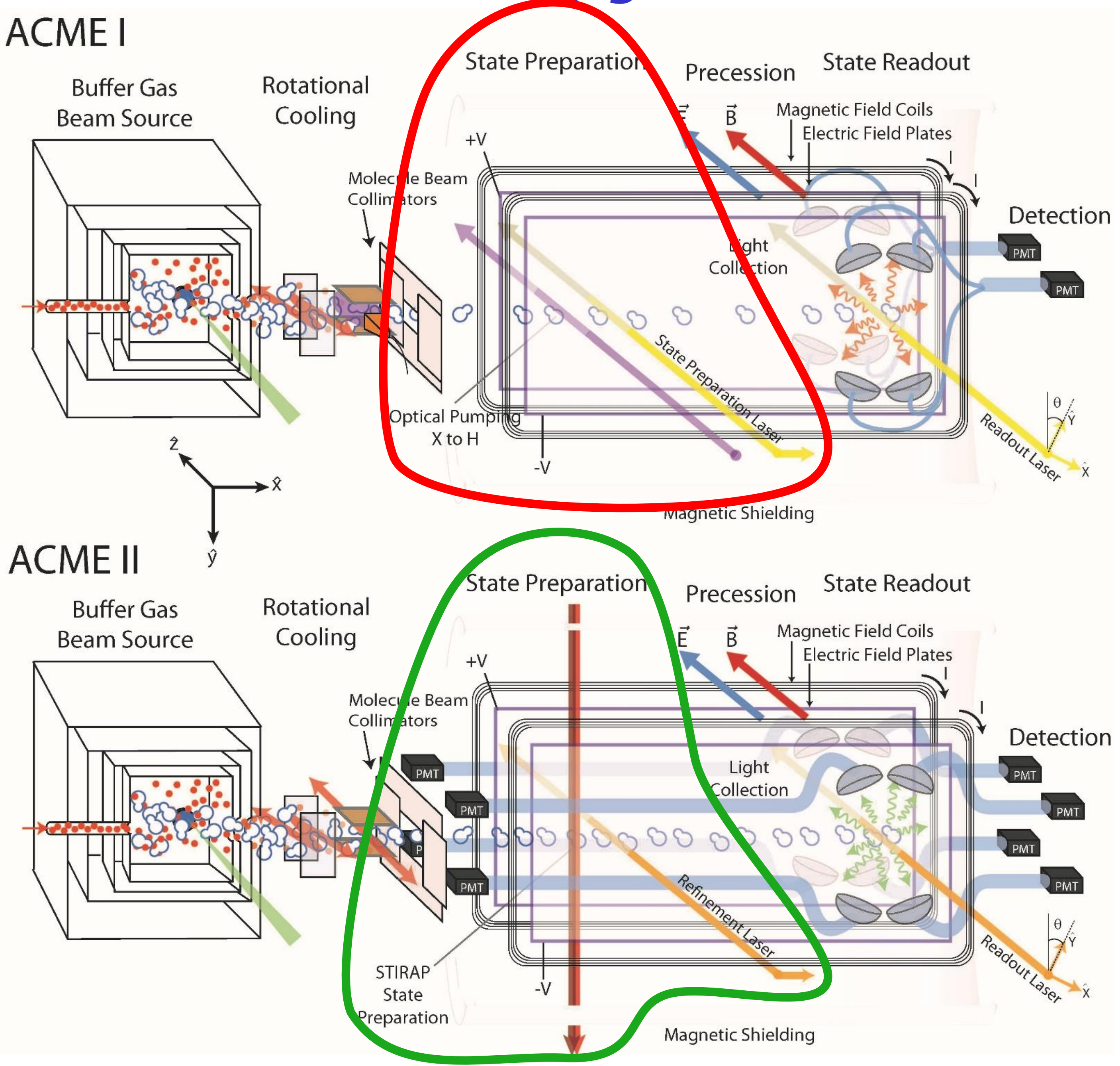


Larger detection region
+ Increased angular acceptance of molecular beam

⇓

x8 Signal

ACME I→ II upgrades to increase signal



Incoherent
(opt. pumping)
state pr

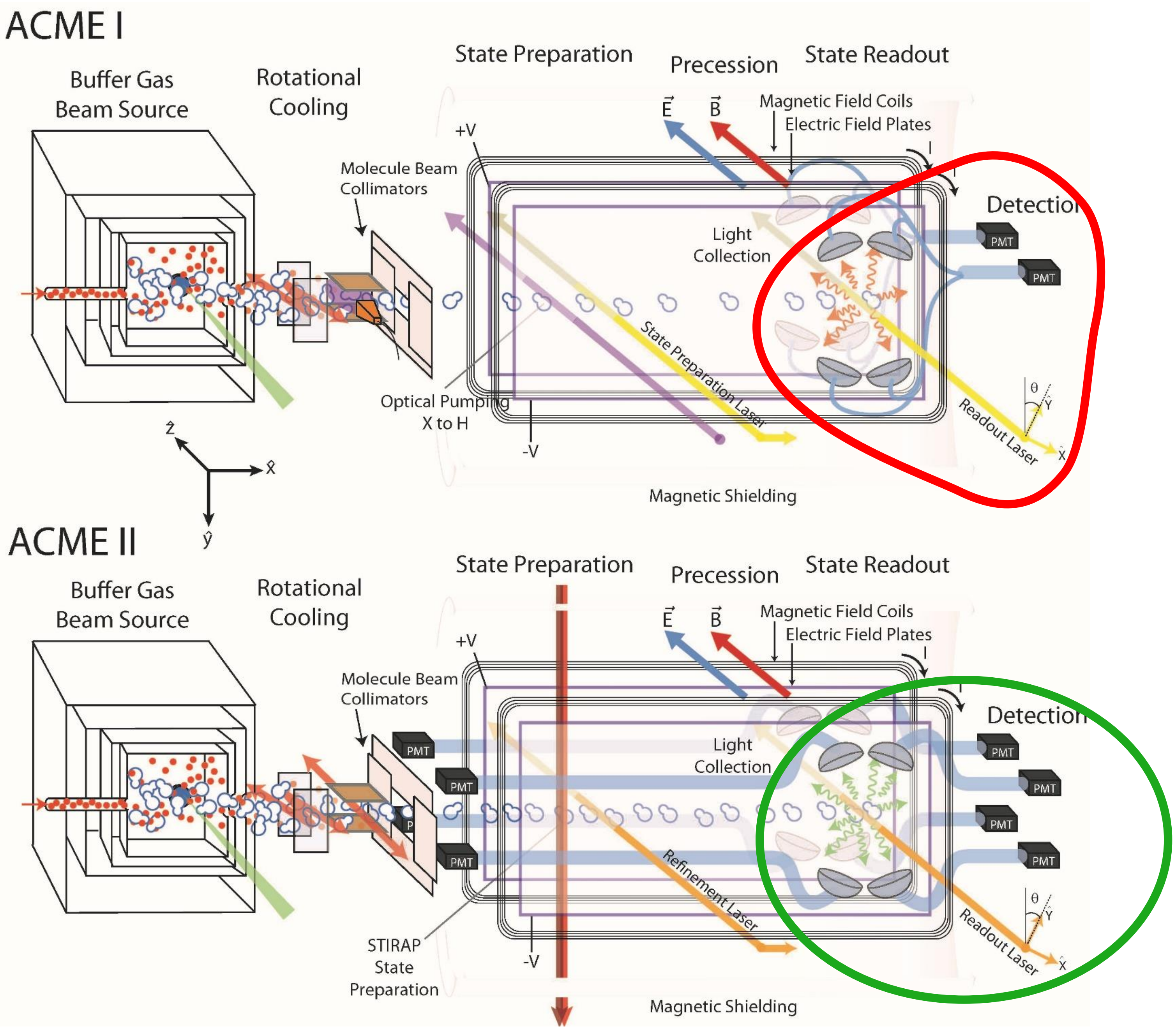
eparation

vs.

Coherent
(STIRAP)
state preparation
⇓
x12 Signal

C. Panda *et al.*, PRA (2016)

ACME I→II upgrades to increase signal

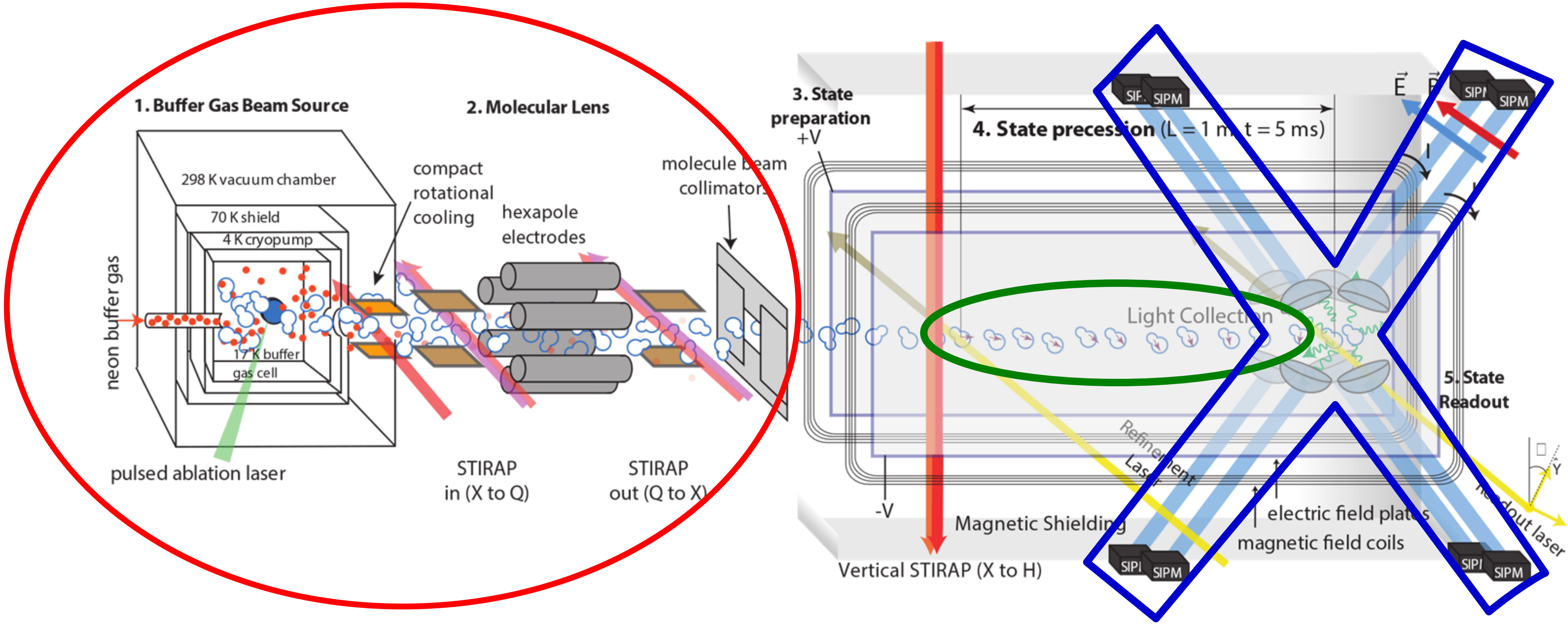


Better detection:
new probe transition →
green vs. red photons,
better PMTs
&
improved optics

⇓

x4 Signal

ACME III approach



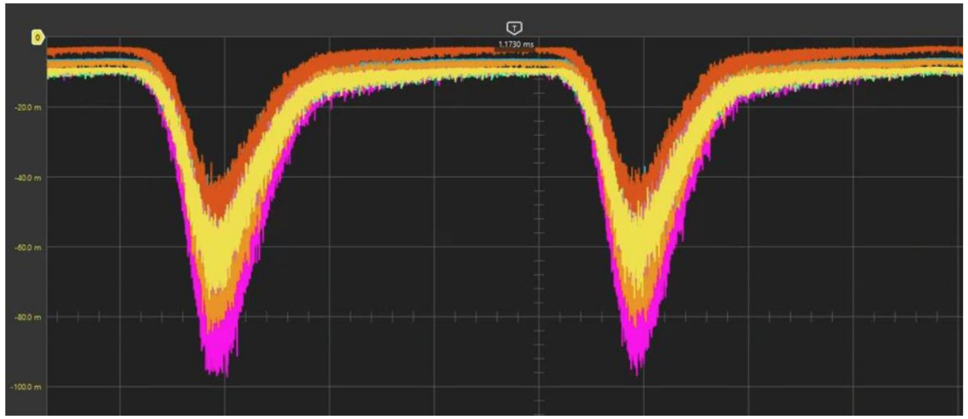
- More useful molecules via molecular lens, improved beam source, etc.
- ~5x longer interaction time via improved state lifetime info
- Better detection efficiency via better detectors & collection optics
- Additional technical improvements to reduce noise & systematic errors

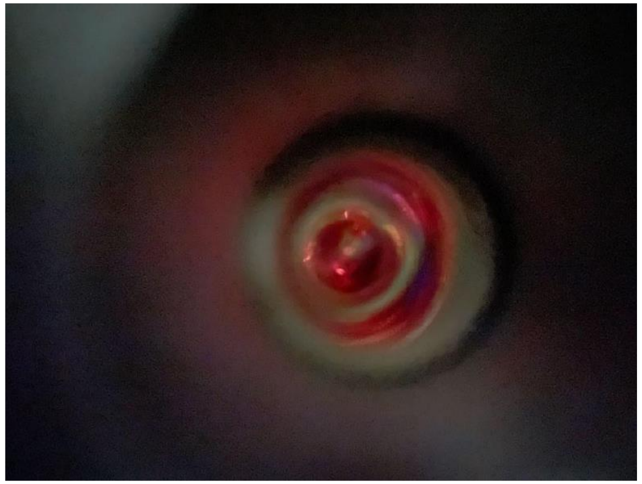
__Bottom line__:

~30× anticipated improvement in sensitivity

Status of ACME III

- Critical new subsystems tested at full-scale, full installation nearly complete
- Control of most important known sources of systematics demonstrated at required levels
- Coming soon: system integration & transition to EDM data taking
- Excellent prospects to probe electron EDM at level $d_e < 3\text{x}10^{-31}\ e\cdot\text{cm}$
  $\Rightarrow$**probe scales > 100 TeV** in simple models
- **~4 weeks ago: first signals from ACME III full beamline, detection rate ≥ anticipated!**

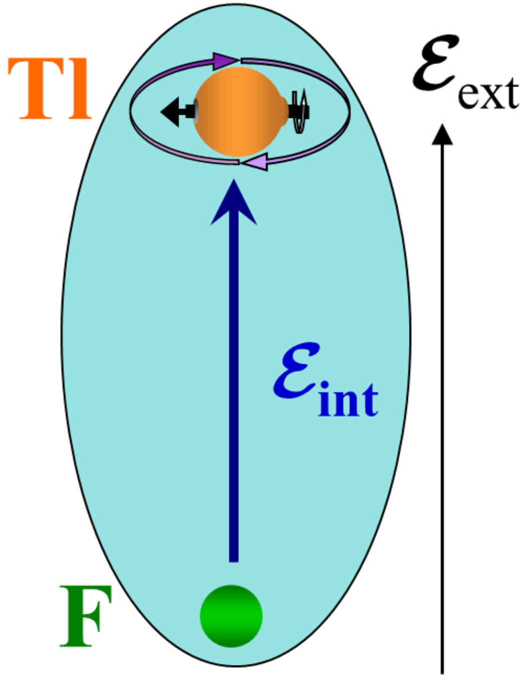




CeNTREX: Cold molecule Nuclear Time-Reversal Experiment

(with T. Zelevinsky [Columbia], D. Kawall [UMass])

New molecule-based search for **nuclear Schiff moment of ^{205}Tl**



- State-of-the-art Schiff moment limit from ^{199}Hg experiment (Seattle)
  Already sensitive to new physics at ~5 TeV scale

- Similar to *e*-EDM, signals amplified vs atoms
  → nuclear spin precession in ^{205}TlF molecules **~10^4× larger than in ^{199}Hg atoms**

- ^{205}Tl nucleus has unpaired proton
  → orthogonal sensitivity to new underlying physics vs. other current experiments
  (e.g. ^{199}Hg, ^{171}Yb, ^{129}Xe have unpaired neutrons)

GOAL: use molecular "enhancement" for
improved sensitivity to ***hadronic*** CP-violating interactions at >TeV scale

CeNTREX Team

Principal Investigators


David DeMille
(Argonne,
U Chicago)


David Kawall
(UMass Amherst)


Tanya
Zelevinsky
(Columbia)

Postdoc


Olivier Grasdijk
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Jianhui Li
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Yuanhang Yang
(UChicago)


Perry Zhou
(Columbia)


Emma McClure
(UChicago)

Former Ph.D. students


Konrad Wenz
(Columbia)

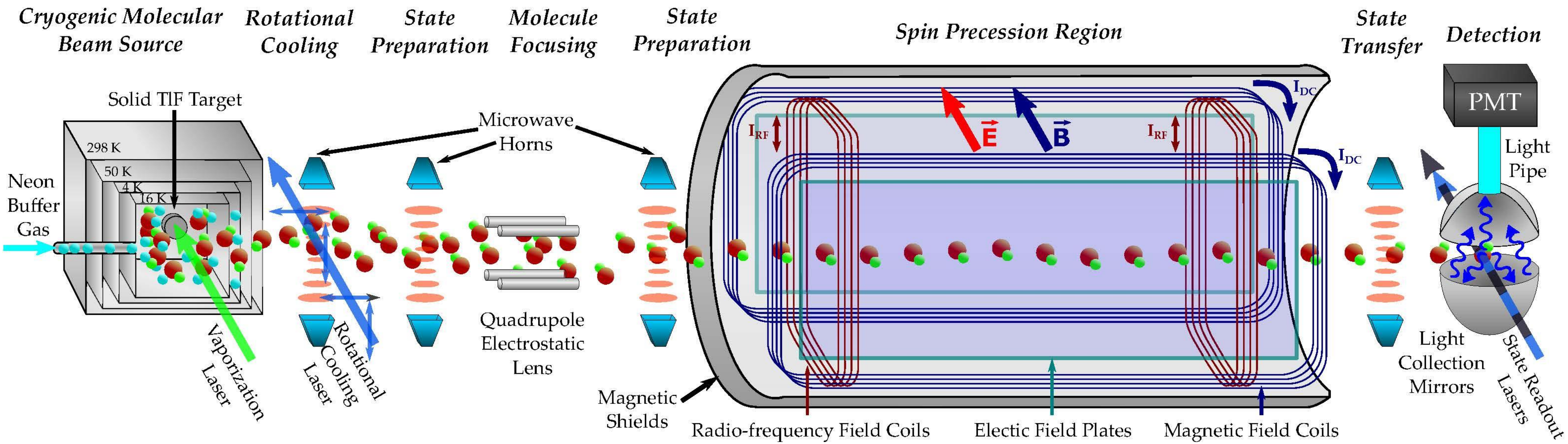

Oskari
Timgren
(Yale+UChicago)


Jakob
Kastelic
(Yale+UChicago)

CeNTREX "proton EDM" experiment schematic

Incorporates *many* methods from ACME III

(slow molecular beam, rotational cooling, adiabatic state transport, etc.)

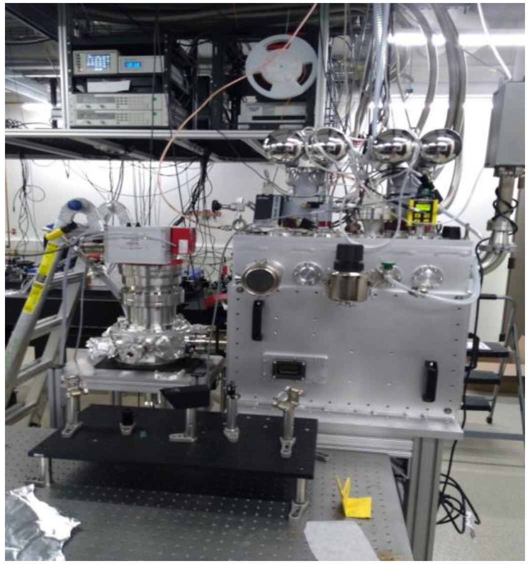


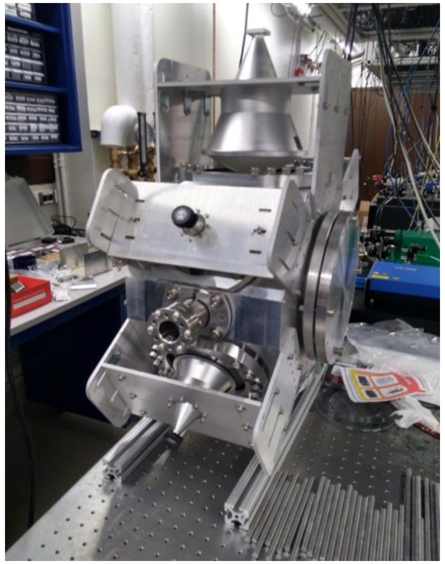
Design nearly complete, construction of most functional modules underway or complete

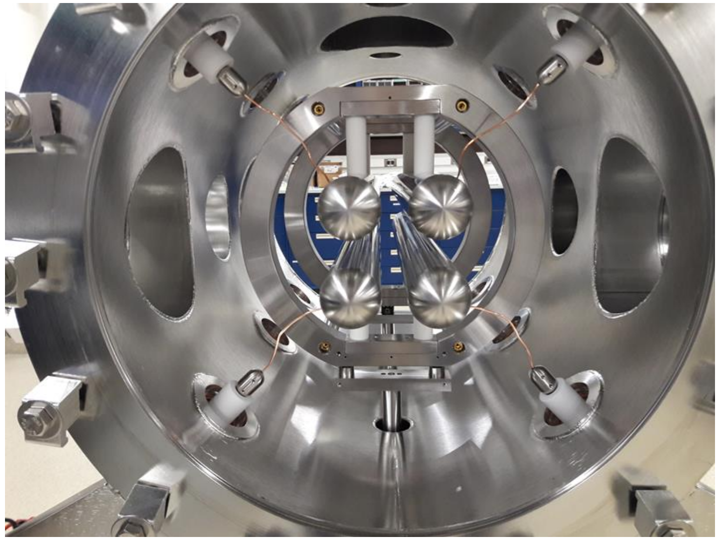
Conceptual details in J.O. Grasdijk *et al.*, Quant. Sci. Technol. (2021)

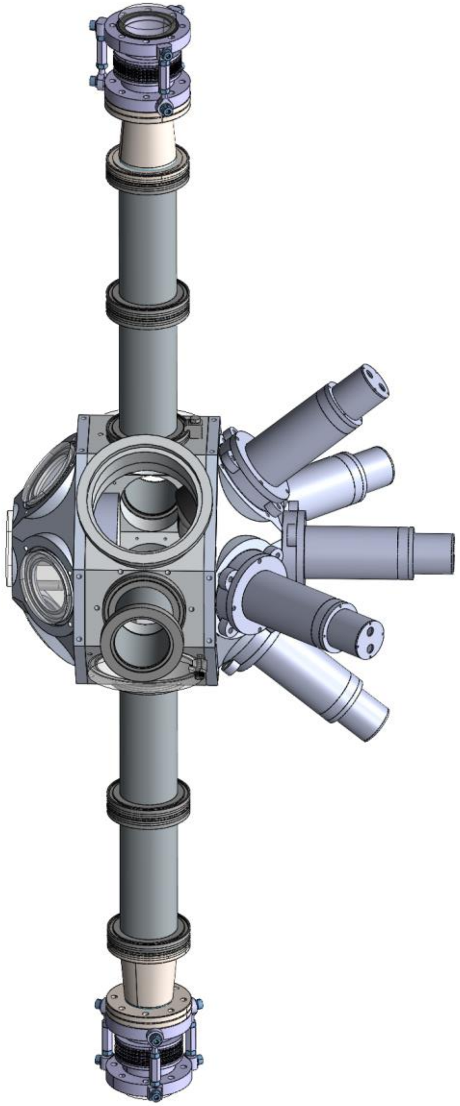
Argonne NATIONAL LABORATORY
U.S. DEPARTMENT OF ENERGY Office of Science
NSF
HEISING-SIMONS FOUNDATION

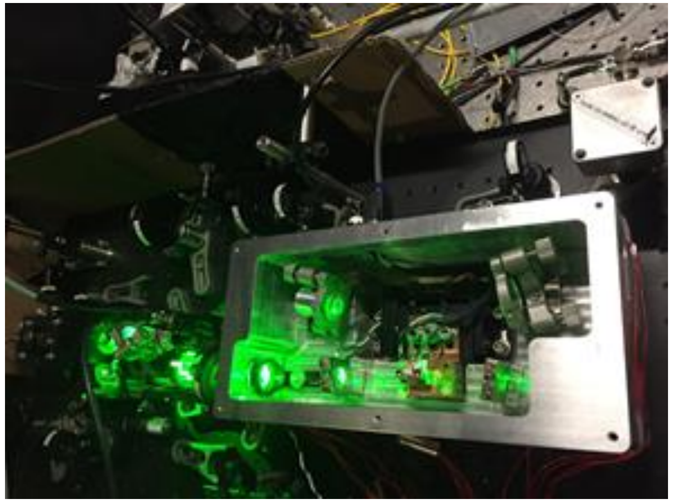
CeNTREX under construction @ Argonne

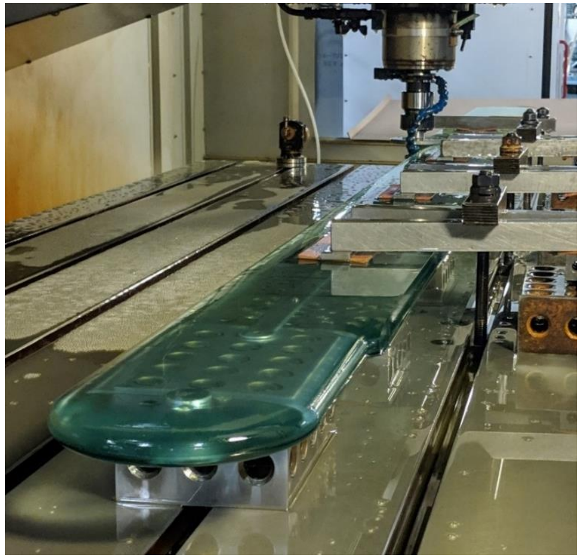

Cryogenic beam source

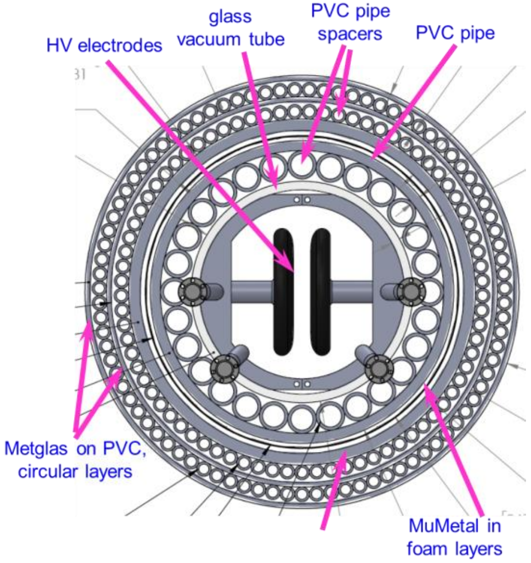

Rotational cooling


Electrostatic quadrupole lens




5x tunable UV laser systems


Machined glass electrodes



Magnetic shielding

Detection Region

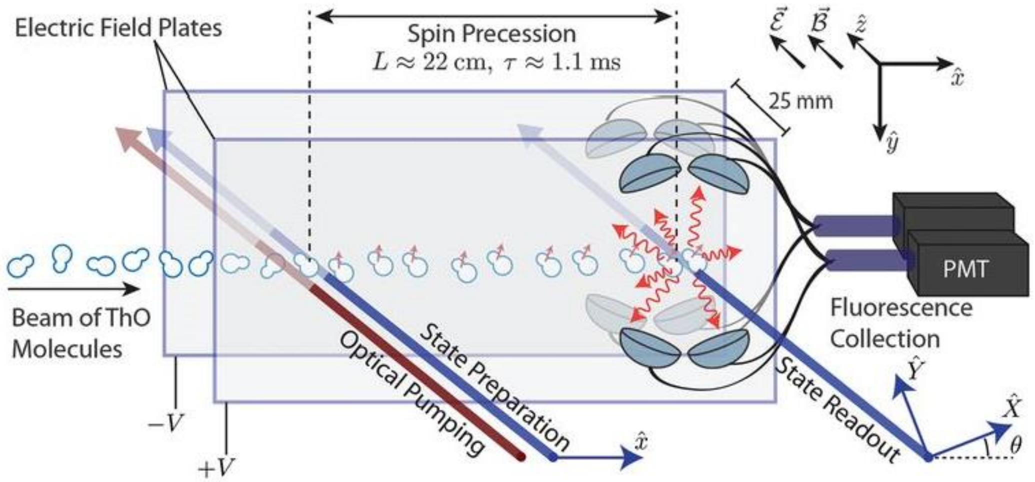
CeNTREX projected sensitivity vs. state of art hadronic EDMs

System	**^{199}Hg**	**n**	**^{205}TlF**	
Latest result	2016	2020	***Projected* ~2026**	***Projected* Improvement**
Sens. to QCD θ param. $\partial\nu/\partial\theta$	0.1 Hz	300 Hz	10^5 Hz	
Sens. to quark chromo-EDM $\partial\nu/\partial\tilde{d}_q$	1×10^{16} Hz/cm	2×10^{18} Hz/cm	2×10^{20} Hz/cm	
Sens. to p/n EDM $\partial\nu/\partial d_{p/n}$	1×10^{15} Hz/[e·cm]	2×10^{18} Hz/[e·cm]	6×10^{18} Hz/[e·cm]	
Limit on θ	$< 1.5\times10^{-10}$	$< 1\times10^{-10}$	$< 1\times10^{-11}$	**×10 (??)**
Limit on $\tilde{d}_q$ (cm)	$\tilde{d}_d$ $< 6\times10^{-27}$	--	$0.8\tilde{d}_u + 0.6\tilde{d}_d$ $< 1\times10^{-27}$	**×5***
Limit on d_n(e·cm)	$<1.6\times10^{-26}$	$<1.8\times10^{-26}$	Not competitive	
Limit on d_p(e·cm)	$<2\times10^{-25}$	Not competitive	$<2\times10^{-26}$	**×10**

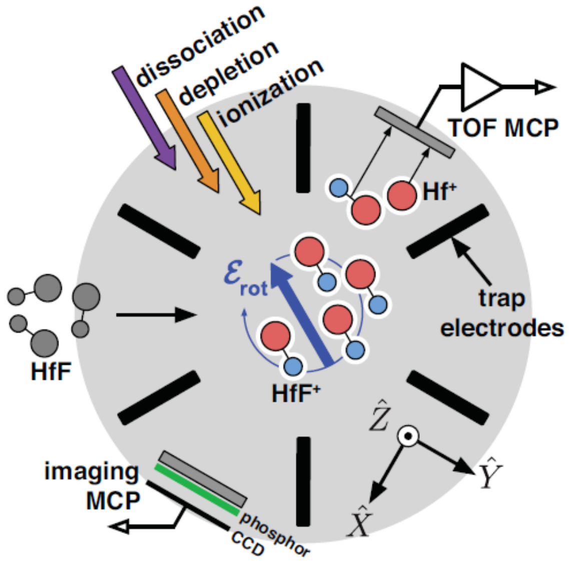
*sensitive to different linear combination of parameters vs comparisons

Cold molecules for EDMs now: $\lesssim$10K, fairly slow, dilute

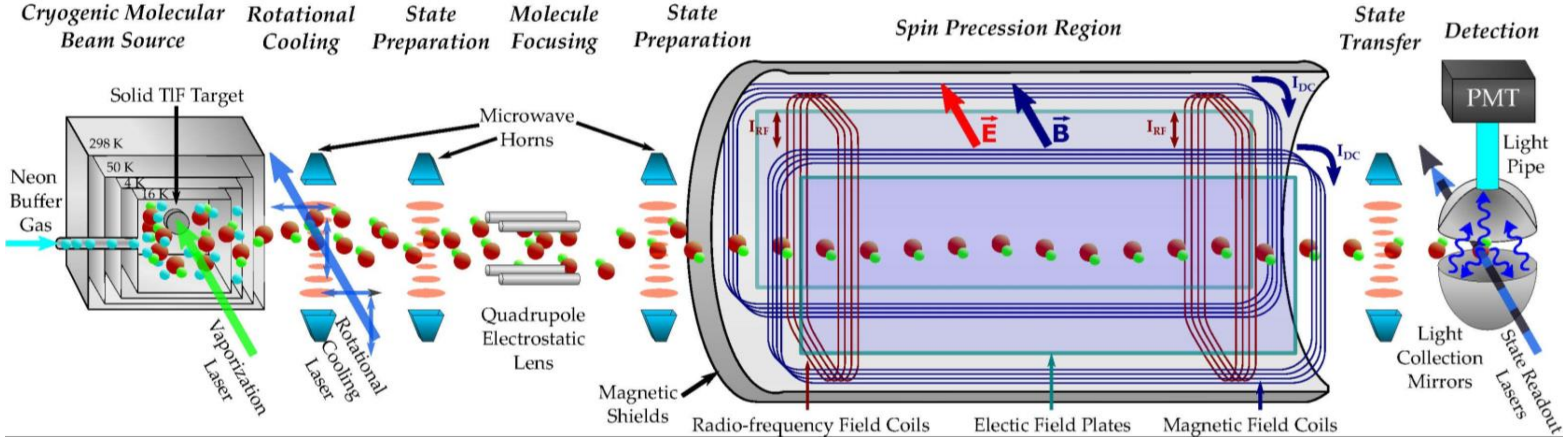
ACME II ThO cryogenic beam for eEDM



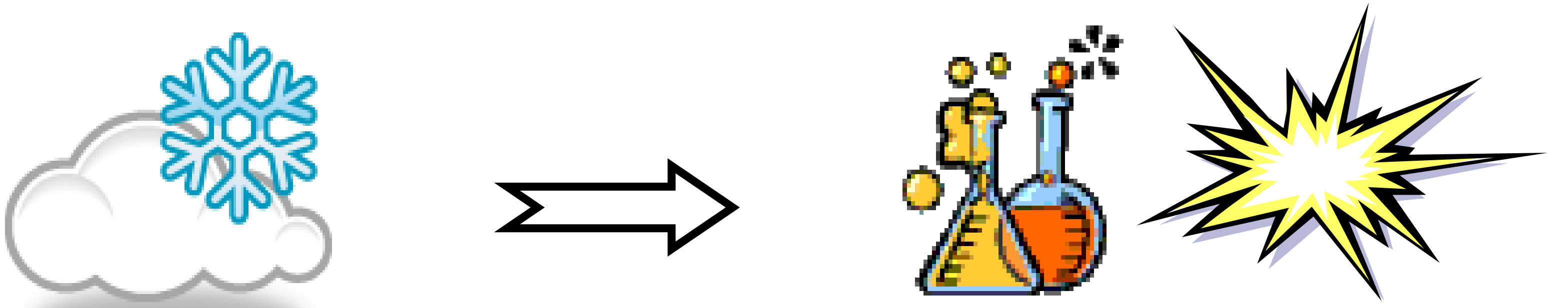
JILA trapped cryogenic HfF$^+$ ions for eEDM



CeNTREX TlF cryogenic beam for ^{205}Tl NSM



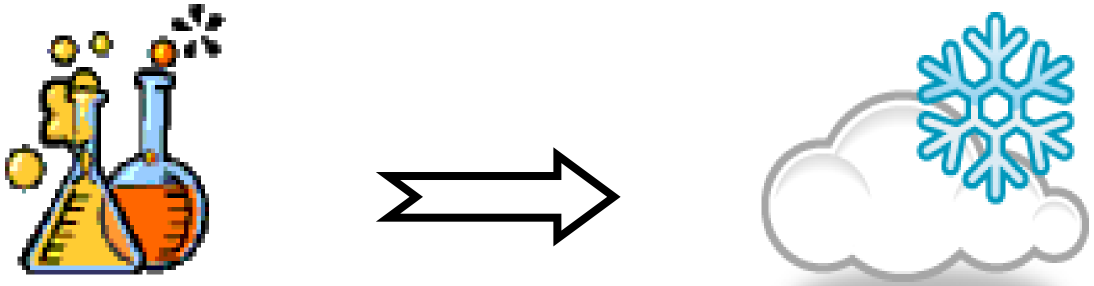
How to make ultracold molecules?



Cooling

Chemistry
(make molecules)

OR



Chemistry
(make molecules)

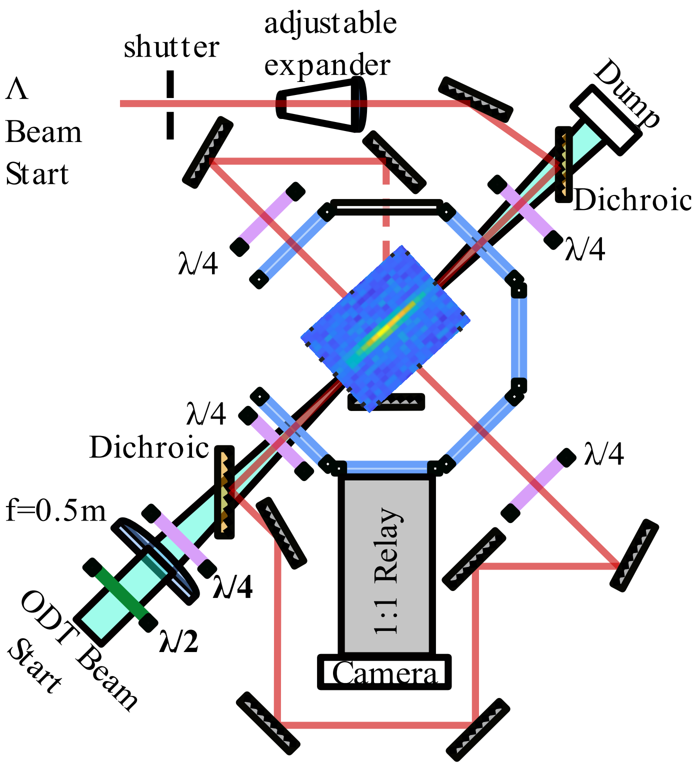
Cooling

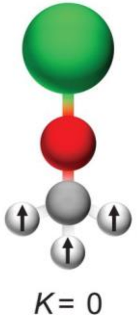
Slide from:
Jun Ye,
JILA

Rapidly advancing: direct laser cooling & trapping of molecules

Optical dipole traps

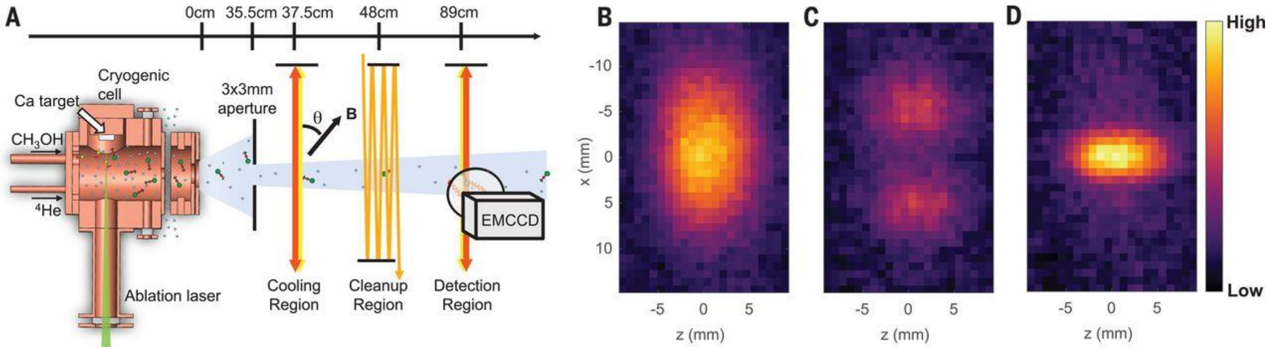
[Harvard Nat.Phys. 2018;
JILA PRX 2020;
UChicago PRL 2021]





Laser cooling + trapping of polyatomics

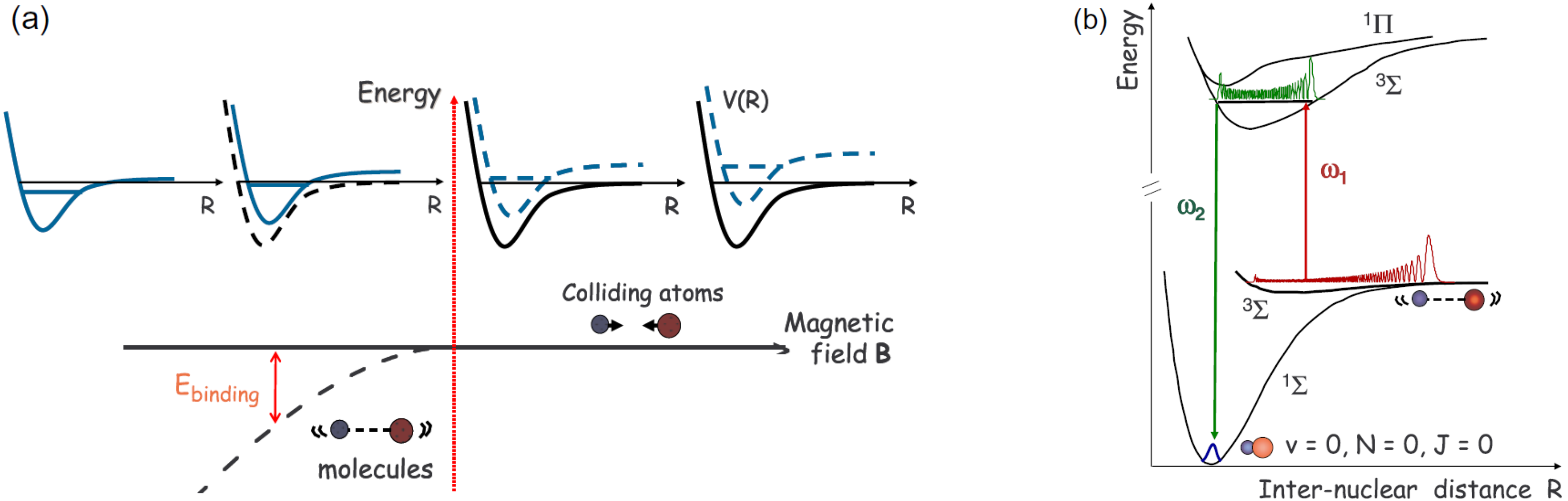
[Harvard Science 2020, Nature 2022, PRL 2023]



Species	PI	Location
SrF/TlF	DeMille	UChicago
CaF/CaOH**/**YbOH/ SrOH/CaOCH$_3$	Doyle	Harvard
CaF/YbF	Tarbutt	Imperial
YO	Ye	JILA
CaF	Cheuk	Princeton
BaH/CaH/CaD	Zelevinsky	Columbia
BaF	Yan	Zhejiang
CaF	Ospelkaus	Hannover
TlF	Hunter	Amherst
AlCl/CH	McCarron	UConn
MgF	Chae	Korea
MgF	Yin	ECN-Shanghai
AlF	Truppe	FHI
AlCl	Hemmerling	UC Riverside
BaF	Langen	Stuttgart
YbOH	Hutzler	Caltech
RaF/RaOH	Garcia-Ruiz, Hutzler, Doyle	

Status: 3 molecular species
in optical traps, $T \sim 10\ \mu\text{K}$,
no heavy species (yet),
only open-shell species (so far)

Most advanced method: "assembly" from ultracold alkali atoms



Efficient, coherent transfer with no added entropy/heating

ultracold atom pairs→weakly-bound molecules→absolute ground state molecules

Most alkali+alkali pairs (closed electron shells) have been assembled:

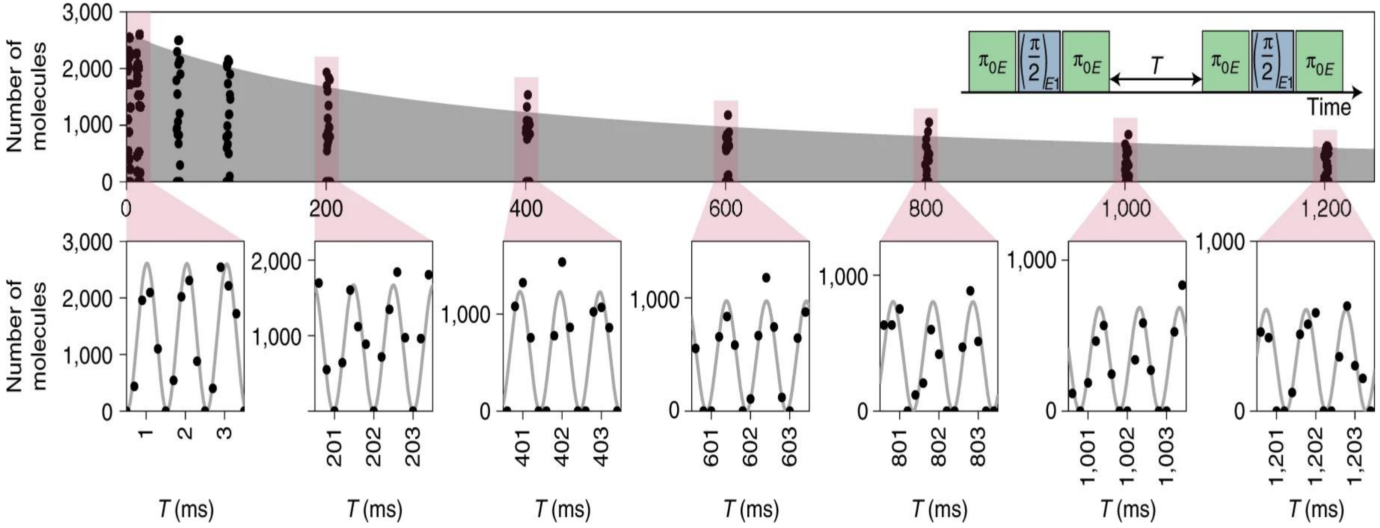
KRb, NaK, RbCs, NaRb, LiNa*, NaCs, … (*BUT* no other polar species, e.g. open shell)

JILA, Innsbruck, Durham, MITx2, CU Hong Kong, Harvard, MPQ Garching, USTC, Hannover, Princeton, Columbia, …

Advanced control with "assembled" polar molecules

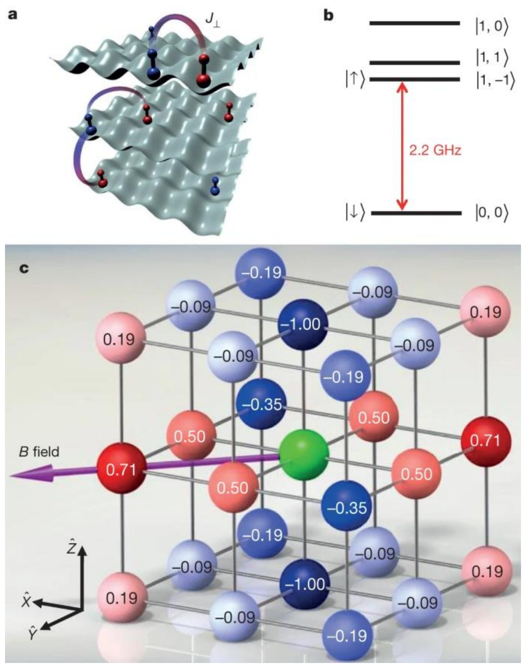
>5s coherence time for nuclear spins

PD Gregory et al.,Nat. Phys 2021



3D volume entanglement via dipole-dipole interactions

JILA
Nature (2013)



Inherit desirable properties from ultracold atom gases:

Efficient use of atoms (~50% from trapped gas), $N_{\text{mol}} > 10^4$ typical

Optically trapped, $T \sim$ 100 nK →

High-fidelity state preparation and readout

Dense & ultracold

Molecular species for next-gen EDM experiments?

Requirements:

- Large intrinsic sensitivity $\propto Z^3 \Rightarrow$ atom w/large Z
- Easily polarized $\Rightarrow$ small energy splittings
- **Correct electron configuration: strong s-p hybridized orbitals**
  $\Rightarrow$ for eEDM: unpaired spins, e.g. $^2\Sigma$ state
  $\Rightarrow$ for NSM: paired spins e.g. $^1\Sigma$ state
- Ultracold (weaker traps → smaller perturbations)

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Proposals for e-EDM via *direct* laser cooling & trapping

--Laser cooled beam/fountain of YbF $^2\Sigma$ for eEDM [Imperial College] $\mathcal{E}_{\text{eff}}$ ~24 GV/cm

--Stark-decelerated/laser cooled BaF $^2\Sigma$ beam for eEDM [Groningen] $\mathcal{E}_{\text{eff}}$ ~8 GV/cm

--Laser-cooled bent polyatomics: YbOH, YbCCCa for eEDM, YbCCAl for SM
[Caltech + Harvard + Toronto] $\mathcal{E}_{\text{eff}}$ ~24 GV/cm

--Laser cooled RaF/RaOH $^2\Sigma$ [MIT, Harvard, Caltech…] $\mathcal{E}_{\text{eff}}$ ~60 GV/cm

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Requirements:

Large *Z*, easily polarized, correct electron configuration, ultracold

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e.g. alkali + alkaline earth $^2\Sigma$ species for electron EDM?

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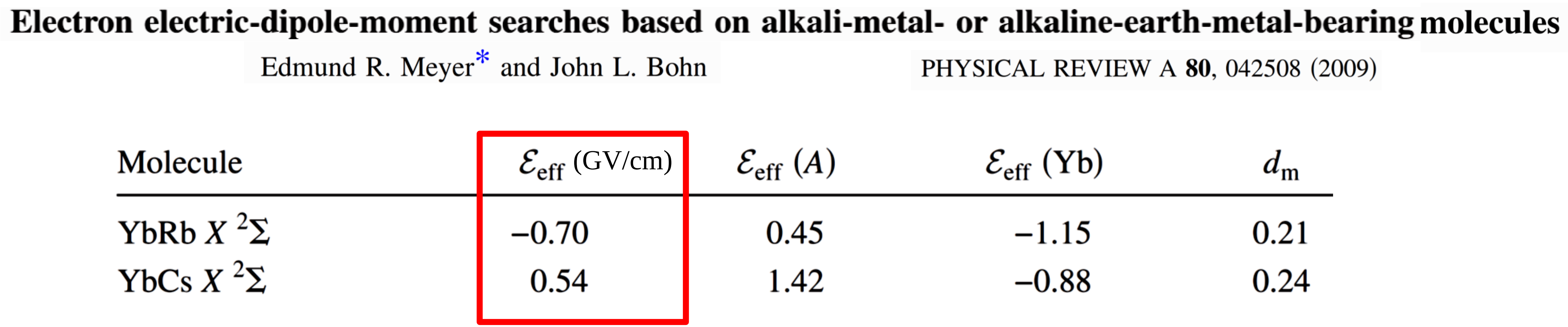
e.g. alkali + alkaline earth $^2\Sigma$ species for electron EDM?

An old tale of woe…



Electron electric-dipole-moment searches based on alkali-metal- or alkaline-earth-metal-bearing molecules

Edmund R. Meyer* and John L. Bohn

PHYSICAL REVIEW A **80**, 042508 (2009)

Molecule	$\mathcal{E}_{\mathrm{eff}}$ (GV/cm)	$\mathcal{E}_{\mathrm{eff}}$ (A)	$\mathcal{E}_{\mathrm{eff}}$ (Yb)	d_{m}
YbRb $X\ ^2\Sigma$	−0.70	0.45	−1.15	0.21
YbCs $X\ ^2\Sigma$	0.54	1.42	−0.88	0.24

Yb-alkali species ⇒ tiny $\mathcal{E}_{\mathrm{eff}}$, ~100x smaller than in ThO* ☹

Molecular species for next-gen EDM experiments

Why not “assembled molecules”?

Observation 1: Best EDM/NSM sensitivity in strongly ionic molecules (*s*-*p* hybridized orbitals)

Molecular species for next-gen EDM experiments

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Observation 2: no laser-cooled atom has large electron affinity → ionic bond…

X	EA(X) [eV]
Li	0.62
Na	0.55
K	0.50
Rb	0.49
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Molecular species for next-gen EDM experiments

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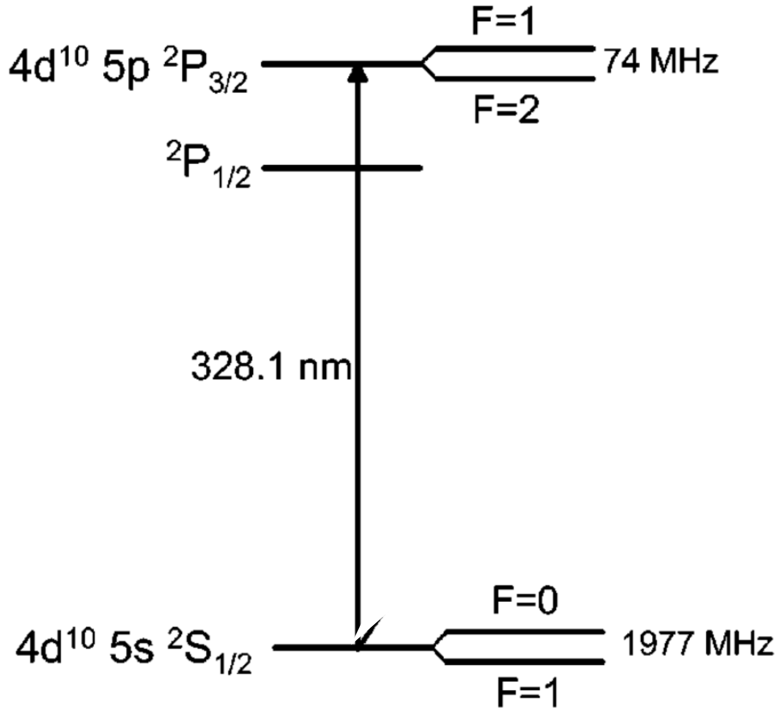
Atomic silver:
alkali-like structure ☺
…but UV lasers 😐

PHYSICAL REVIEW A, VOLUME 62, 063404

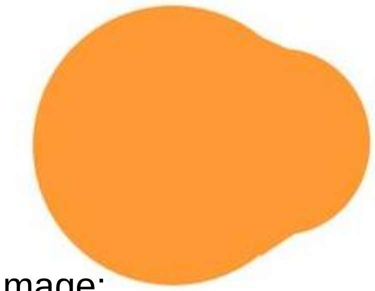
Magneto-optical trapping of silver atoms

G. Uhlenberg, J. Dirscherl,* and H. Walther



Enhanced nuclear Schiff moment in pear-shaped nuclei

Auerbach, Flambaum, Spevak
PRA 1996



(Image:
V.V. Flambaum)

Intrinsic Schiff moment:

$$S_{\text{intr}} \approx eZR_N^3 \frac{9\beta_2\beta_3}{20\pi\sqrt{35}}$$

$\beta_2 \approx 0.2$ **- quadrupole deformation**

$\beta_3 \approx 0.1$ **- octupole deformation**

- Rotational structure of octupole-deformed nucleus analogous to rotational structure of molecules
→large polarizabilty →**enhanced nuclear Schiff moment**

- Compared to ^{199}Hg (state of the art),
~1000x larger signals expected for a few heavy nuclei:
^{223}Rn, ^{223}Ra, ^{225}Ra, ^{227}Ac, **^{223}Fr**, …

Assembled silver + alkaline-earth molecules for "ultimate" eEDM search?

T. Fleig, Toulouse

Theoretical aspects of radium-containing molecules amenable to assembly from laser-cooled atoms for new physics searches

T. Fleig and DD 2021
New J. Phys. **23** 113039

Existence Proof: RaAg

- Ra (alkaline earth) = heaviest laser-cooled atom (Z=90) [Argonne]
--Long-lived ^{226}Ra available in macroscopic quantities

- Ra^+Ag^- $^2\Sigma$ ground state w/valence electron on Ra^+ (Z=90)
$\Rightarrow$ Large $\mathcal{E}_{\text{eff}}$, similar to 60 GV/cm in RaF

- Large dipole moment [ionic bond] & small rotational constant [Ag is heavy]
$\Rightarrow$ Small $\mathcal{E}$-field sufficient for polarization

All expectations for AgRa verified: $\mathcal{E}_{\text{eff}}$ = 65 GV/cm

μ = 5.4D; B_e = 630 MHz $\Rightarrow$ **$\mathcal{E}_{\text{pol}}$ = 260 V/cm**

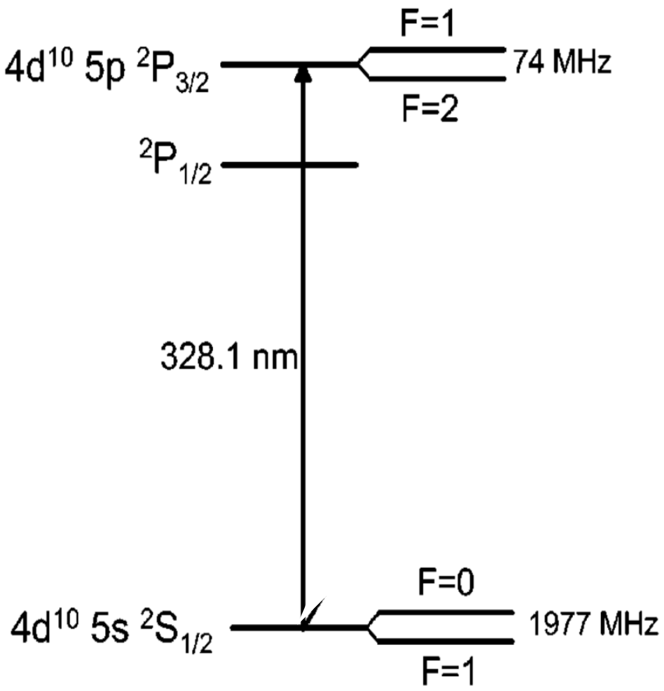
Assembled molecules for next-gen nuclear Schiff moment

^{223}FrAg identified as VERY promising

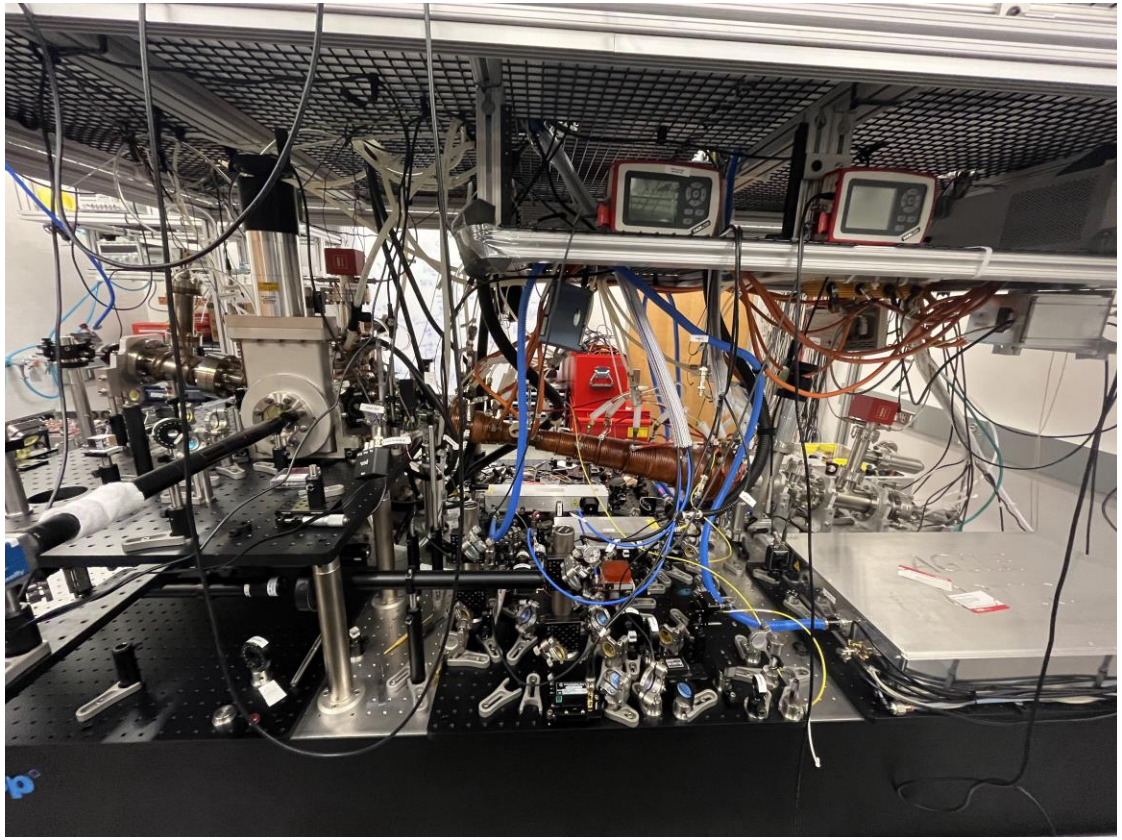
A. Marc, M. Hubert, T. Fleig
PRA **108**, 062815 (2023)

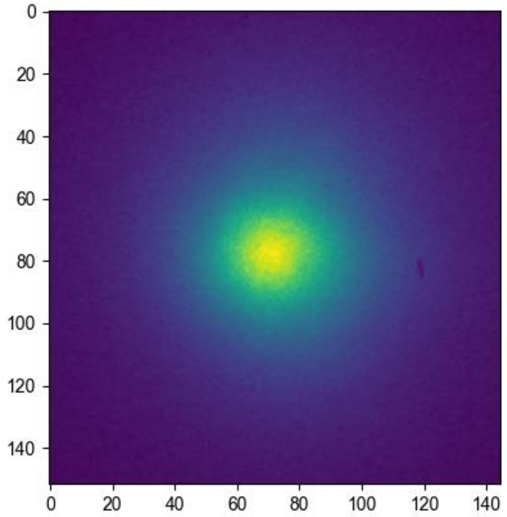
- Atom cooling & molecule assembly analogous to other bi-alkalis, ***N* ~ 10^4 ALREADY TYPICAL**
- In bialkali RbCs: nuclear spin **coherence time >5 s ALREADY DEMONSTRATED**
- ^{223}Fr ($t_{1/2}$ = 22 min) has nuclear octupole deformation → **~300-1000x enhanced Schiff moment**
- ***Efficient* collection & trapping of radioactive alkalis: established**
- Can get continuous ^{223}Fr flux from decay of long-lived ^{227}Ac (20 yr)
  → ***no online beam time needed***

First magneto-optic trap of silver atoms in 20+ years @ UChicago!



Atomic silver:
alkali-like structure ☺
…but UV lasers 😐





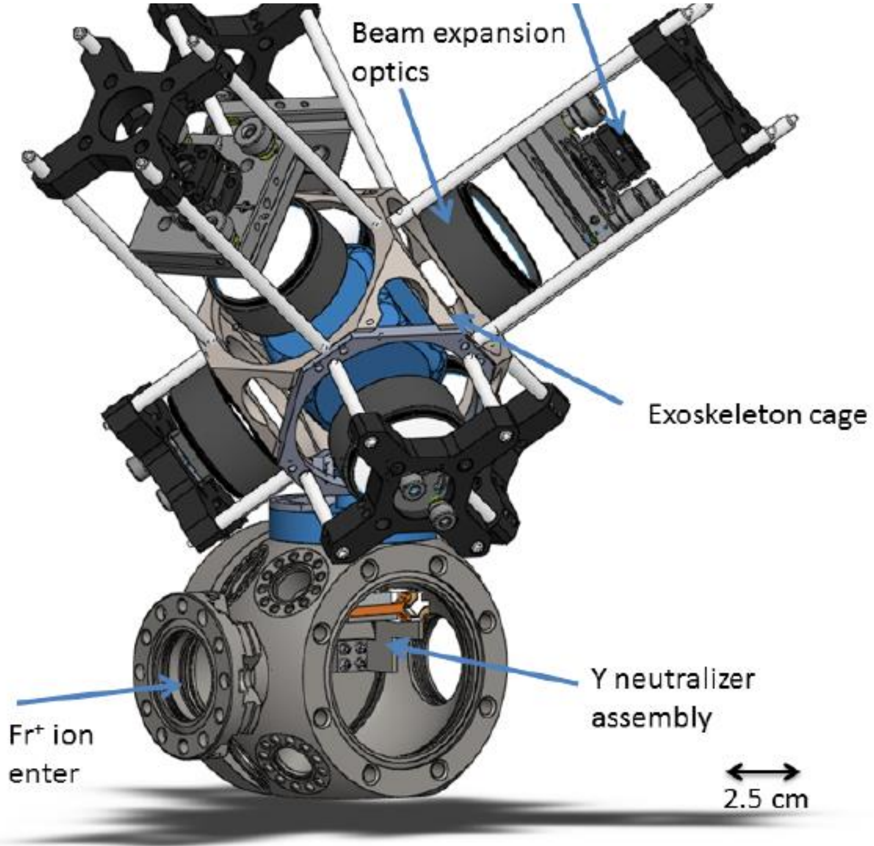
NEXT: measure Ag-Ag ultracold scattering properties

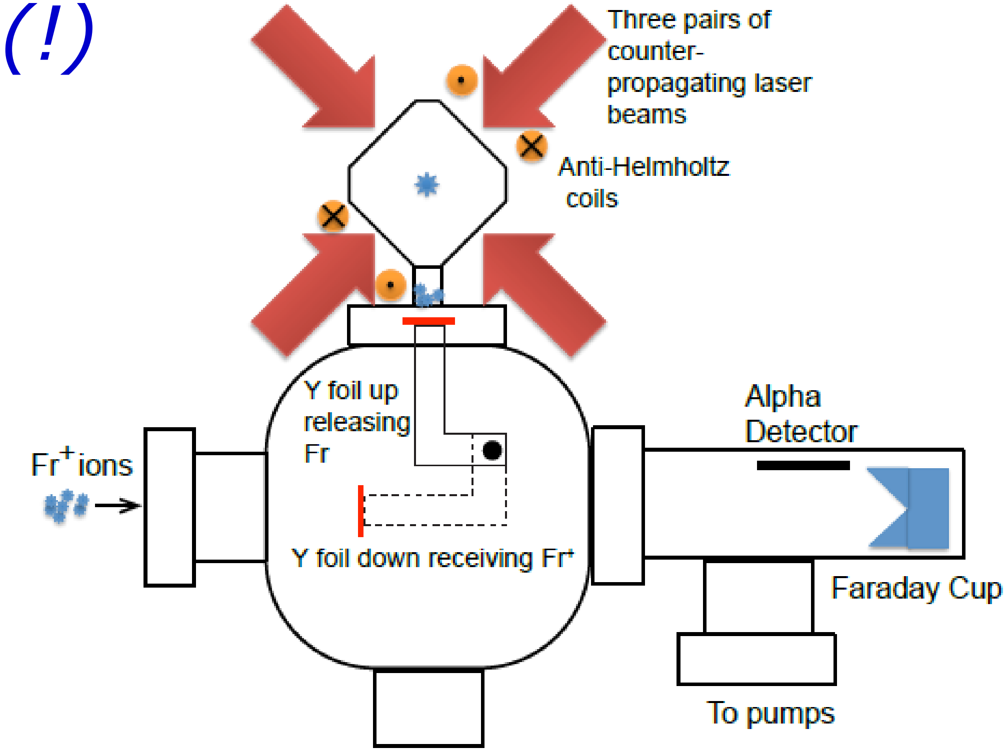
Aside: Ag-alkali molecules have huge dipole moments: good for qubits, quantum simulation, etc.

Trapping Fr atoms: a solved problem(!)

Commissioning of the Francium Trapping Facility at TRIUMF *JINST* 8 P12006

M. Tandecki,[a] J. Zhang,[b] R. Collister,[c] S. Aubin,[d] J.A. Behr,[a] E. Gomez,[e] G. Gwinner,[c] L.A. Orozco [b,1] and M.R. Pearson[a]





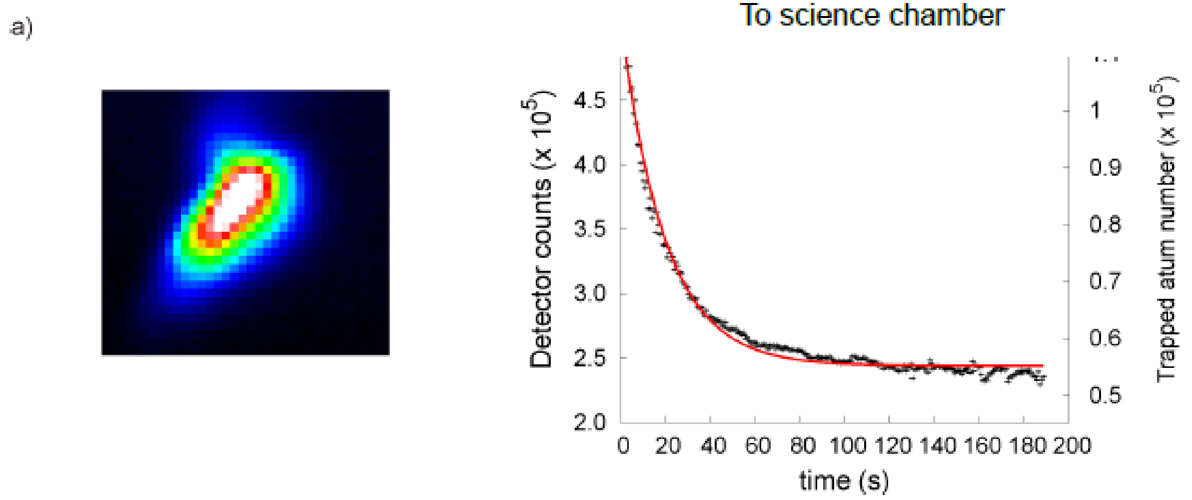
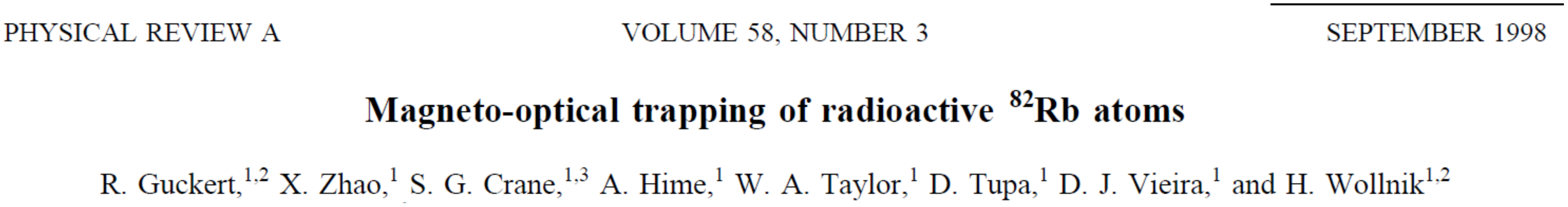


Figure 8. Fr MOT performance. a) False color CCD image of the MOT fluorescence of a cloud of about 10^5 ^{209}Fr trapped at the FTF. The pixel size of the camera is $6.7 \times 6.7\ \mu m^2$; an area of $0.86 \times 0.86\ mm^2$ is shown.

Making ^{223}*Fr* without *accelerator beam time*

Approaches to ^{223}Fr^{+} beam for 1st-gen NSM search

- Demonstrated method with shorter-lived Fr and Rb isotopes:
  Use decay from longer-lived precursor + standard alkali-ion extraction

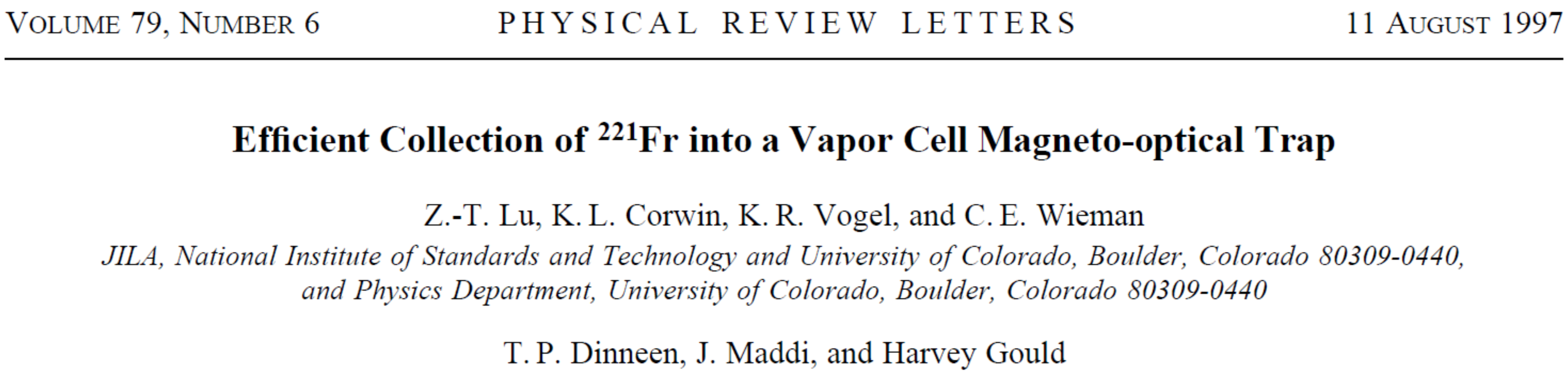


PHYSICAL REVIEW A VOLUME 58, NUMBER 3 SEPTEMBER 1998

Magneto-optical trapping of radioactive ^{82}Rb atoms

R. Guckert,[1,2] X. Zhao,[1] S. G. Crane,[1,3] A. Hime,[1] W. A. Taylor,[1] D. Tupa,[1] D. J. Vieira,[1] and H. Wollnik[1,2]

10^{8}/s ^{82}Rb^{+} ions/s from 10 mCi ^{82}Sr



VOLUME 79, NUMBER 6 PHYSICAL REVIEW LETTERS 11 AUGUST 1997

Efficient Collection of ^{221}Fr into a Vapor Cell Magneto-optical Trap

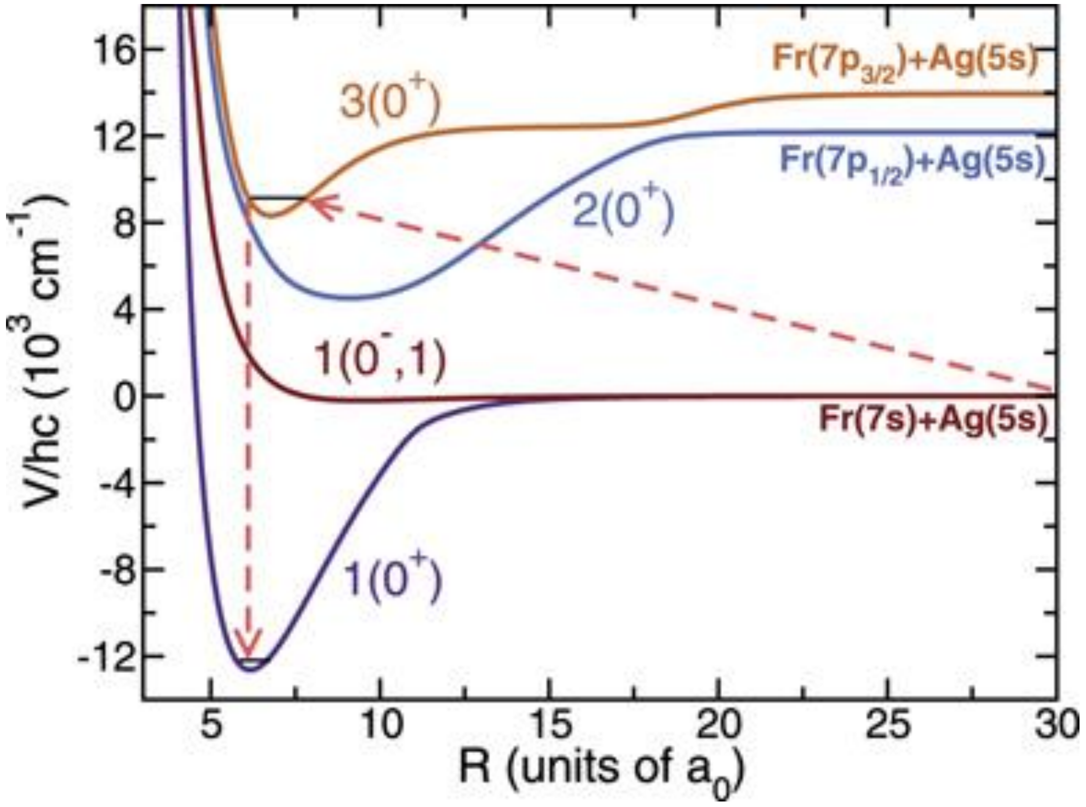
Z.-T. Lu, K. L. Corwin, K. R. Vogel, and C. E. Wieman

JILA, National Institute of Standards and Technology and University of Colorado, Boulder, Colorado 80309-0440,
and Physics Department, University of Colorado, Boulder, Colorado 80309-0440

T. P. Dinneen, J. Maddi, and Harvey Gould

~4x10^{4}/s ^{221}Fr atoms/s from 50 μCi ^{225}Ac

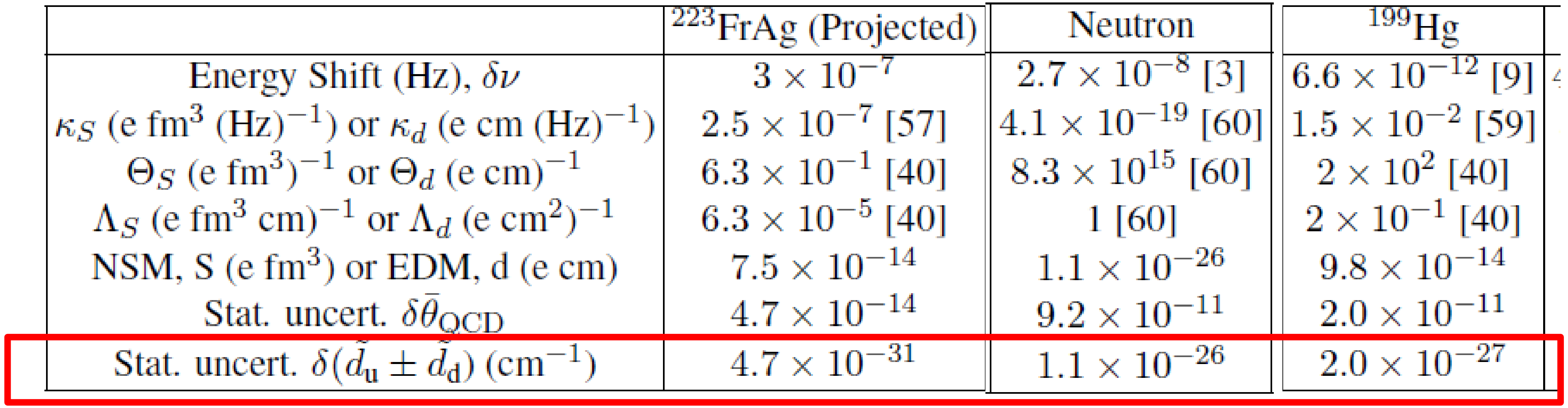
Theory: assembling ^{223}FrAg molecules is ~just like other bi-alkalis



J. Klos, H. Li, **E. Tiesinga, S. Kotochigova**
NJP 24, 025005 (2022)

- As anticipated, electronic state structure entirely analogous to other bi-alkalis
- Standard Fano-Feshbach resonance structure for “magneto-association”
- Strong “assembly” transitions with convenient laser wavelengths
- All looks fine, but experimental measurements needed to sort out details

Projected sensitivity of 1st-gen ^{223}FrAg NSM search



	^{223}FrAg (Projected)	Neutron	^{199}Hg
Energy Shift (Hz), $\delta\nu$	3×10^{-7}	2.7×10^{-8} [3]	6.6×10^{-12} [9]
κ_S (e fm^3 (Hz)$^{-1}$) or κ_d (e cm (Hz)$^{-1}$)	2.5×10^{-7} [57]	4.1×10^{-19} [60]	1.5×10^{-2} [59]
Θ_S (e fm^3)$^{-1}$ or Θ_d (e cm)$^{-1}$	6.3×10^{-1} [40]	8.3×10^{15} [60]	2×10^{2} [40]
Λ_S (e fm^3 cm)$^{-1}$ or Λ_d (e cm^2)$^{-1}$	6.3×10^{-5} [40]	1 [60]	2×10^{-1} [40]
NSM, S (e fm^3) or EDM, d (e cm)	7.5×10^{-14}	1.1×10^{-26}	9.8×10^{-14}
Stat. uncert. $\delta\bar{\theta}_{\mathrm{QCD}}$	4.7×10^{-14}	9.2×10^{-11}	2.0×10^{-11}
Stat. uncert. $\delta(\tilde{d}_{\mathrm{u}}\pm\tilde{d}_{\mathrm{d}})$ (cm^{-1})	4.7×10^{-31}	1.1×10^{-26}	2.0×10^{-27}

- **Included in Estimate:**
  - --300x NSM enhancement
  - --near-ideal molecular structure
  - --τ_{coh} ~10 s [Cornish, Zwierlein, etc.]
  - -- ~100% detection efficiency
  - --n = 10^4 molecules
  - -- ~10% efficiency ^{223}Fr atoms into trap

All these parameters *ALREADY DEMONSTRATED* with stable bi-alkalis (!)

ALREADY DEMONSTRATED **with radioactive alkali atoms**

⇒ ~4000x projected improvement vs. ^{199}Hg state of the art

What is needed to make ^{223}FrAg NSM search happen?

From the AMO community:

-- Measure & control ultracold scattering properties Ag-Ag

-- Measure & control ultracold scattering properties Fr-Fr

-- Measure & control ultracold scattering properties Fr-Ag

-- Assemble excited FrAg* molecules in optical trap

-- Transfer FrAg* molecules to absolute ground state in trap

What is needed to make ^{223}FrAg NSM search happen?

From the NP/Isotopes experimental community:

-- Off-line source of ^{223}Fr for development & first-generation data

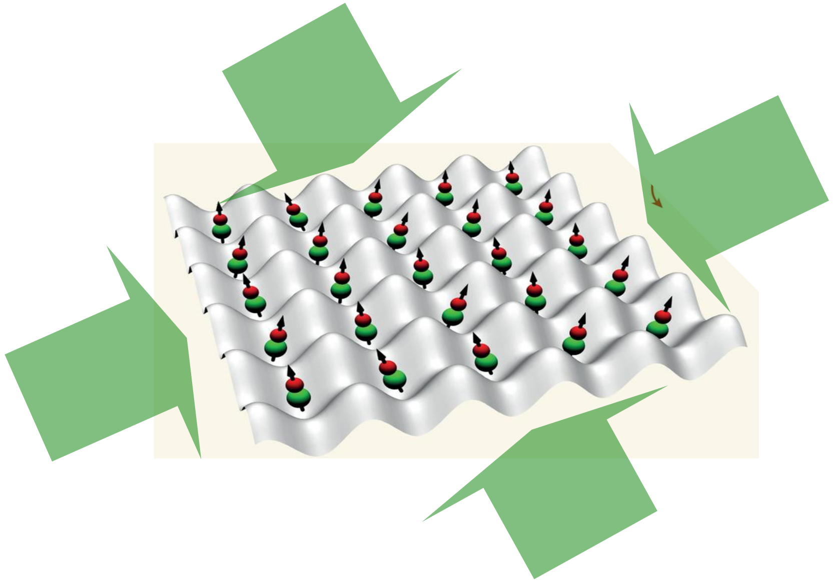
-- On-line access to Fr isotopes for initial AMO studies (TRIUMF)

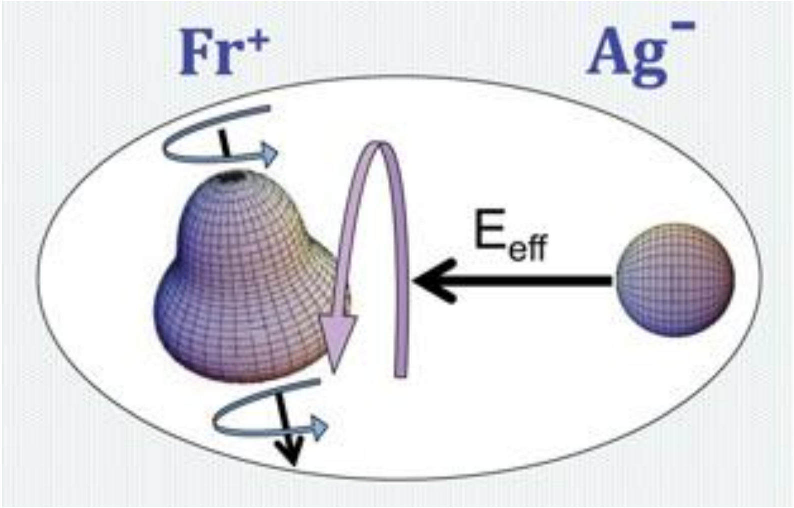
-- High-flux on-line ^{223}Fr from ISOL for ultimate EDM statistics?

From the NP theory community:

--Quasi-reliable calculations of Schiff moment in ^{223}Fr!

“Ultimate” Schiff moment experiment with ^{223}FrAg?





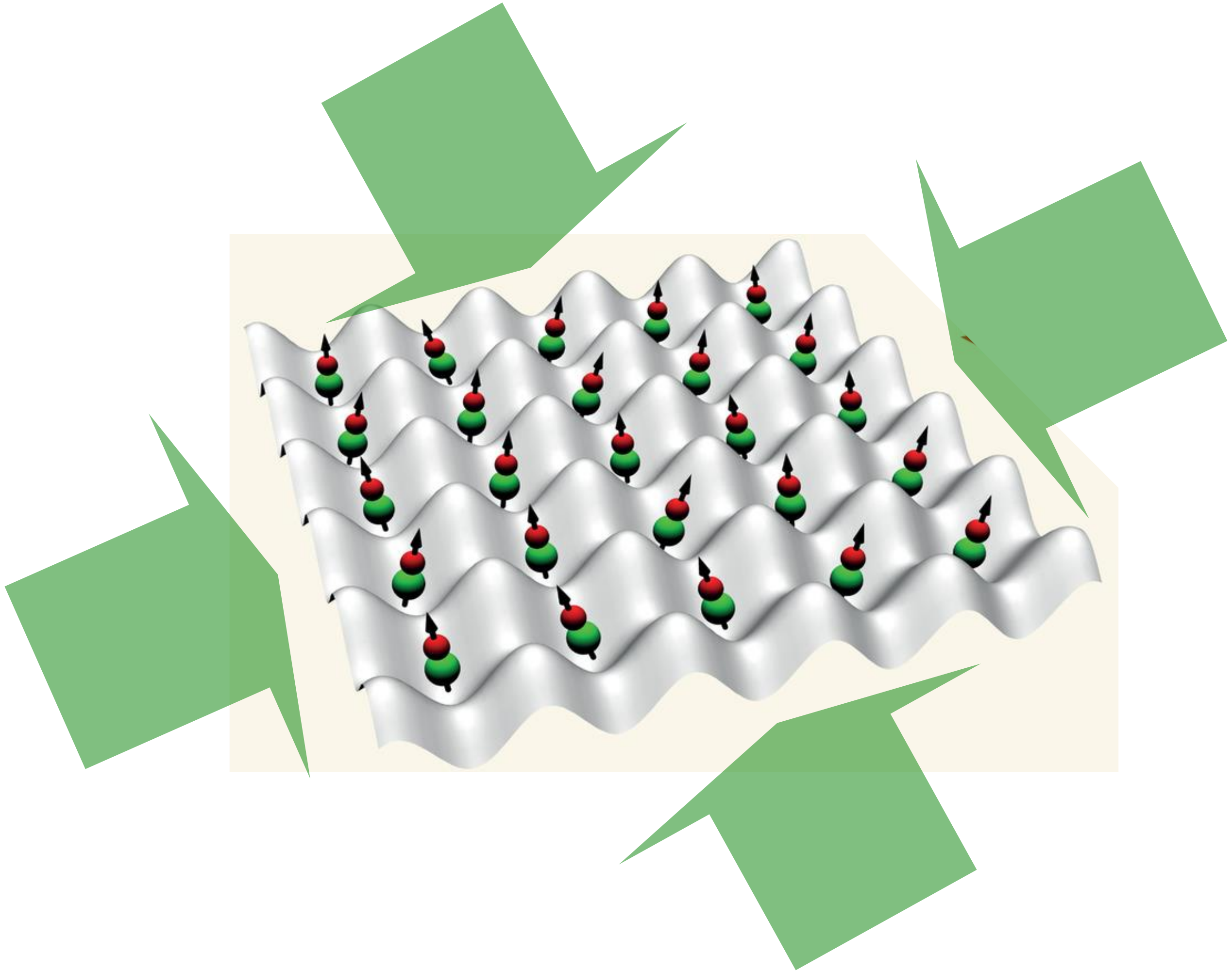
REDRUM COLLABORATORS

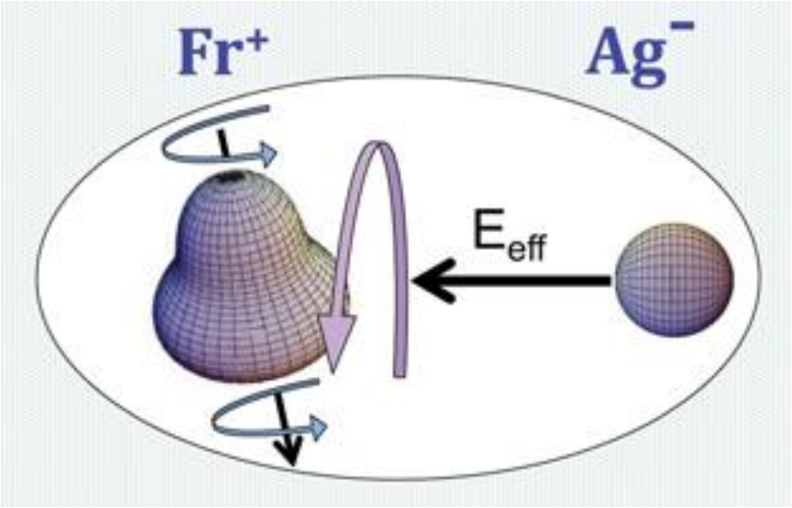
- UChicago
  - Mohit Verma
  - Shaozhen Yang
  - Wesley Cassidy
  - Dr. Thomas Langin
- Univ. of Waterloo
  - Prof. Alan Jamison
- TRIUMF+
  - Gerald Gwinner (U. Manitoba)
  - John Behr
  - Stephan Malbrunot
  - Andrea Teigelhofer
  - Luis Orozco (U. Maryland)
  - Kirk Madison (UBC)
- Univ. of Utah
  - Tara Mastren
- Temple U./NIST
  - Svetlana Kotochigova

REDRUM Collaboration PIs:

- TRIUMF: Fr trapping & spectroscopy
  --Stephan Malbrunot-Ettenauer,
  John Behr
- Univ. of British Columbia
  --Kirk Madison
- Univ. of Manitoba:
  --Gerald Gwinner
- Univ. of Maryland
  --Luis Orozco
- Univ. of Utah: Actinide electrochemistry
  --Tara Mastren
- Univ. of Waterloo: Fr & Ag &FrAg spectroscopy
  --Alan Jamison
- Temple U./NIST: Theory of molecular structure
  --Svetlana Kotochigova
- Univ. of Chicago: all aspects
  --Dave DeMille

"Ultimate" Schiff moment experiment with ^{223}FrAg?





1st Generation ^{223}FrAg:

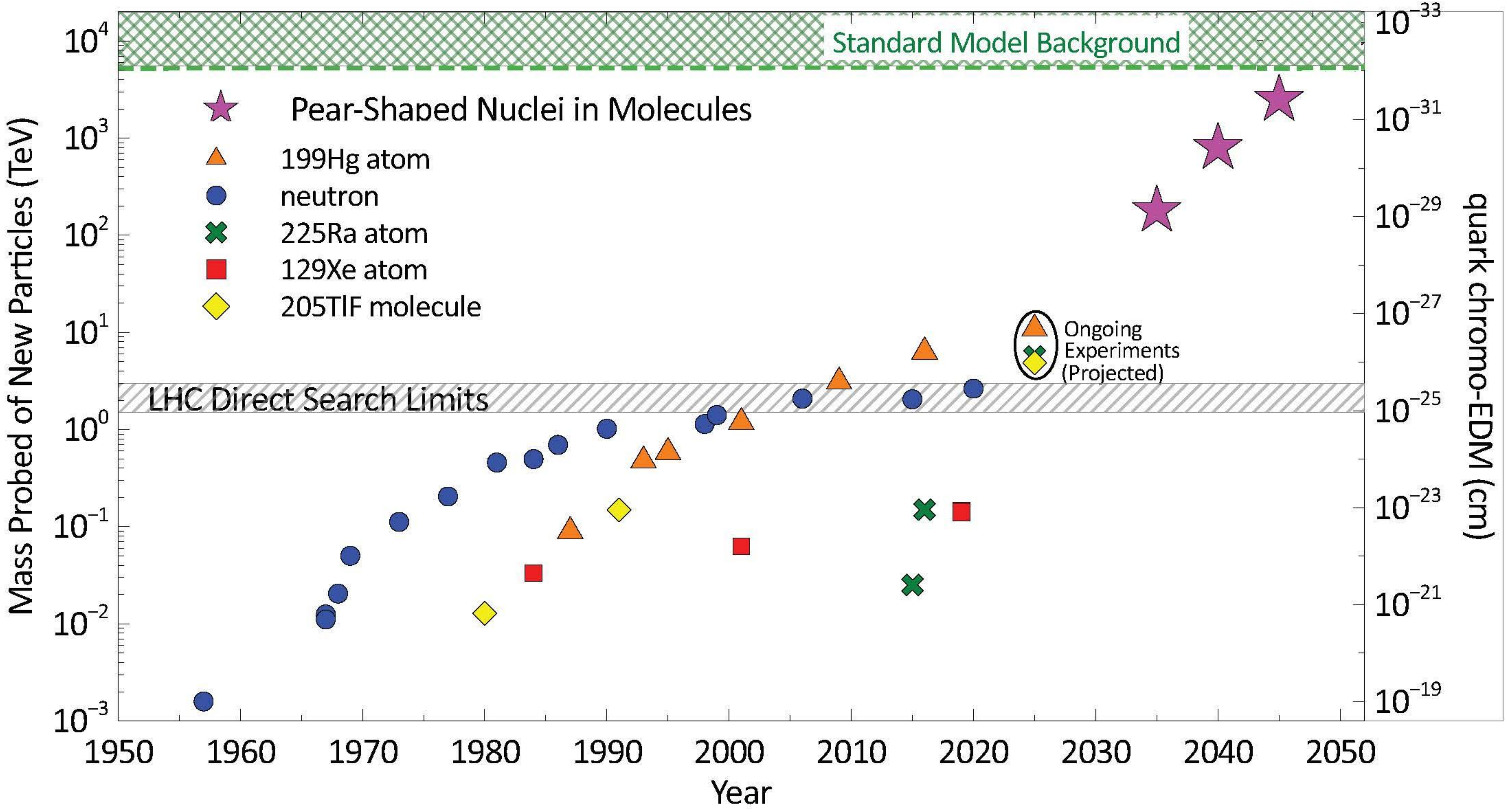
~4000x projected improvement vs. state of the art assuming only demonstrated performance parameters from other bi-alkali species

→ sufficient to probe >100 TeV

+Long-term potential for dramatic further improvements:

Longer τ_{coh}, larger N, …

Trajectory: probing >1000 TeV with chromo-EDMs



ZOMBIES: nuclear spin-dependent parity violation in molecules*

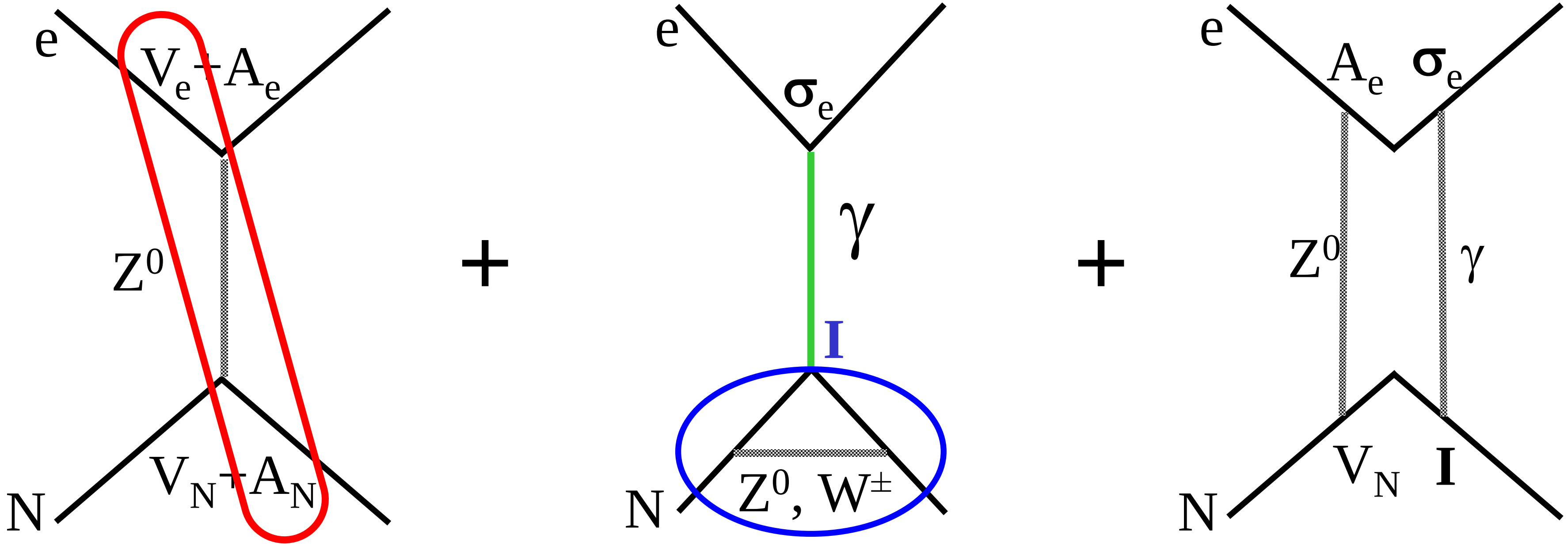
*stable

- Underlying physics of NSD-PV
- ZOMBIES approach & projected sensitivity
- Long term outlook

Postdoc
Mangesh Bhattarai

Argonne NATIONAL LABORATORY

Mechanisms for NSD-PV in atoms and molecules



Tree-level
NSD-PV
from suppressed
V_eA_N term:
C_2 subject to QCD
renormalization
similar to g_A

HPV interactions
inside nucleus induce
nuclear
"anapole moment":
couples to electron
magnetically

Coherent sum of
weak charge Q_W
and
electromagnetic
hyperfine interaction

$$H_{NSD-PV} \propto \left(\kappa_2' + \kappa_a' + \kappa_Q'\right) G_F \left(\vec{\sigma}\cdot\vec{I}\right)\left(\vec{\sigma}\cdot\vec{p}\right)\delta^3(\vec{r})$$

3 contributions to NSD-PV: scaling with *Z* & *A*

$$H_{NSD-PV} \propto \left(\kappa'_2 + \kappa'_a + \kappa'_Q\right) G_F \left(\vec{\sigma}\cdot\vec{I}\right)\left(\vec{\sigma}\cdot\vec{p}\right)\delta^3(\vec{r})$$

Overall Z^2

$\kappa'_{2P} = -\kappa'_{2N} \approx -.05$

$$\kappa'_a \approx .05 g_{eff}\left(\frac{A}{50}\right)^{2/3}$$

$(g_{eff,P} \cong 4,\ g_{eff,N} \lesssim 1)$

$$|\kappa'_Q| << |\kappa'_2 + \kappa'_a|$$

(Well understood, calculable, and small: ignore κ'_Q

Heavy atoms: anapole term dominates: $|\kappa'_a| > |\kappa'_2|$

(Collective enhancement causes radiative correction > tree level…!)

Light atoms: tree-level Z exchange term dominates: $|\kappa'_a| > |\kappa'_2|$

$|\kappa_a| \approx |\kappa_2|$ for $A \approx 10$ (odd proton)

$A \approx 100$ (odd neutron)

3 contributions to NSD-PV: scaling with *Z* & *A*

$$H_{NSD-PV} \propto \left(\kappa_2 + \kappa_a + \kappa_Q\right) G_F \left(\vec{\sigma}\cdot\vec{I}\right)\boxed{\left(\vec{\sigma}\cdot\vec{p}\right)\delta^3(\vec{r})}$$

Overall Z^2

$$\kappa'_{2P} = -\kappa'_{2N} \approx -.05$$

$$\kappa'_a \approx .05 g_{eff}\left(\frac{A}{50}\right)^{2/3}$$

$$(g_{eff,P} \cong 4,\ g_{eff,N} \lesssim 1)$$

Challenge for atomic/molecular approaches:

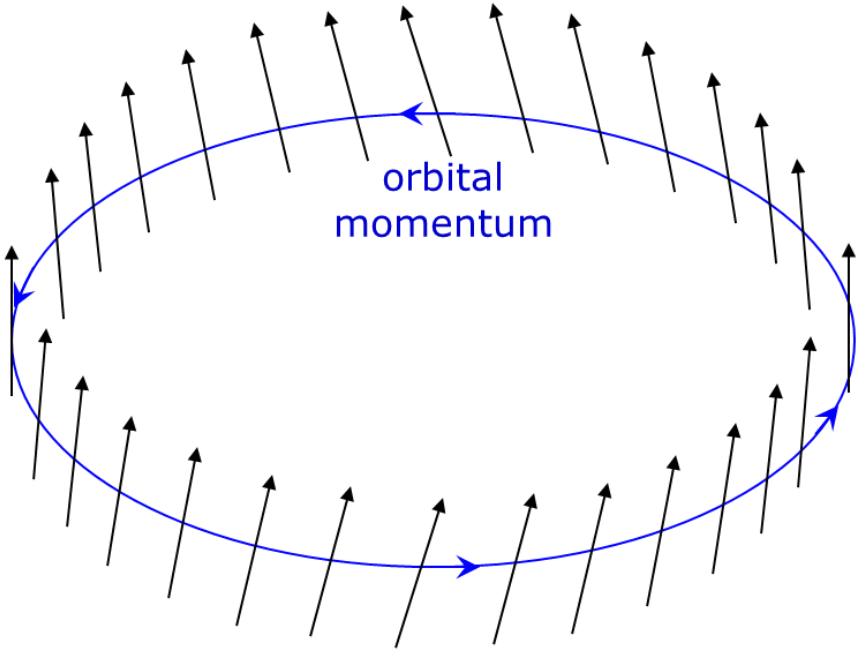
Signals easiest to detect with high Z & A

BUT

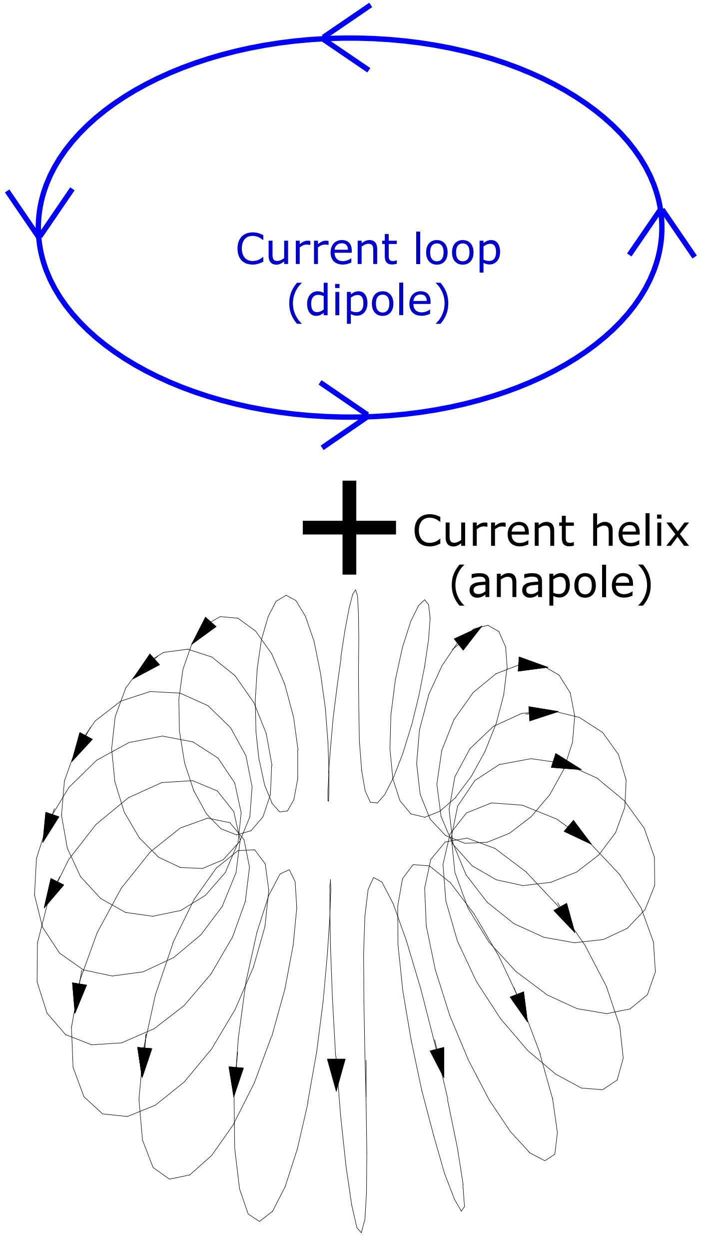
Much easier to interpret with lowest Z & A

Purely hadronic PV in nucleus induces nuclear spin helix = anapole moment

$H_{HPV} \propto \vec{\sigma}_N \cdot \vec{p}_N \;\Rightarrow$ nucleon spin tilted along momentum



$=$



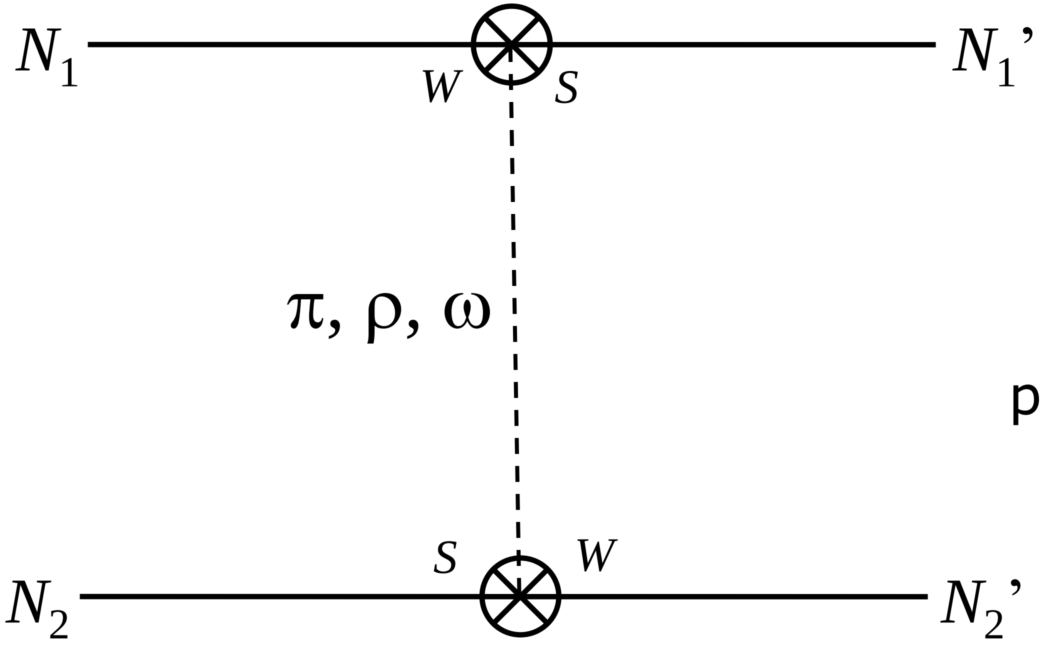
Simple model for nuclear anapole

(valence nucleon + constant-density core):

$$\vec{a} \propto g_{eff} A^{2/3} \hat{I}$$

Microscopic physics of nuclear anapole moment

Nucleon-nucleon HPV interactions perturb nuclear structure:



DDH
description of HPV:
effective meson
exchange
parameterizes range &
isospin structure

Hamiltonian for unpaired nucleon interacting with paired core gives spin-momentum correlation

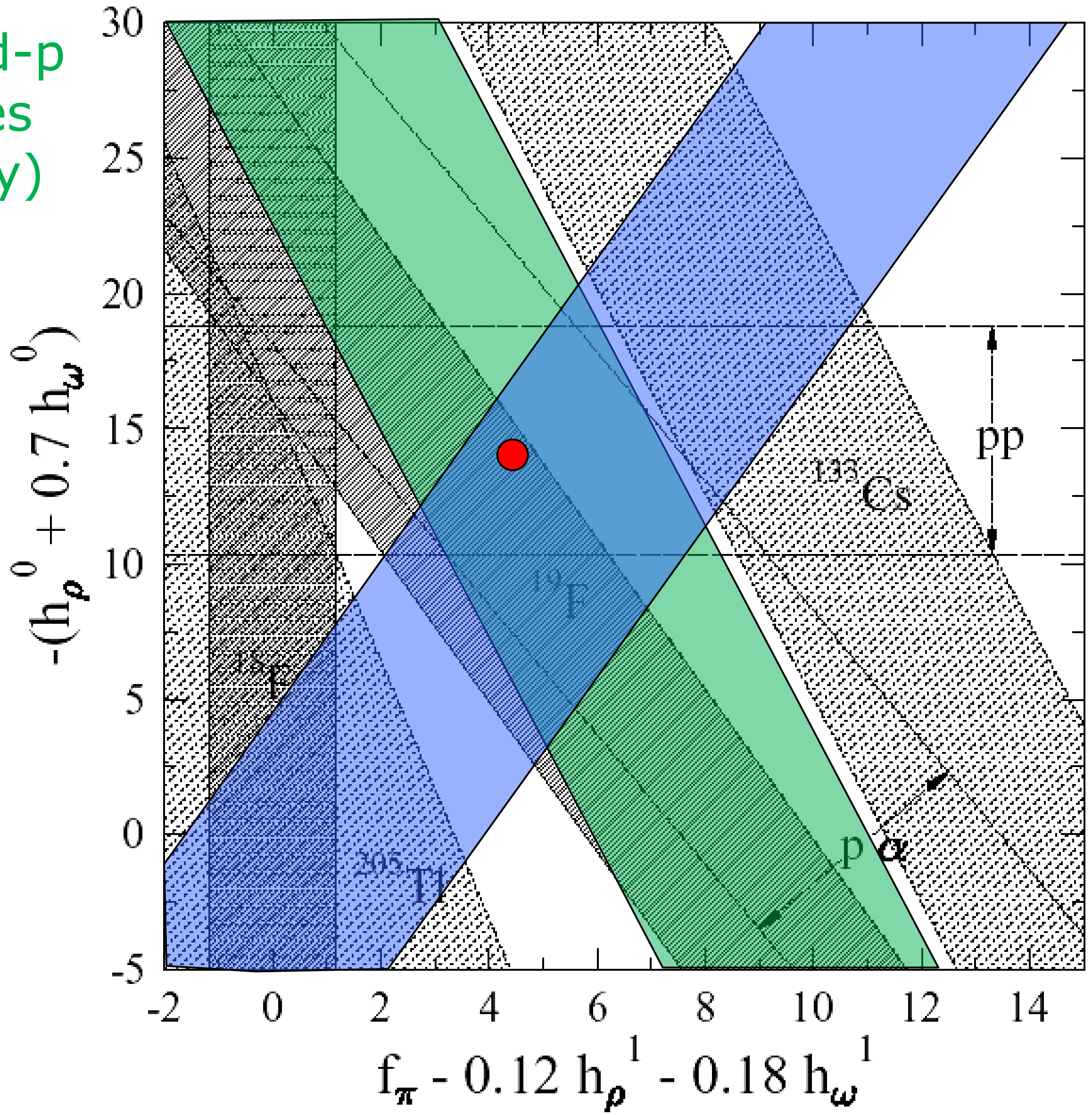
$$H_{HPV} \sim G_F \left(\vec{\sigma}_N \cdot \vec{p}_N\right) \sum_i g_{\mathrm{eff},i} F_i(\vec{r},\vec{\tau})$$

5 terms in principle at low q^2

2 linear combinations estimated important for anapole

HPNC measurements including anapole moments (past & future)

New odd-p isotopes (× many)



New odd-n isotopes (×many)

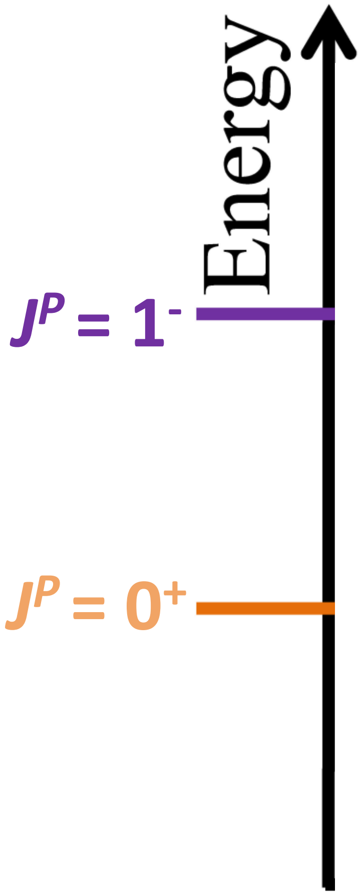
Single prior anapole measurement from atomic NSD-PV in ^{133}Cs

[C. Wieman group, JILA, 1997]

Assumes ~30% uncertainty in nuclear structure calculations…?

NOTE: new data (NPDGamma, *n* spin rotation in ^{4}He) + theory advances (EFT, large N_c)
→ Hope for convergence in HPV parameters

Enhanced NSD-PV mixing in simple molecules [$^2\Sigma$, *s-p* hybridized orbital]

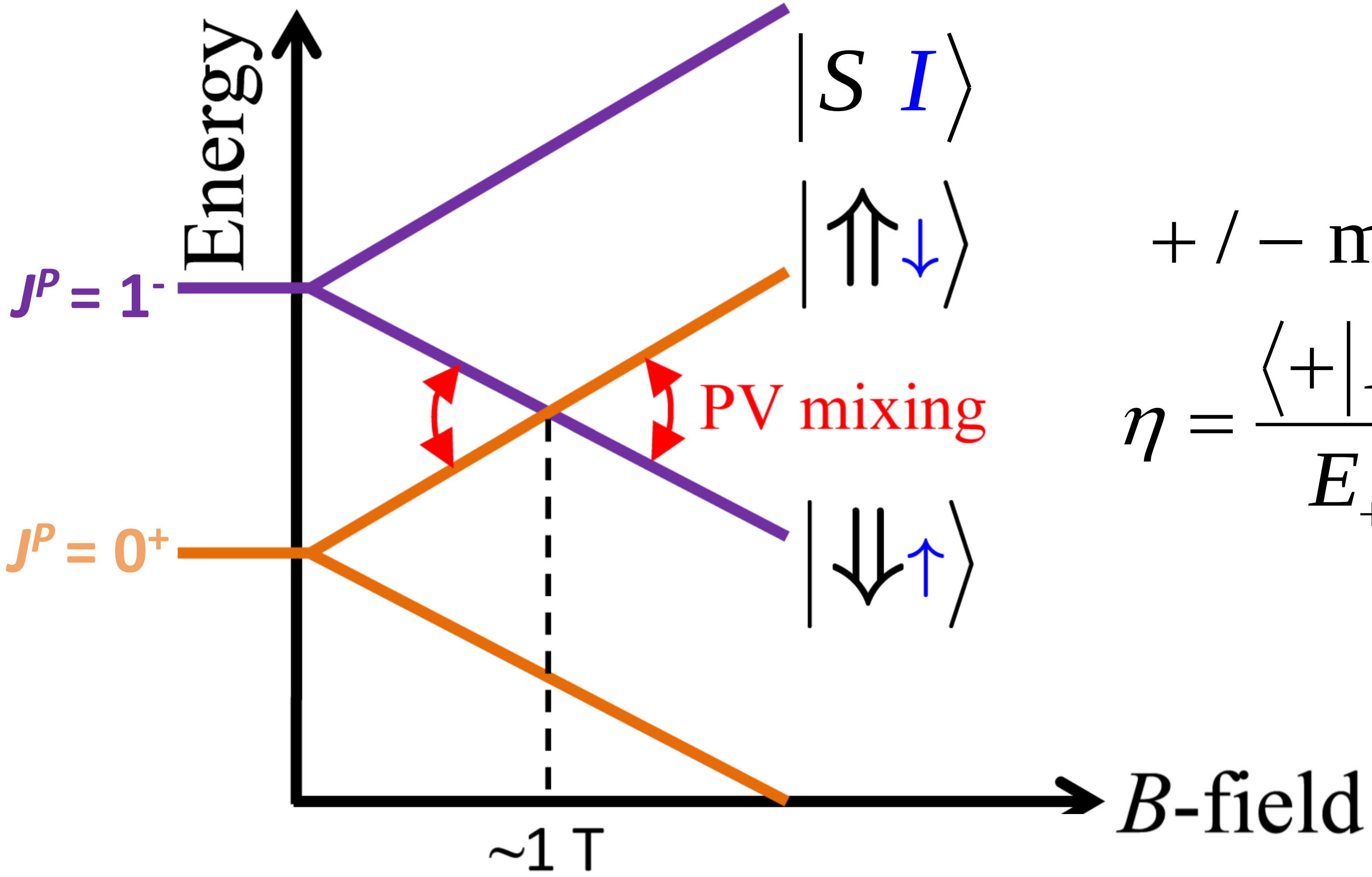


$$+/-\ \text{mixing}$$

$$\eta = \frac{\langle + | H_{PV} | - \rangle}{E_+ - E_-}$$

Naturally small rotational splitting (~10^{-4} eV vs. ~1 eV in atoms)

Enhanced NSD-PV mixing in simple molecules [$^2\Sigma$, *s-p* hybridized orbital]

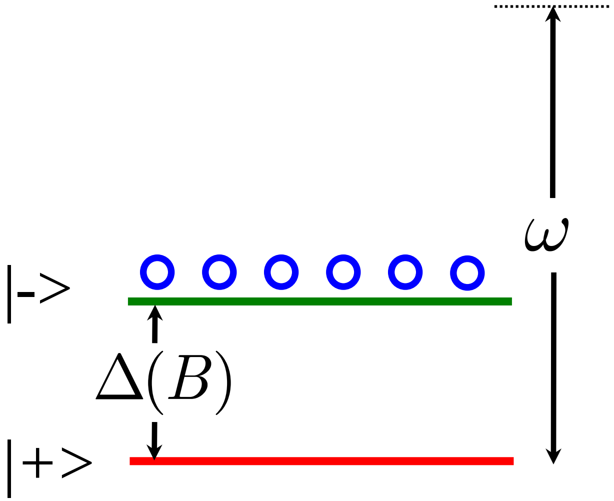


$+/-$ mixing

$$\eta = \frac{\langle + | H_{PV} | - \rangle}{E_+ - E_-}$$

Naturally small rotational splitting (~10^{-4} eV vs. ~1 eV in atoms)
can be bridged w/Zeeman shift:
$\gtrsim$ **10^{11} enhanced PV mixing vs. classic experiments with atoms**

Detecting PV in near-degenerate levels: AC Stark shift

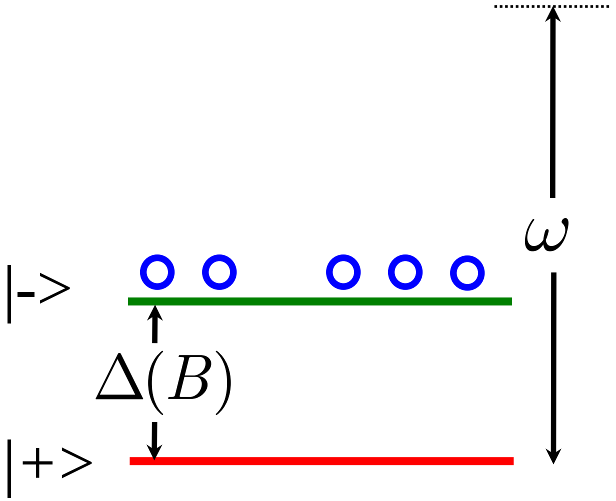


$$H = \begin{pmatrix} 0 & iW + d\mathcal{E}(t) \\ -iW + d\mathcal{E}(t) & \Delta \end{pmatrix}$$

D.D., S.B. Cahn, *et al.*
PRL **100,** 023003 (2008)

Nguyen, DD, … D. Budker,
PRA **56**, 3453 (1997)

Detecting PV in near-degenerate levels: AC Stark shift



PV mixing iW encodes physics of interest

$$H = \begin{pmatrix} 0 & \boxed{iW} + d\mathcal{E}(t) \\ \boxed{-iW} + d\mathcal{E}(t) & \Delta \end{pmatrix}$$

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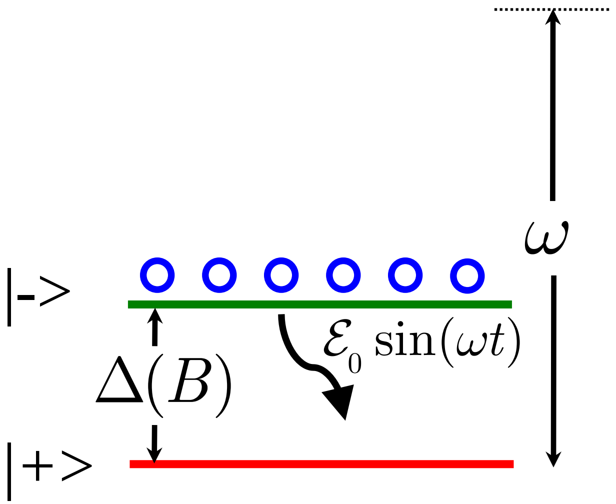

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$$H = \begin{pmatrix} 0 & iW + d\mathcal{E}(t) \\ -iW + d\mathcal{E}(t) & \Delta \end{pmatrix}$$

Apply oscillating $\mathcal{E}$-field, 1 cycle:

$$\mathcal{E}(t) = \mathcal{E}_0 \sin(\omega t) \qquad \begin{bmatrix} \omega \gg \Delta,\ d\mathcal{E}_0; \\ T = 2\pi / \omega \end{bmatrix}$$

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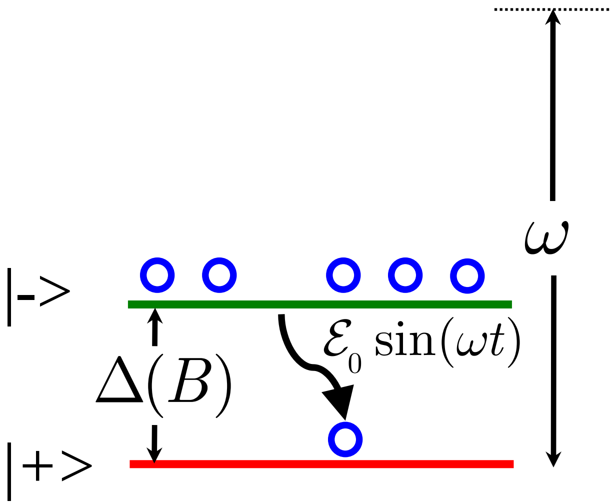

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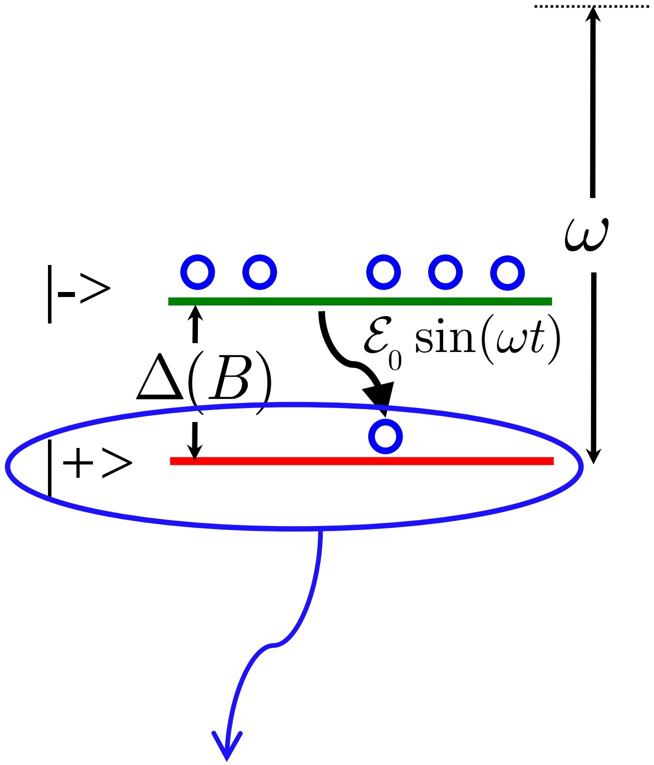

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$$S = \left|\langle + | \psi(T) \rangle\right|^2 = 4\sin^2\left(\frac{\Delta T}{2}\right)\left[\left(\frac{d\mathcal{E}_0}{\omega}\right)^2 + 2\frac{W}{\Delta}\frac{d\mathcal{E}_0}{\omega}\right]$$

D.D., S.B. Cahn, *et al.*
PRL **100,** 023003 (2008)

Nguyen, DD, … D. Budker,
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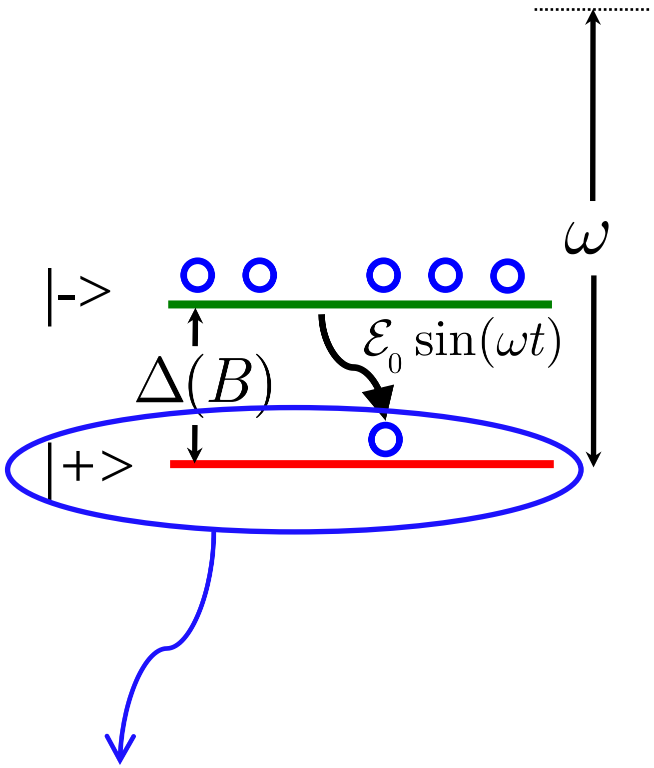
Detecting PV in near-degenerate levels: AC Stark shift



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"Large"
Stark Term
Even in $\mathcal{E}_0$

Small
PV Term
Odd in $\mathcal{E}_0$

D.D., S.B. Cahn, *et al.*
PRL **100,** 023003 (2008)

Nguyen, DD, … D. Budker,
PRA **56**, 3453 (1997)

Signal, Asymmetry, Sensitivity

- - Measure signal $S \;\; \mathcal{E}_0 \;\; \approx 4N_0 \sin^2\left(\frac{\Delta T}{2}\right)\left[\left(\frac{d\mathcal{E}_0}{\omega}\right)^2 + 2\frac{W}{\Delta}\frac{d\mathcal{E}_0}{\omega}\right]$

with opposite-sign $\mathcal{E}$-fields $+\mathcal{E}_0, -\mathcal{E}_0$

- - Form asymmetry to extract W in terms of known quantities :

$$\mathcal{A} = \frac{S(+\mathcal{E}_0) - S(-\mathcal{E}_0)}{S(+\mathcal{E}_0) + S(-\mathcal{E}_0)} \approx 2\frac{W}{\Delta}\frac{\omega}{d\mathcal{E}_0}$$

Dispersion-like
function
of detuning Δ

Statistical Uncertainty

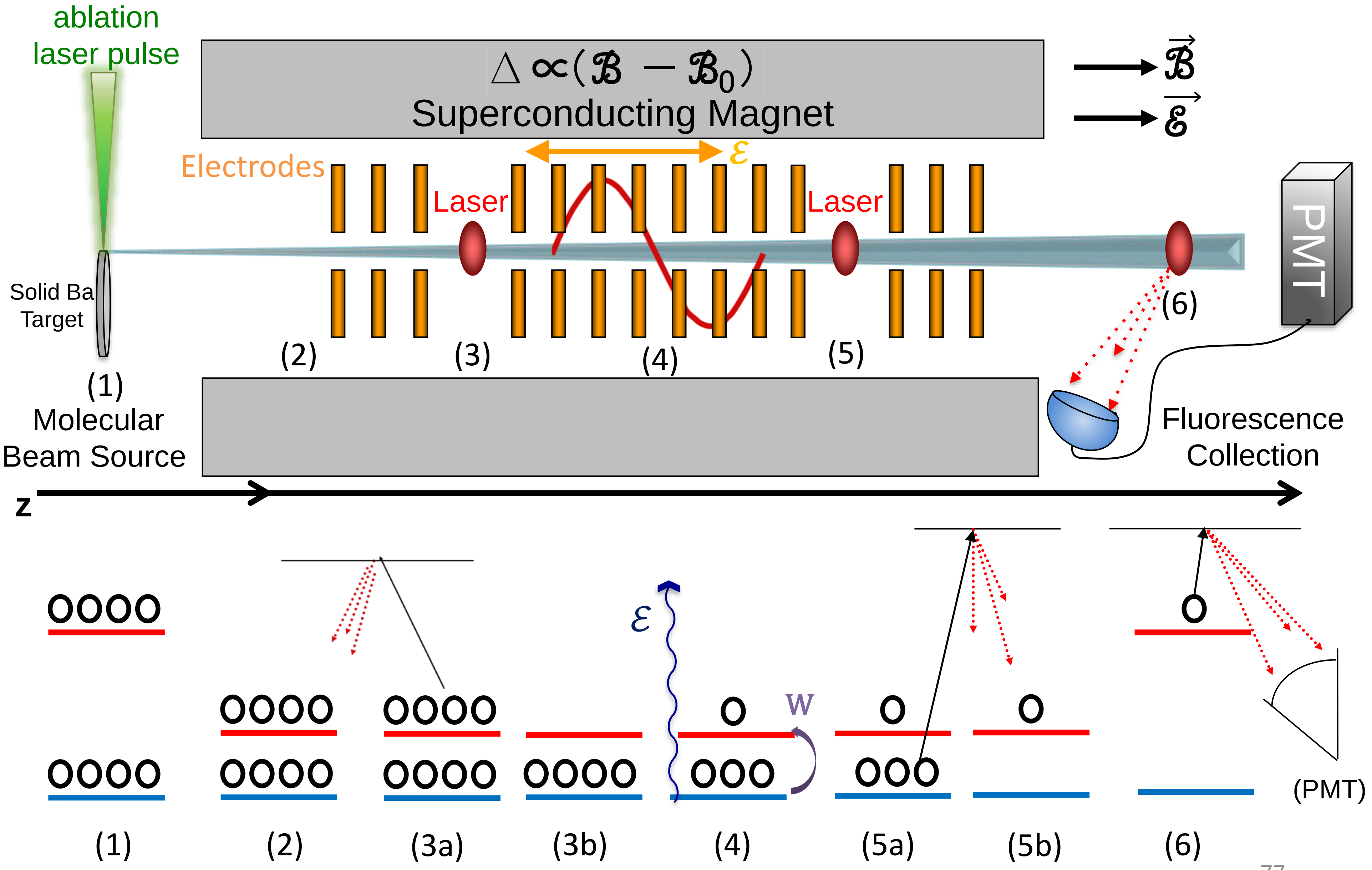
$$\delta W = \frac{1}{2\sqrt{2}}\frac{1}{\sqrt{N_0}}\frac{1}{T}$$

Best sensitivity from
large interaction time T

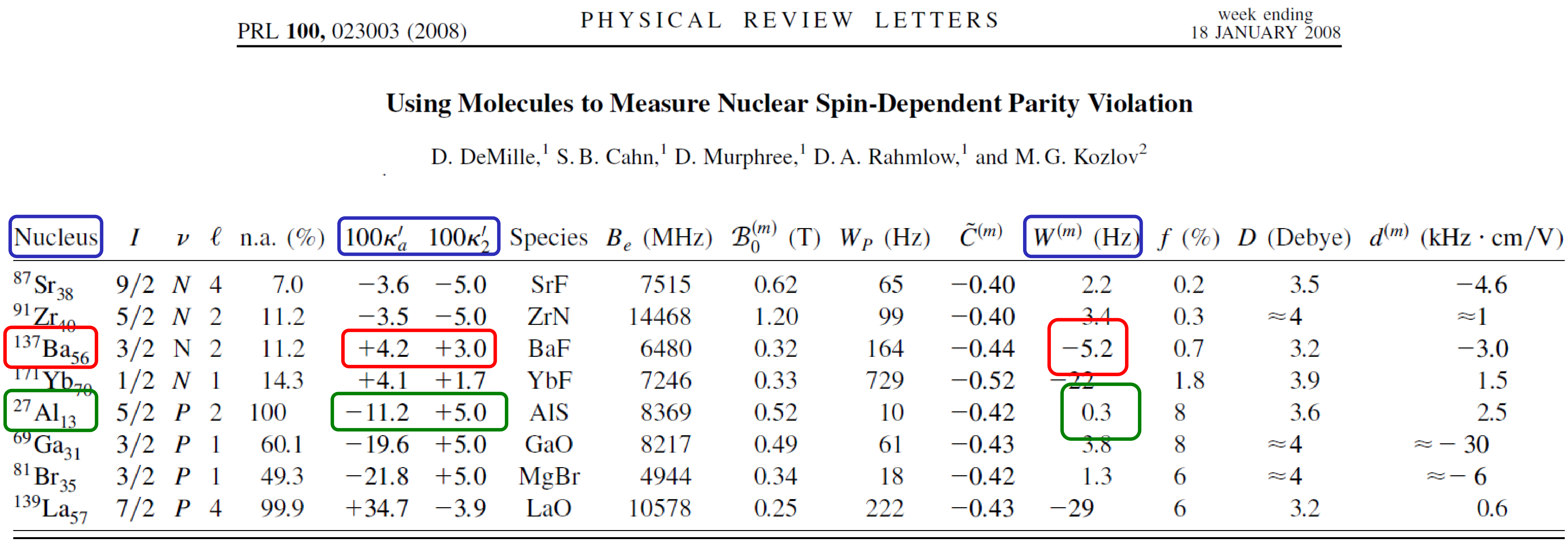
Equivalent to measuring W as generic energy shift at Standard Quantum Limit

N. Fortson, PRL **70**, 2383 (1993)

ZOMBIES experimental schematic



ZOMBIES: general-purpose technique, applicable to many isotopes



PRL **100,** 023003 (2008) PHYSICAL REVIEW LETTERS week ending 18 JANUARY 2008

Using Molecules to Measure Nuclear Spin-Dependent Parity Violation

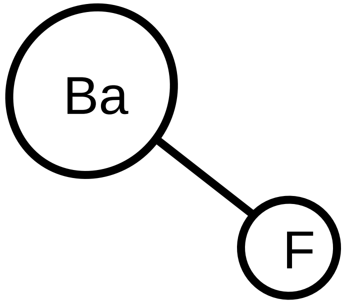
D. DeMille,[1] S. B. Cahn,[1] D. Murphree,[1] D. A. Rahmlow,[1] and M. G. Kozlov[2]

Nucleus	I	ν	ℓ	n.a. (%)	$100\kappa'_a$	$100\kappa'_2$	Species	B_e (MHz)	$\mathcal{B}_0^{(m)}$ (T)	W_P (Hz)	$\tilde{C}^{(m)}$	$W^{(m)}$ (Hz)	f (%)	D (Debye)	$d^{(m)}$ (kHz · cm/V)
$^{87}\mathrm{Sr}_{38}$	9/2	N	4	7.0	−3.6	−5.0	SrF	7515	0.62	65	−0.40	2.2	0.2	3.5	−4.6
$^{91}\mathrm{Zr}_{40}$	5/2	N	2	11.2	−3.5	−5.0	ZrN	14468	1.20	99	−0.40	3.4	0.3	≈4	≈1
$^{137}\mathrm{Ba}_{56}$	3/2	N	2	11.2	+4.2	+3.0	BaF	6480	0.32	164	−0.44	−5.2	0.7	3.2	−3.0
$^{171}\mathrm{Yb}_{70}$	1/2	N	1	14.3	+4.1	+1.7	YbF	7246	0.33	729	−0.52	−22	1.8	3.9	1.5
$^{27}\mathrm{Al}_{13}$	5/2	P	2	100	−11.2	+5.0	AlS	8369	0.52	10	−0.42	0.3	8	3.6	2.5
$^{69}\mathrm{Ga}_{31}$	3/2	P	1	60.1	−19.6	+5.0	GaO	8217	0.49	61	−0.43	3.8	8	≈4	≈ − 30
$^{81}\mathrm{Br}_{35}$	3/2	P	1	49.3	−21.8	+5.0	MgBr	4944	0.34	18	−0.42	1.3	6	≈4	≈ − 6
$^{139}\mathrm{La}_{57}$	7/2	P	4	99.9	+34.7	−3.9	LaO	10578	0.25	222	−0.43	−29	6	3.2	0.6

Improved understanding of molecular structure since 2008

→ many more viable molecule species to study many different isotopes

ZOMBIES I: NSD-PV with BaF



Initial physics goal: NSD-PV with ^{137}BaF

- Odd neutron (vs. ^{133}Cs w/odd proton)
- Heavy → large effect, anapole largest term
- Large enough natural abundance (barely)
- Required lasers = simple, cheap diodes

Recently completed: proof of principle using ^{138}Ba^{19}F

- Larger natural abundance (~75% vs ~11% for ^{137}Ba)
- Uses same beam source, lasers, magnet, etc. as ^{137}BaF
- $W(^{138}\text{Ba}) = 0$ Hz (no unpaired nucleons = no NSD-PV)
  $W(^{19}\text{F}) \approx 0.002$ Hz ≈ 0 (light, small electron spin density in BaF)
- **Test for practical sensitivity& systematics with known answer**

Uncertainties in proof-of-principle with ^{138}BaF

Strategy

- Deliberately exaggerate imperfection by known, large factor
- Measure effect on the NSD-PV matrix element *W* from coupling to ambient imperfections in the experiment

Parameter	Shift	Systematic Uncertainty $\delta W_{\rm sys}$ (Hz)
Bipolar $\mathcal{E}_{nr}$ Pulses		0.12
Unipolar $\mathcal{E}_{nr}$ Pulses		0.16
$\mathcal{B}$-Field Inhomogeneities		0.24
$\delta\nu_{L2}$ and $\mathcal{E}_{nr}$ at and near Gap 22	-0.04	0.21
Total Systematic	-0.04	0.38

Final Error Budget with ^{138}Ba^{19}F

Crossing	$W/(2\pi)$ (Hz)	C	d (Hz/(V/cm))	$W_{\rm mol} = \kappa' W_P/(2\pi)$ (Hz)
A	$0.28 \pm 0.49_{\rm stat} \pm 0.38_{\rm sys}$	-0.41	3360	$-0.68 \pm 1.20_{\rm stat} \pm 0.93_{\rm sys}$
F	$0.01 \pm 0.51_{\rm stat} \pm 0.38_{\rm sys}$	$+0.39$	3530	$0.03 \pm 1.30_{\rm stat} \pm 0.97_{\rm sys}$
Weighted Average	-	-	-	$-0.36 \pm 0.88_{\rm stat} \pm 0.95_{\rm sys}$

~170 h data
~6x10^7 molecules total

$$W_{mol} = 2\pi \times \left(-0.36 \pm 1.29\right)\ \text{Hz}$$

What does the ^{138}Ba^{19}F result mean?

$$W_{mol} \equiv \left(\kappa'_2 + \kappa'_a\right) W_P = 2\pi \times \left(-0.36 \pm 1.29\right)\ \text{Hz}$$

Most useful comparison:

$$W_P\left({}^{137}\text{Ba in BaF}\right) = 2\pi \times 160\ \text{Hz}$$

Same experimental uncertainty in ^{137}BaF would mean

$$\underbrace{\delta\kappa'\left({}^{137}\text{Ba}\right) = 0.008 \quad \text{vs. } \kappa'\left({}^{137}\text{Ba}\right)\left[\text{shell model}\right] \approx 0.07}_{\textbf{\textasciitilde10\% of predicted value}}$$

Compares favorably to JILA ^{133}Cs result: $\kappa'\left({}^{133}\text{Cs}\right) = 0.39 \pm 0.06$

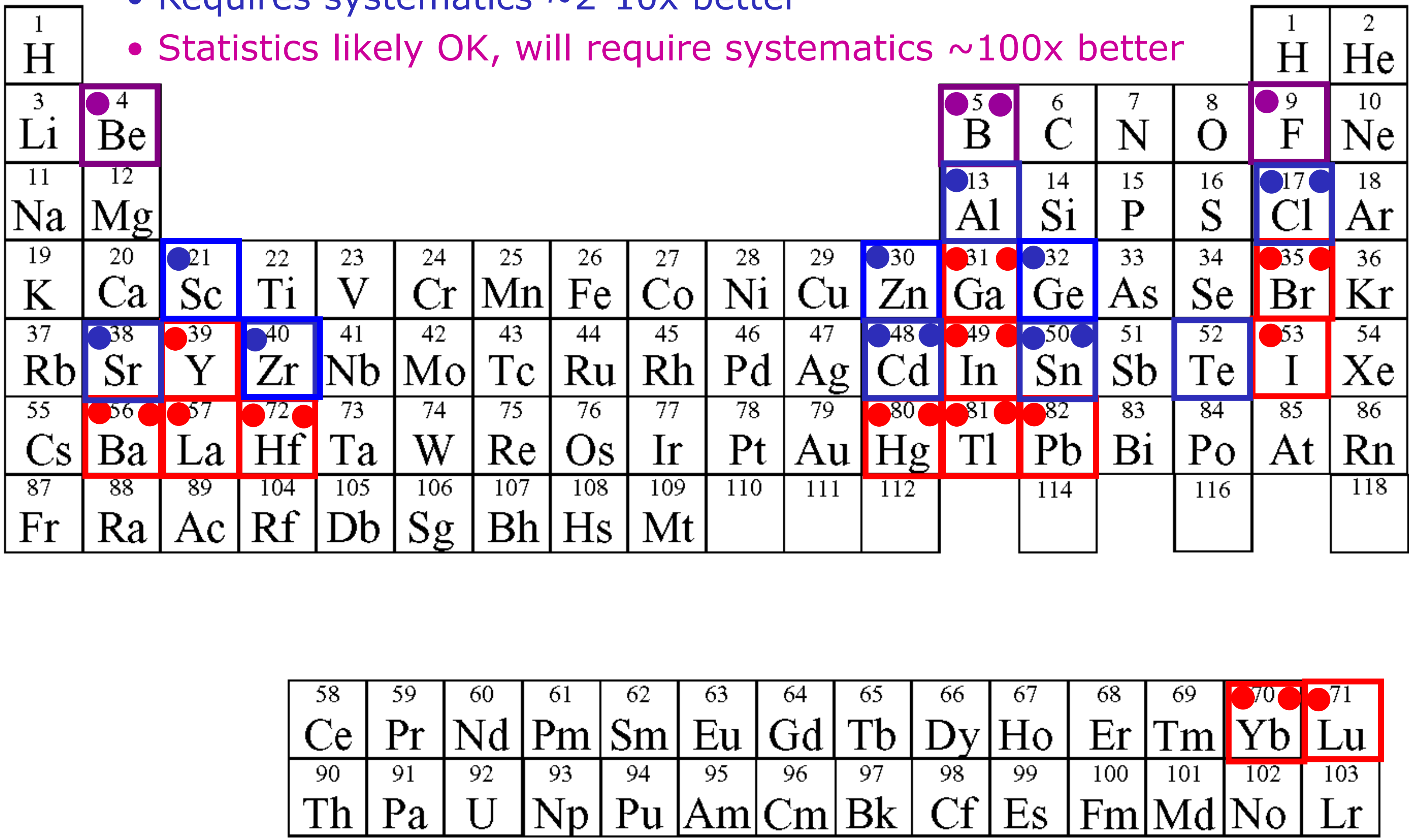
C.S. Wood *et al.*, Science **275**, 1759 (1997)

- Unprecedented sensitivity to NSD-PV
- General technique enables measurements in broad range of nuclei

E. Altuntas, J. Ammon, S.B. Cahn, DD PRL **120**, 142501 (2018); PRA **97**, 042101 (2018)

Viable nuclei for anapole/NSD-PV measurement with ZOMBIES

- 10% measurement possible with demonstrated sensitivity, $\lesssim$100 h data
- Requires systematics ~2-10x better
- Statistics likely OK, will require systematics ~100x better



Questions/requests for theorists

--Anapole moment calculations with new HPV χEFT parameterization

--Calculate anapole moments & weak axial coupling for mid-mass nuclei

--Quantitative uncertainties on calculations!

--Could C_2 values be extracted reliably from mid-mass nuclei with existing HPV data & understanding?

--Is consistency check between isotopes in heavier nuclei useful?

--Are there special cases of particular interest/particularly easy to calculate?

--Can anapole measurements (with known HPV inputs) shed light on other related calculations
e.g. Schiff moment, $0\nu\beta\beta$ decay, …?

--Generally: modern theory perspective on anapole moments *URGENTLY* needed
(>10 years since last dedicated nuclear theory paper)

--"quick and dirty" calculations of molecular sensitivities for "new" species
(e.g. ^{133}CsMg, Xe^{19}F, …)

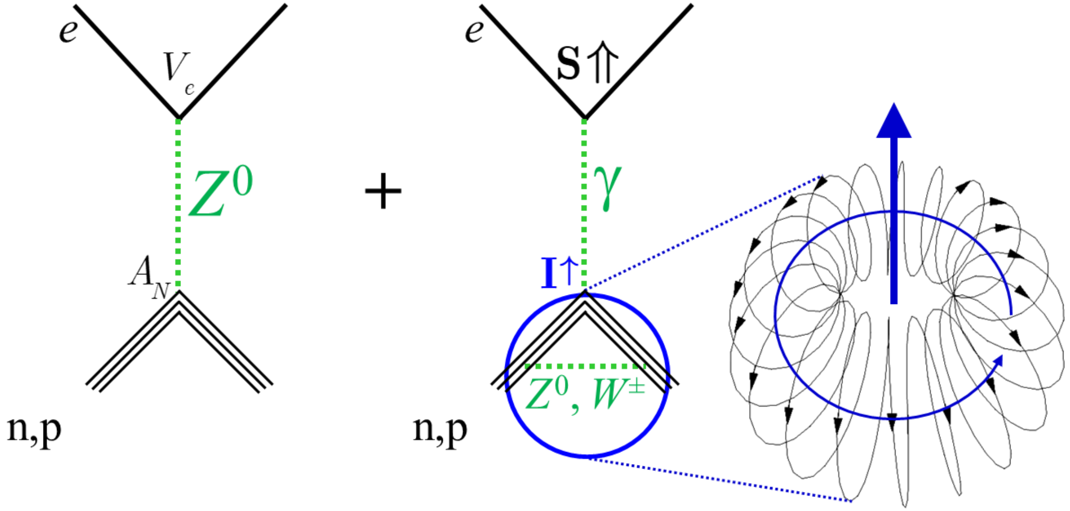
ZOMBIES: Summary & Outlook

--New era in NSD-PV = anapole + V_eA_N measurements beginning

--Sensitivity & accuracy of molecular systems likely to enable measurements on many nuclei, including light-ish species, with <10% uncertainty

--Complementary to SoLID/PVDIS @ JLab & other hadronic PV experiments

--long term vision: unify understanding of hadronic & semileptonic PV interactions in strongly-interacting environment, across wide range of scales



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