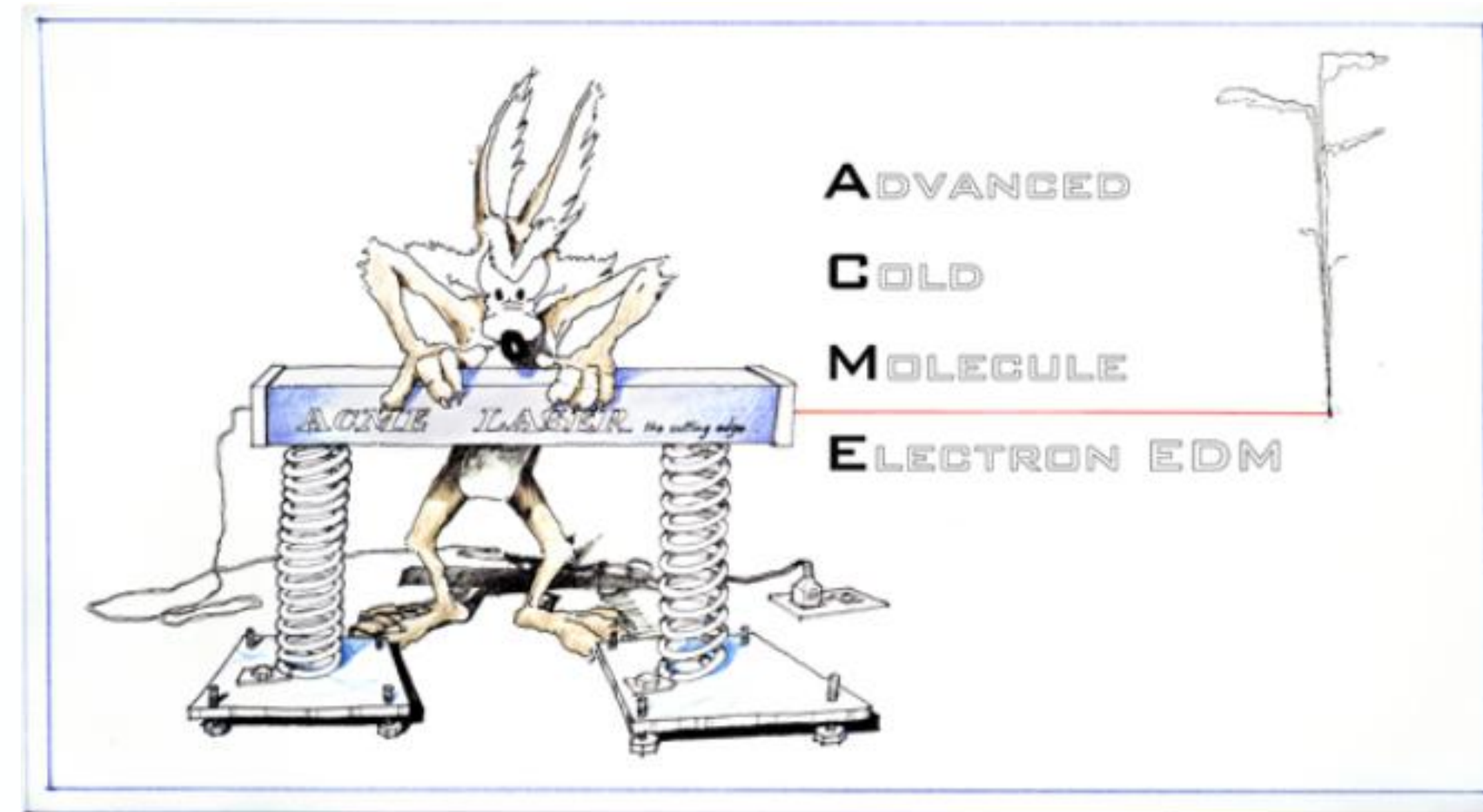


ACME III: electron EDM search using cold ThO molecular beam



Takahiko MASUDA on behalf of the ACME collaboration



Research Institute for Interdisciplinary science,
Okayama University,

ACME Collaboration



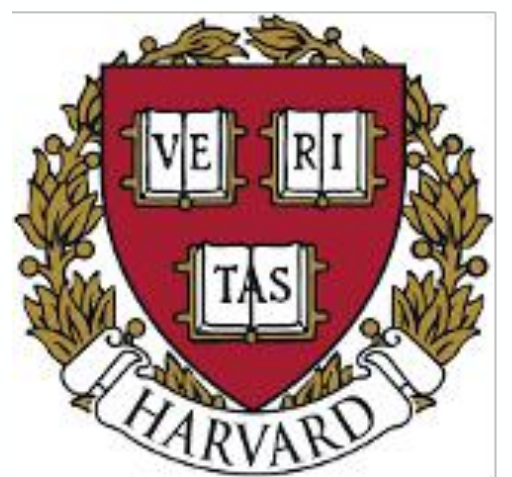
David DeMille



John Doyle



Gerald Gabrielse



Northwestern University



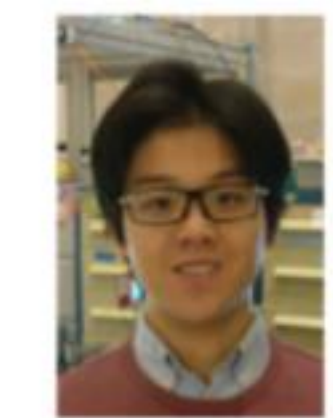
THE UNIVERSITY OF CHICAGO



OKAYAMA UNIVERSITY

Caltech

Berkeley UNIVERSITY OF CALIFORNIA



Zhen Han



Xing Wu



Xing Fan



Siyuan Liu



Collin Diver



Nick Hutzler



Peiran Hu



Daniel Ang



Cole Meisenhelder



Maya Watts



Koji Yoshimura



Satoshi Uetake



Naboru Sasao



Takahiko Masuda



Ayami Hiramoto



Cris Panda

Acknowledgements:

- National Science Foundation
- Gordon & Betty Moore Foundation
- Alfred P. Sloan Foundation
- JSPS Kakenhi
- JST SICORP
- Matsuo Foundation

- Physics background and motivations
- Quick review of ACME:
 - Experimental approach and prior results from ACME II
- Improvements with ACME III
 - Statistics improvements
 - Systematics improvements

Baryonic asymmetry

Matter universe

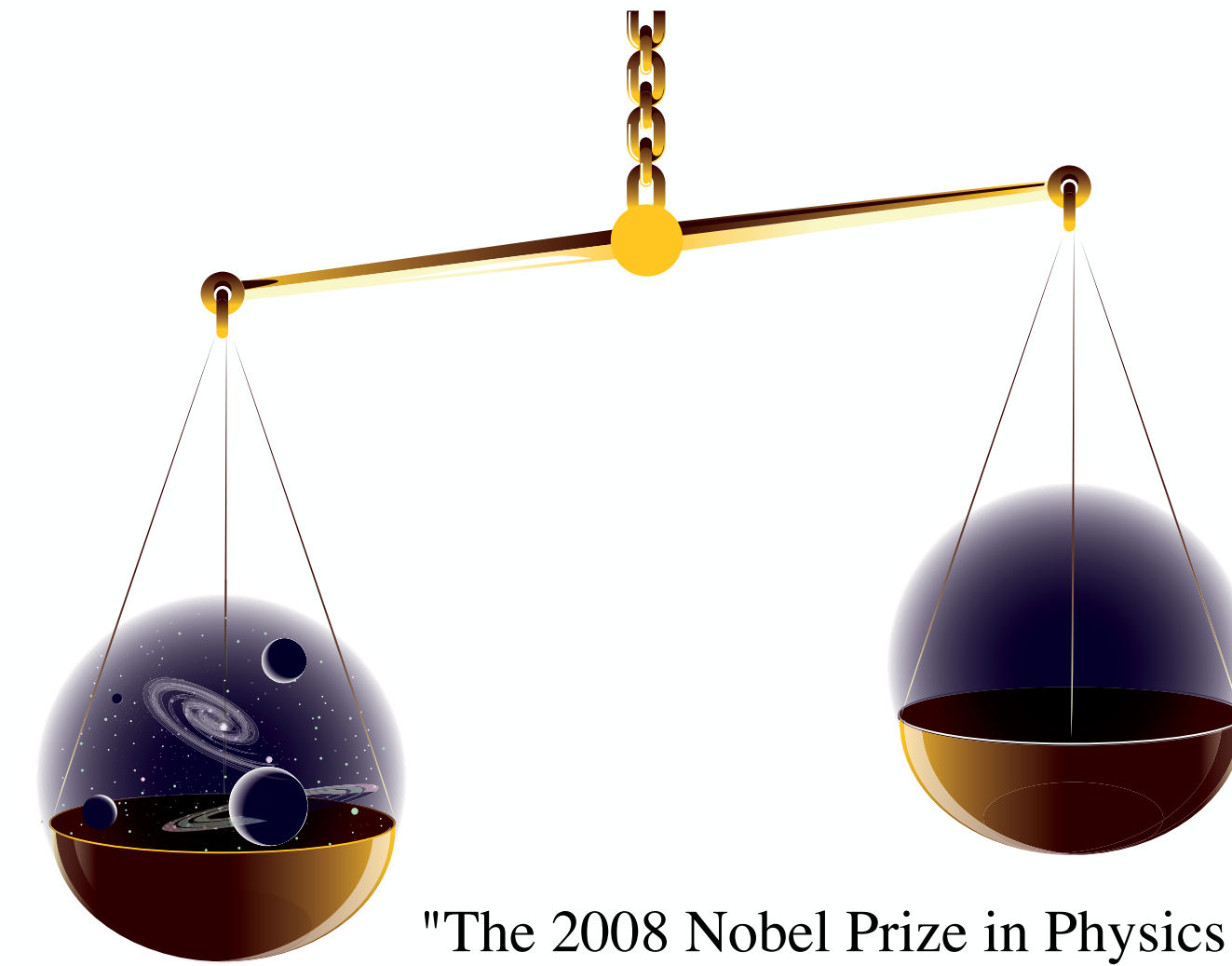
Why is the universe dominated by matter?

Observation : $n_B/n_\gamma \sim 6 \times 10^{-10}$

P.A. Zyla *et al.* (PDG), PTEP **2020**, 083C01 (2020).

Calculation : $n_B/n_\gamma \sim 10^{-18-20}$

8-10 orders
different!



"The 2008 Nobel Prize in Physics - Popular Information".
Nobelprize.org. Nobel Media AB 2013. Web. 25 Nov 2013.

http://www.nobelprize.org/nobel_prizes/physics/laureates/2008/popular.html

Sakharov conditions

Sakharov A. D., JETP **5** pp.24-26 (1967)

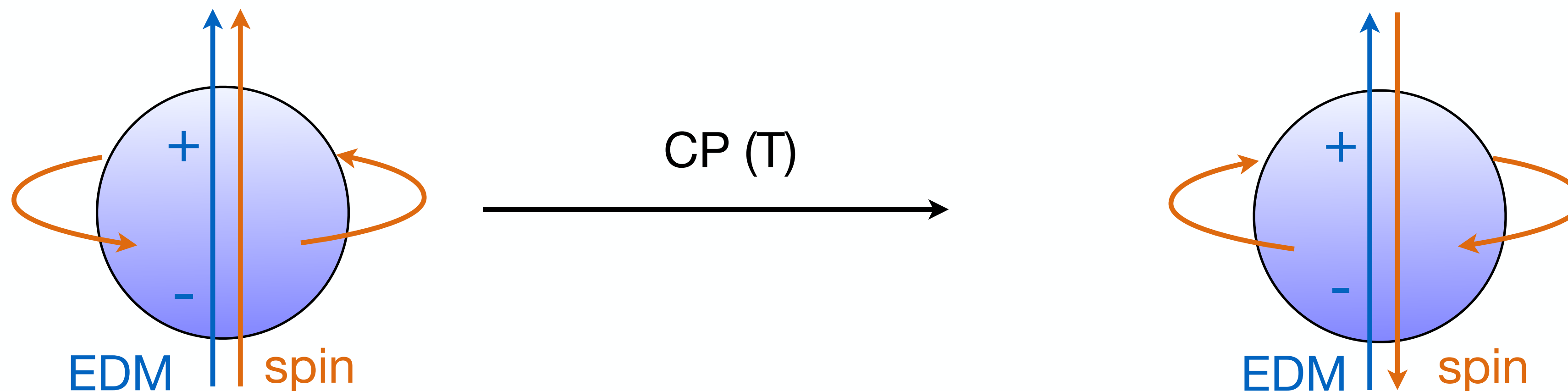
- Baryon number ***B*** violation
- ***C***-symmetry and ***CP***-symmetry violation
- Out of thermal equilibrium

→ There must be a term that violates ***CP***-symmetry beyond the Standard Model

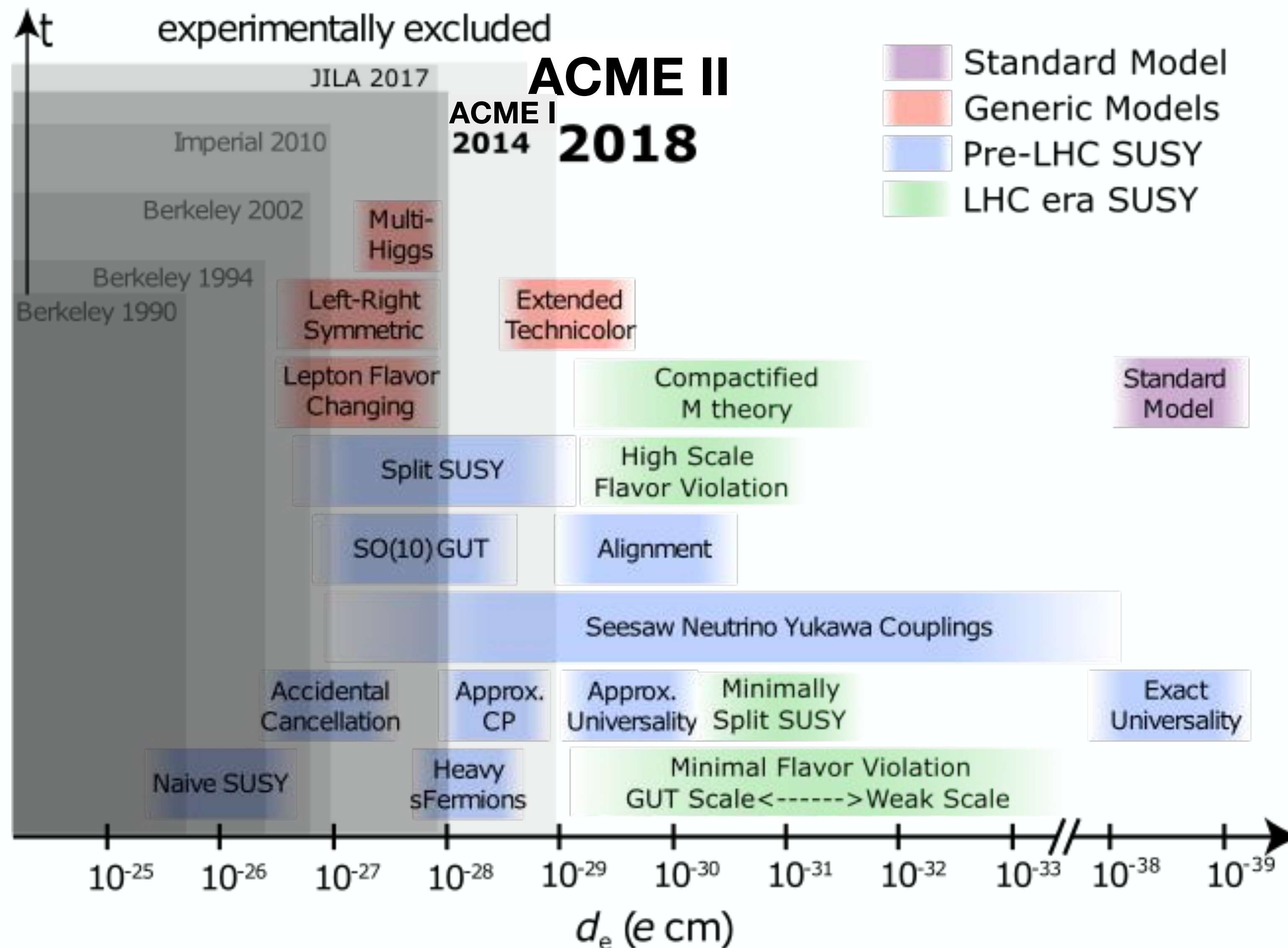
EDM : electric dipole moment

EDM : Permanent electric polarization of internal charge

If a spin 1/2 particle has finite EDM, it **violates** the CP symmetry.

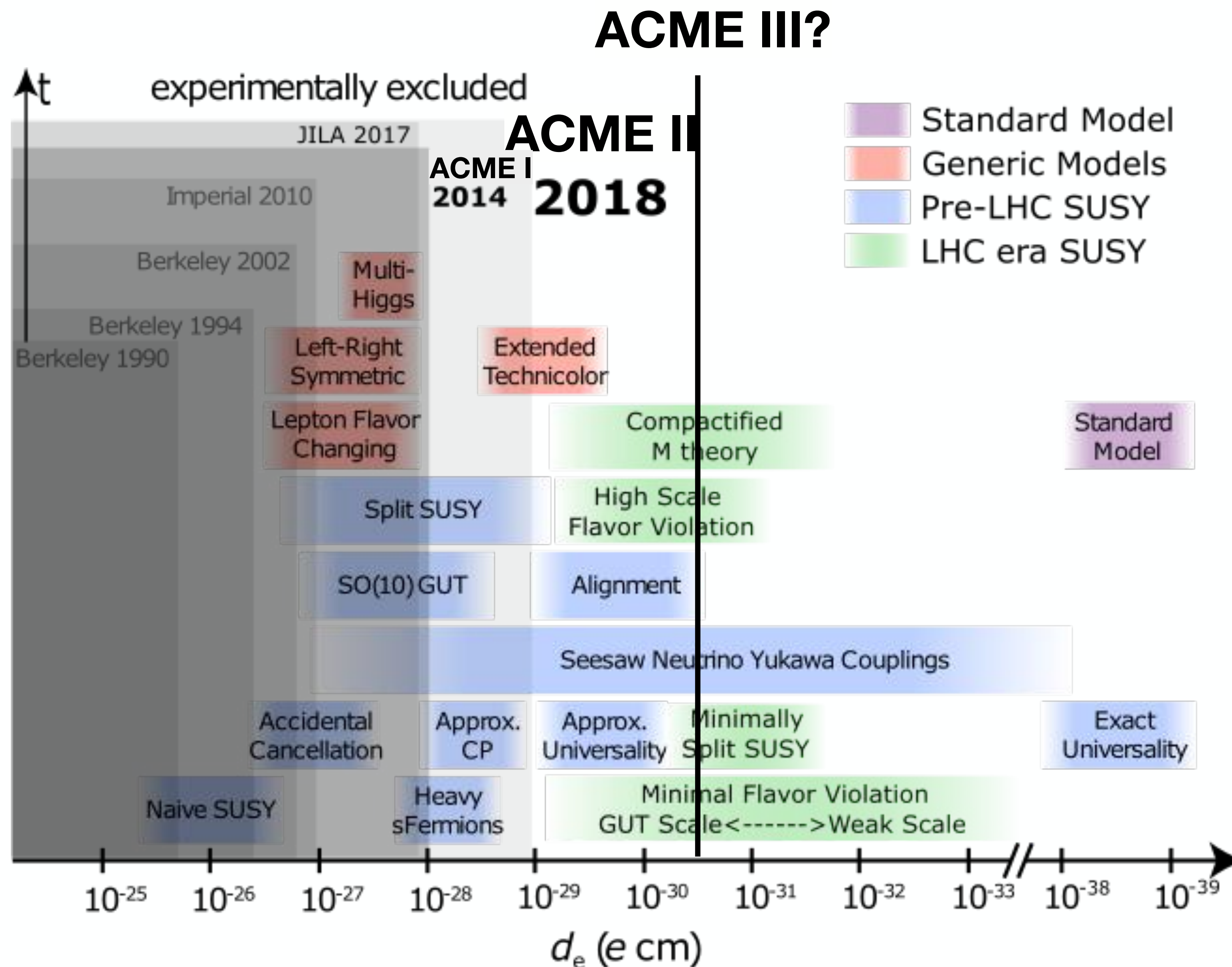


Electron EDM experiments



ACME II : $|d_e| < 1.1 \times 10^{-29}$ e cm
 ACME Collab. Nature **562** (2018) 355.

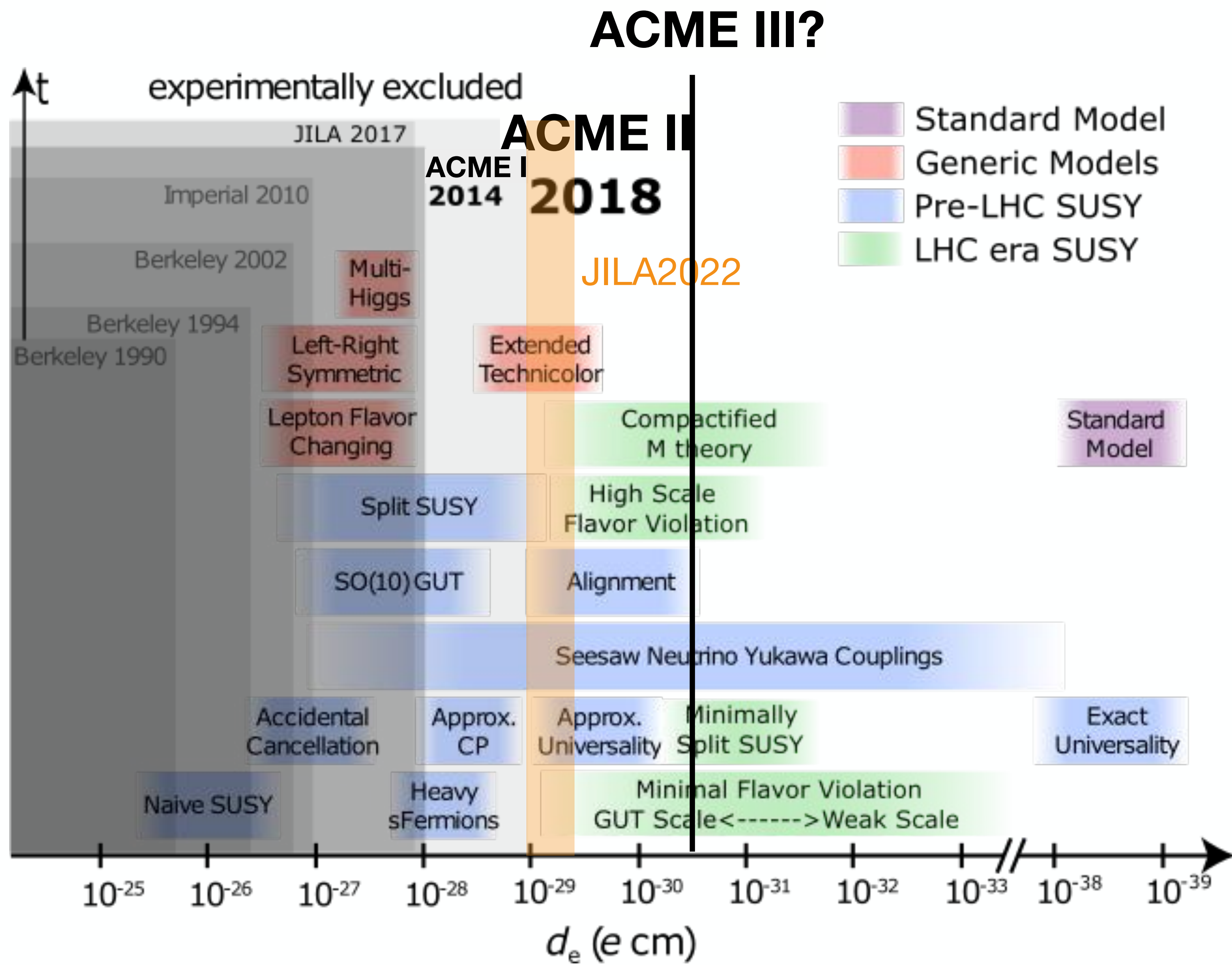
Electron EDM experiments



ACME II : $|d_e| < 1.1 \times 10^{-29}$ e cm
 ACME Collab. Nature **562** (2018) 355.

ACME III goal:
 $\times 30$ improvement
Today's topic

Electron EDM experiments

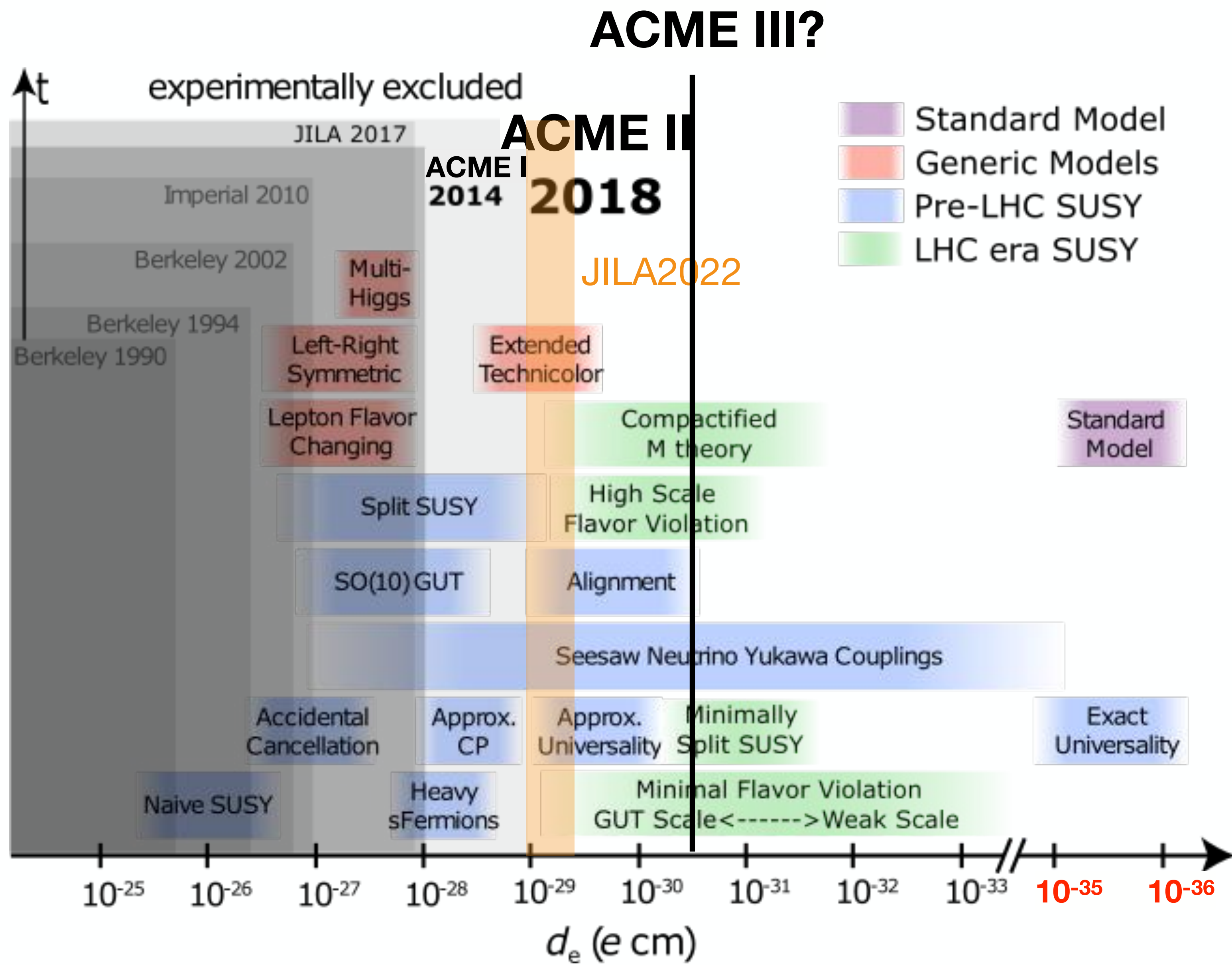


JILA group has recently reported $|d_e| < 4.1 \times 10^{-30}$ e cm on arXiv:2212.11841

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ACME Collab. Nature **562** (2018) 355.

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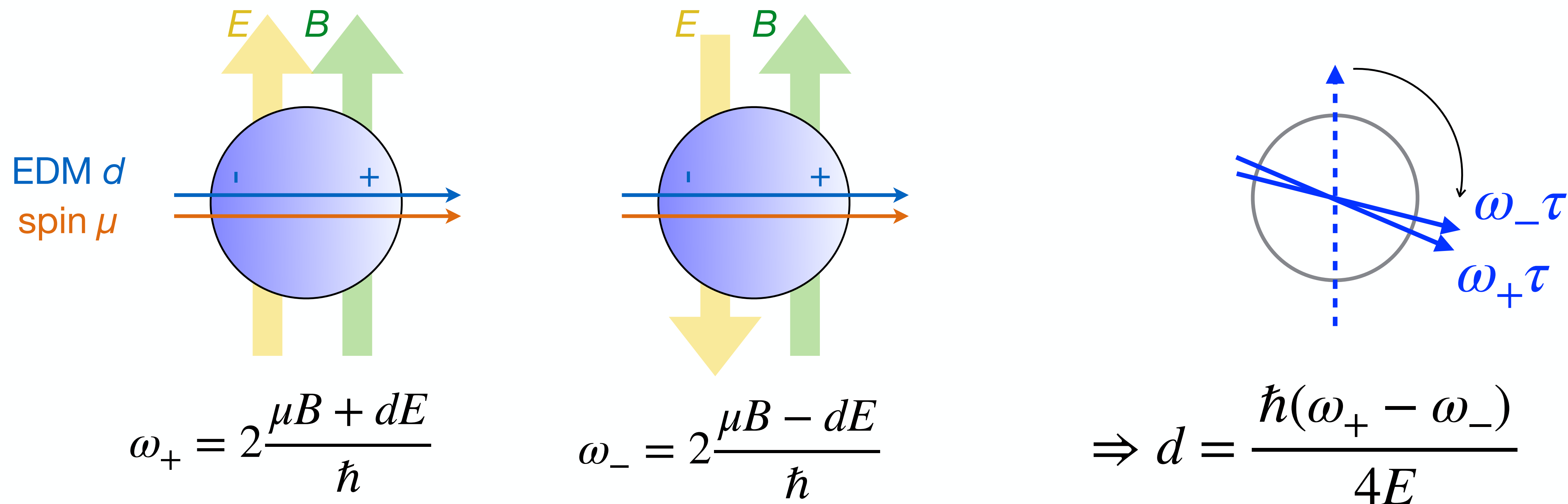
ACME III goal:
× 30 improvement
Today's topic

SM background has been updated.
Y. Ema *et al.*, PRL**129**, 231801 (2022)

Principle of the EDM measurement

Spin precession measurement:

EDM changes precession frequency depending on E field reversal.



Statistical precision : $\Delta d_e \sim \frac{\hbar}{E \tau} \frac{1}{\sqrt{nT}}$

Electric field

precession time

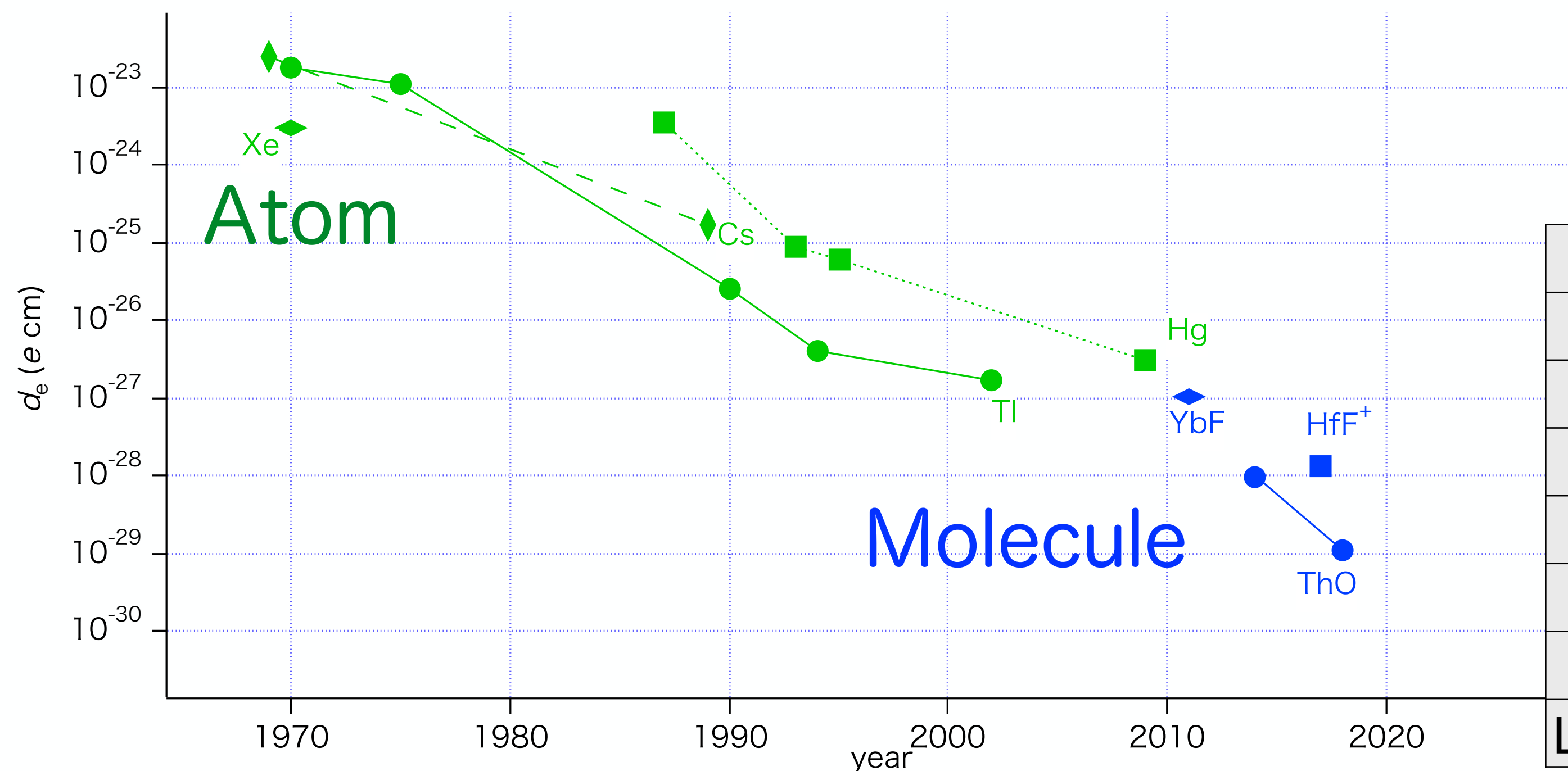
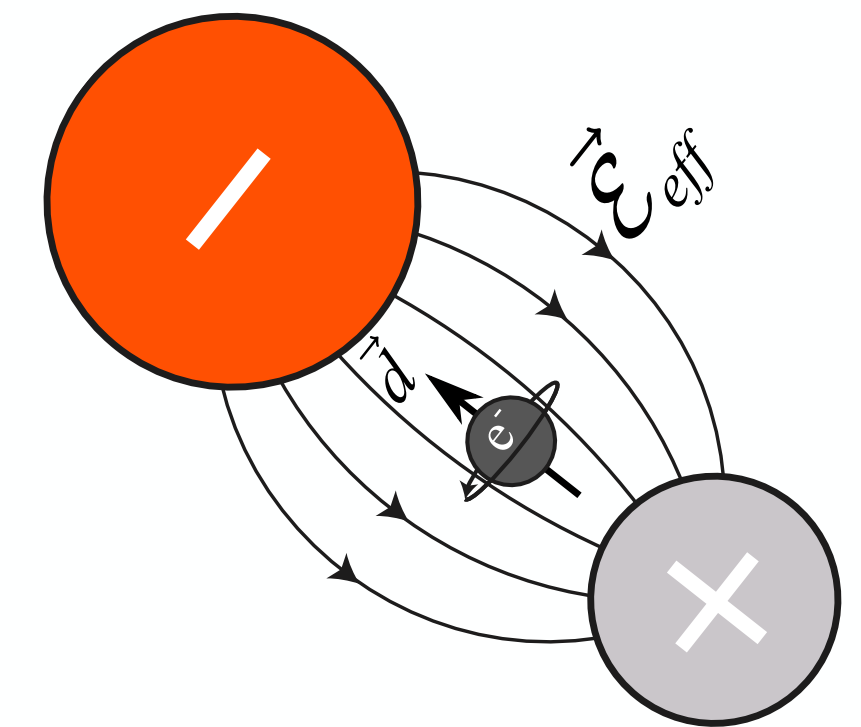
Number of signals

Electron EDM (eEDM, d_e)

eEDM measurement in polar molecules.

Sensitivity for d_e can be amplified in polar molecules due to its large internal field.

$$\Delta d_e \sim \frac{\hbar}{E\tau} \frac{1}{\sqrt{\dot{n}T}} \quad \Rightarrow \quad \Delta d_e \sim \frac{\hbar}{E_{\text{eff}}\tau} \frac{1}{\sqrt{\dot{n}T}}$$



	E_{eff} [GV/cm]	Reference
YbF	25	J. Phys. B 30 , L607 (1997)
ThO	78	J. Chem. Phys. 145 , 214301 (2016)
HfF ⁺	23	Phys. Rev. A 96 , 040502 (2017)
ThF ⁺	35	New J. Pays. 17 , 043005 (2015).
PbO	25	Phys. Rev. Lett. 89 , 133001 (2002).
LrO, LrF ⁺ , LrH ⁺	250-340	Phys. Rev. A 104 , 062801 (2021).

Advantage of ThO

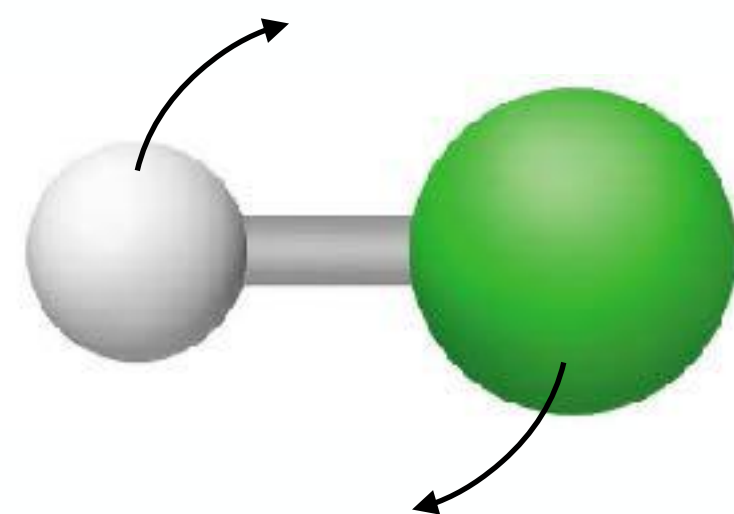
Ω -doublet *Nearly degenerated parity doublet*

Modest external E field can generate extremely **strong internal E field (GV/cm)**

Rotational energy

$$\Delta E \sim \text{GHz}$$

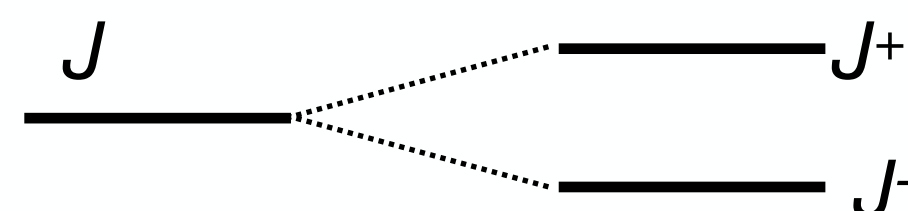
$$P \sim O(1) \text{ @ } 10 \text{ kV/cm}$$



Ω -doublet case

$$\Delta E \sim \text{MHz}$$

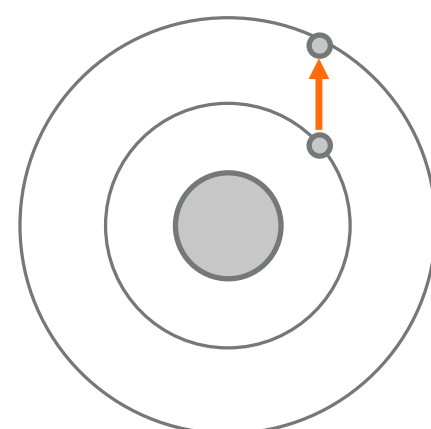
$$P \sim O(1) \text{ @ } 10 \text{ V/cm}$$



(Ref.) Atom case

$$\Delta E \sim \text{THz}$$

$$P \sim O(10^{-3}) \text{ @ } 10 \text{ kV/cm}$$

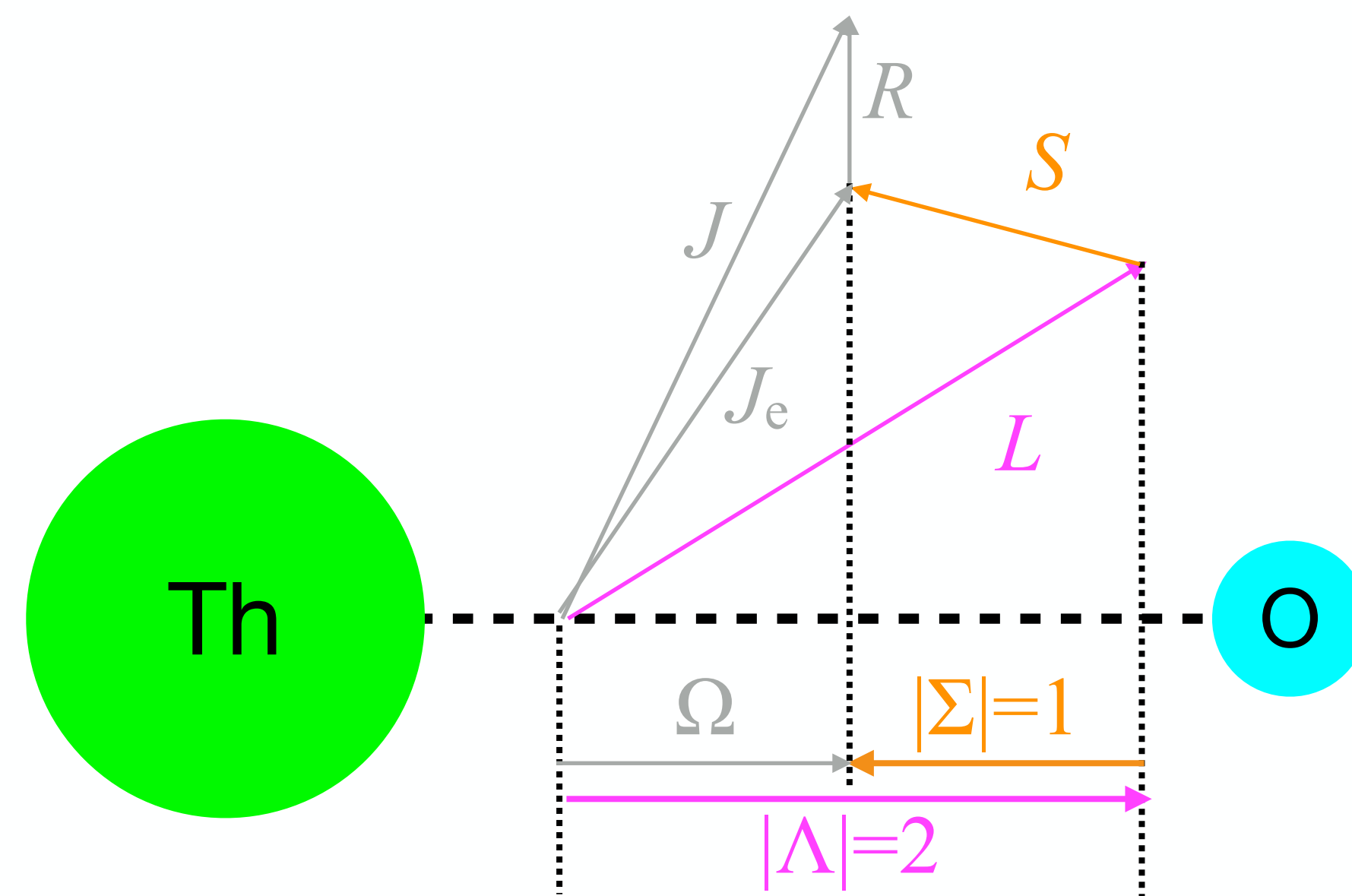


$^3\Delta_1$ *State without magnetic moment*

Electron's angular momentum ($\Lambda=+2$) and spin ($\Sigma=-1$) **vanish** the net magnetic moment

→ **Insensitive** to the magnetic field

$$\mu = g_L \underset{2}{\Lambda} \mu_B + g_S \underset{-2}{\Sigma} \mu_B \simeq 0$$



EDM in a ThO molecule

EDM measurement state : $H^3\Delta_1$ state

Strong internal E field (Ω doublet) : $E_{\text{eff}} \sim 78$ GV/cm

- 10 V/cm external field can saturate the polarization

L.V. Skripnikov, J. Chem. Phys. **145** 214307 (2016).

Tiny magnetic moment ($^3\Delta_1$): $\mu=0.0044\mu_B$

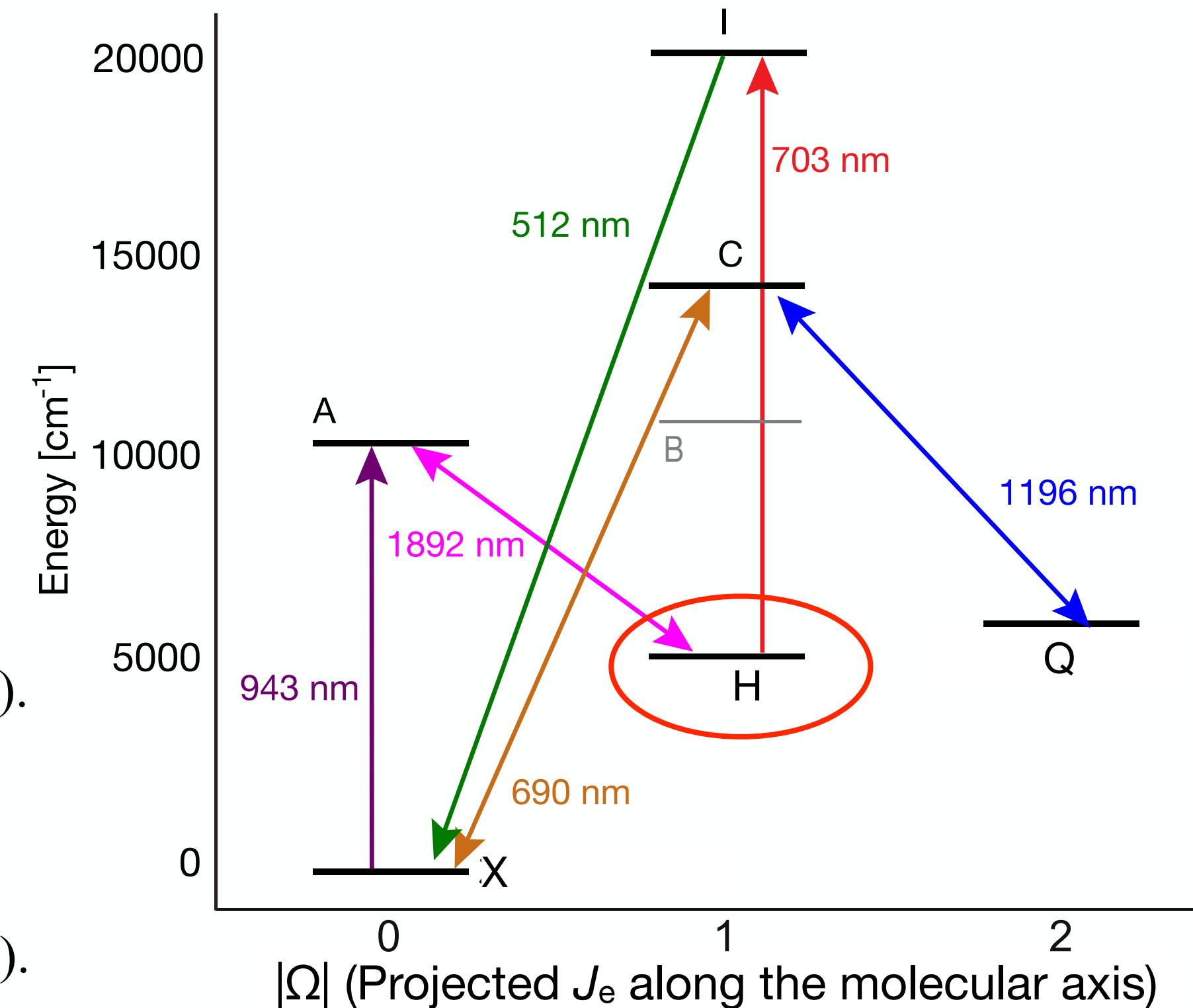
- Insensitive to the magnetic field

L.V. Skripnikov *et al.*, J. Chem. Phys. **139** 221103 (2013).

- Long lifetime (spin precession time) : $\tau = 4.2 \pm 0.5$ ms

D. Ang *et al.*, PRA**106**, 022808 (2022).

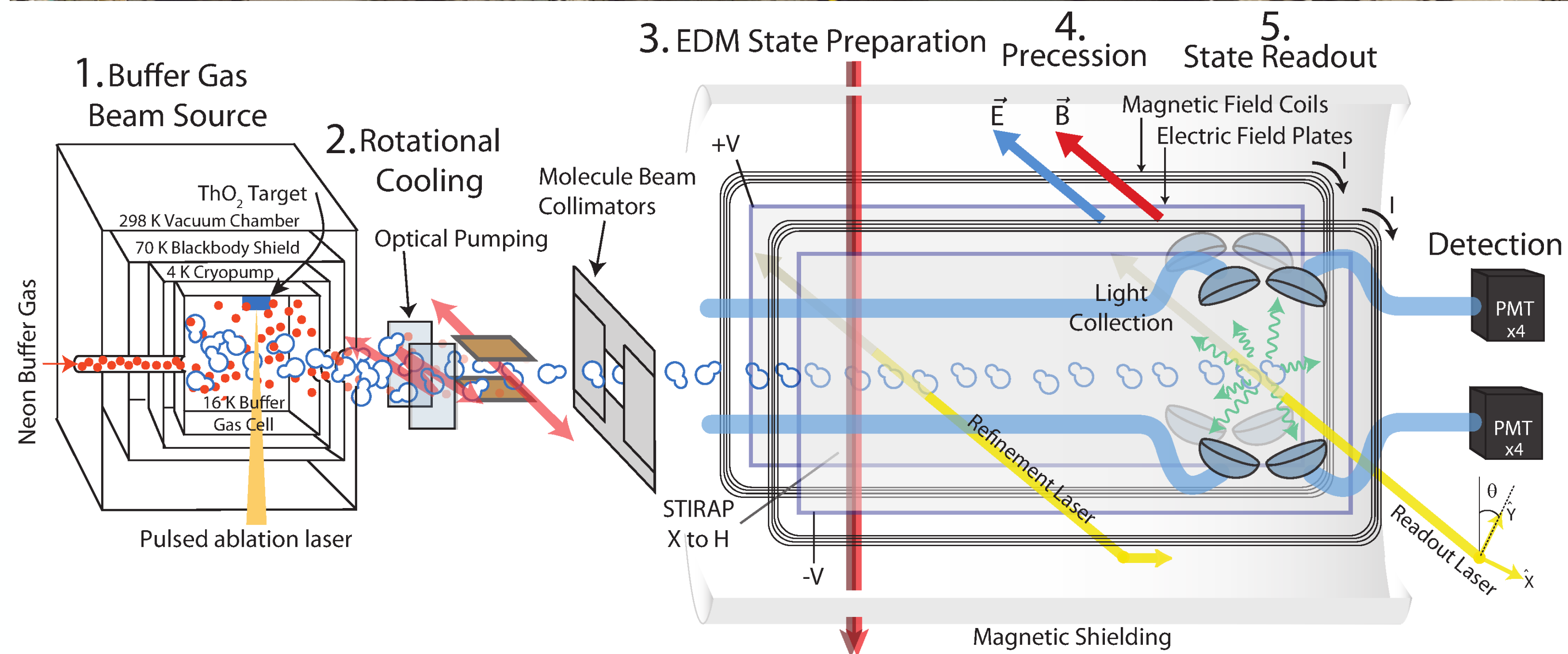
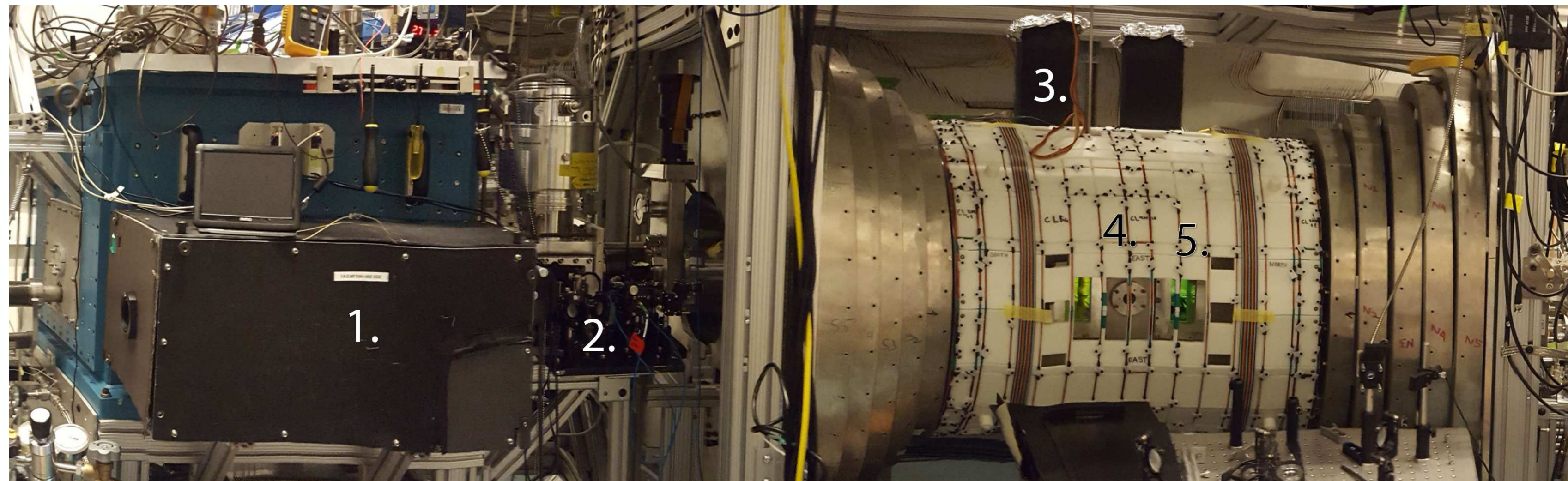
- Diatomic, Even nucleus, for spectroscopic simplicity
- Affordable laser : Red - NIR
- Efficiently producible in a beam by laser ablation
- $^{232}\text{Th}^{16}\text{O}$ Natural abundance >99.7%
- etc.



$$\Delta d_e \sim \frac{\hbar}{E_{\text{eff}} \tau} \frac{1}{\sqrt{nT}}$$

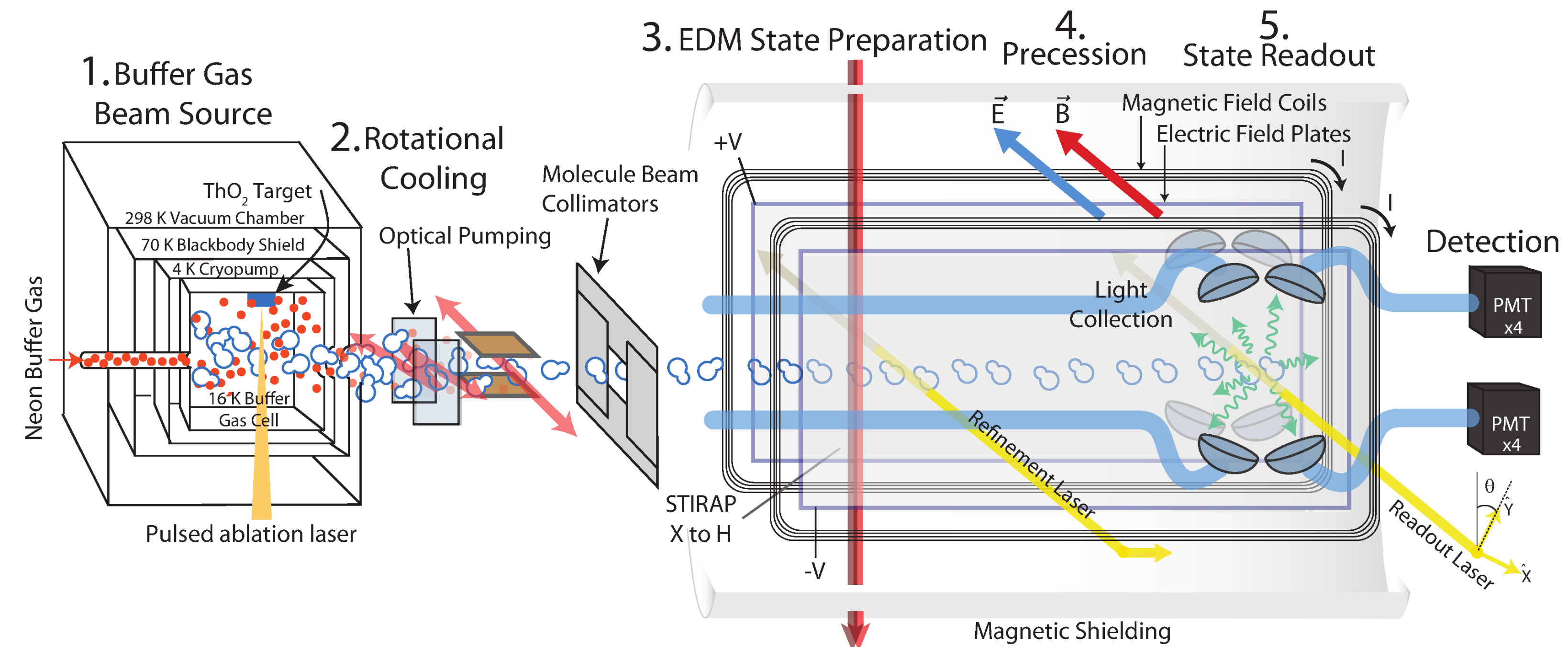
Quick review on ACME II (2018)

Zack Laser, Ph.D thesis. (2019)



Quick review on ACME II (2018)

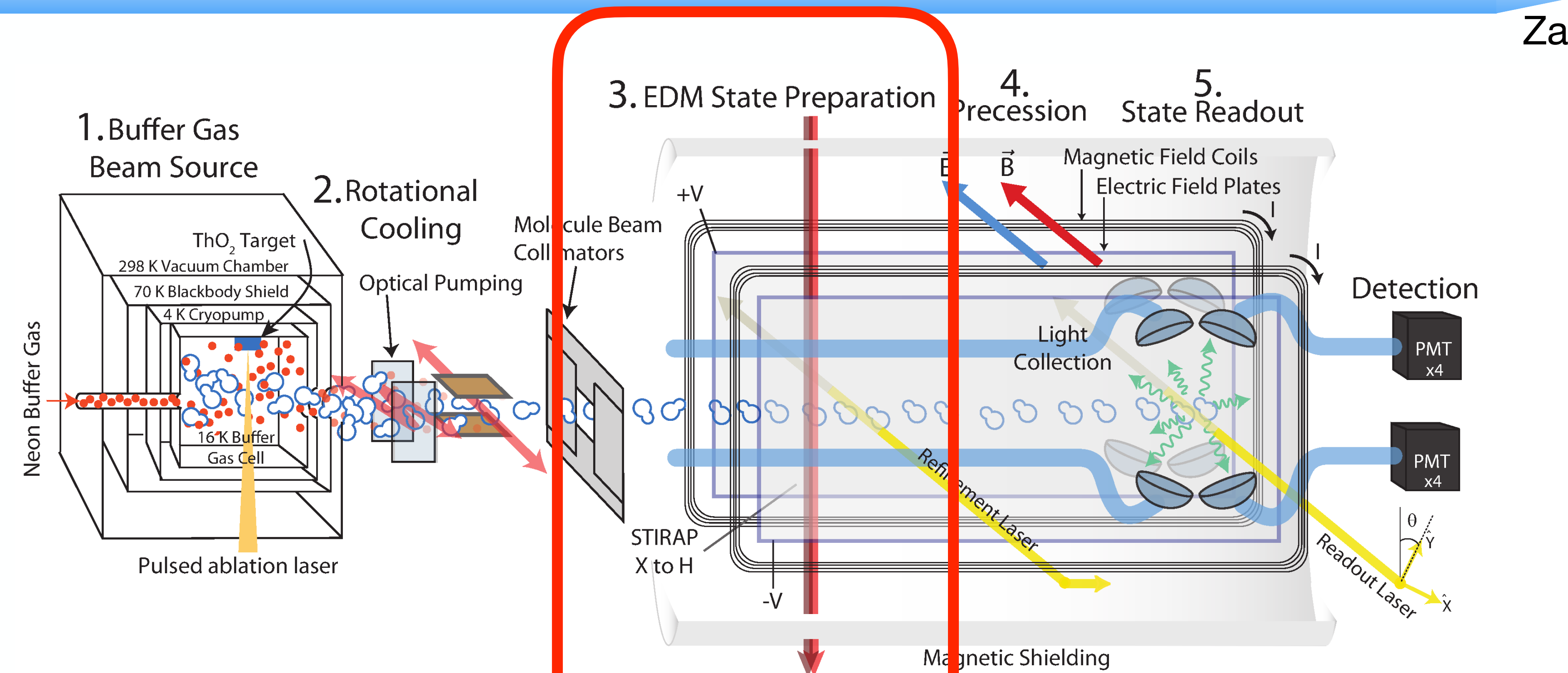
Zack Laser, Ph.D thesis. (2019)



	1. Beam Source	2a. Rotational Cooling	2b. Rotational Cooling	3a. EDM State Preparation	3b. State Refinement	4. Spin Precession	5. State Readout
State	$ X\rangle$	$ X, J=0,1\rangle$	$ X, J=0\rangle$	$ H, J=1\rangle$	$\frac{1}{\sqrt{2}}(M=-1\rangle + M=+1\rangle)$	$\frac{1}{\sqrt{2}}(e^{i\phi} M=-1\rangle + e^{-i\phi} M=+1\rangle)$	
Laser/field direction		\otimes	$\uparrow \epsilon$	\downarrow	\otimes	ϵ, \mathcal{B}	\otimes
Spin Orientation		\otimes	\otimes	\otimes	\otimes	\otimes	\otimes
Laser beam polarization		x2				ϕ	θ
Energy Level Diagram							

Quick review on ACME II (2018)

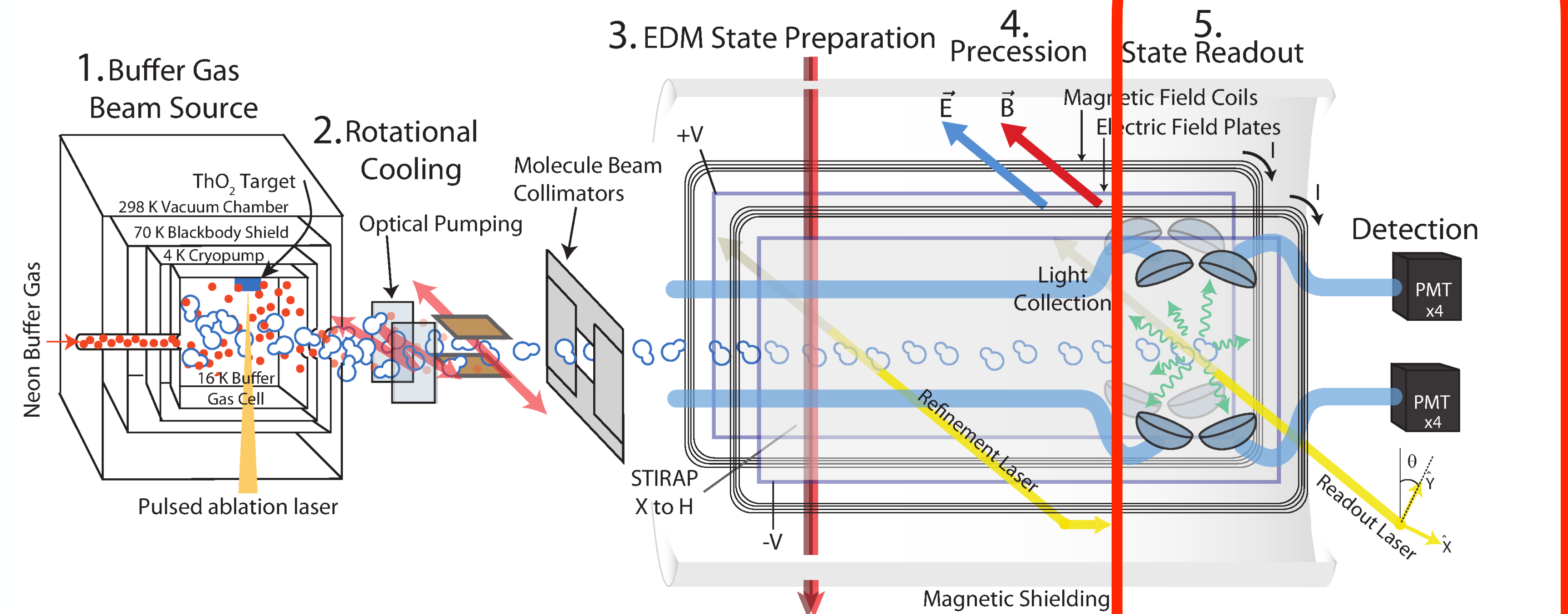
Zack Laser, Ph.D thesis. (2019)



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Laser/field direction		\otimes	$\uparrow \epsilon$	\downarrow	\otimes	ϵ, \mathcal{B}	\otimes
Spin Orientation		\otimes	\otimes	\odot	\otimes	\odot	\otimes
Laser beam polarization		x2	x2	\odot	\otimes	ϕ	θ
Energy Level Diagram							

Quick review on ACME II (2018)

Zack Laser, Ph.D thesis. (2019)



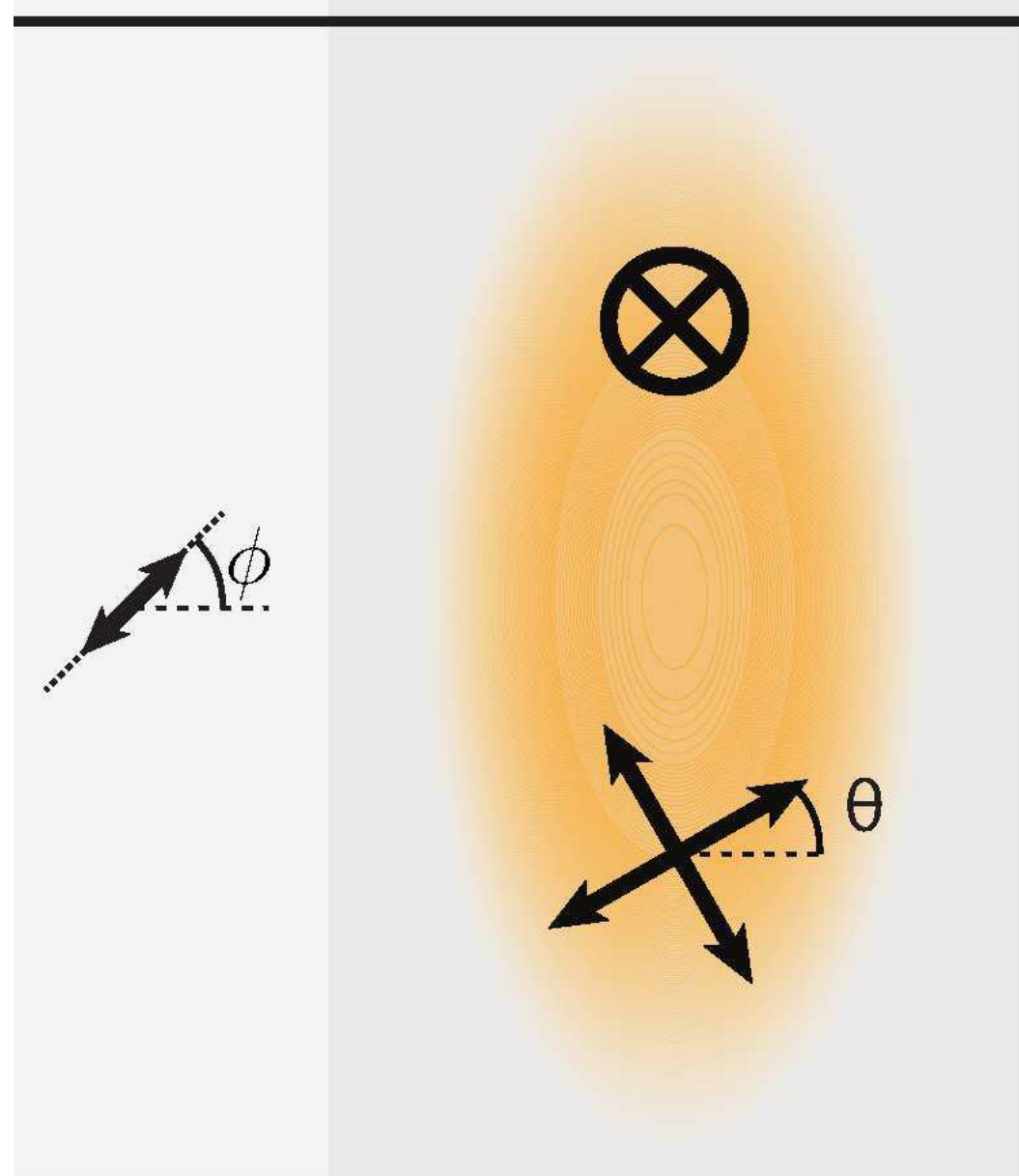
	1. Beam Source	2a. Rotational Cooling	2b. Rotational Cooling	3a. EDM State Preparation	3b. State Refinement	4. Spin Precession	5. State Readout
x y							
	$ X\rangle$	$ X, J=0,1\rangle$	$ X, J=0\rangle$	$ H, J=1\rangle$	$\frac{1}{\sqrt{2}}(M=-1\rangle + M=+1\rangle)$	$\frac{1}{\sqrt{2}}(e^{i\phi} M=-1\rangle + e^{-i\phi} M=+1\rangle)$	
Laser/field direction		\otimes	\otimes	\downarrow	\otimes	\otimes	\otimes
Spin Orientation		\uparrow	\uparrow	\odot	\leftrightarrow	\leftrightarrow	\leftrightarrow
Laser beam polarization		$x2$				ϕ	θ
		overlapped					
I C H X	1090 512 690 703	690 nm	690 nm	690 nm 1090 nm	703 nm	$-2(\vec{B}\mu_{Bg}B_z + \vec{N}\tilde{\mathcal{E}}d_e\mathcal{E}_{eff}) = 2\phi/\tau$	512 nm 703 nm
	$4+ \quad 3 \quad 2 \quad 1 \quad 0$	$X \quad 3 \quad 2 \quad 1 \quad 0$	$X \quad 2 \quad 1 \quad 0$	$X \quad 0 \quad H \quad 1 \quad = \quad =$	$H \quad 1 \quad = \quad =$	$M = -1 \quad M = +1$	$H \quad 1 \quad = \quad =$

Example of spin rotation fringe

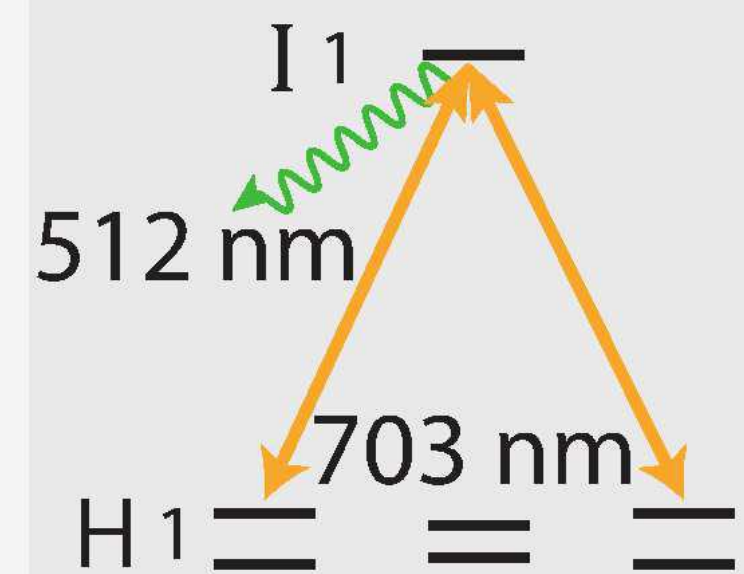
Cris Panda, Ph.D thesis. (2018)

5. State Readout

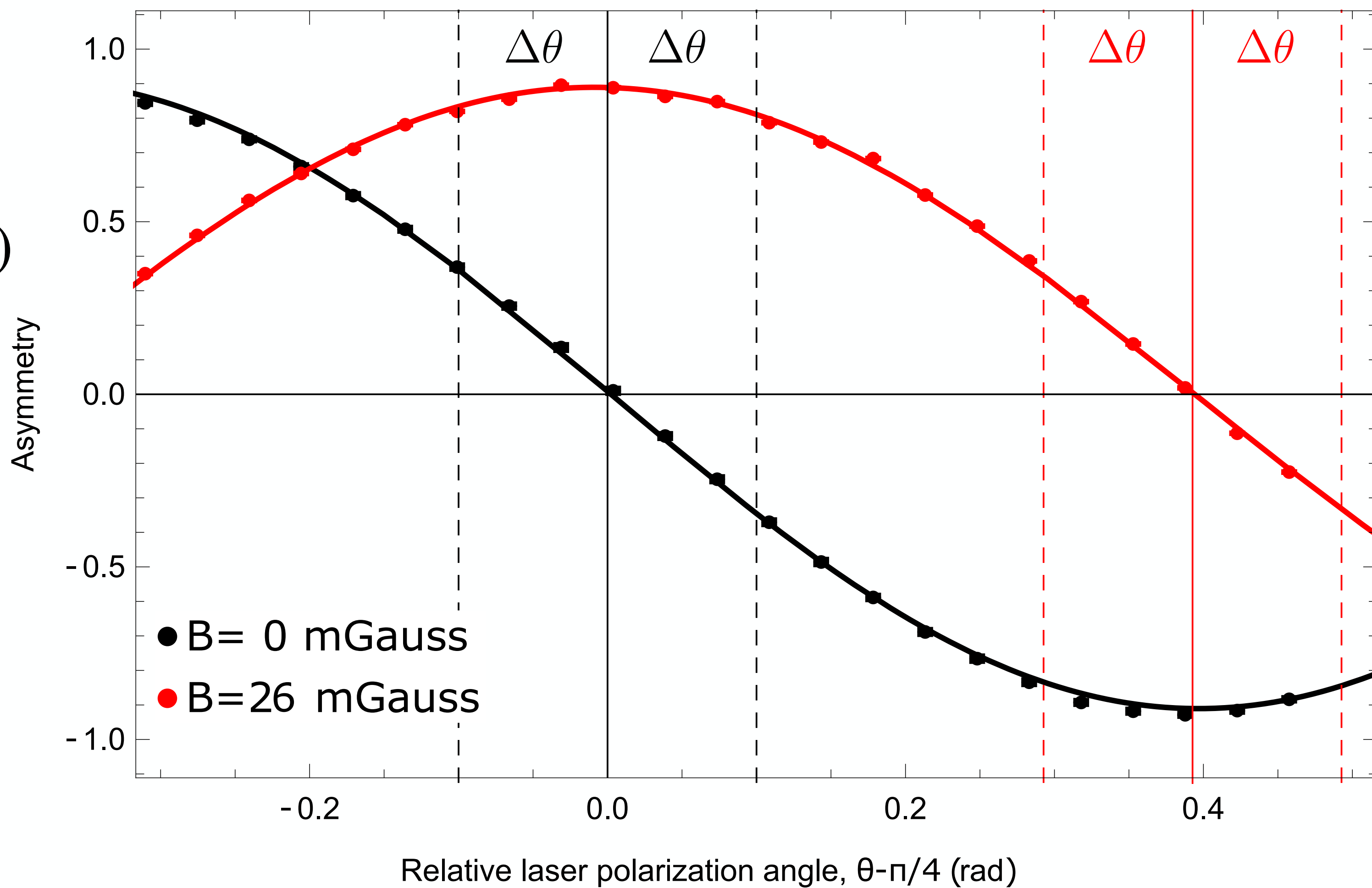
$|m = +1\rangle\rangle$



$= 2\phi/\tau$



$$A = \frac{F_x + F_y}{F_x - F_y} \propto \cos 2(\phi - \theta)$$



ACME II result

$$d_e = (4.3 \pm 3.0_{\text{stat}} \pm 2.6_{\text{syst}}) \times 10^{-30} e \cdot \text{cm}$$

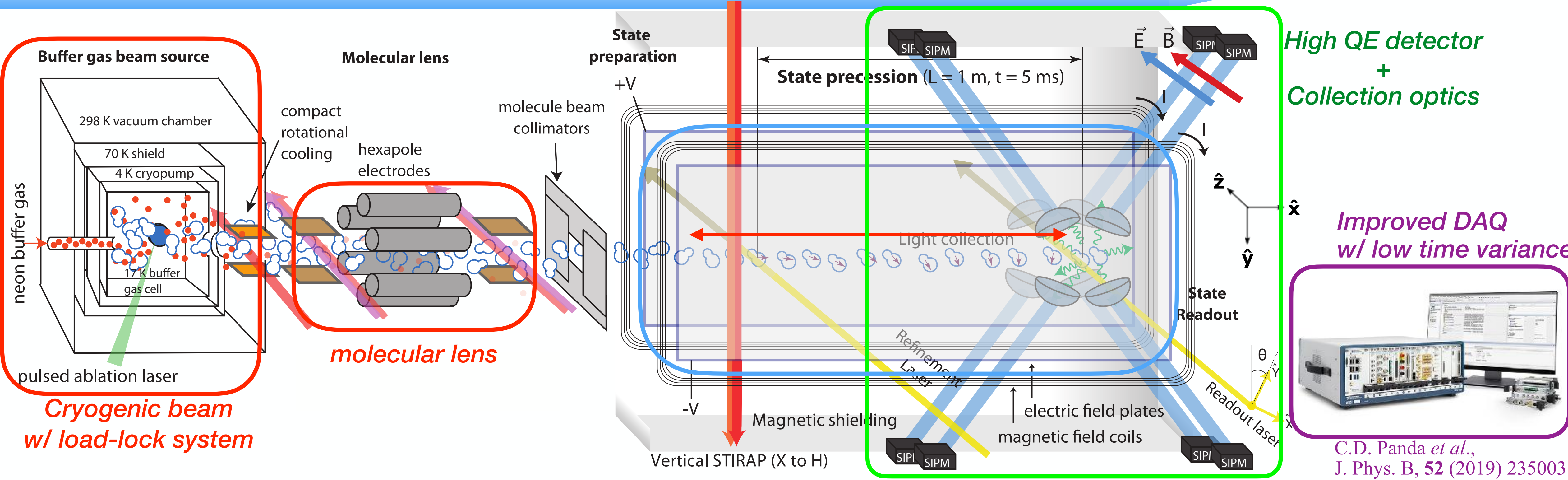
based on

- 2×10^{14} ThO molecules detected
- 6×10^6 / pulse \times 3×10^7 pulses
- 50Hz, 10 weeks
- 3×10^5 p.e./pulse (5% eff.)

Parameter	Shift	Uncertainty
$\partial \mathcal{B}_z / \partial z$ and $\partial \mathcal{B}_z / \partial y$	7	59
$\omega_{\text{ST}}^{\mathcal{N}\mathcal{E}}$ (via $\theta_{\text{ST}}^{\text{H-C}}$)	0	1
$P_{\text{ref}}^{\mathcal{N}\mathcal{E}}$	–	109
\mathcal{E}^{nr}	–56	140
$ \mathcal{C} ^{\mathcal{N}\mathcal{E}}$ and $ \mathcal{C} ^{\mathcal{N}\mathcal{E}\mathcal{B}}$	77	125
$\omega^{\mathcal{E}}$ (via $\mathcal{B}_z^{\mathcal{E}}$)	1	1
Other magnetic-field gradients (4)	–	134
Non-reversing magnetic field, $\mathcal{B}_z^{\text{nr}}$	–	106
Transverse magnetic fields, $\mathcal{B}_x^{\text{nr}}, \mathcal{B}_y^{\text{nr}}$	–	92
Refinement- and readout-laser detunings	–	76
$\tilde{\mathcal{N}}$ -correlated laser detuning, $\Delta^{\mathcal{N}}$	–	48
Total systematic	29	310
Statistical uncertainty		373
Total uncertainty		486

Values are shown in $\mu\text{rad s}^{-1}$. All uncertainties are added in quadrature. For $\mathcal{E}_{\text{eff}} = 78 \text{ GV cm}^{-1}$, $d_e = 10^{-30} e \text{ cm}$ corresponds to $|\omega^{\mathcal{N}\mathcal{E}}| = \mathcal{E}_{\text{eff}} d_e / \hbar = 119 \mu\text{rad s}^{-1}$.

Statistics improvements in ACME III



5x long precession time (length)

Statistical sensitivity:

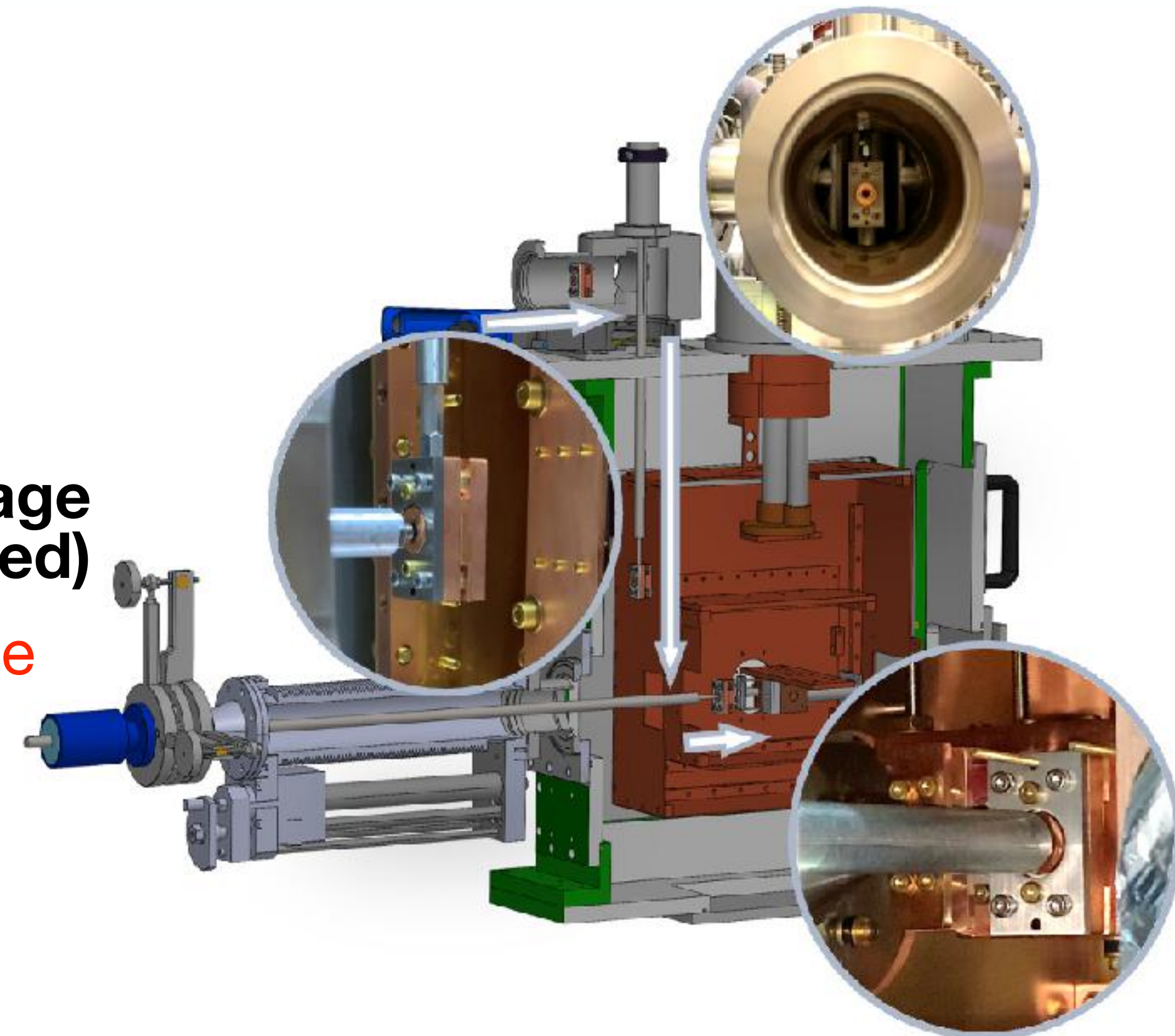
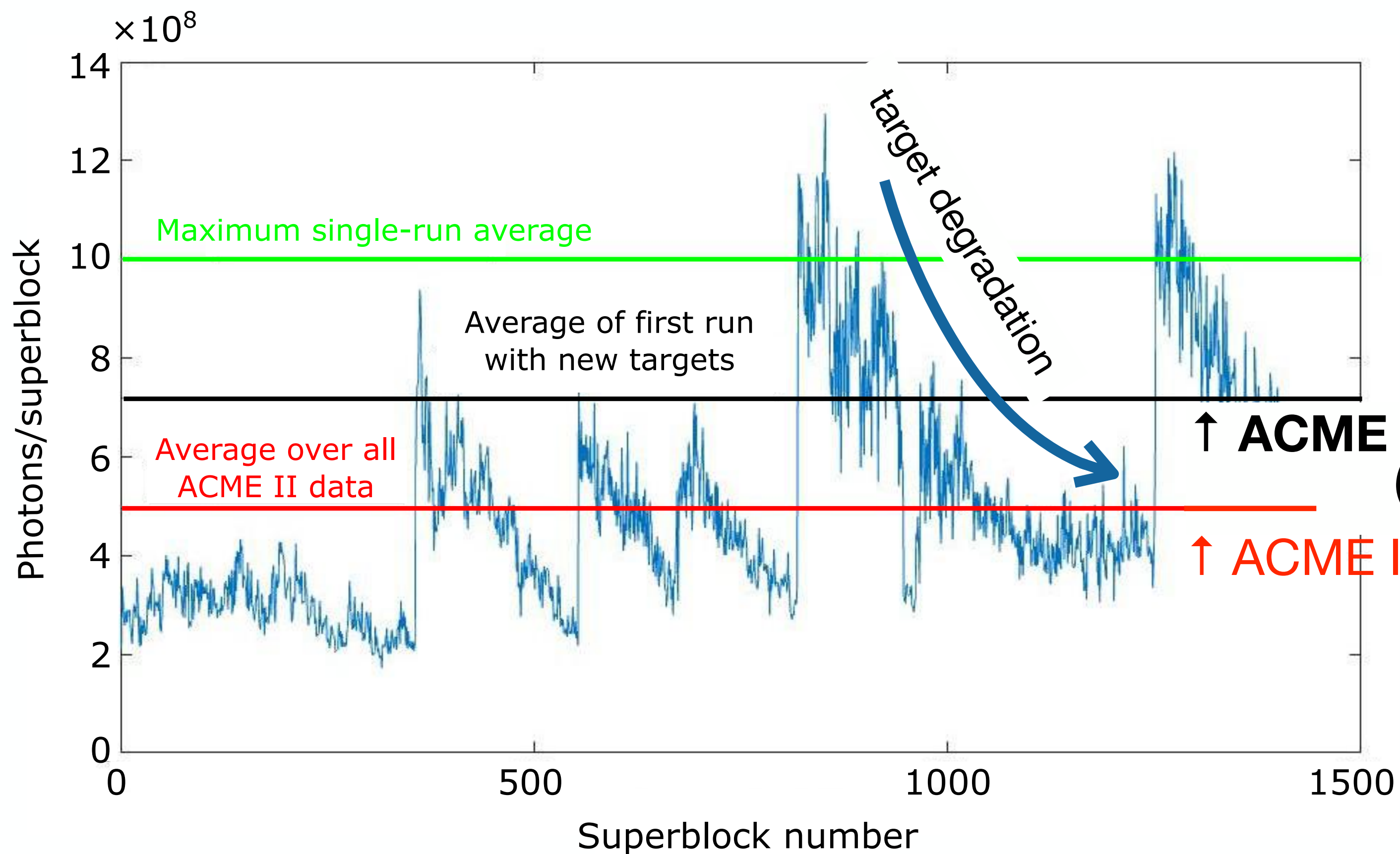
$$\Delta d_e \sim \frac{\hbar}{E_{\text{eff}} \tau} \frac{1}{\sqrt{\dot{n}_{\text{mol}} T}} \sqrt{\frac{F}{\epsilon_{\text{det}}}}$$

Improvement	Signal gain	EDM sensitivity gain
New beam source	1.5	1.2
Electrostatic lens	12	3.5
Longer precession time	0.3	2.6
Detector upgrade	2.7	1.6
Collection optics	1.7	1.3
Timing jitter reduction	1	1.7
Total	25	39

New cryogenic beam source w/ load-lock

Ablation target Load-lock system : enabling daily *in-situ* target replacement

- Ablation targets were usually replaced after ~2 weeks in ACME II.
 - Replacement takes more than one day (warm up, vacuum break, radioactive work, cool down)
- Load-lock system can increase the average statistics.



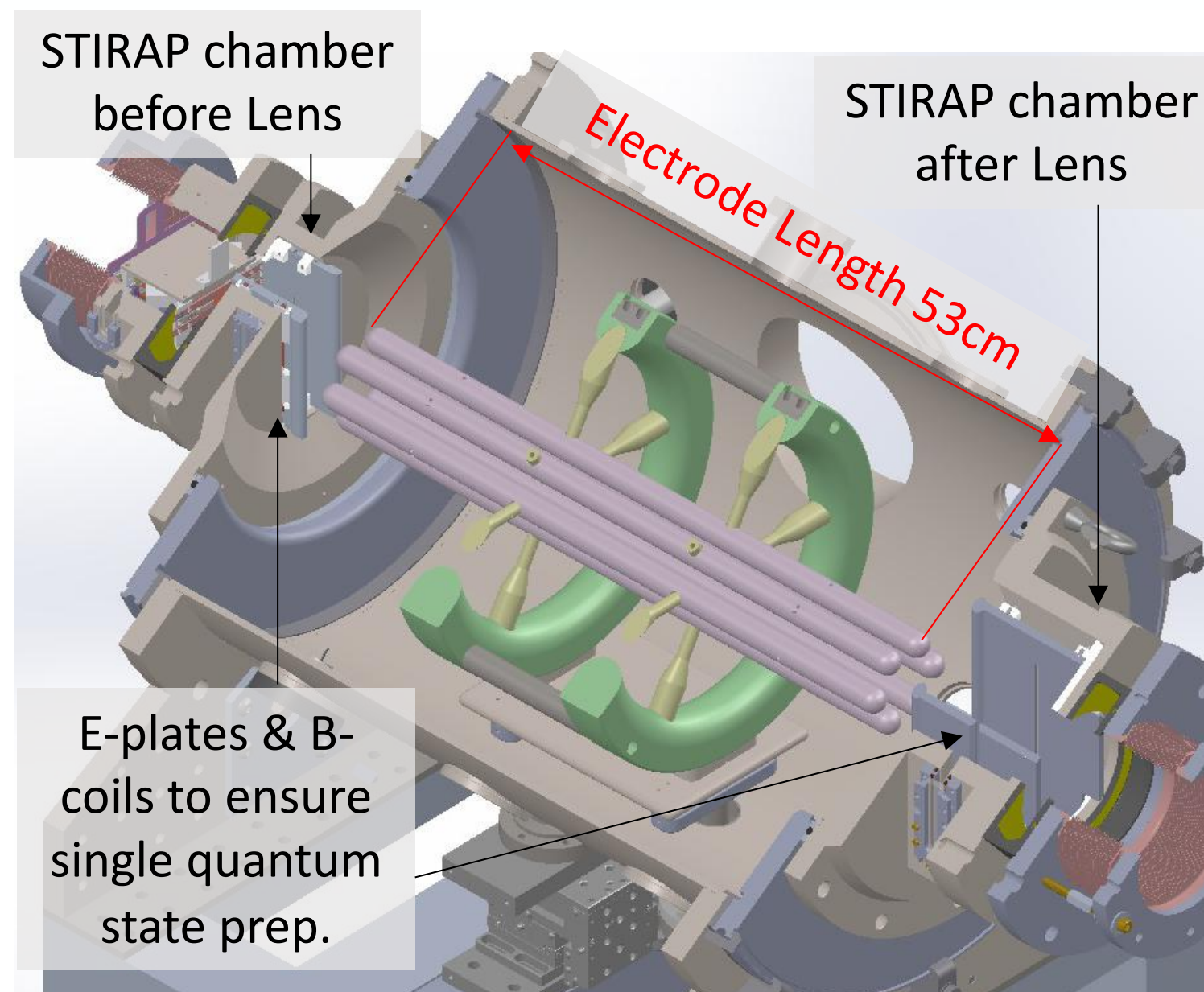
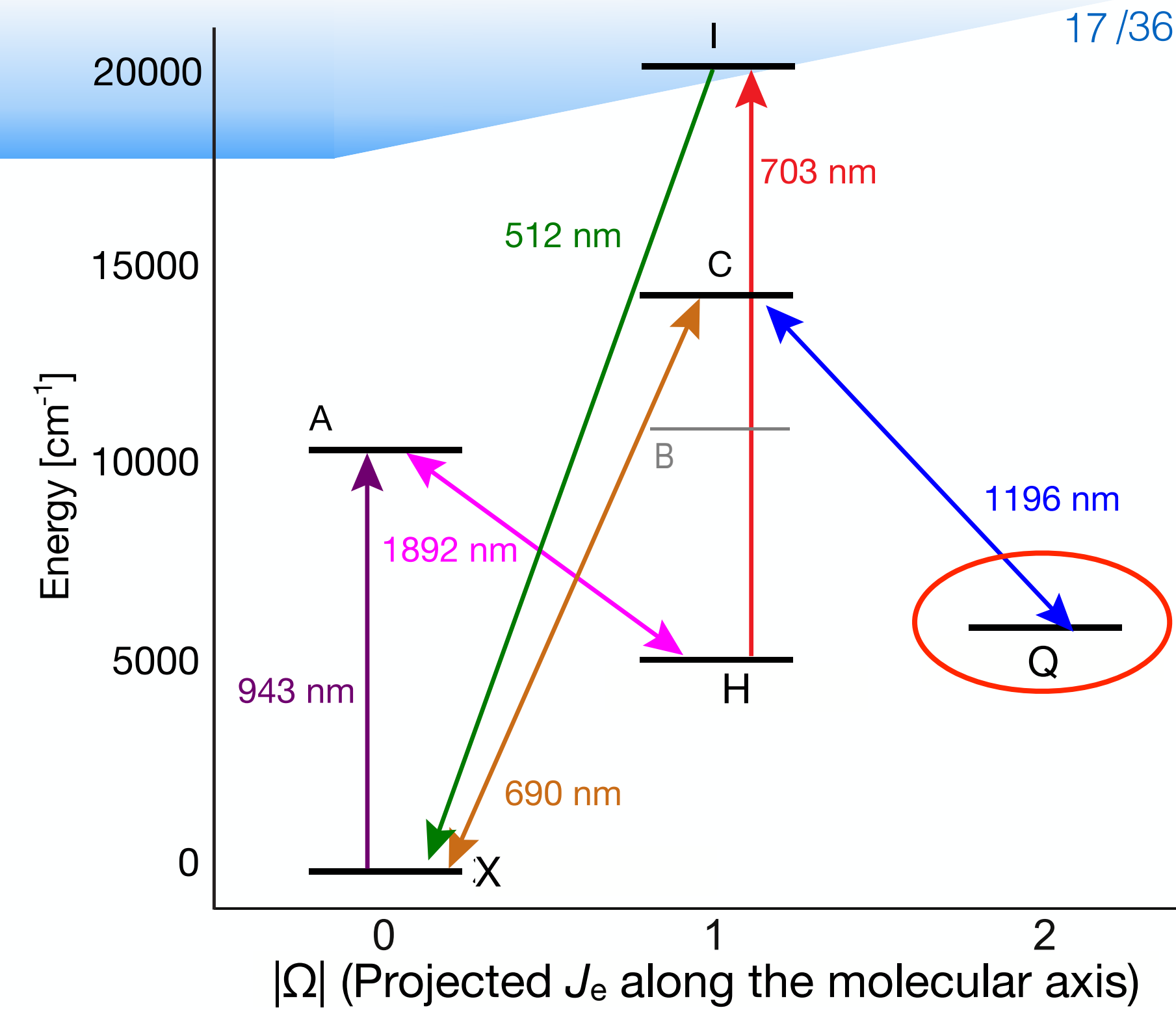
Electrostatic molecular lens

ThO beam is collimated by electrostatic potential

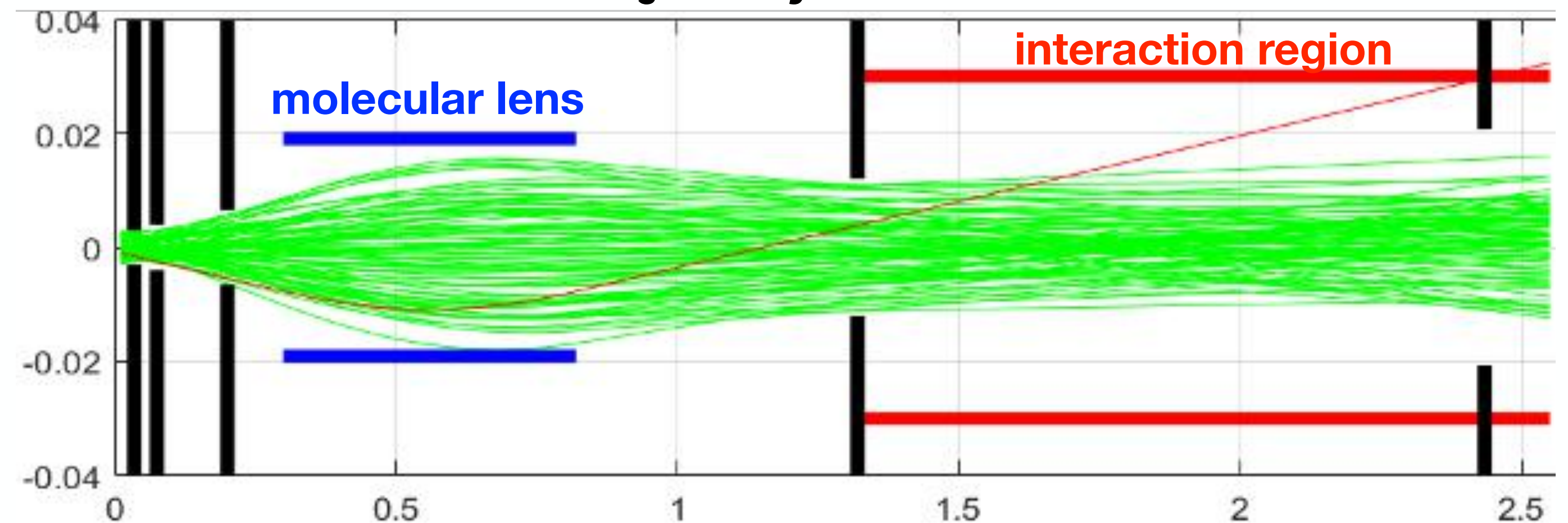
- Increasing the number of available ThO
- Reducing the loss of extended flight distance

Q state X. Wu *et al.*, New J. Phys. **22** (2020) 023013.

- Large Stark shift: $D_Q = 4.1D$ *deep electric potential is available*
- Transition strength: $d_{Q-C} = 1.0 D$ *~100% transfer $C \rightleftharpoons Q$ is feasible*
- Life time: $\tau > 62$ ms *lossless during the collimation*



Trajectory simulation

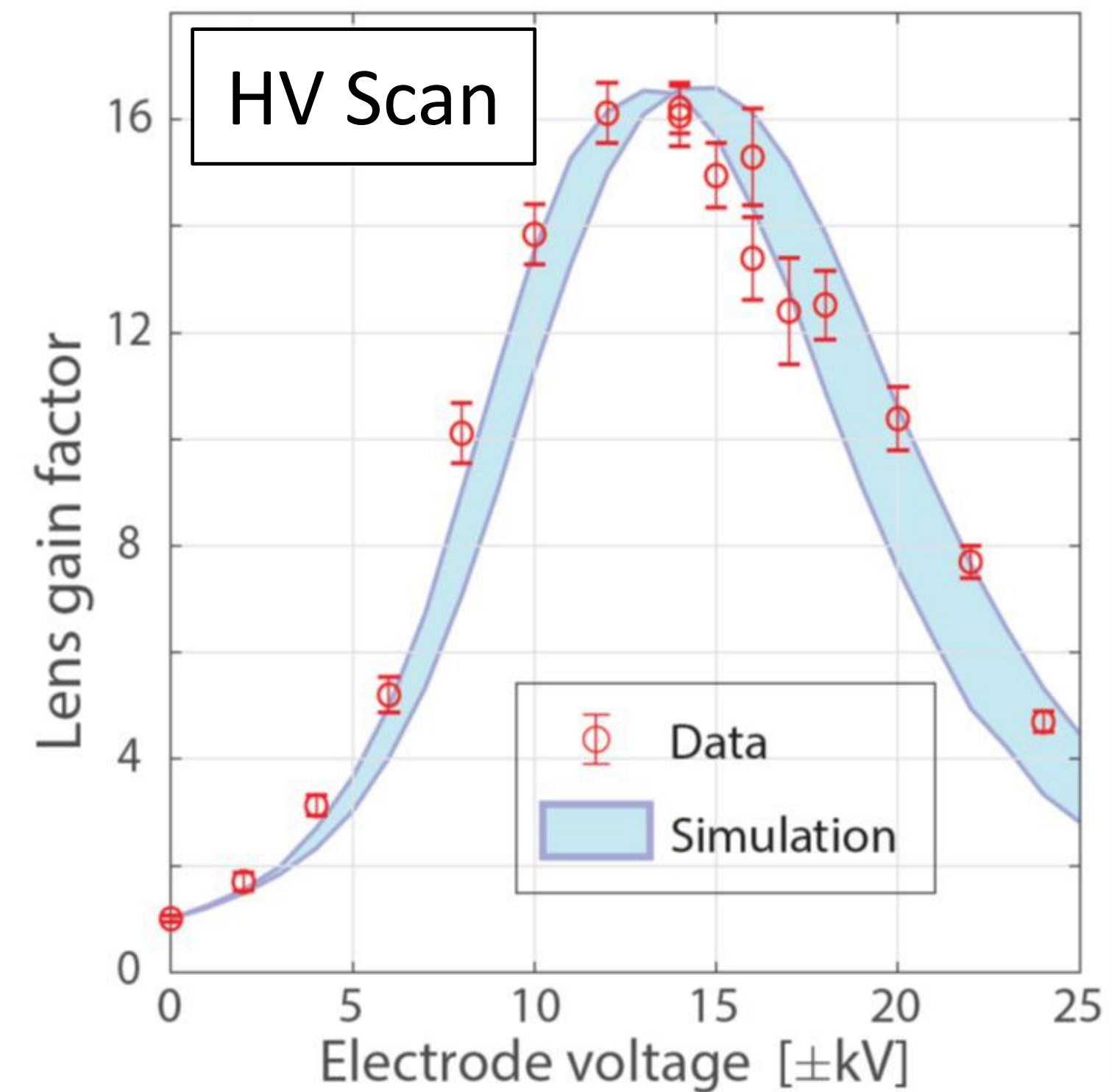
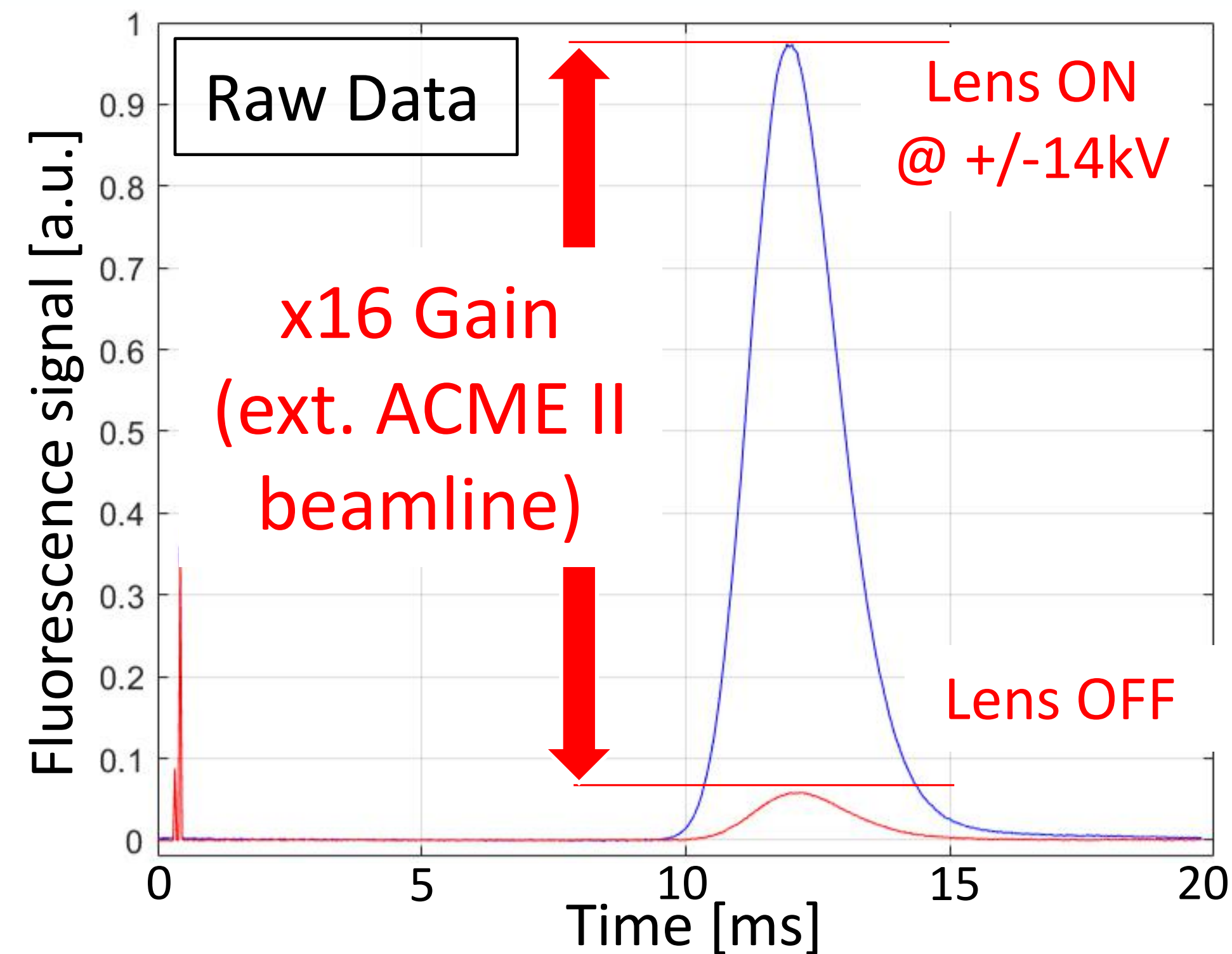


Electrostatic molecular lens

X. Wu et al., New J. Phys. **24** 073043 (2022).

×16 improvements has been demonstrated.

(consistent with expectations from the simulations)

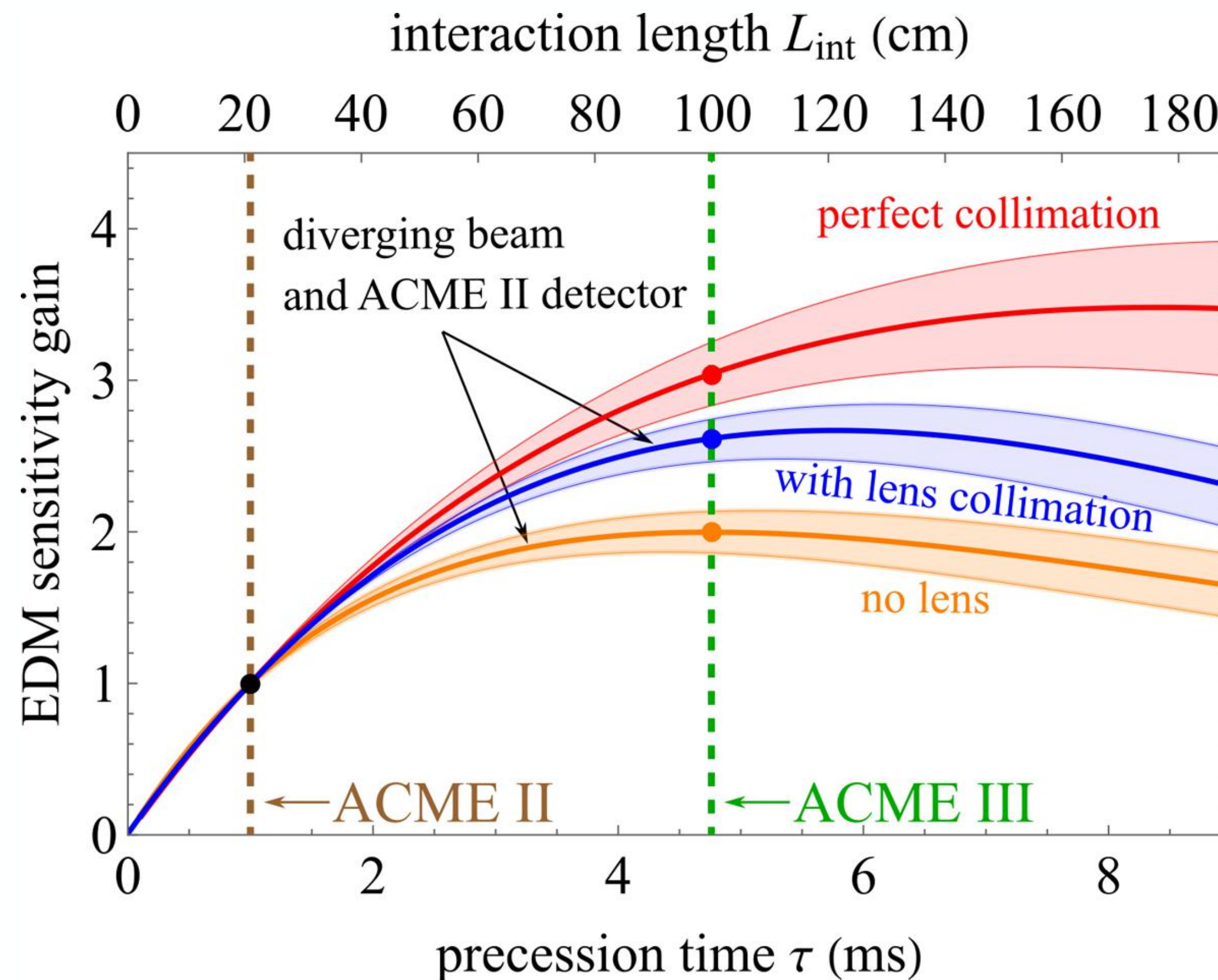


Longer precession time : 1 → 5 ms

D. Ang et al., PRA **106**, 022808 (2022).

Possible precession time is limited by the lifetime of the H state τ_H

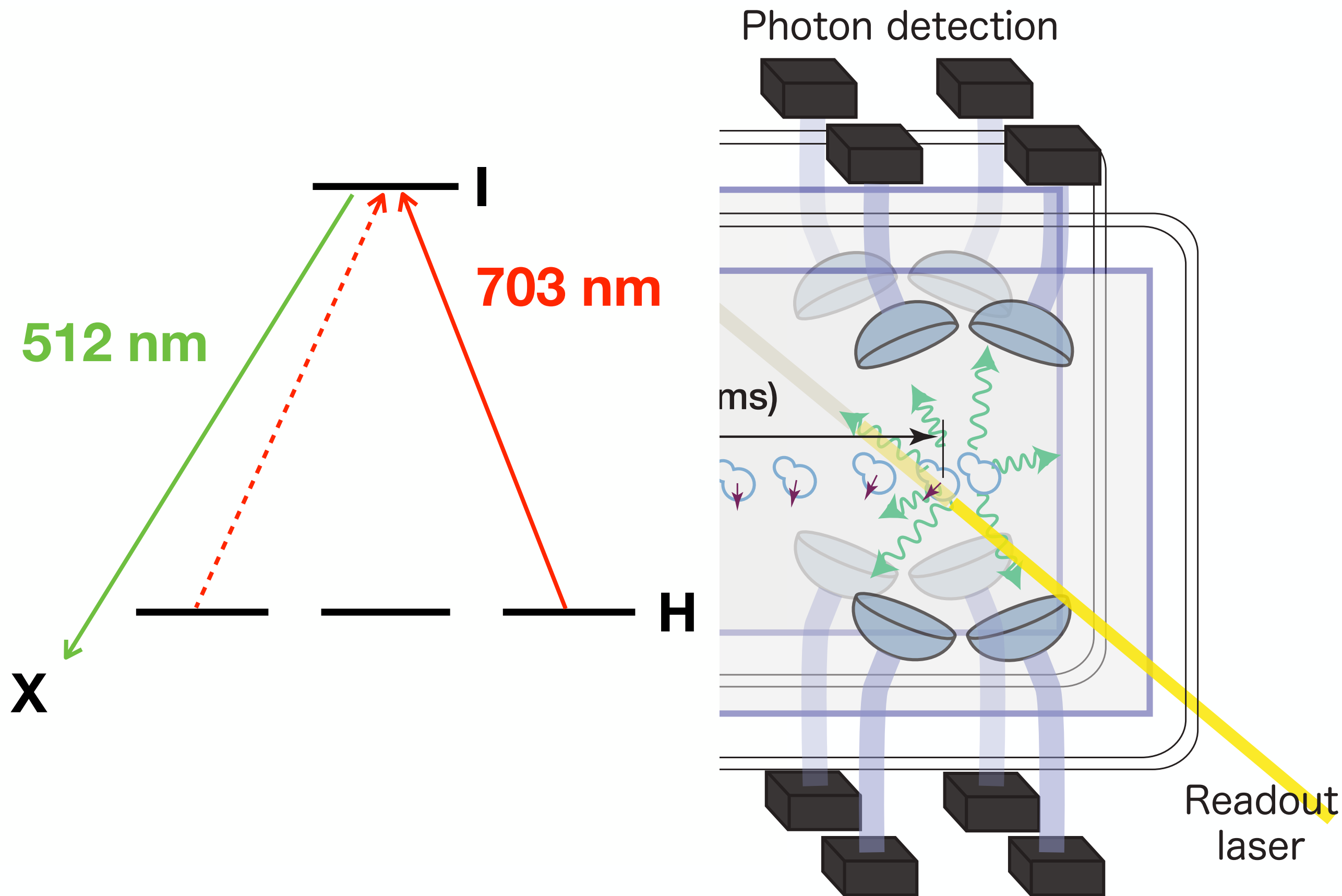
- Previous measurement : $\tau_H > 1.8$ ms → ACME II used $\tau=1$ ms
(A. Vutha et al., J. Phys. B 43 (2010) 074007.)
- Recent measurement : $\tau_H = 4.2$ ms → ACME III will use $\tau=5$ ms



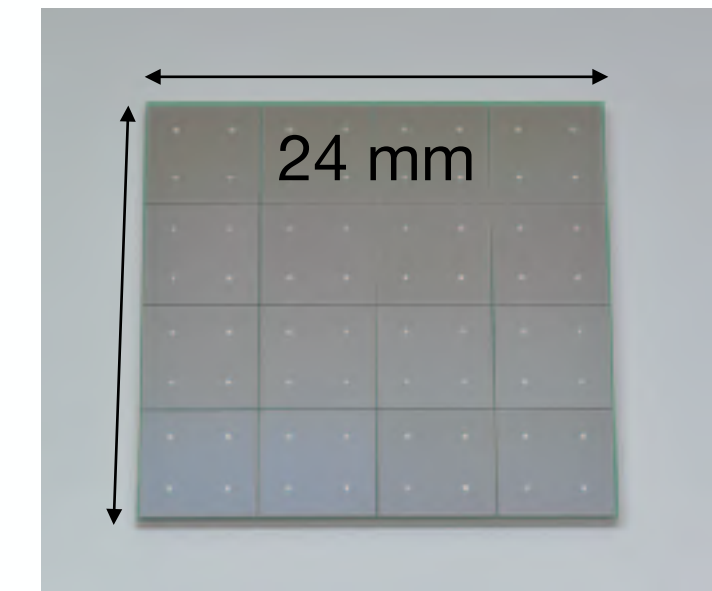
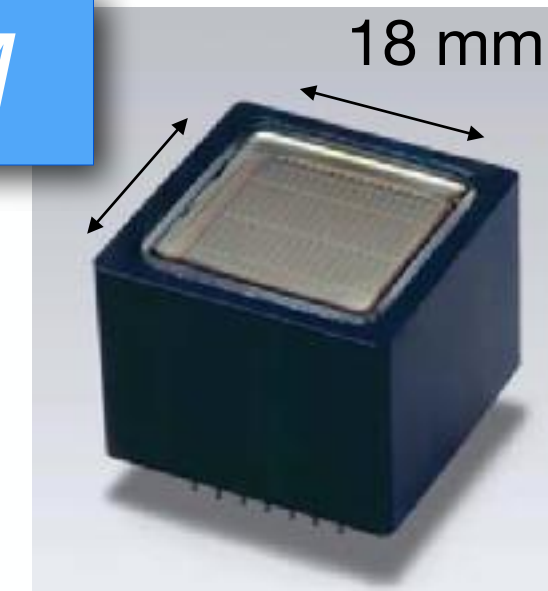
$$\Delta d_e \sim \frac{\hbar}{E_{\text{eff}} \tau} \frac{1}{\sqrt{\dot{n}_{\text{mol}} T}} \sqrt{\frac{F}{\epsilon_{\text{det}}}}$$

Photodetector upgrade : PMT → SiPM

Final state readout



PMT vs. SiPM



	ACME II PMT	Advanced ACME SiPM
Part No.	R7600U-300	S13361-6075NE-04
Sensitive area	18×18 mm ²	24×24 mm ² (16 ch.)
Q.E. @ 512 nm	~25%	~45%
Excess noise <i>F</i>	~1.2	~1.2 (depend on CT & AP)
Q.E. @ 703 nm	~0.6%	~ 20%
Dark count @ 25°C	~ 3 kcps	~ 2 Mcps/ch
Capacitance	few pF	1.4 nF

} pros

} cons

Changing from PMTs to SiPMs will increase the PDE by a factor of ~2 and sensitive area as well.

SiPM module

T. Masuda *et al.*, Opt. Express **29** 16914 (2021).

Suppress DCR

Cooling (-20°C), Vacuum ($<10\text{ Pa}$)

Suppress Optical Crosstalk and Stray light

Optimized triple filtering scheme

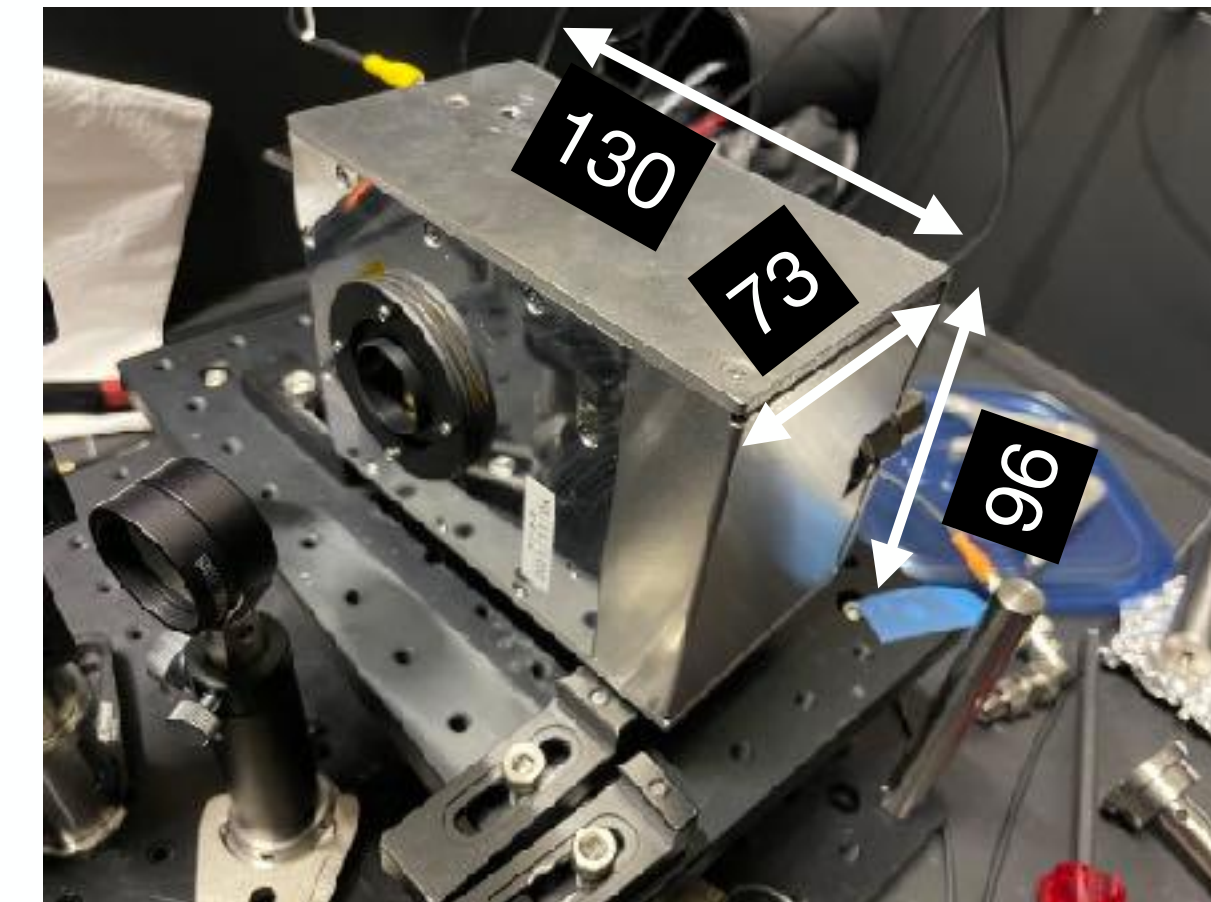
Dedicated electronics

Molecular pulse duration $\sim 4\text{ ms}$

Readout laser pulse duration $\sim 1\text{ }\mu\text{s}$

I state lifetime 115 ns

16ch analog summing



Cut-out view

BPF (Semrock FF01-520/70)

BPF, window
(Schott BG39)

BPF, OCT suppressor
(Schott BG40)

Aluminum PCB

TEC element

Aluminum vacuum chamber

Water-cooled heatsink

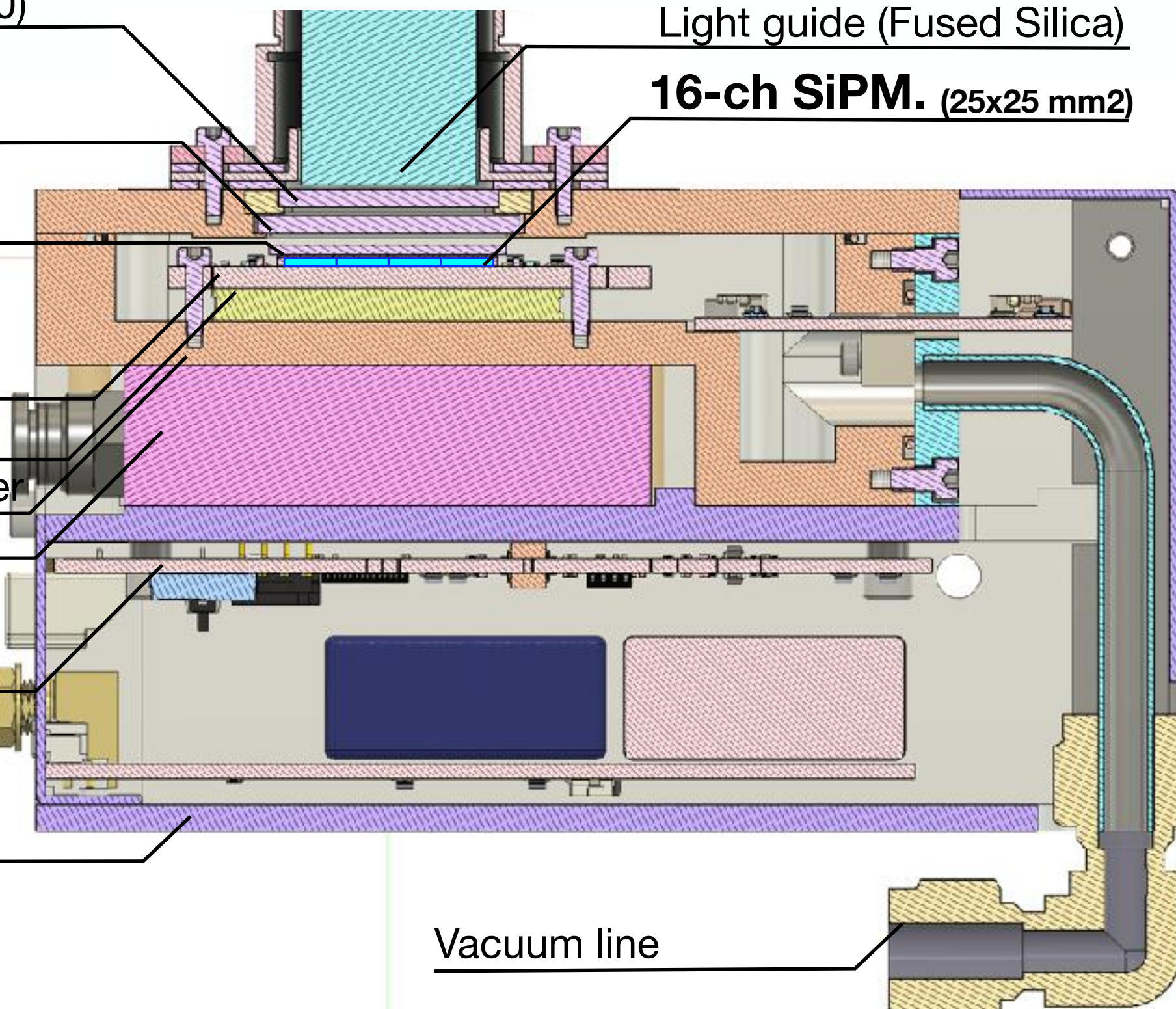
Preamplifier PCB

Electronics box

Light guide (Fused Silica)

16-ch SiPM. (25x25 mm²)

Vacuum line

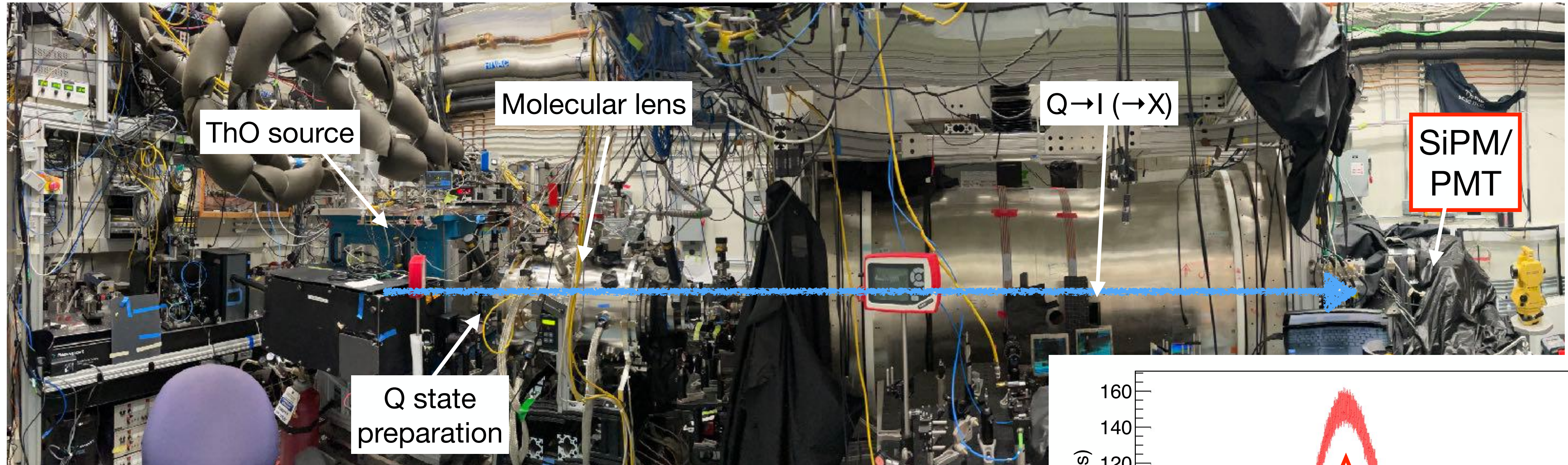


Beam test (2021 August)

A. Hiramoto et al., Nucl. Instrum. Meth. A, **1045** (2023) 167513.

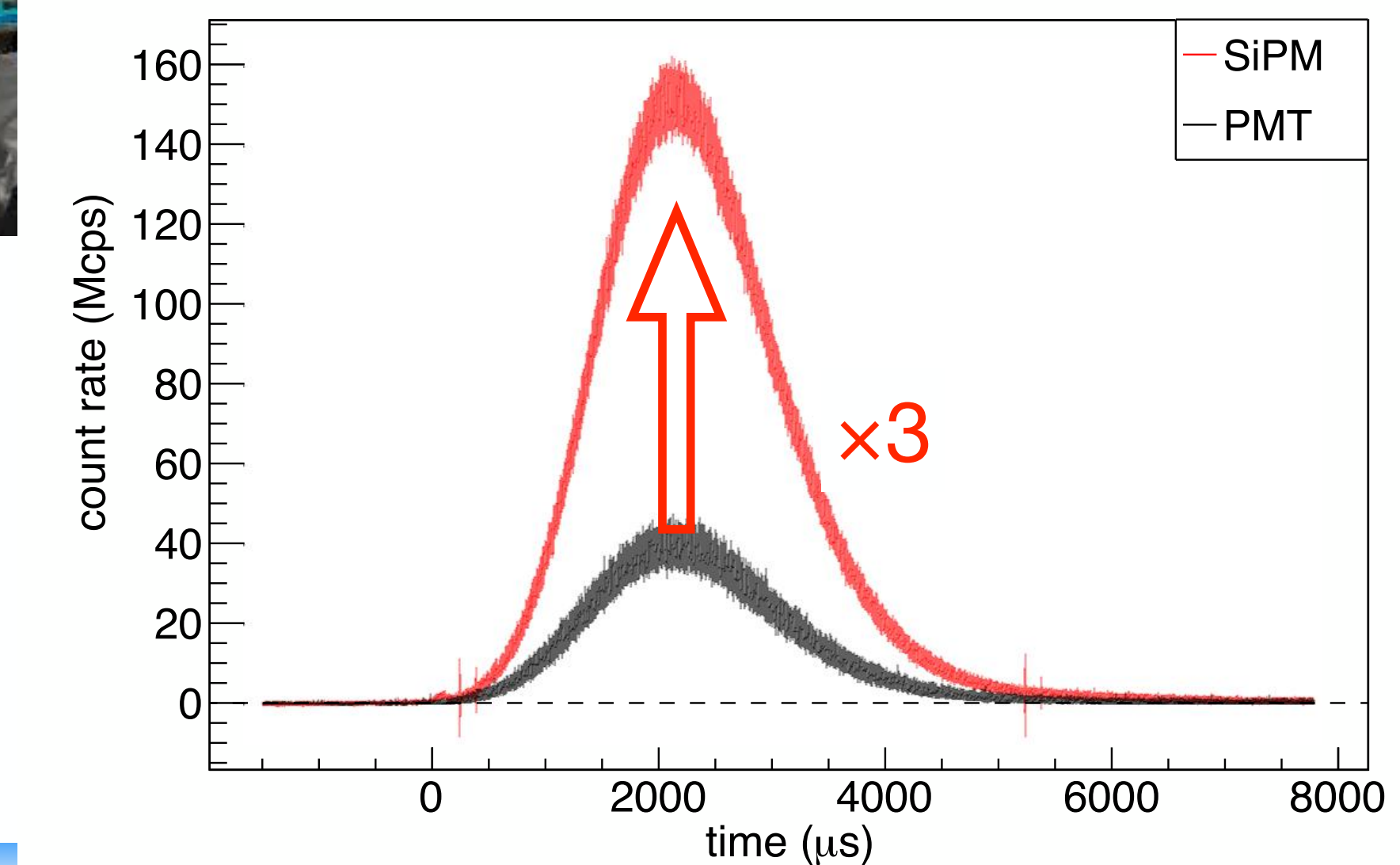
T. Masuda et al., Opt. Express **31** 1943 (2023).

ThO fluorescence detection test @ ACME II beam line



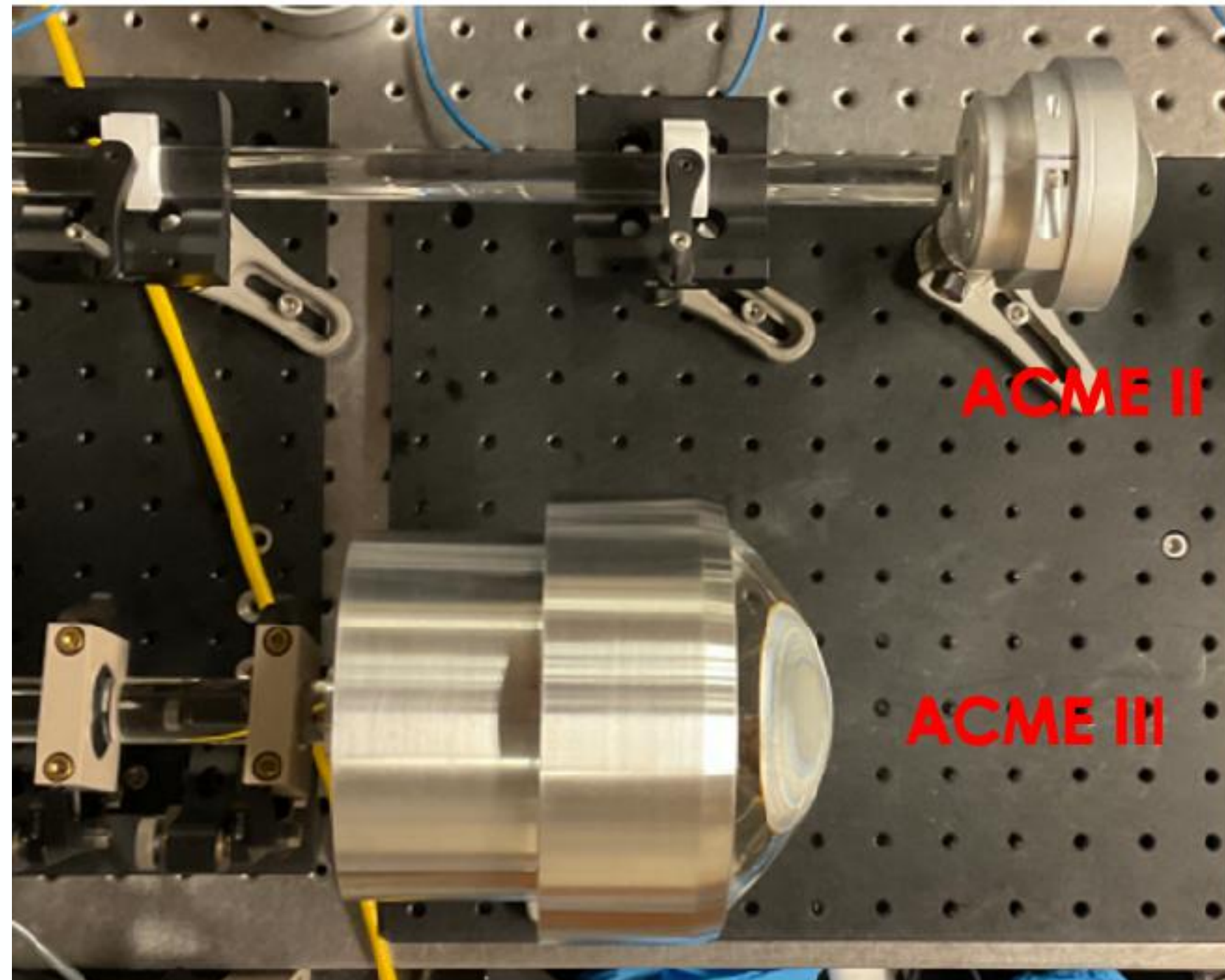
Aug. 27-30, 2021

First signal from ThO with the SiPM module!



Collection optics

Bigger lens increases the collection efficiency: 5% \rightarrow 8%



optics	detector	detection rate (Mcps)
ACME2	PMT	8.55
ACME2	SiPM	23.4
ACME3	SiPM	39.3

$\times 2.74$: detector effect

$\times 1.68$: optics effect

Summary of the statistics improvements

Improvement	Signal gain	EDM sensitivity gain	Reference
New beam source	1.5	1.2	
Electrostatic lens	12	3.5	X. Wu <i>et al.</i> , New J. Phys. 22 , 023013 (2020) X. Wu <i>et al.</i> , New J. Phys. 24 , 073043 (2022)
Longer precession time	0.3	2.6	D.G. Ang <i>et al.</i> , Phys. Rev. A 106 , 022808 (2022)
Detector upgrade	2.7	1.6	T. Masuda <i>et al.</i> , Opt. Exp. 29 , 16914 (2021) T. Masuda, A. Hiramoto <i>et al.</i> , Opt. Exp. 31 , 1943 (2023)
Collection optics	1.7	1.3	
Timing jitter reduction	1	1.7	C.D. Panda <i>et al.</i> , J. Phys. B 52 , 235033 (2019)
Total	25	39	

$$\Delta d_e \sim \frac{\hbar}{E_{\text{eff}} \tau} \frac{1}{\sqrt{\dot{n}_{\text{mol}} T}} \sqrt{\frac{F}{\epsilon_{\text{det}}}}$$

ACME III anticipated statistical sensitivity

$$\delta d_e \sim 3 \times 10^{-31} \text{ e} \cdot \text{cm} / \sqrt{\text{day}}$$

Systematics improvements in ACME III

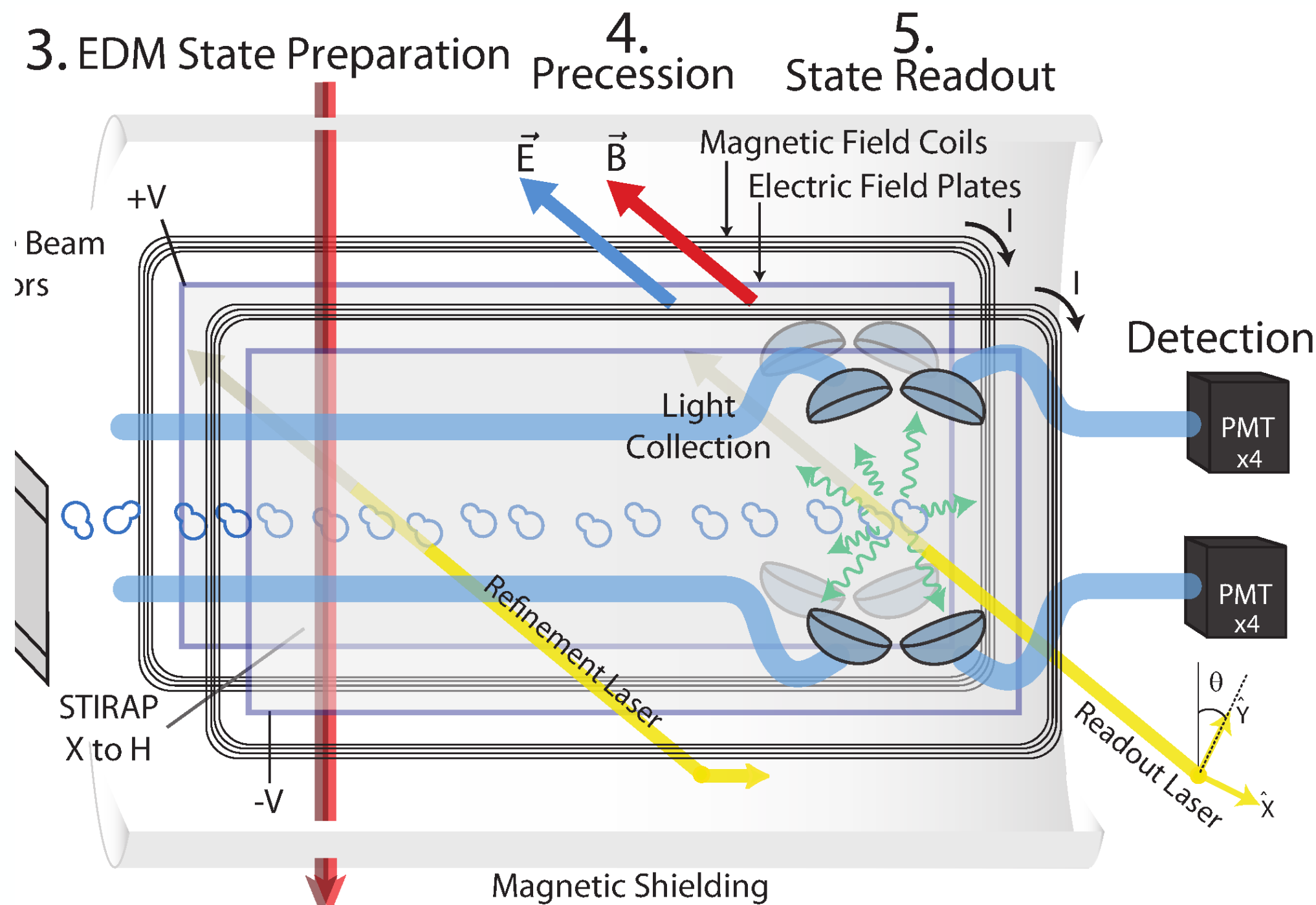
Parameter	Shift	Uncertainty
$\partial B_z/\partial z$ and $\partial B_z/\partial y$	7	59
ω_{ST}^{NE} (via θ_{ST}^{H-C})	0	1
P_{ref}^{NE}	–	109
\mathcal{E}^{nr}	–56	140
$ C ^{NE}$ and $ C ^{NEB}$	77	125
$\omega^{\mathcal{E}}$ (via $B_z^{\mathcal{E}}$)	1	1
Other magnetic-field gradients (4)	–	134
Non-reversing magnetic field, B_z^{nr}	–	106
Transverse magnetic fields, B_x^{nr}, B_y^{nr}	–	92
Refinement- and readout-laser detunings	–	76
$\tilde{\mathcal{N}}$ -correlated laser detuning, $\Delta^{\mathcal{N}}$	–	48
Total systematic	29	310
Statistical uncertainty		373
Total uncertainty		486

Imperfection of laser polarization

Imperfection of the magnetic field

Values are shown in $\mu\text{rad s}^{-1}$. All uncertainties are added in quadrature. For $\mathcal{E}_{\text{eff}} = 78 \text{ GV cm}^{-1}$, $d_e = 10^{-30} \text{ e cm}$ corresponds to $|\omega^{NE}| = \mathcal{E}_{\text{eff}} d_e / \hbar = 119 \mu\text{rad s}^{-1}$.

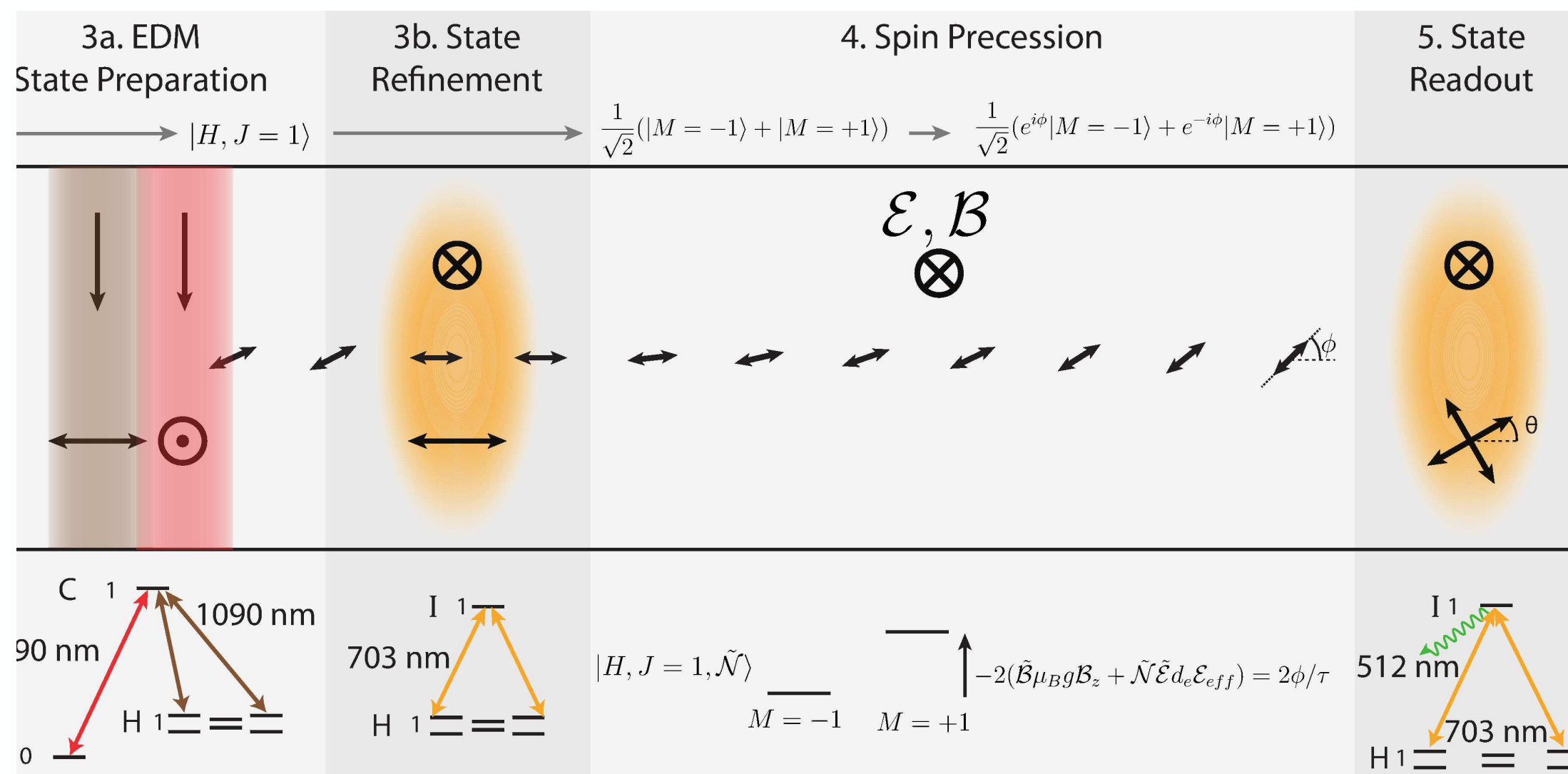
Ellipticity gradients in the laser polarization



*State preparation and readout are depending on the **laser polarization**.*

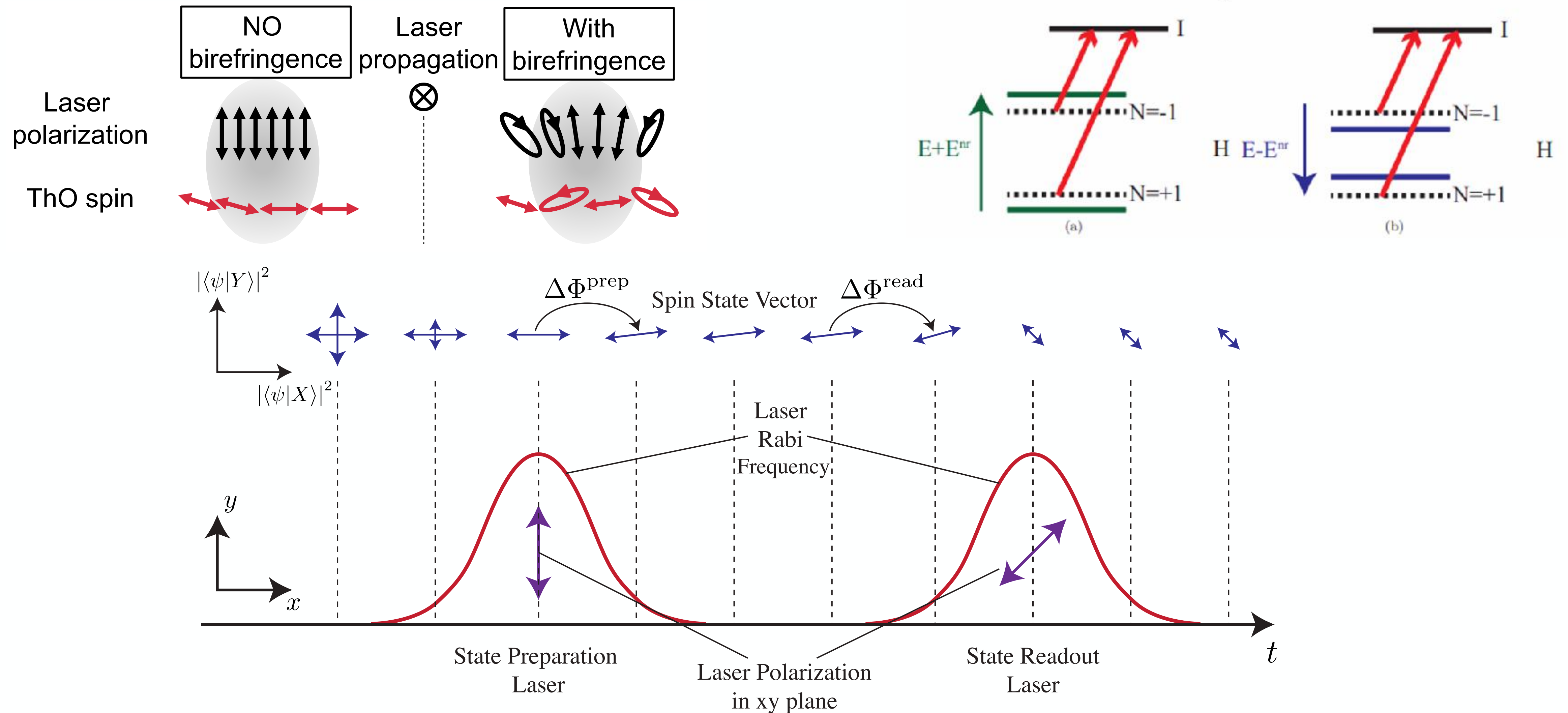


If the laser polarization is not perfect, the spin precession could be disturbed.



Cause of systematics

False EDM : Ellipticity gradient (laser imperfection)
+ AC-Stark shift gradient (E-field imperfection)



Ellipticity gradient due to glass

*Laser polarization is varying
due to birefringence gradient of the optical components along the laser pass.*

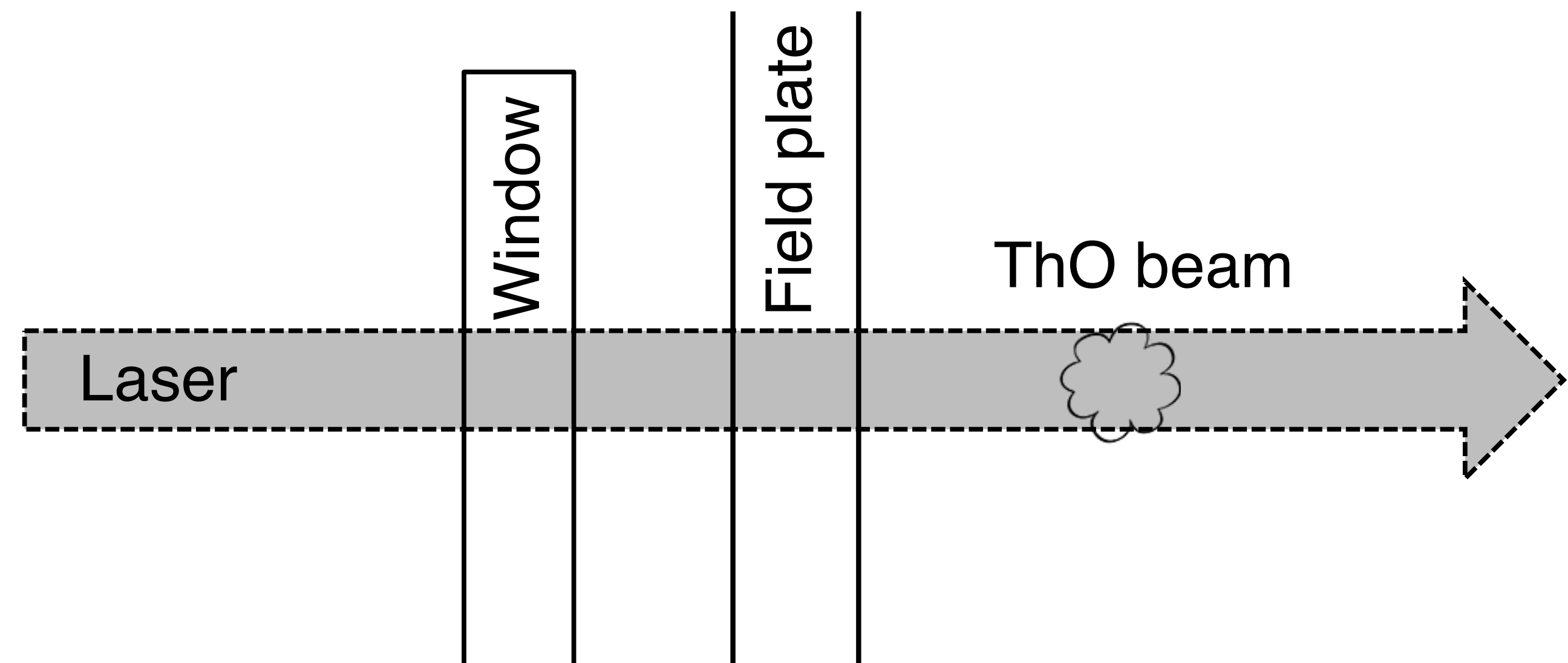
Birefringence Gradients in *Vacuum window* & *Field plate*:

ACME II. 0.1%/mm

ACME III. 0.01%/mm

mainly caused by stress

wikipedia

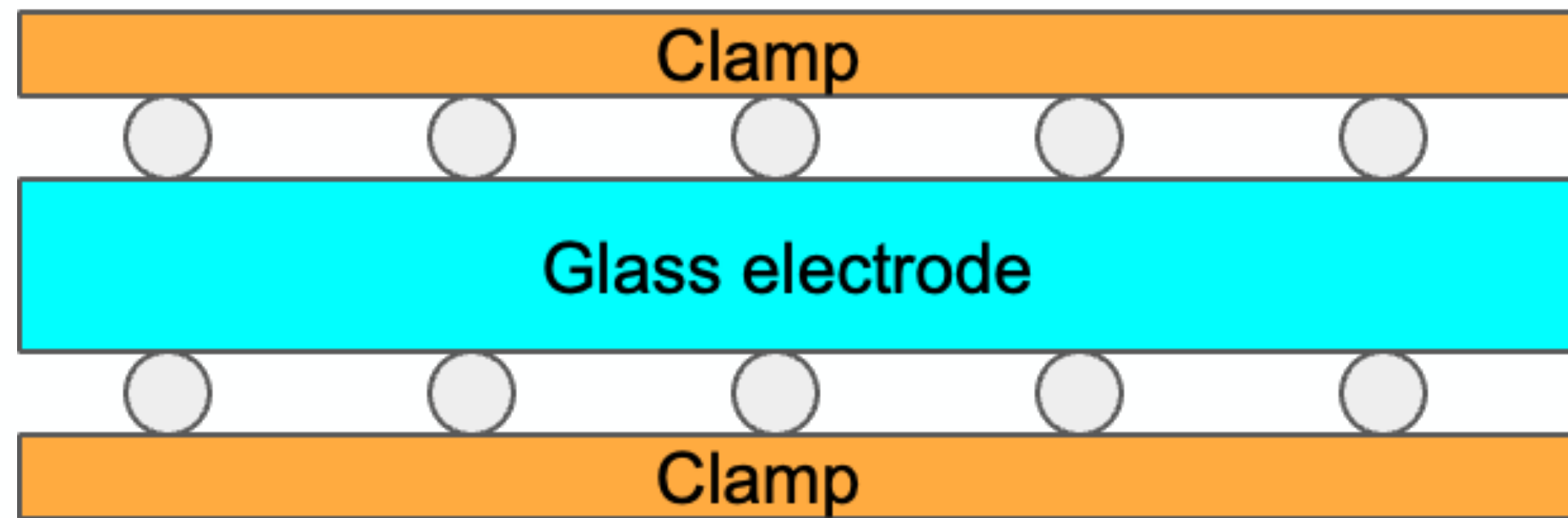
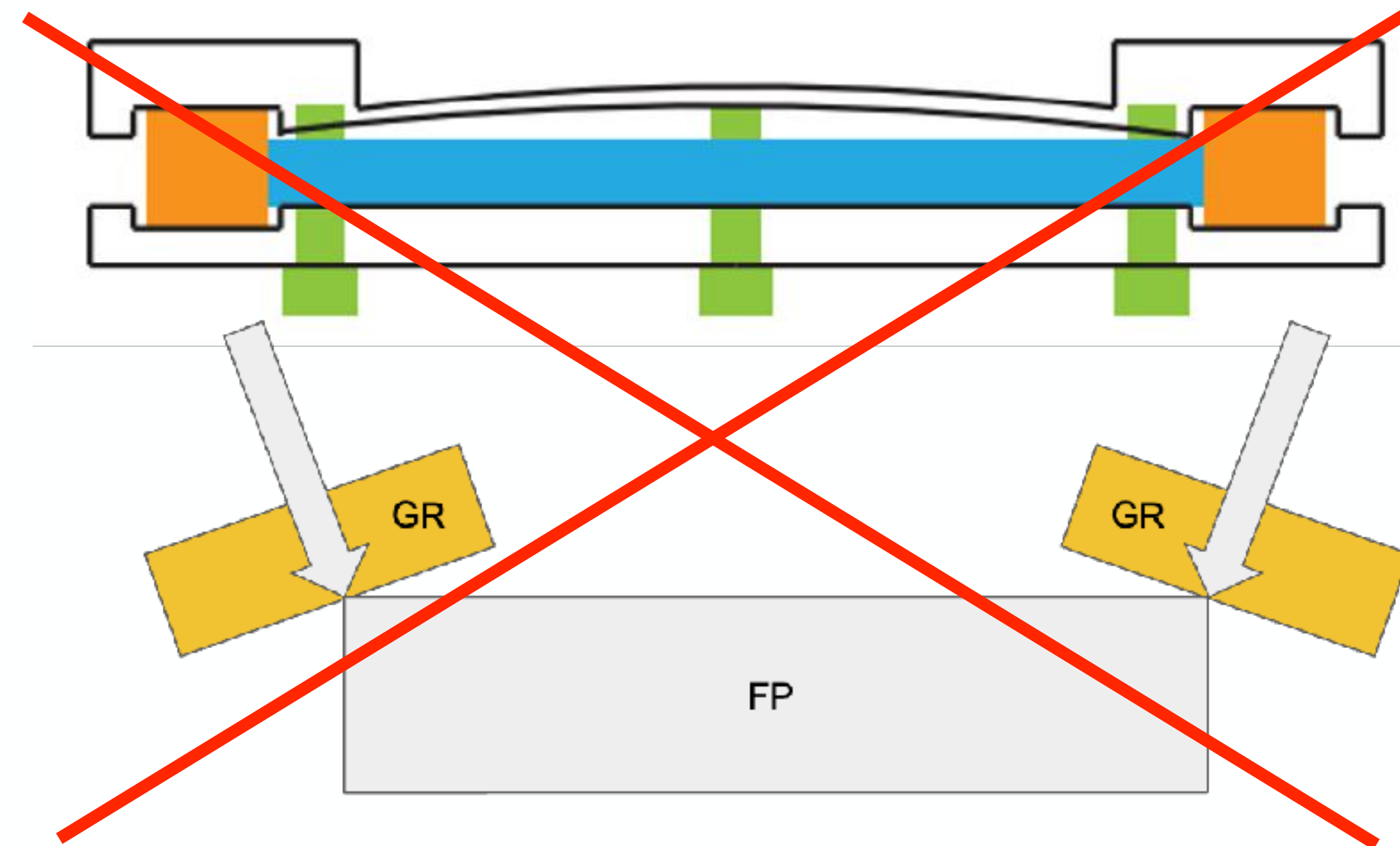


Field plate

Birefringence gradients in ACME II field plates were dominated by tangential **clamping stress**.



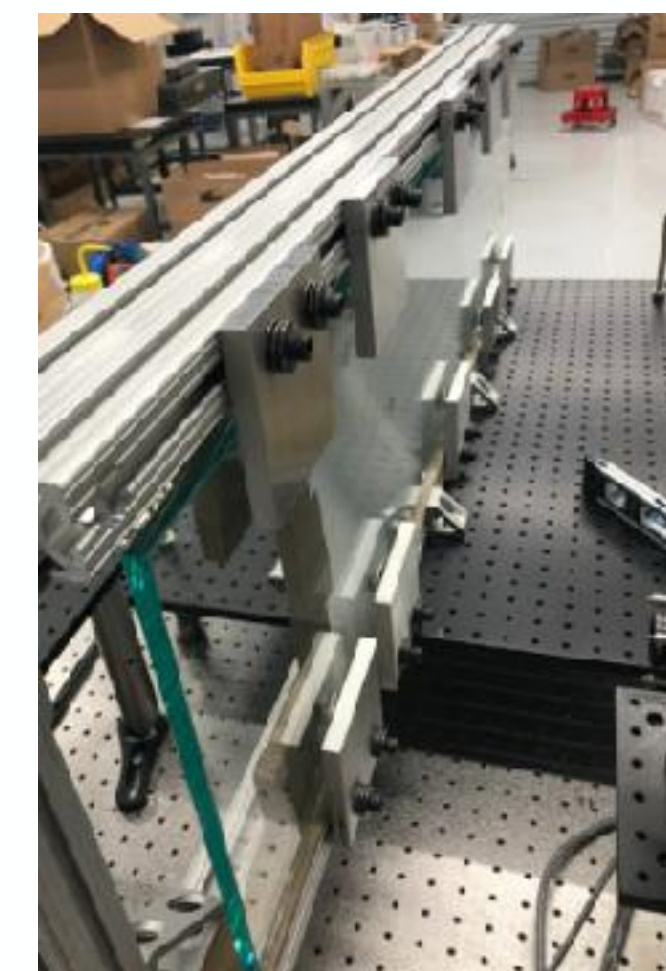
Stress-free mounting has been designed.



Birefringence free mounting scheme



Proof of principle clamping modification on ACME II



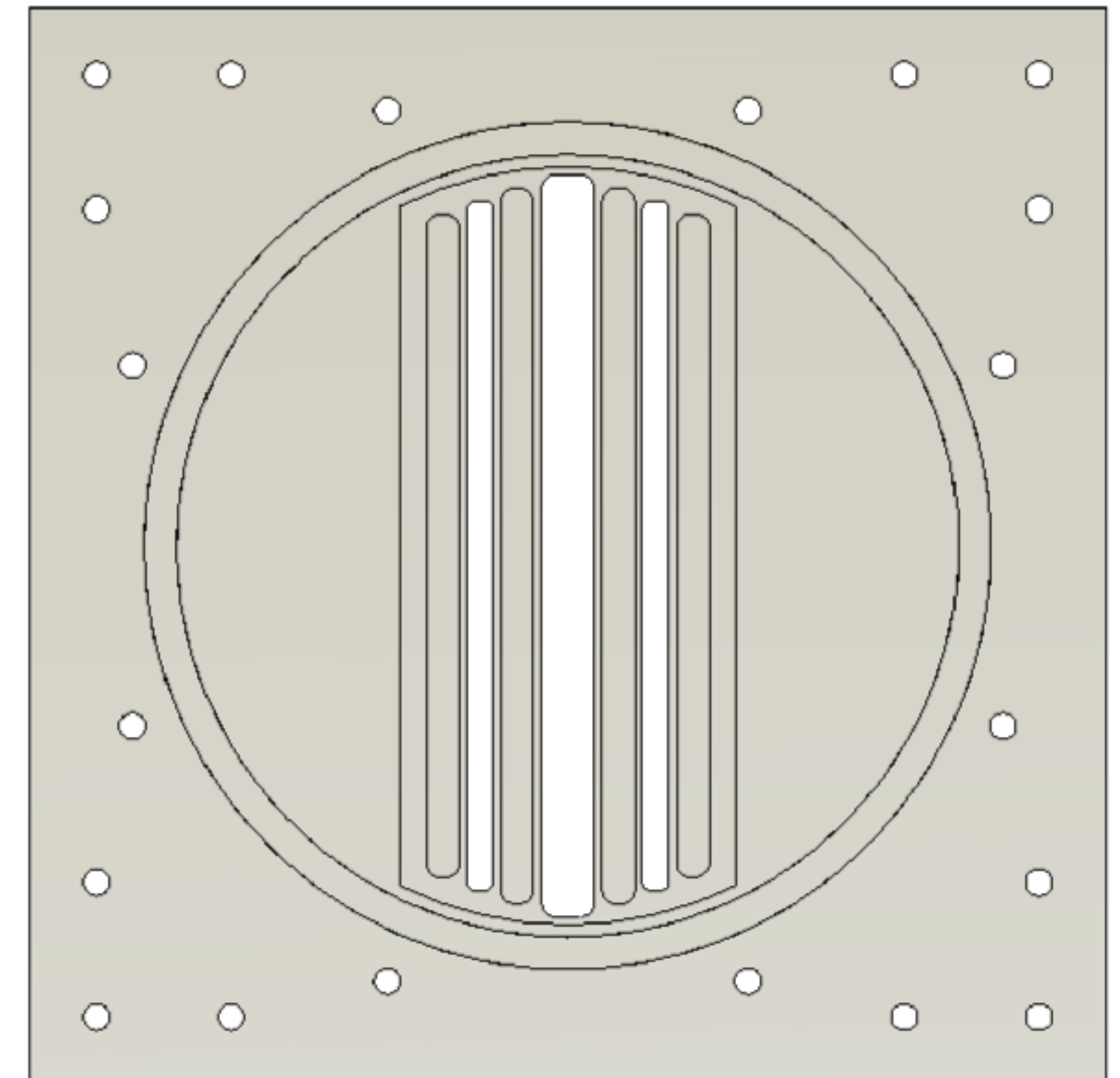
clamping test

Vacuum window

Vacuum window is suffered huge stress due to atmospheric pressure.

→ Use ***ultra-low stress-optic glass*** as window material
& ***Thin slit*** vacuum opening to minimize vacuum stress

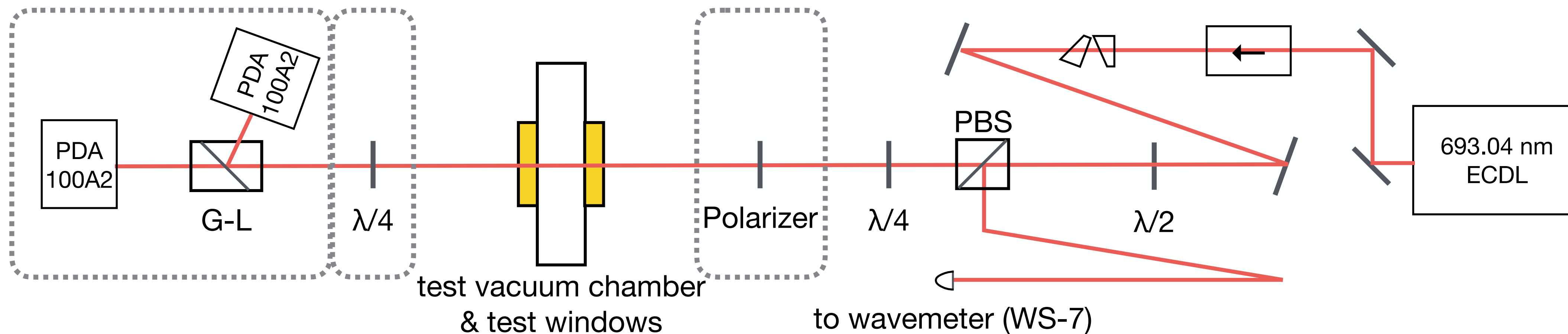
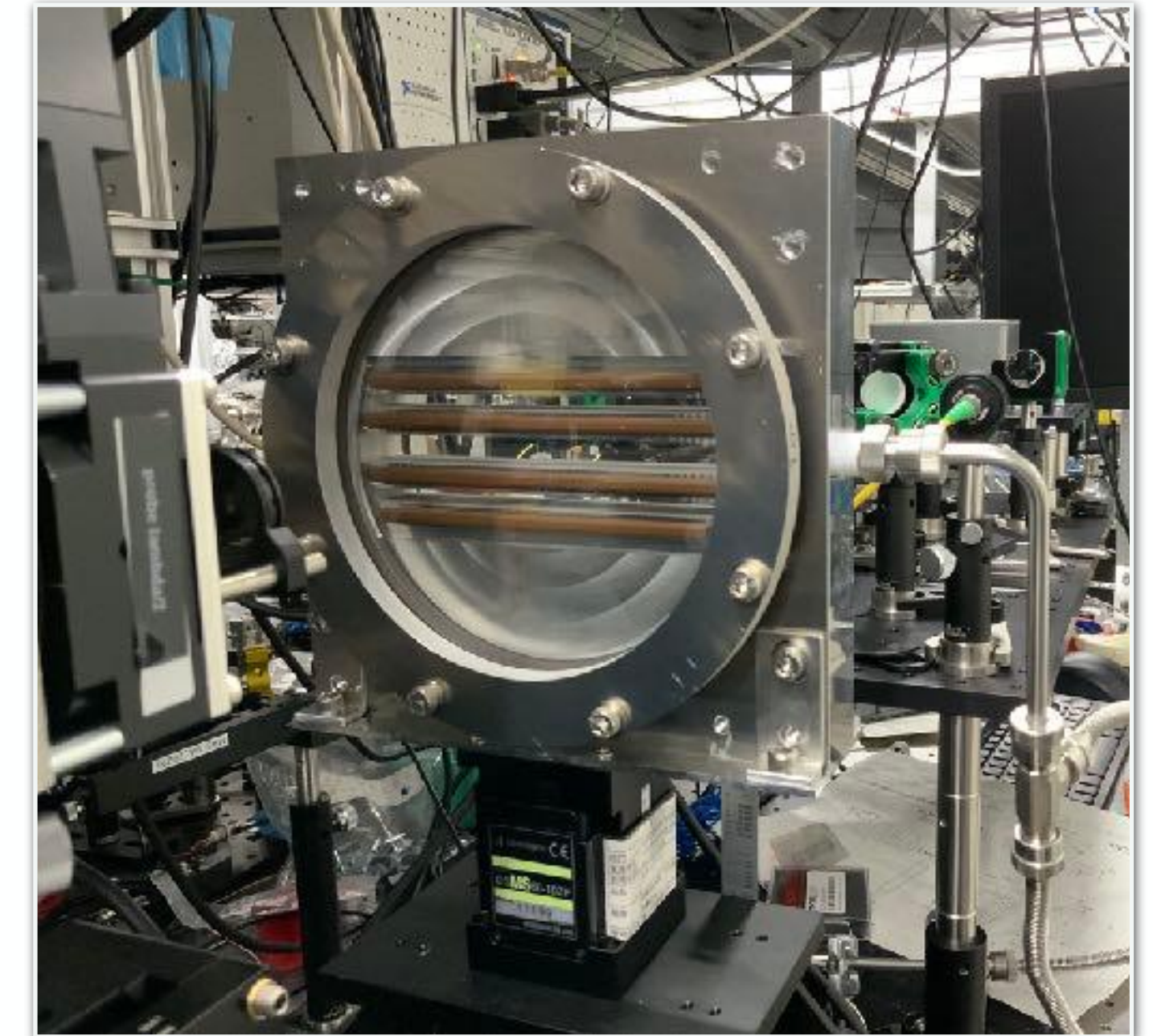
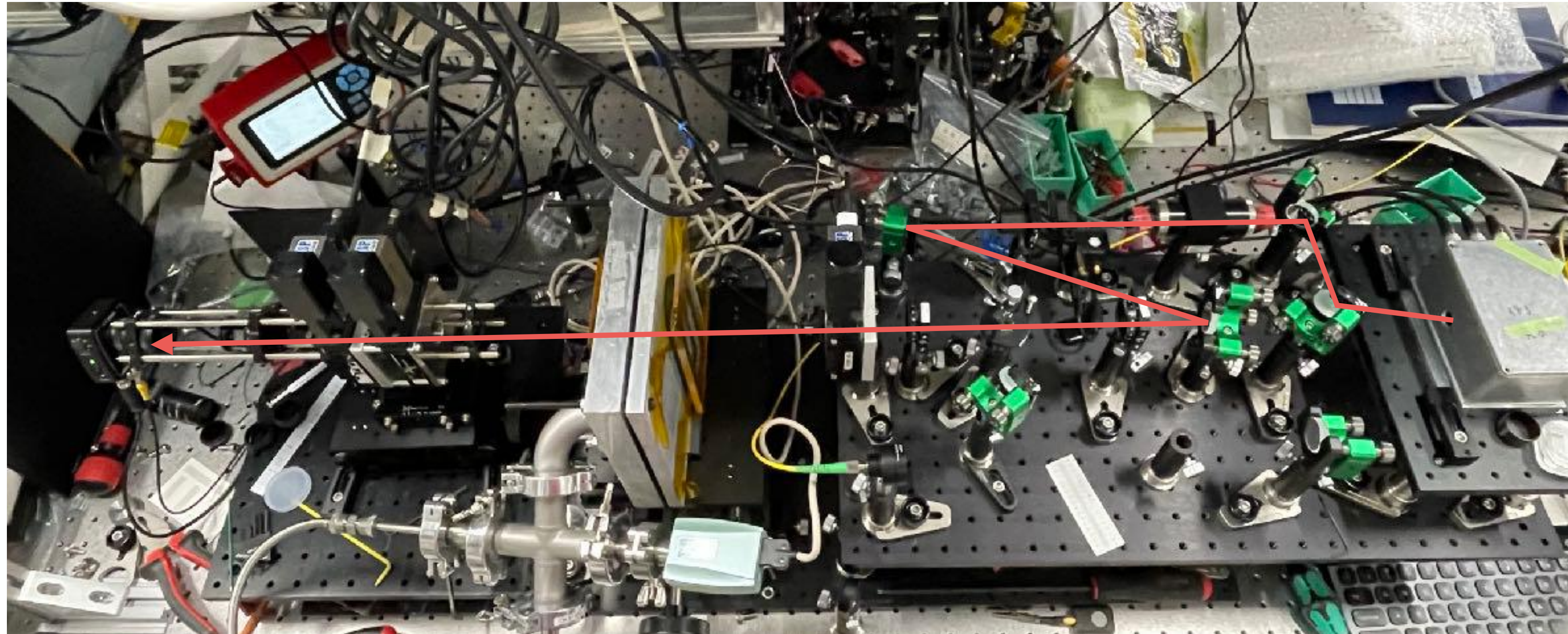
Glass	Stress-optical coeff. (10^{-3} /GPa)
Borofloat	4.0
N-BK7	2.77
Corning 7980 (Fused silica)	3.5
Shott SF57HTultra	0.07



Birefringence measurement @ Okayama

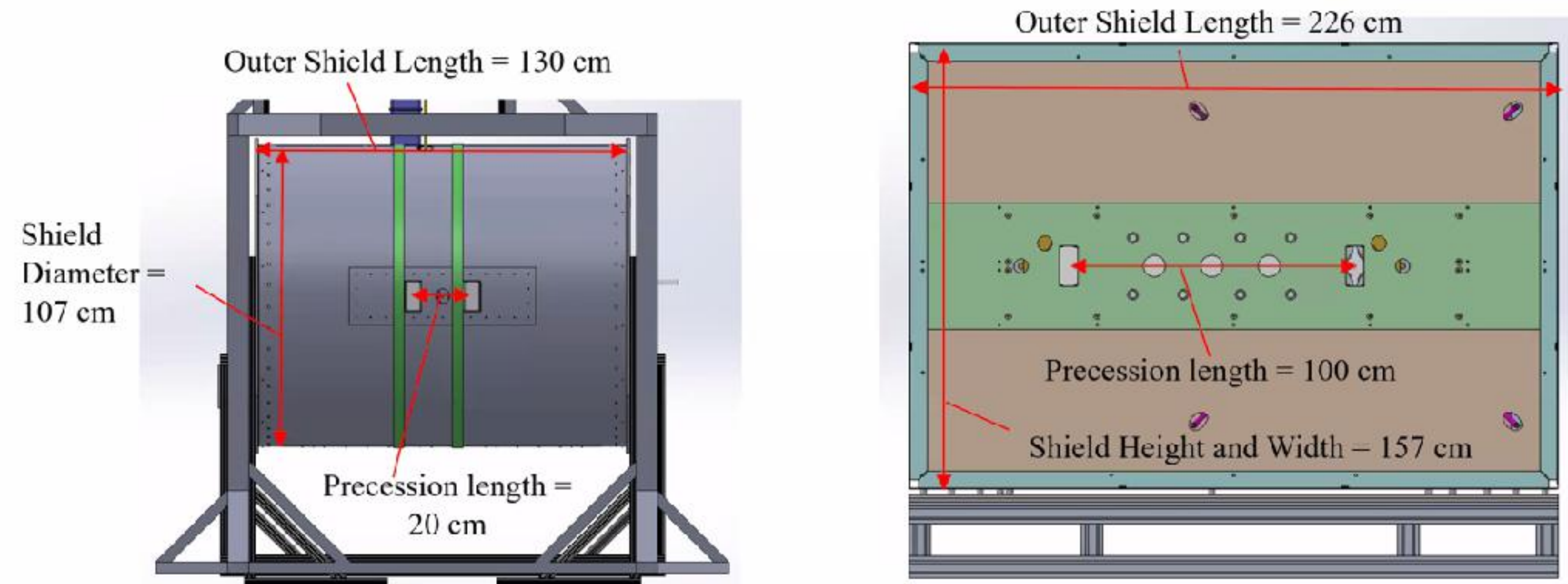
V. Wirthl *et al.*, OSA Continuum 4 2949 (2021).

Evaluation is on-going w/ a dedicated polarimeter.



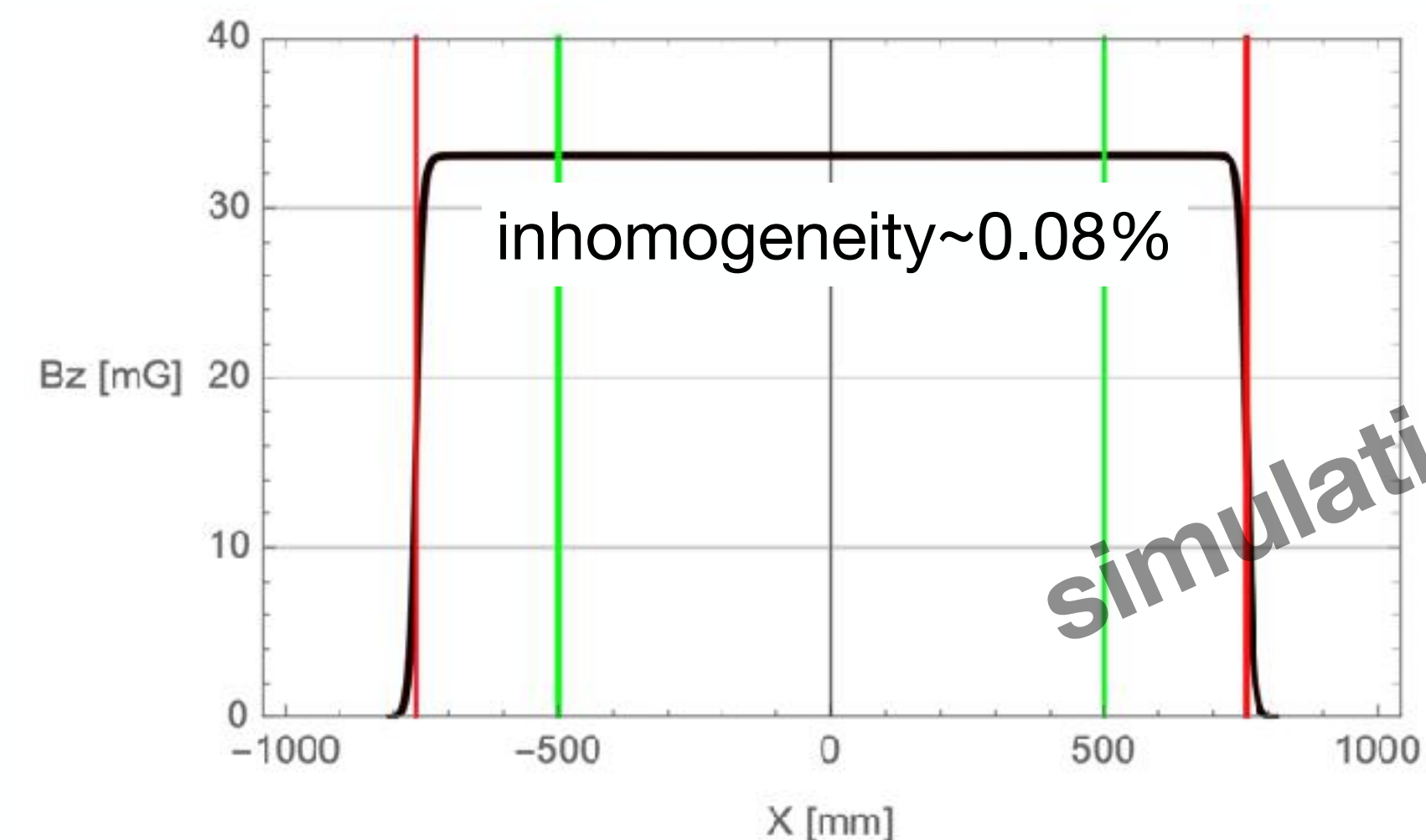
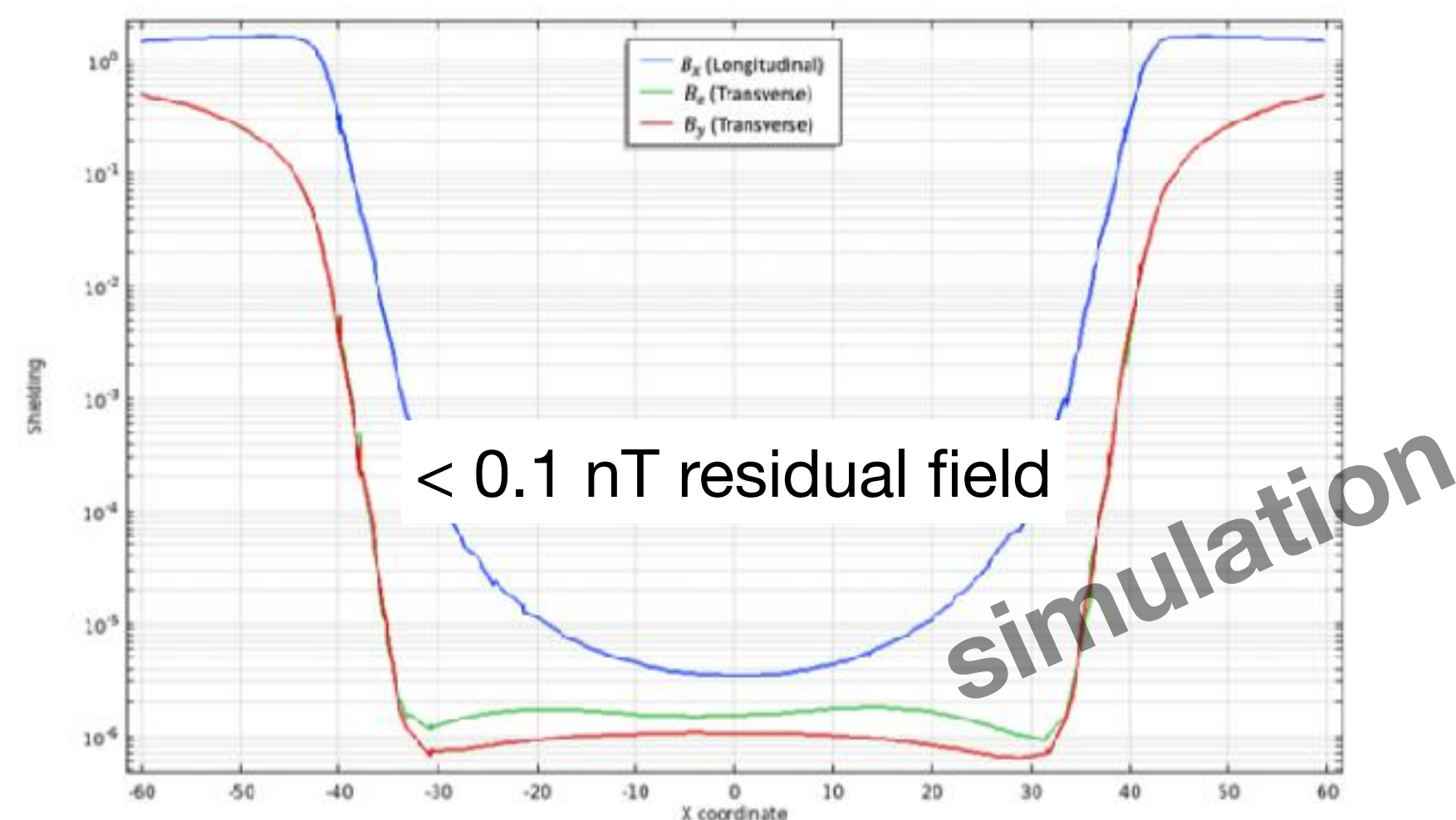
magnetic field

- Enlarged magnetic shield
- Applied magnetic field ~ 10 nT
- Required residual field & field gradient
 < 0.1 nT, < 0.1 nT/cm



- 3-layer mu-metal shield + 108 degaussing coils + Self shielded $\text{Cos}\theta$ coils

Siyuan Liu (Northwestern), DAMOP2022



Magnetic shield

Assembly work and performance evaluation on-going at Northwestern.

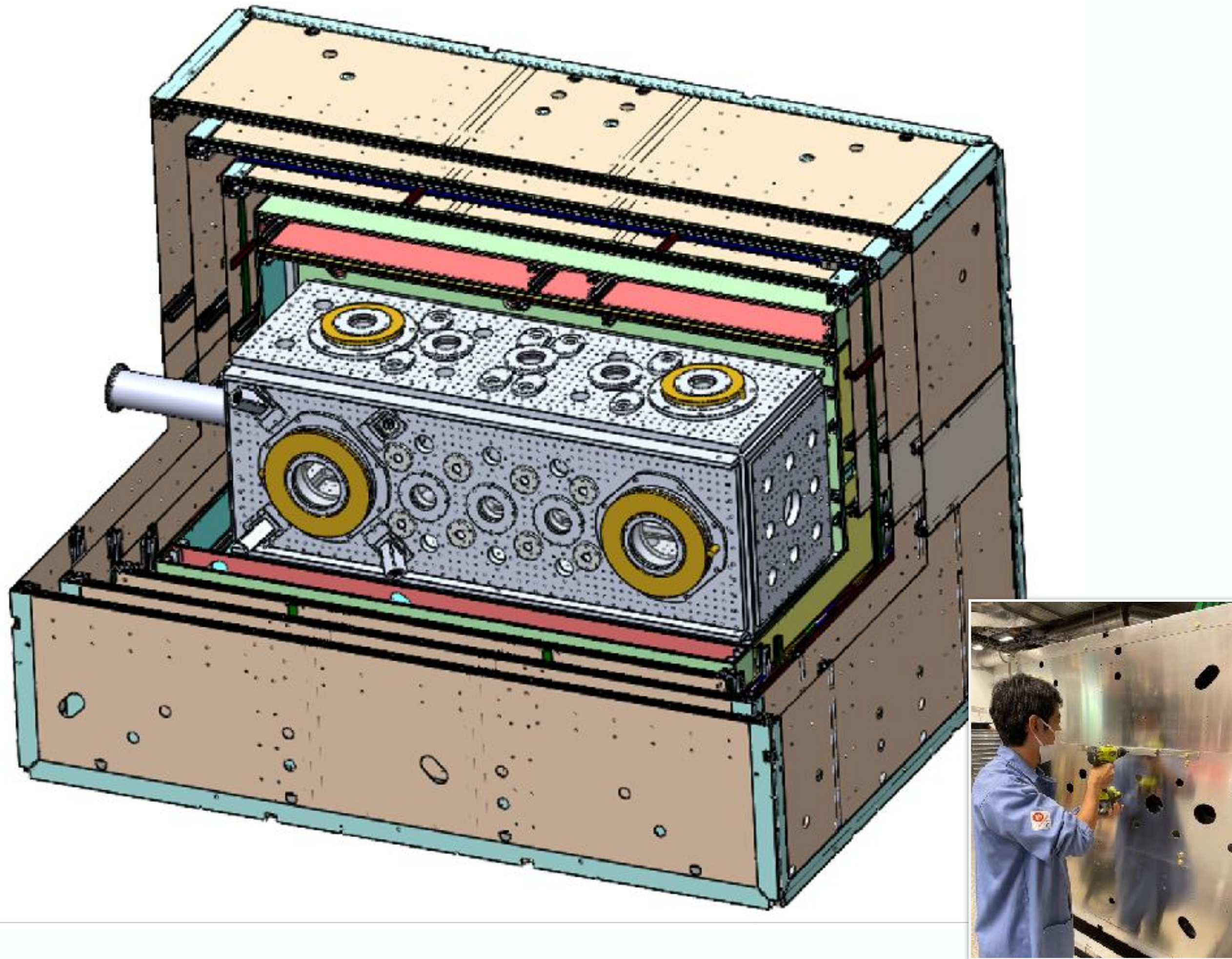
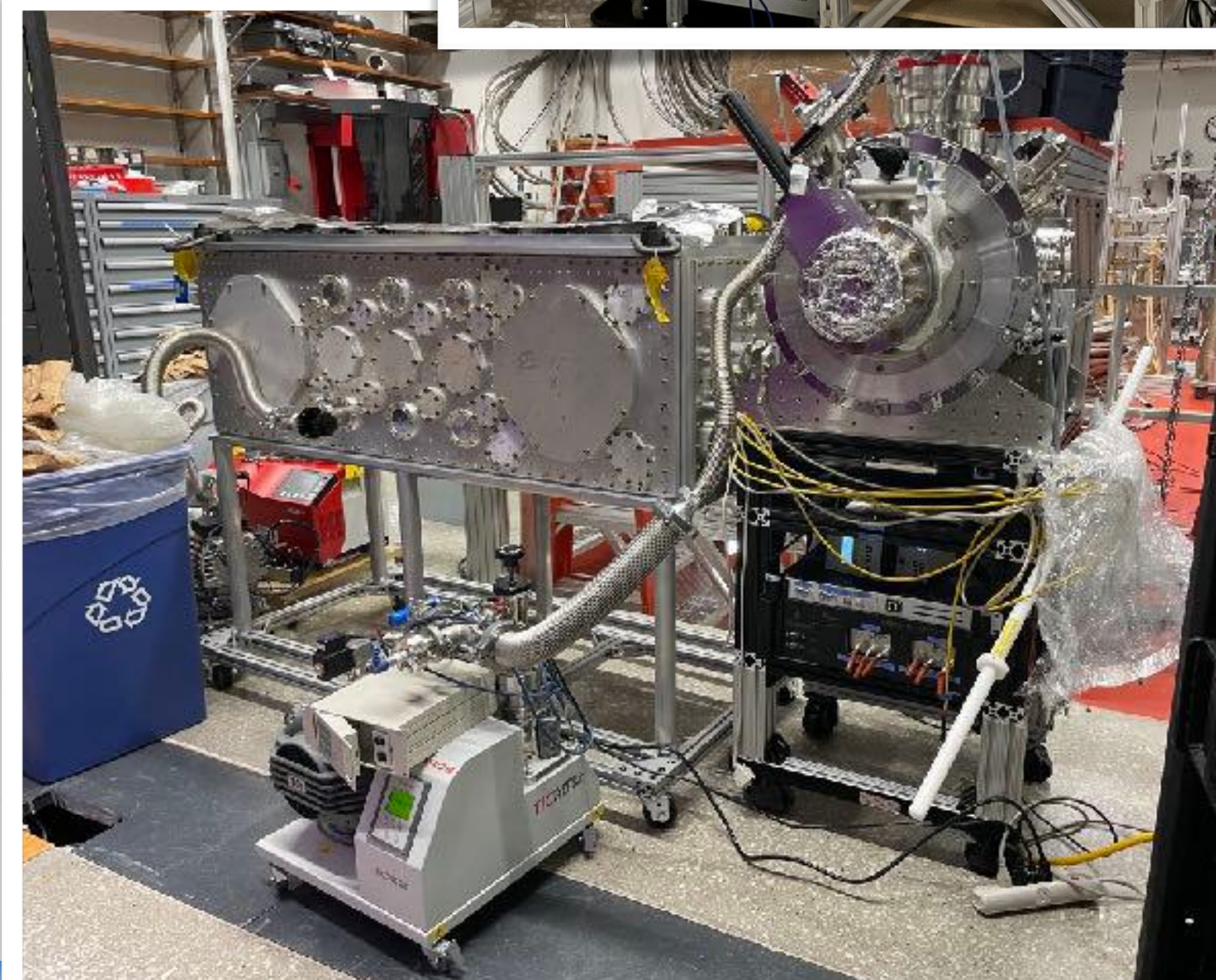
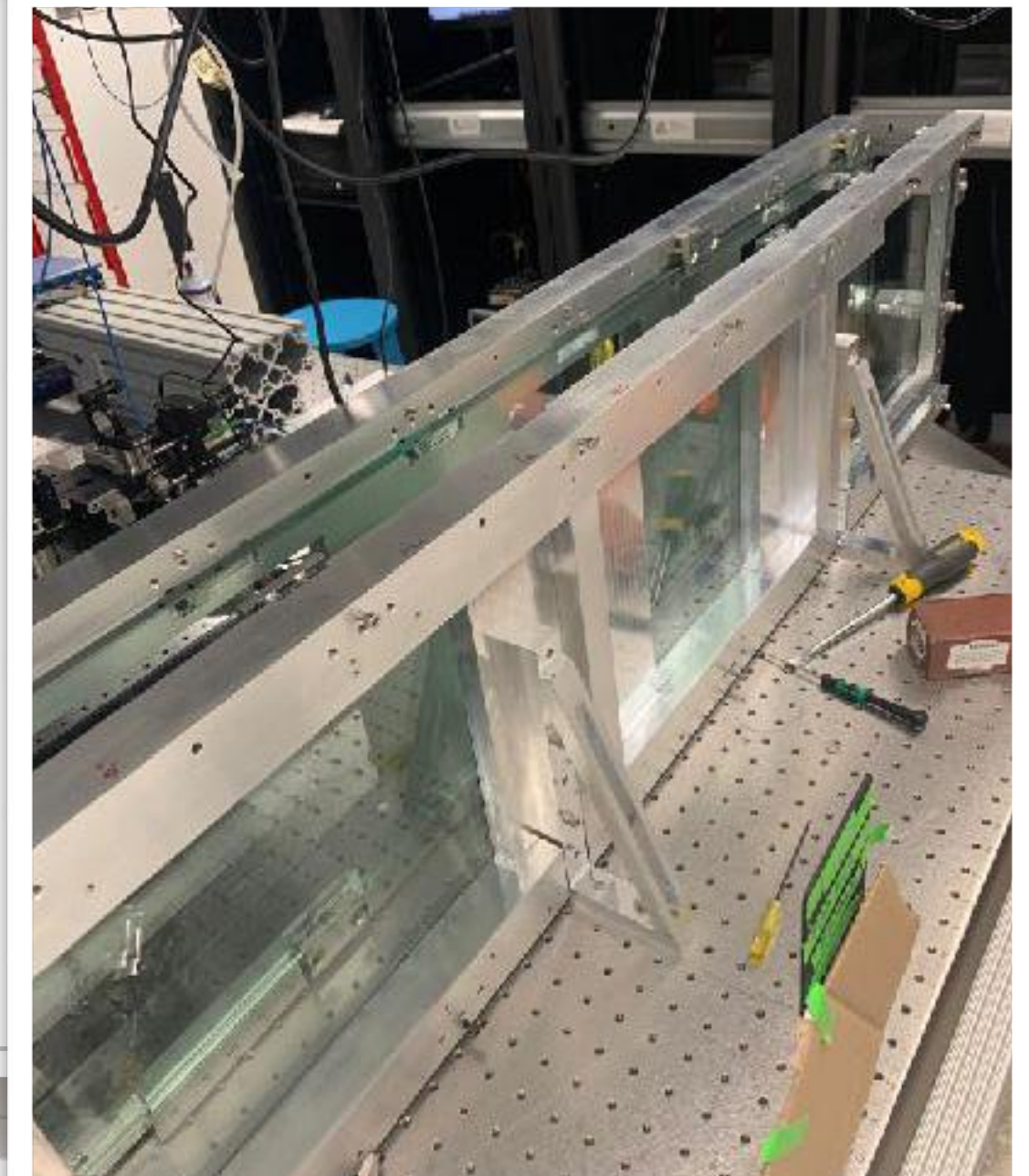
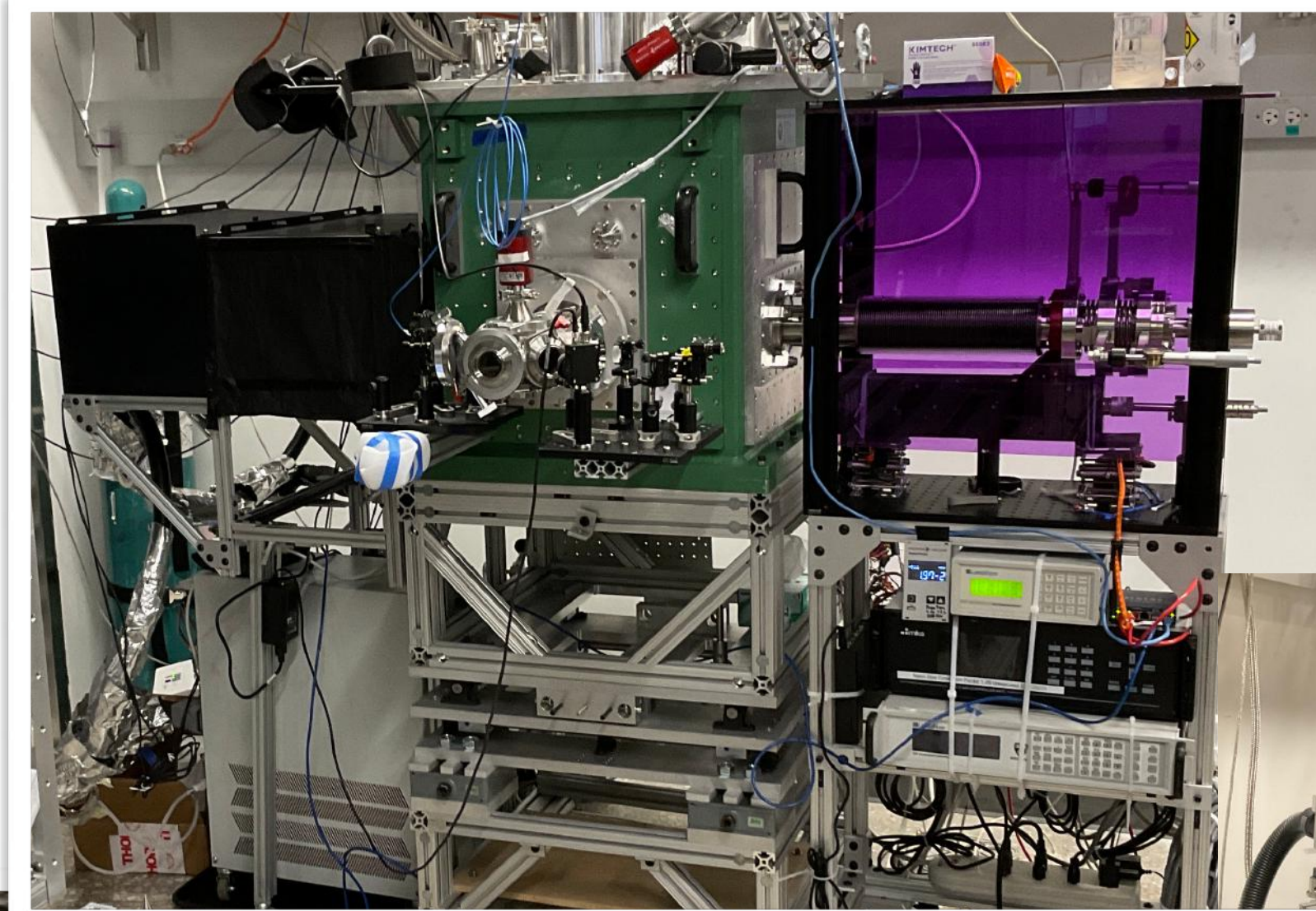


Photo gallery at Northwestern

2022-2023

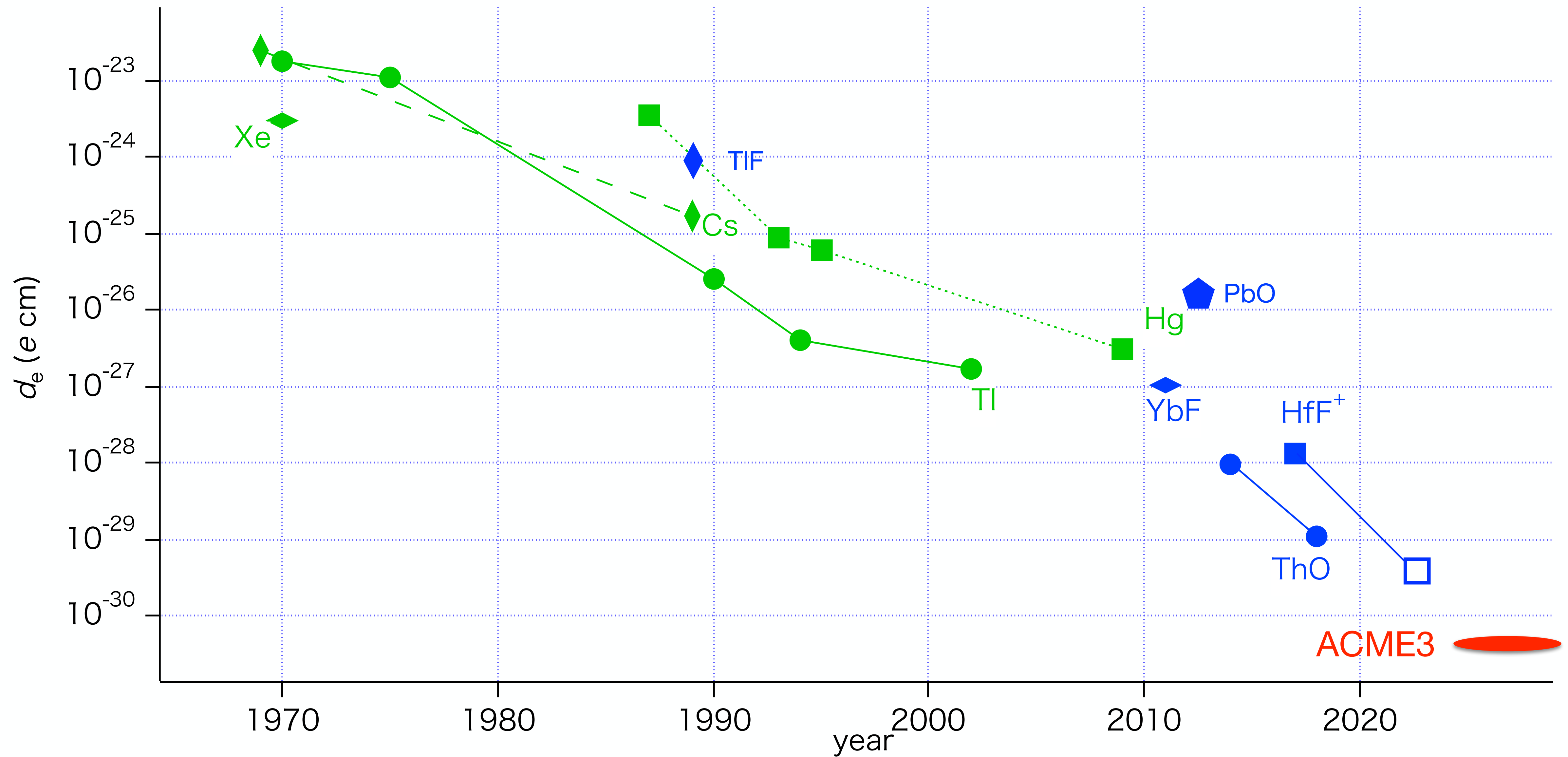
March 2021

Clean up the ACME space



***ACME construction
in progress !***

Outlook



- Electron EDM (eEDM) is a powerful tool to search for a new physics beyond the standard model.
- Many R&D works for ACME3 are going to improve the current upper limit of eEDM by a factor of ~ 30 .