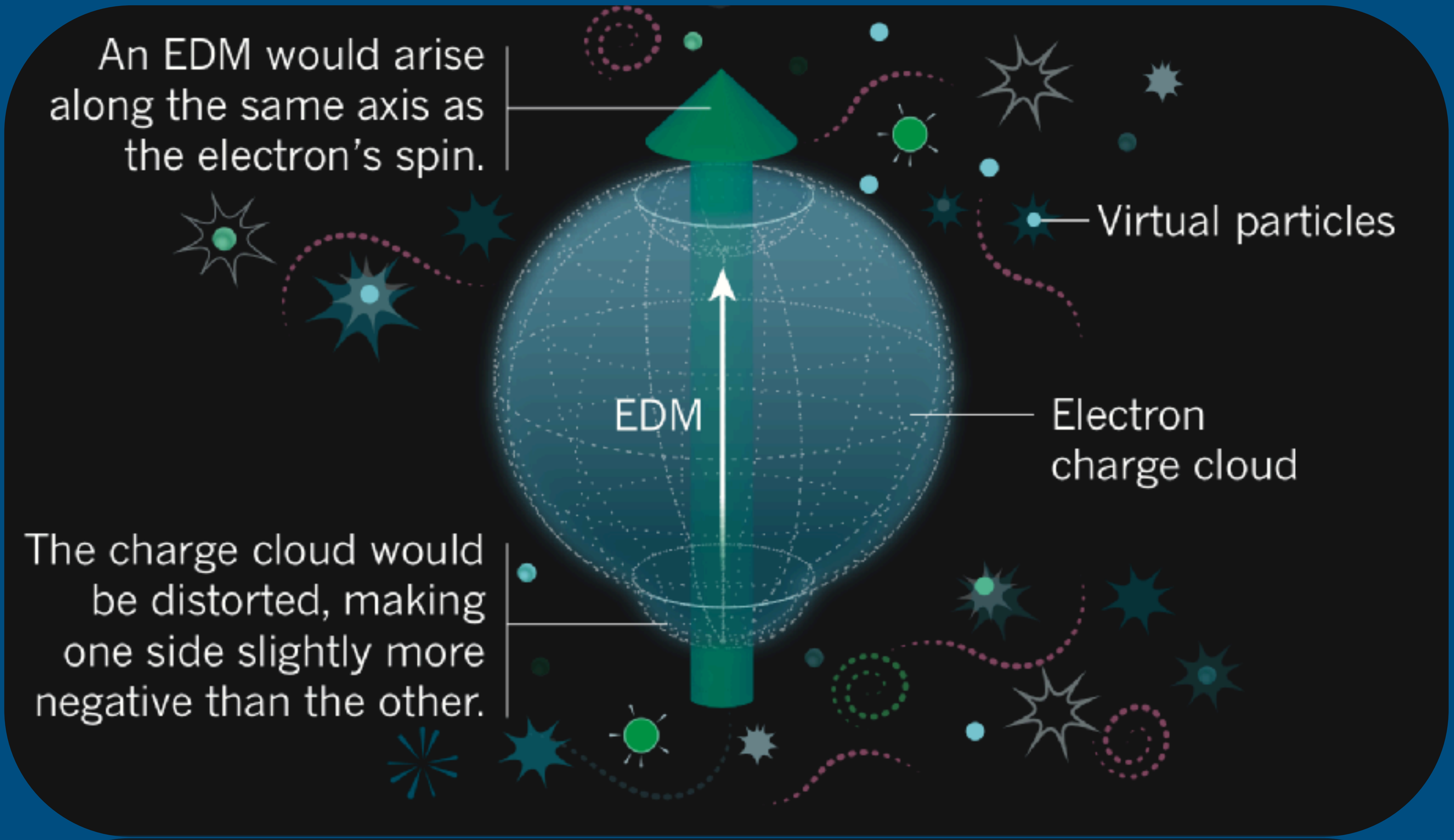
Probing the electron's EDM

using cold and slow molecules

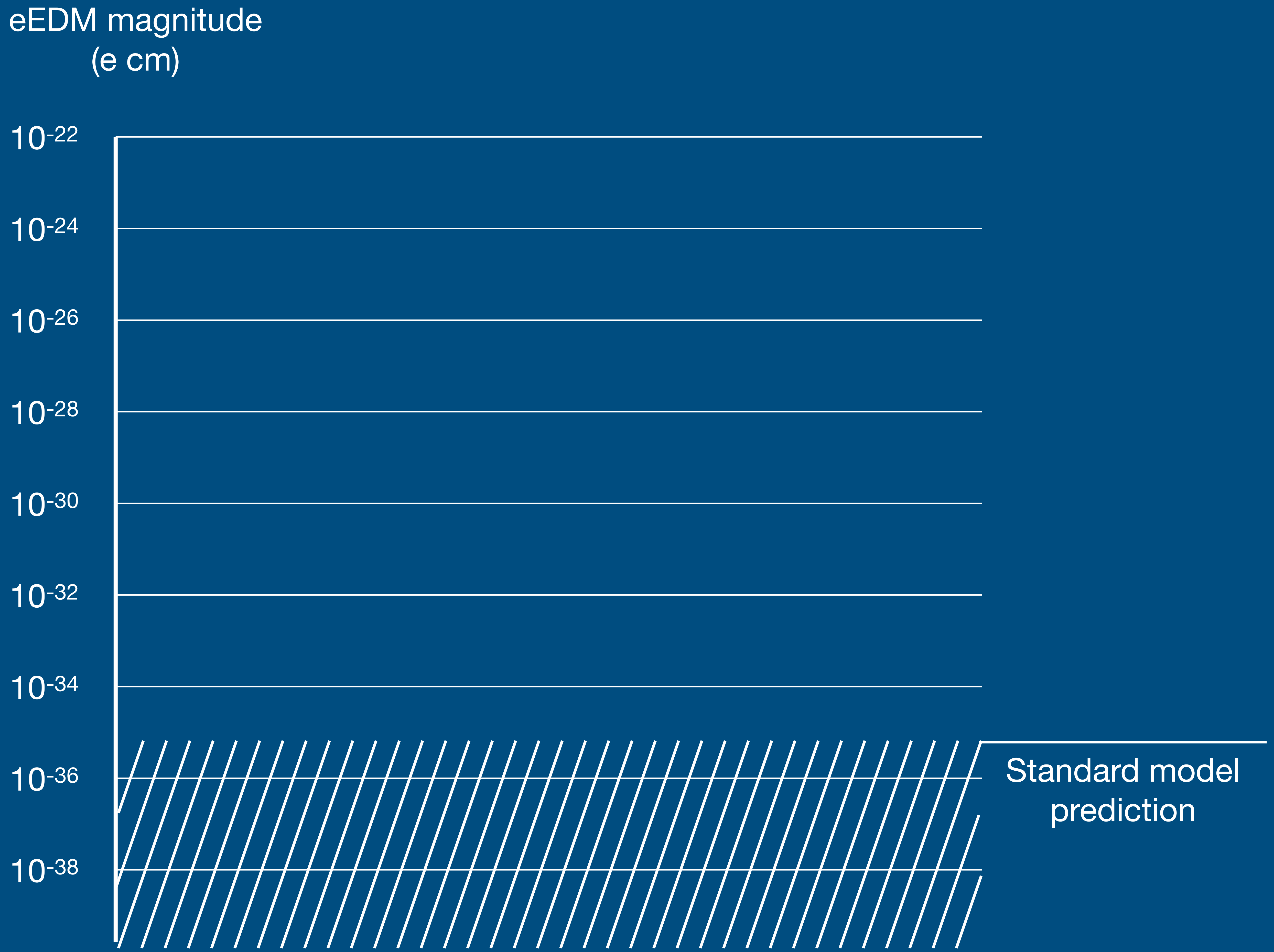
Steven Hoekstra, University of Groningen and Nikhef, The Netherlands

The electric dipole moment of the electron

Probing (or eliminating) new physics

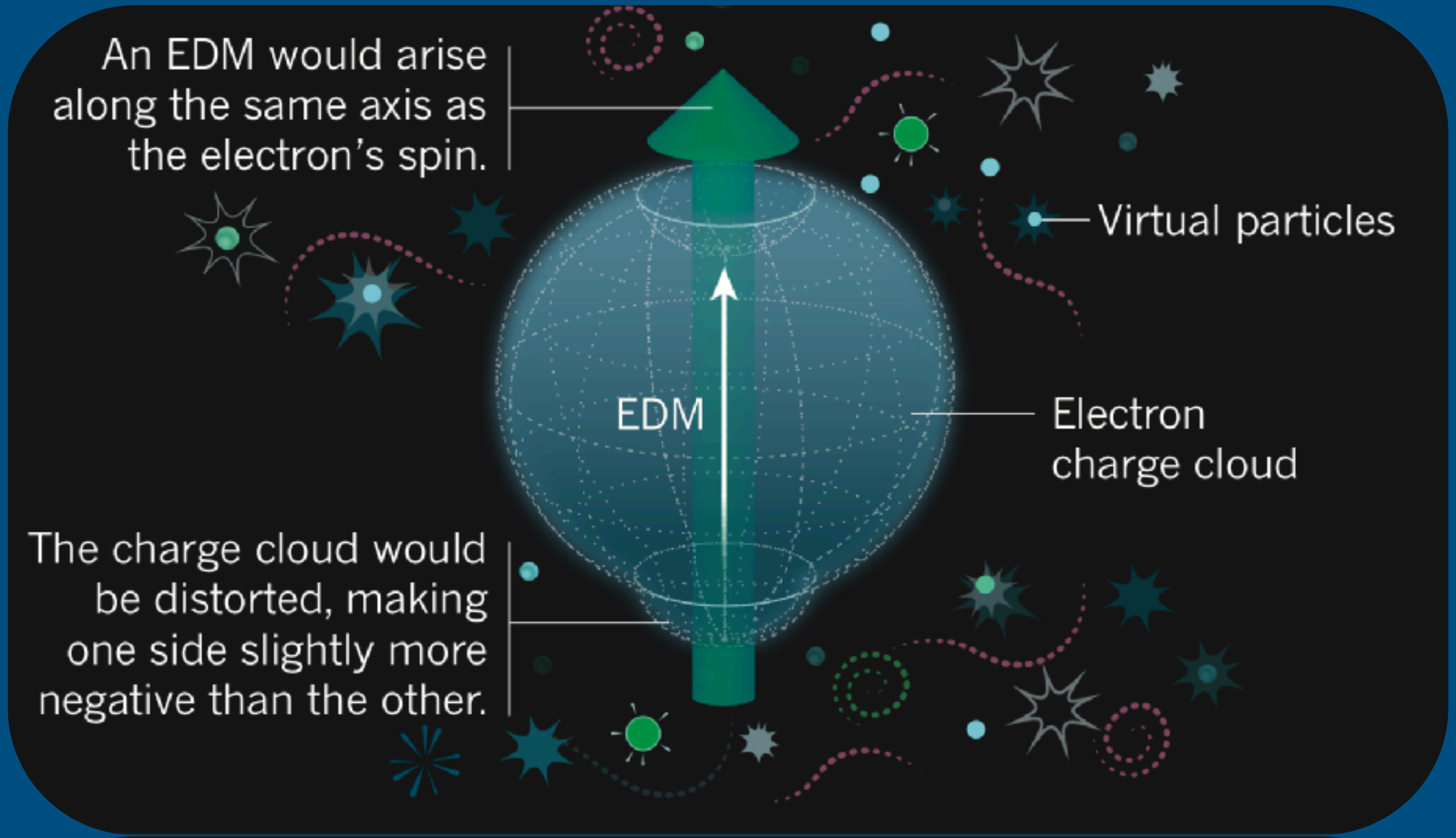


eEDM violates P, T and CP symmetry
(provided CPT holds)

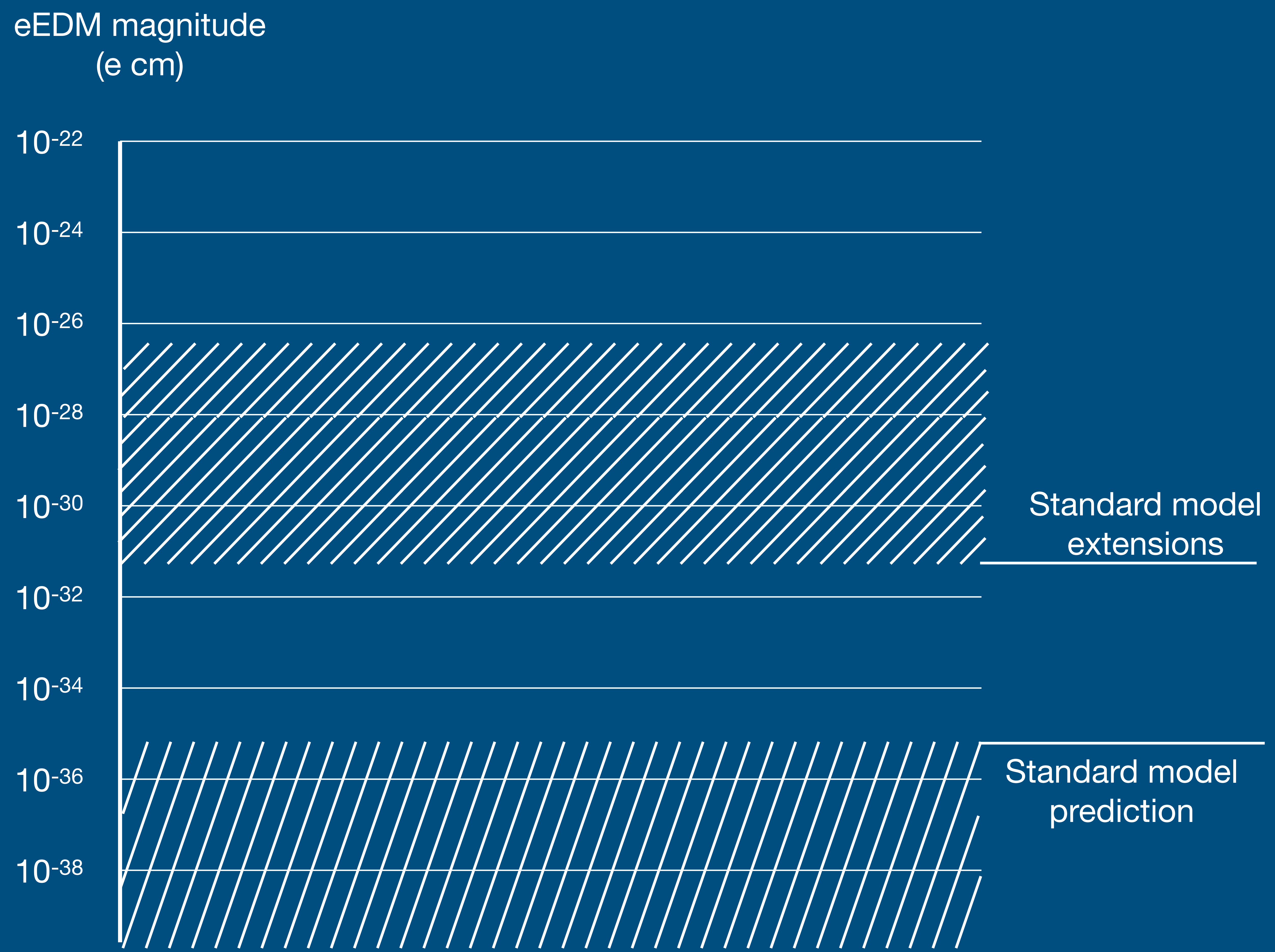


The electric dipole moment of the electron

Probing (or eliminating) new physics

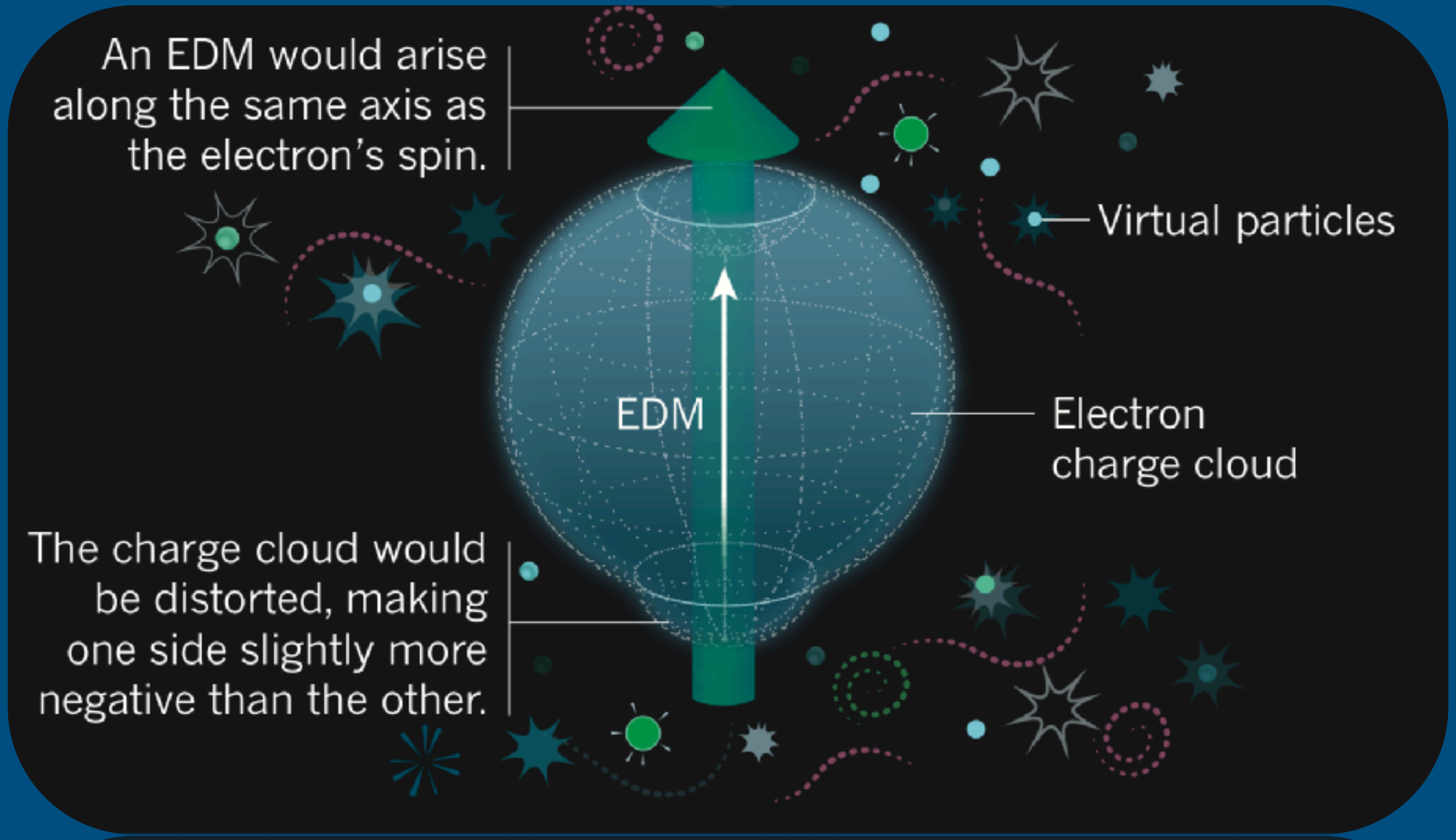


eEDM violates P, T and CP symmetry (provided CPT holds)

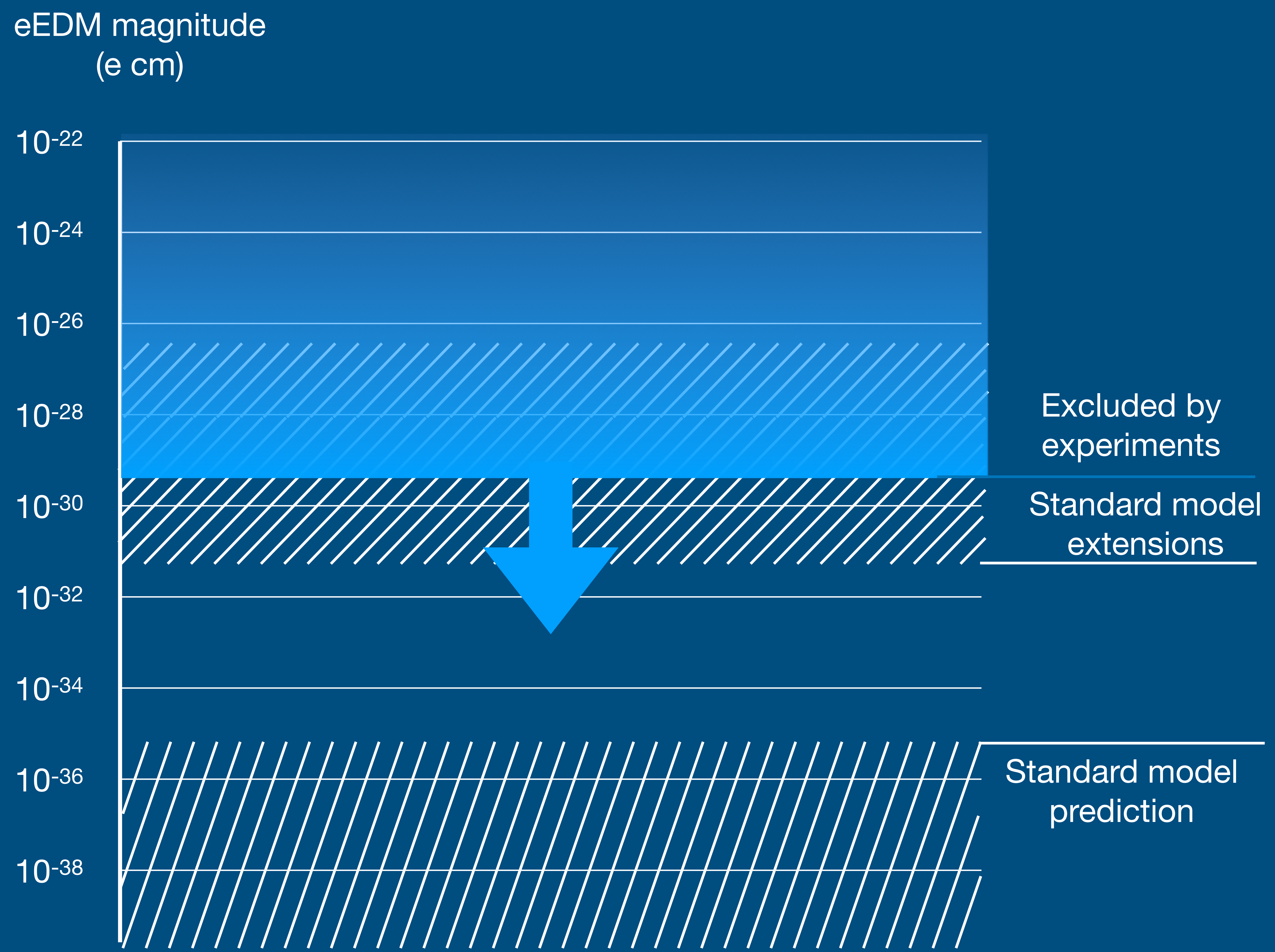


The electric dipole moment of the electron

Probing (or eliminating) new physics

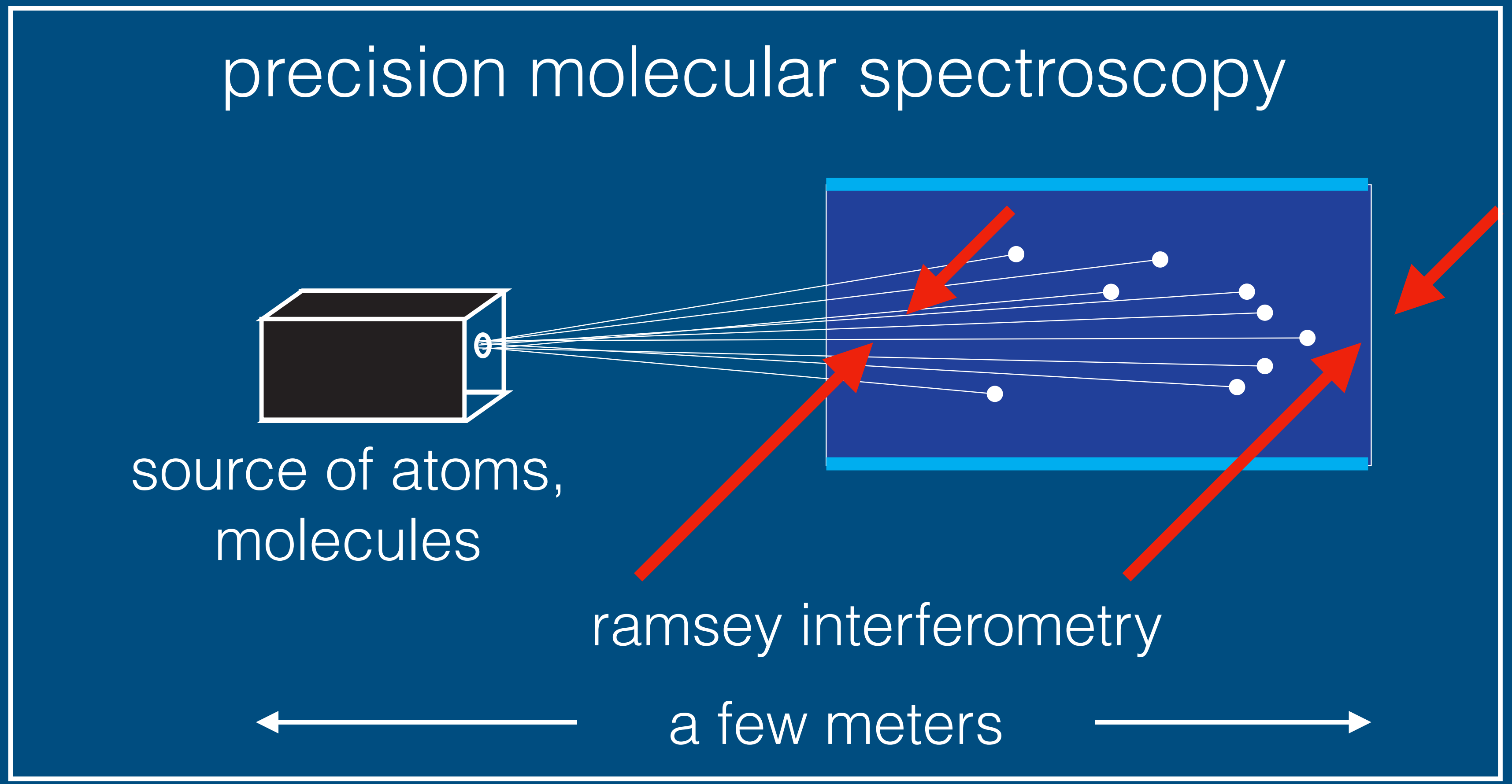


eEDM violates P, T and CP symmetry
(provided CPT holds)



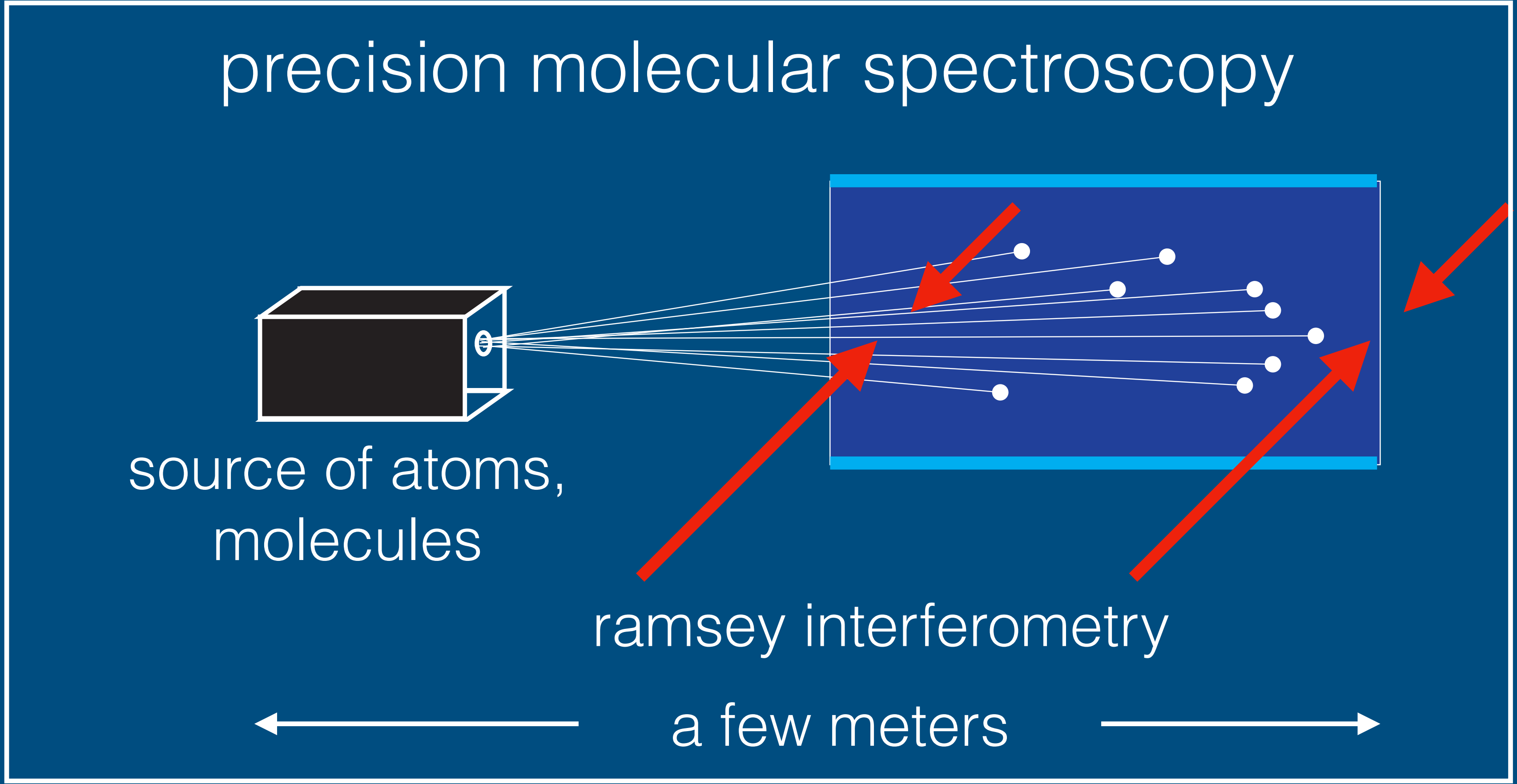
Precision measurements with molecules

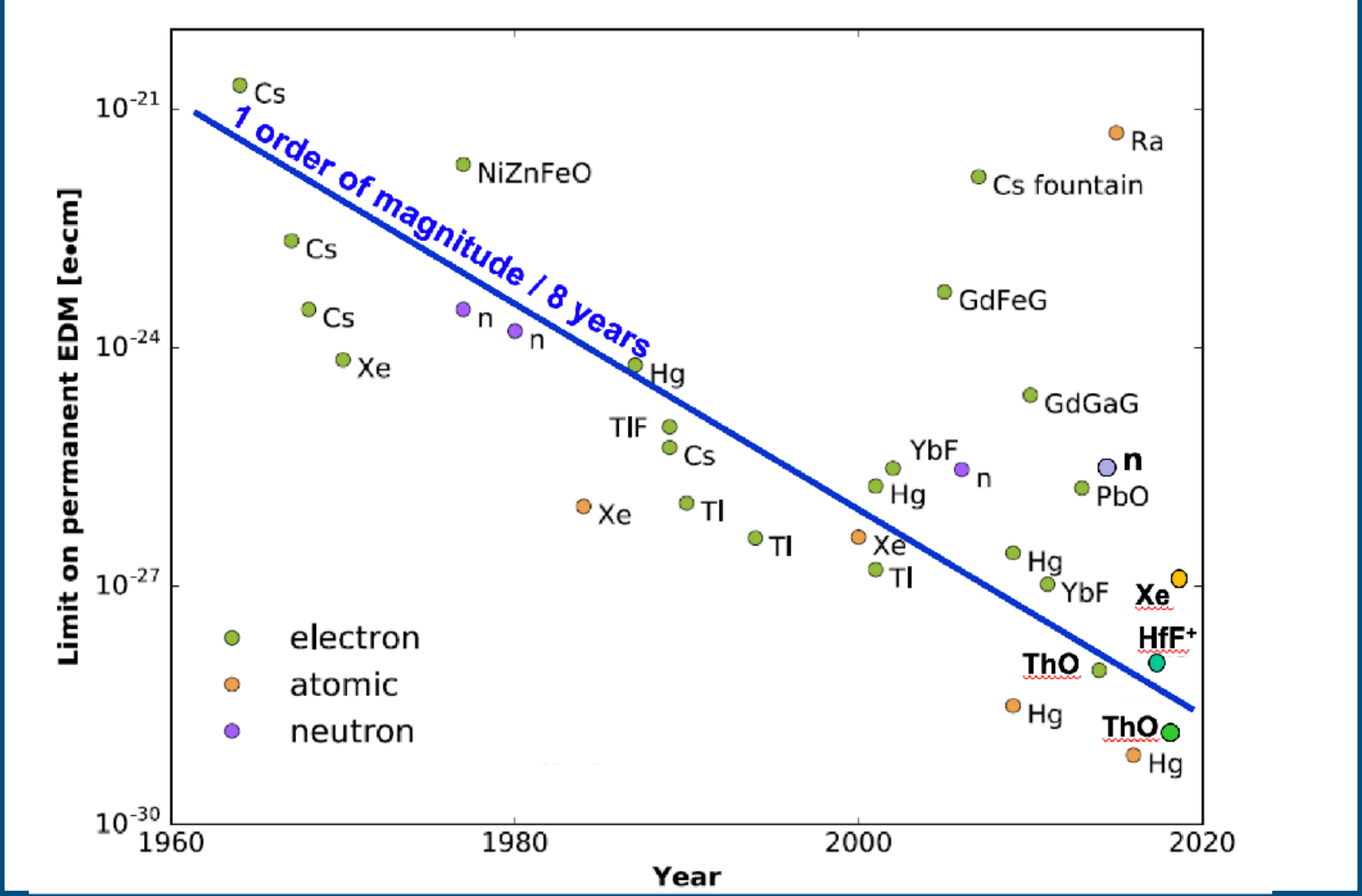
Complex quantum systems with an advantage



Precision measurements with molecules

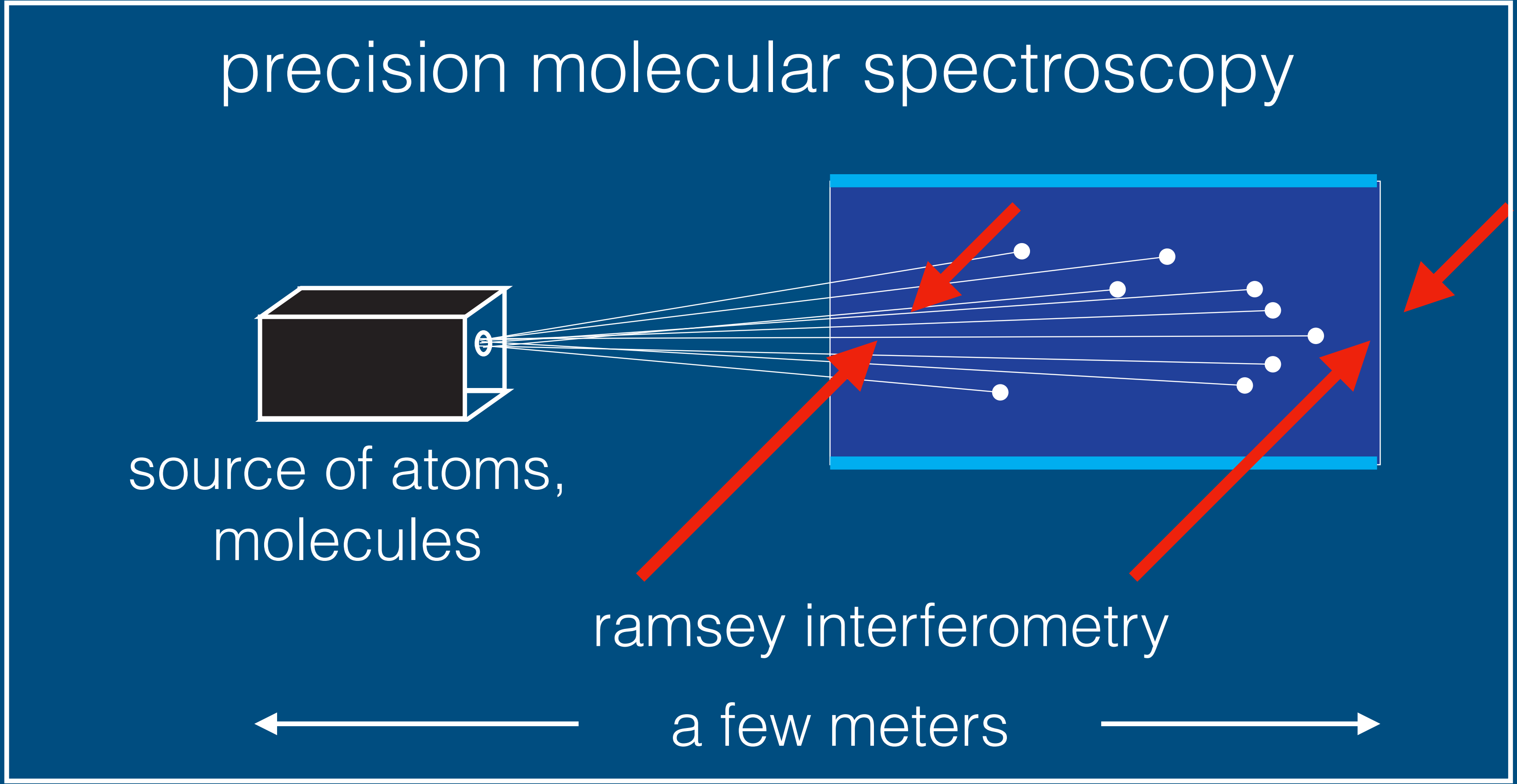
Complex quantum systems with an advantage

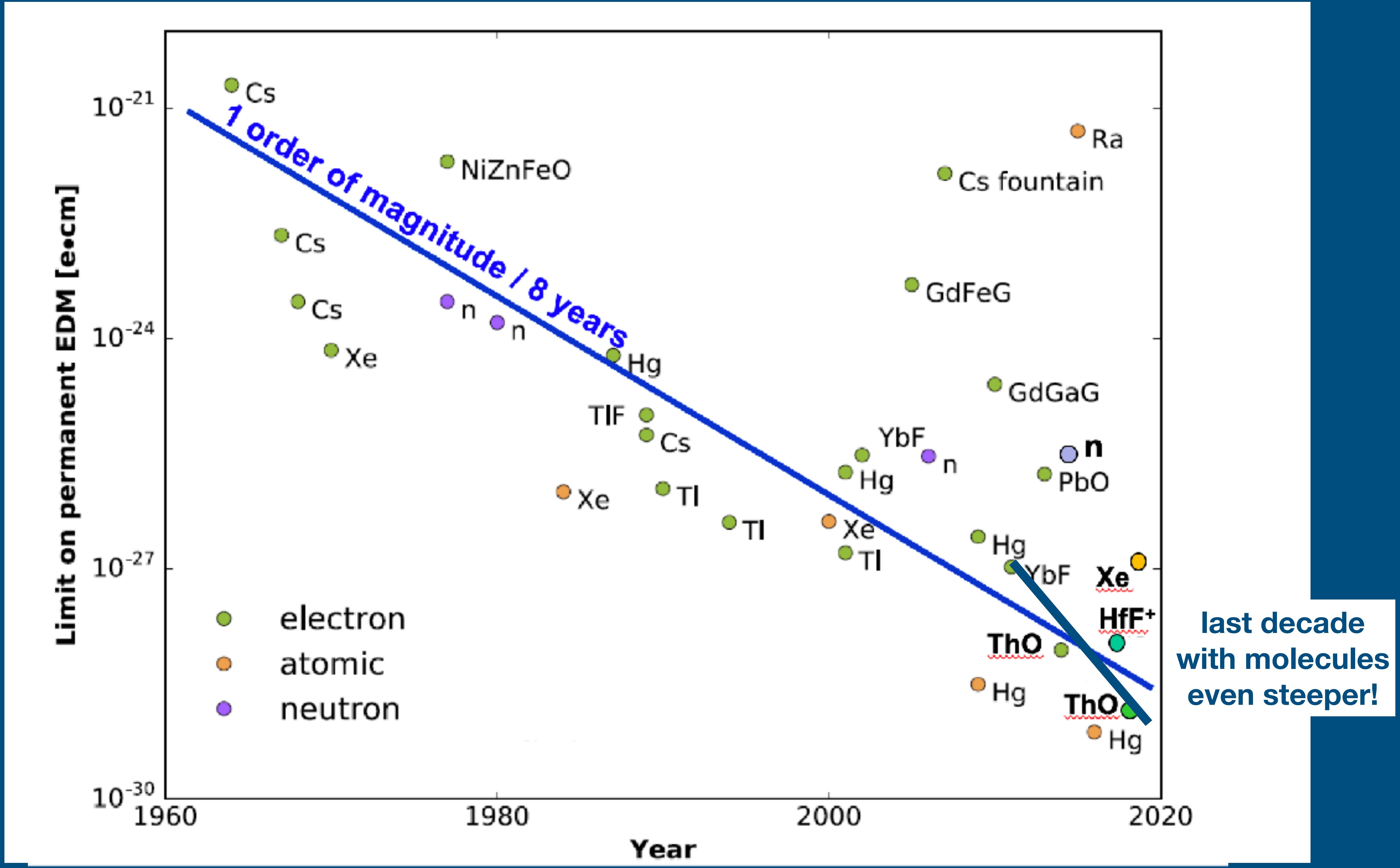




Precision measurements with molecules

Complex quantum systems with an advantage

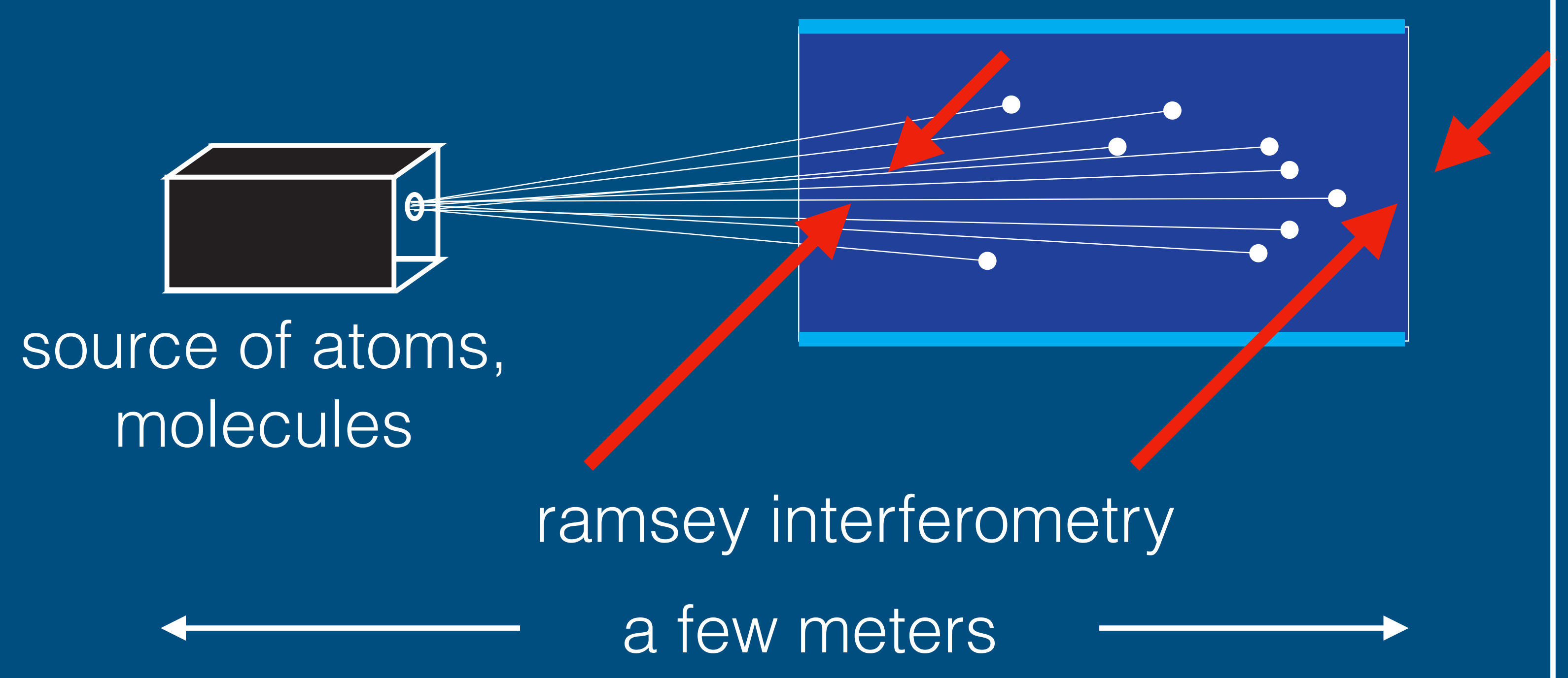




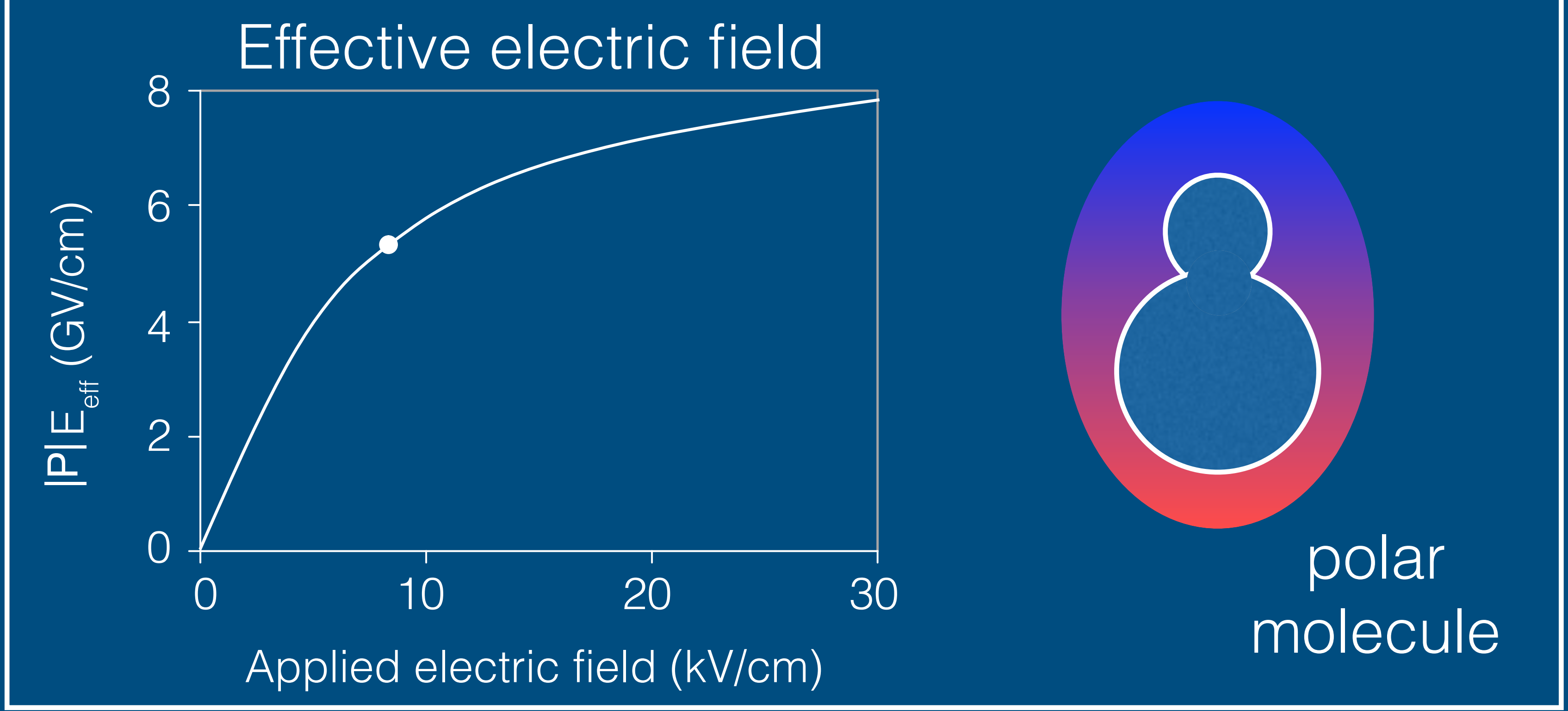
Precision measurements with molecules

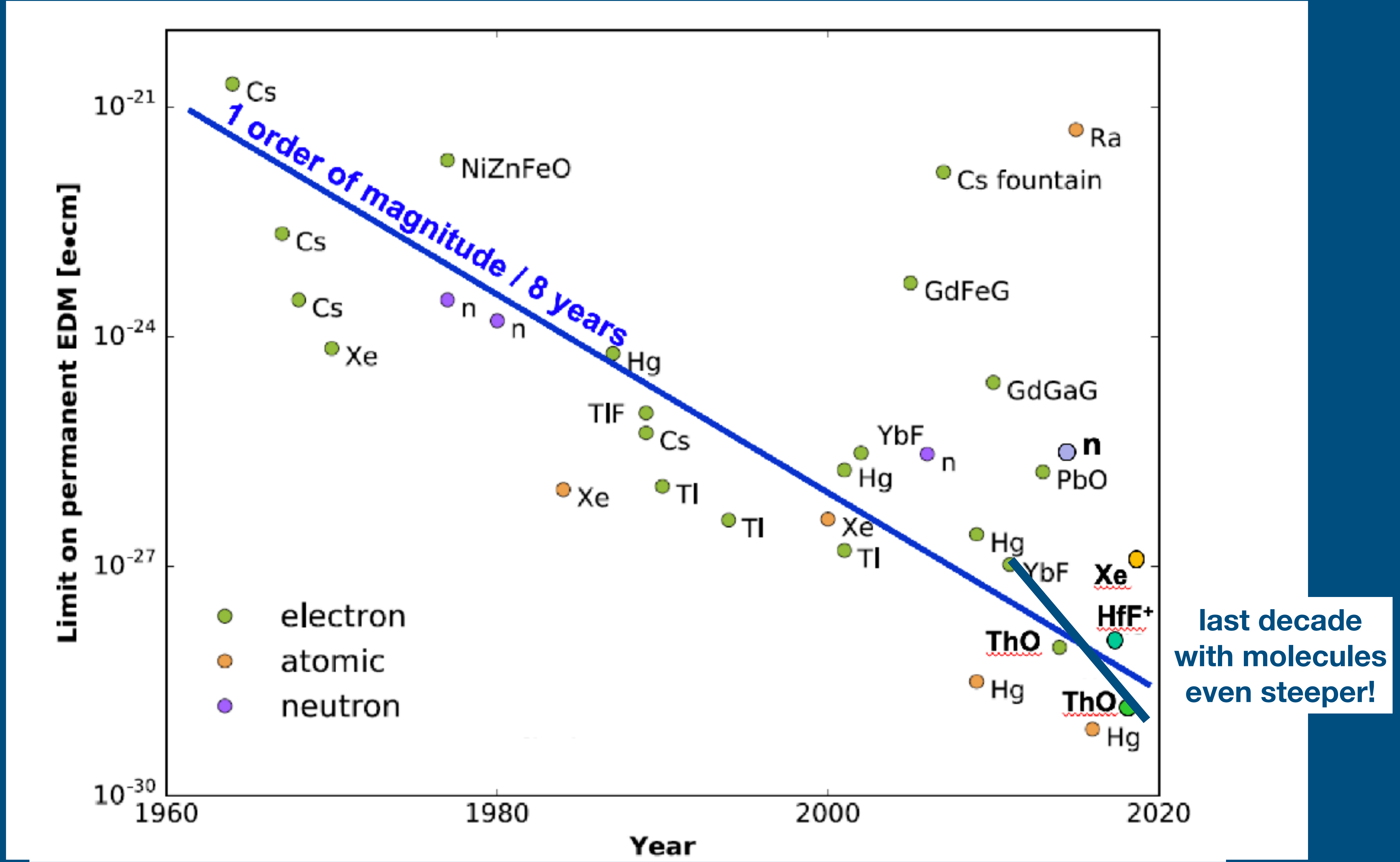
Complex quantum systems with an advantage

precision molecular spectroscopy



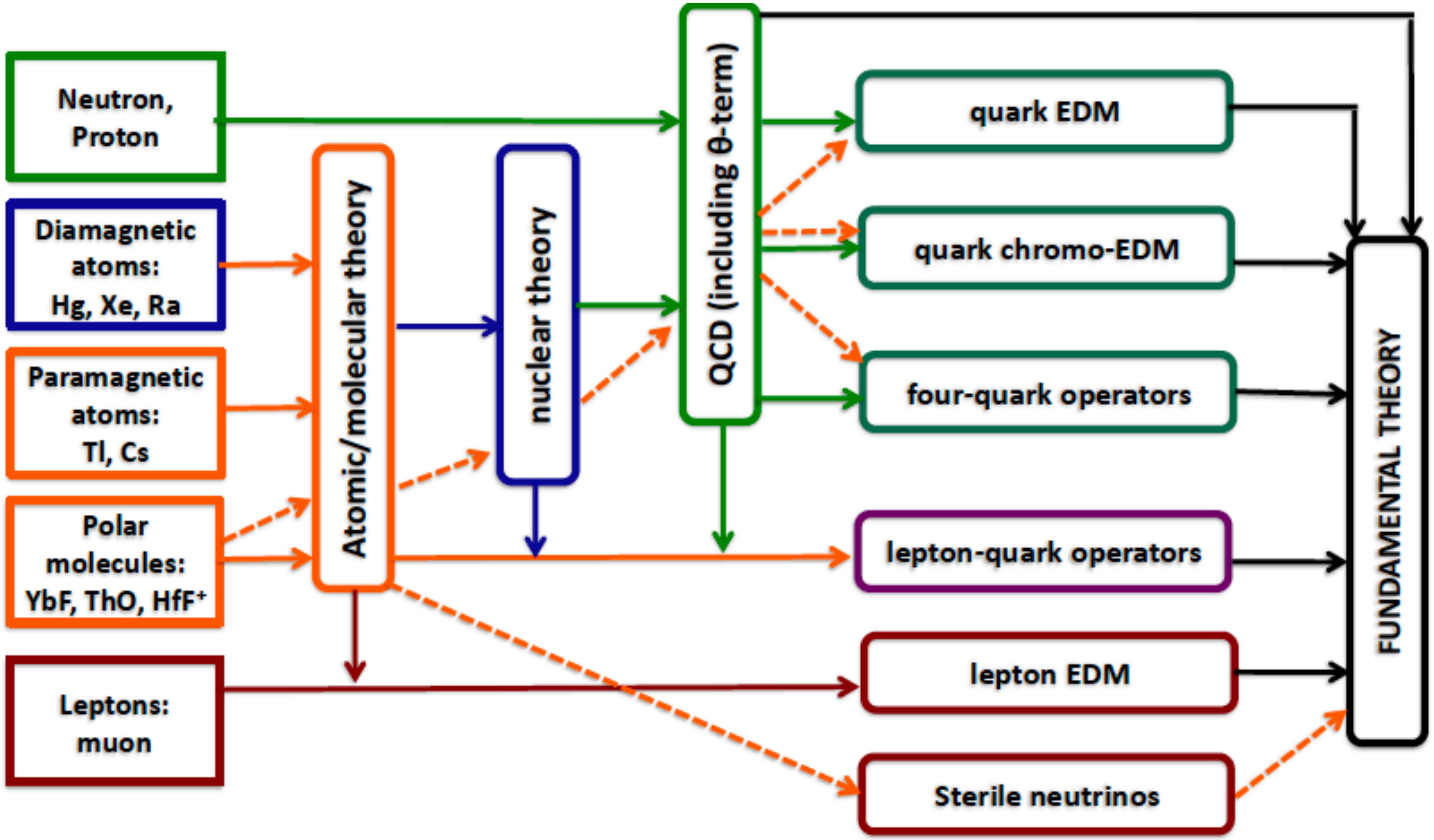
huge enhancement of E-field in molecule





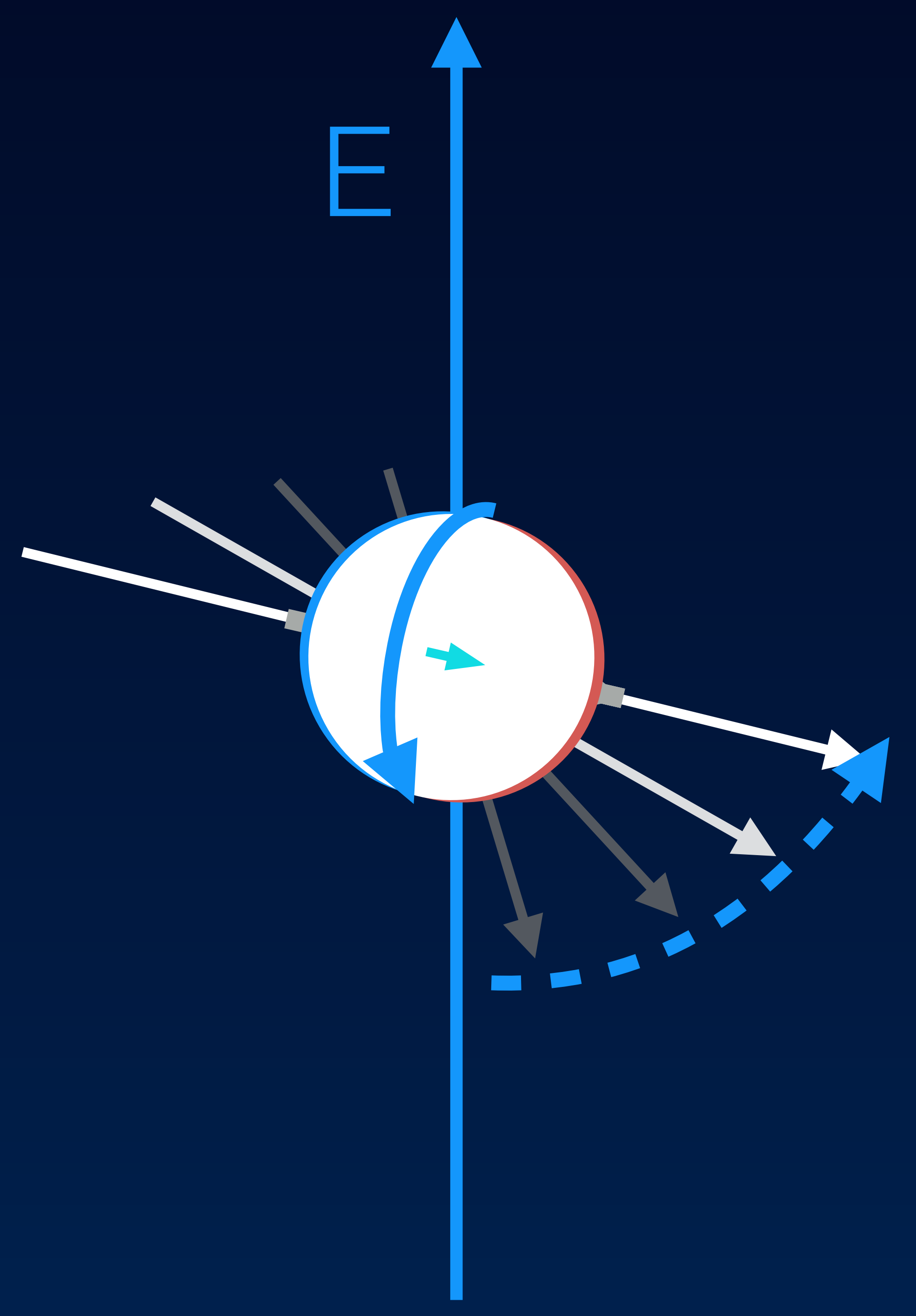
Crucial role of theory

Experiments such as neutrinoless double beta decay, EDMs, Dark Matter searches involve particle, hadronic, nuclear, molecular physics

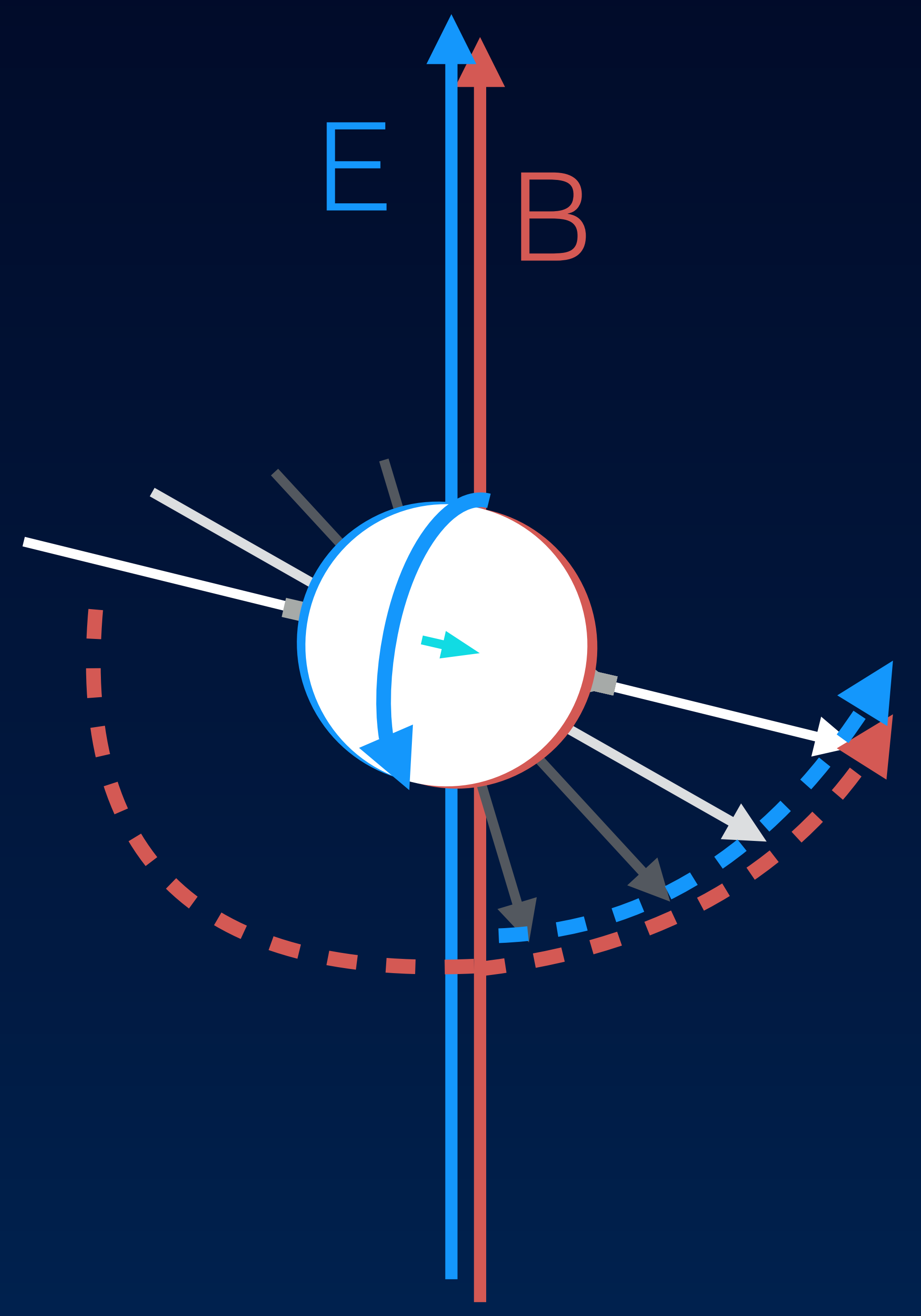


Collaboration between nuclear, AMO, condensed matter theorists and experimentalists

How to measure a dipole moment?

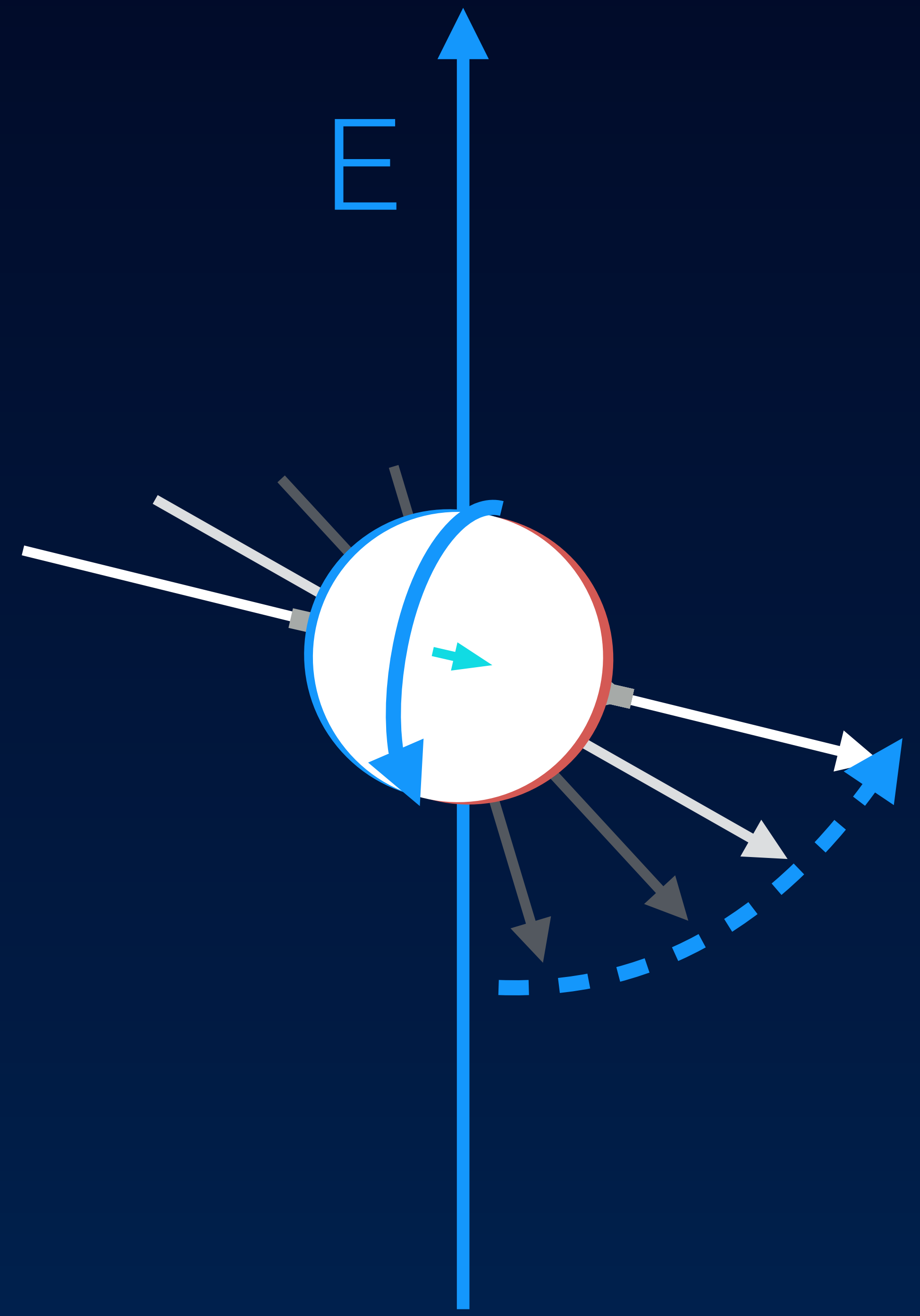


precession!

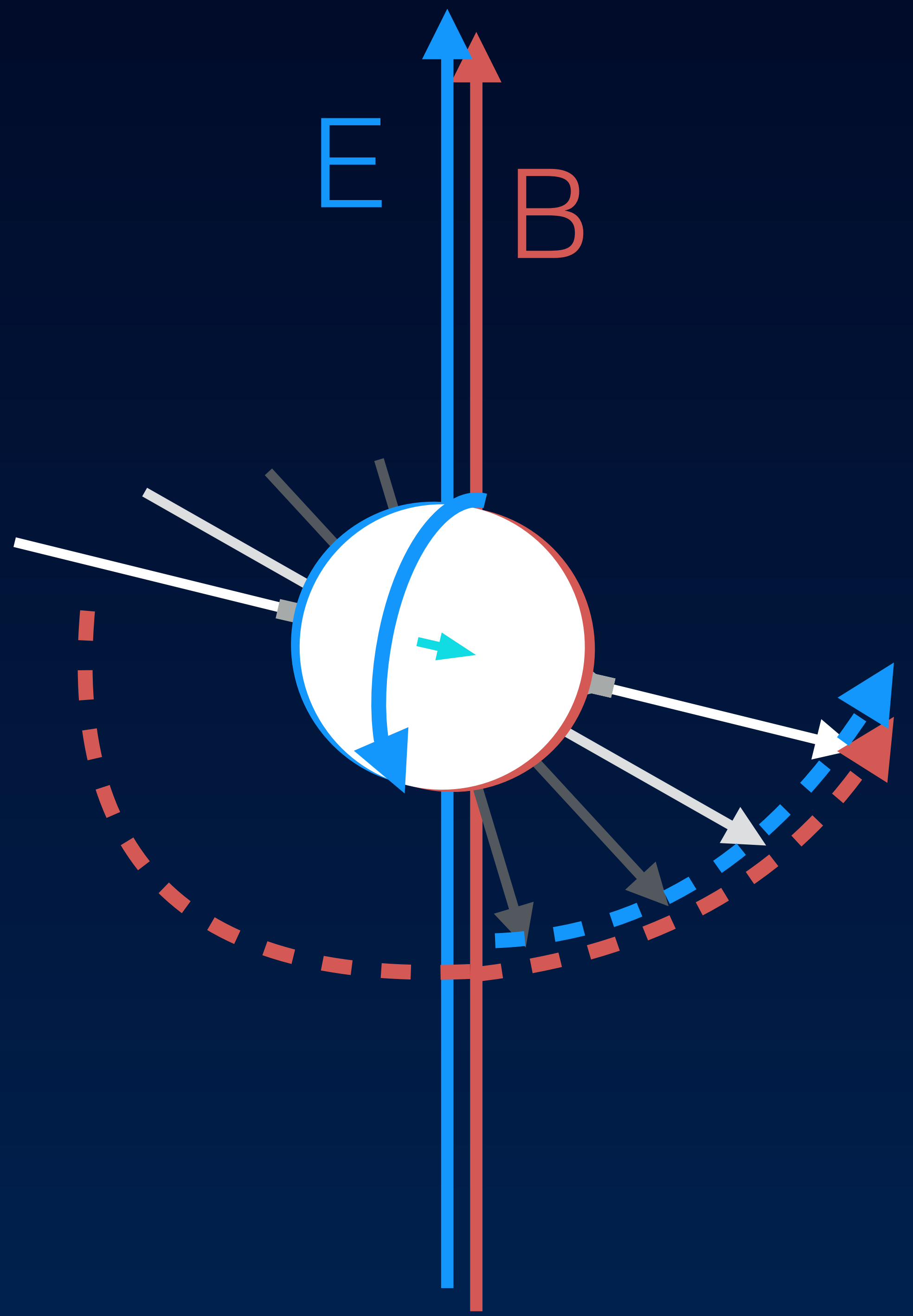


However, electron also has
magnetic dipole moment
(and charge!)

How to measure a dipole moment?

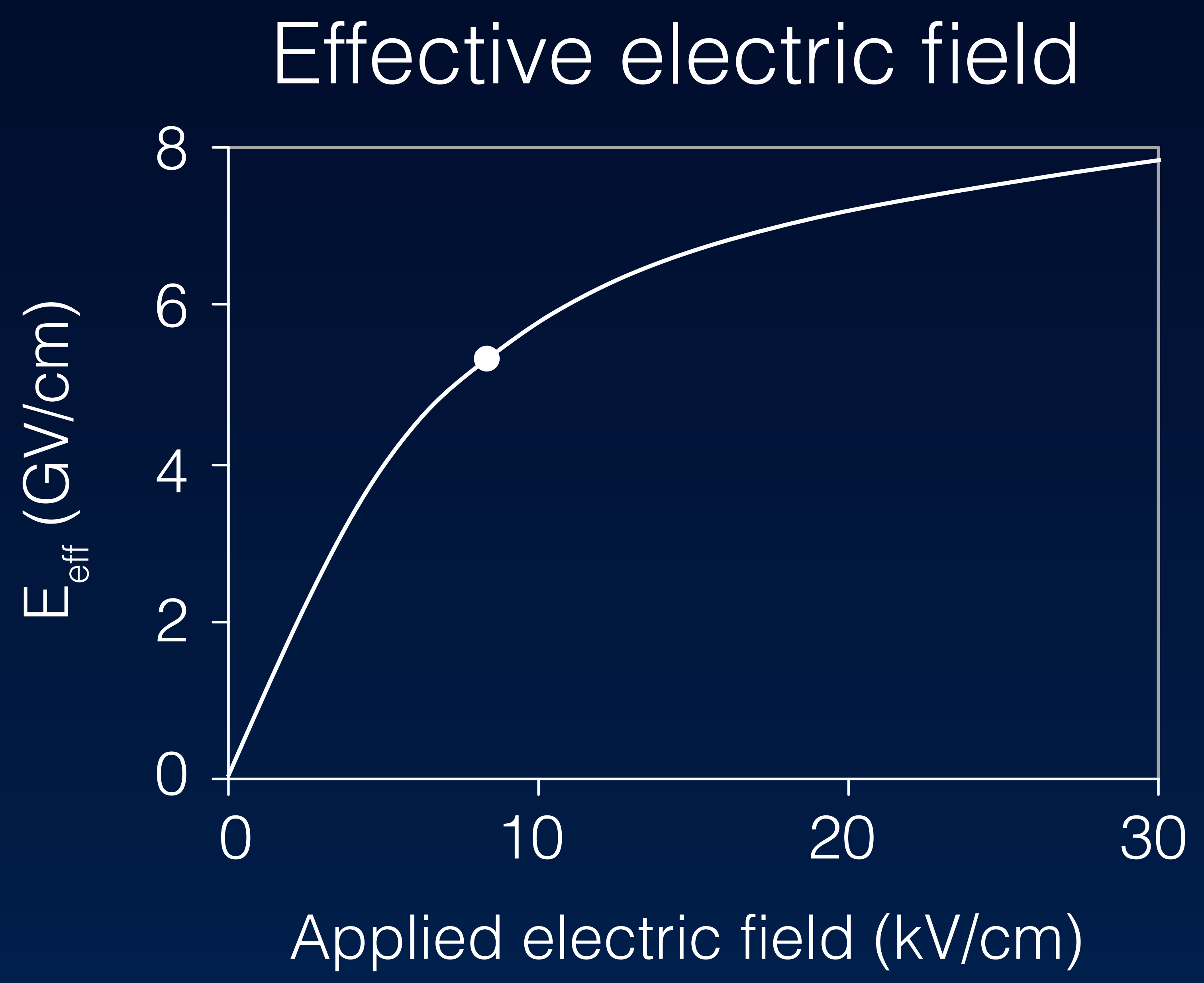


precession!



However, electron also has magnetic dipole moment (and charge!)

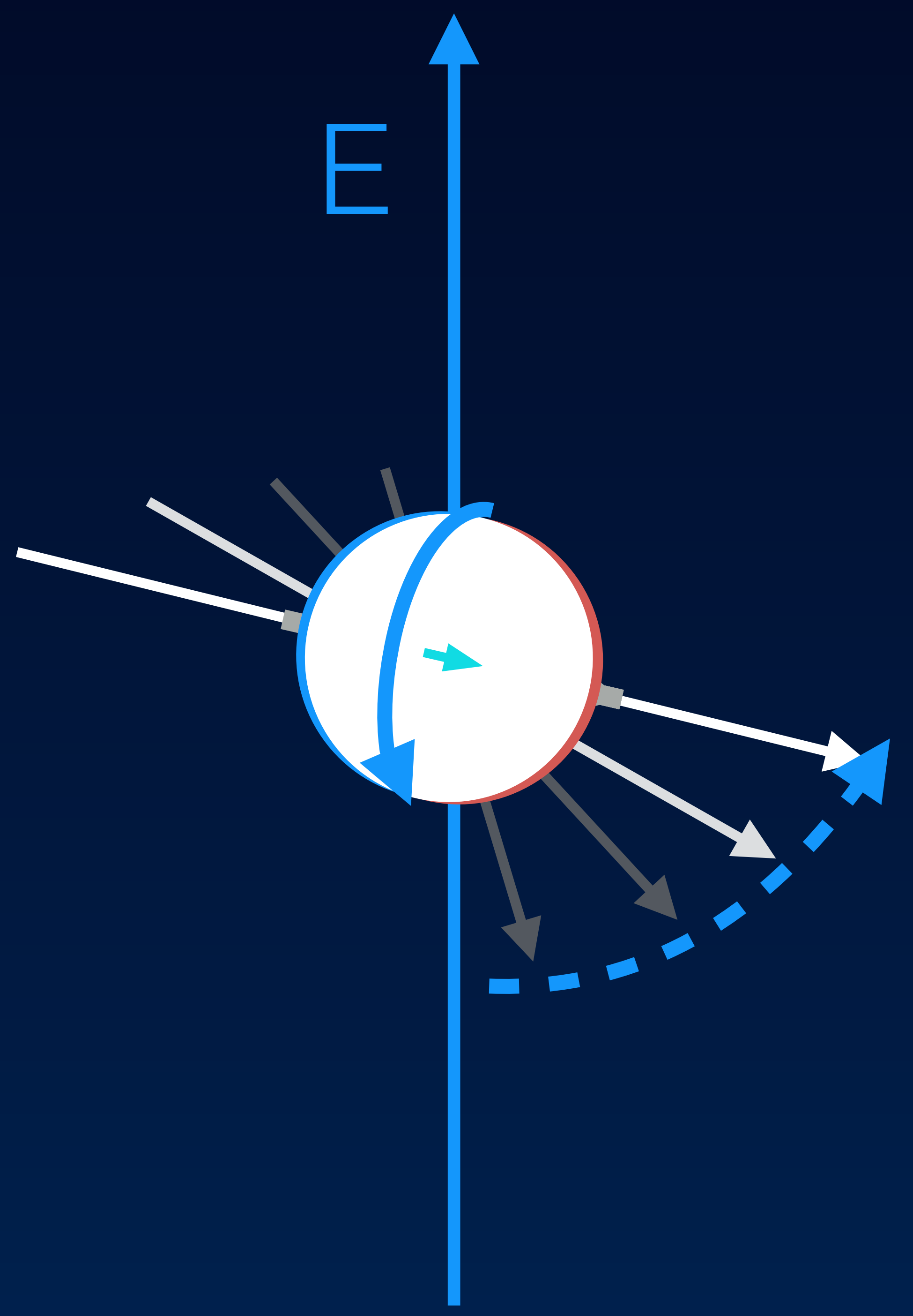
polar molecule

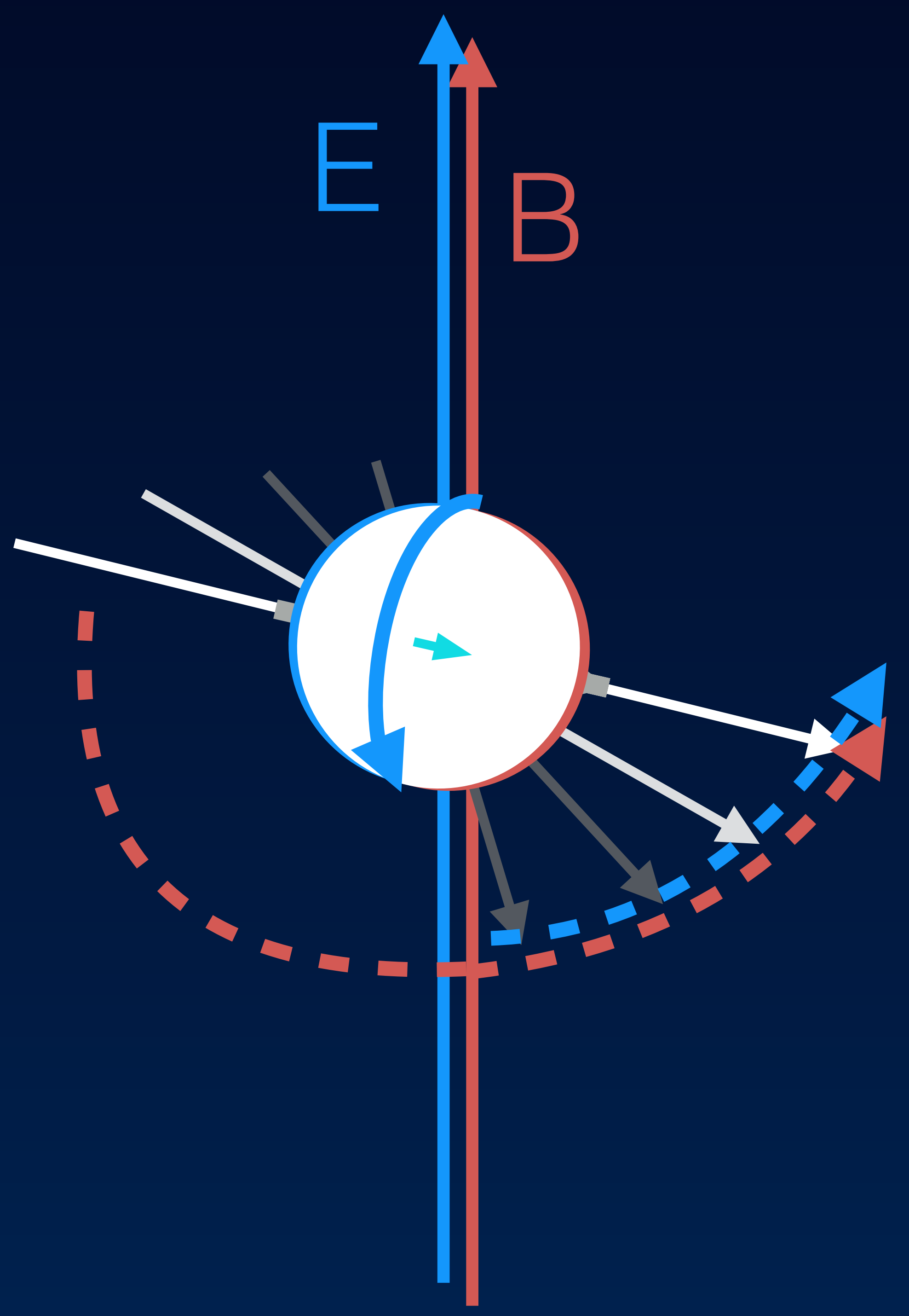


Solution:
use electron embedded in a polar molecule!

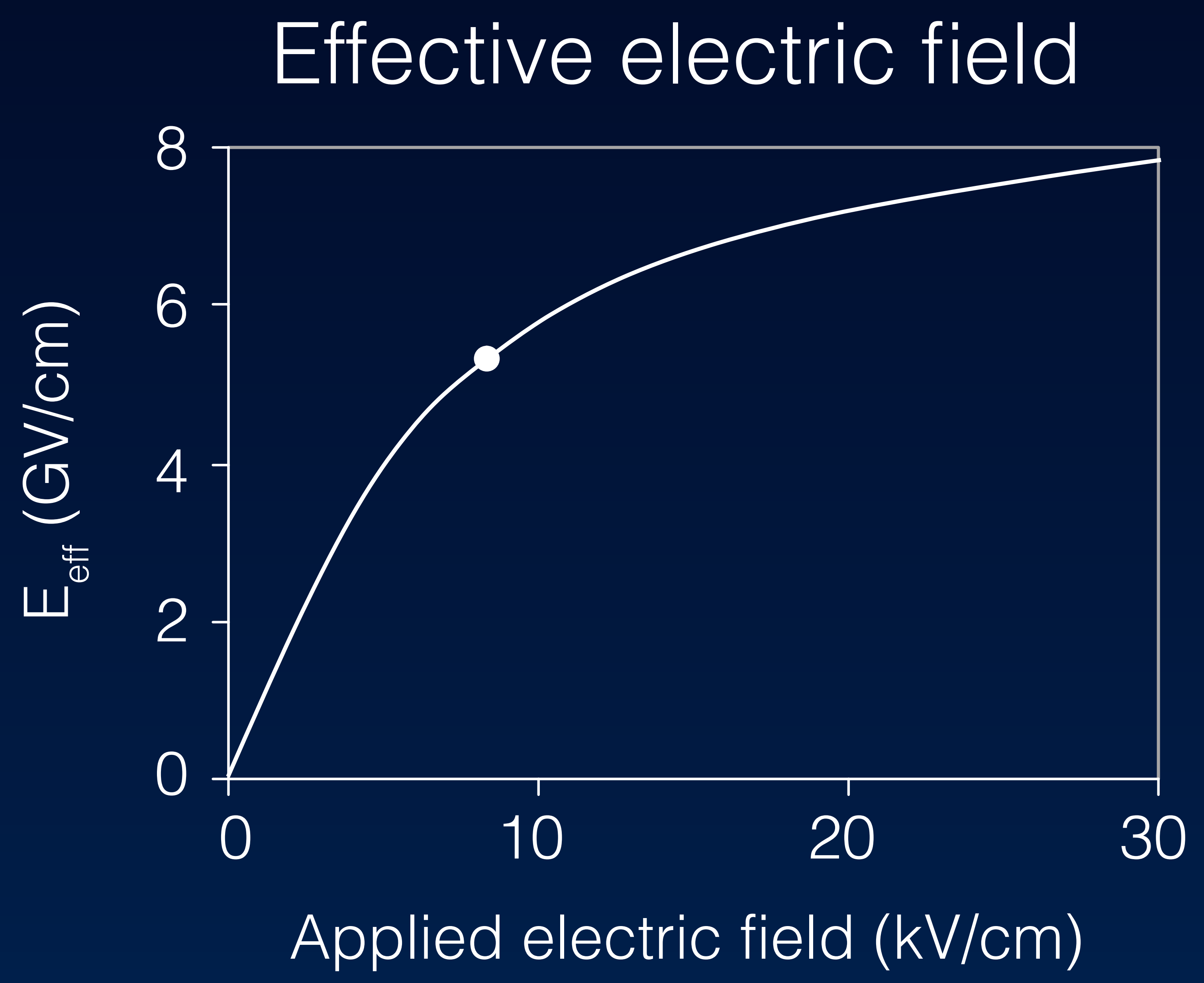
Enhances E
Shields B

How to measure a dipole moment?





polar
molecule



precession!

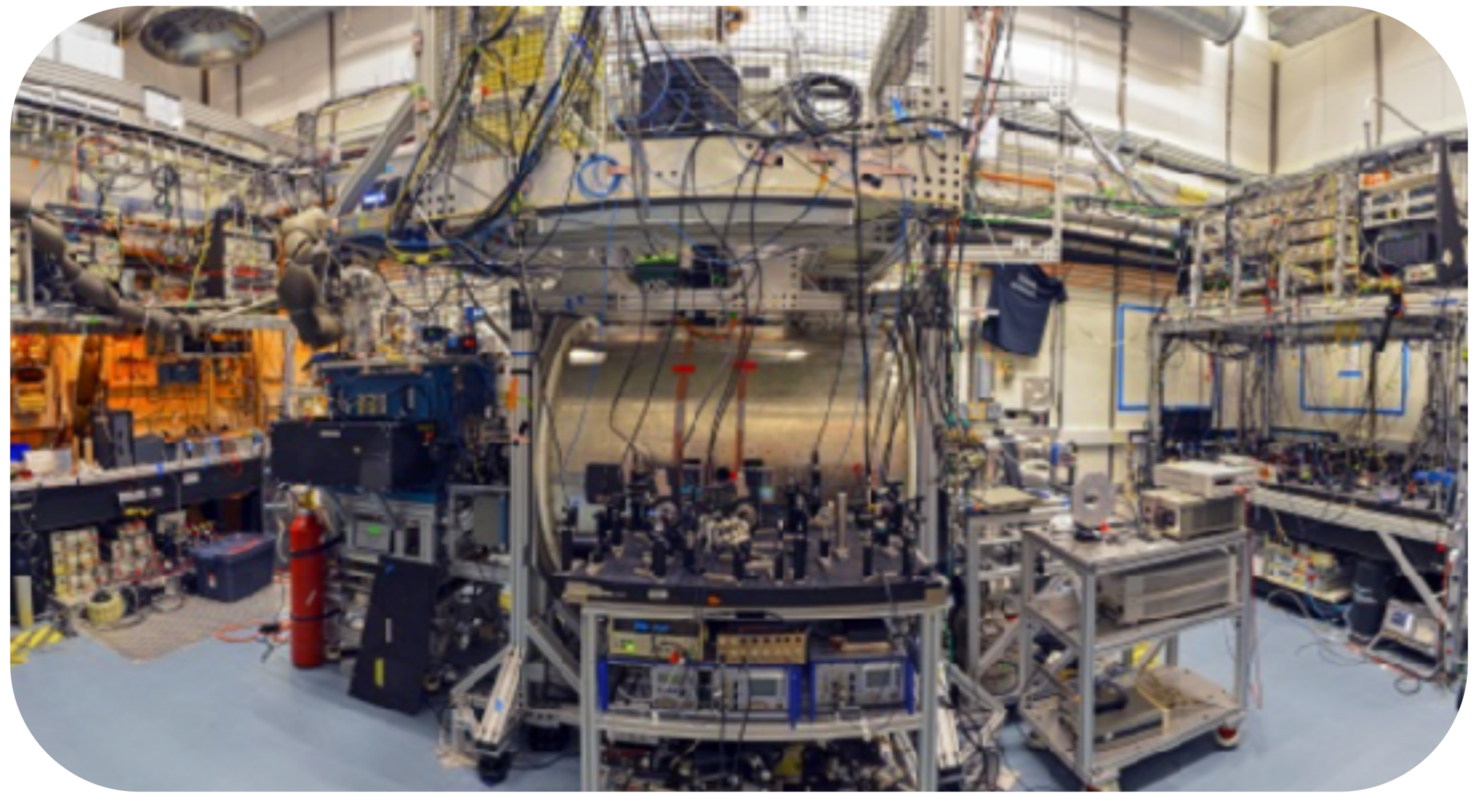
However, electron also has
magnetic dipole moment
(and charge!)

Solution:
use electron embedded
in a polar molecule!

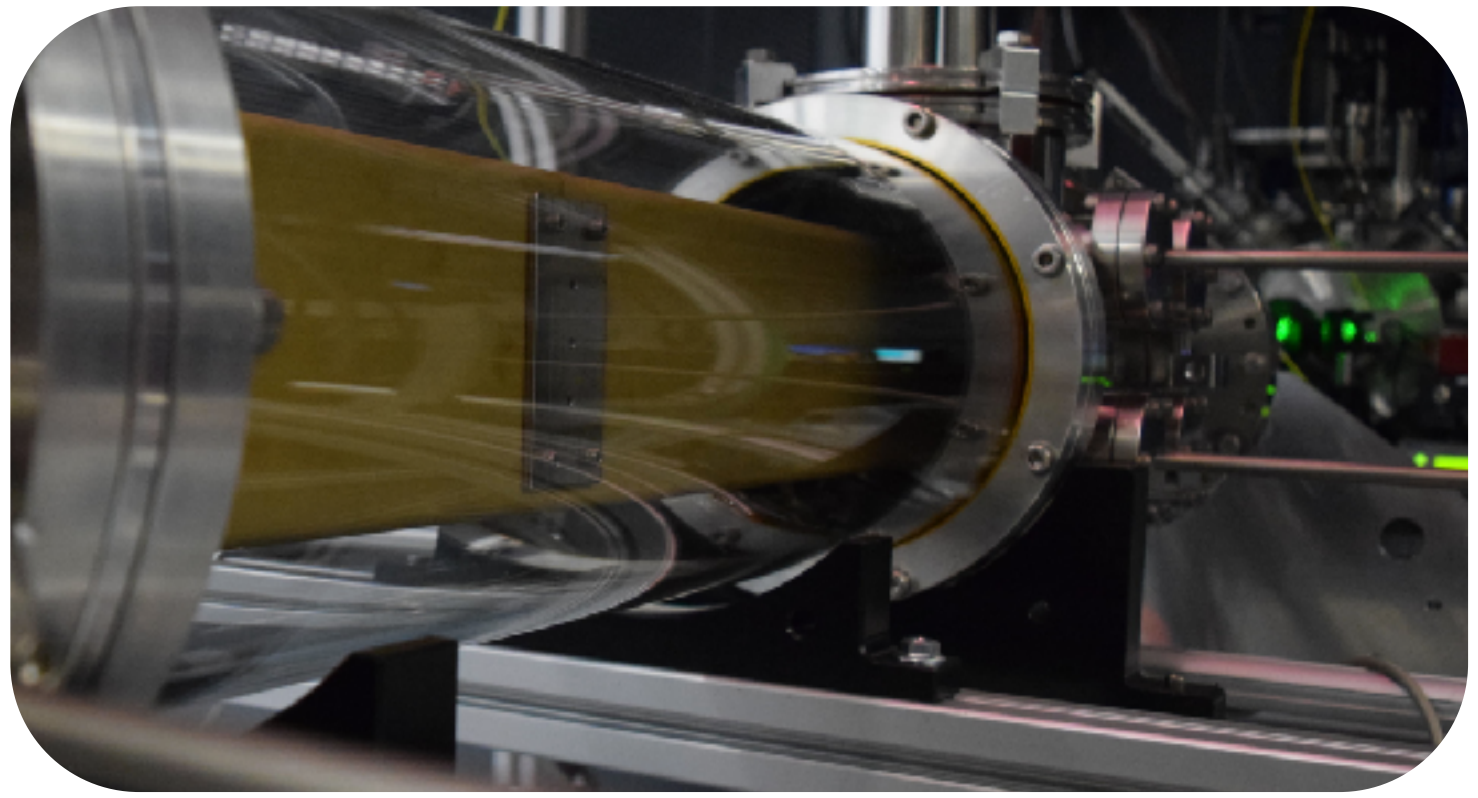
Enhances E
Shields B

General recipe: look for correlation with E-field reversal

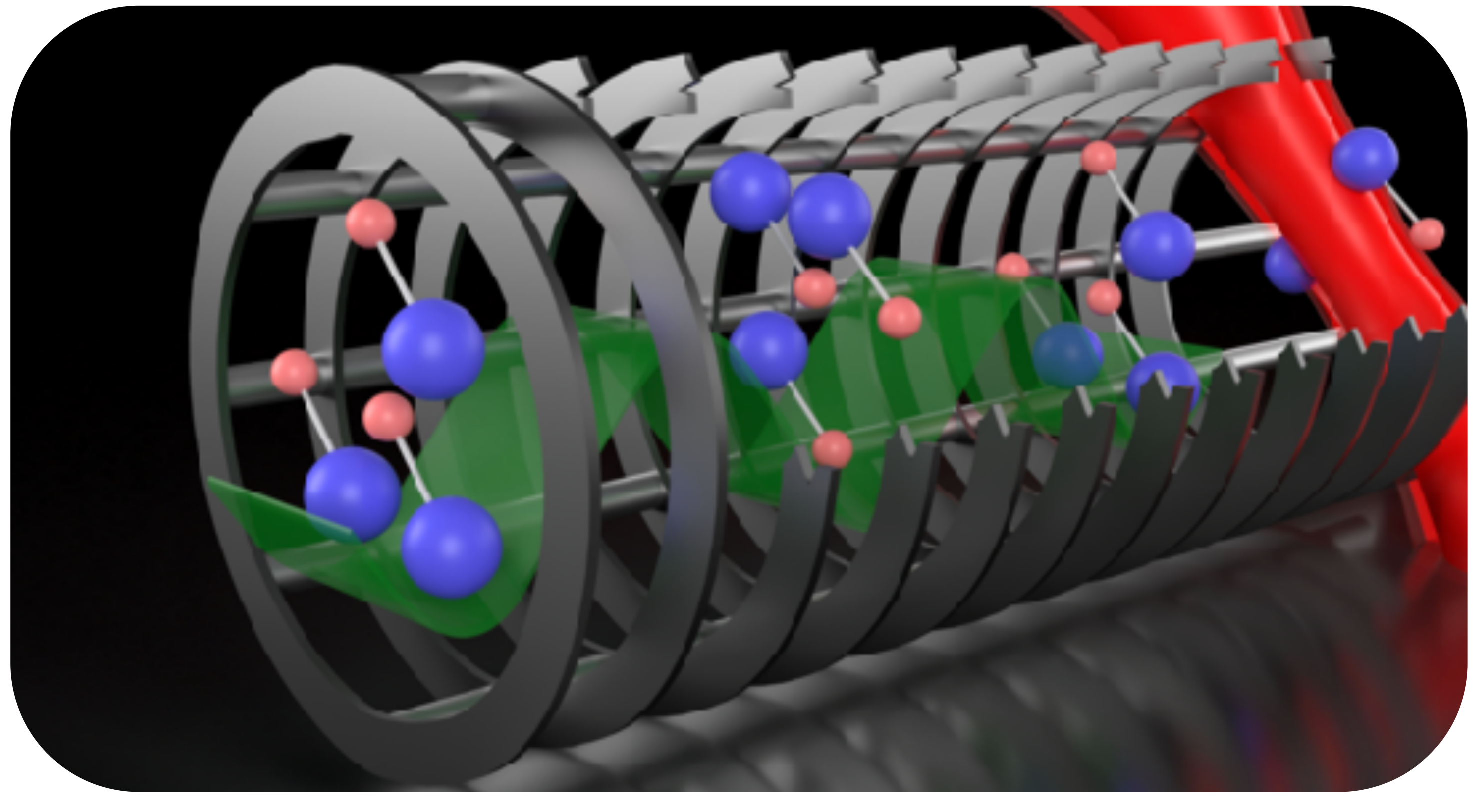
eEDM experiments using molecules



ACME - beam of ThO molecules
John Doyle, David DeMille,
Gerald Gabrielse

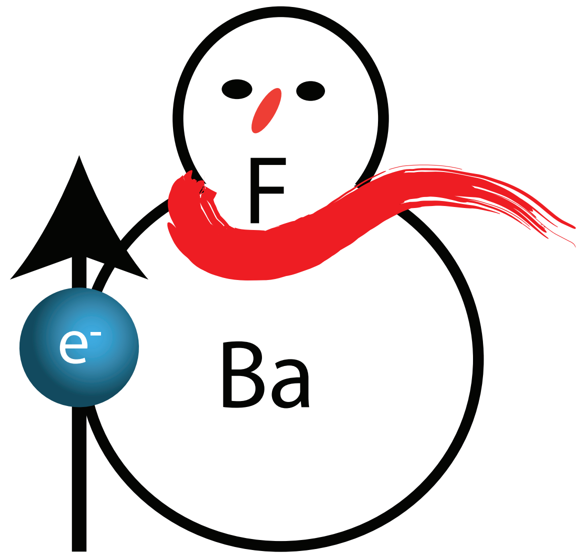


Imperial College London - beam of
YbF molecules
Mike Tarbutt, Ben Sauer, Ed Hinds



JILA - trapped HfF^+ ions
Eric Cornell, Jun Ye

Others are being set up:



Slow and cold BaF
The Netherlands
since 2018
(NL-eEDM)

PolyEDM
Polyatomic Electron EDM Search

Search for eEDM in cryogenic crystals
PHYDES:
Para-Hydrogen and Diatomic for eEDM Study
Università degli Studi di Ferrara
INFN
Istituto Nazionale di Fisica Nucleare

- York University
- Michigan State University
- University of Toronto

YORK UNIVERSITÉ UNIVERSITY
MICHIGAN STATE UNIVERSITY
UNIVERSITY OF TORONTO

EDM^3 Collaboration

Electric Dipole Measurements using molecules within a matrix

Statistical sensitivity for eEDM

$$\sigma = \frac{\hbar}{2|\langle\Omega\rangle|W_{\mathrm{d}}\tau\sqrt{N_{\mathrm{p}}}},$$

statistical error

state sensitivity

molecular sensitivity
(effective E-field)

Choice of molecule

total #
detected particles

coherent interaction time
of spin precession

Experimental approach

In addition to this,
control of systematic
effects is crucial!

Coherent interaction time

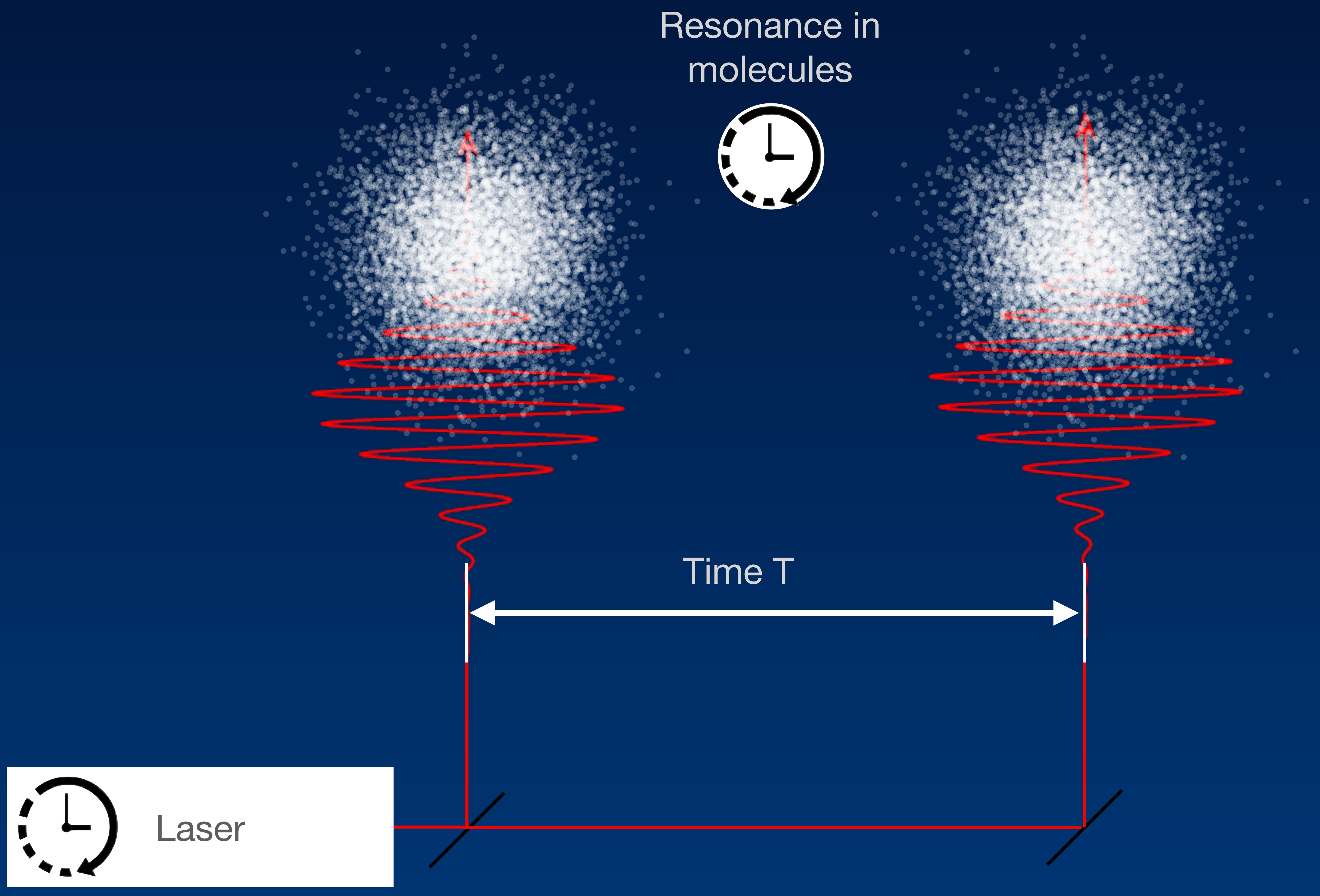
Key technique: Ramsey spin interferometer

laser pulse 1:

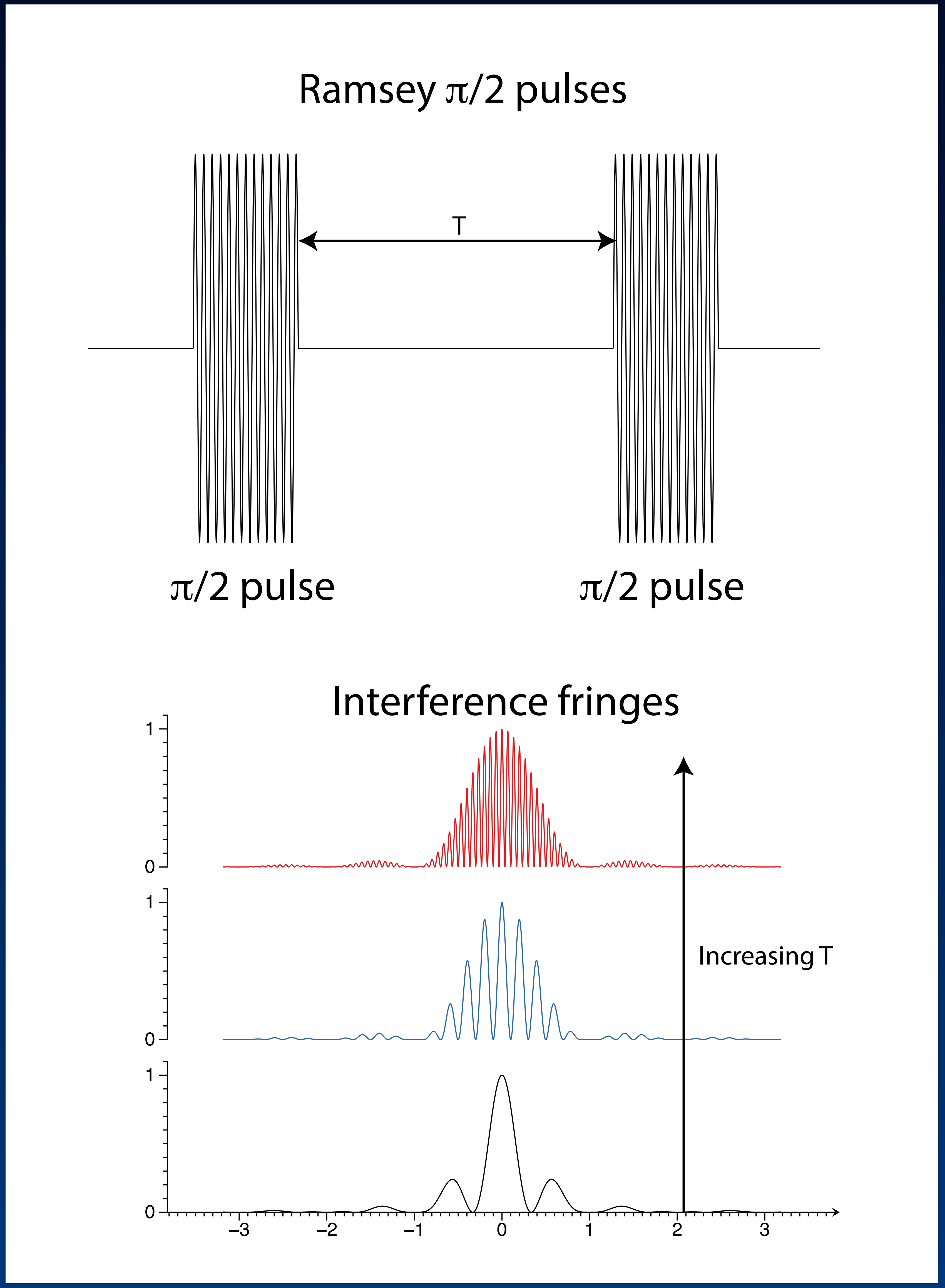
Creates a quantum superposition, creating coherent excitation of all molecules

laser pulse 2:

Measures state of the molecules through interference



Frequency set by external reference, tuned to molecular resonance



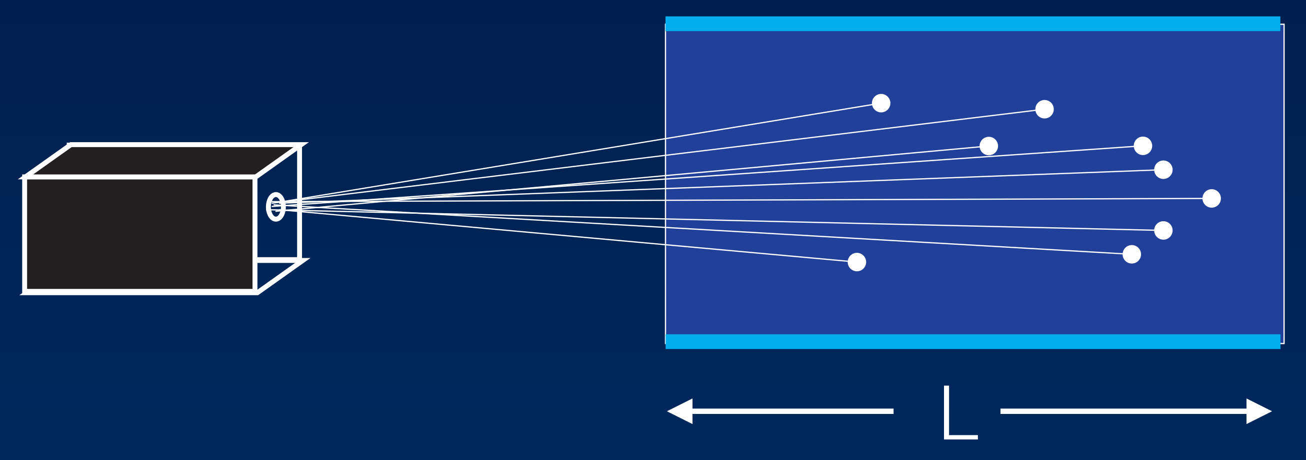
Towards longer coherent interaction times

fast beam

τ ~ 1-2 ms

L ~ 0.5 m

v ~ 250-500 m/s



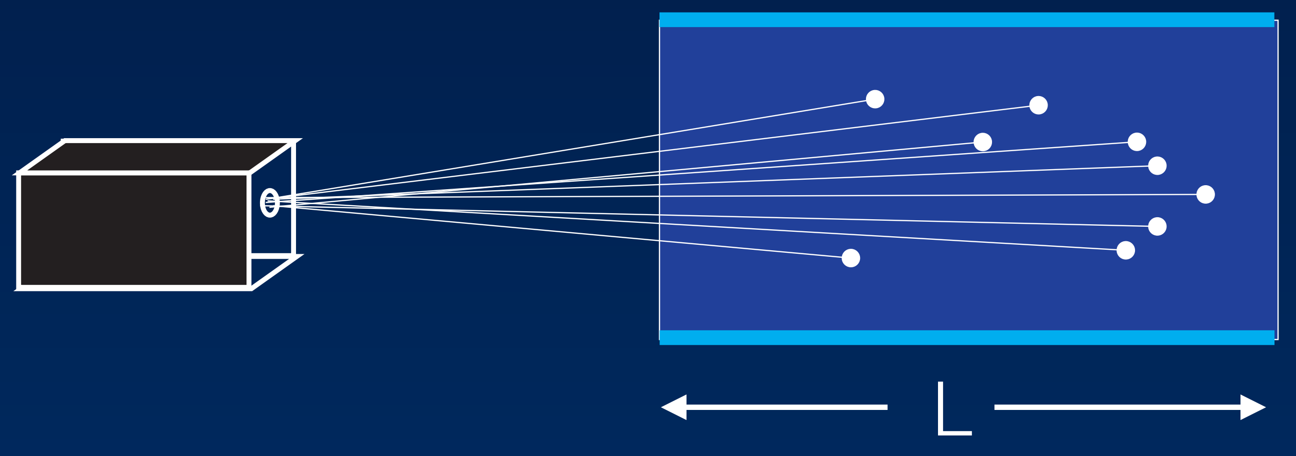
Towards longer coherent interaction times

fast beam

τ ~ 1-2 ms

L ~ 0.5 m

v ~ 250-500 m/s

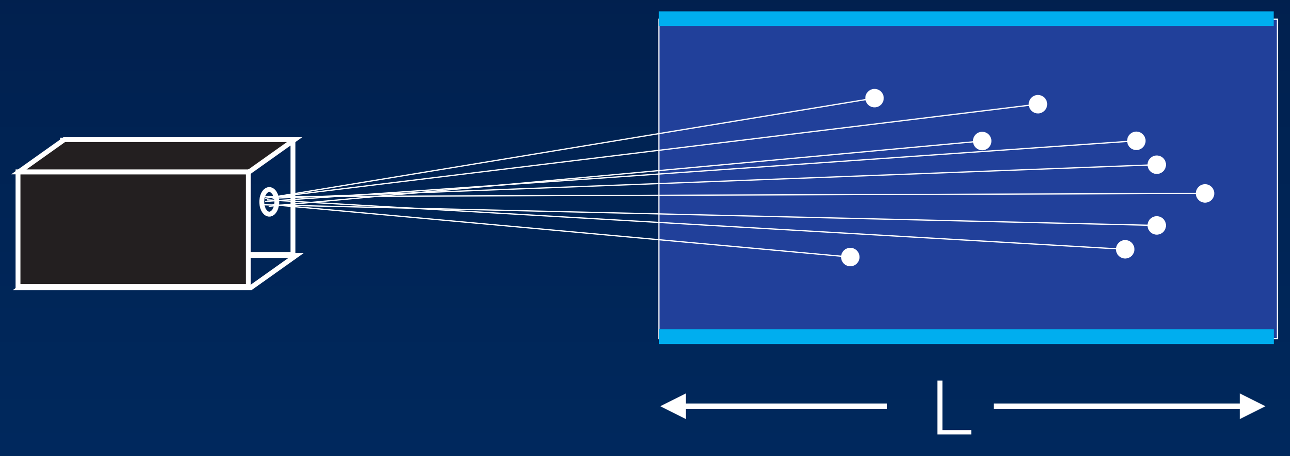


slow beam

τ ~ 15 ms

L ~ 0.5 m

v ~ 30 m/s



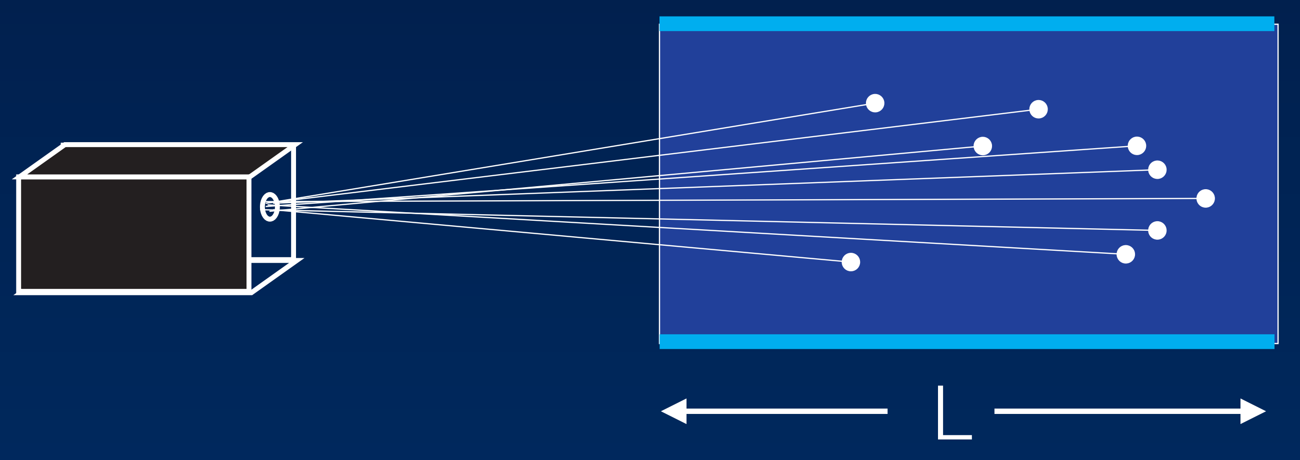
Towards longer coherent interaction times

fast beam

τ ~ 1-2 ms

L ~ 0.5 m

v ~ 250-500 m/s

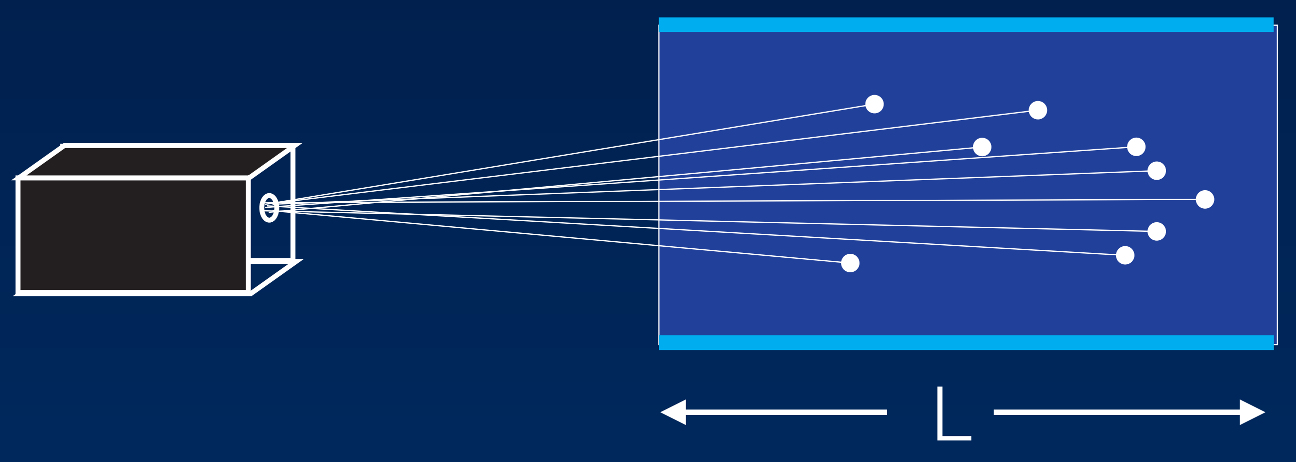


slow beam

τ ~ 15 ms

L ~ 0.5 m

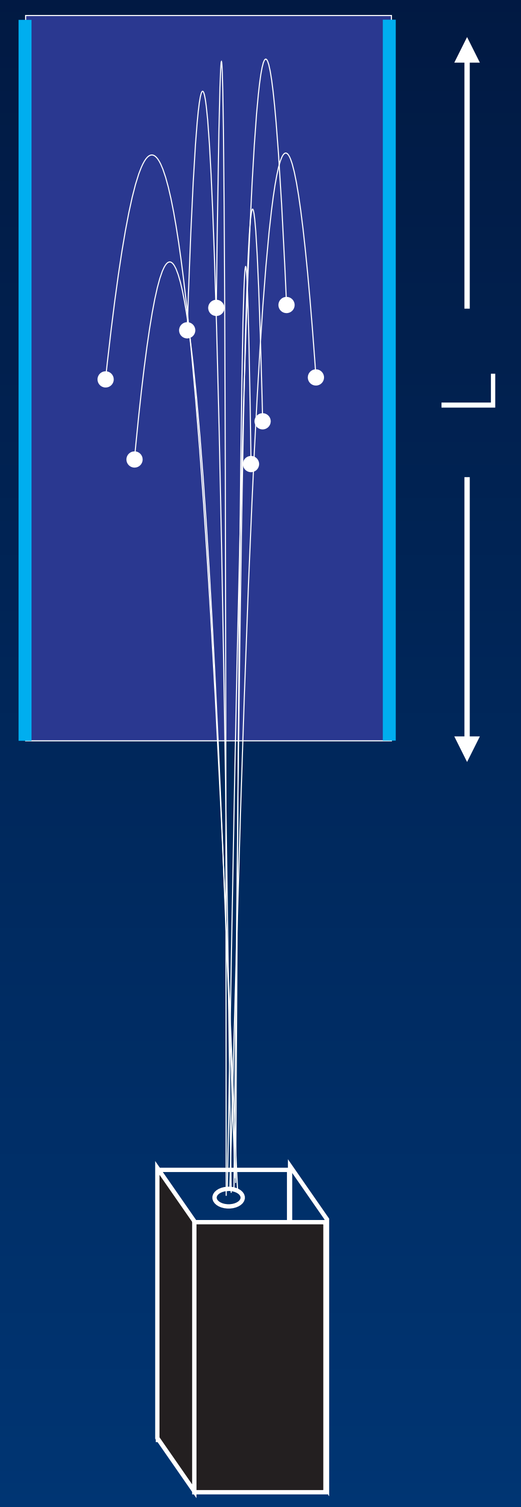
v ~ 30 m/s



fountain

τ ~ 100 ms

L ~ 0.5 m

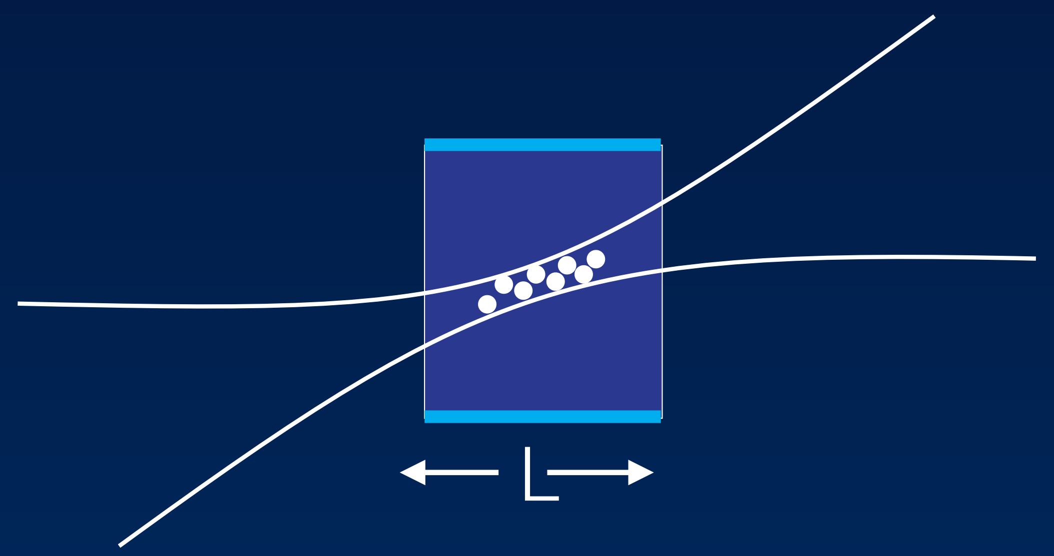


slow vertical beam

trap

τ ~ 1-10 s

L ~ 0.5 mm



molecules trapped in laser focus

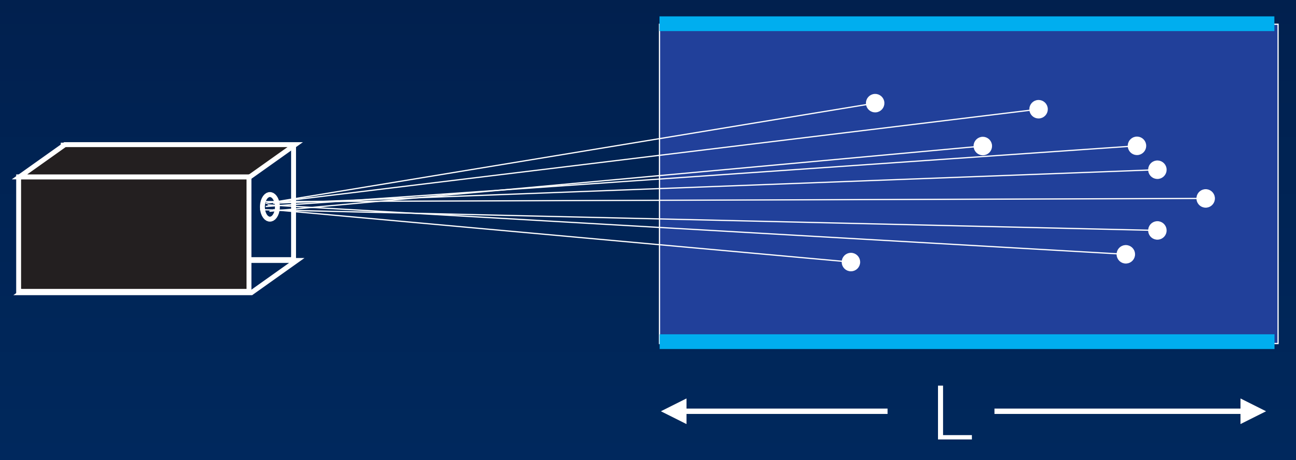
Towards longer coherent interaction times

fast beam

τ ~ 1-2 ms

L ~ 0.5 m

v ~ 250-500 m/s

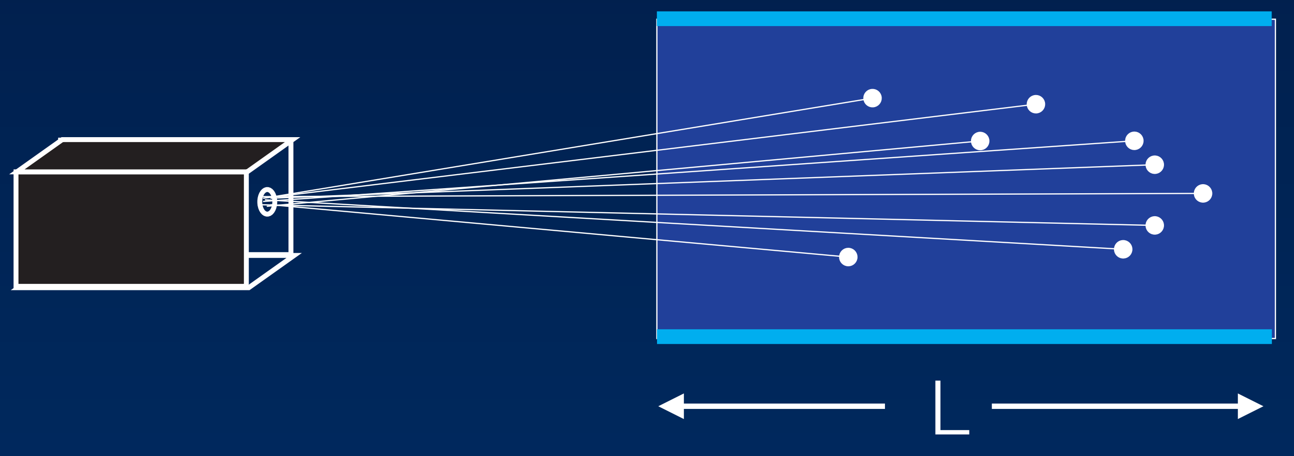


slow beam

τ ~ 15 ms

L ~ 0.5 m

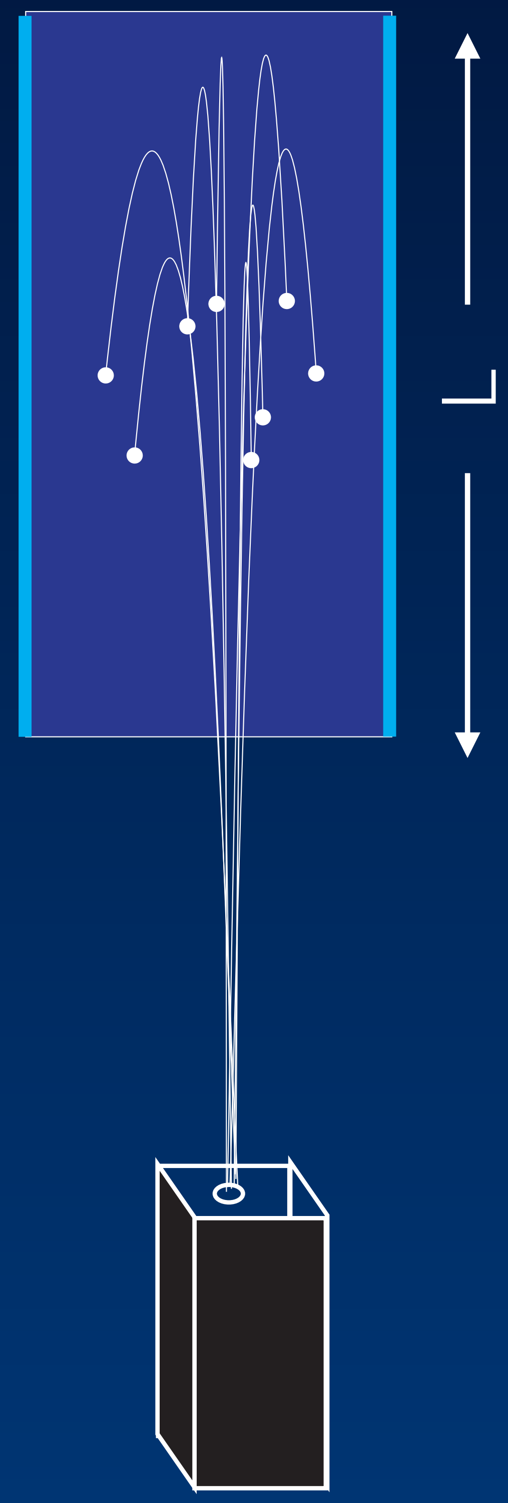
v ~ 30 m/s



fountain

τ ~ 100 ms

L ~ 0.5 m

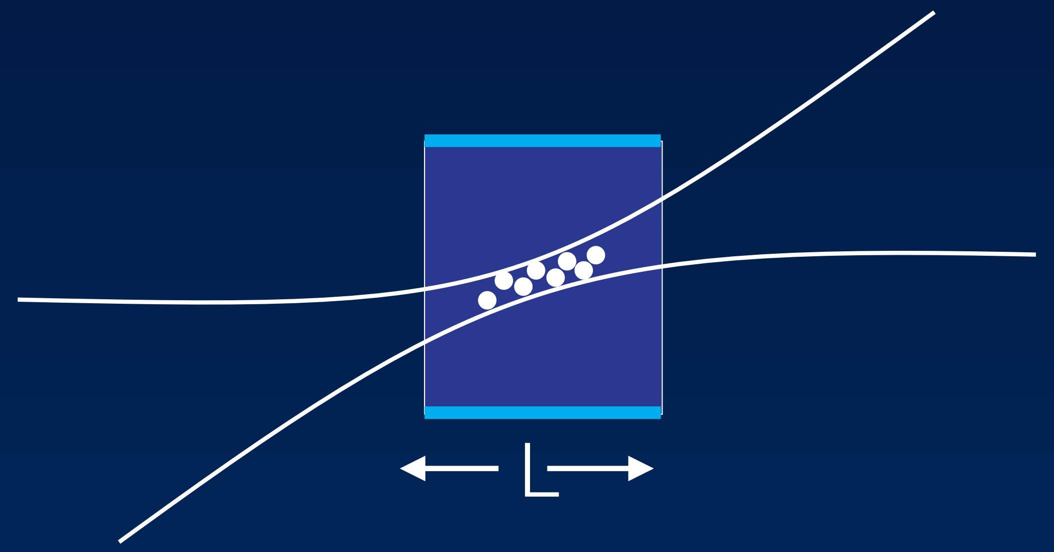


slow vertical beam

trap

τ ~ 1-10 s

L ~ 0.5 mm



molecules trapped in laser focus

Main challenge:
how to maintain N while increasing τ

Strongly connected to choice of molecule!

Statistical sensitivity for eEDM

$$\sigma = \frac{\hbar}{2|\langle\Omega\rangle|W_{\mathrm{d}}\tau\sqrt{N_{\mathrm{p}}}},$$

statistical error

state sensitivity

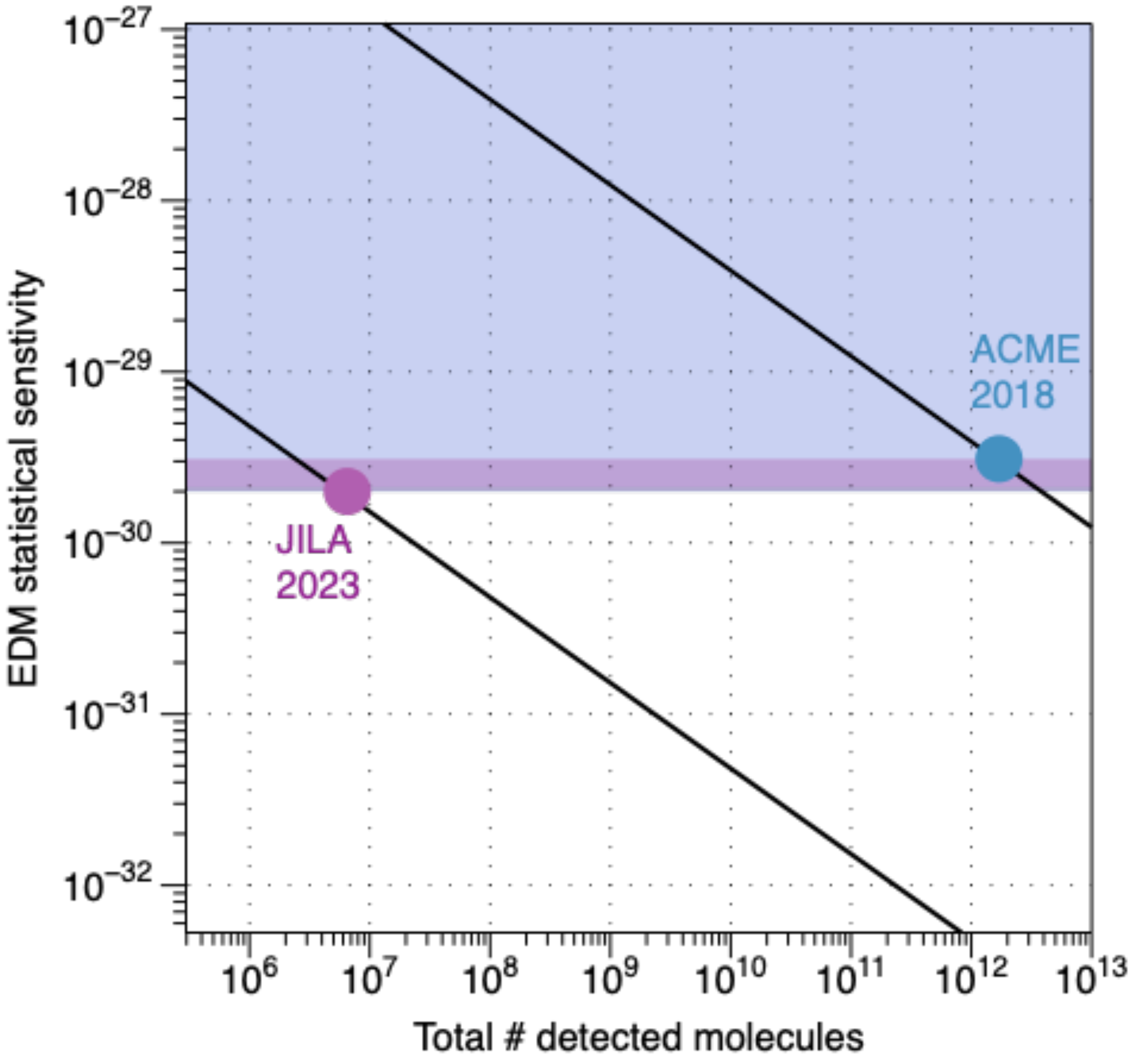
molecular sensitivity
(effective E-field)

Choice of molecule

total #
detected particles

coherent interaction time
of spin precession

Experimental approach



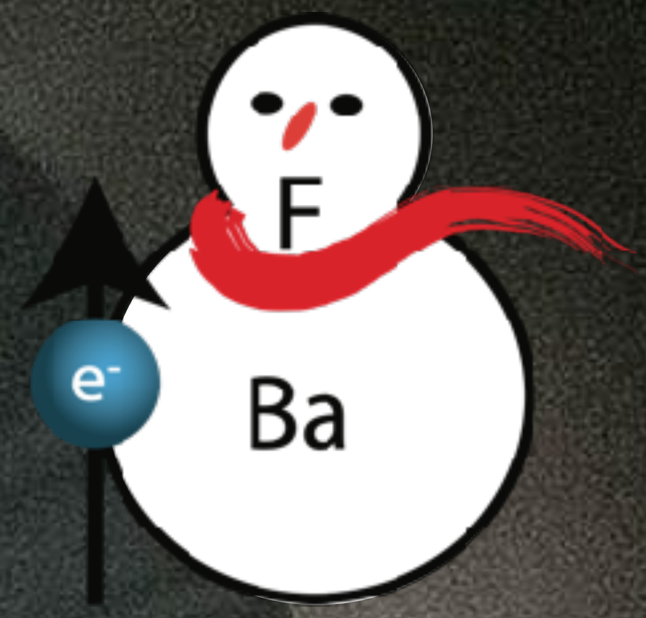
Community input to the European Strategy on particle physics: Searches for Permanent Electric Dipole Moments

edited by M. Athanasakis-Kaklamanakis, M. Au, R. Berger, S. Degenkolb, J. De Vries, S. Hoekstra, A. Keshavarzi, D. Ries, P. Schmidt-Wellenburg, and M. Tarbutt,

Document in preparation
soon open for community input

Name	Species	Method	$\lvert E_{\mathrm{eff}}\rvert$ (GV/cm)	T (ms)	N (approx.)	Status	90% CL (10^{-29} e cm)	Ref
Imperial I	YbF ($^2\Sigma$)	Beam	14.5	0.64	10^{11}	Complete	105	[60]
ACME I	ThO ($^3\Delta_1$)	Beam	78	1.1	4×10^{10}	Complete	9.4	[61]
ACME II	ThO ($^3\Delta_1$)	Beam	78	1.1	10^{13}	Complete	1.1	[4]
JILA I	HfF^+ ($^3\Delta_1$)	Ion trap	23	700	3×10^6	Complete	13	[62]
JILA II	HfF^+ ($^3\Delta_1$)	Ion trap	23	3000	10^8	Complete	0.41	[5]
ACME III	ThO ($^3\Delta_1$)	Beam	78	5	8×10^{14}	Commissioning		[63]
JILA III	ThF^+ ($^3\Delta_1$)	Ion trap	36	20000	10^7	Commissioning		[64]
Imperial II	YbF ($^2\Sigma$)	μK beam	18	20	10^{13}	Commissioning		[65]
Imperial III	YbF ($^2\Sigma$)	Lattice	18	3000	10^{10}	Construction		[65]
NL-eEDM I	BaF ($^2\Sigma$)	Slow beam	5	15	10^{13}	Commissioning		[66]
NL-eEDM II	BaOH ($^2\Sigma$)	Lattice	5	1000	10^{10}	Construction		[67]
PolyEDM	SrOH ($^2\Sigma$)	Lattice	2.2	1000	10^{10}	Construction		[68]
EDM^3	BaF ($^2\Sigma$)	Matrix	6	100	10^{20}	Construction		[69]
PHYDES	BaF ($^2\Sigma$)	Matrix	6	100	10^{20}	Construction		[70]

The NL-eEDM team



Particle physics theory	Quantum chemistry	Experiments	
Jordy de Vries Heleen Mulder **Rob Timmermans**	**Anastasia Borschevsky** Lukas Pastecka Agustin Aucar Yuly Chamorro Eiffion Prinsen	**Steven Hoekstra** **Lorenz Willmann** **Rick Bethlem** **Steve Jones** **Wim Ubachs** Roman Bause Lucas van Sloten Jelmer Levenga	Joost van Hofslot Maarten Mooij Ginny Marshall Anno Touwen Bart Schellenberg Ties Fikkers Nithesh Balasubramanian

university of groningen

VU VRIJE UNIVERSITEIT AMSTERDAM

Nikhef

Dutch National Institute for (astro)Particle Physics

UvA

phased approach

F
Ba
e⁻

university of groningen
van swinderen institute for particle physics and gravity

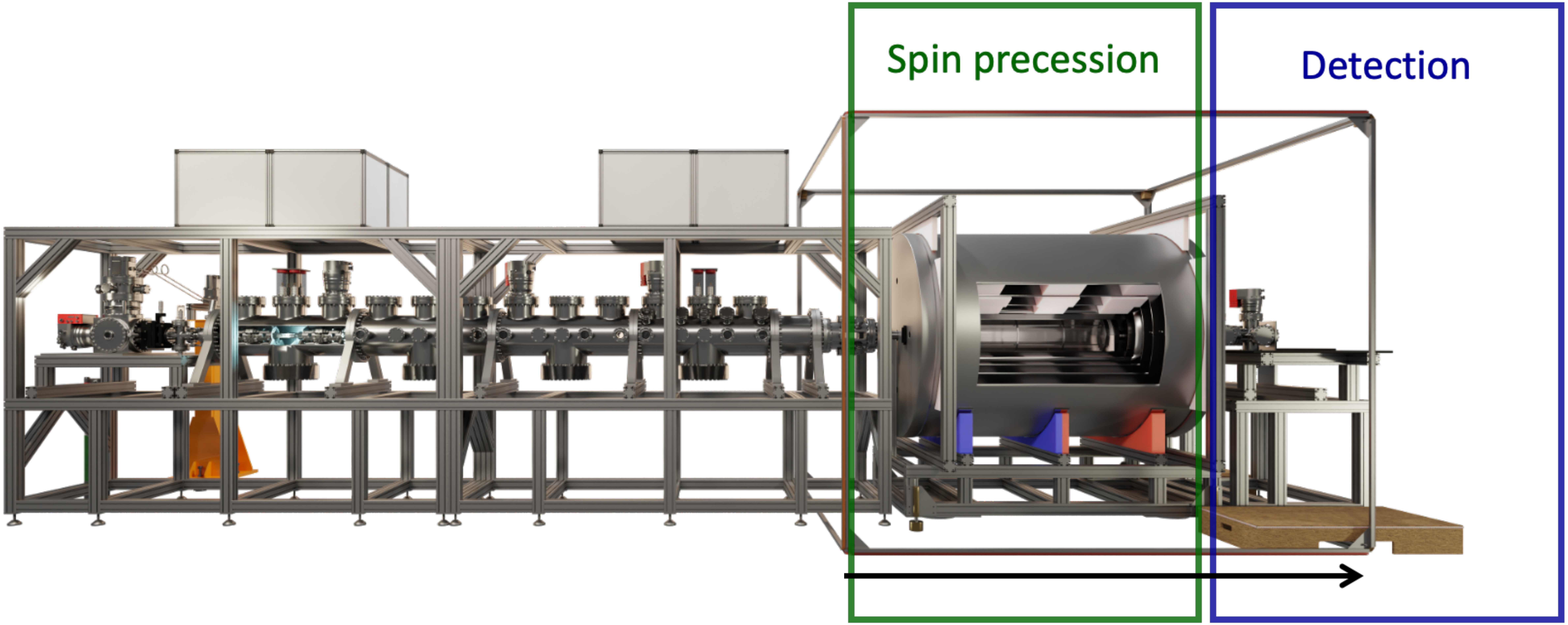
Nikhef
Dutch National Institute for (astro)Particle Physics

VU VRIJE UNIVERSITEIT AMSTERDAM

UvA

Key ingredients

Key ingredients of our approach



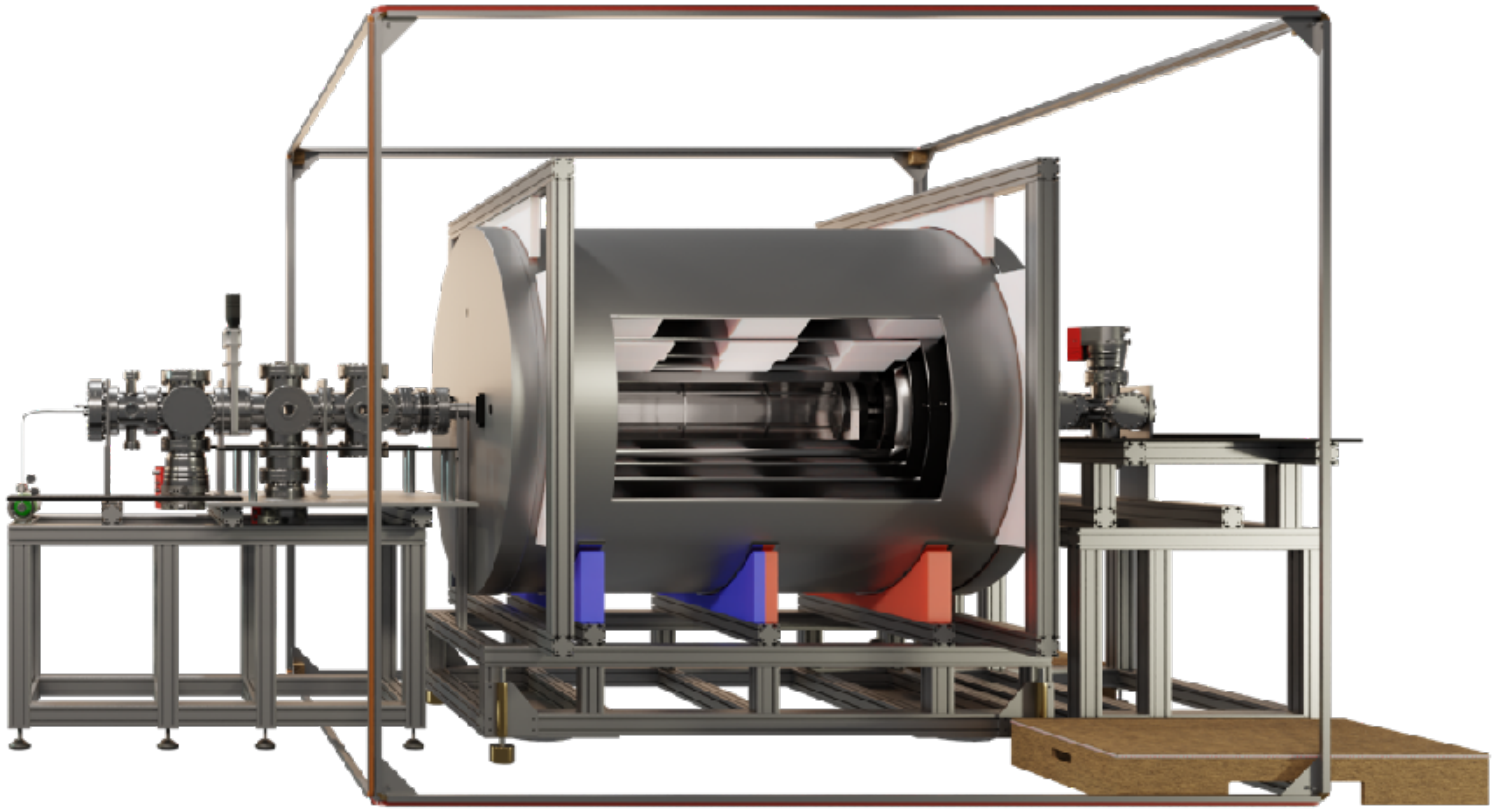
Phase 1: Fast beam

Supersonic beam (600 m/s)

Controlled field environment

Explore molecular structure

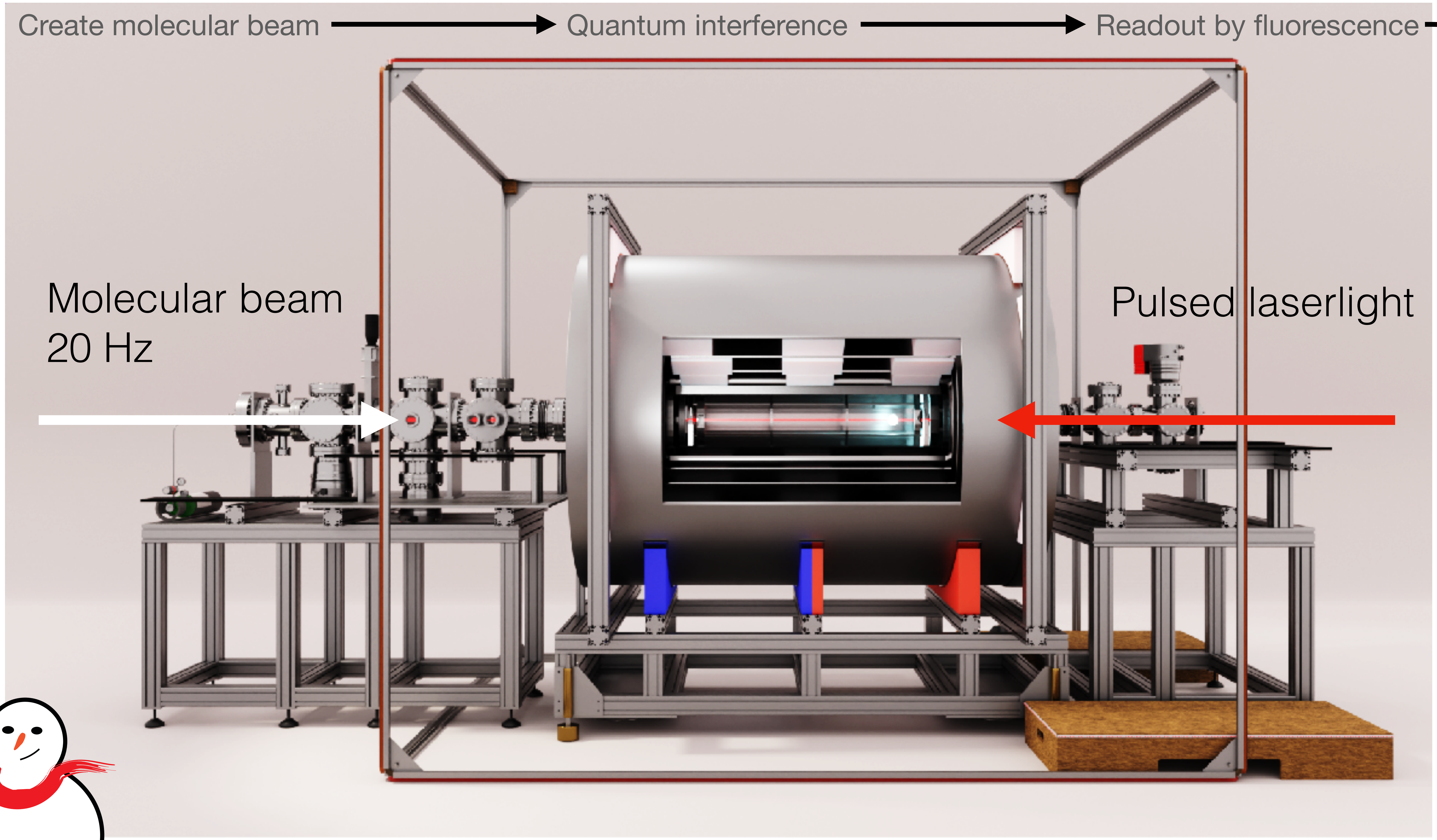
Spin interferometer measurement



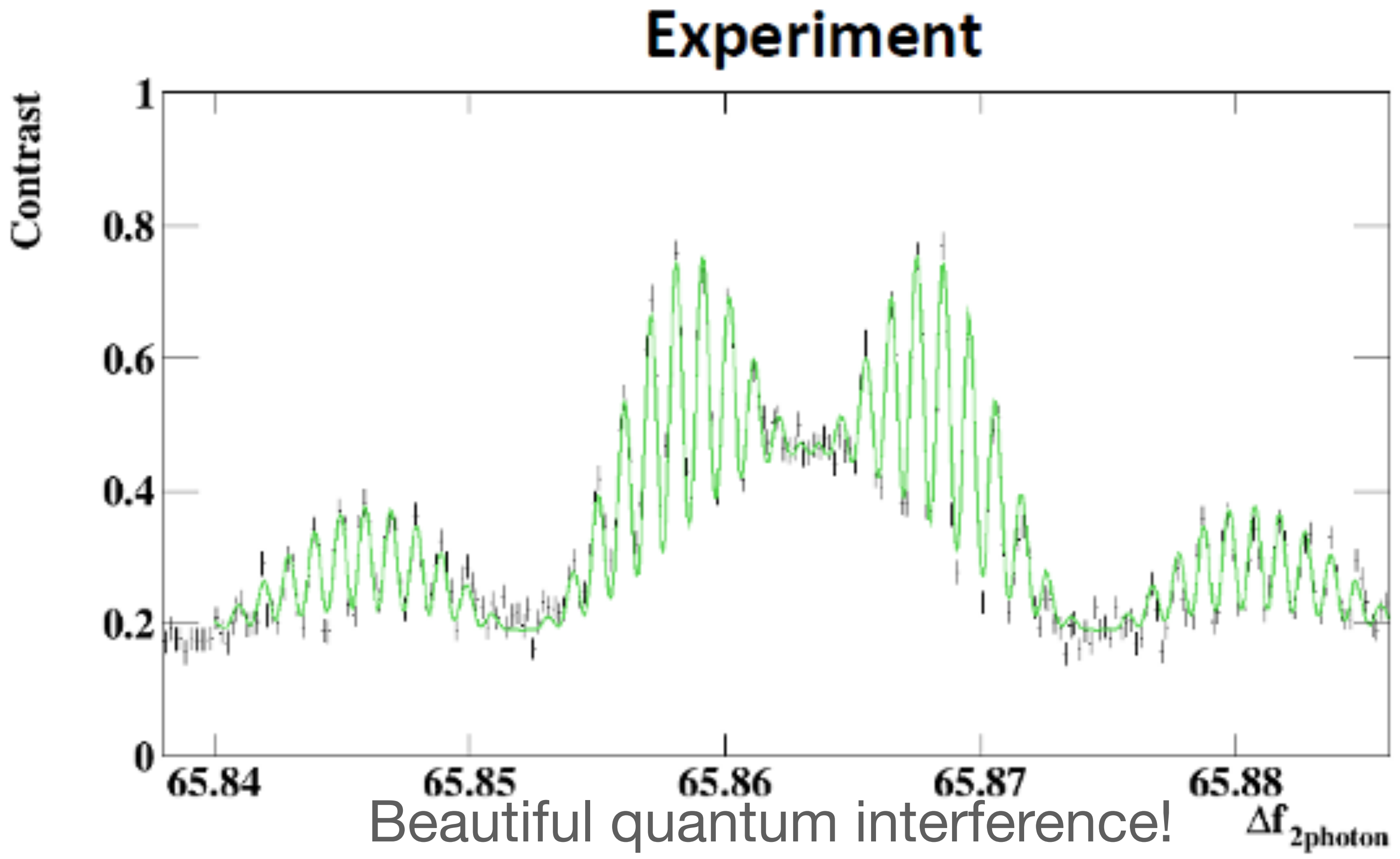
Understand systematics

Interference data using fast molecular beam

to demonstrate control over systematic effects



Compare to theory that includes the full interaction of the molecule with light, electric and magnetic fields (optical Bloch equations)



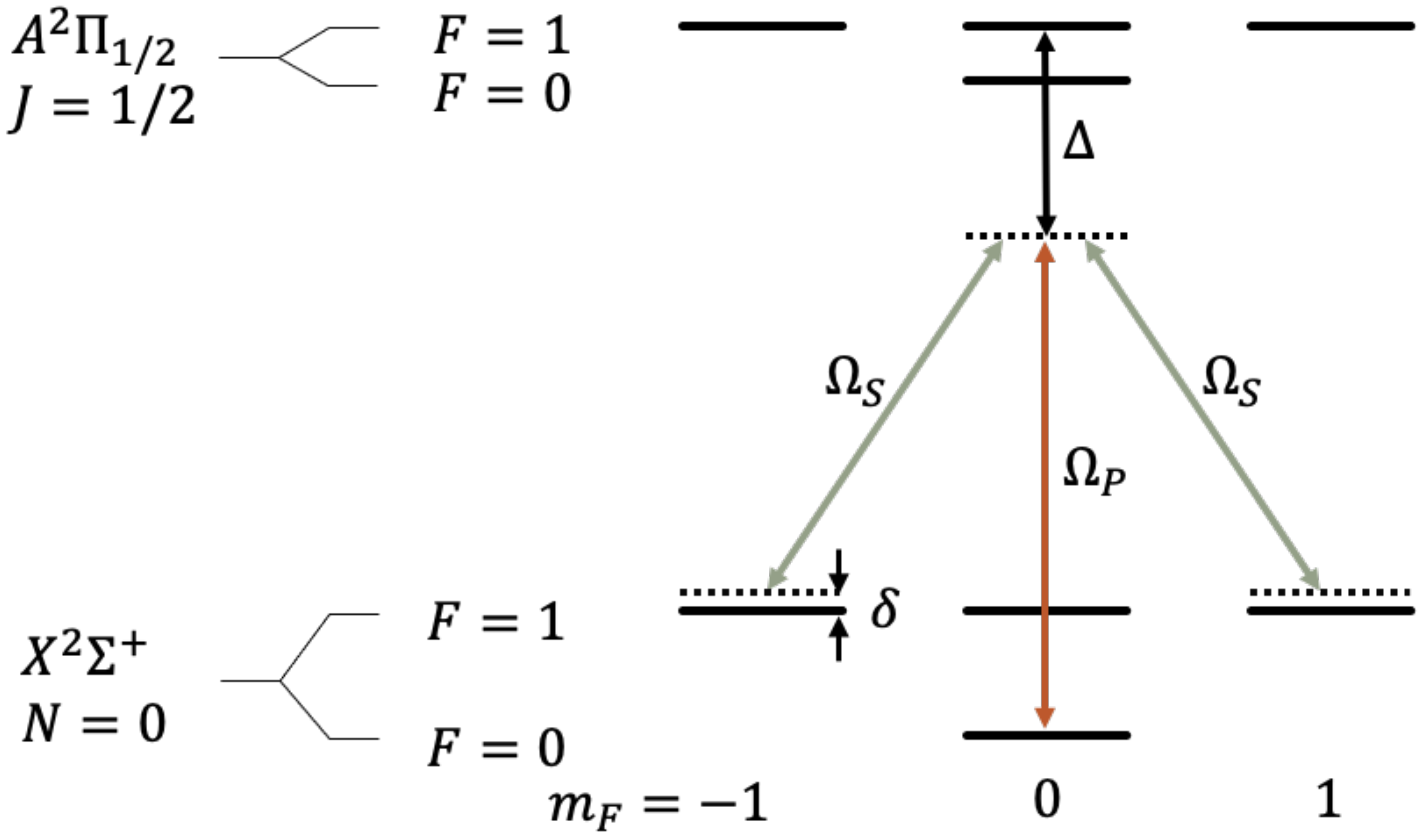
Beautiful quantum interference!

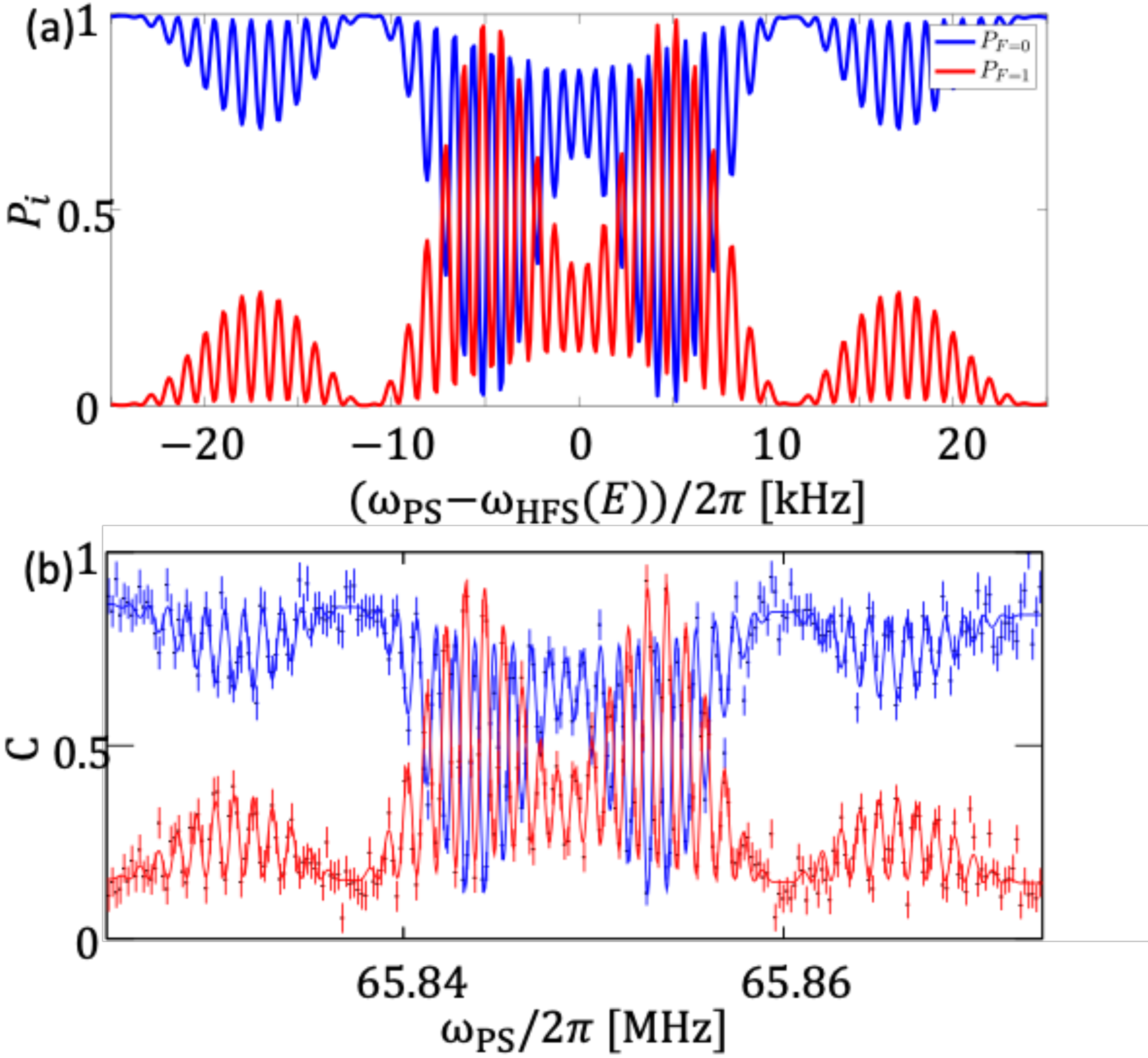
Contains all relevant experimental parameters
Crucial for reduction of systematic effects
*(A.Boeschoten et al, NL-eEDM collaboration, PRA **110** L010801 (2024))*

NL-eEDM

Experiment and theory

Optical Bloch equations

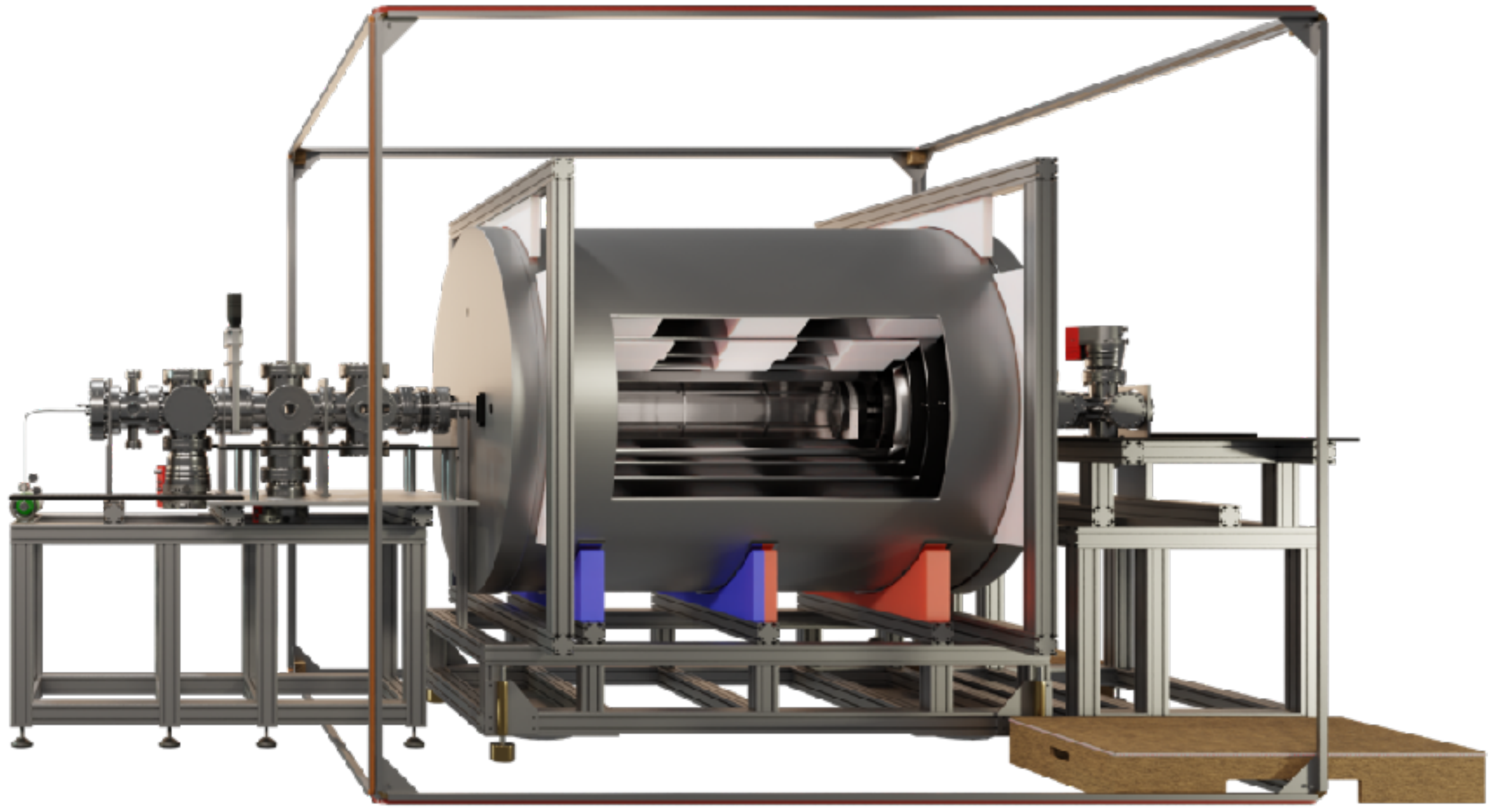




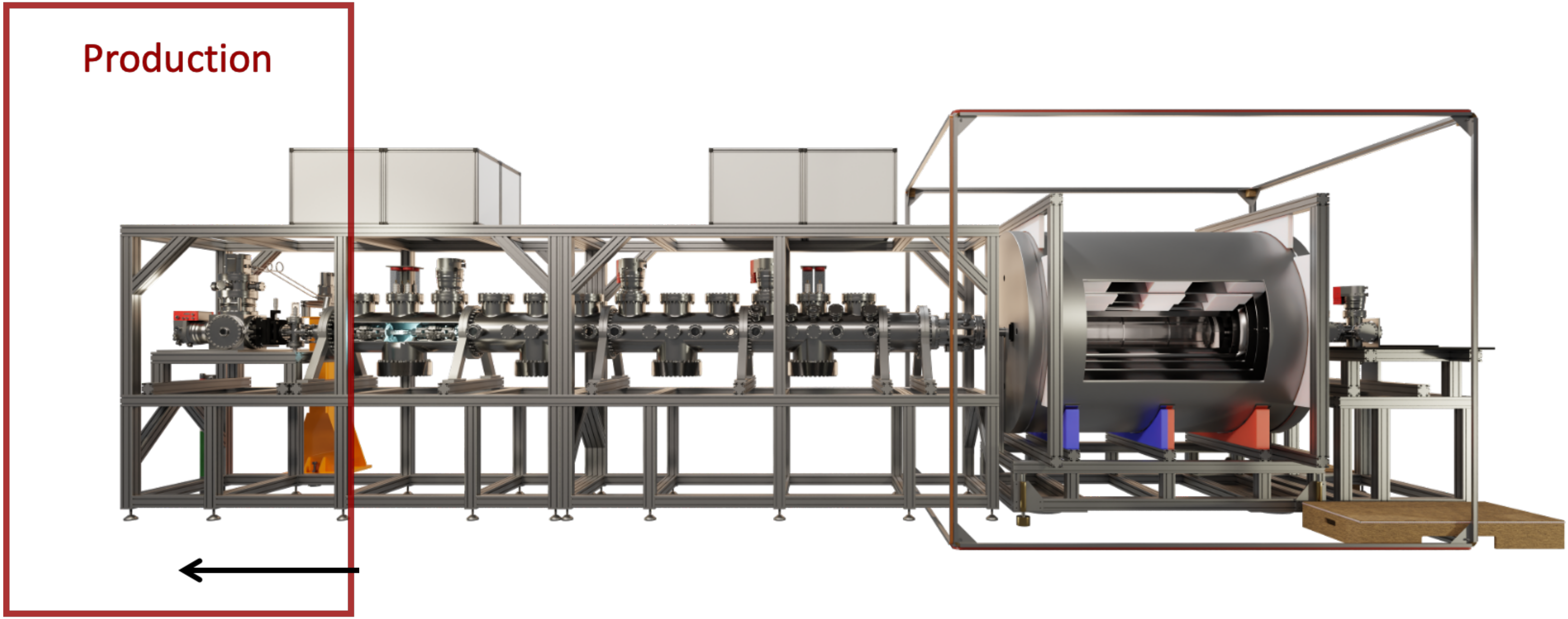
Current status

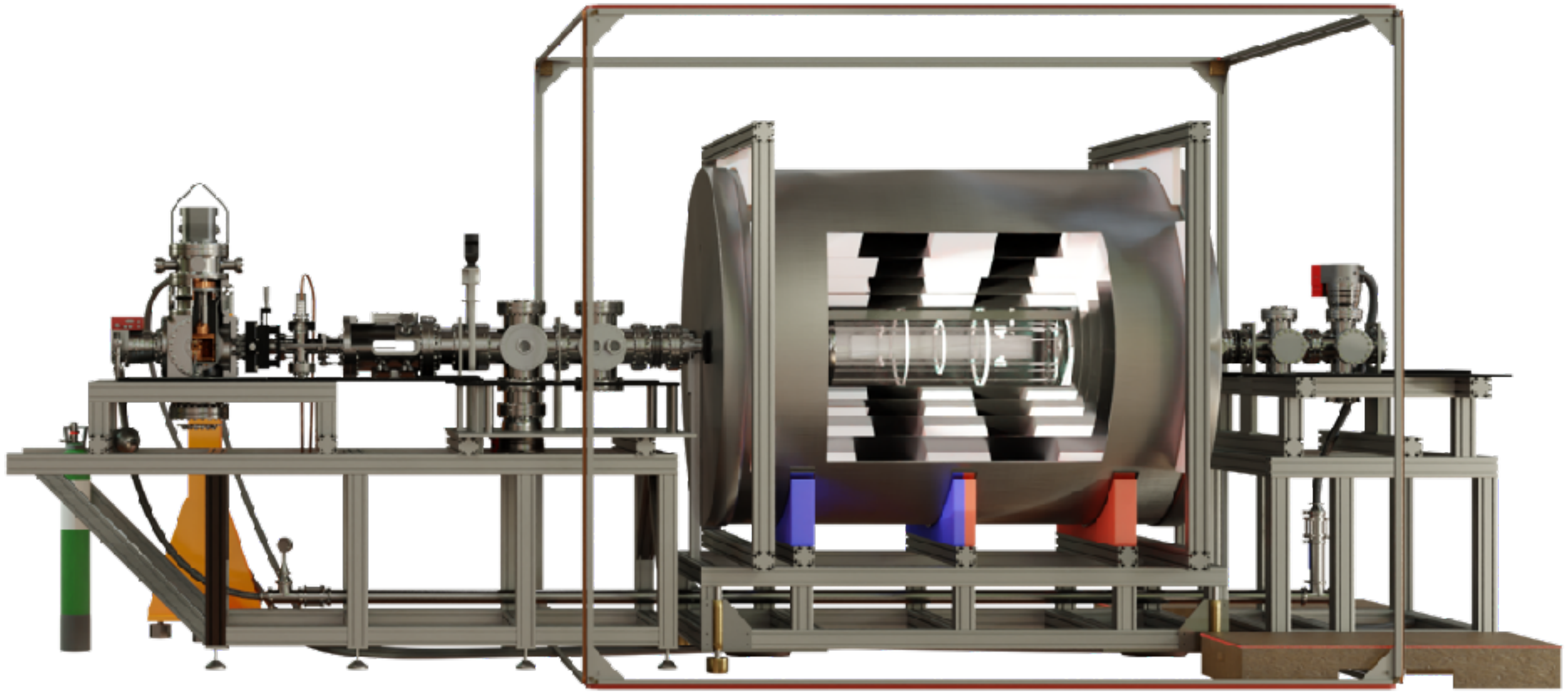
Phase 1: Fast beam

- Construction completed
  - source, lasers, magnetic shielding, DAQ, interference fringes
- Routinely taking data - and recently moved to new lab….
- Analysing for eEDM limit (expect at ~YbF level)



Key ingredients of our approach





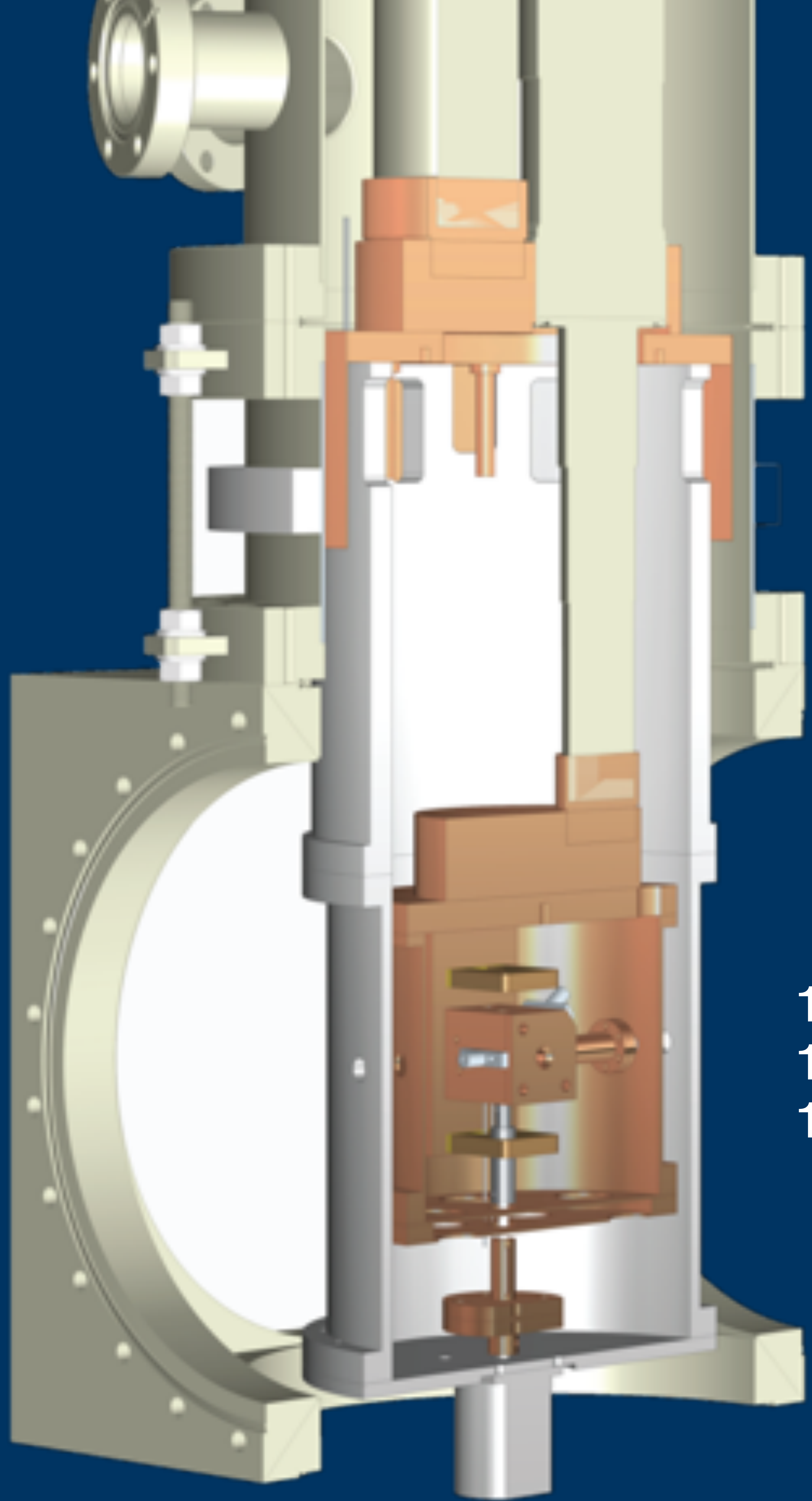
Increase statistics

Phase 2: Slow beam

Cryogenic beam (150 m/s)

Hexapole focussing

Transverse laser cooling

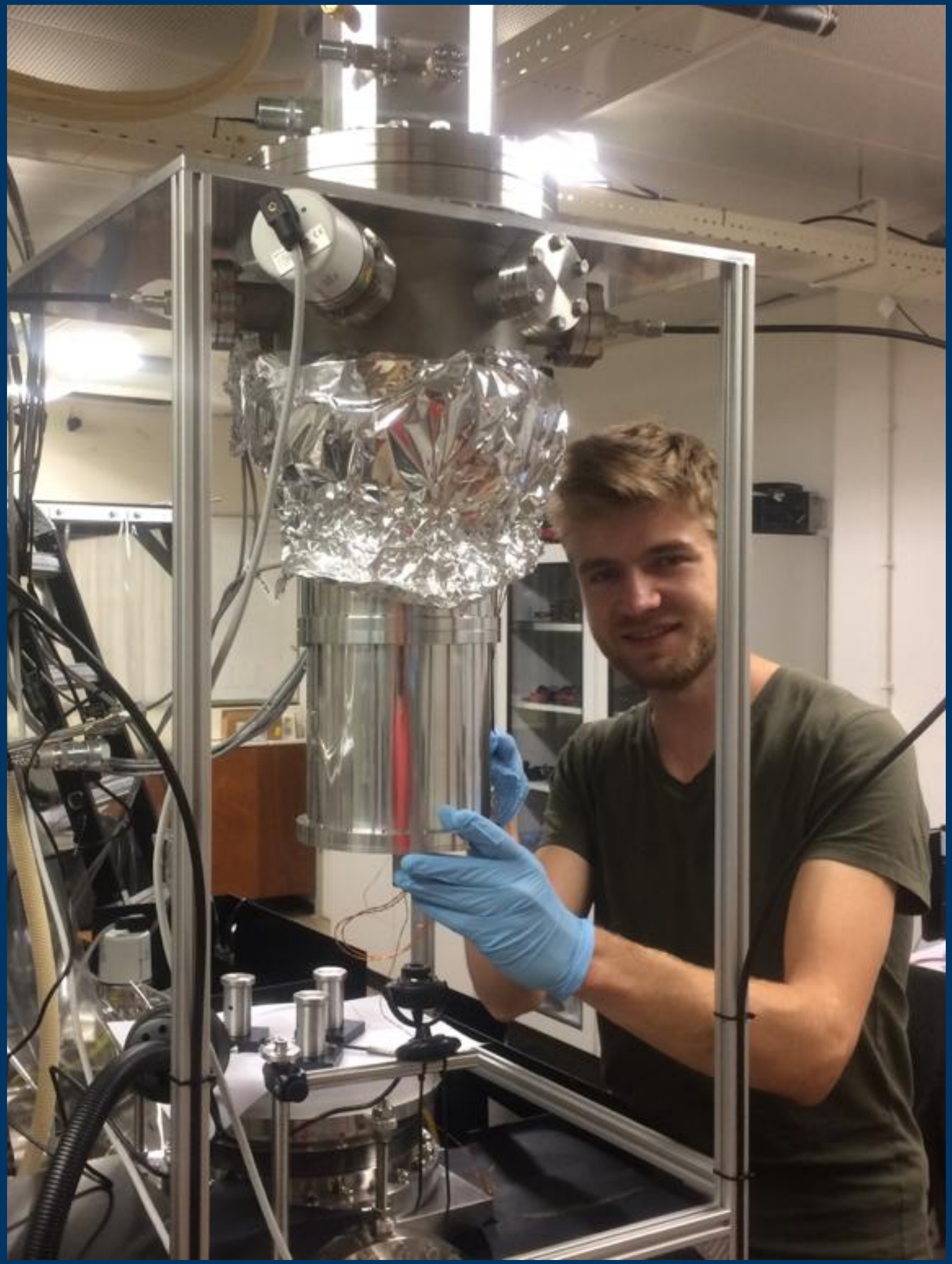


Cryogenic beam

- Evaporating metal target
- Neon carrier gas + SF_6
- Velocity 150-200 m/s

Goal:
Make the most intense source of slow molecules

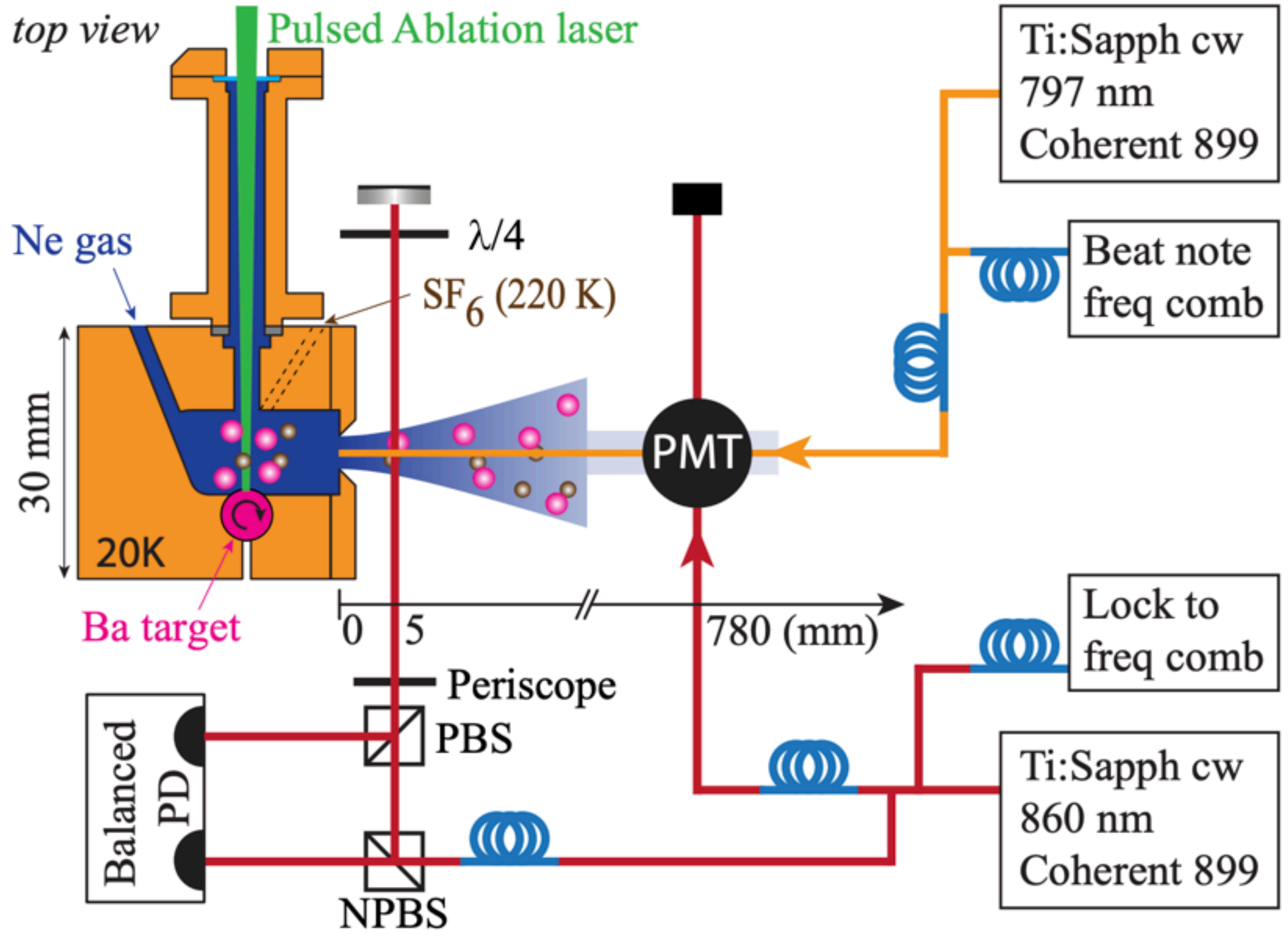
1 in Groningen (SrF, BaF, production)
1 in A'dam (BaF, optimisation)
1 in Groningen (polyatomic molecules)

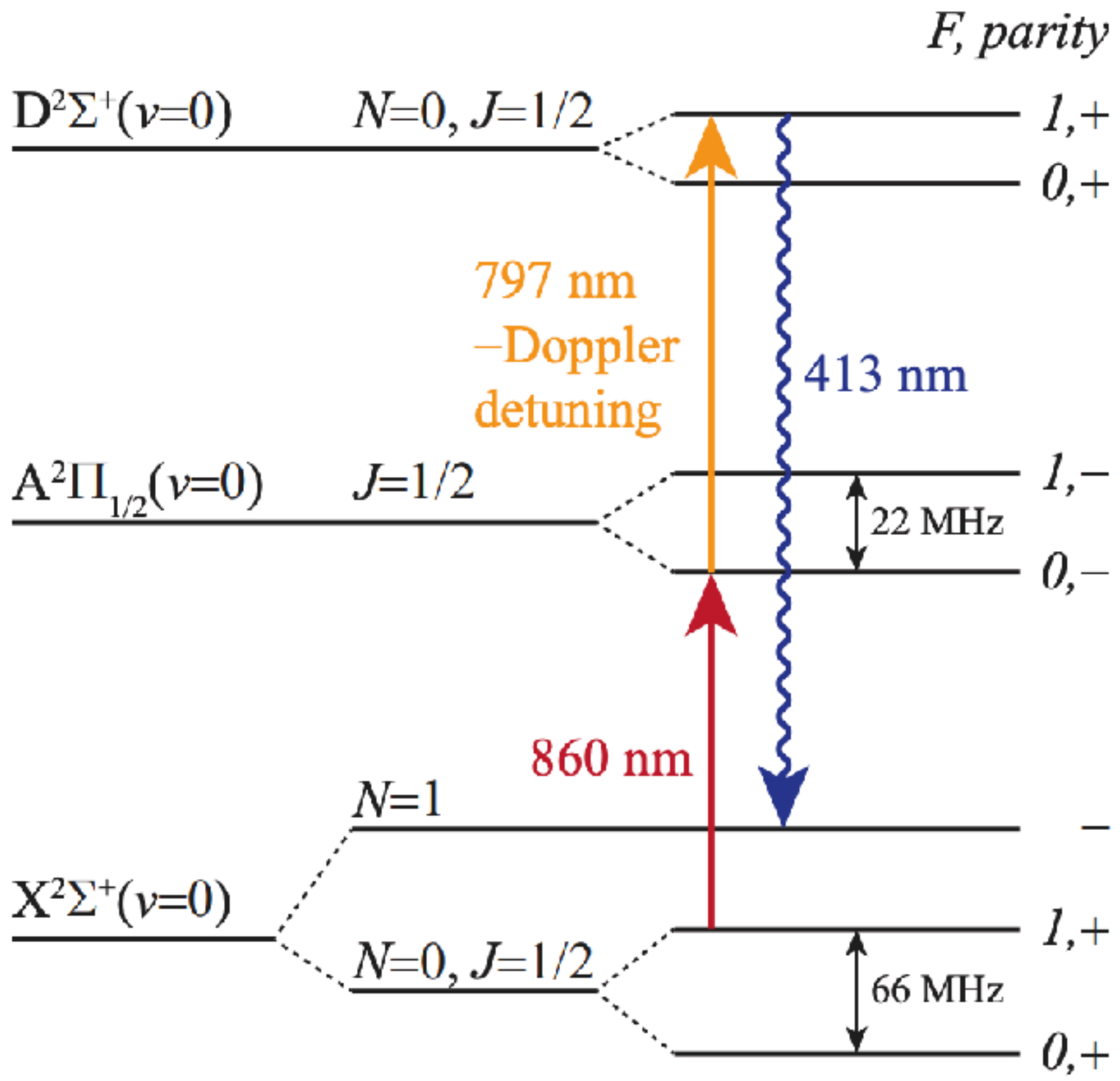


Maarten Mooij, Rick Bethlem @ VU Amsterdam

Optimising the molecular beam source

Mooij et al, NJP **26** 053009 (2024) and J. Phys. B **58** 015303 (2025)





Example: beam cell length

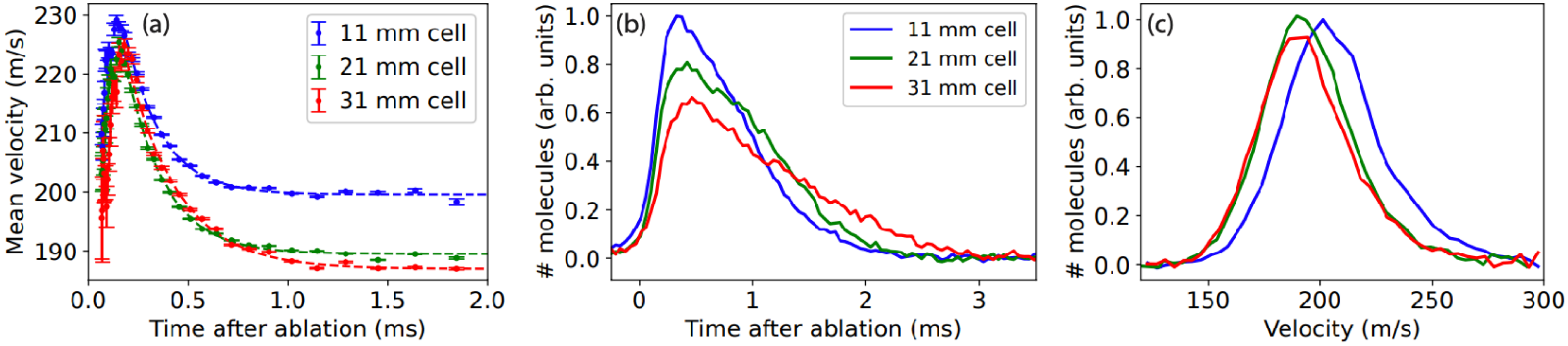
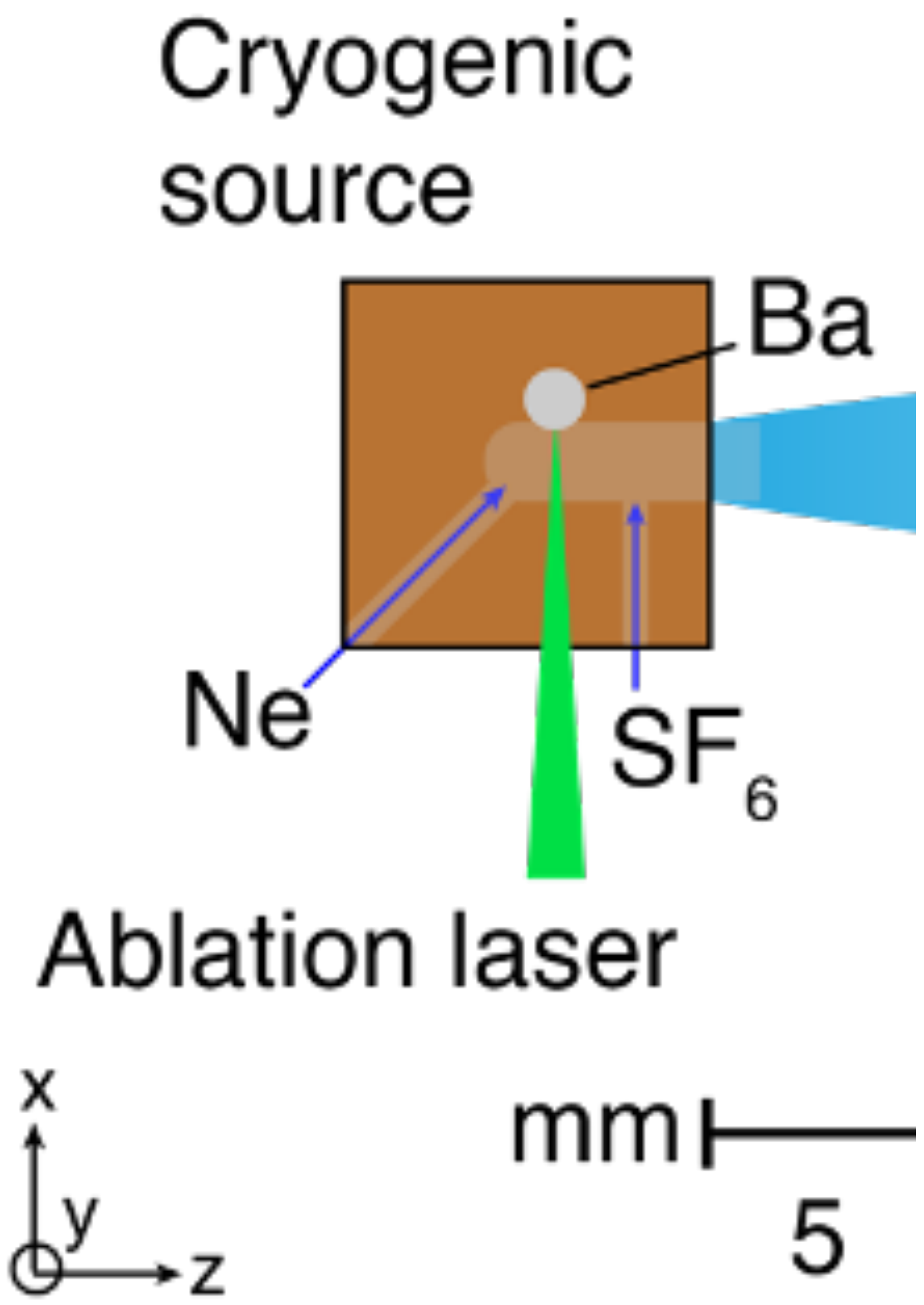


Figure 7. (a) Mean velocity as a function of time, (b) time-of-flight and (c) velocity distribution for three different cell lengths. The velocity in the tail of the molecular pulse is seen to decrease significantly, while the intensity is comparable.

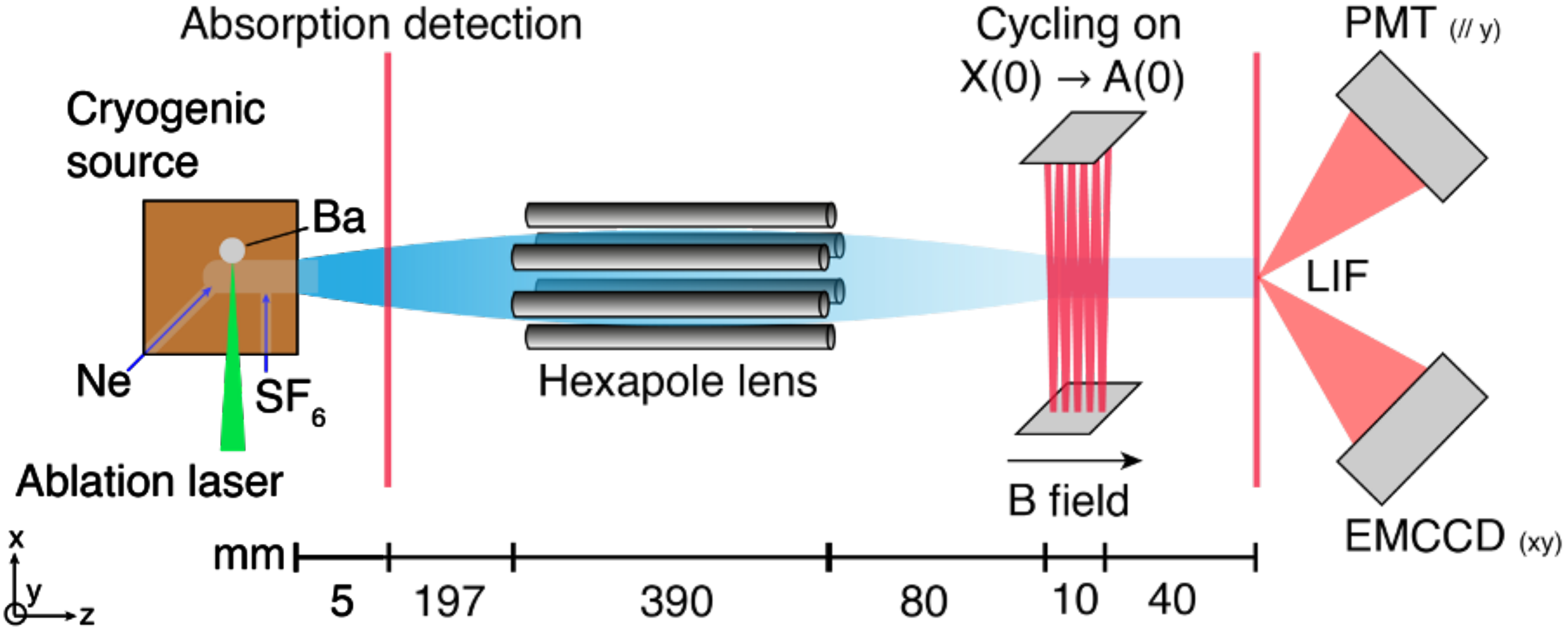
Beam divergence

Hexapole and laser cooling



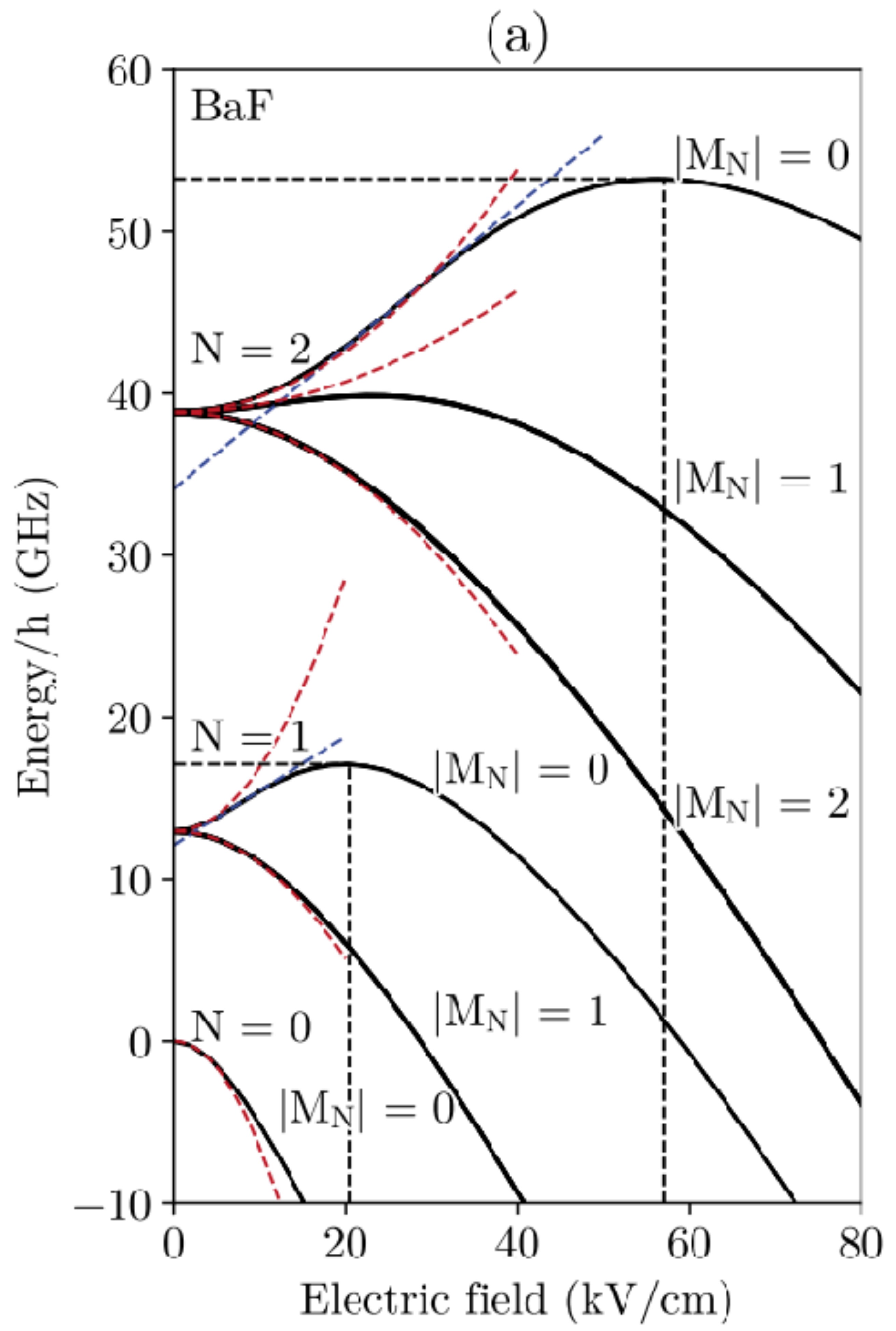
Beam divergence

Hexapole and laser cooling



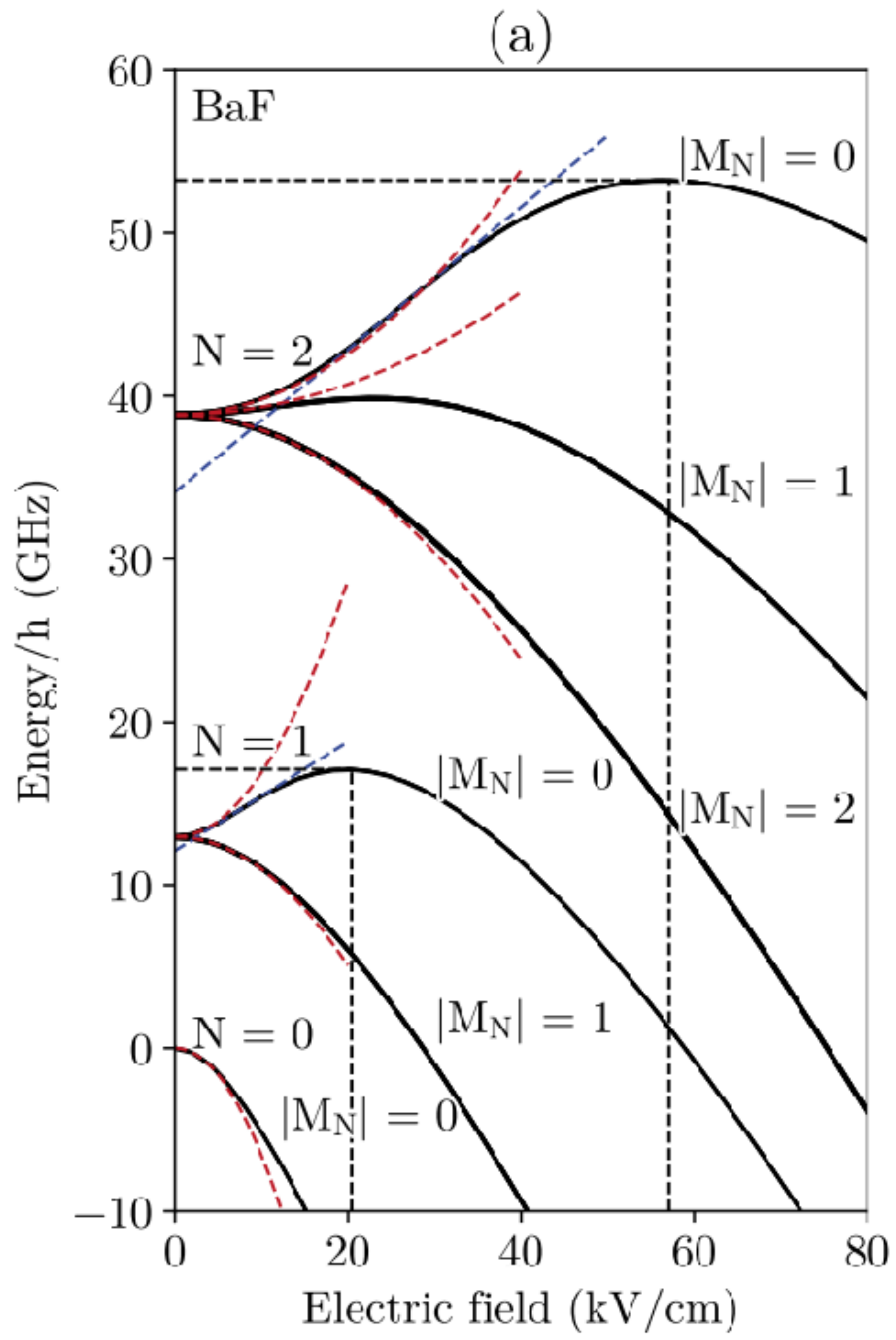
BaF in electric fields

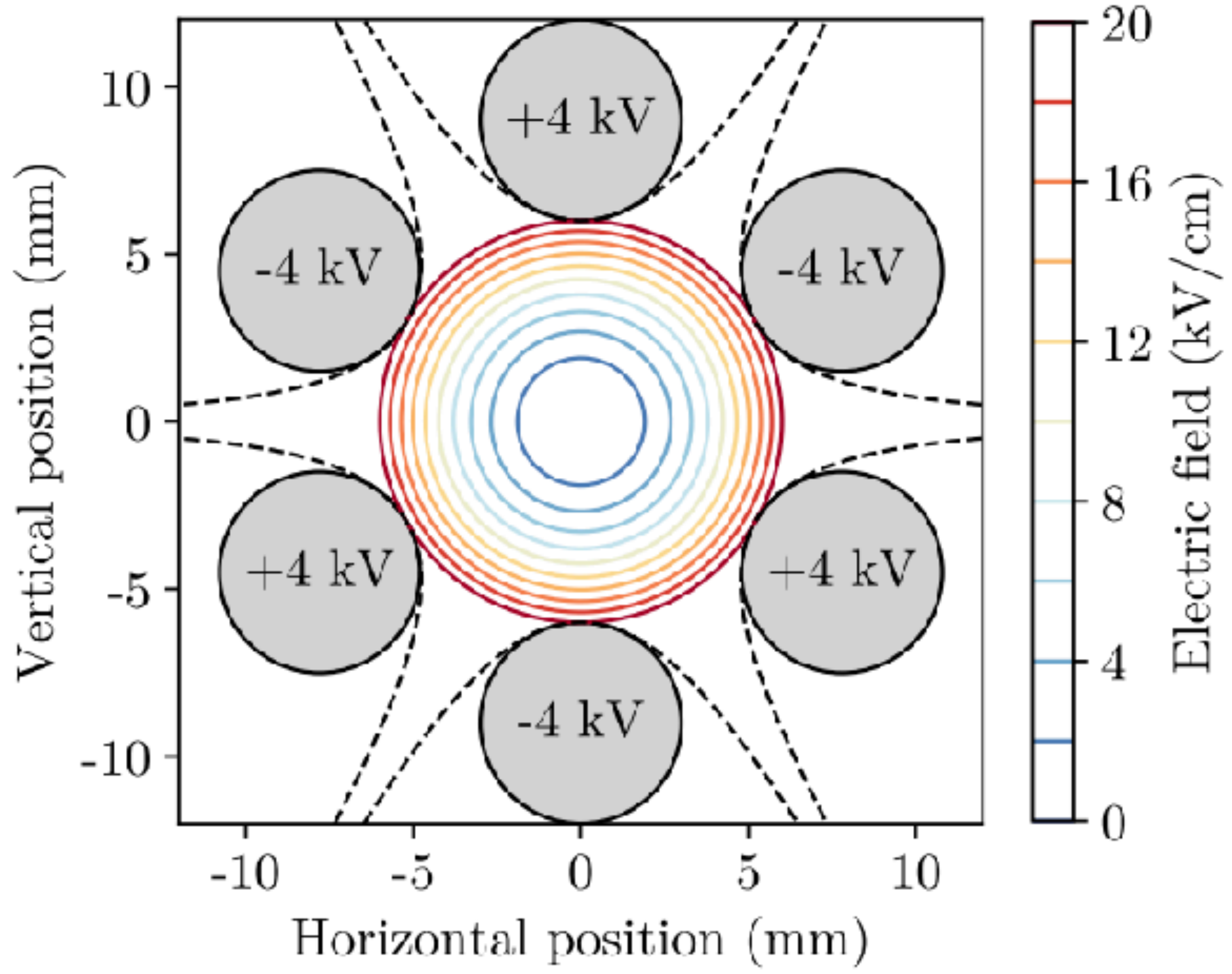
Hexapole (static fields) can focus a beam of neutral molecules



BaF in electric fields

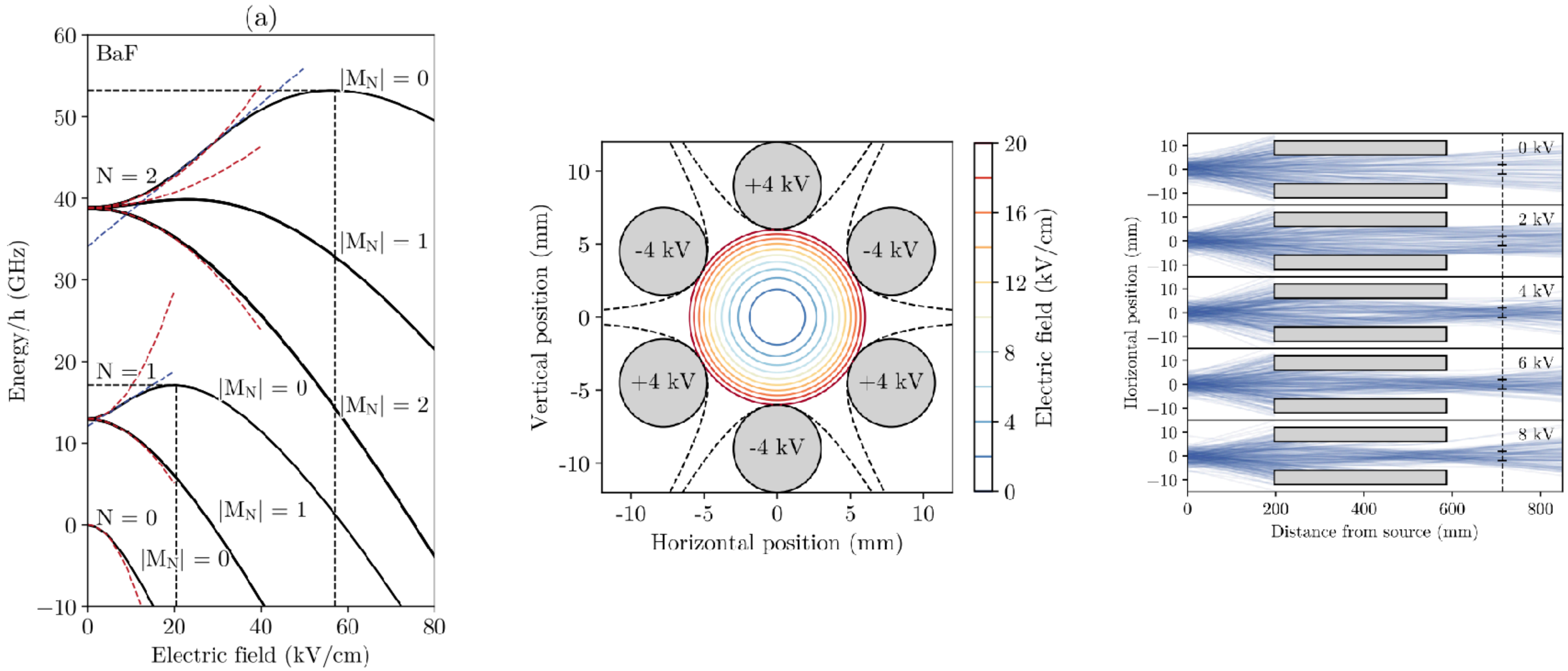
Hexapole (static fields) can focus a beam of neutral molecules





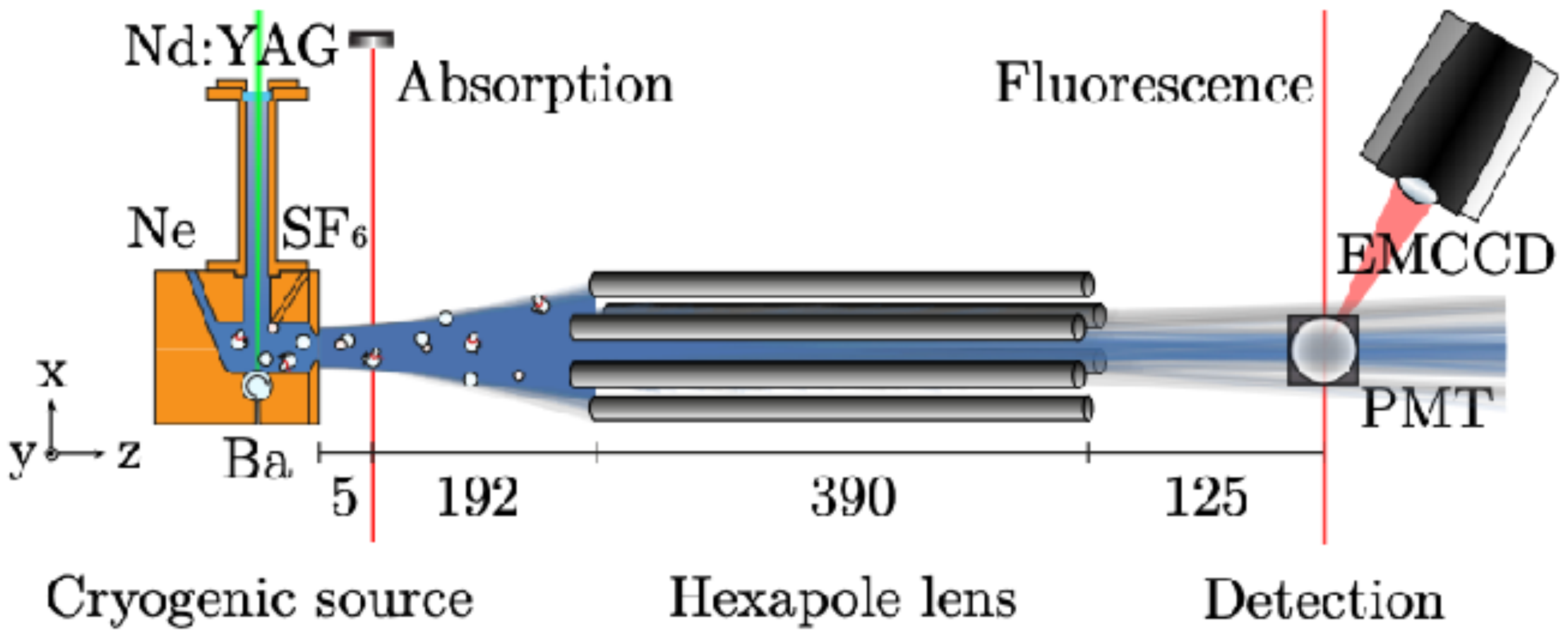
BaF in electric fields

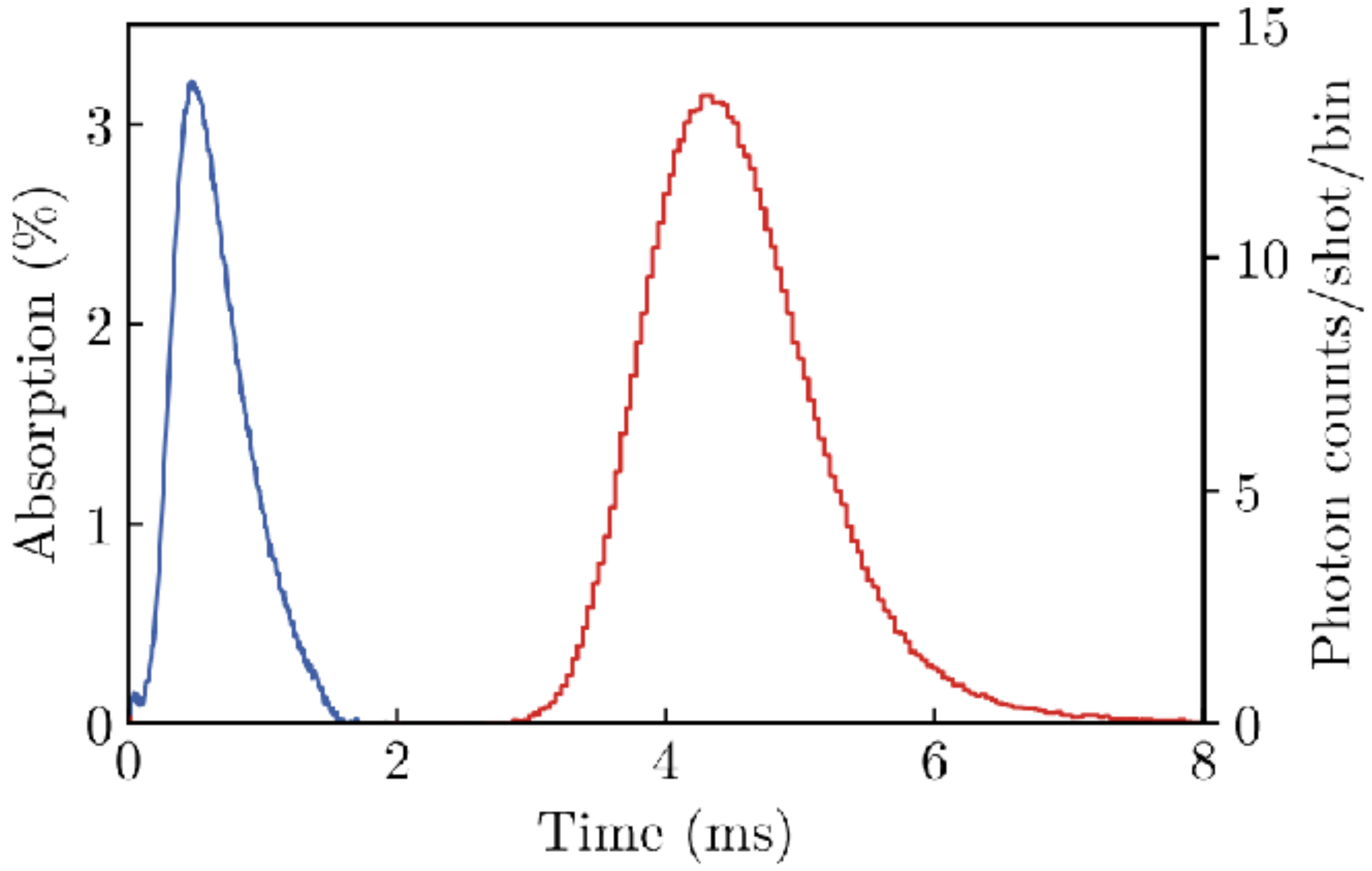
Hexapole (static fields) can focus a beam of neutral molecules

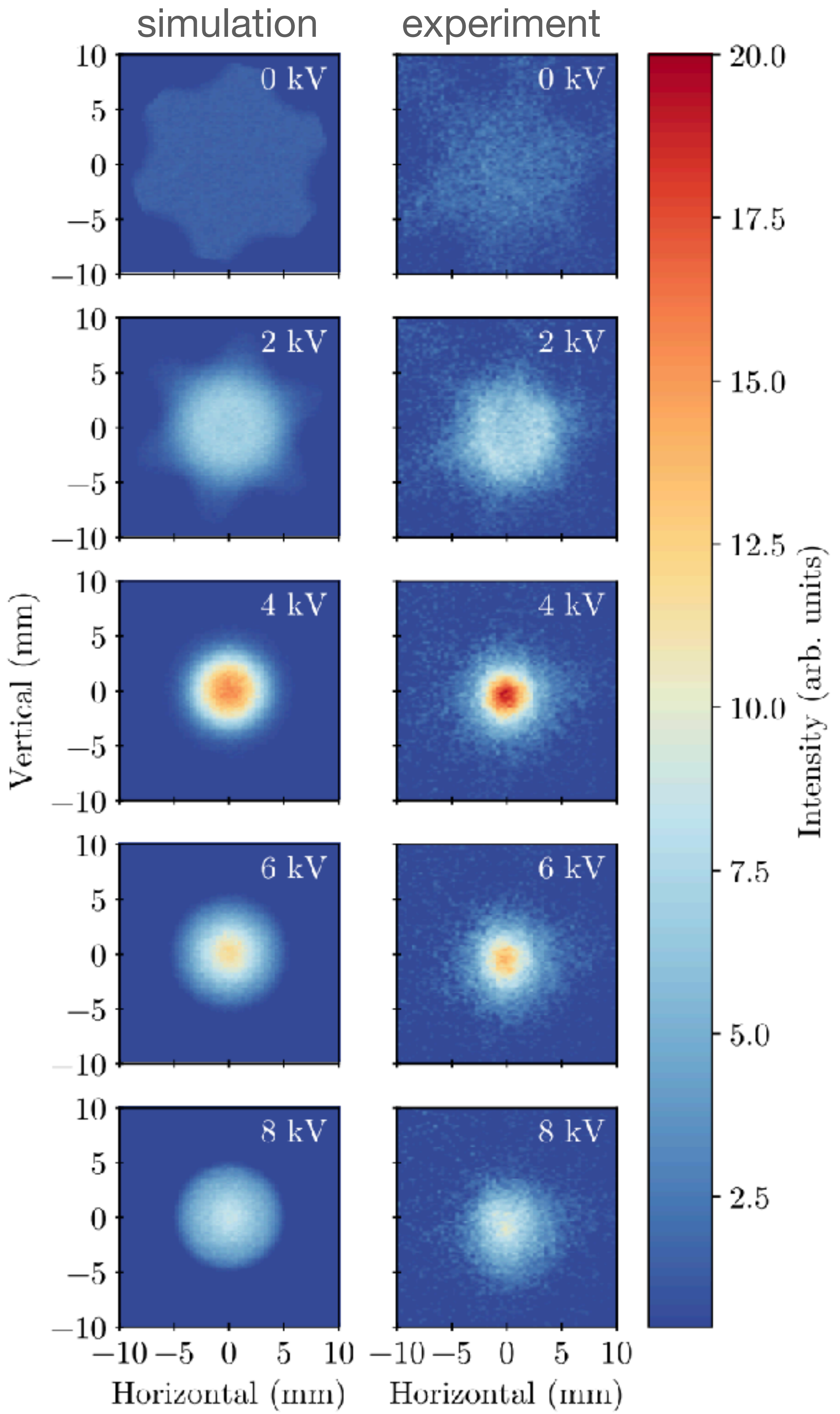


Hexapole focussing

Anno Touwen et al, NJP **26** 073054 (2024)







A few words on laser cooling

‘molecule X can be lasercooled’

J. Chem. Phys. 151, 034302 (2019)

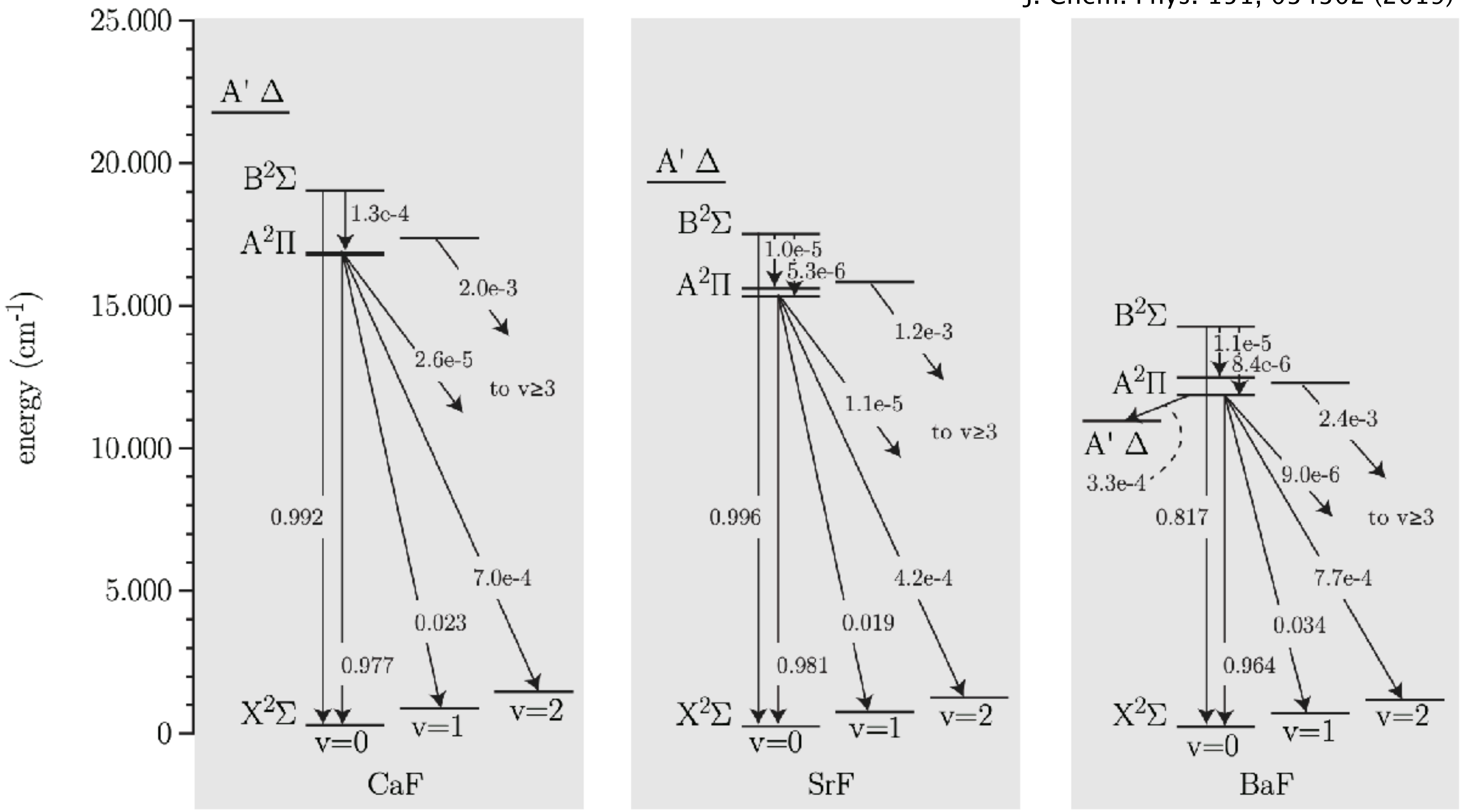
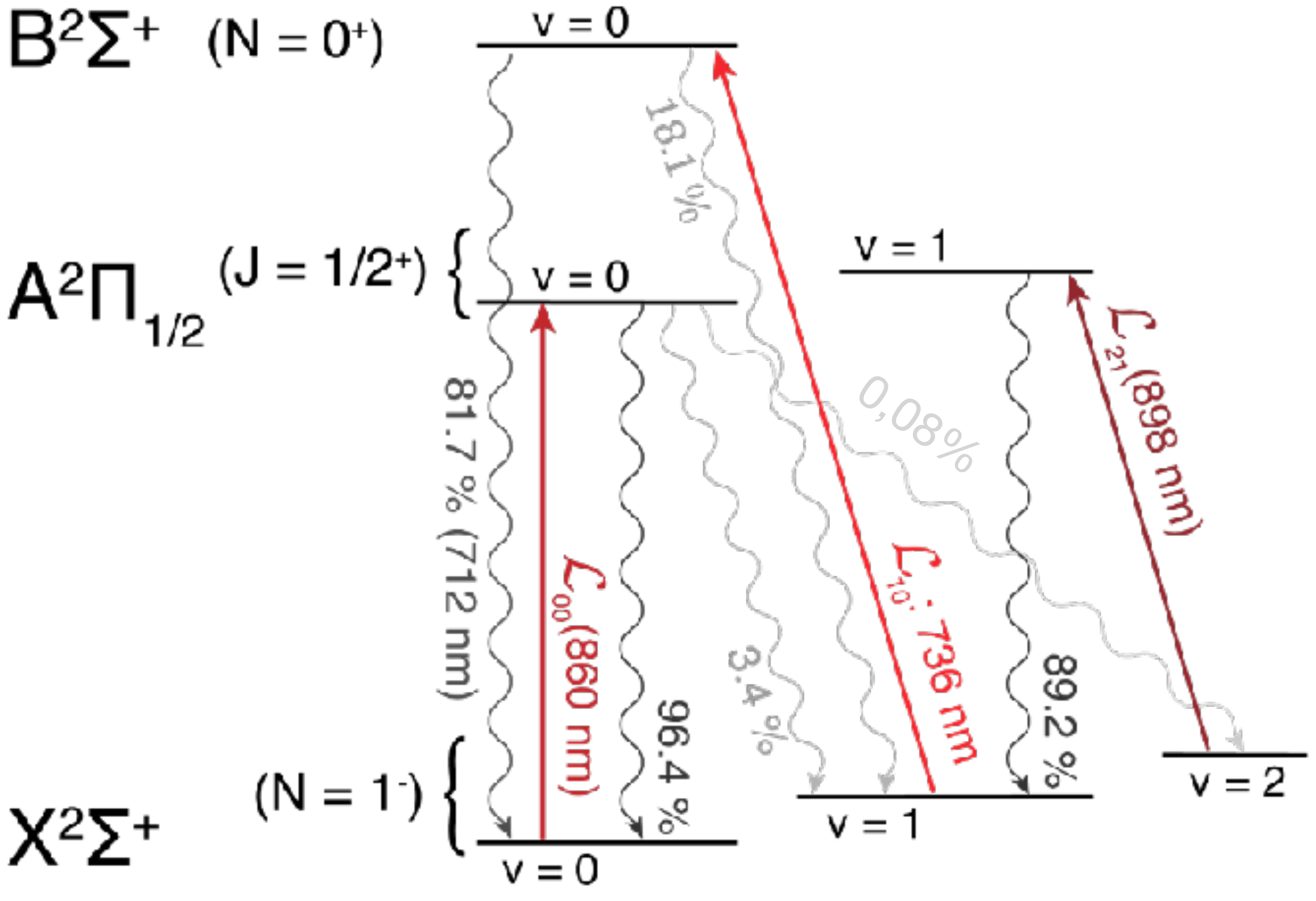
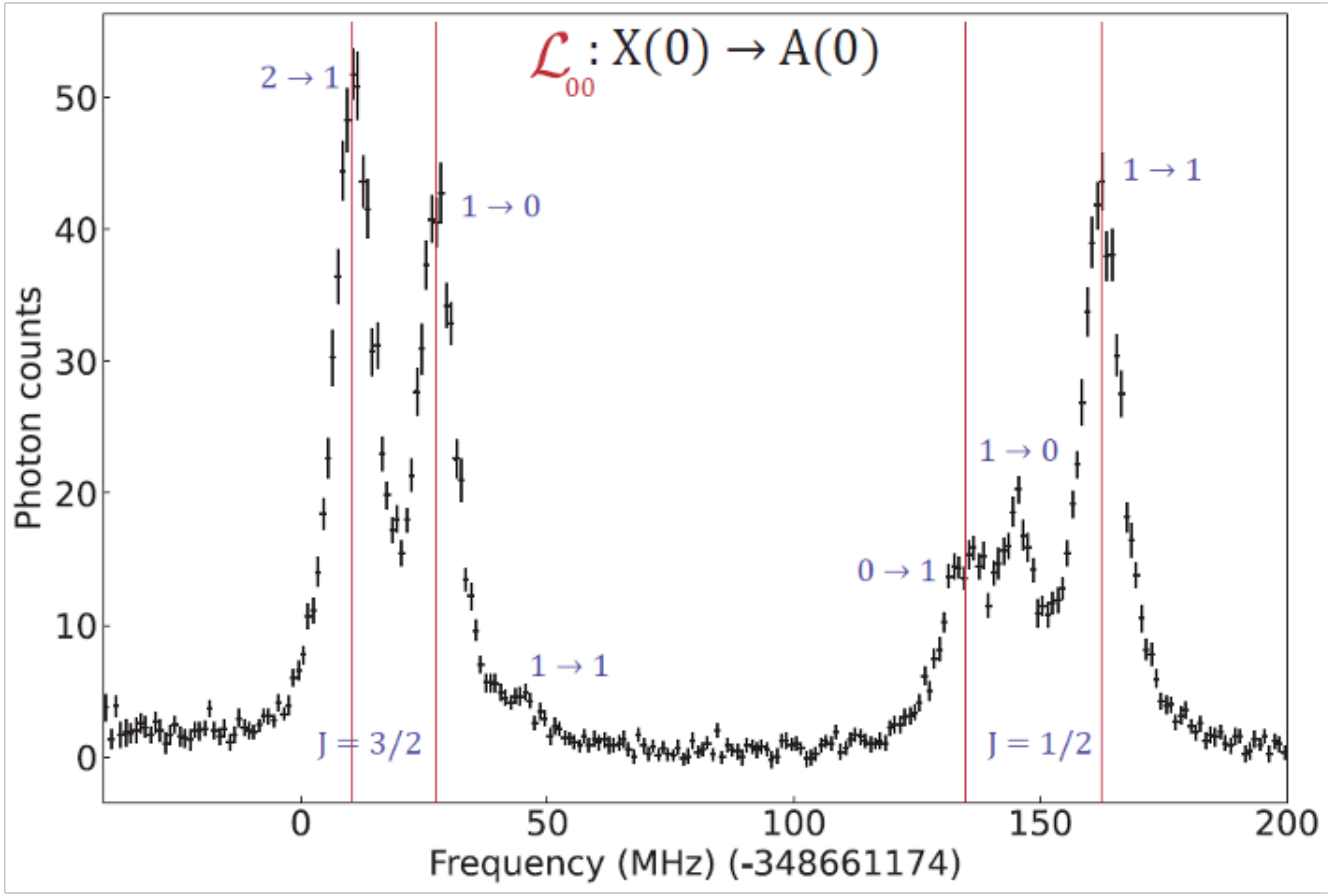


FIG. 5: The most important energy levels for laser-cooling and the calculated relative decay fractions for CaF, SrF, and BaF.

Transitions for laser cooling

In the presence of hyperfine structure





2D laser cooling

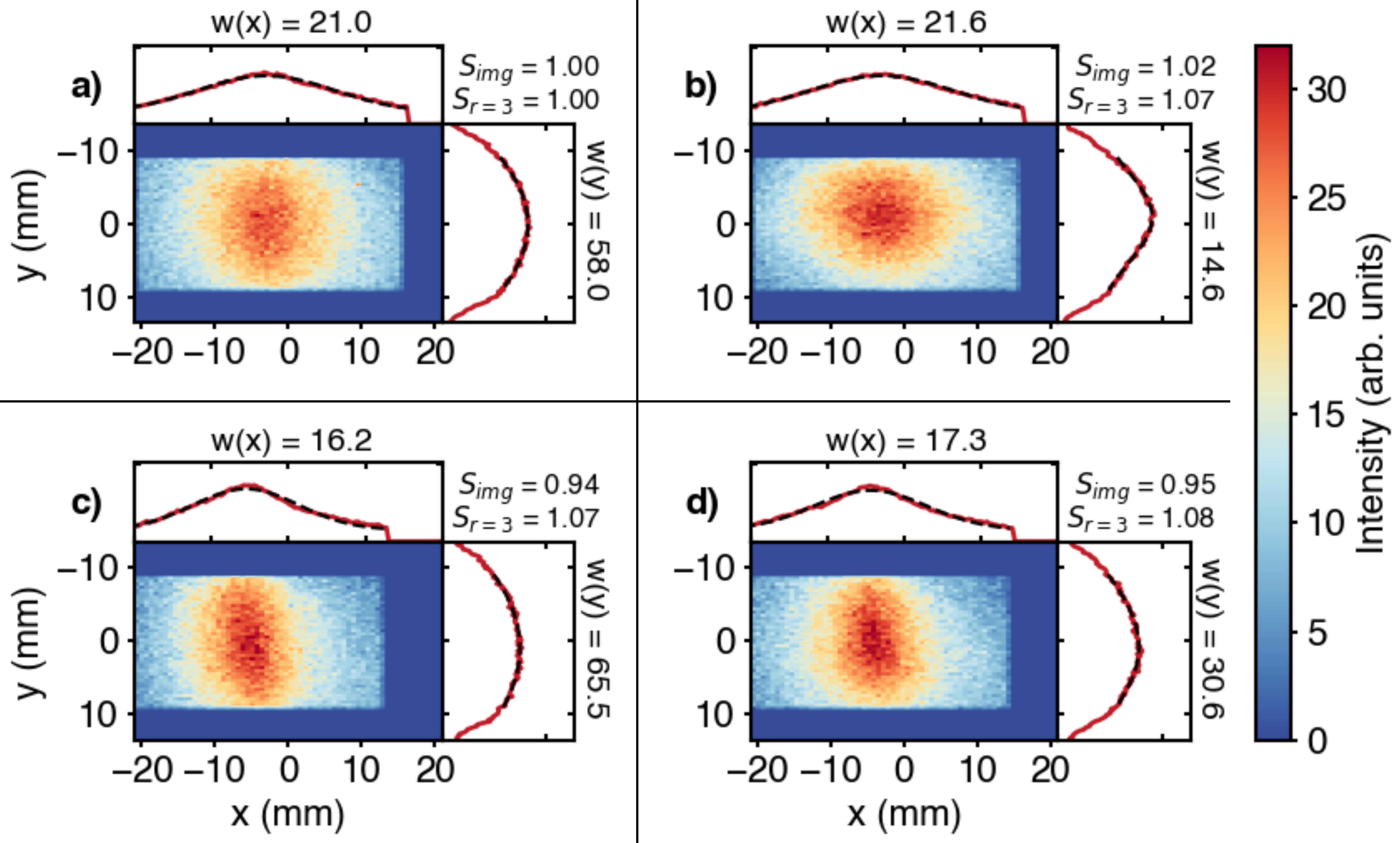
large capture velocity in combination with hexapole

promising results

ongoing work

y-cooling

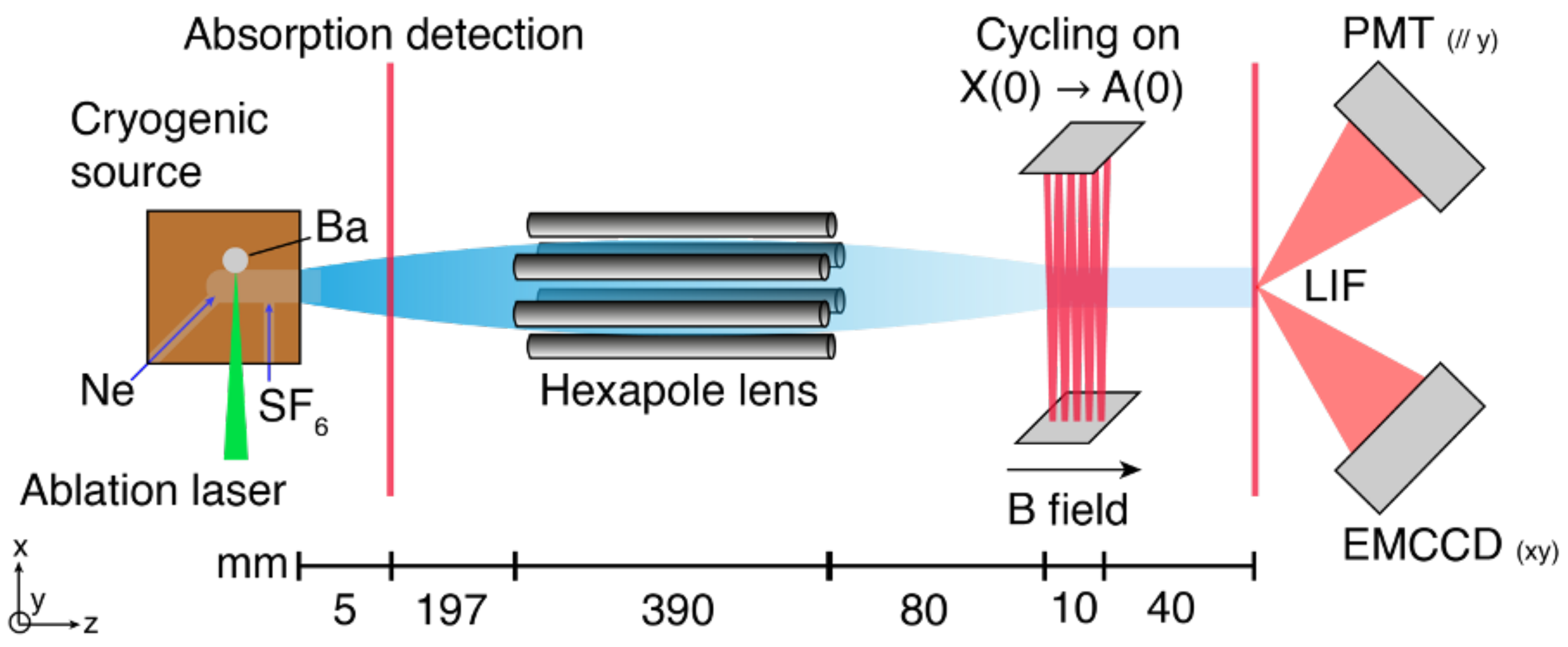
x-cooling



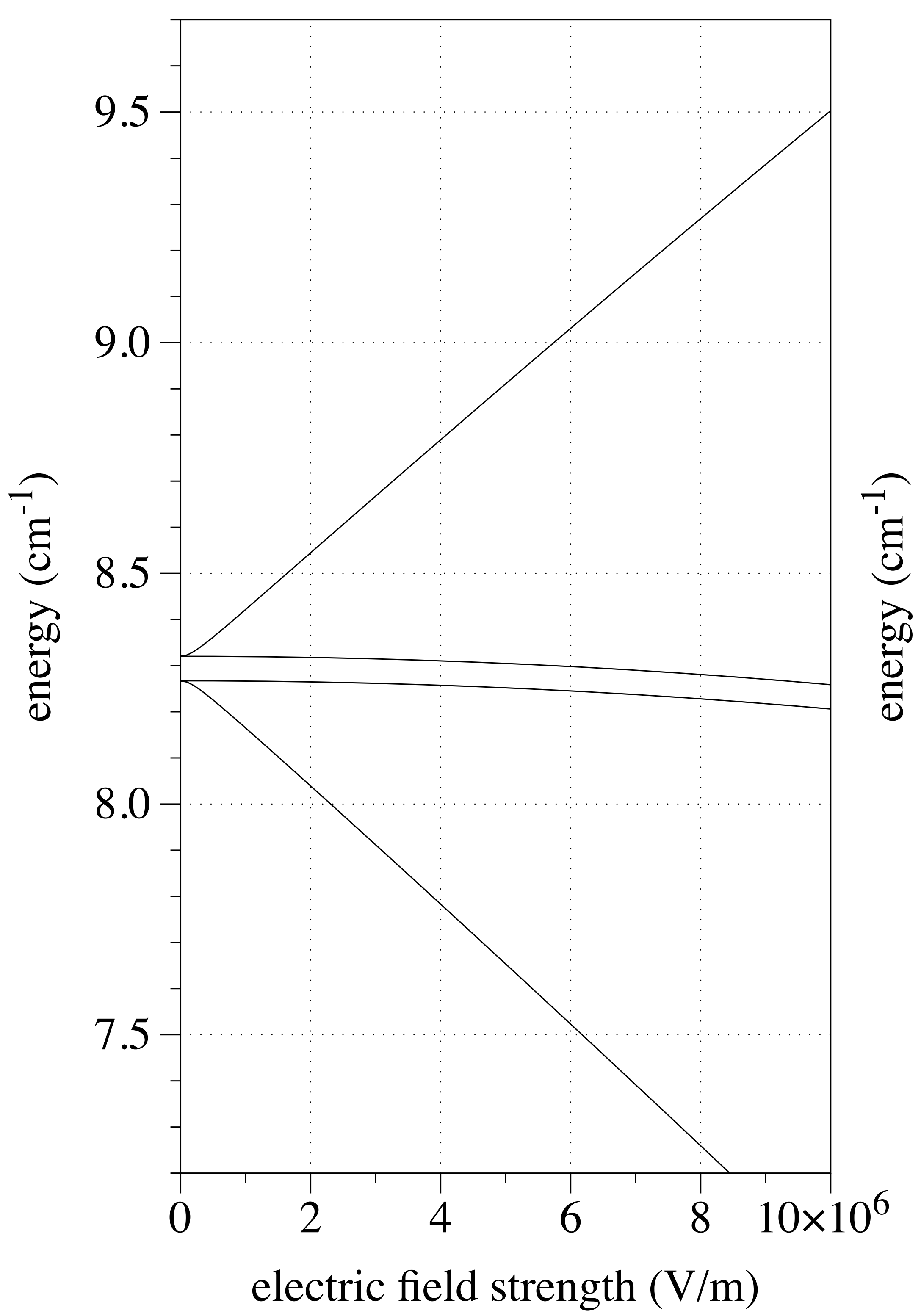
Current status

Phase 2: 150 m/s beam

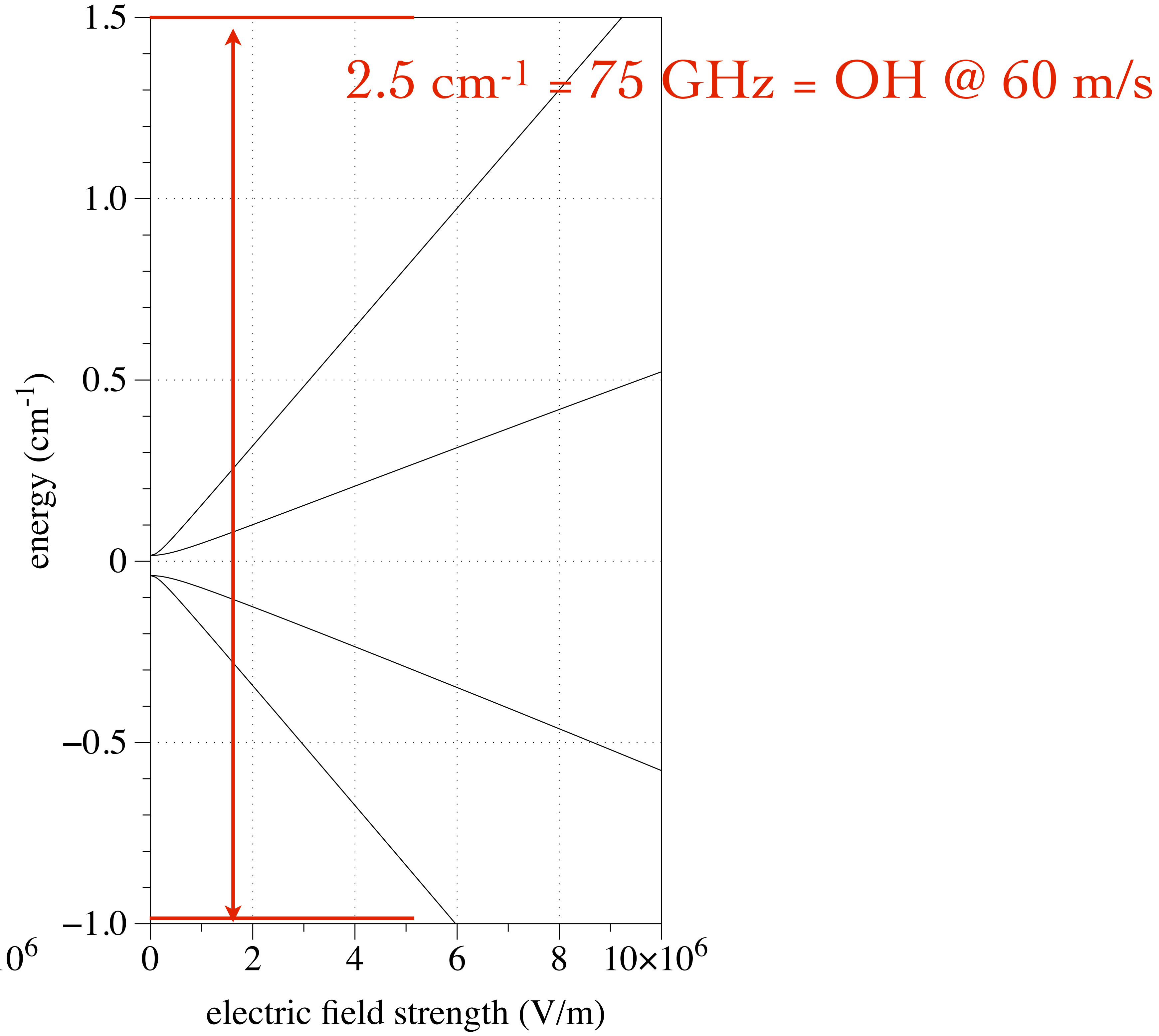
- Cryogenic beams optimised, ~10^{11} molecules/sr/shot in eEDM state
- Hexapole implemented, gain factor ~5
- Laser cooling setup completed
- Currently optimising 2D transverse cooling
- Combine with interaction zone this year



Even slower: use Stark shift to decelerate

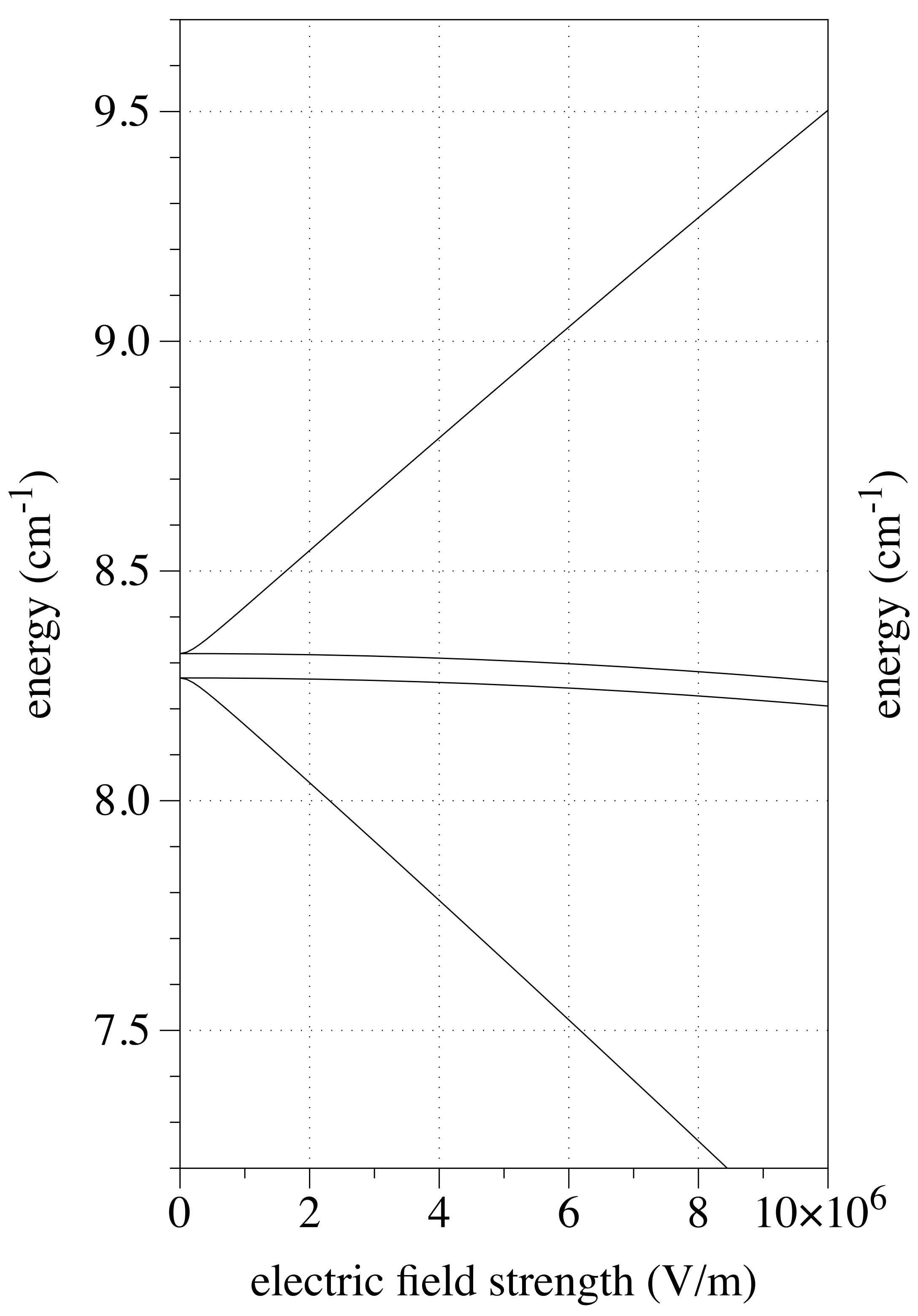


ND3

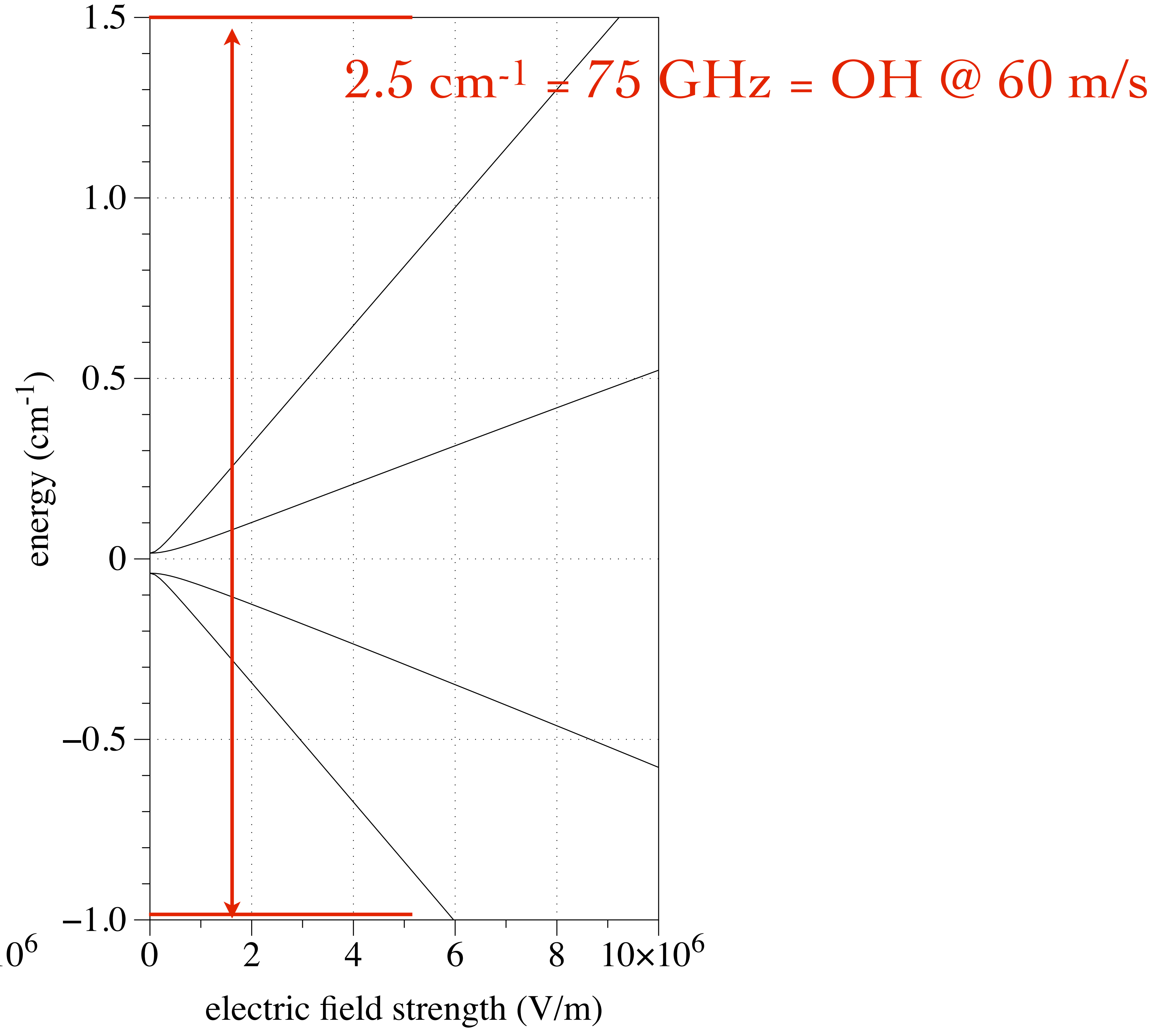


OH

Even slower: use Stark shift to decelerate



ND_3



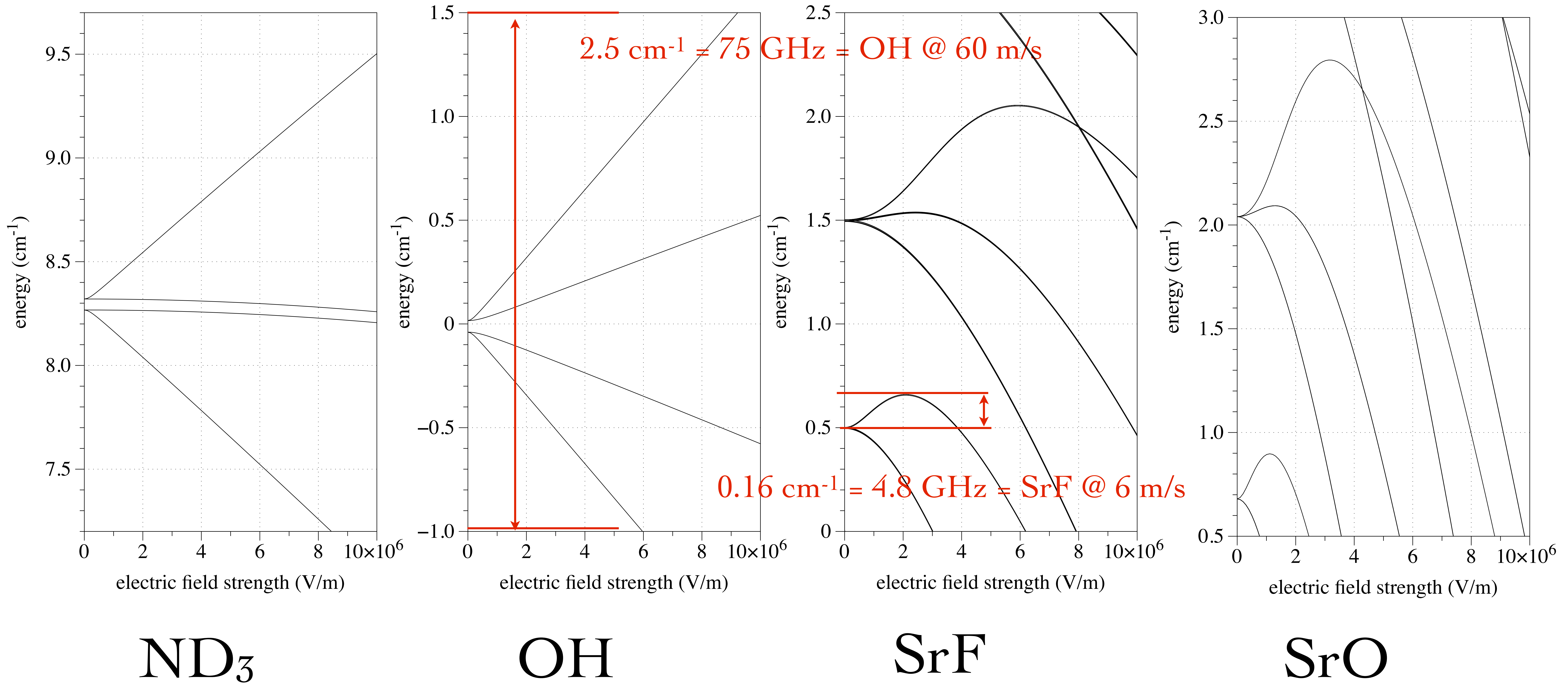
OH

Deceleration, trapping, collision studies, lifetime measurements

Demonstrated for light molecules: OH, CO, NH_3, NH

Science 313 5793 (2006), PRL 98 133001 (2007), PRL 110 133003 (2013)

Even slower: use Stark shift to decelerate



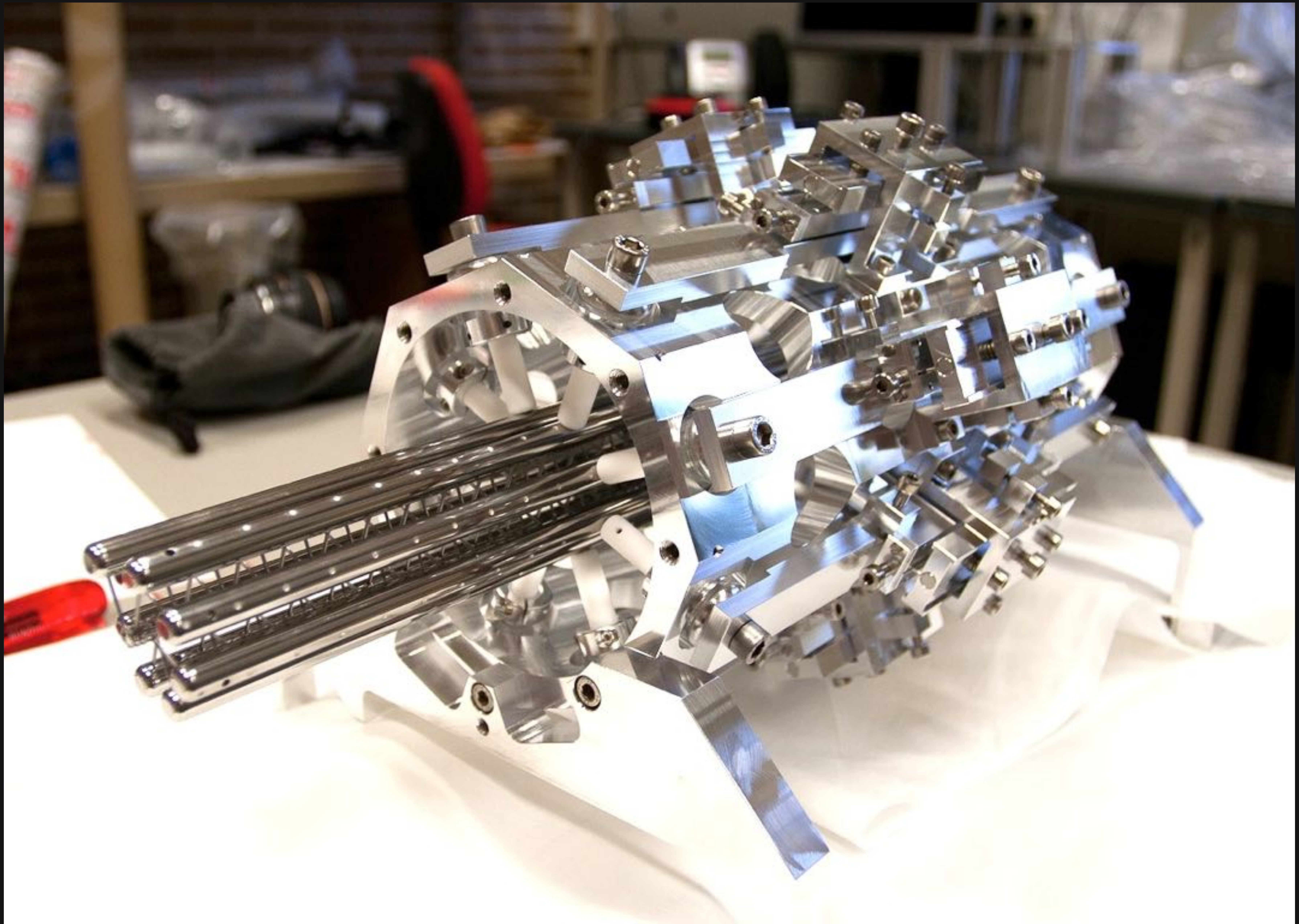
Challenge: extend this technique to heavier species

Deceleration, trapping, collision studies, lifetime measurements

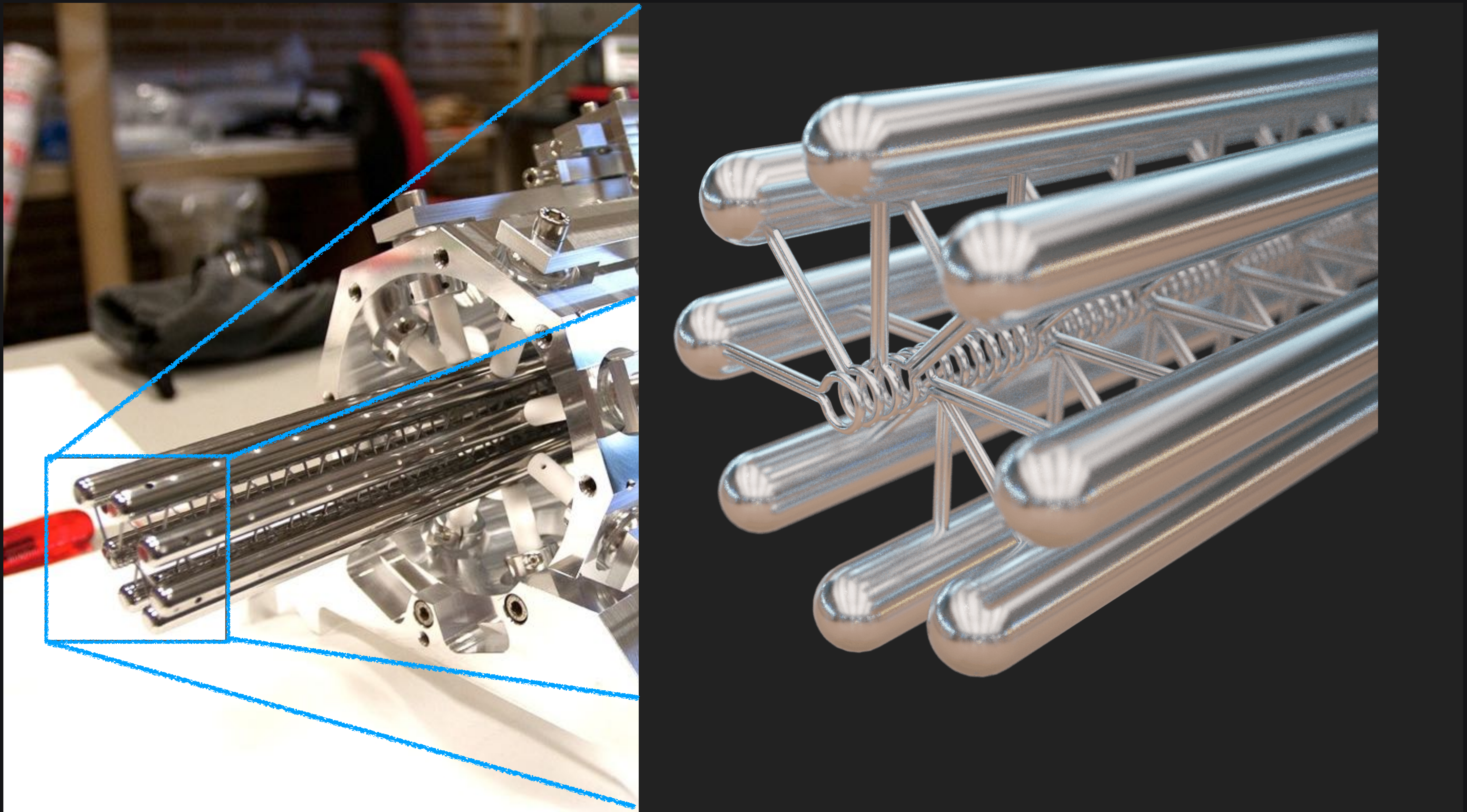
Demonstrated for light molecules: OH, CO, NH_3, NH

Science 313 5793 (2006), PRL 98 133001 (2007), PRL 110 133003 (2013)

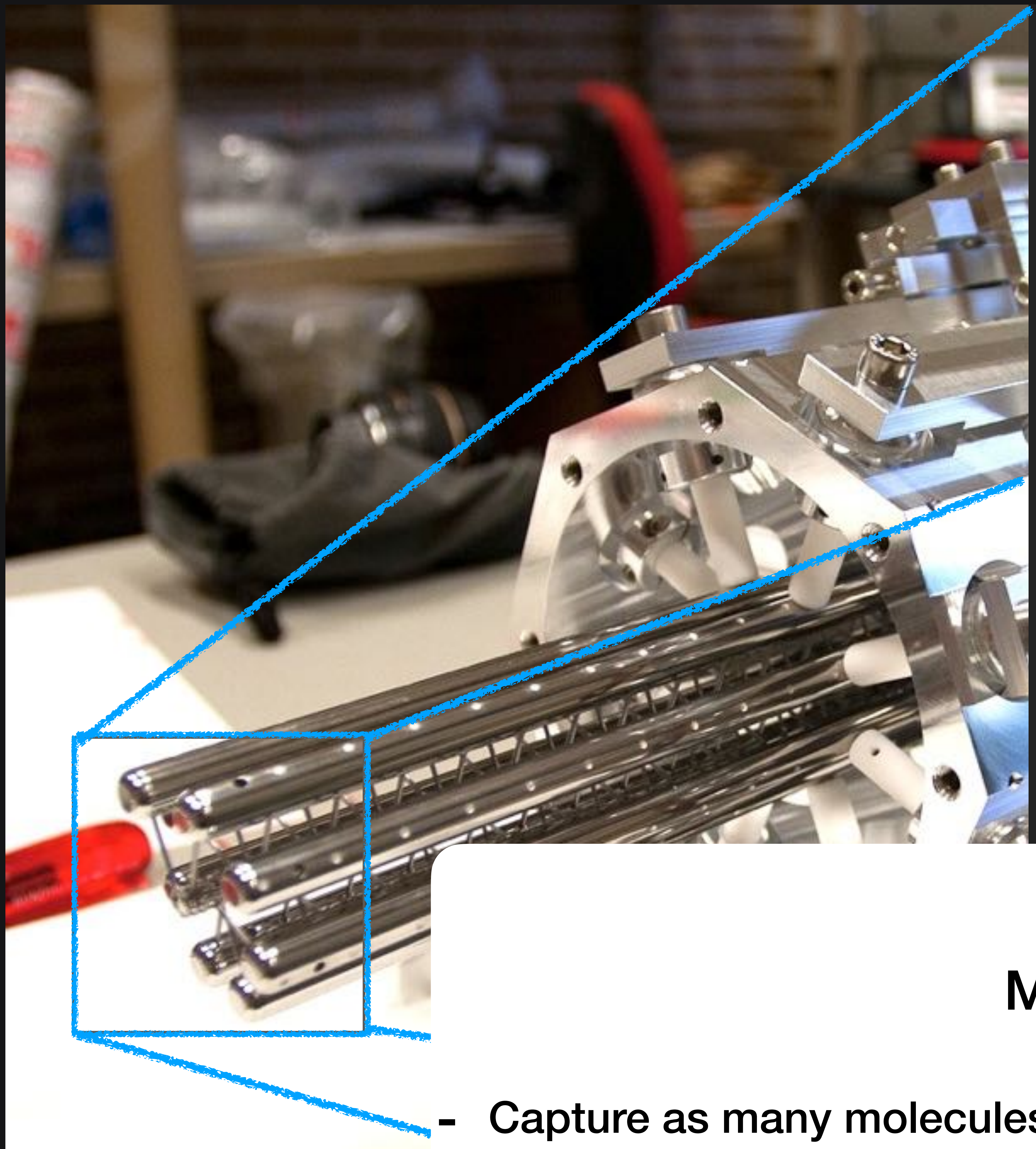
Traveling-wave decelerator: decelerate or completely stop molecules

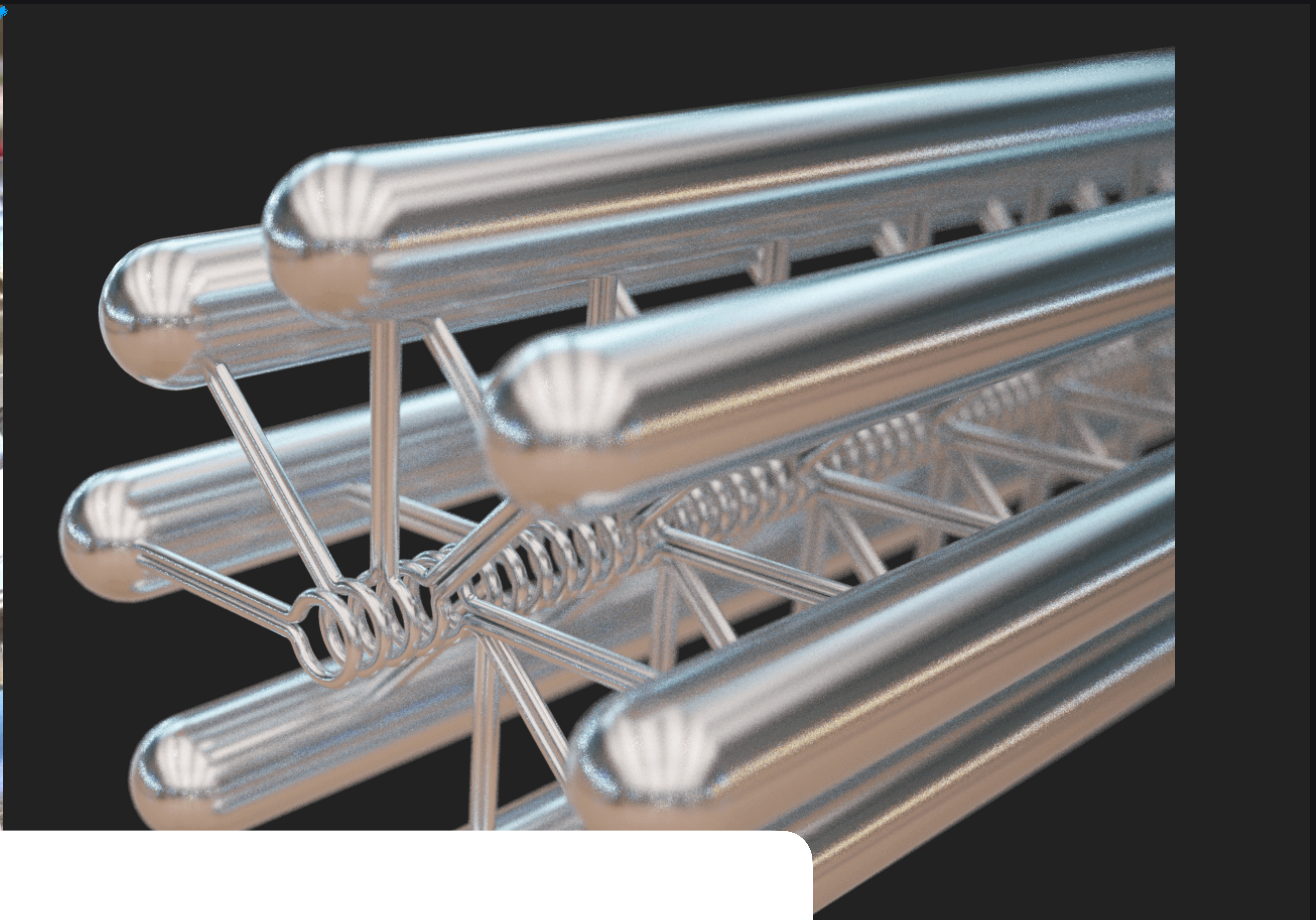


Traveling-wave decelerator: decelerate or completely stop molecules



Traveling-wave decelerator: decelerate or completely stop molecules

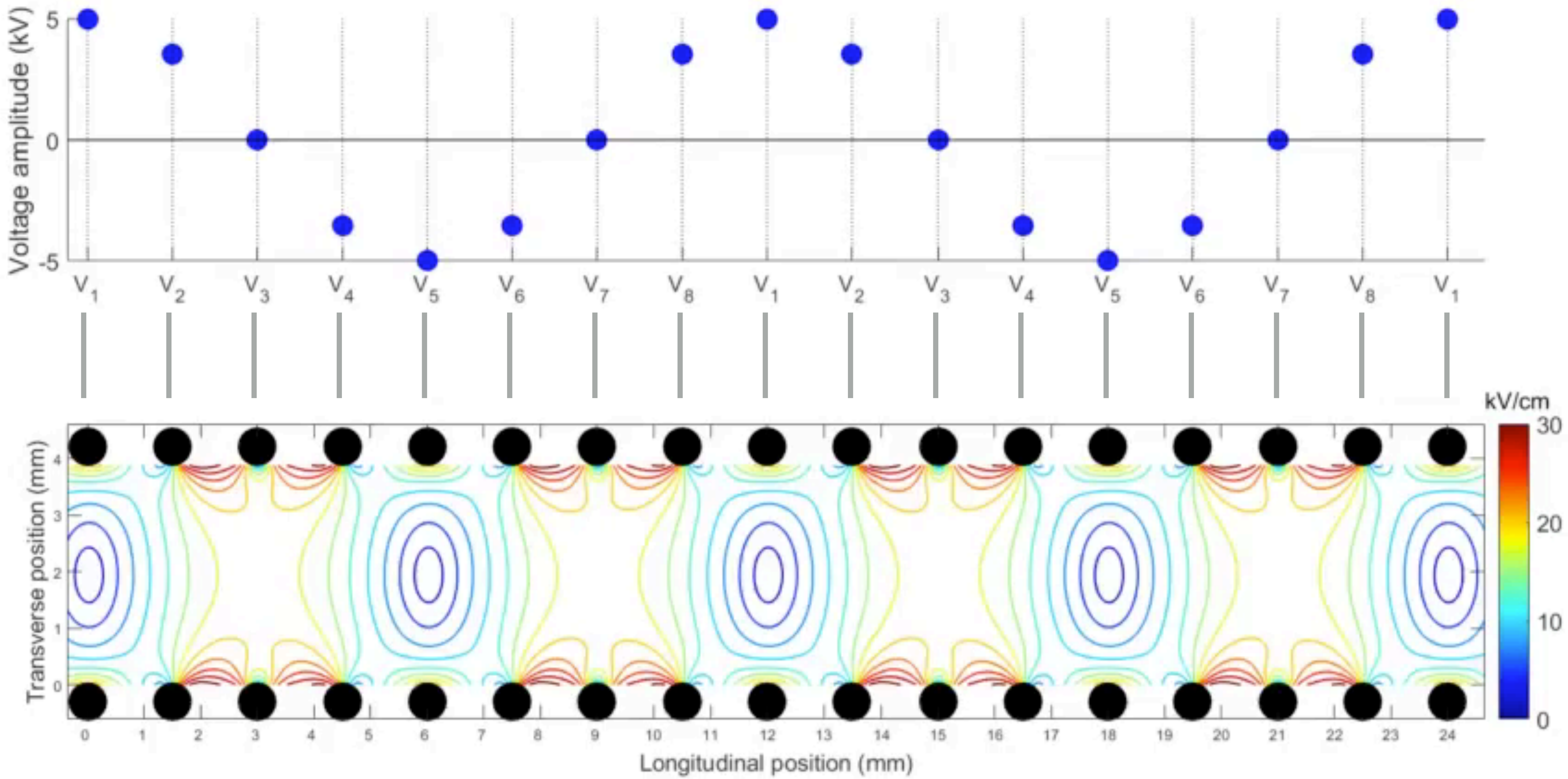




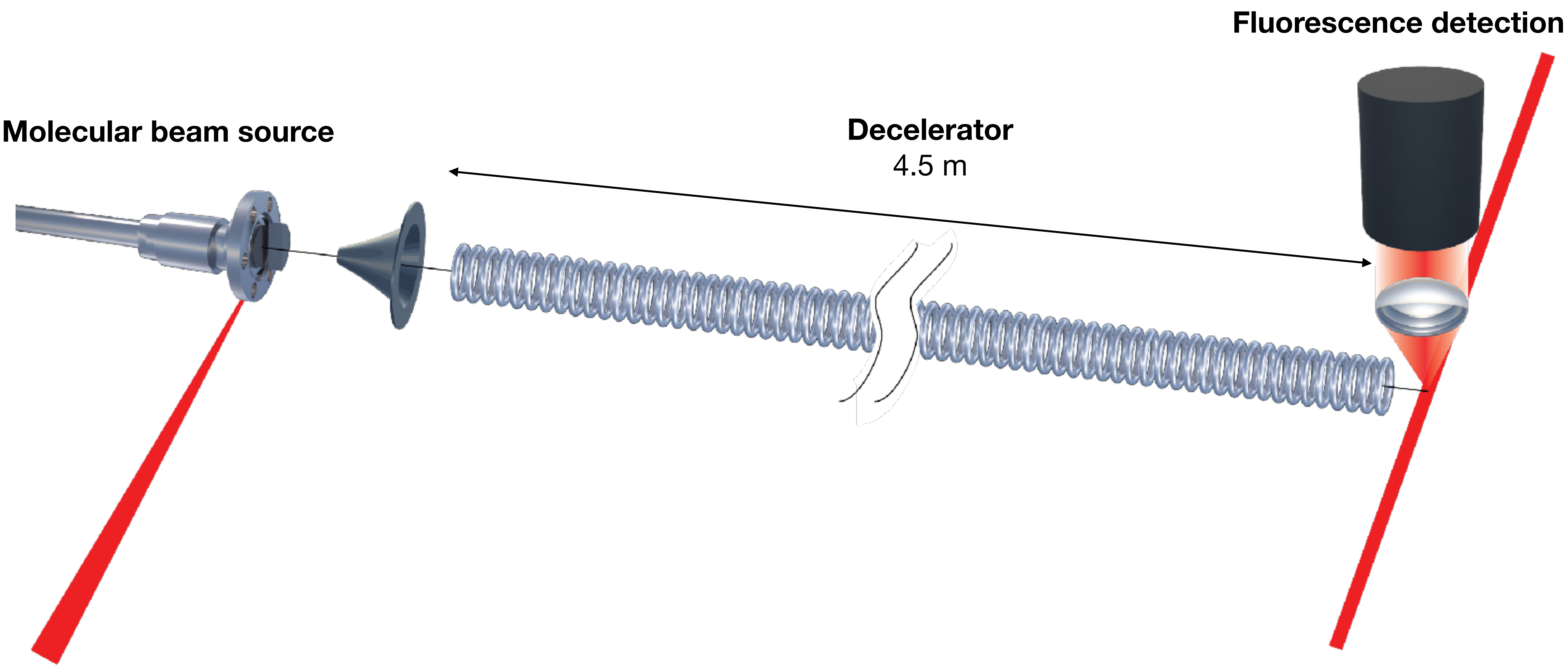
Main aims:

- Capture as many molecules as possible from molecular beam
- Maintain N during deceleration

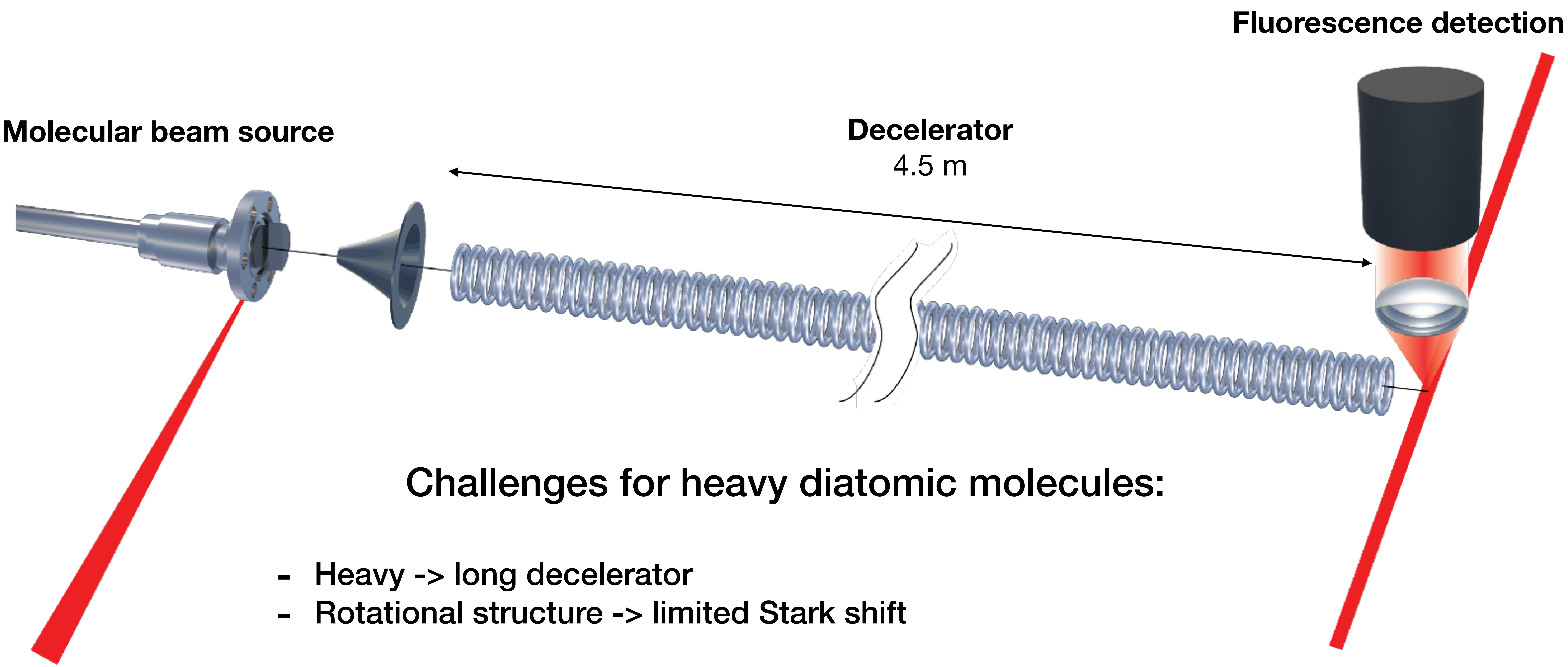
Traveling-wave decelerator



Traveling-wave decelerator



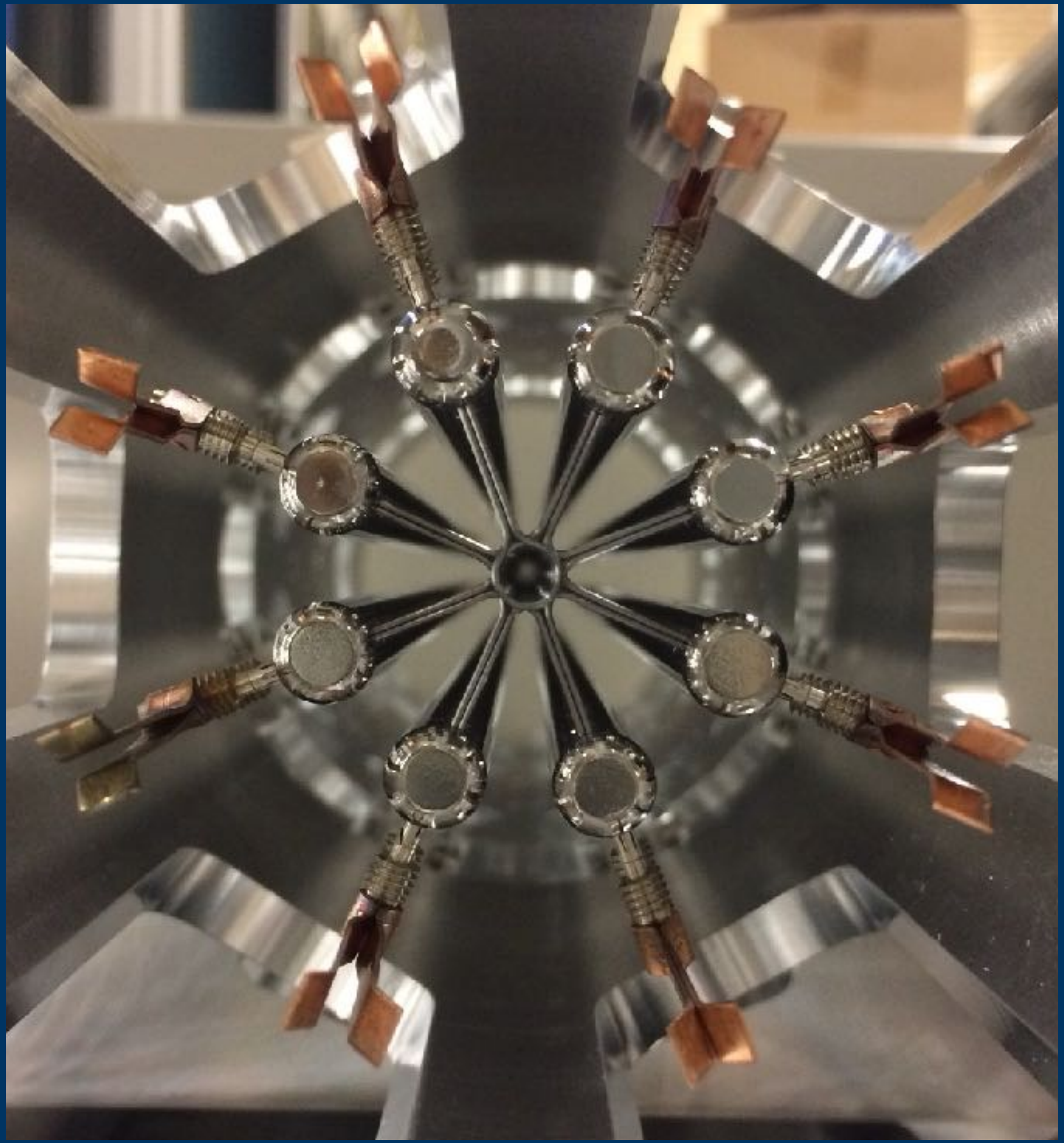
Traveling-wave decelerator



Challenges for heavy diatomic molecules:

- Heavy -> long decelerator
- Rotational structure -> limited Stark shift

Modular traveling-wave decelerator

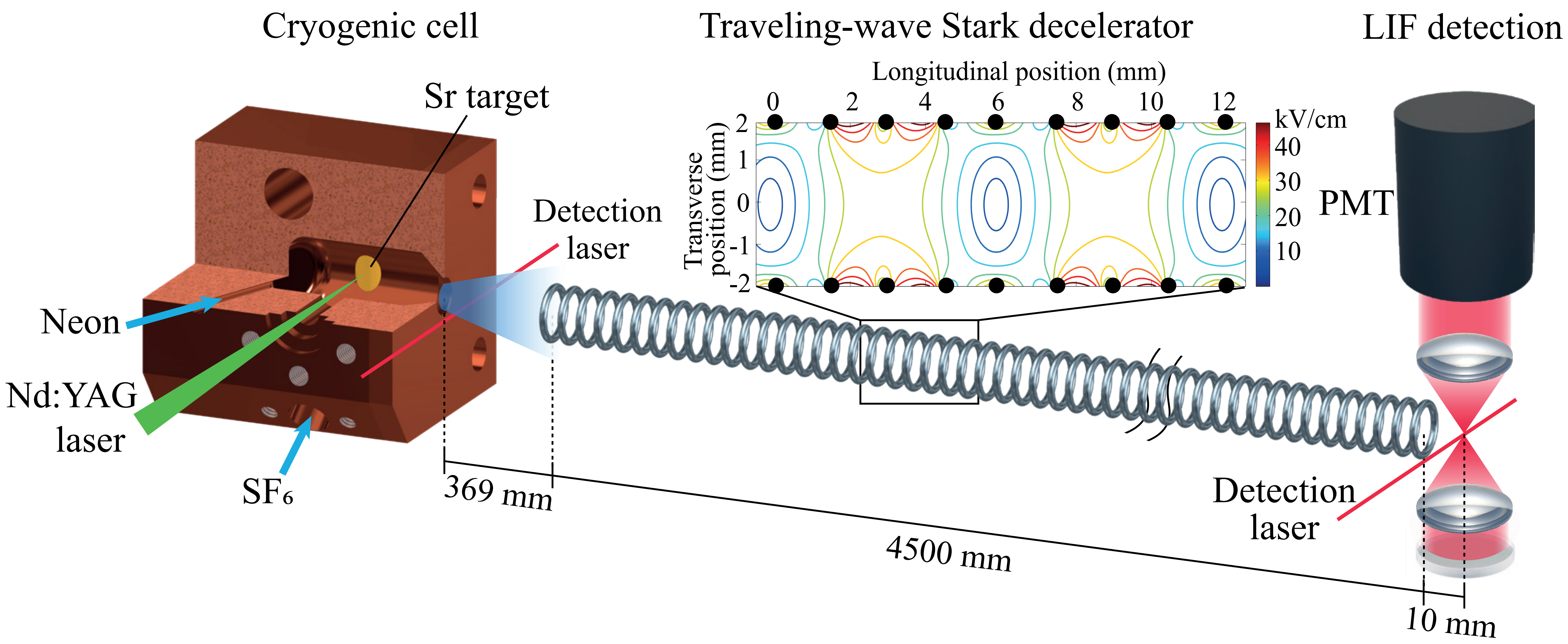


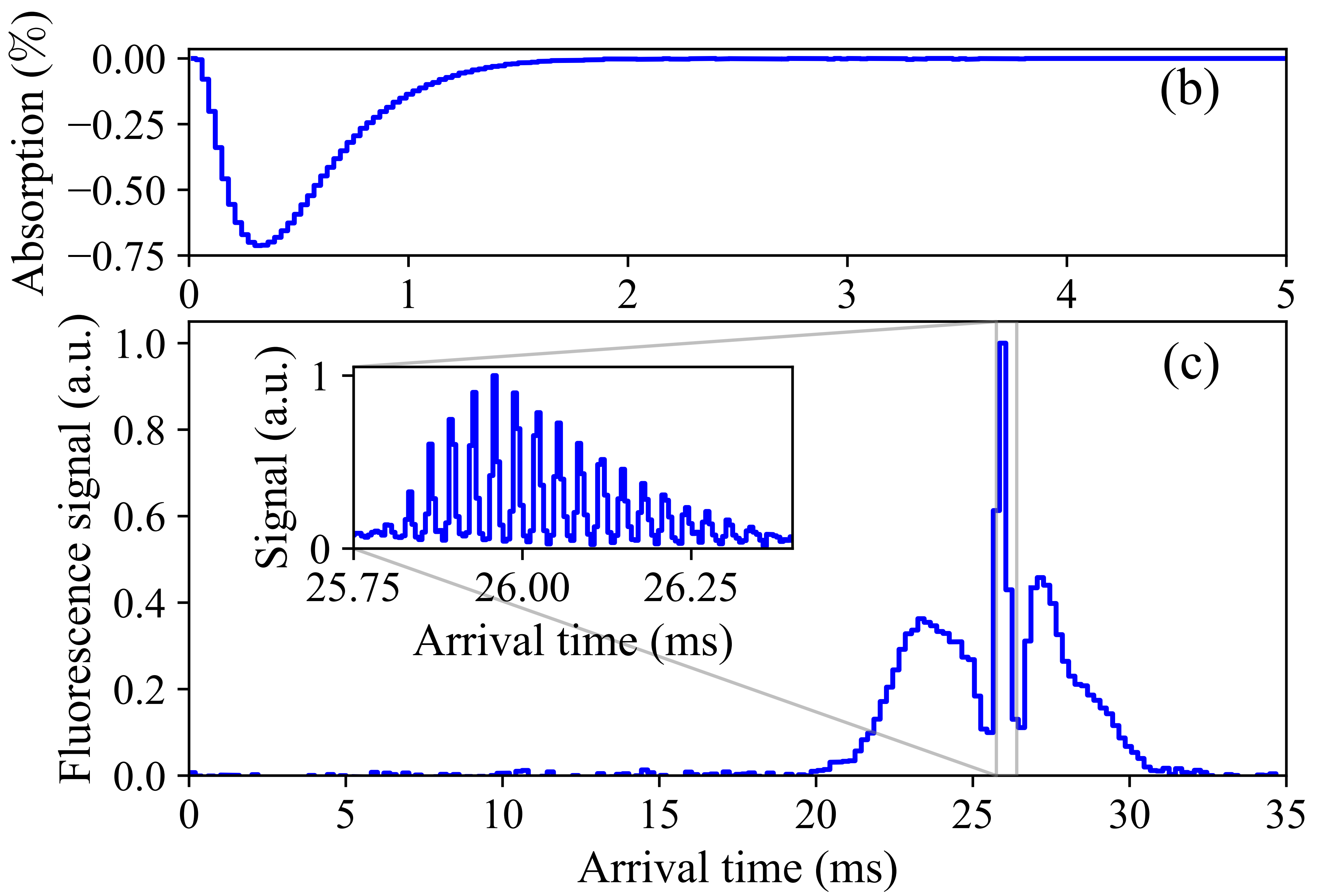
Traveling-wave decelerator

A slow beam of molecules

SrF: First combination of deceleration and cryogenic source

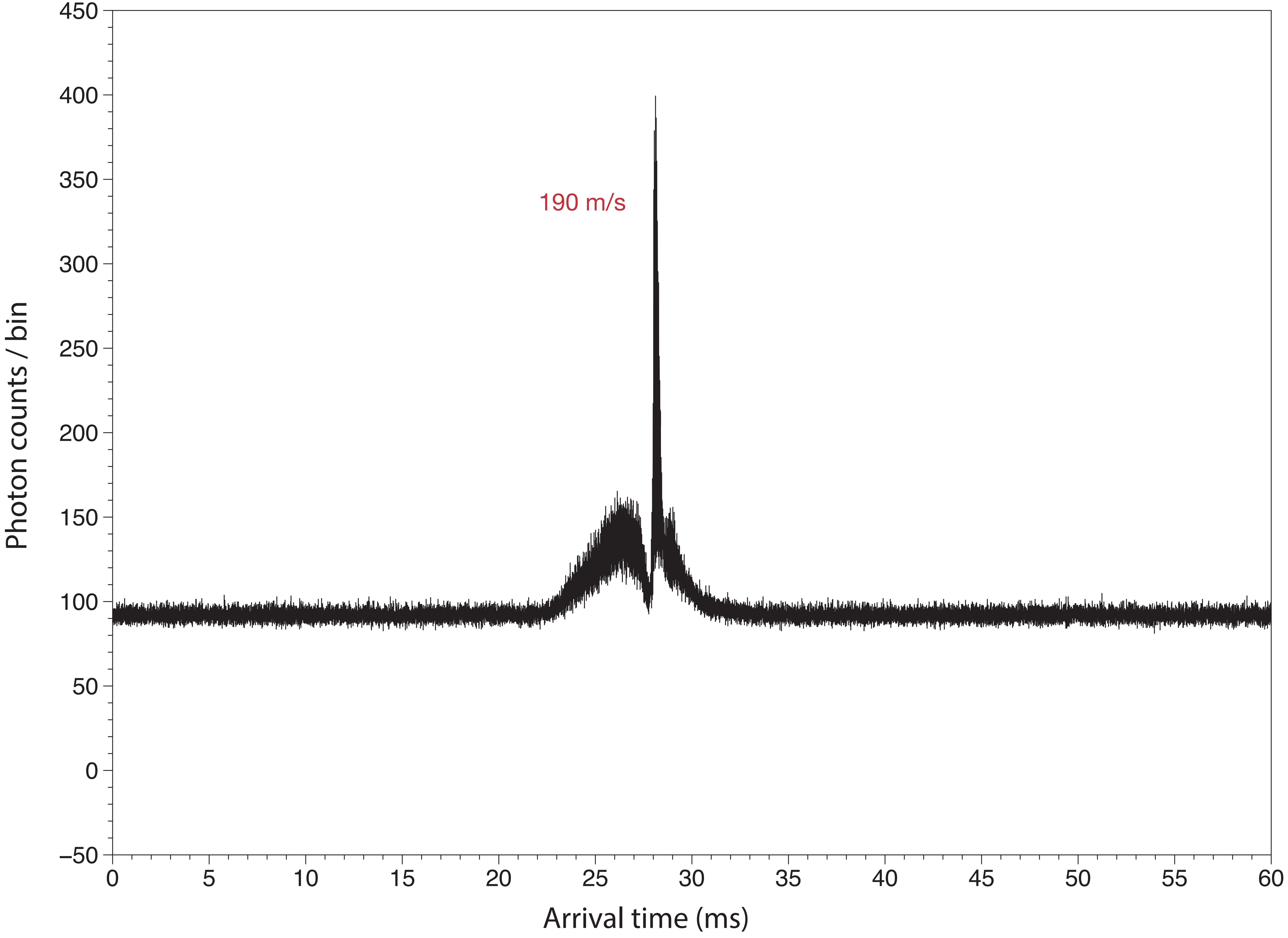
Aggarwal at al, PRL **127** 173201 (2021)





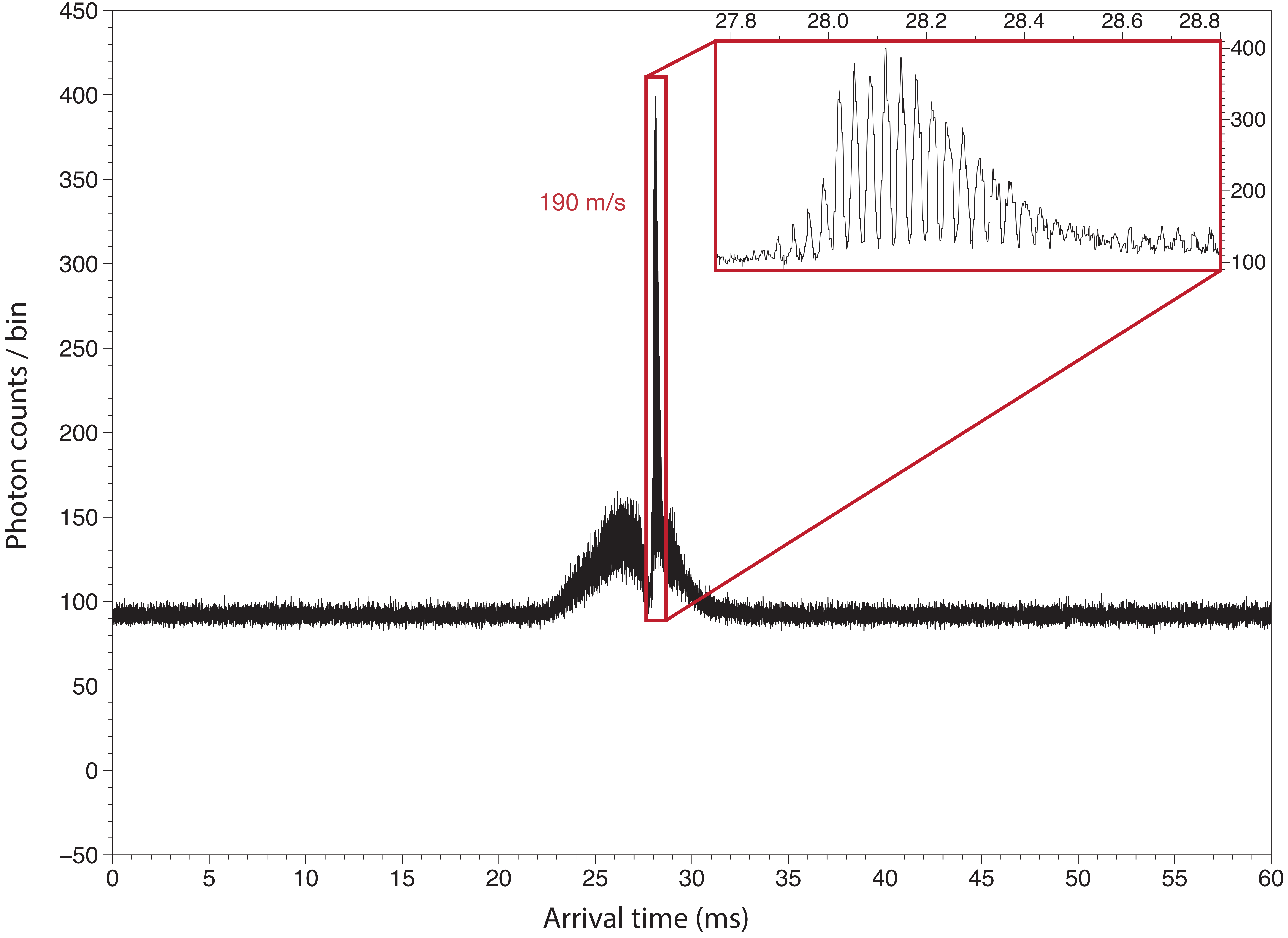
Decelerate to 30 m/s,

or bring to complete standstill



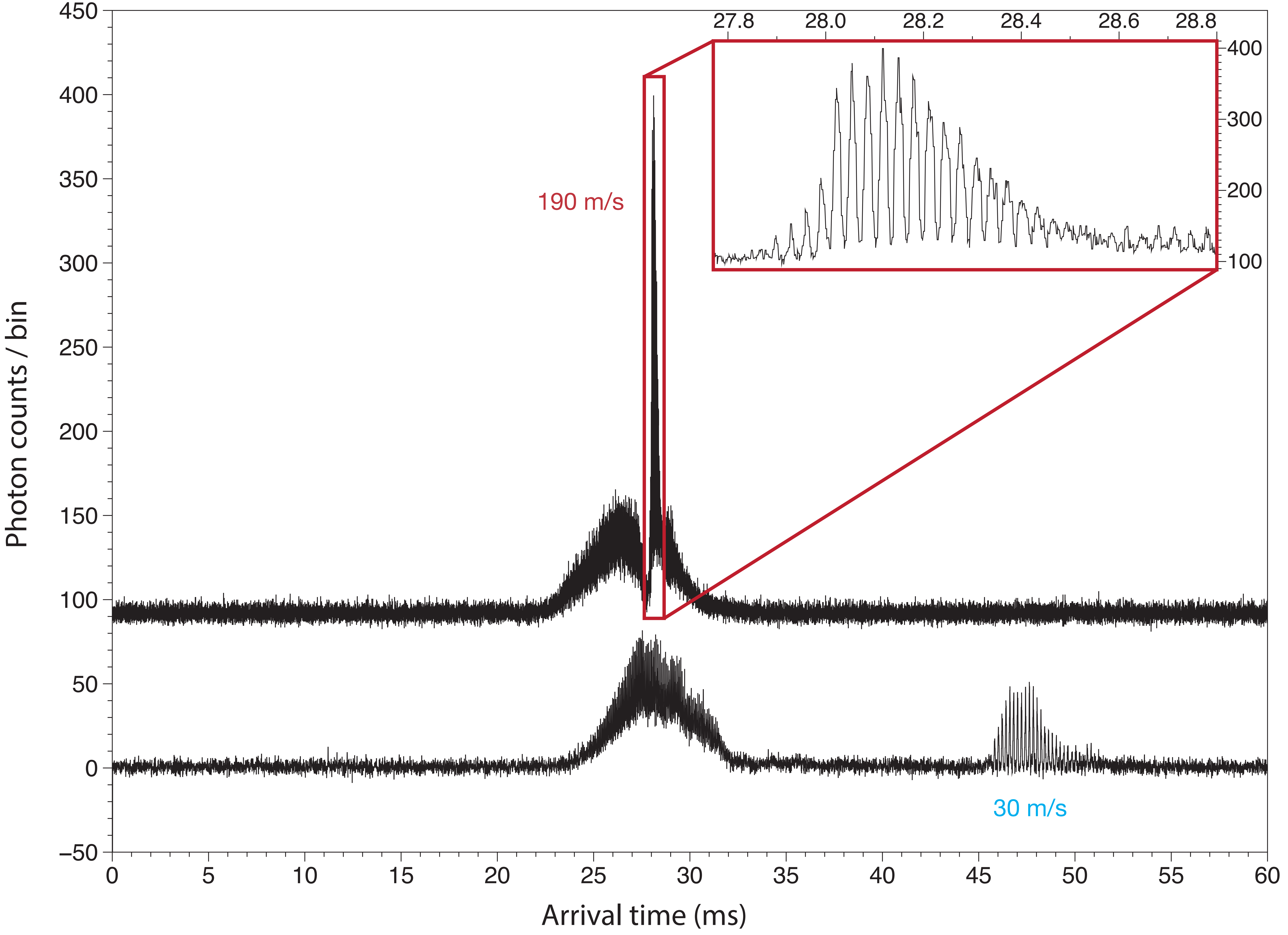
Decelerate to 30 m/s,

or bring to complete standstill



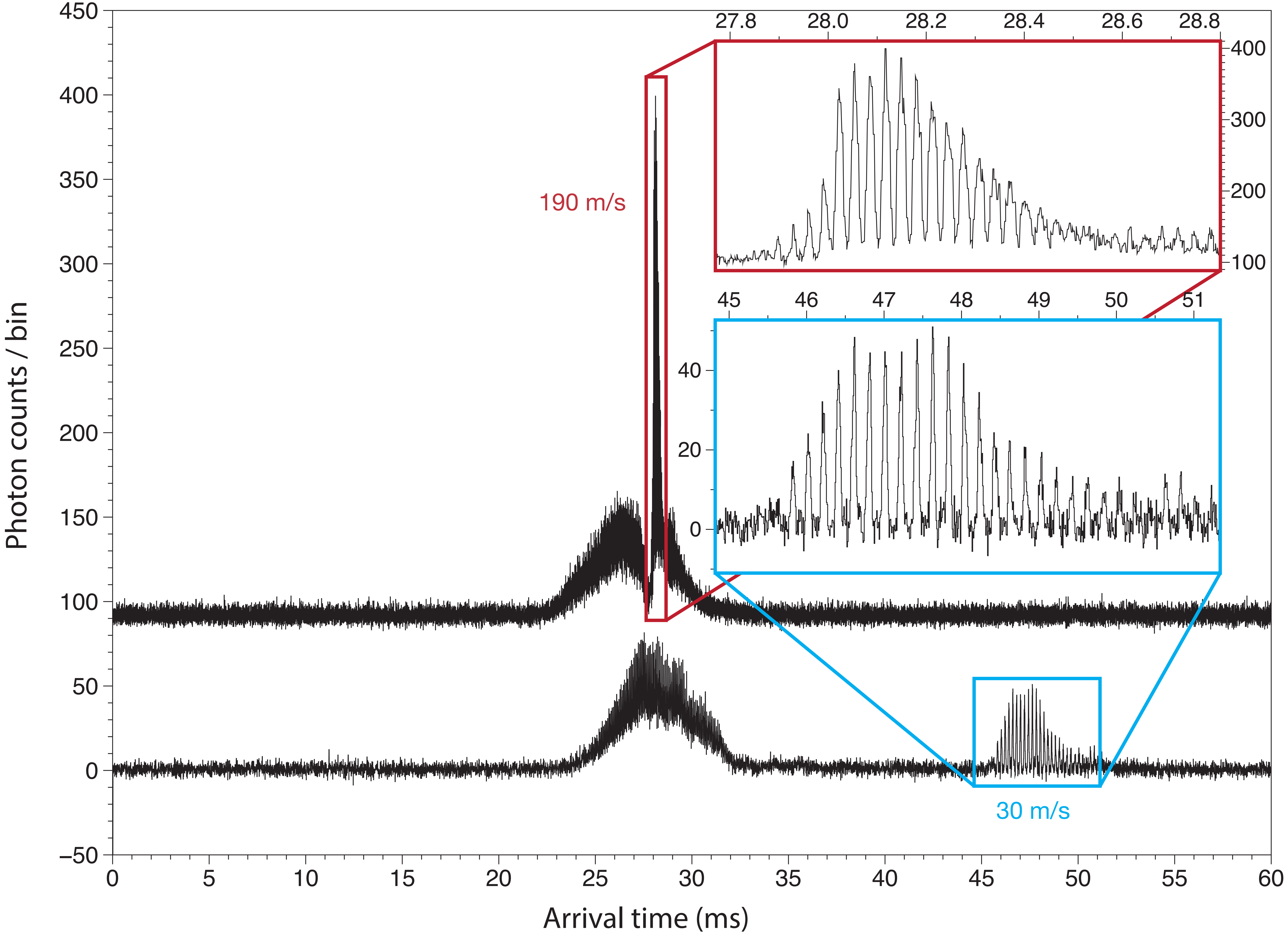
Decelerate to 30 m/s,

or bring to complete standstill



Decelerate to 30 m/s,

or bring to complete standstill

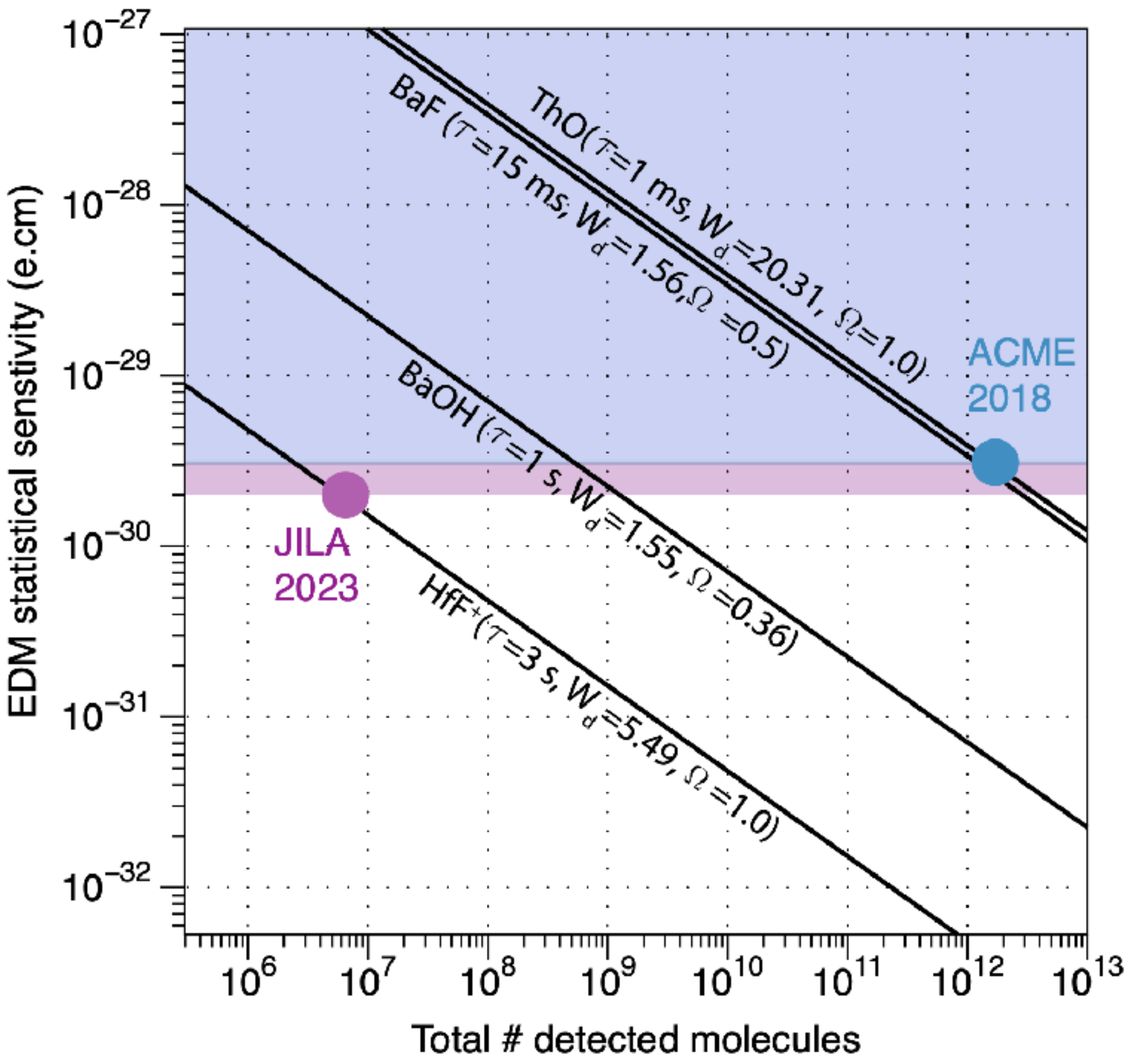


Future directions

- Radioactive molecules
  - Hold great promise due to extra enhancements
  - *How to get molecules into precision environment?*
- Polyatomic systems
  - Complexity brings advantages
  - *Can laser cooling still be done?*
- Trapped samples
  - Even longer interaction times
  - *What about the systematics?*

Outlook: even longer interaction times

Phase 3: Trapped molecules

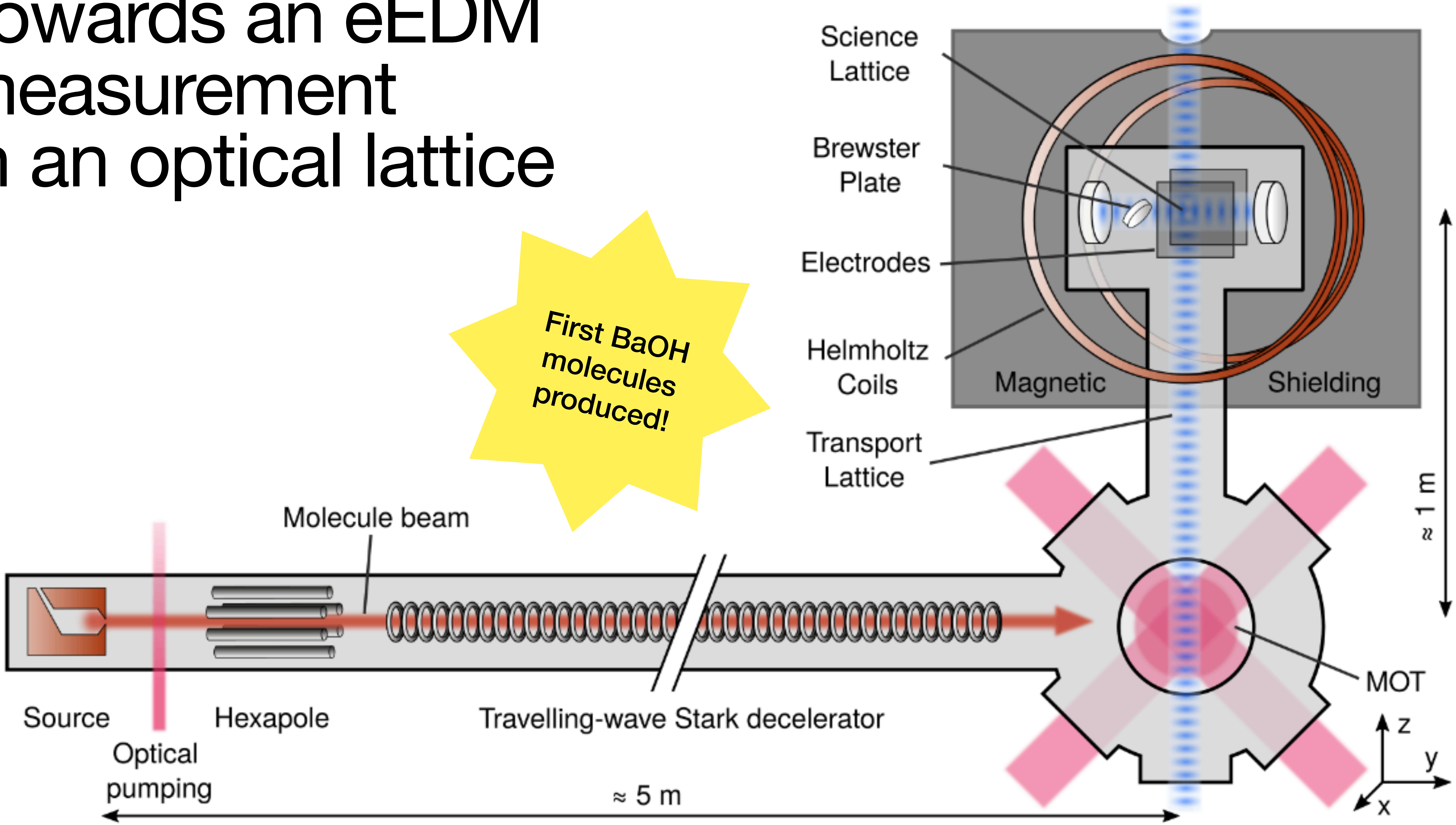


Bause et al,

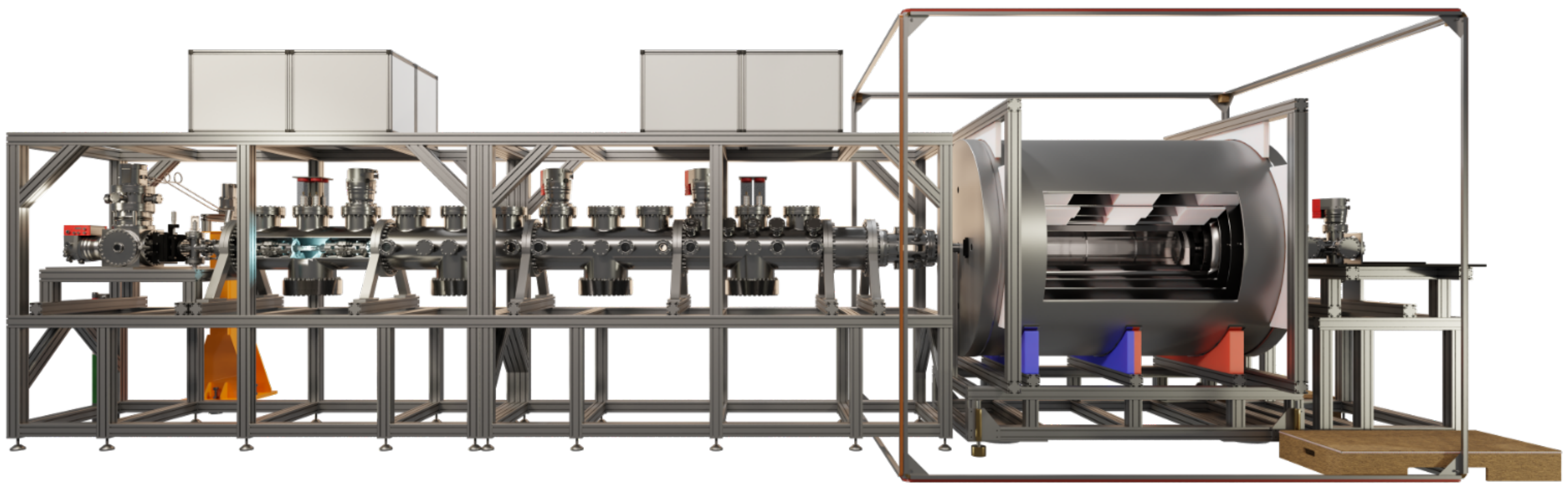
Prospects for measuring the electron's electric dipole moment with polyatomic molecules in an optical lattice,

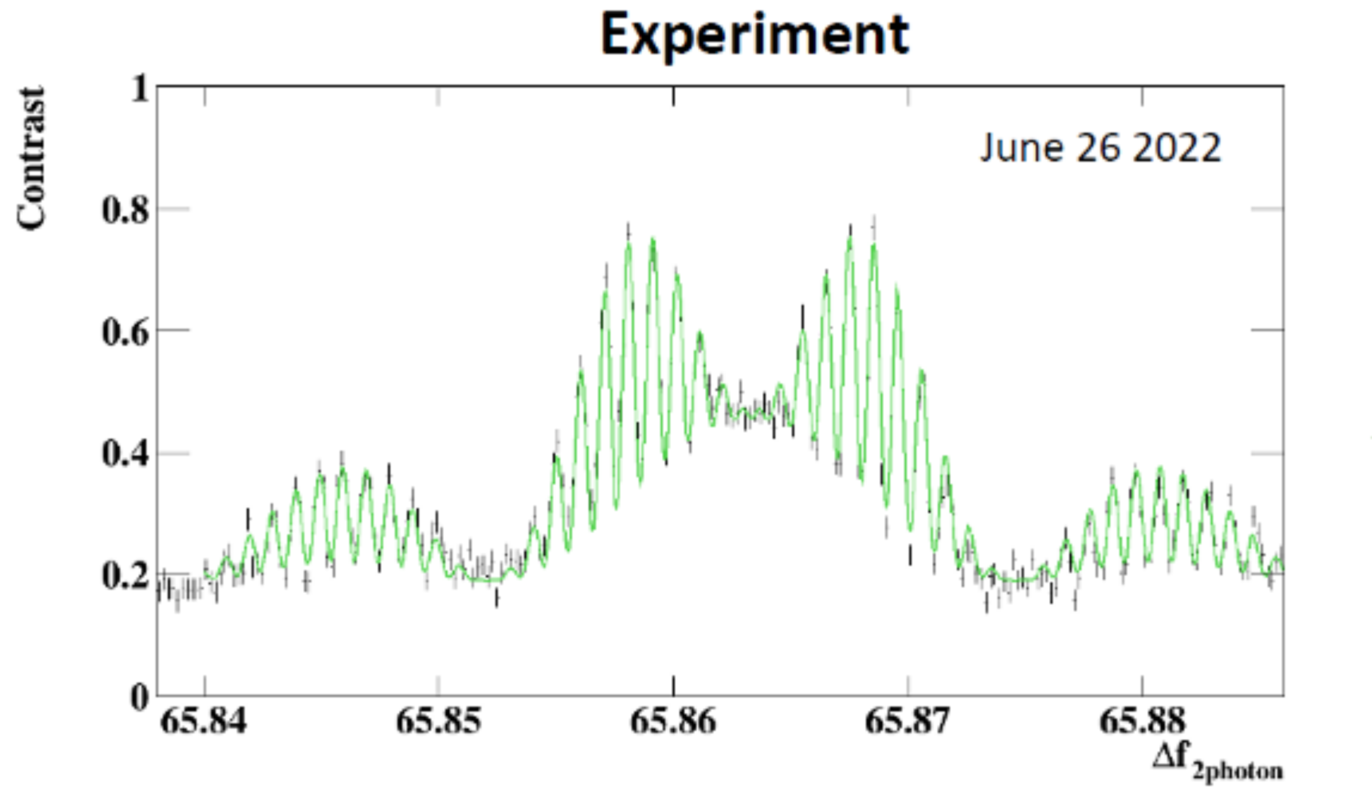
arXiv:2411.00441 (2024)

Towards an eEDM measurement in an optical lattice

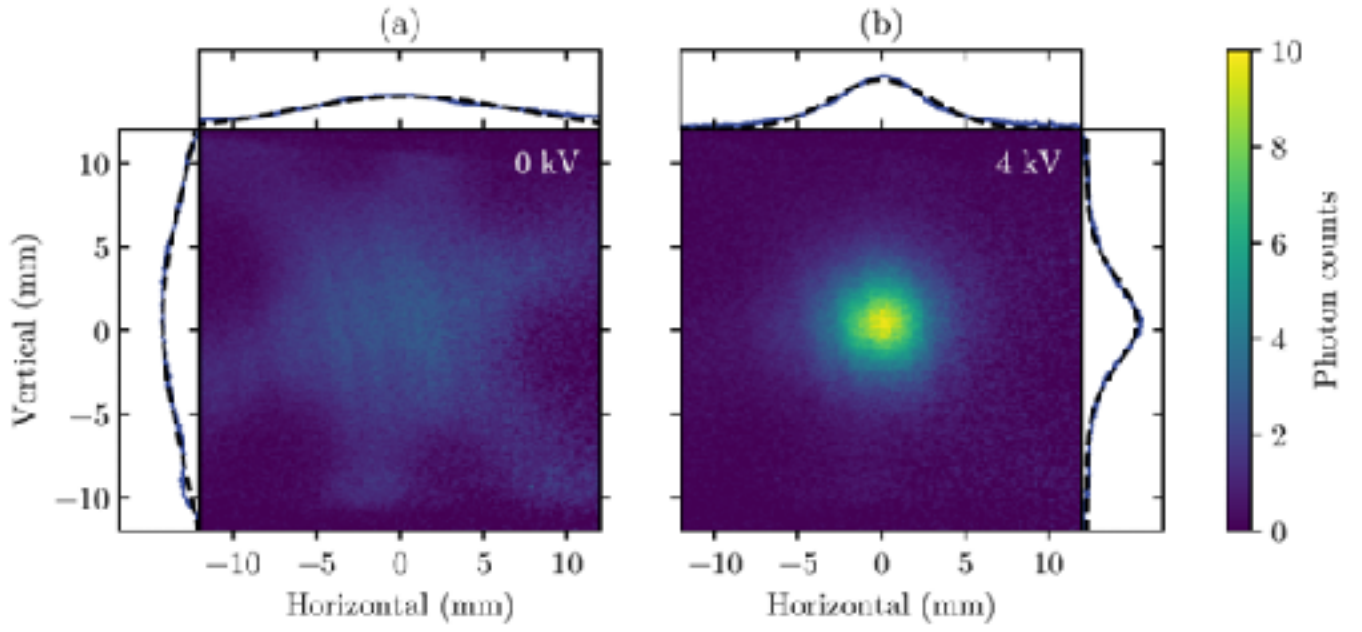


Summary

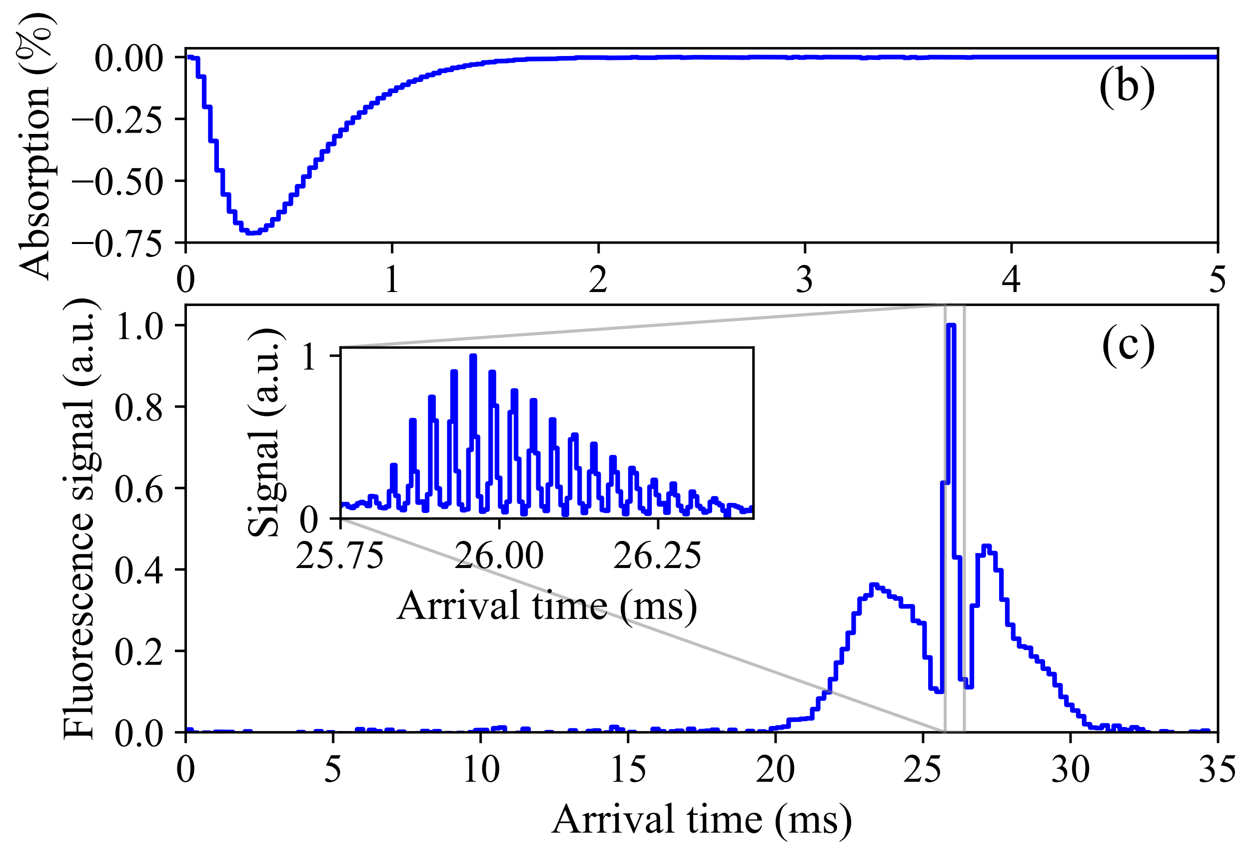




Spin interference demonstrated and understood



Intense and bright slow beam



Deceleration demonstrated

(part of) the team

university of groningen
van swinderen institute for particle physics and gravity

Nikhef
Dutch National Institute for (astro)Particle Physics

VU VRIJE UNIVERSITEIT AMSTERDAM

UvA