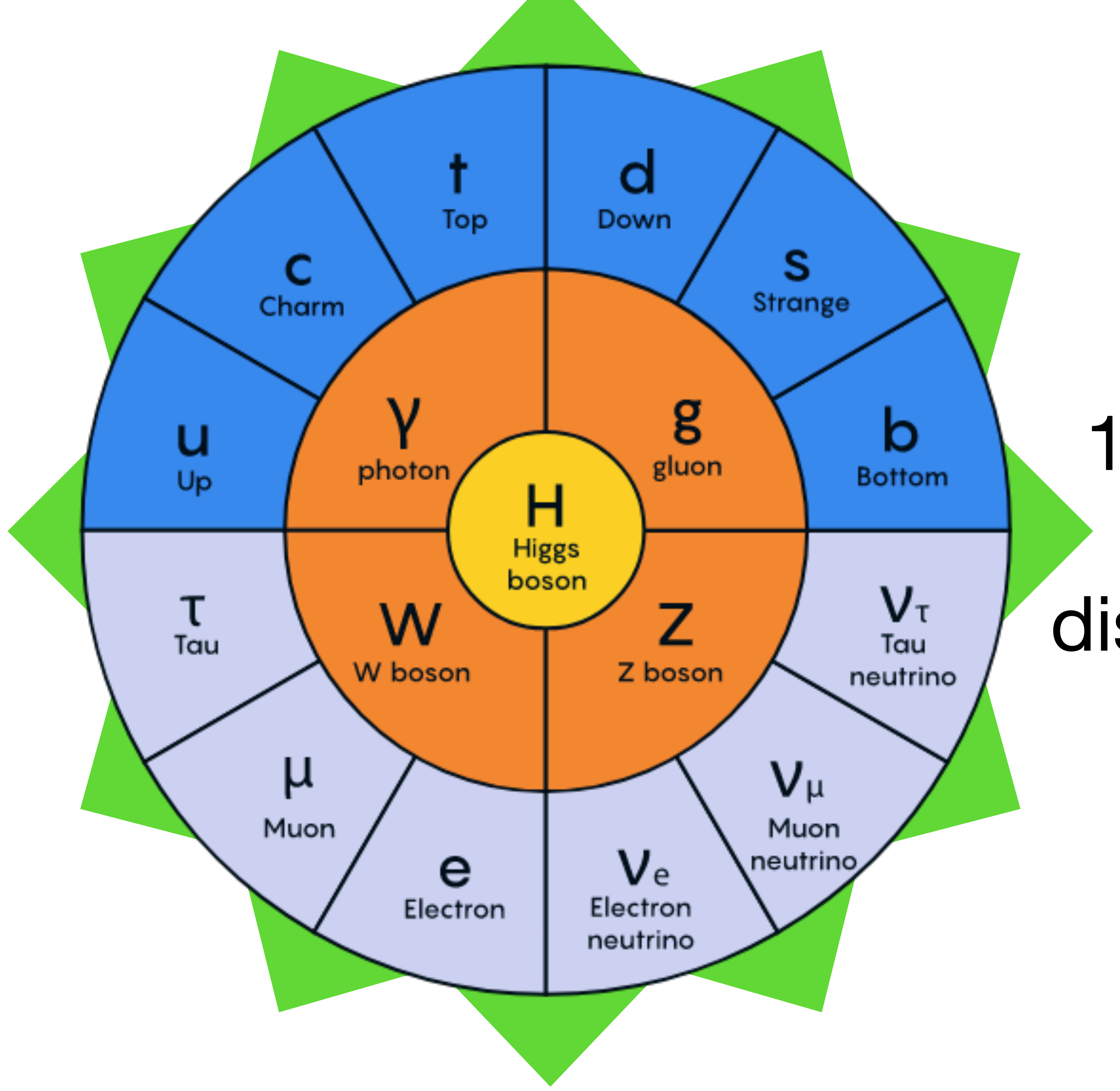
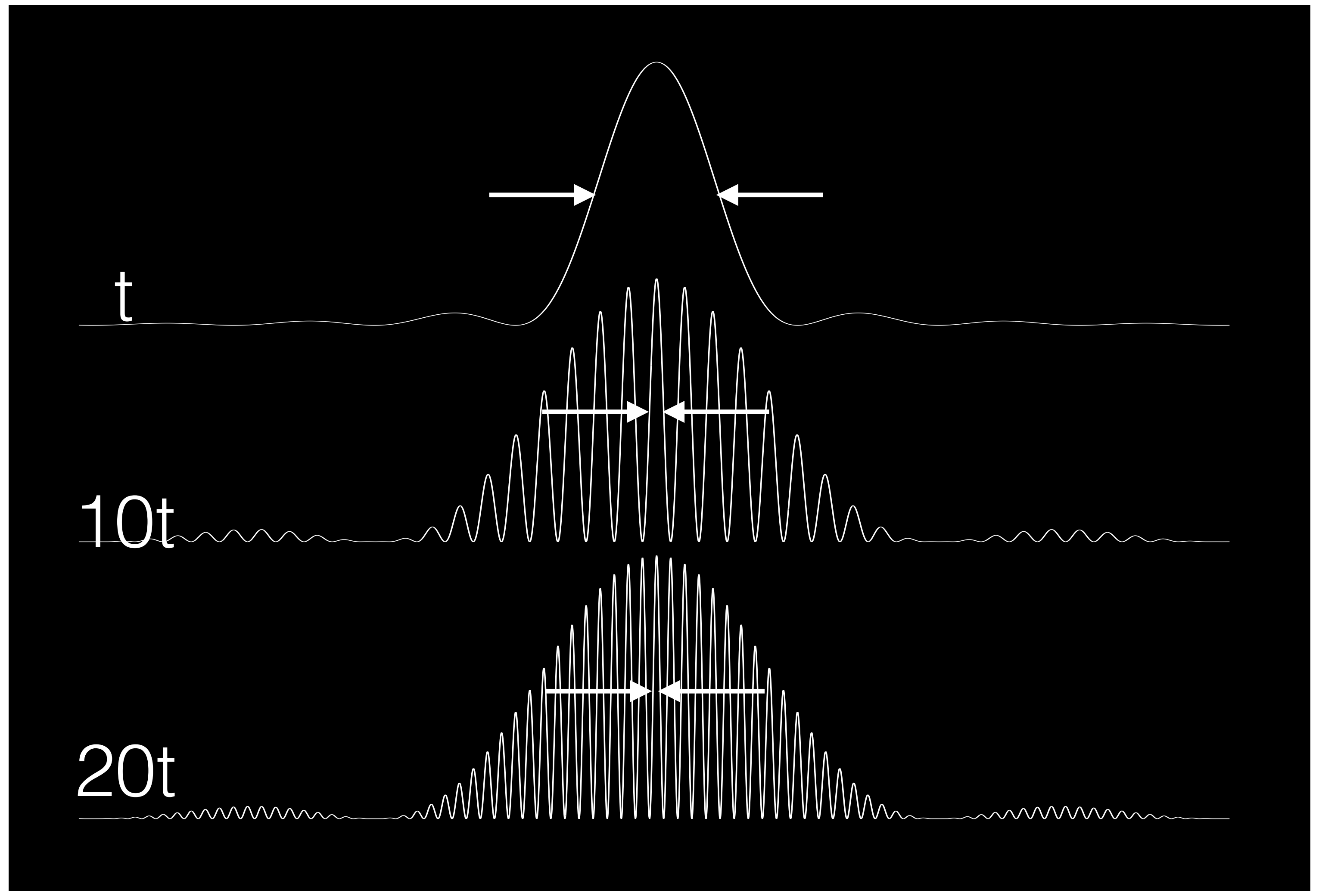
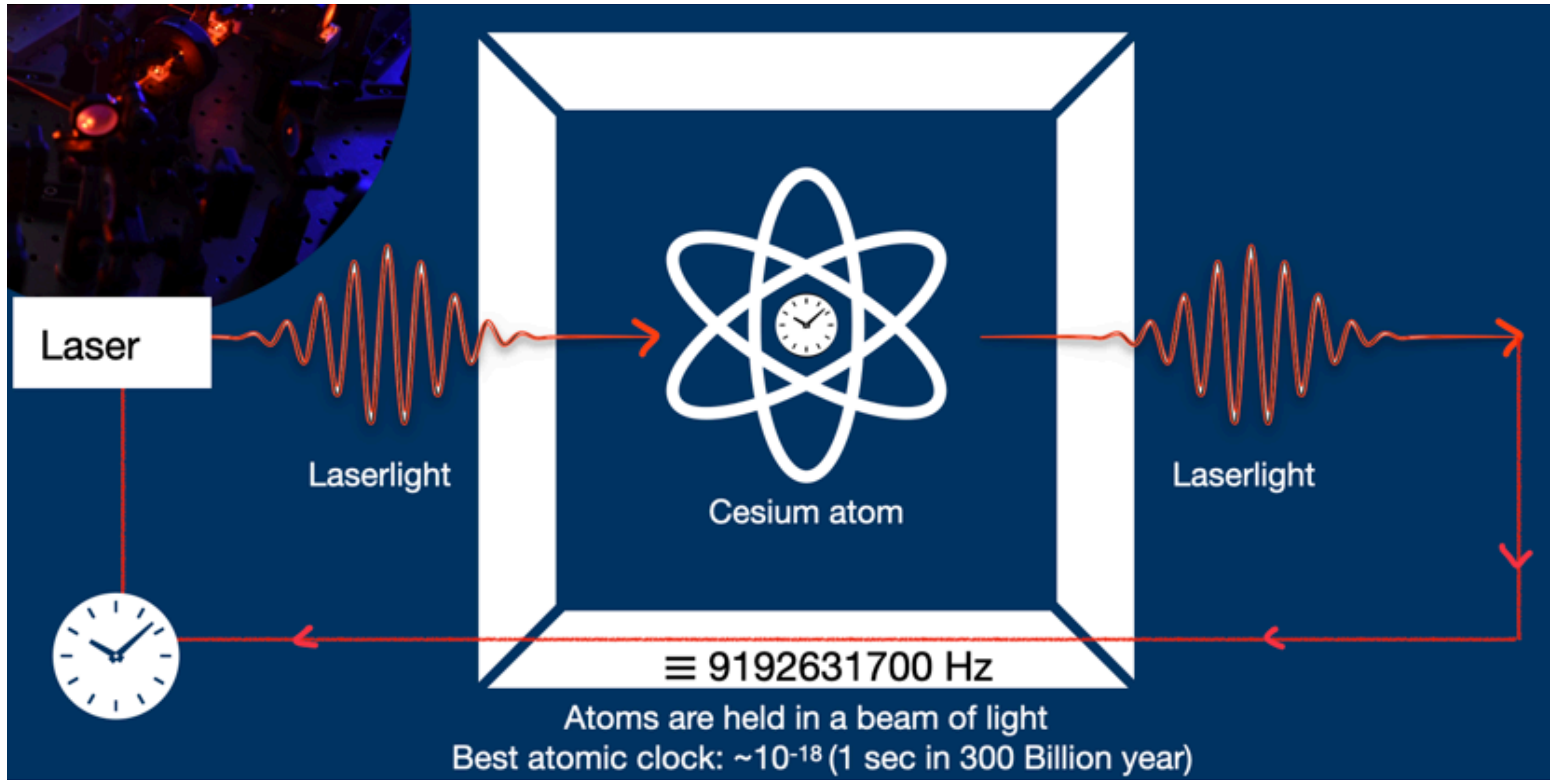
Summary of lecture 1

- We need to probe Standard model extensions
- Inspired by atomic clock precision, we would like to use atoms and molecules to experimentally probe ‘new physics’
- Ramsey interference is the most precise method of spectroscopy



12 elementary particles, their interaction, discrete symmetries C,P,T





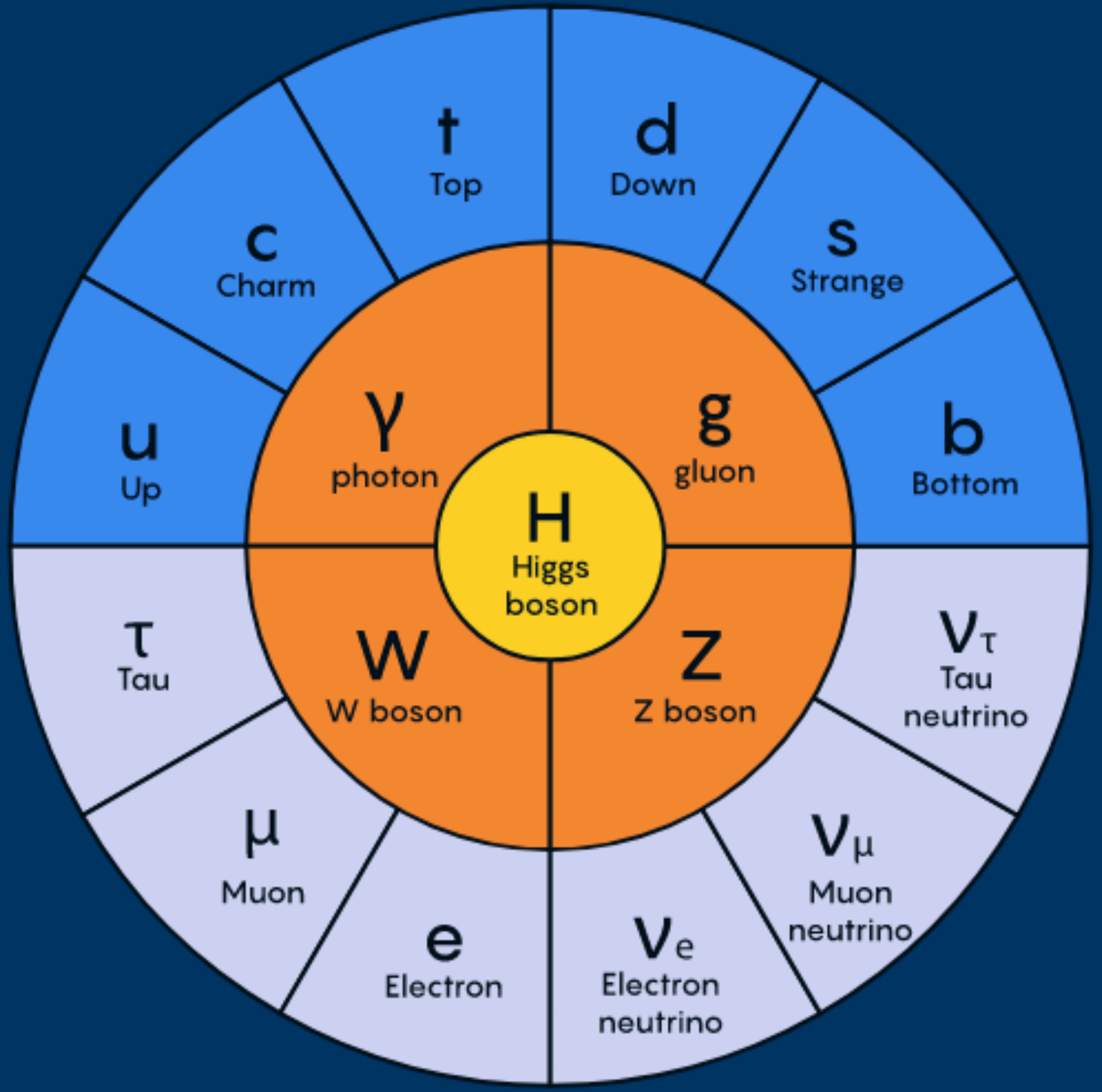
Part 2 - the electric dipole moment of the electron

and using molecules to probe it

- Precision measurements using molecules
- The electron’s electric dipole moment
- Experimental approaches

What makes a molecular clock tick?

The Standardmodel
of particle physics



12 elementary particles,
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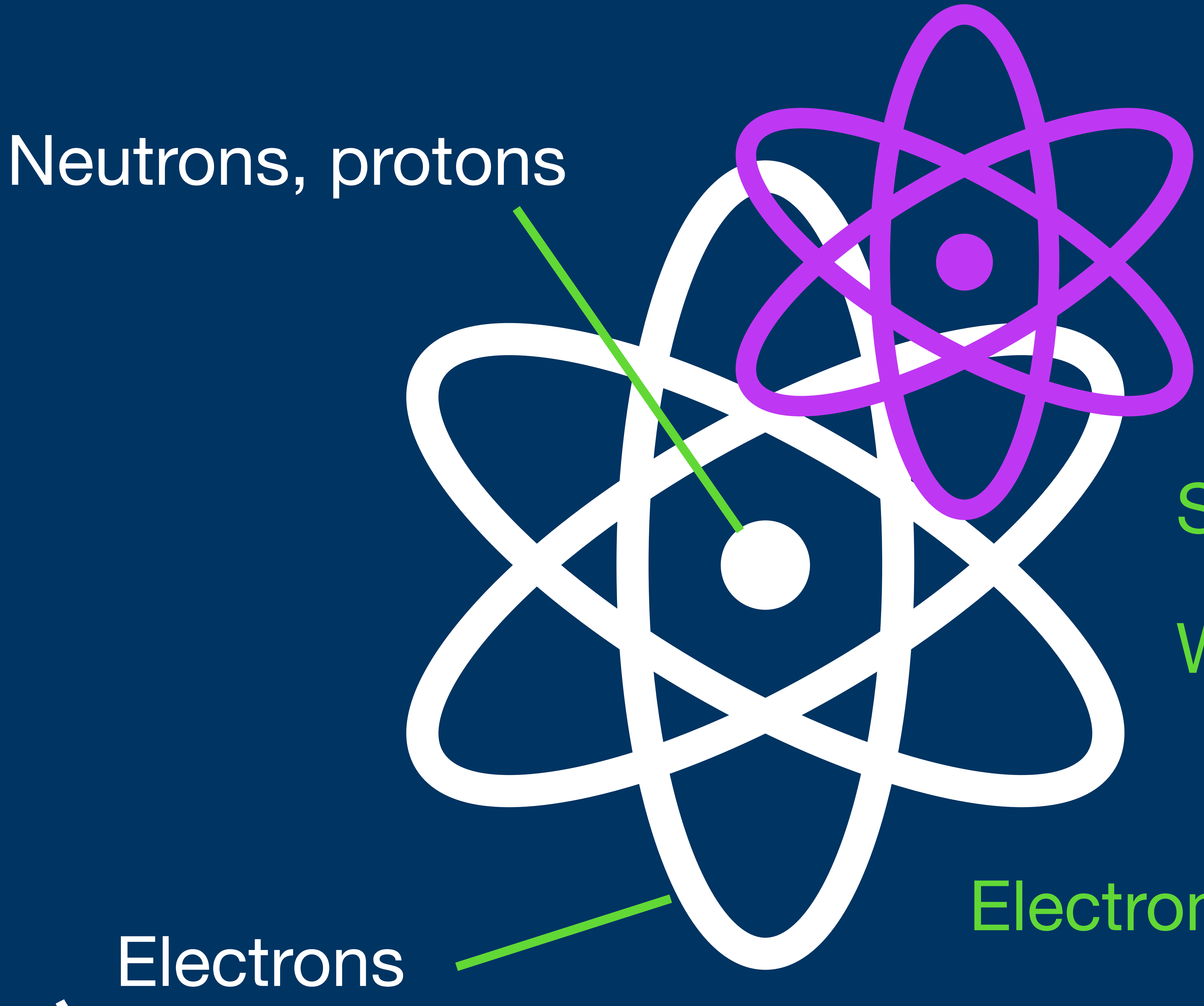


Strong force

Weak force

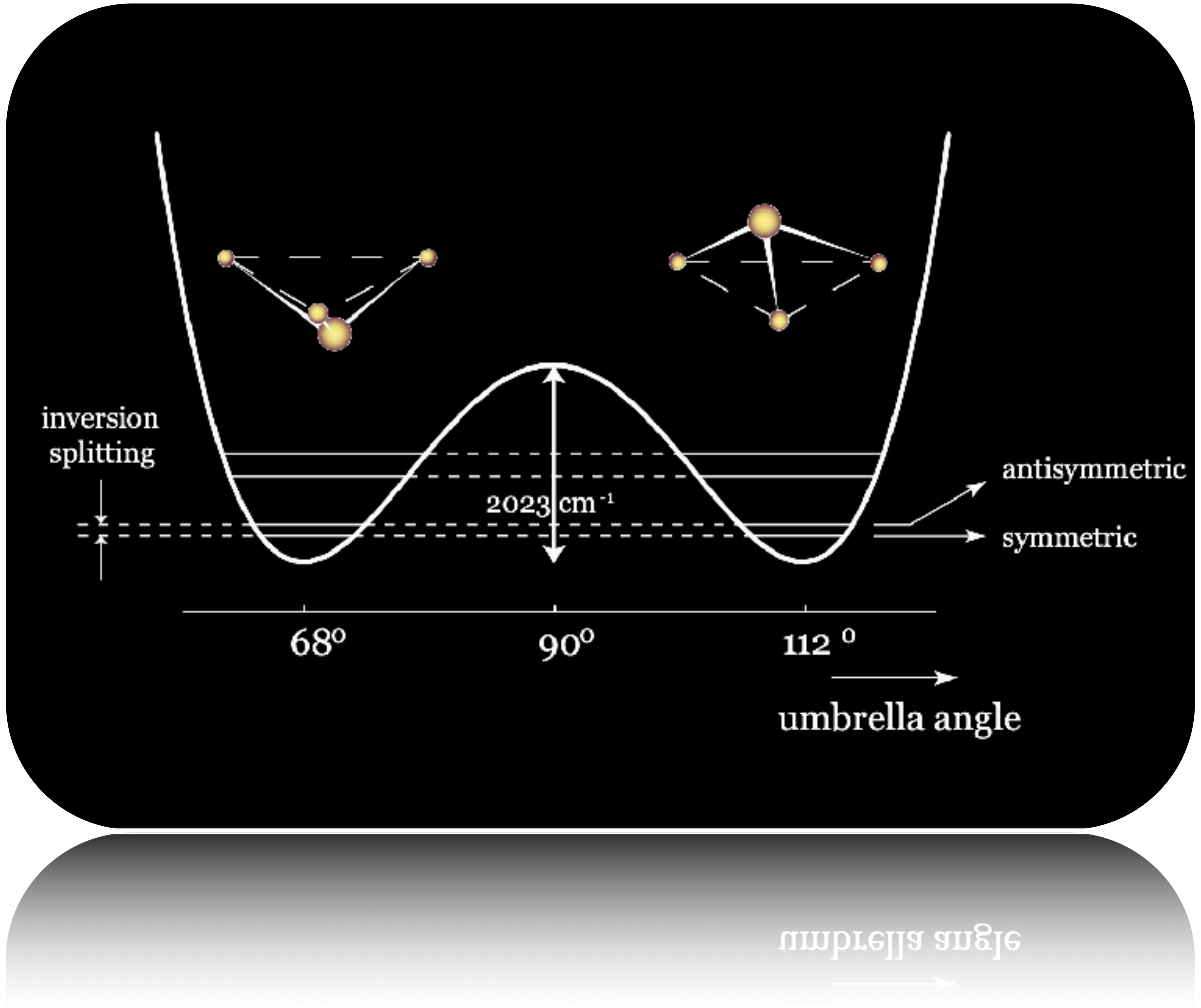
Electromagnetic force

The forces and symmetries in the molecule
reflect those of the Standardmodel

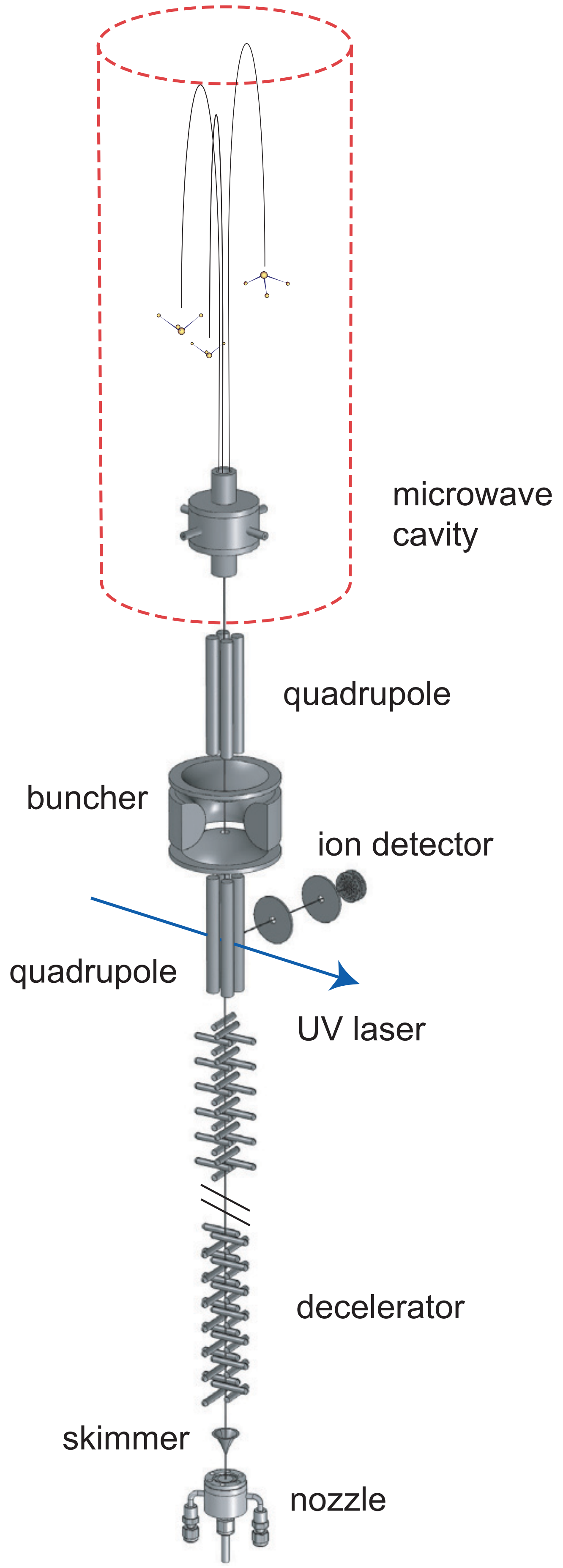
Precision measurements with molecules

Complex quantum systems with an advantage

Example 2: Variation of constants



Very sensitive to proton / electron mass ratio



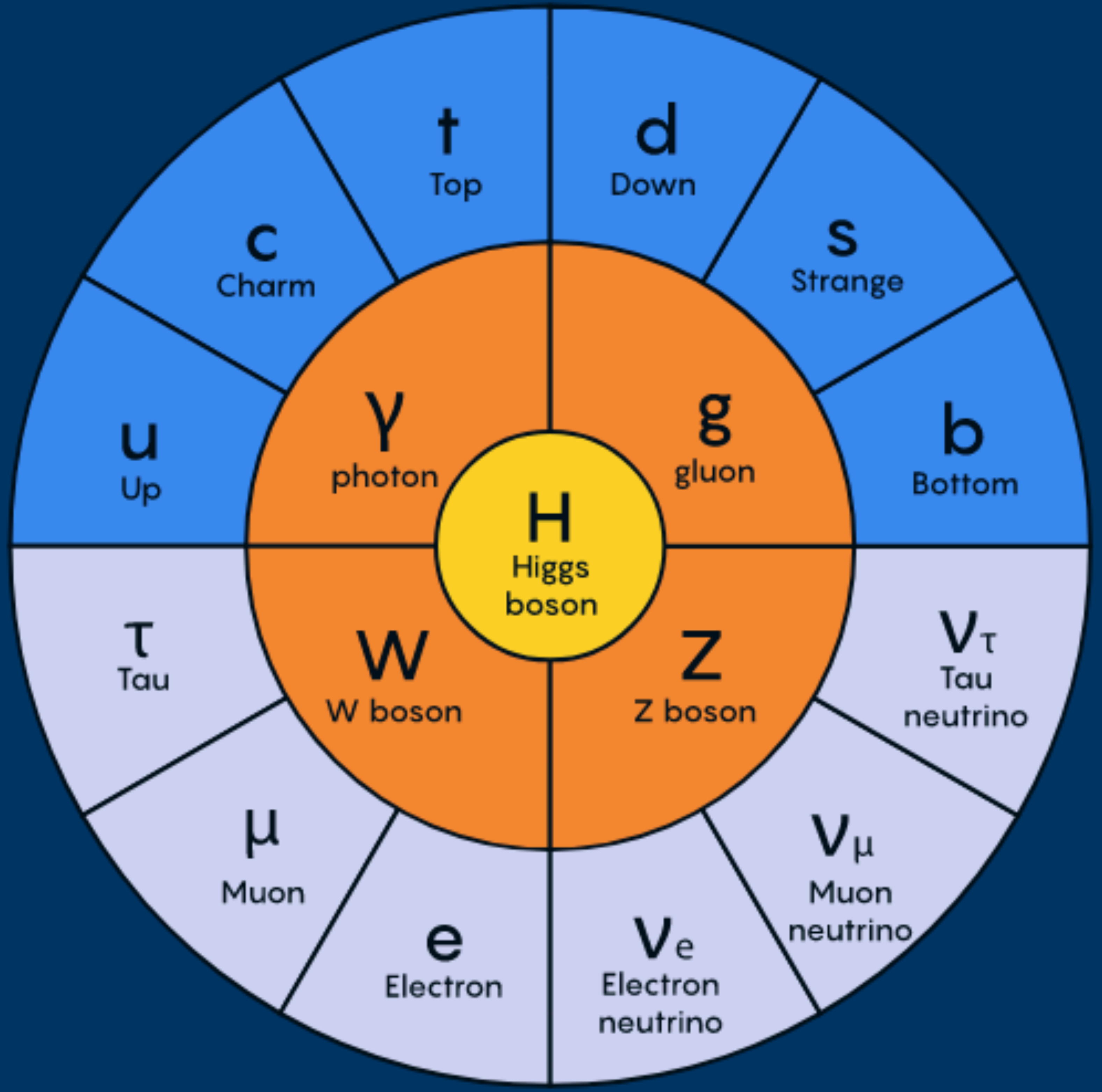
proposed experiment

?

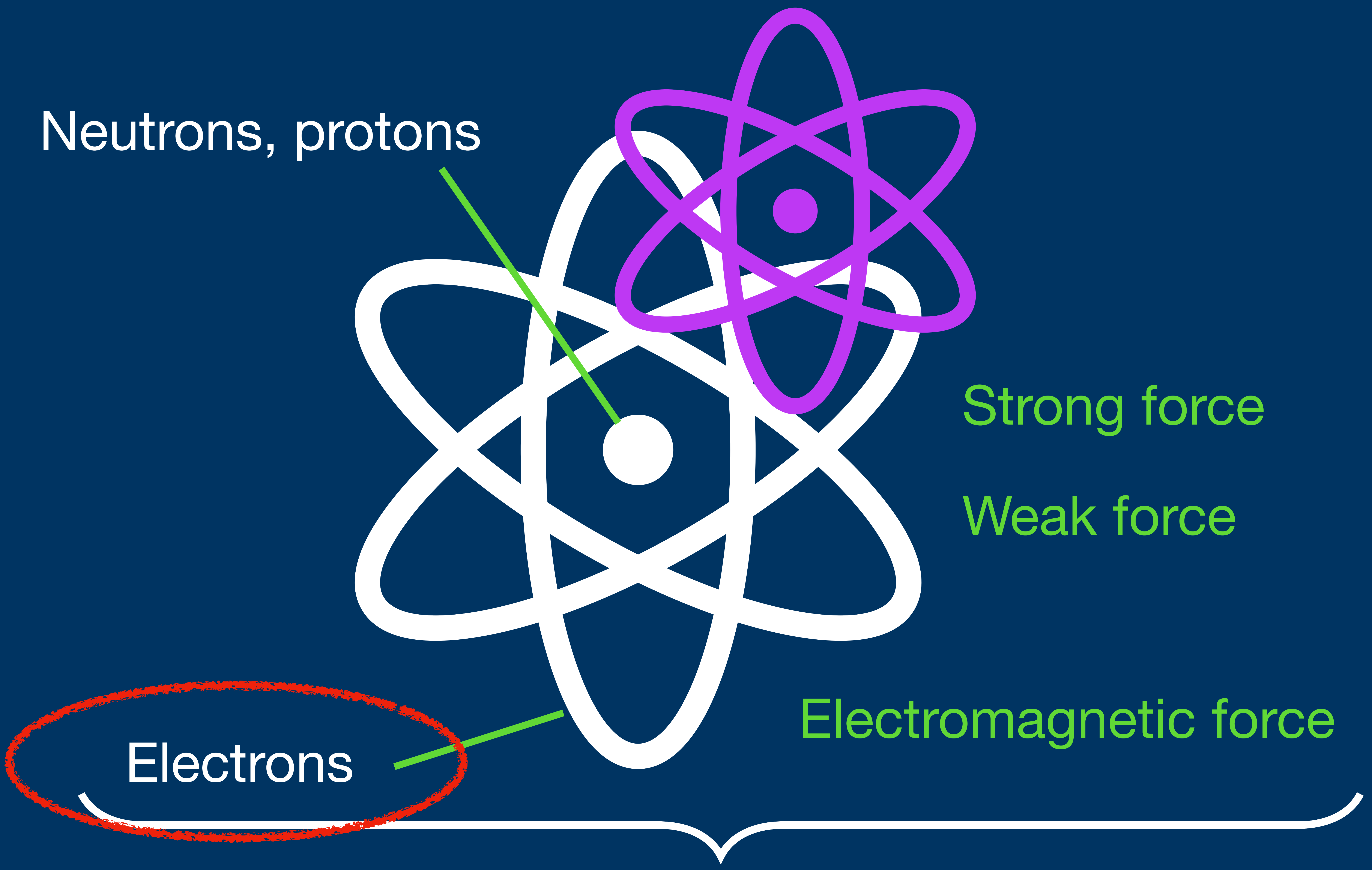
Theory

What makes a molecular clock tick?

The Standardmodel
of particle physics



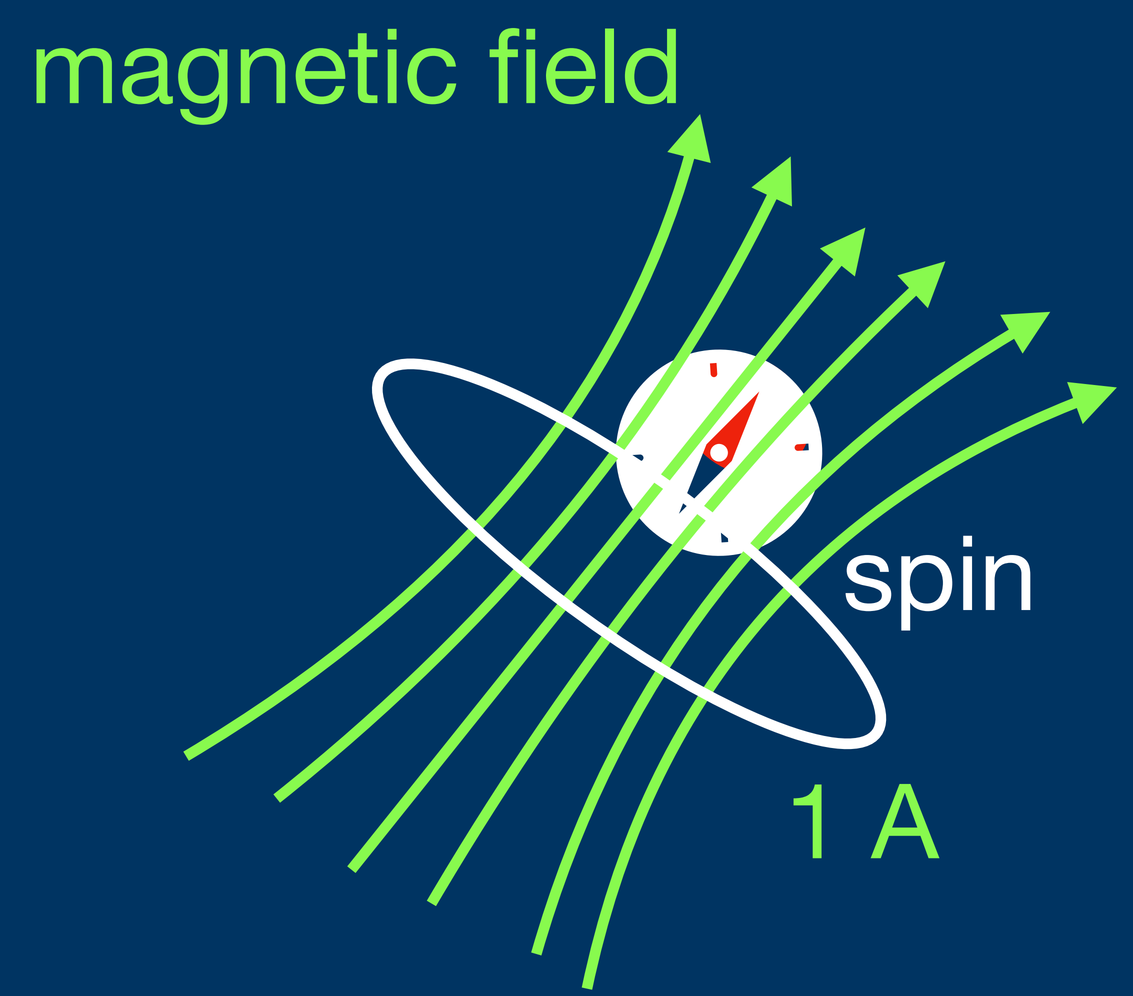
12 elementary particles,
their interaction,
discrete symmetries C,P,T

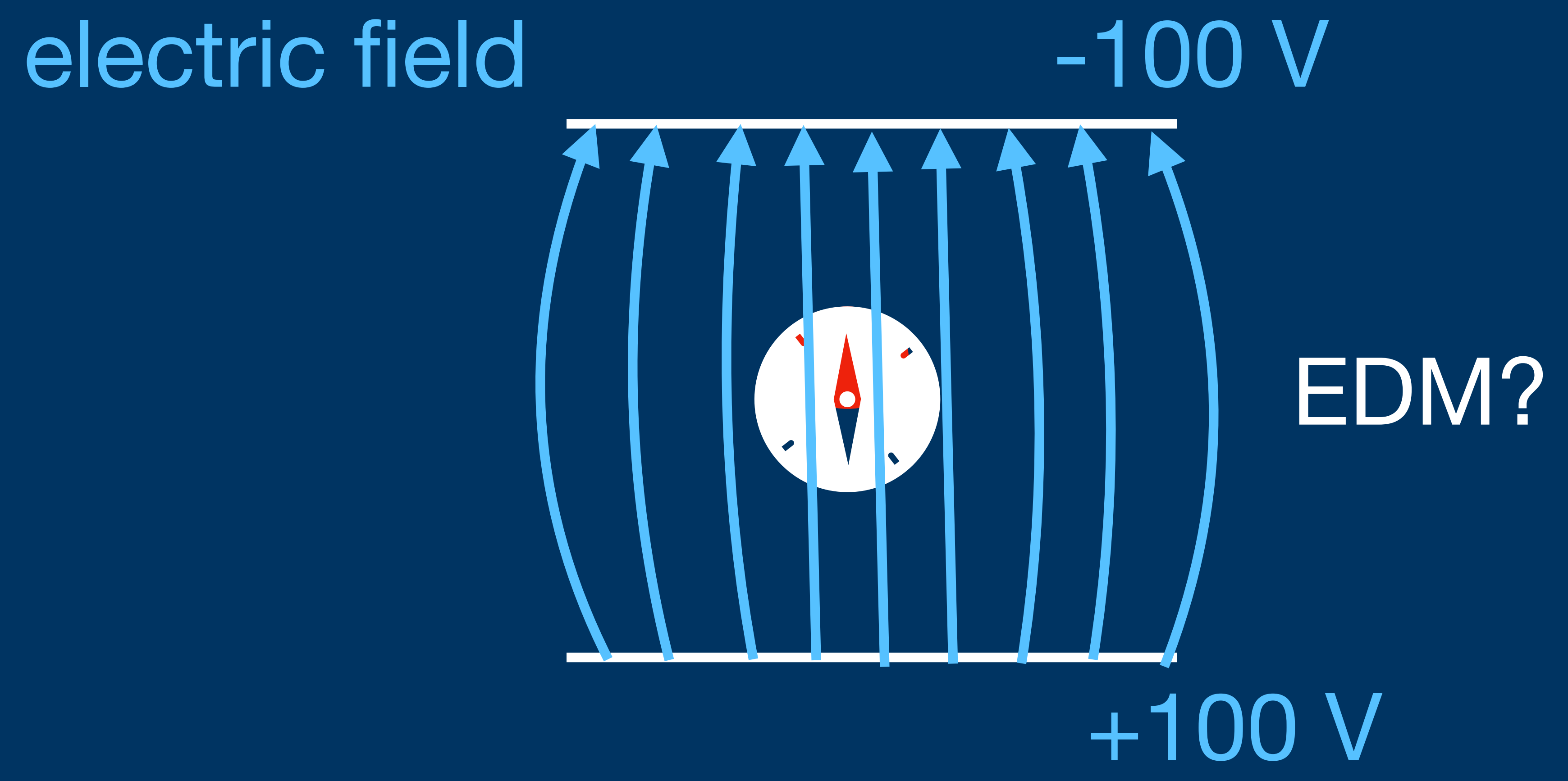


The forces and symmetries in the molecule
reflect those of the Standardmodel

The dipole moments of the electron

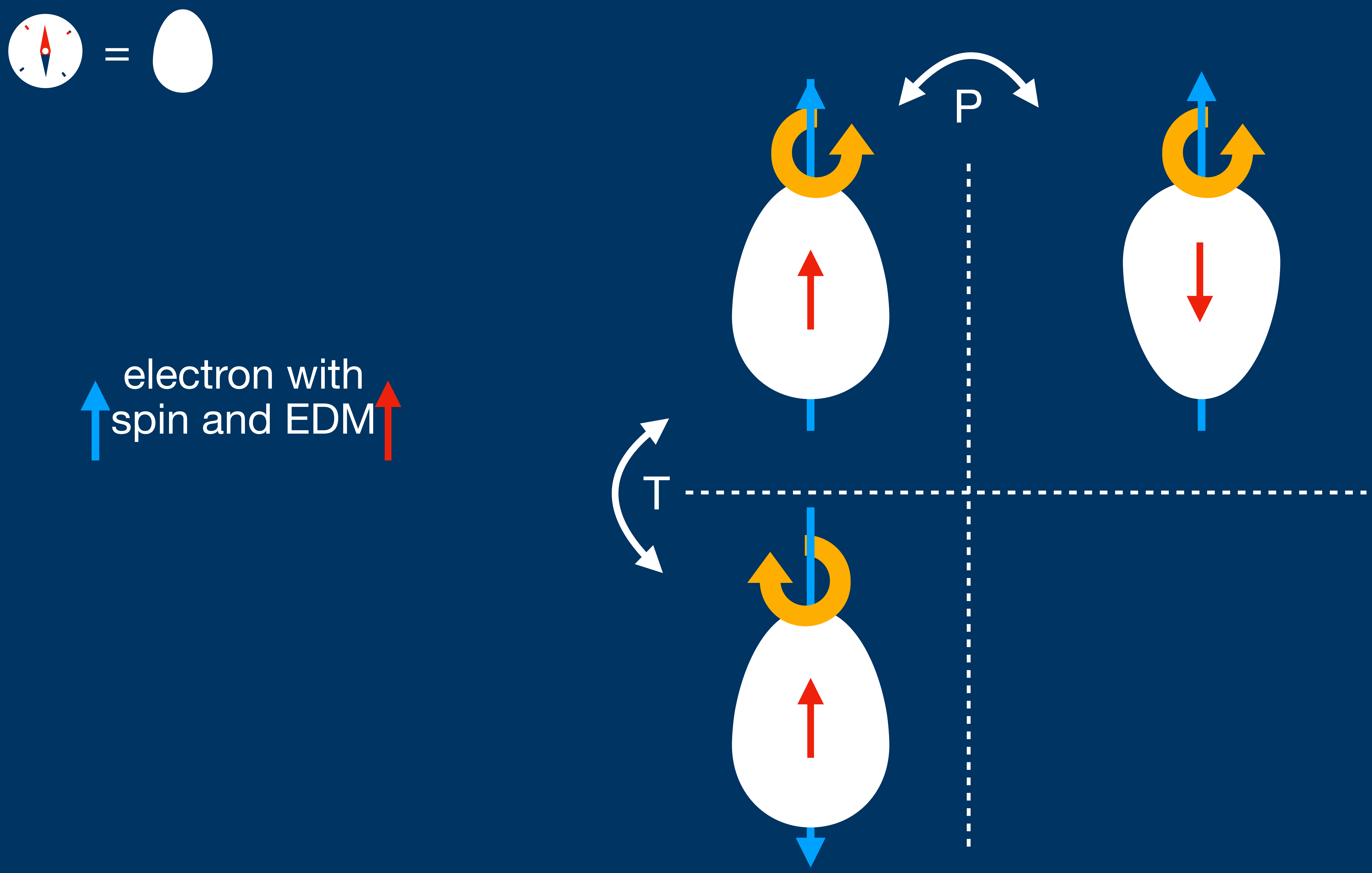
Besides its magnetic dipole moment (spin), an electron could have an *electric dipole moment (eEDM).*





an eEDM violates Time reversal symmetry (T) and is direct proof of new physics!

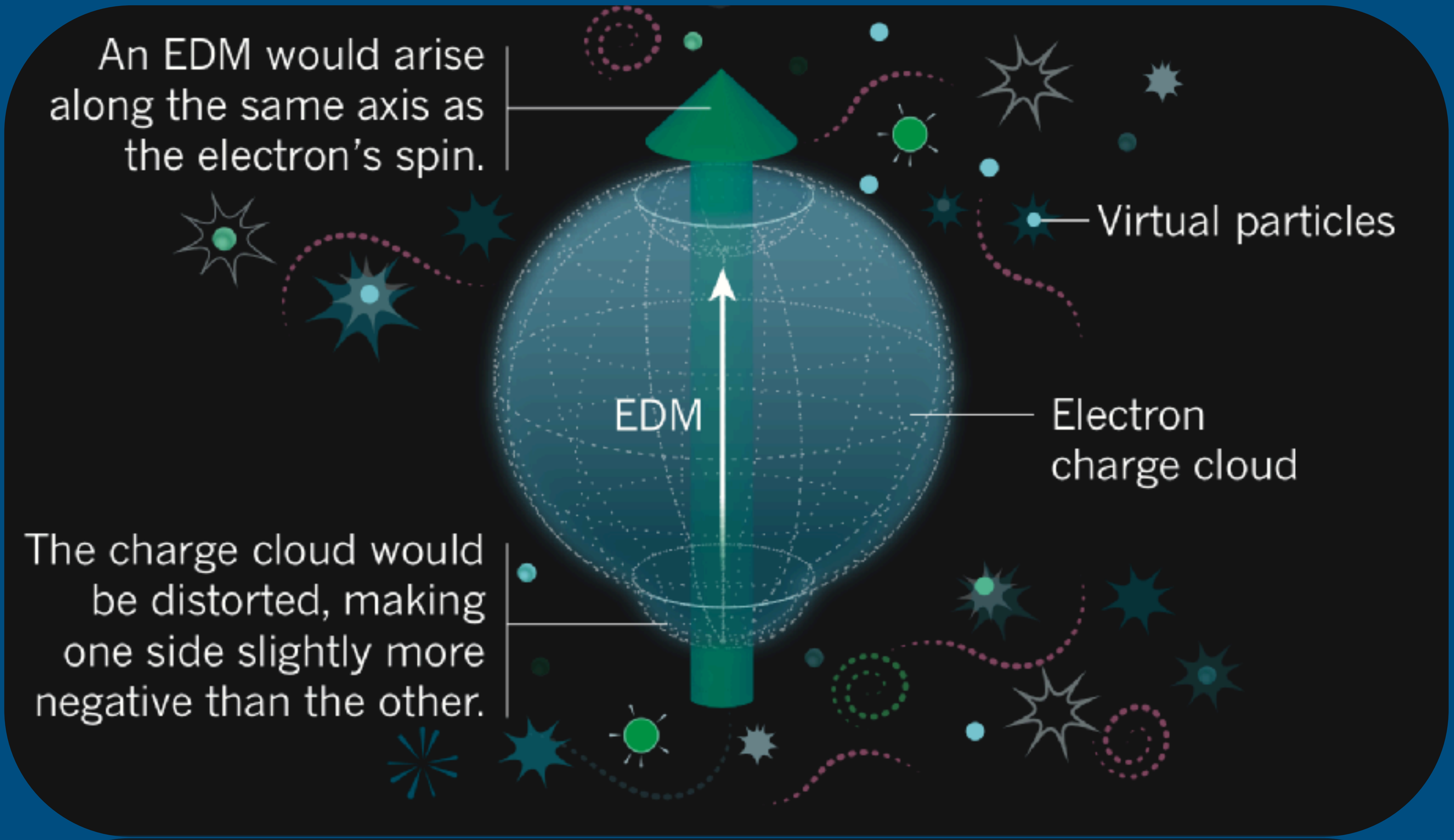
An eEDM violates T (and P) symmetry



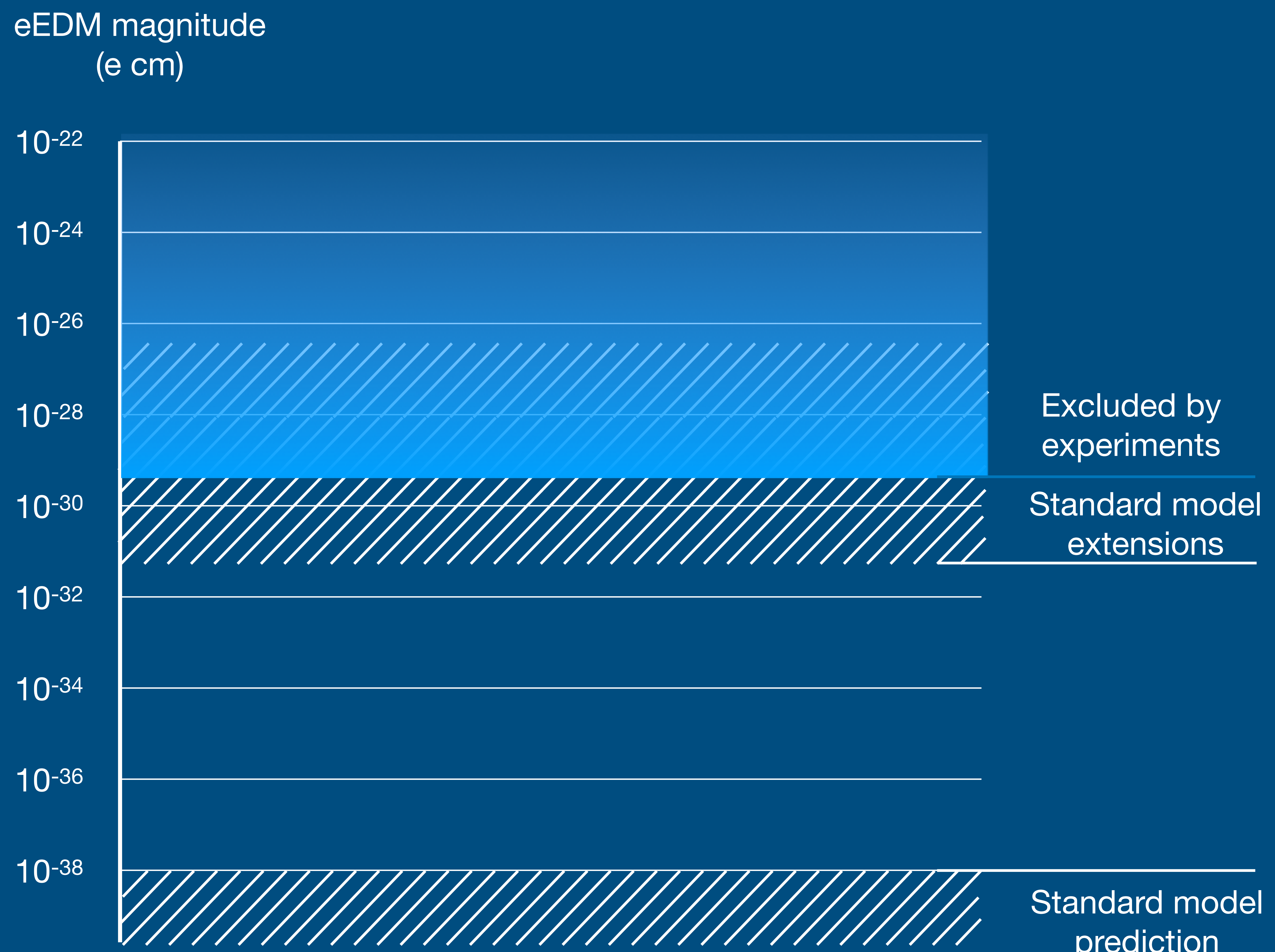
Precision measurements with molecules

Complex quantum systems with an advantage

Example 3: The electric dipole moment of the electron

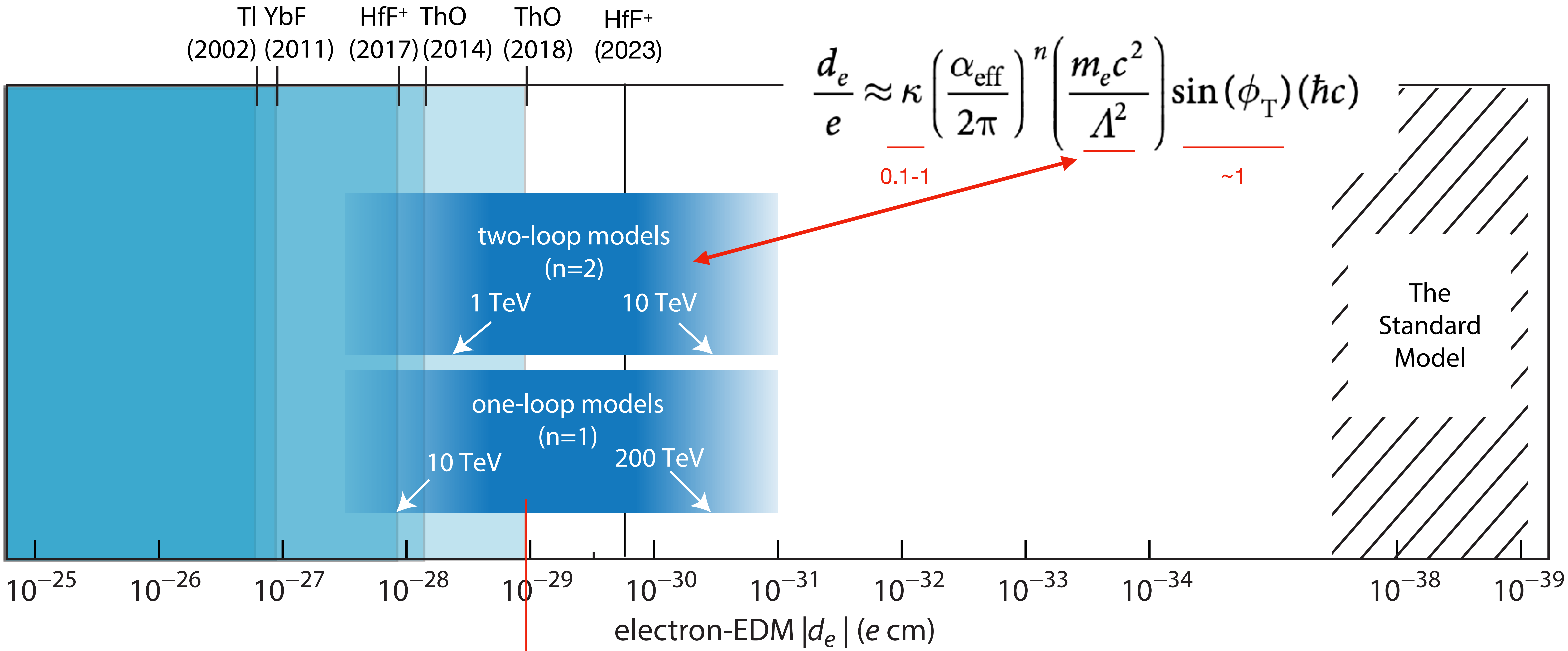


eEDM violates P, T and CP symmetry
(provided CPT holds)



The electron's electric dipole moment (eEDM)

Effectively a background-free method to probe new physics



The ThO result limits time-reversal-symmetry-violating new physics to energy scales above $\Lambda \approx 30$ TeV or $\Lambda \approx 3$ TeV, for n=2 or 1, respectively

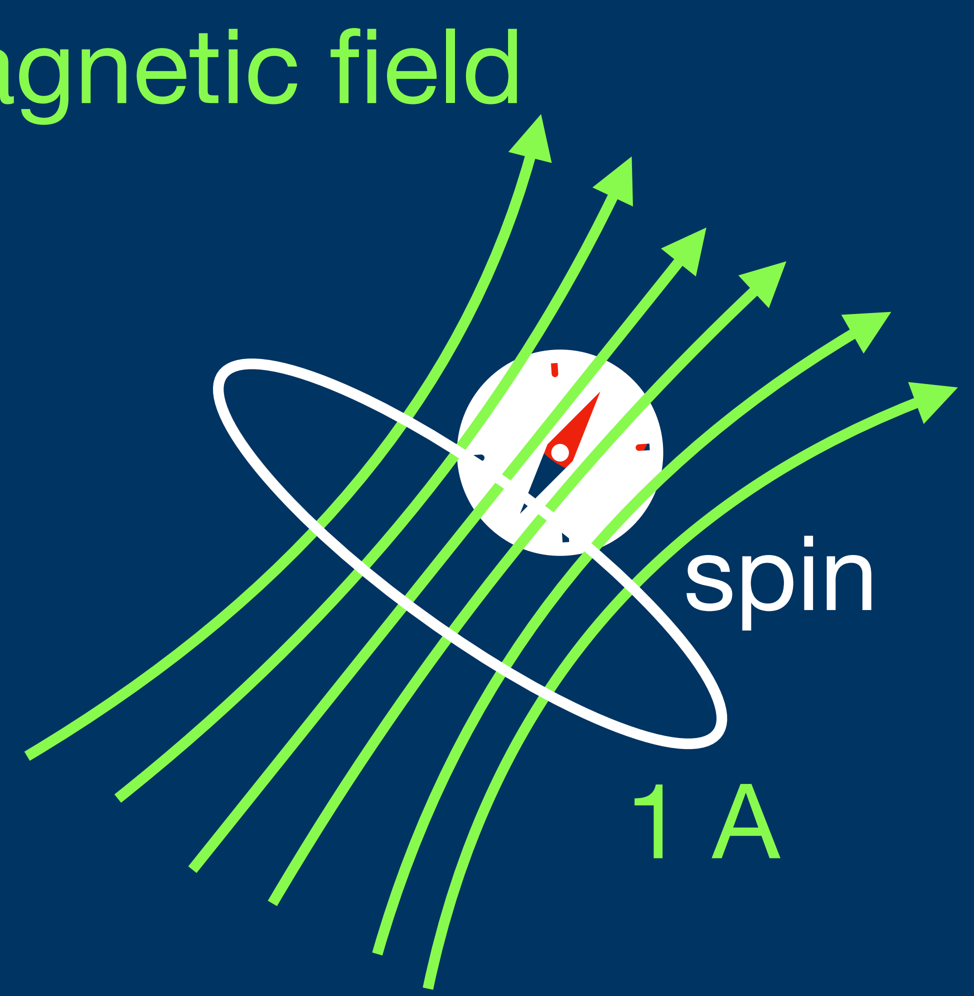
Measuring *zero* is something very special!

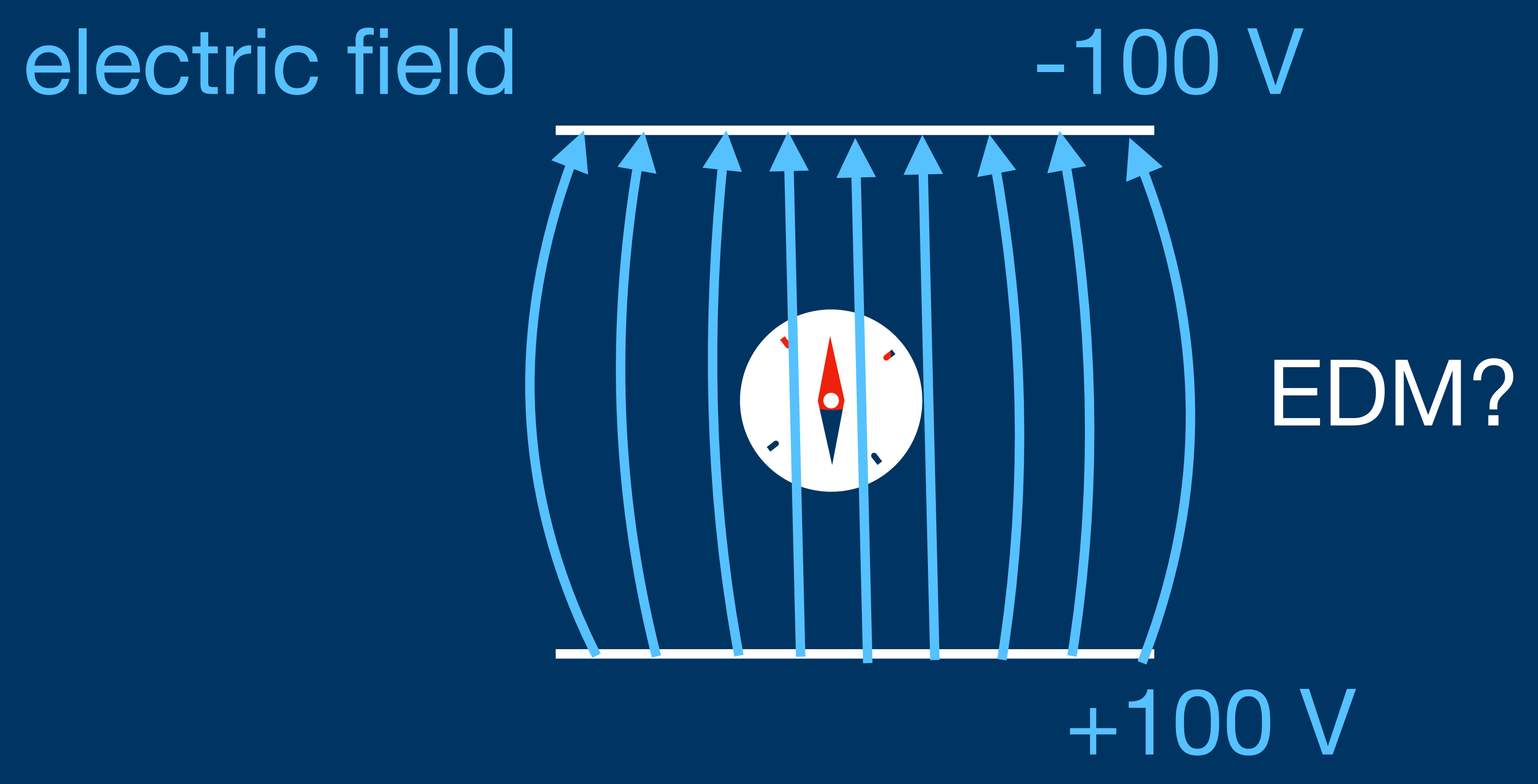
Suppose you measure this:
f_{Al^+}/f_{Hg^+} = 1.052871833148990438(55)
Did you discover new physics?

Suppose you measure this:
eEDM = 0.0000000000000000153(2)
This is direct proof of physics beyond the Standardmodel!

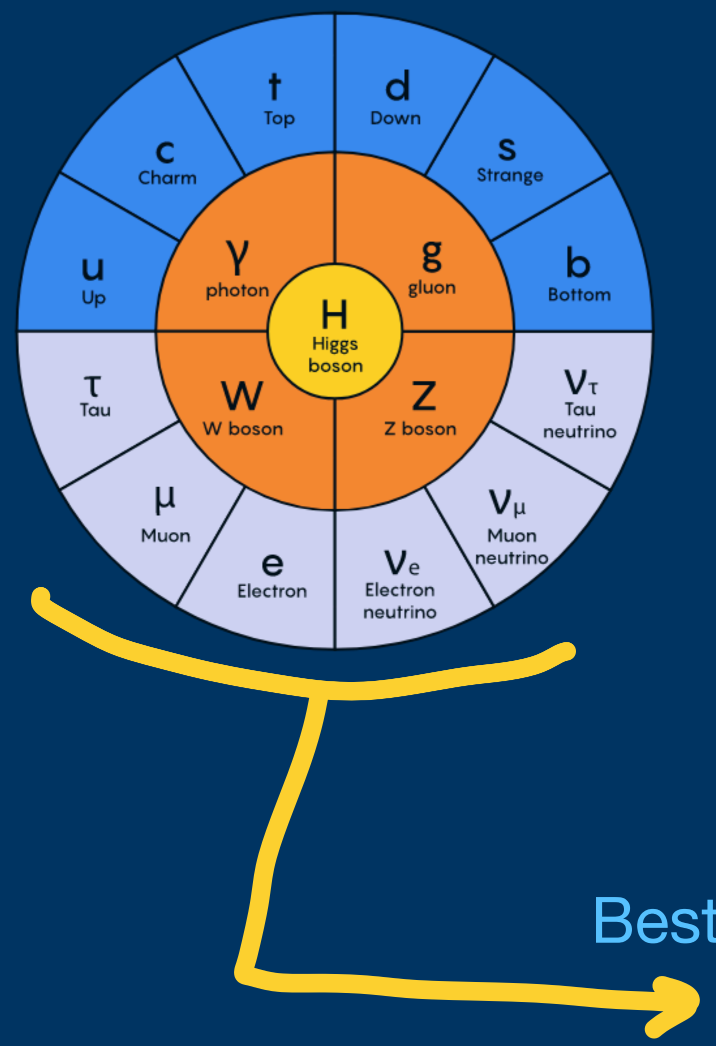
The dipole moments of the electron

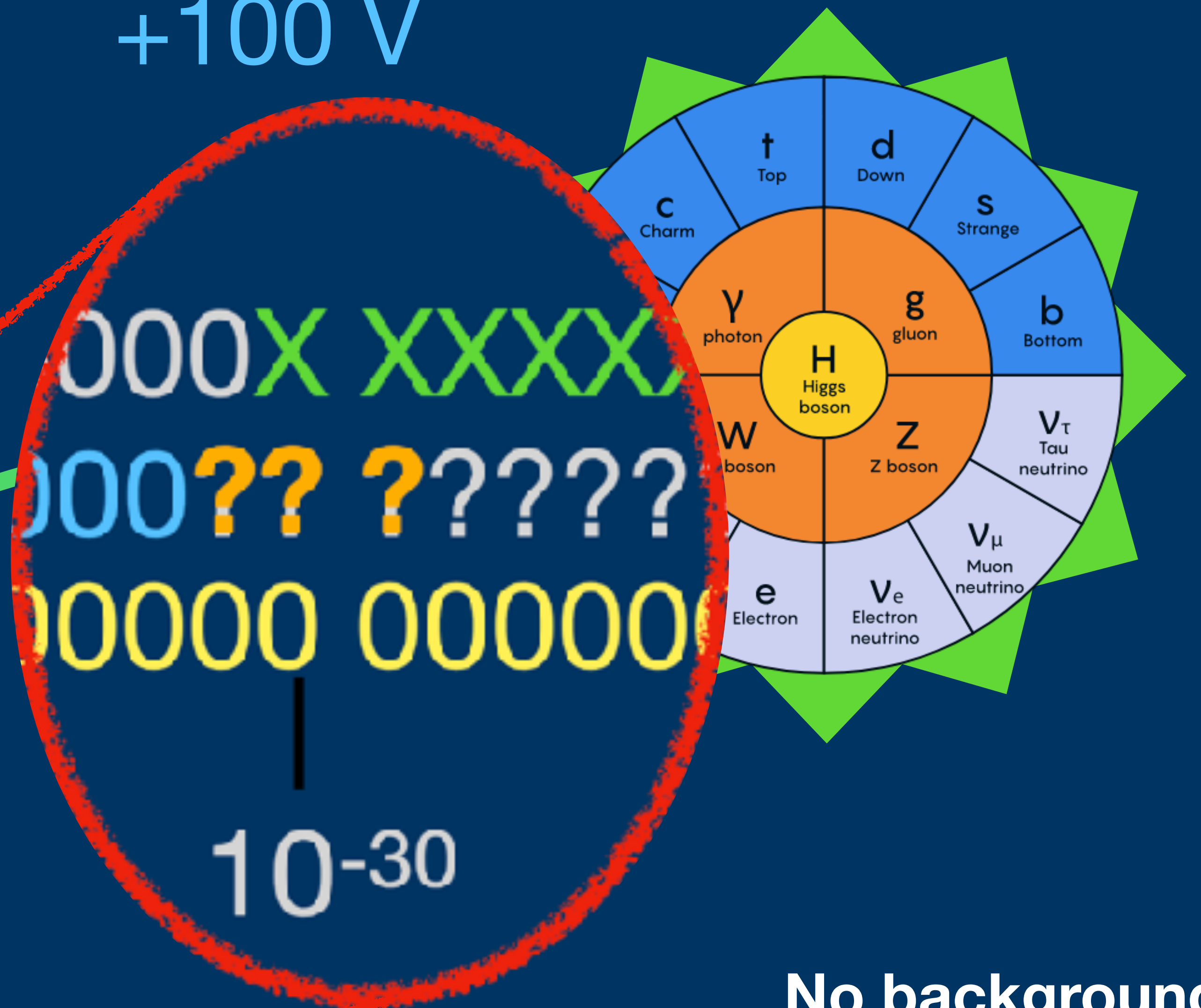
Besides its magnetic dipole moment (spin), an electron could have an *electric dipole moment (eEDM).*

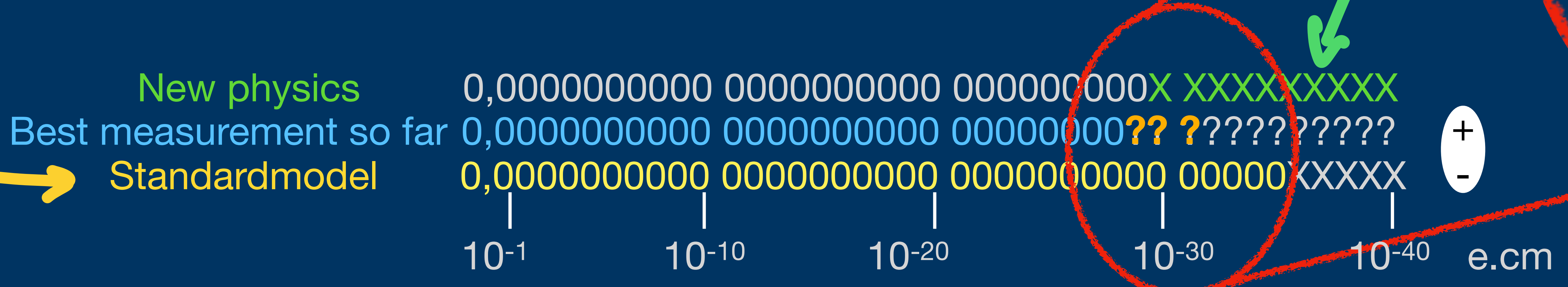




an eEDM violates Time reversal symmetry (T) and is direct proof of new physics!

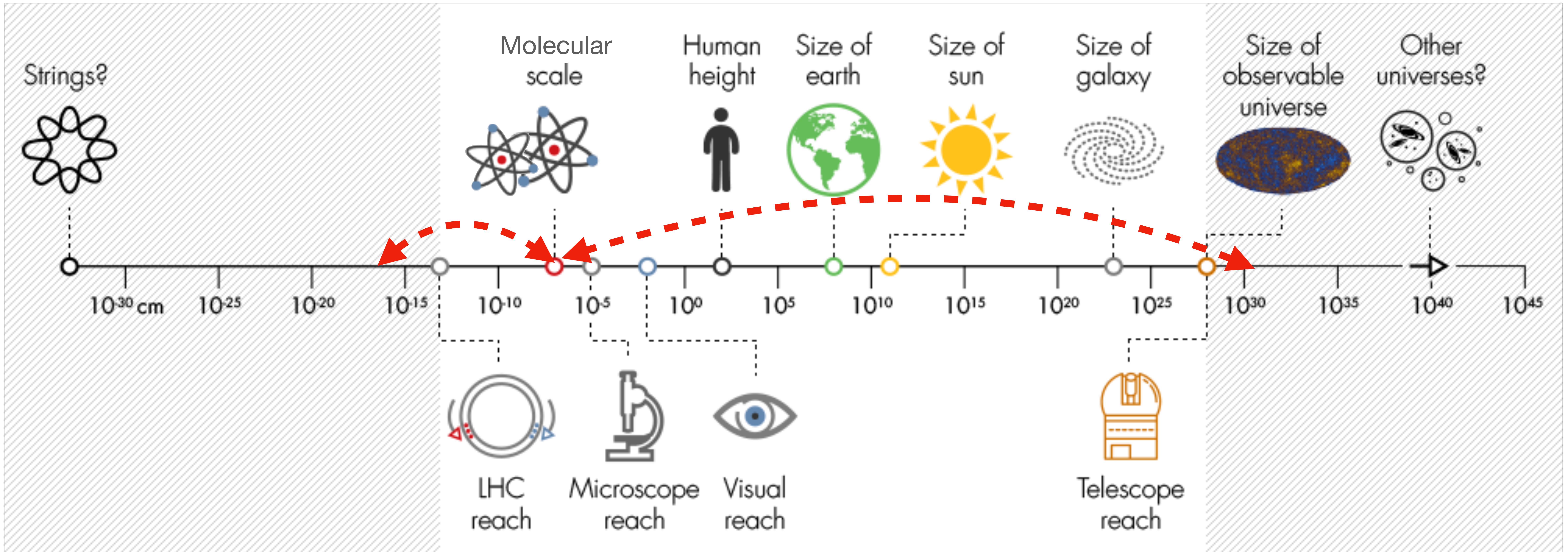






No background: fantastic possibility to discover new physics!

Quantum sensing!



source: Quanta Magazine, 2015

Approximate and neglect: essential in physics. But with this extreme precision we can measure *new physics* which ‘trickles through’ to these last significant digits!

So we want to measure the eEDM!

- Questions:
  - How large is the interaction energy of a non-zero electron-EDM of order 1e-29 e*cm in a strong laboratory electric field??
  - How large is the interaction energy of the electron spin with the earth magnetic field?

Q How large is the interaction energy due to an EDM of $\sim 1 \cdot 10^{-29}$ e·cm, in a stray field in a lab?

$$E = \vec{d}_e \cdot \vec{E}$$

$\vec{E} \rightarrow 100$ kV/cm $\sim 10^7$ V/m

$\vec{d}_e \hookrightarrow 10^{-29}$ e cm

$10^{-29} \cdot 10^{-19}$ C $\cdot 10^{-2}$ m $= 10^{-50}$ C m

[J = C·V]

so 10^{-43} J

Q How large is the interaction energy for an electron spin in the earth magnetic field?

$$E = g \vec{\mu} \cdot \vec{B}$$

$\vec{B} \rightarrow \sim G \sim 10^{-4}$ T

$\hookrightarrow s = \frac{1}{2} \rightarrow \mu = \frac{1}{2} \mu_B$

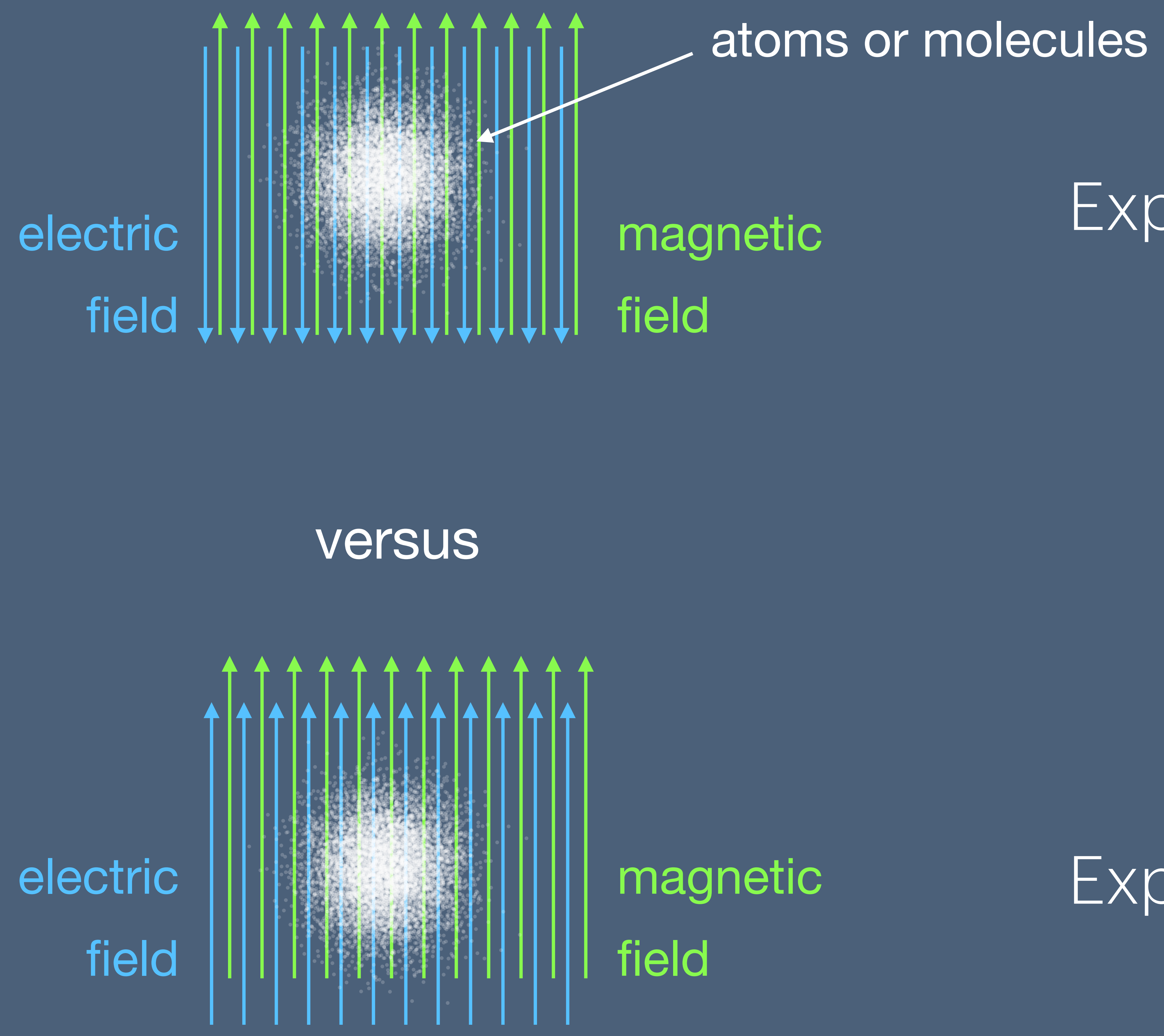
$\hookrightarrow 1 \cdot 10^{-23}$ J/T

$g_s = 2$

so $1 \cdot 10^{-23} \cdot 10^{-4} = 10^{-27}$ J

General approach:

Flip fields:
looking for an energy shift that correlates with the electric field switch



Experiment 1 (do many times)

Experiment 2 (do many times)

Look for a difference

Can we estimate the magnitude of the shifts?

Magnitude of energy shifts

base system: e^- doesn't work

atom: Thallium: enhanced E-field by $\sim 585\times$
$\sim 10^3$

Frequency shifts of an energy level in the atom

$$B: \quad \Delta\nu_\mu = \frac{10^{-27}\,\mathrm{J}}{h} \Bigg\} \sim 10^{6}\,\mathrm{Hz} \sim 1\,\mathrm{MHz}$$

$h \to 6.6\cdot 10^{-34}\,\mathrm{Js} \sim 10^{-33}\,\mathrm{Js}$

$$E \quad \Delta\nu_{edm} \sim \frac{10^{-43}\cdot 10^{3}}{10^{-33}} \sim 10^{-7}\,\mathrm{Hz} \sim 0.1\,\mu\mathrm{Hz}$$

History: EDM measurements with atoms

New limit on the electron electric dipole moment

B. C. Regan,* Eugene D. Commins,† Christian J. Schmidt,‡ and David DeMille§
Physics Department, University of California, and Lawrence Berkeley National Laboratory, Berkeley, California 94720
(Dated: August 8, 2001)

We present the result of our most recent search for T-violation in ^{205}Tl, which is interpreted in terms of an electric dipole moment of the electron d_e. We find $d_e = (6.9 \pm 7.4) \times 10^{-28}\, e$ cm. The present apparatus is a major upgrade of the atomic beam magnetic-resonance device used to set the previous limit on d_e.

Long interaction zone

Mu-metal shielding

Co-magnetometers

Ramsey interferometry

Intense beams

Interference detection - just like the Cs experiment

This was the best experiment for over a decade!

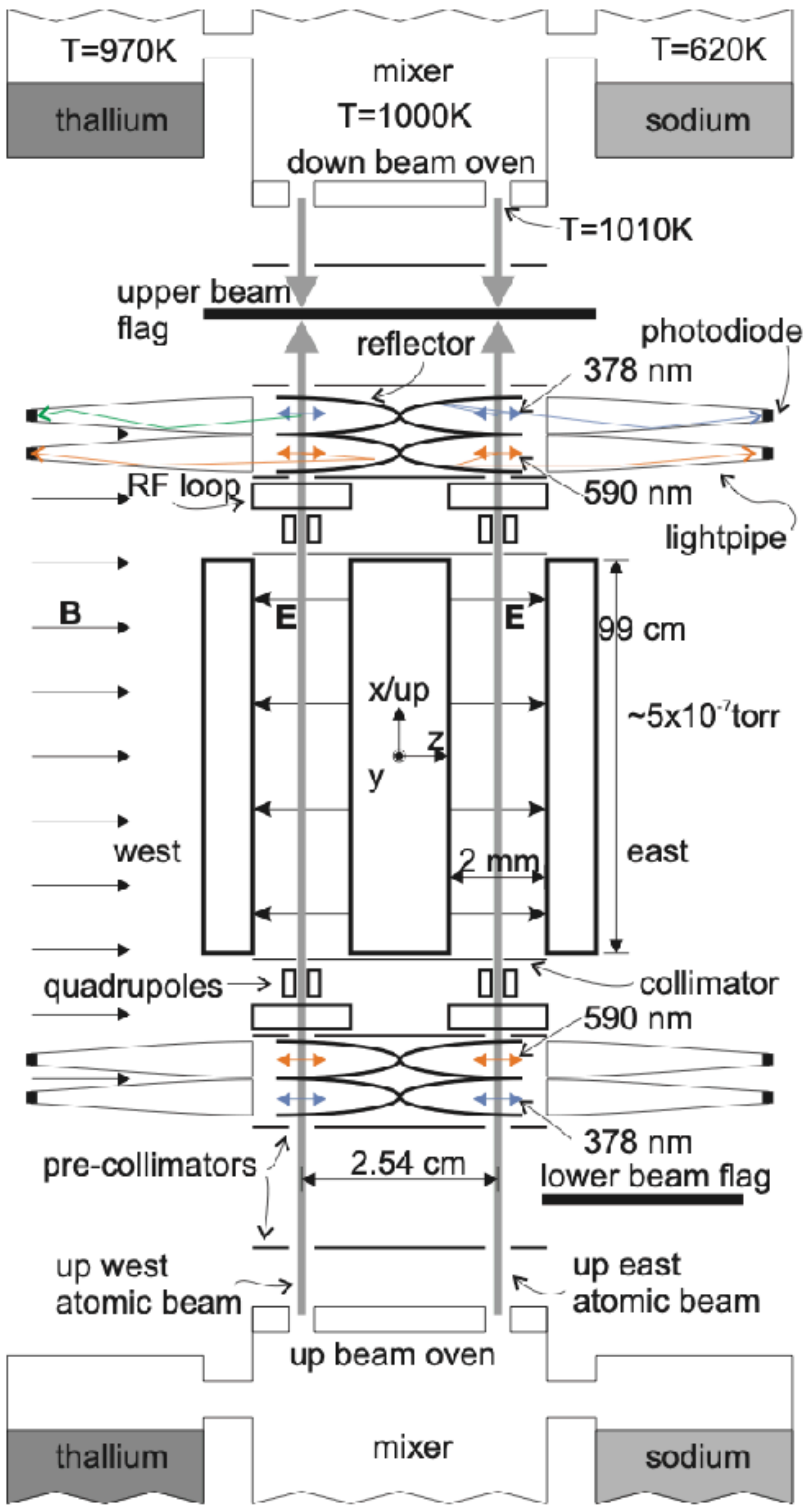


FIG. 1: Schematic depiction of the experiment, not to scale.

Statistical sensitivity of the measurement

Heisenberg uncertainty $\Delta E \Delta t = \hbar$

(Δt → observation time T)

$\delta_e E_{eff} = \delta_e \cdot E_{nh} \cdot E_{lab}$

E_{nh}: 585 Tl atom; for molecules, can be $\sim 10^6$

E_{lab}: 100 kV/cm

$$\delta_e = \frac{\hbar}{E_{eff} T}$$

let's use many atoms / molecules: $$\delta_e = \frac{\hbar}{E_{eff} T \sqrt{N}}$$

How can we do better?

The statistical sensitivity equation

Statistics

longer interaction times (T in Ramsey scheme) - *cooling techniques*

higher enhancement factors *molecules to the rescue!*

more atoms, longer measurement time

Systematics

v x E effect - *molecules to the rescue!*

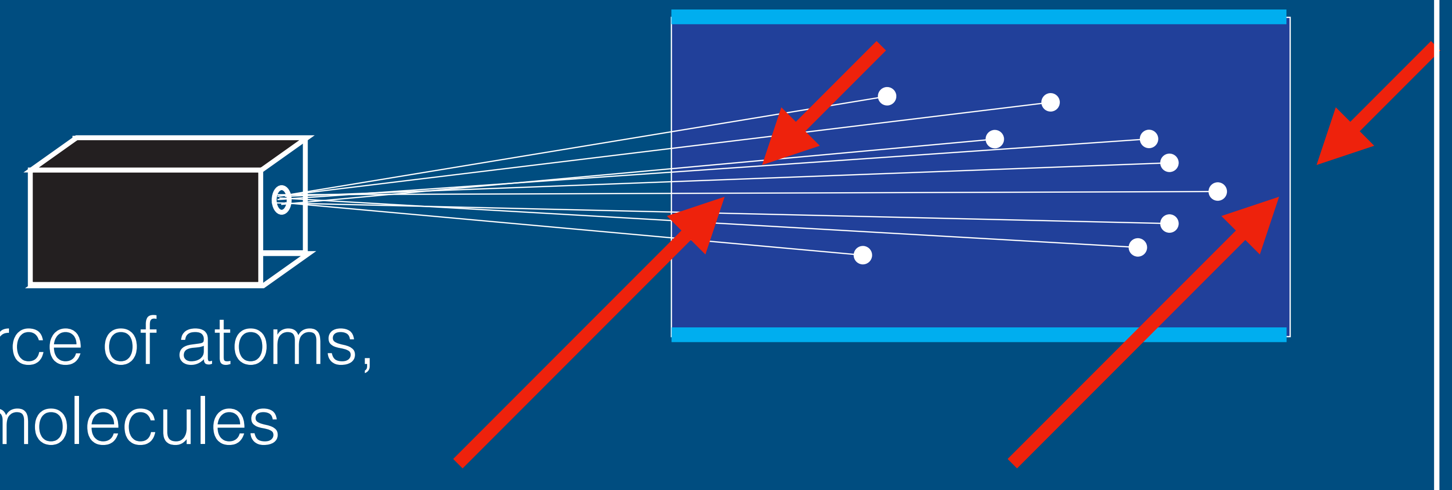
reduced magnetic field sensitivity - *molecules to the rescue!*

Precision measurements with molecules

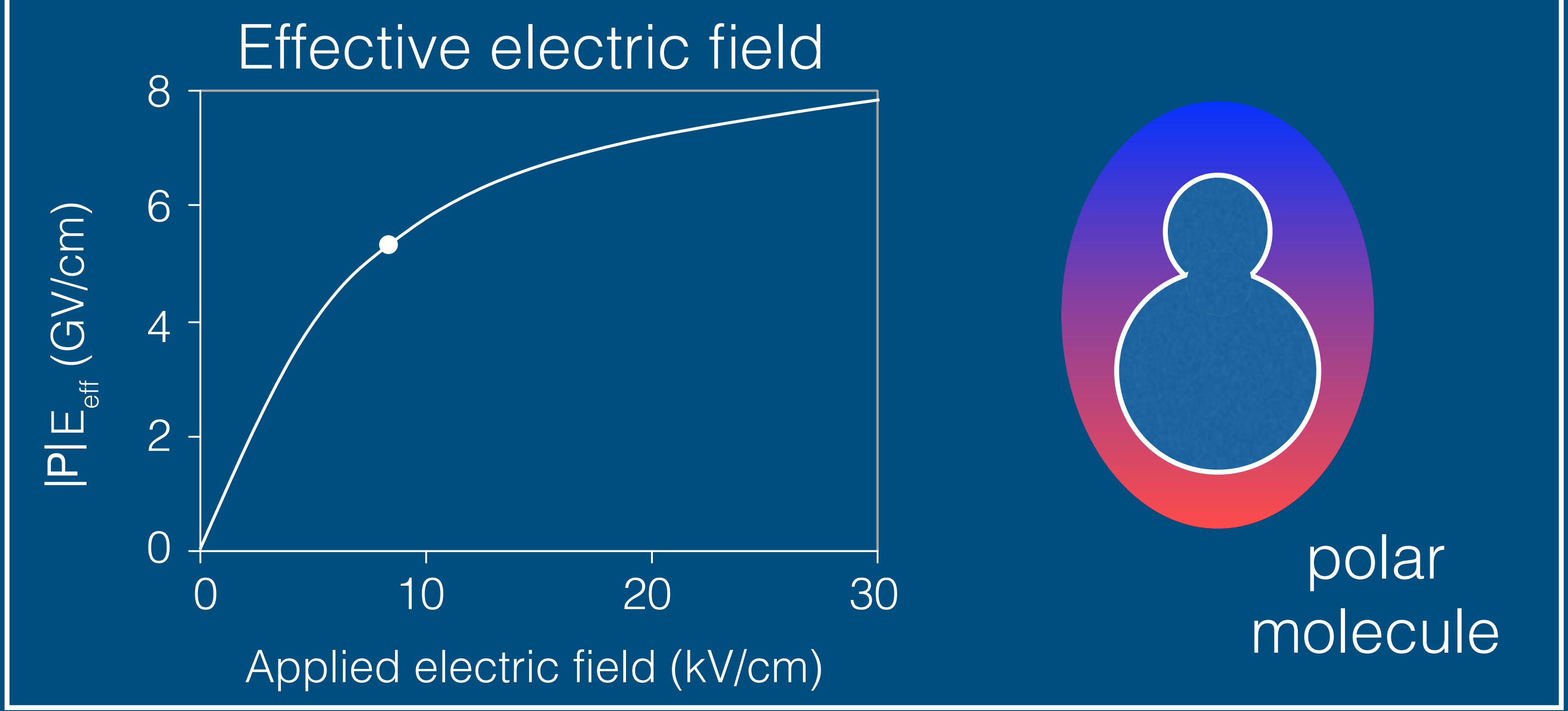
Complex quantum systems with an advantage

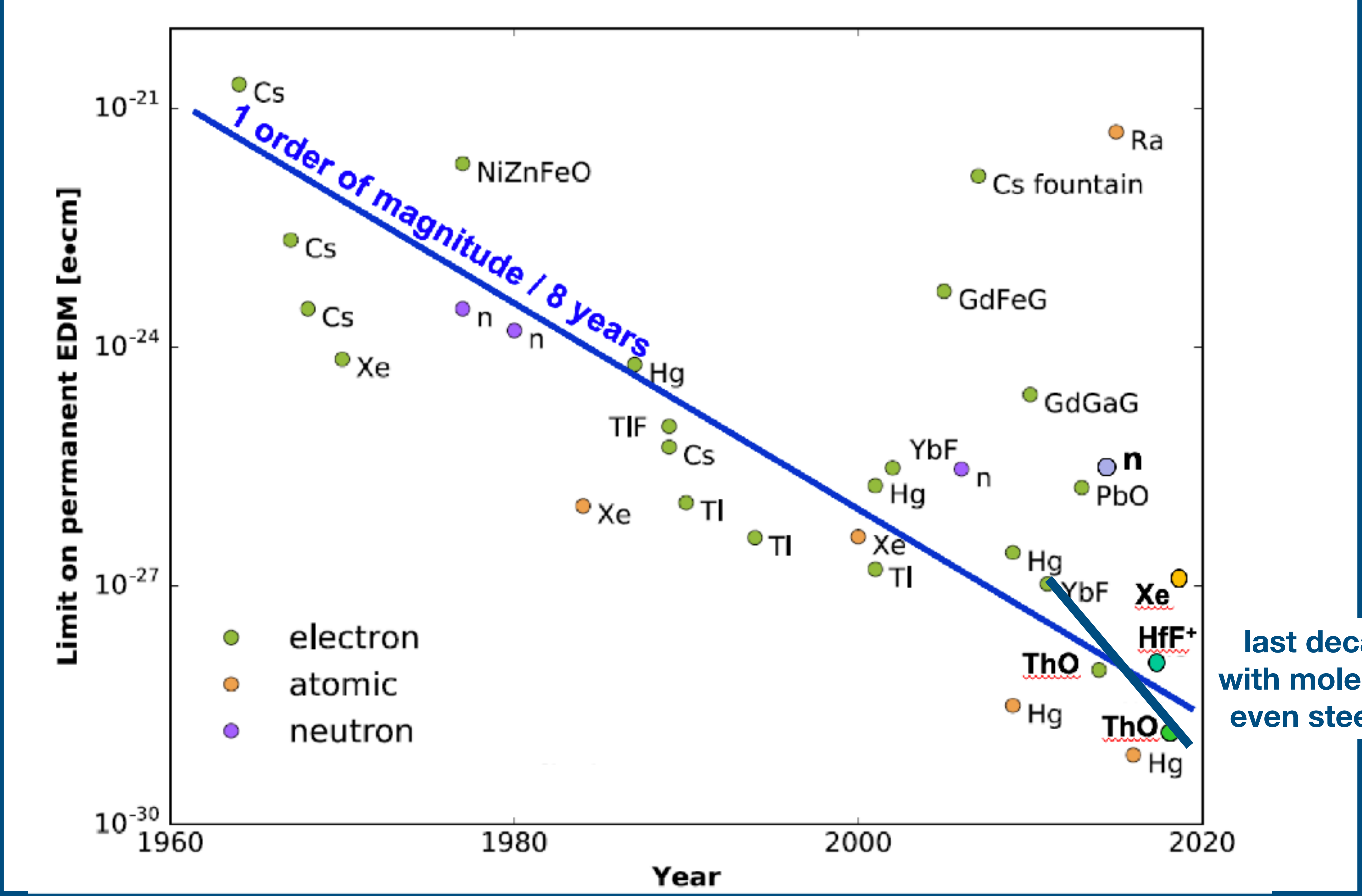
Example 4: The electric dipole moment of the electron

precision molecular spectroscopy



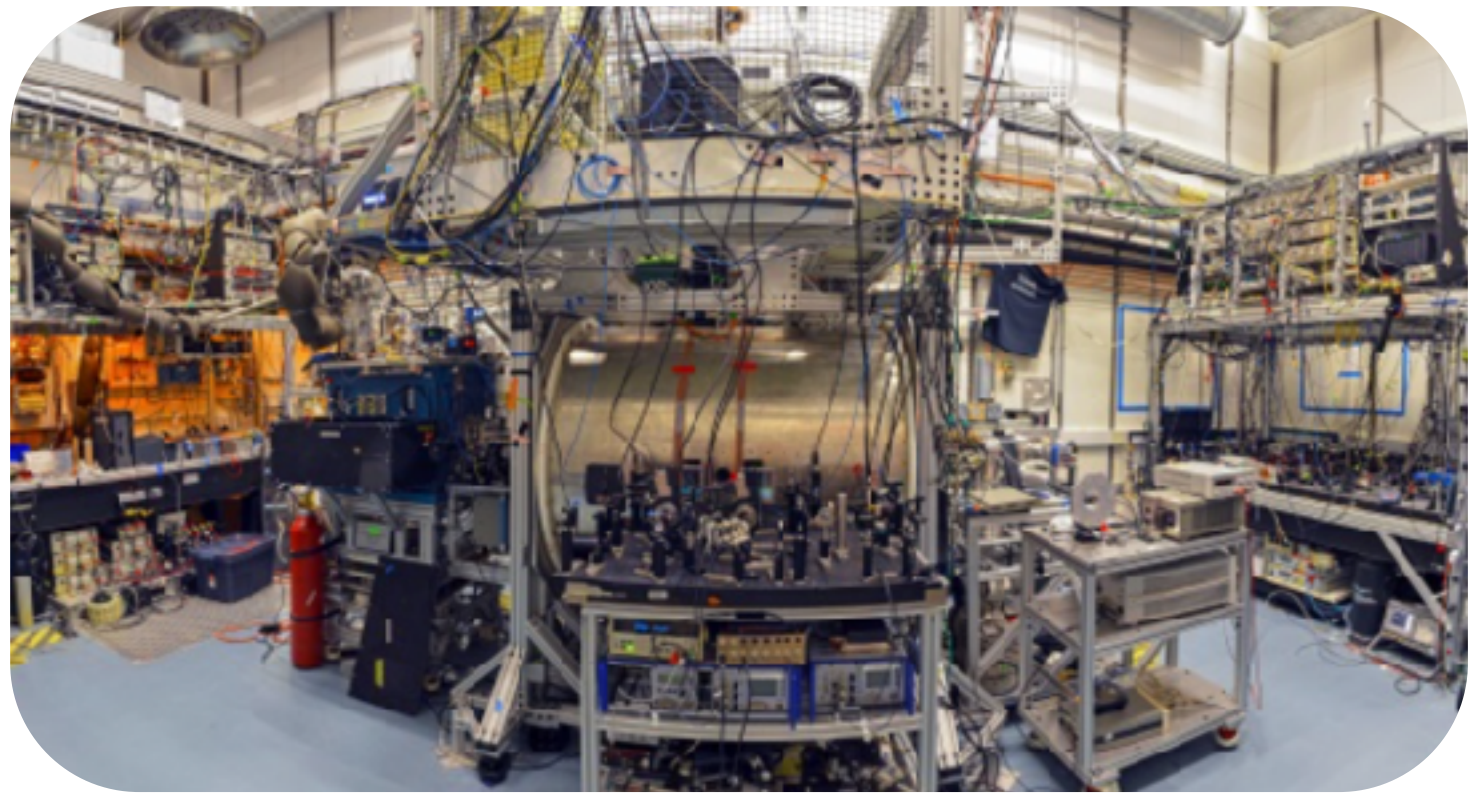
huge enhancement of E-field in molecule



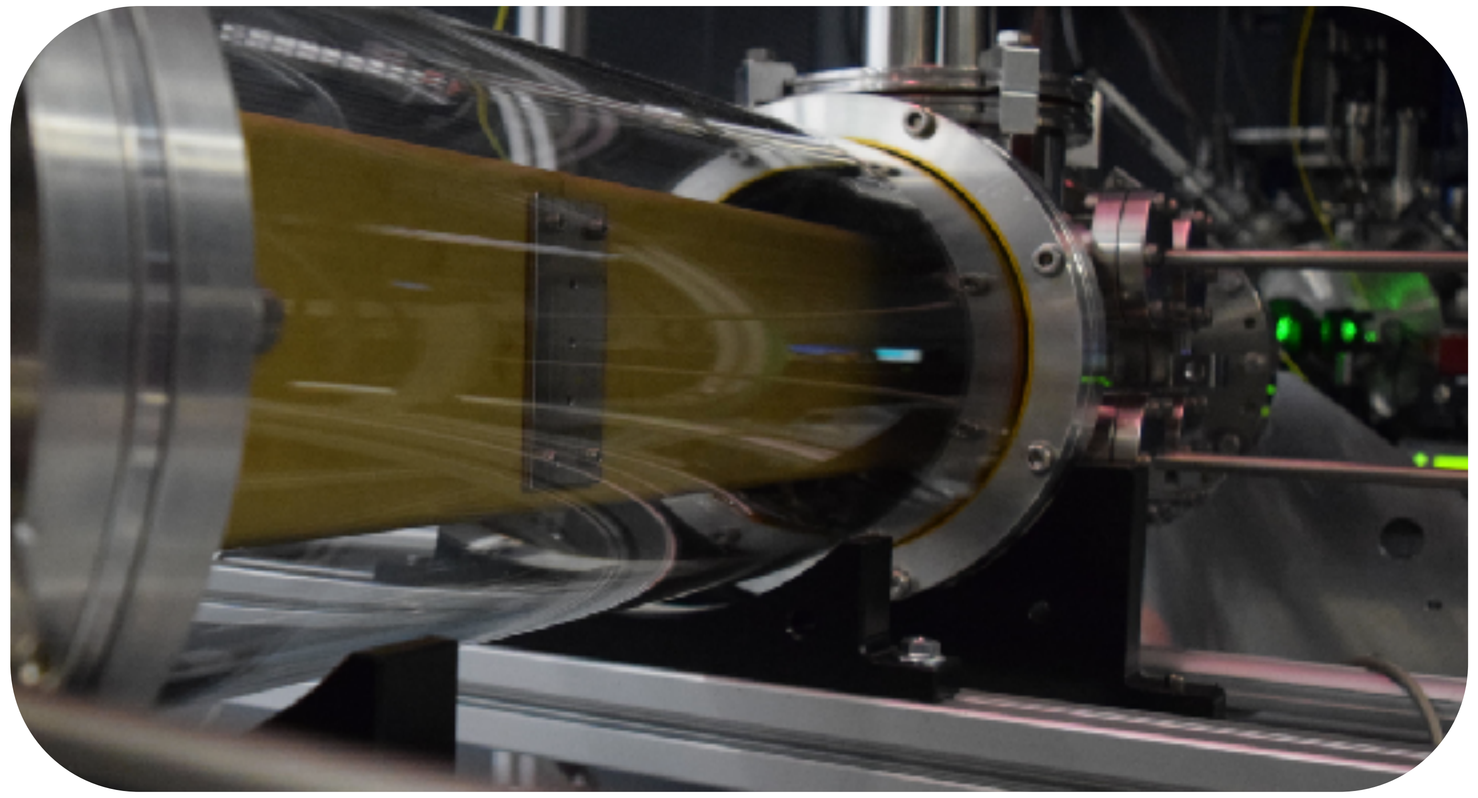


last decade with molecules even steeper!

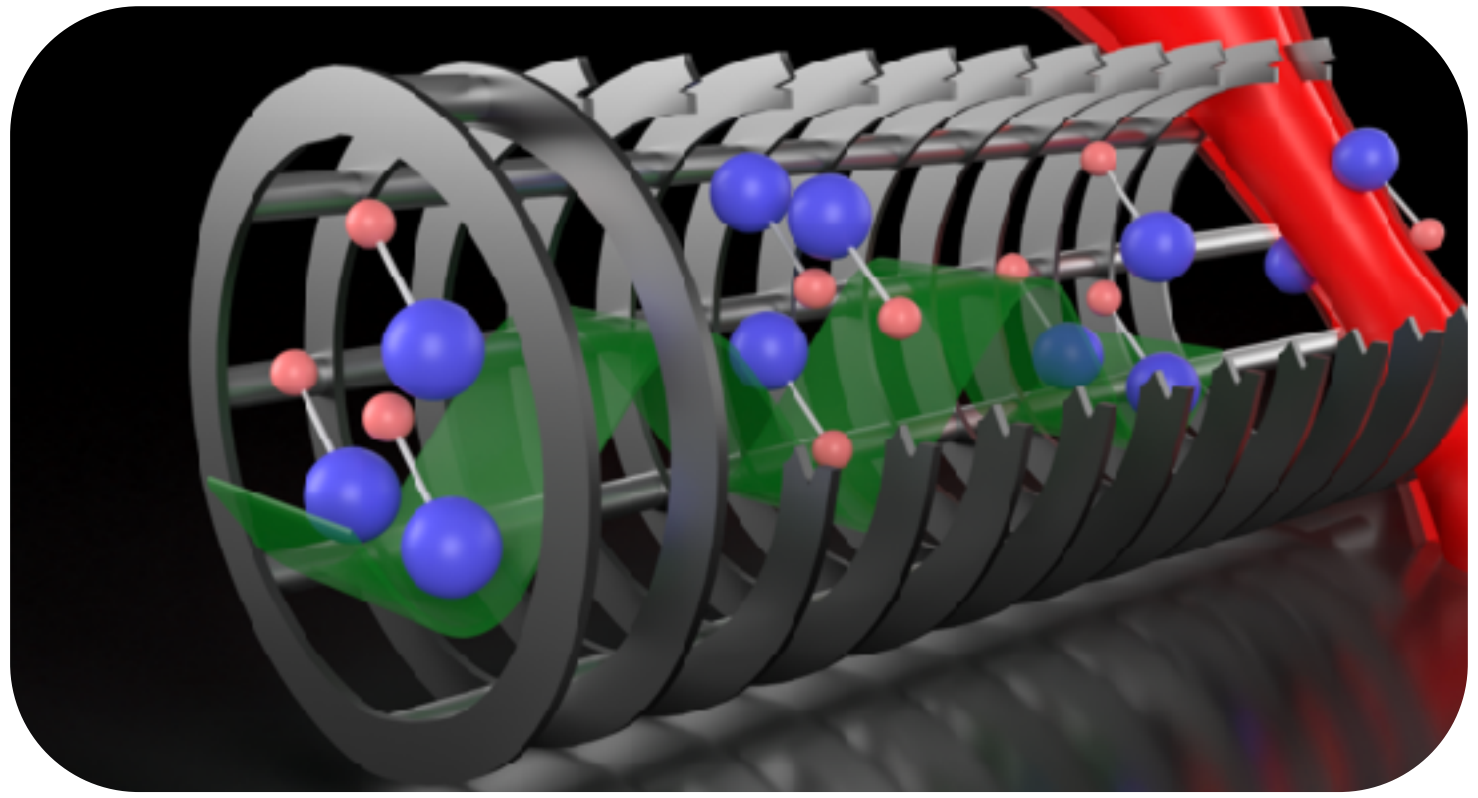
eEDM experiments with molecules



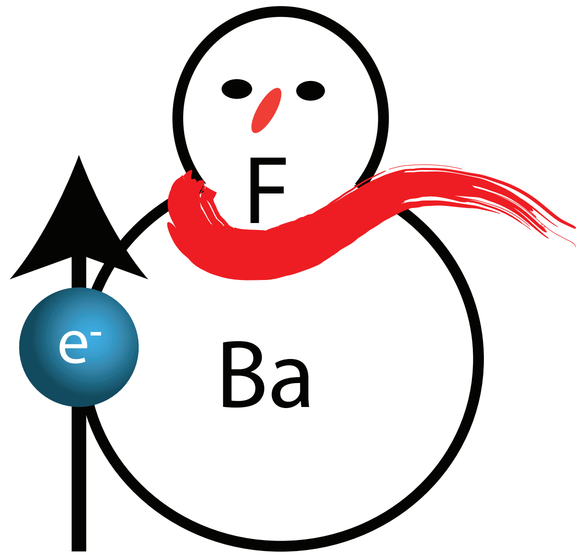
ACME, Harvard
beam of ThO molecules



Imperial College London
beam of YbF molecules



JILA, Boulder
trapped HfF^+ ions



Cold BaF molecules in Groningen, NL
since 2017 (NL-eEDM)

EDM measurements with molecules in a frozen noble gas:

Search for eEDM in cryogenic crystals

PHYDES/DOCET

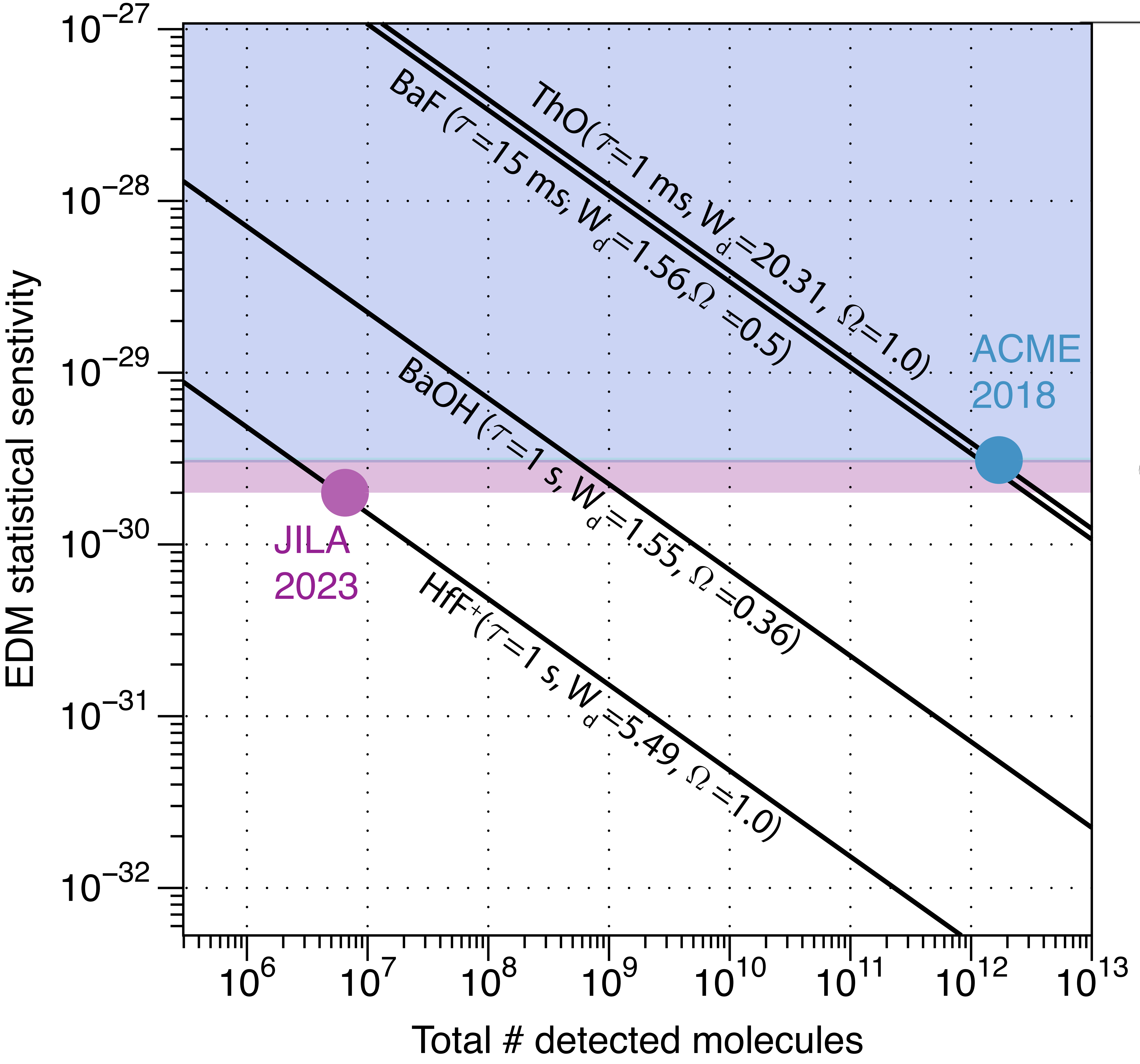
Para-Hydrogen and Diatomic for eEDM Study

Università degli Studi di Ferrara

INFN Istituto Nazionale di Fisica Nucleare

- York University
- Michigan State University
- University of Toronto

EDM³ Collaboration



PHYSICAL REVIEW A **111**, 062815 (2025)

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

Roman Bause,[1,2,*] Nithesh Balasubramanian,[1,2] Ties Fikkers,[1,2] Eifion H. Prinsen,[1,2] Kees Steinebach,[3] Arian Jadbabaie,[4] Nicholas R. Hutzler,[5] I. Agustín Aucar,[1,2,6] Lukáš F. Pašteka,[1,2,7] Anastasia Borschevsky,[1,2] and Steven Hoekstra[1,2,†]

[1]Van Swinderen Institute for Particle Physics and Gravity, University of Groningen, The Netherlands
[2]Nikhef, National Institute for Subatomic Physics, Amsterdam, The Netherlands
[3]LaserLaB, Vrije Universiteit Amsterdam, The Netherlands
[4]Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA
[5]Division of Physics, Mathematics, and Astronomy, California Institute of Technology, Pasadena, California 91125, USA
[6]Instituto de Modelado e Innovación Tecnológica (UNNE-CONICET), Facultad de Ciencias Exactas y Naturales y Agrimensura, Universidad Nacional del Nordeste, Corrientes, Argentina
[7]Department of Physical and Theoretical Chemistry, Comenius University, Bratislava, Slovakia

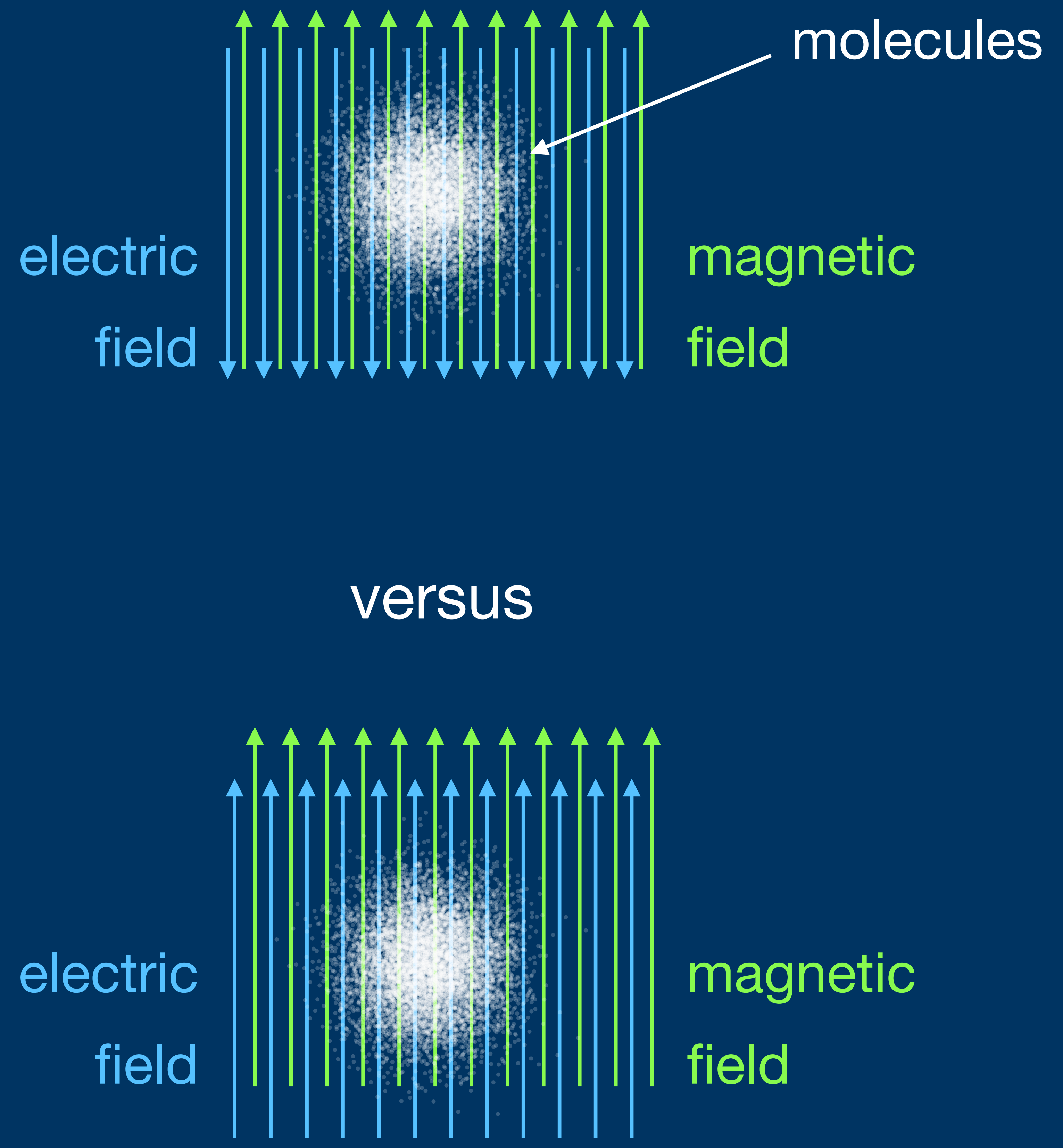
(Received 1 November 2024; accepted 3 June 2025; published 17 June 2025)

We present the conceptual design of an experiment to measure the electron's electric dipole moment (eEDM) using ^{138}BaOH molecules in an optical lattice. The BaOH molecule is laser-coolable and highly sensitive to the eEDM, making it an attractive candidate for such a precision measurement, and capturing it in an optical lattice offers potentially very long coherence times. We study possibilities and limitations of this approach, identify the most crucial limiting factors and ways to overcome them. The proposed apparatus can reach a statistical error of 10^{-30} e cm by measuring spin precession on a total number of 5×10^9 molecules over a span of 120 days.

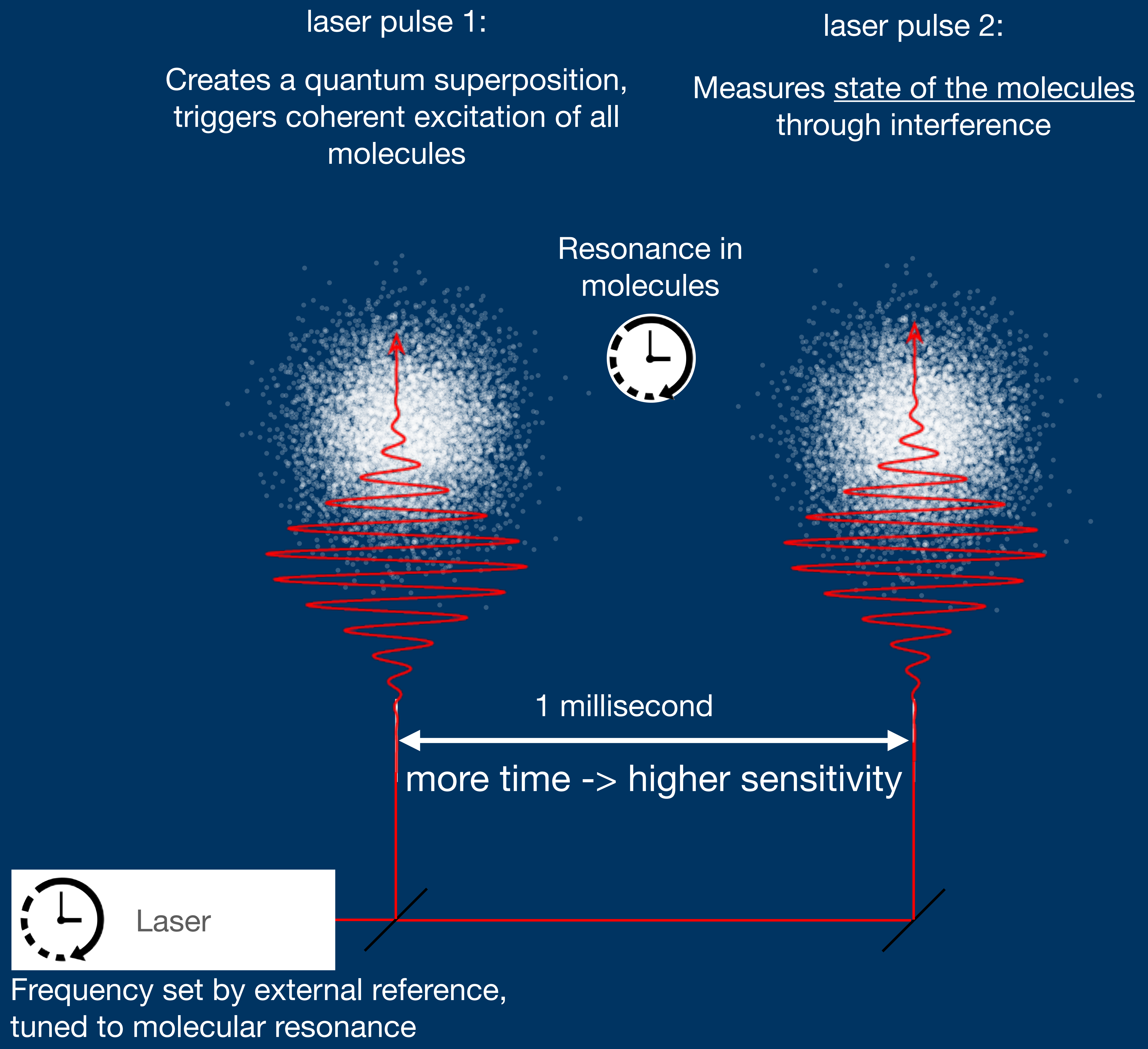
DOI: 10.1103/8ltl-7wsb

Extra slides with experimental details

Ingredients of all eEDM experiments:



Flip fields:
looking for an asymmetry



Ramsey interferometry

New labs in Groningen

How to read out small energy shifts: spin interferometer

How to read out small energy shifts: spin interferometer

$$|\pm\rangle = (|-1\rangle \pm |+1\rangle)/\sqrt{2}$$

$$|\pm 1\rangle = e^{\pm i\varphi}|\pm 1\rangle$$

$$\varphi = \frac{(\mu B \pm dE)T}{\hbar}$$

How to read out small energy shifts: spin interferometer

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$$\left|\pm 1\right\rangle = e^{\pm i\varphi}\left|\pm 1\right\rangle$$

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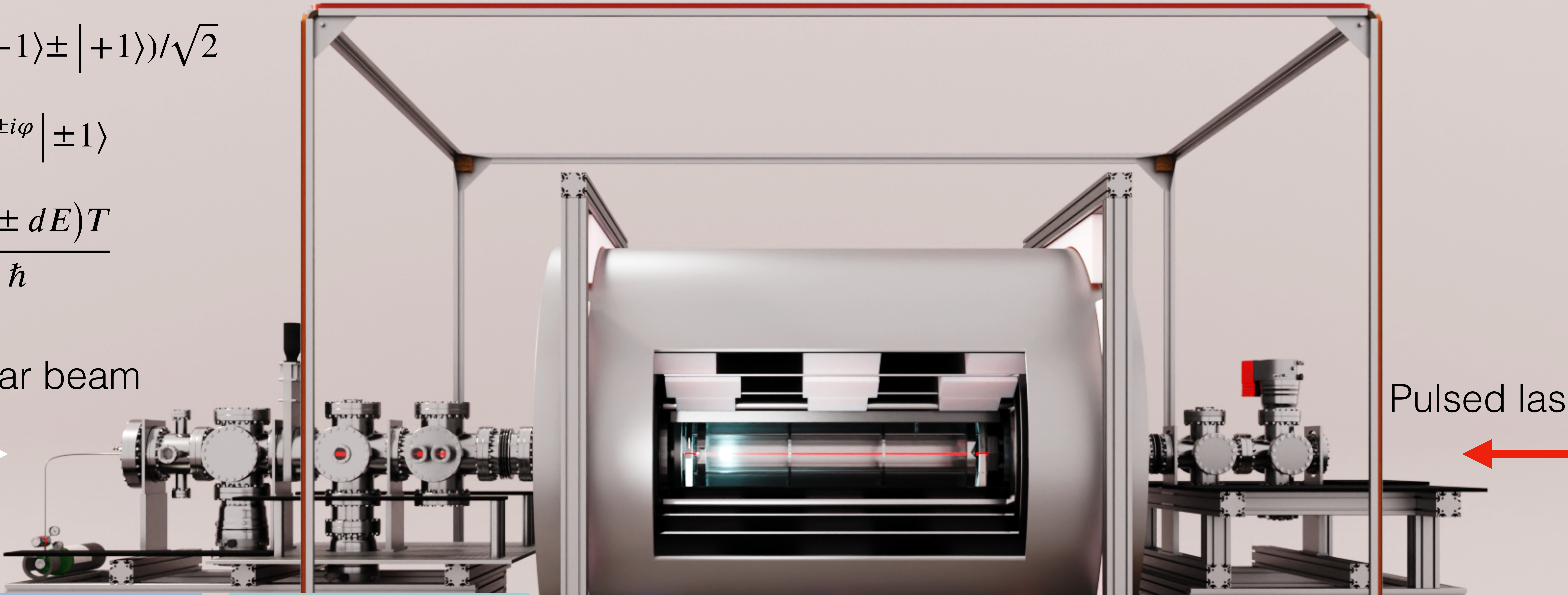
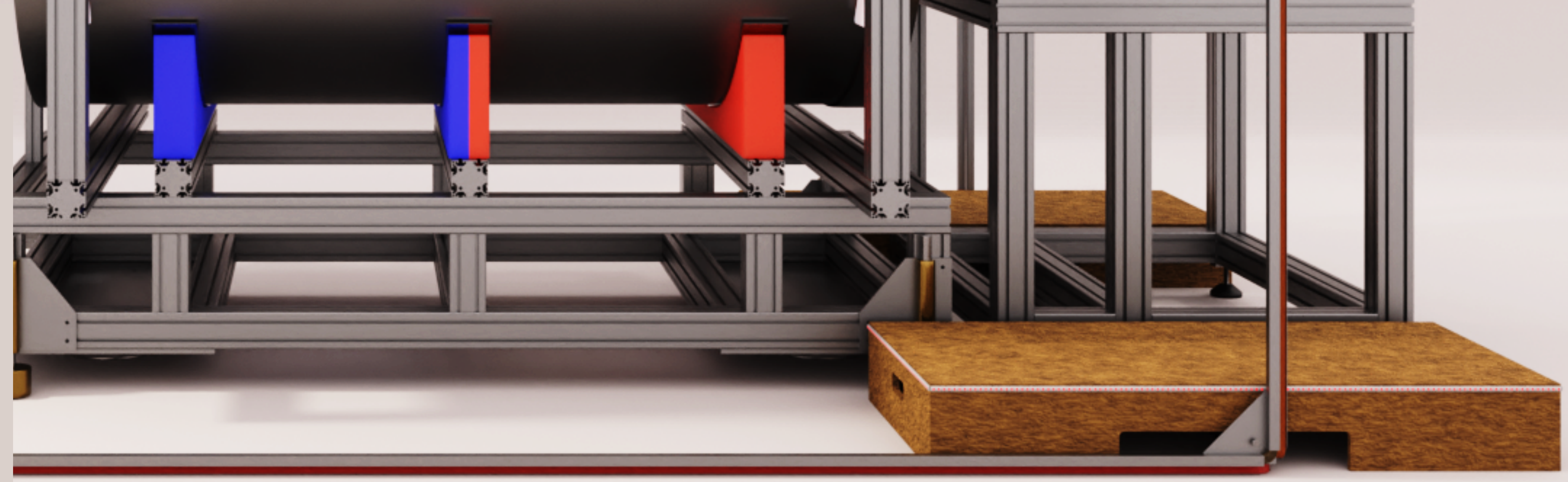
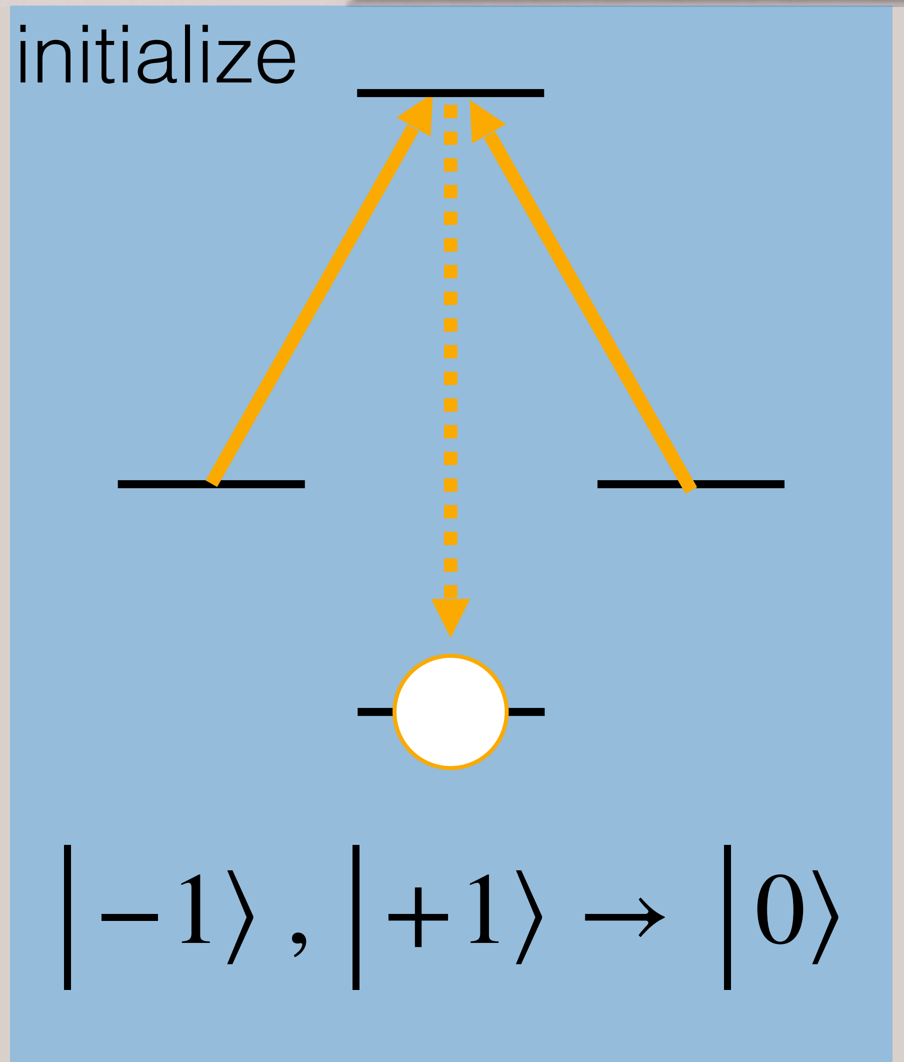
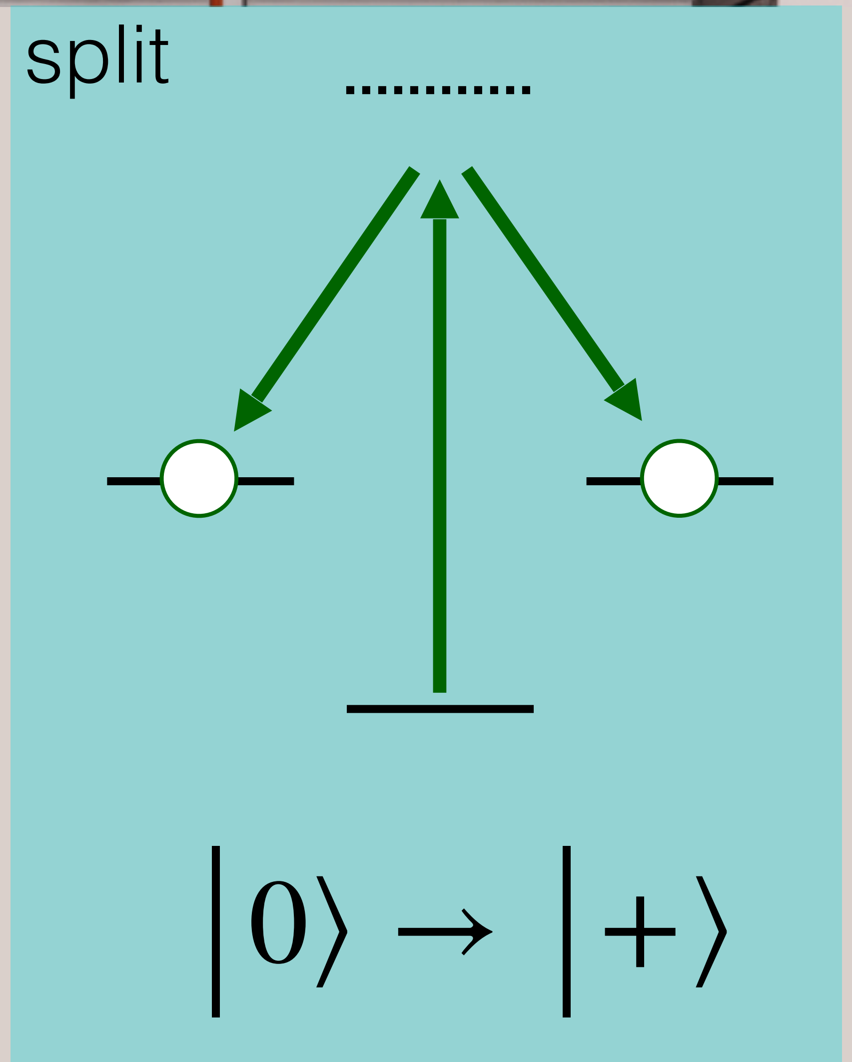
Molecular beam
20 Hz

Pulsed laserlight





How to read out small energy shifts: spin interferometer

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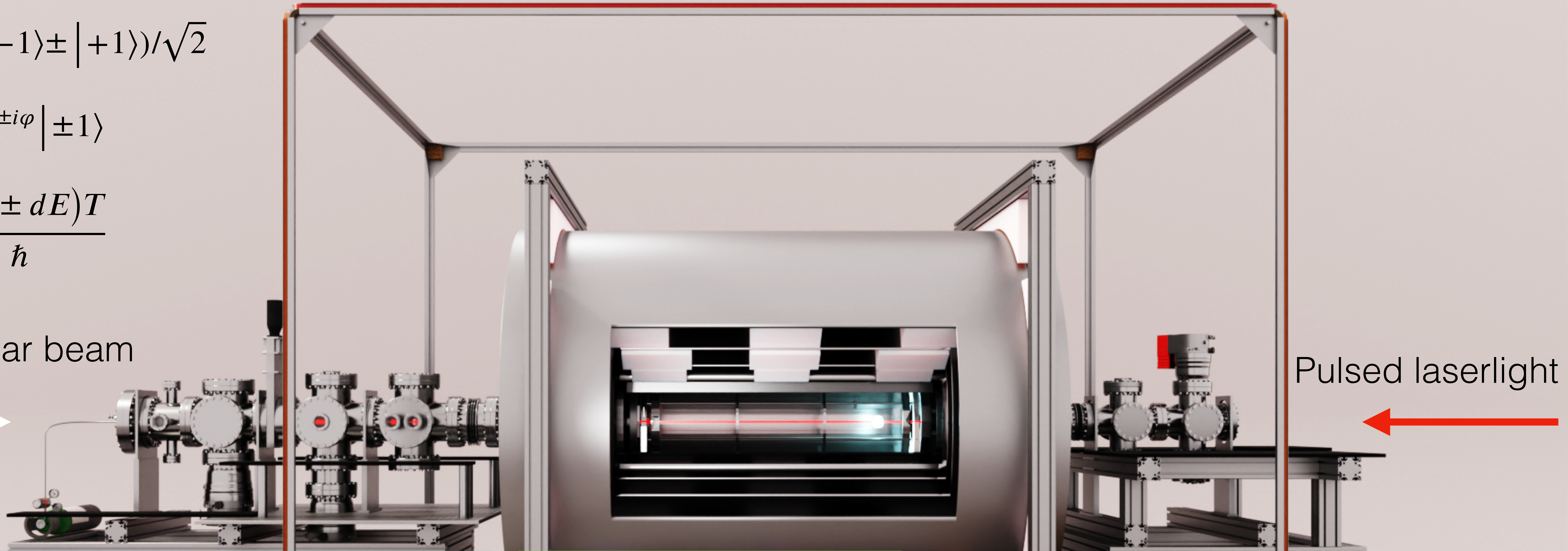
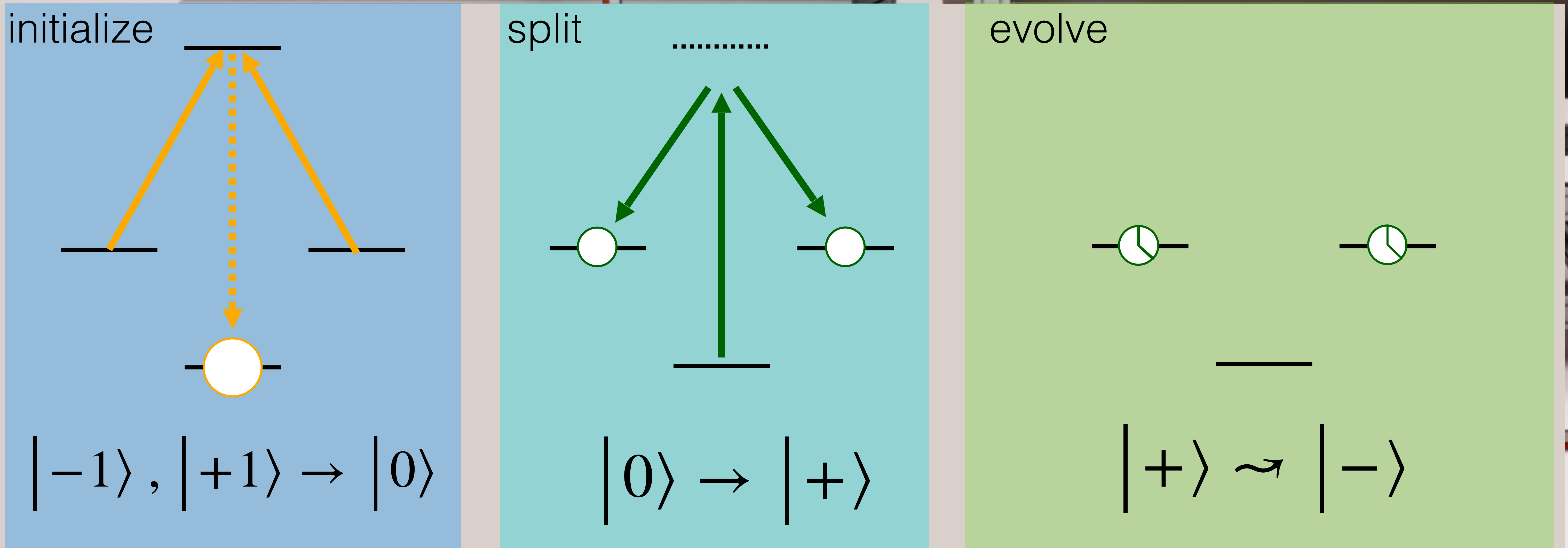
Molecular beam
20 Hz





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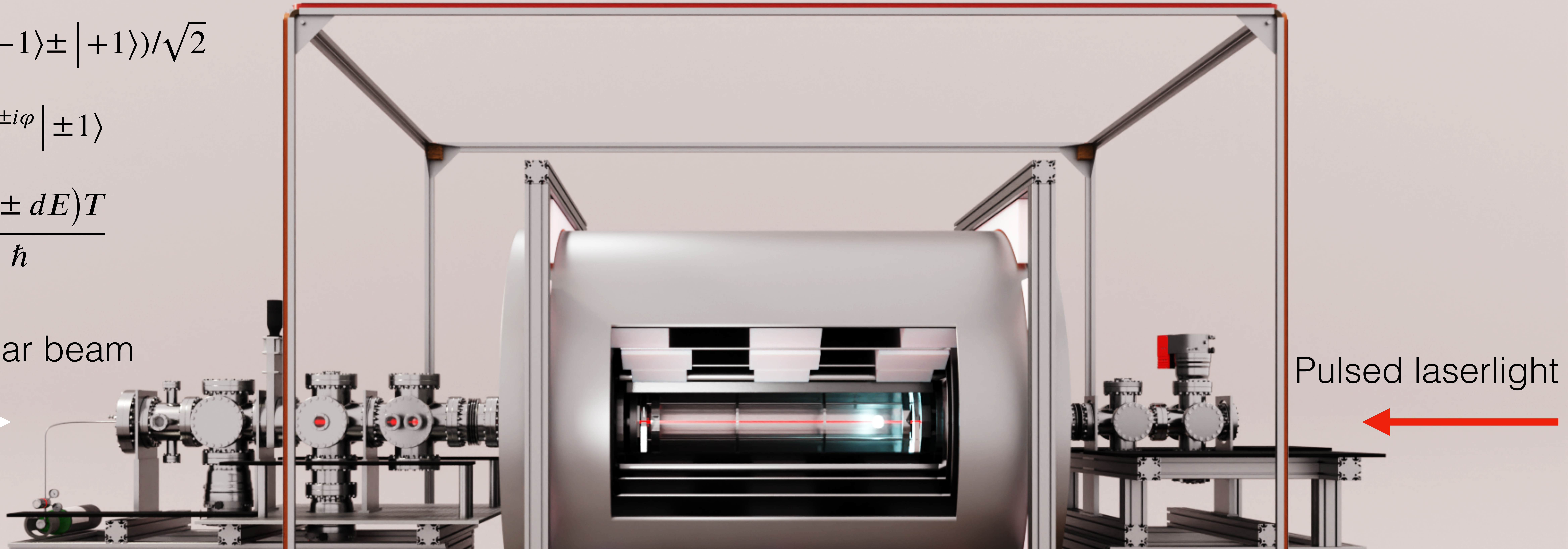

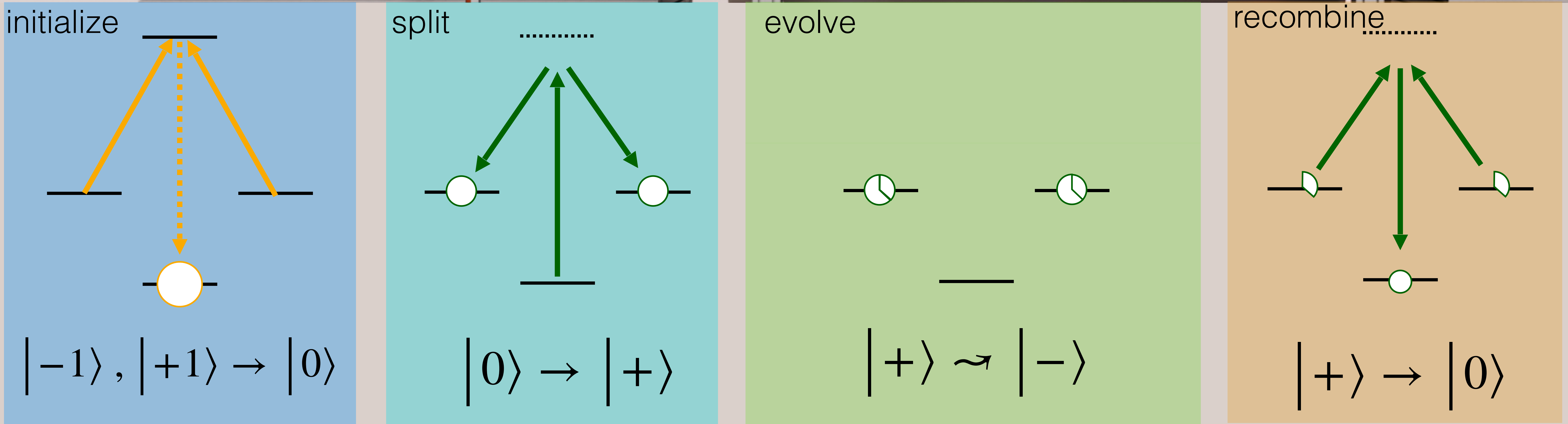

How to read out small energy shifts: spin interferometer

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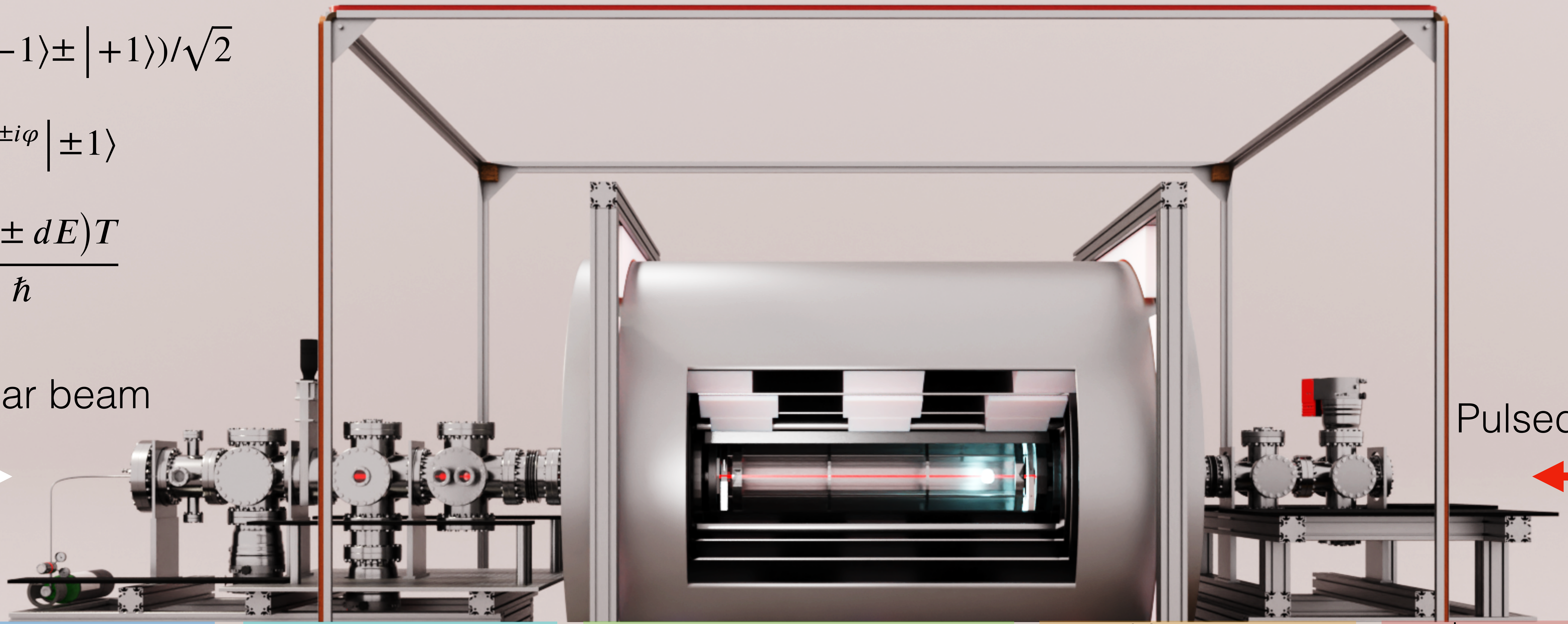
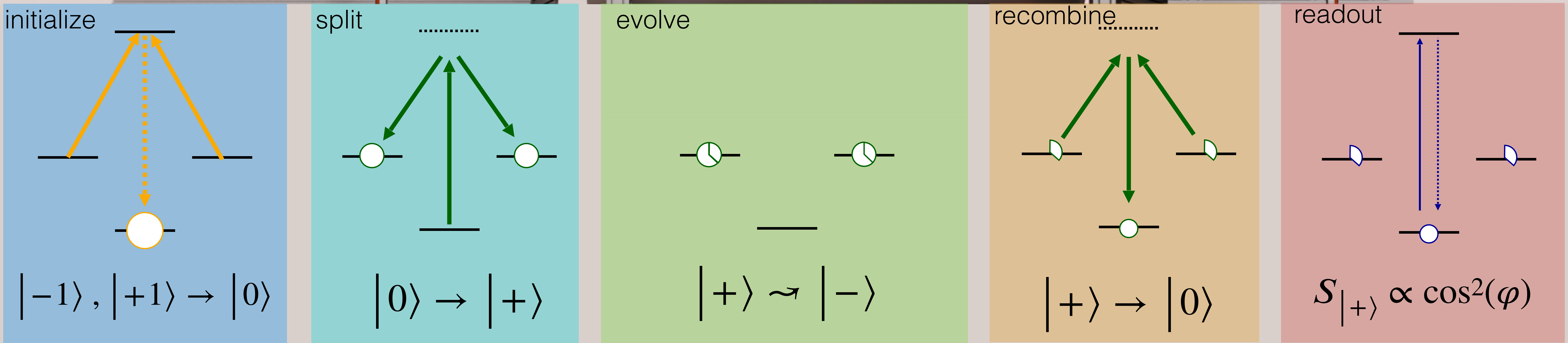
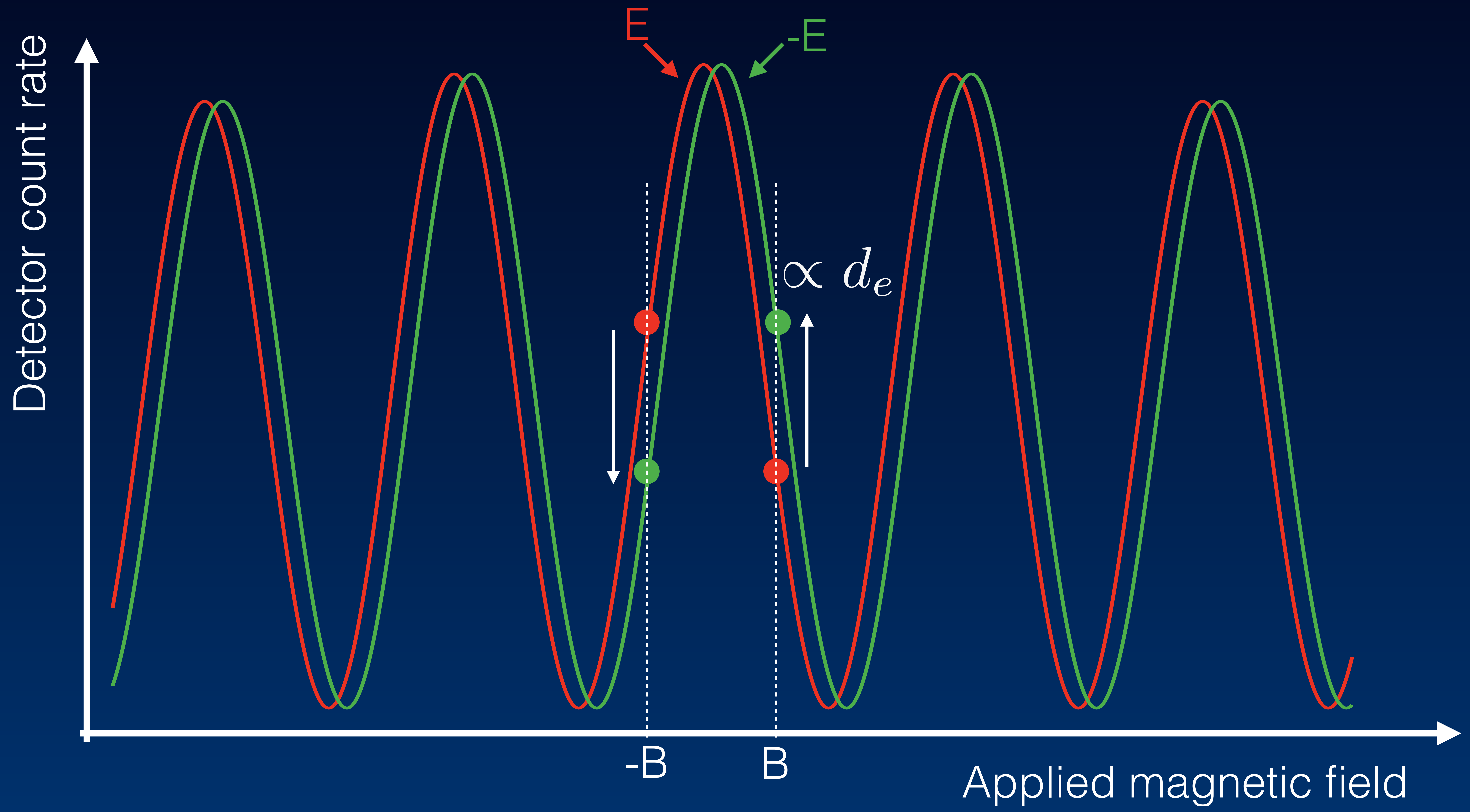
Molecular beam
20 Hz



Pulsed laserlight

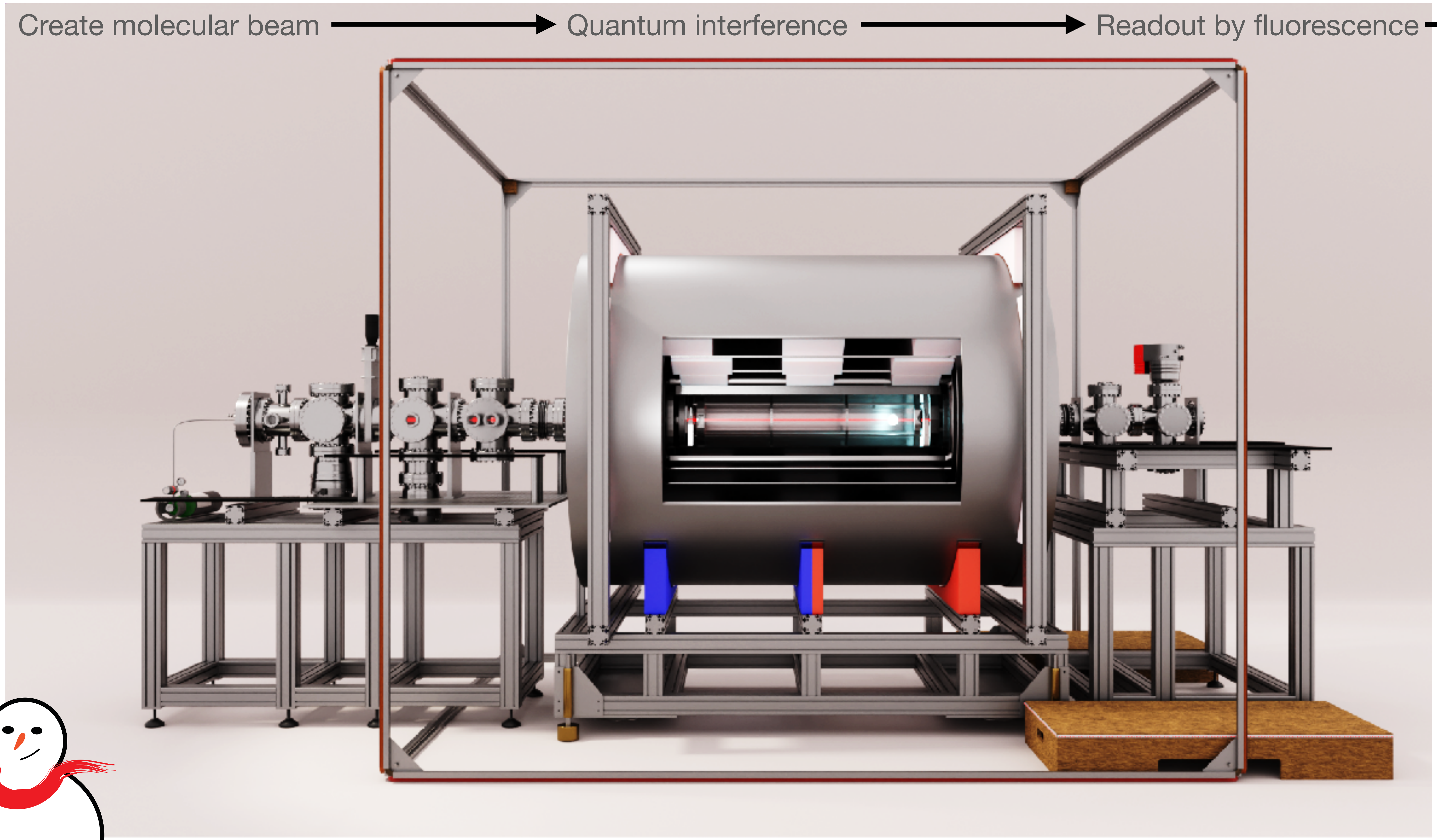


Interferometer phase $\phi = (\pm d_e E_{eff} \mp \mu_B B)T/\hbar$

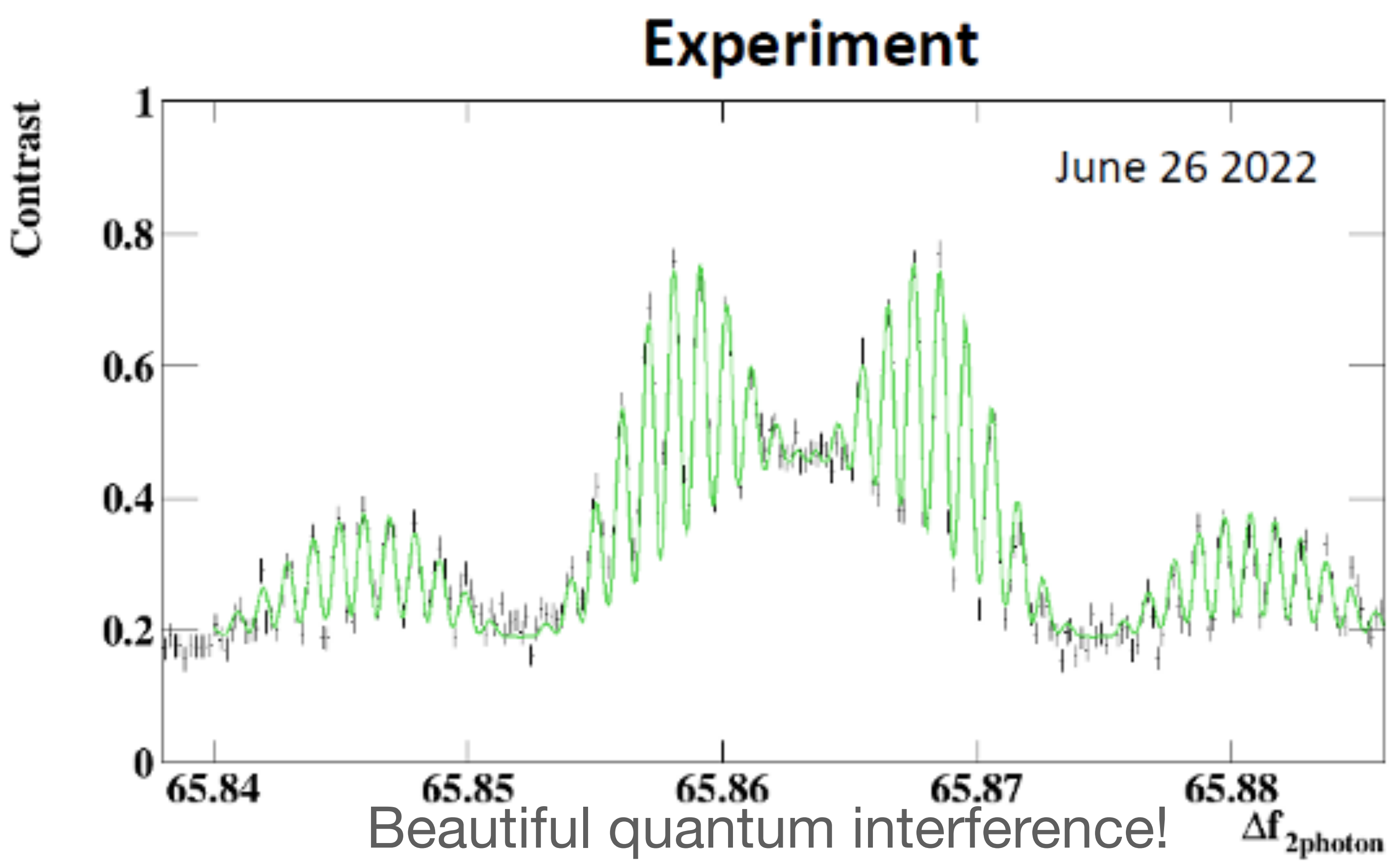


Interference data using fast molecular beam

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, Phys. Rev. A 110, L010801 (2024))

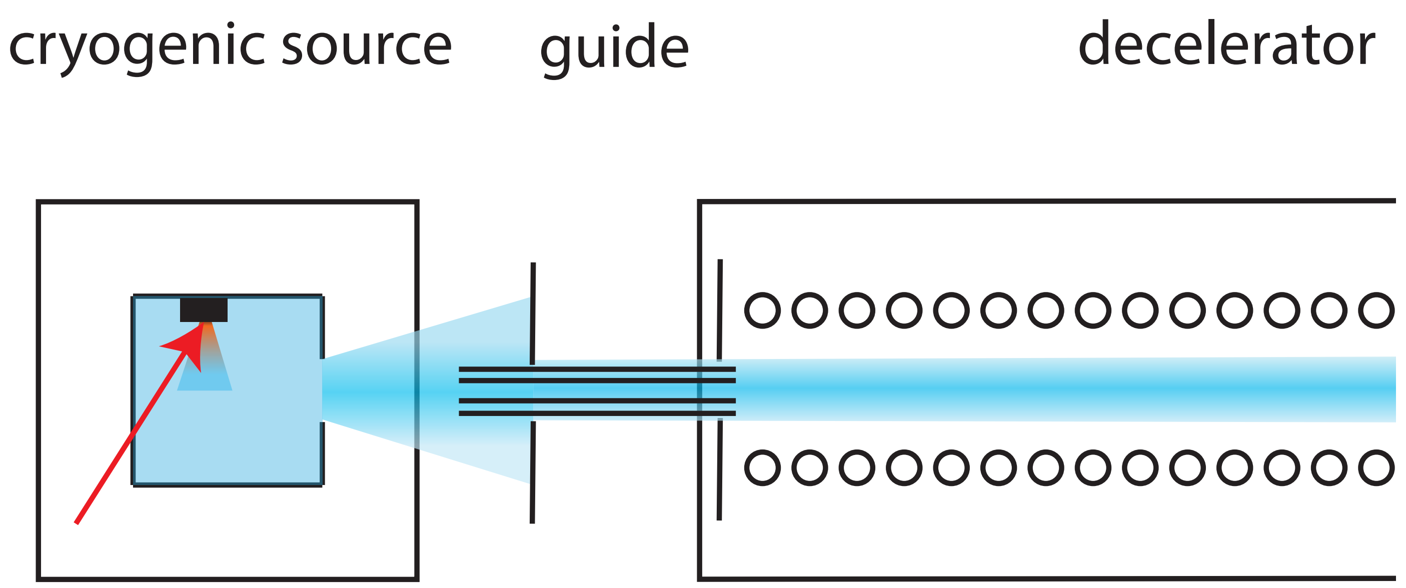
NL-eEDM

Some key experimental techniques

- Intense and slow molecular beam sources
- Hexapole lens and laser cooling
- Stark deceleration
- Combining it all: the ultimate experiment

An intense beam of molecules

How-to: source



Supersonic

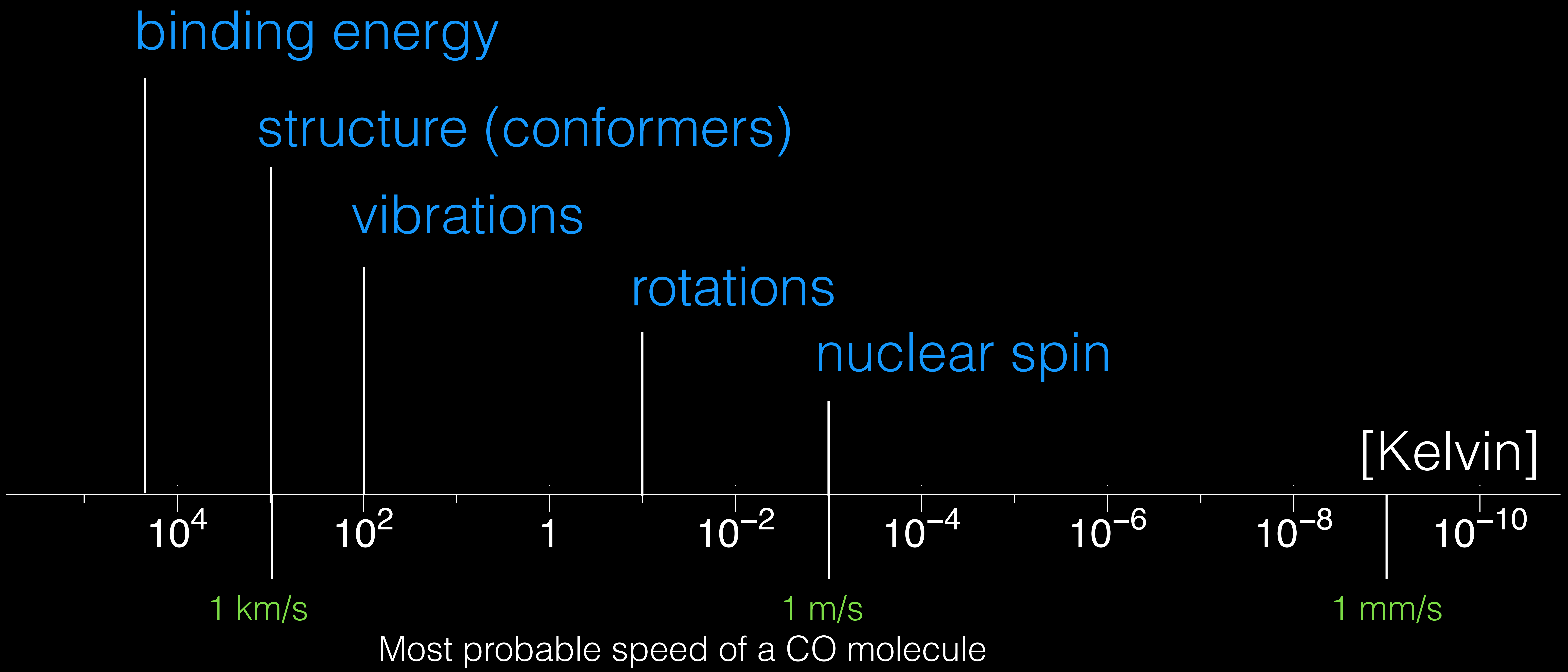
Aims:
- Intense, fast beam (600 m/s)
- Short pulse
- Test lasers systems, state manipulation and interaction zone

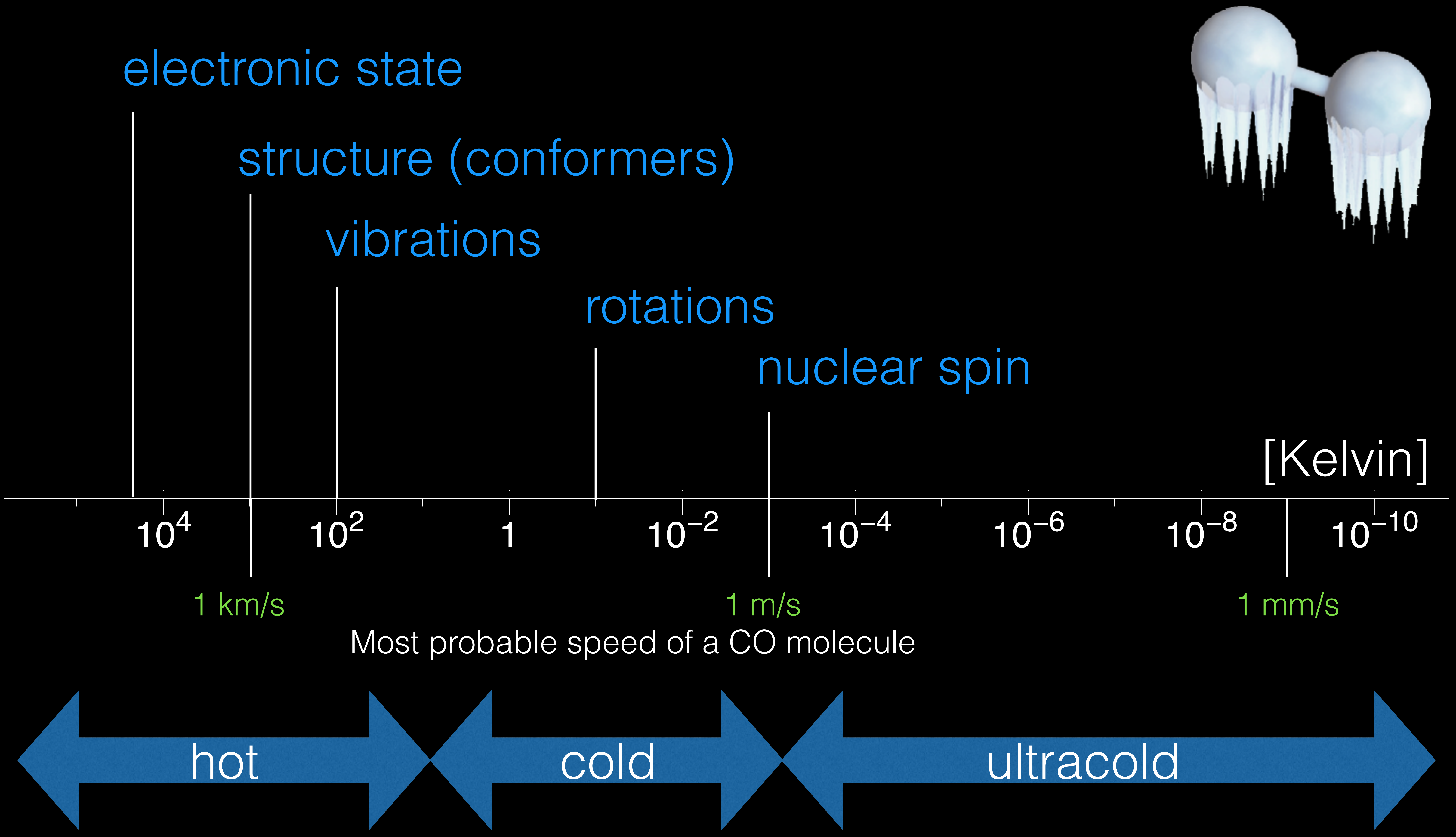
Cryogenic

Aims:
- slow beam (~180 m/s)
- High N: $4x10^9$/shot in the desired state
- Use for eEDM measurement

Why cold molecules?

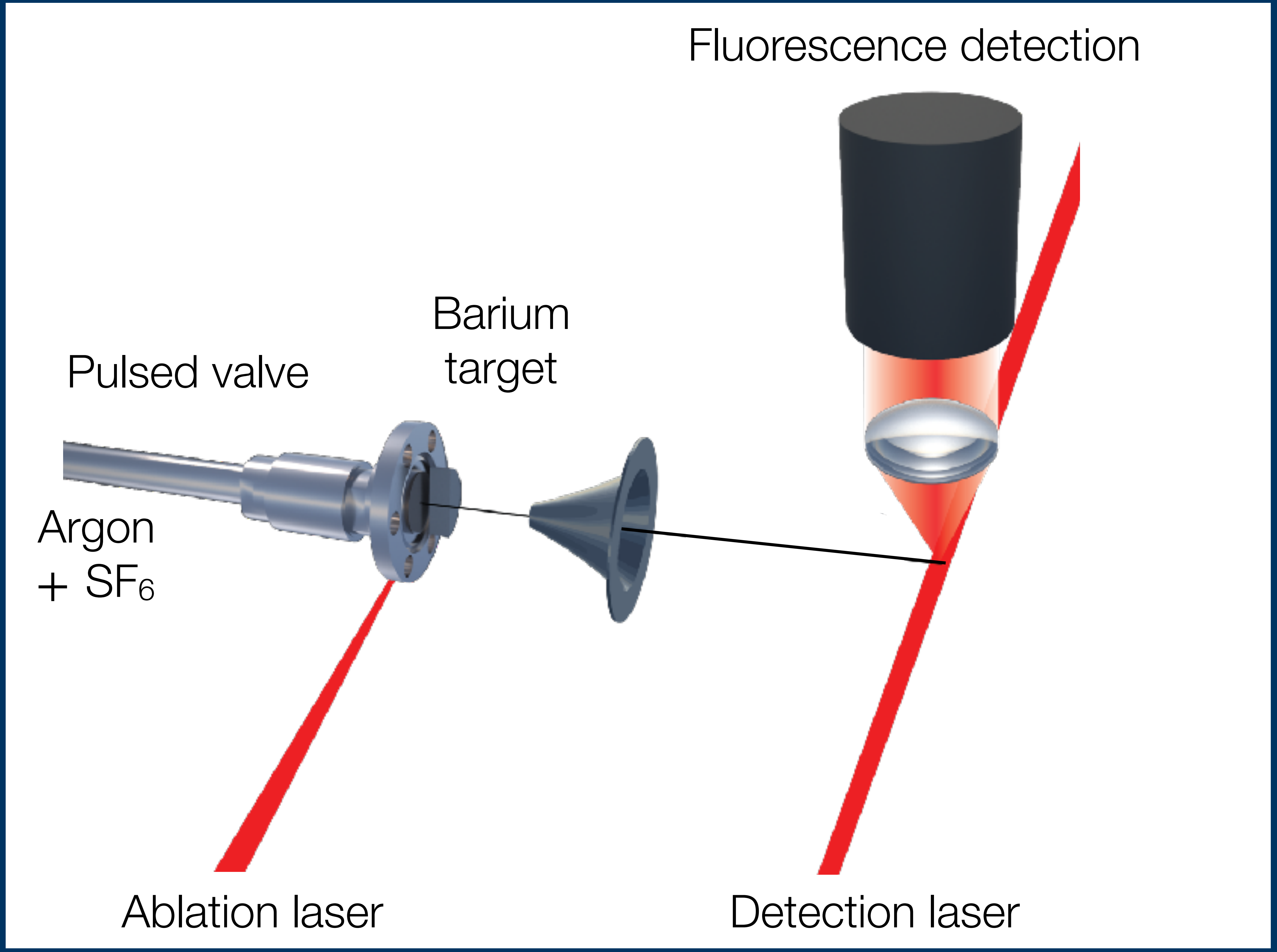
The temperature describes the statistical distribution of the *motional* degrees of freedom, but also the *internal* degrees of freedom





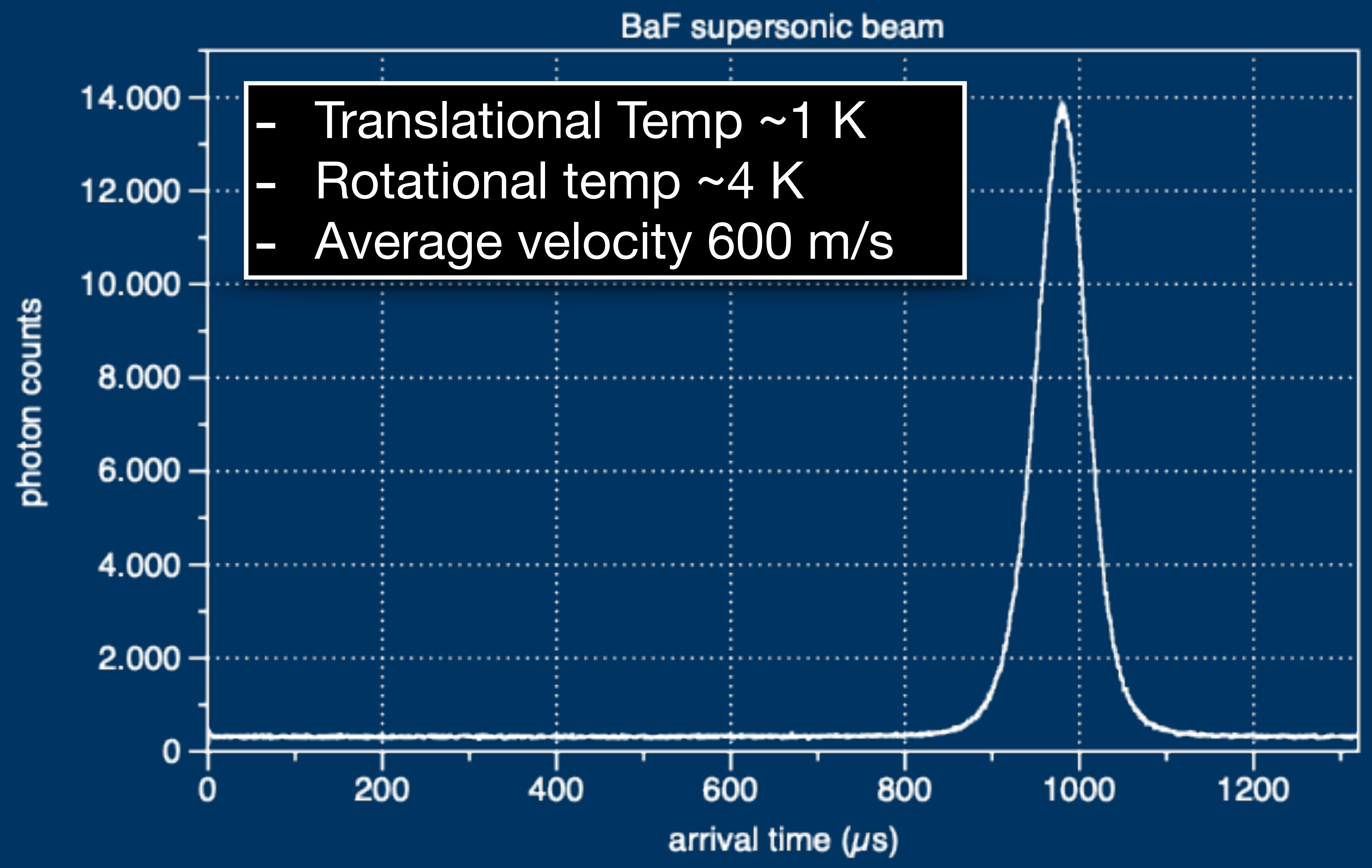
It is a big challenge to extend the control and precision of atomic physics to molecules.

Supersonic beams of SrF and BaF molecules

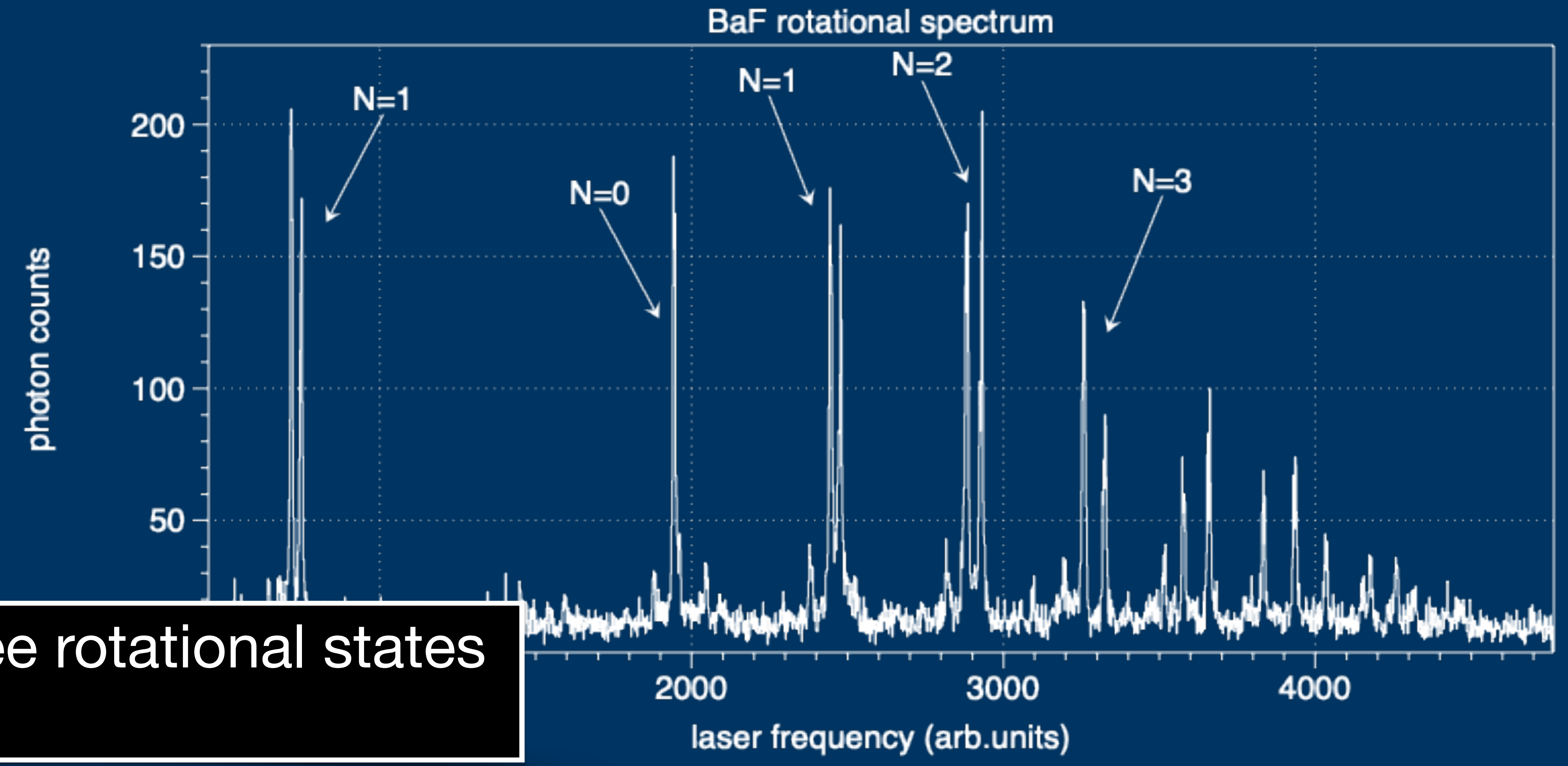


P. Aggarwal et al. A supersonic laser ablation beam source with narrow velocity spreads. Rev Sci Instrum 92, 033202 (2021).

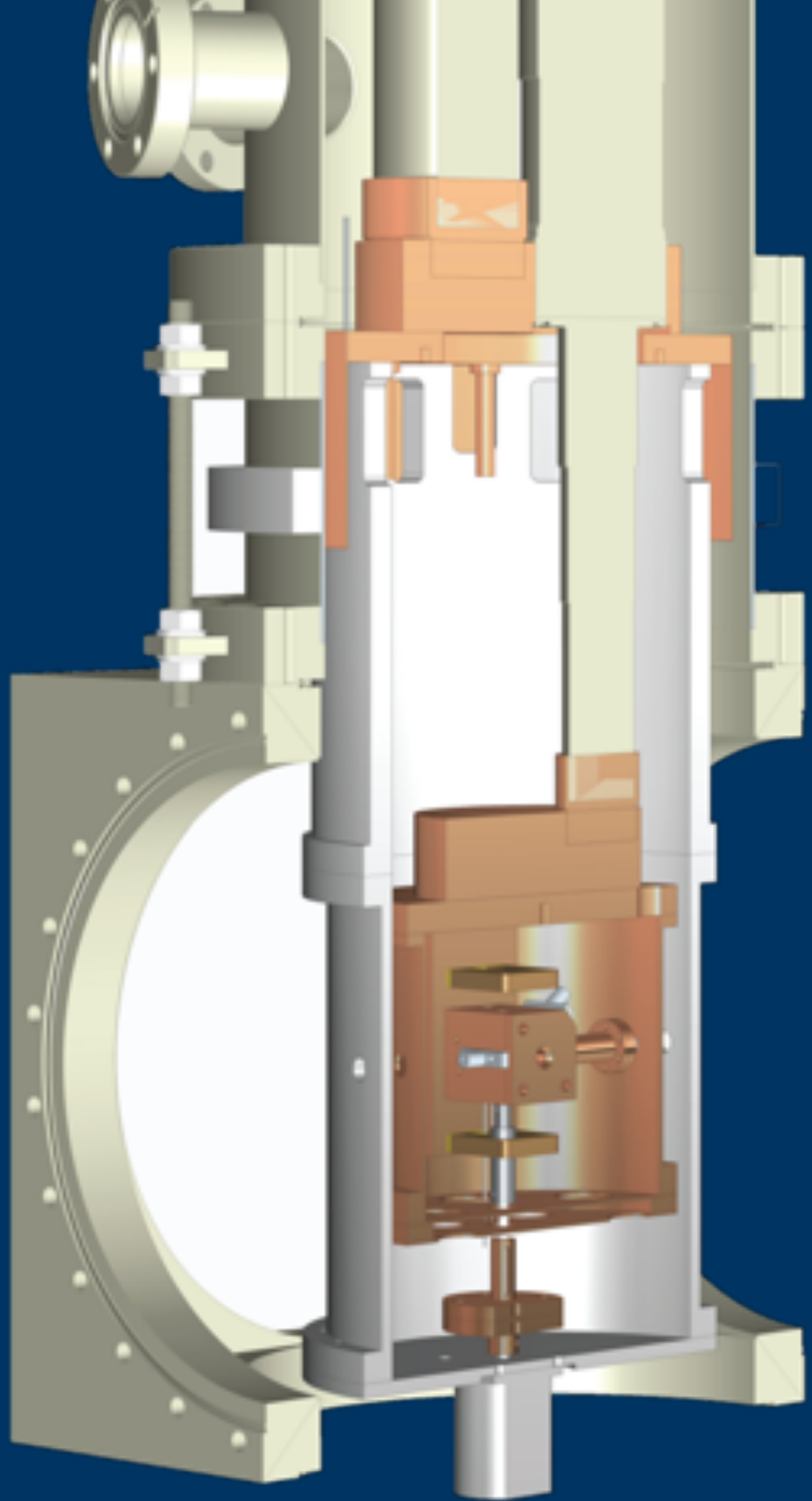
P. Aggarwal et al. Lifetime measurements of the A $^2\Pi_{1/2}$ and A $^2\Pi_{3/2}$ states in BaF. Phys Rev A 100, 052503 (2019).



- Translational Temp ~1 K
- Rotational temp ~4 K
- Average velocity 600 m/s



Lowest three rotational states populated



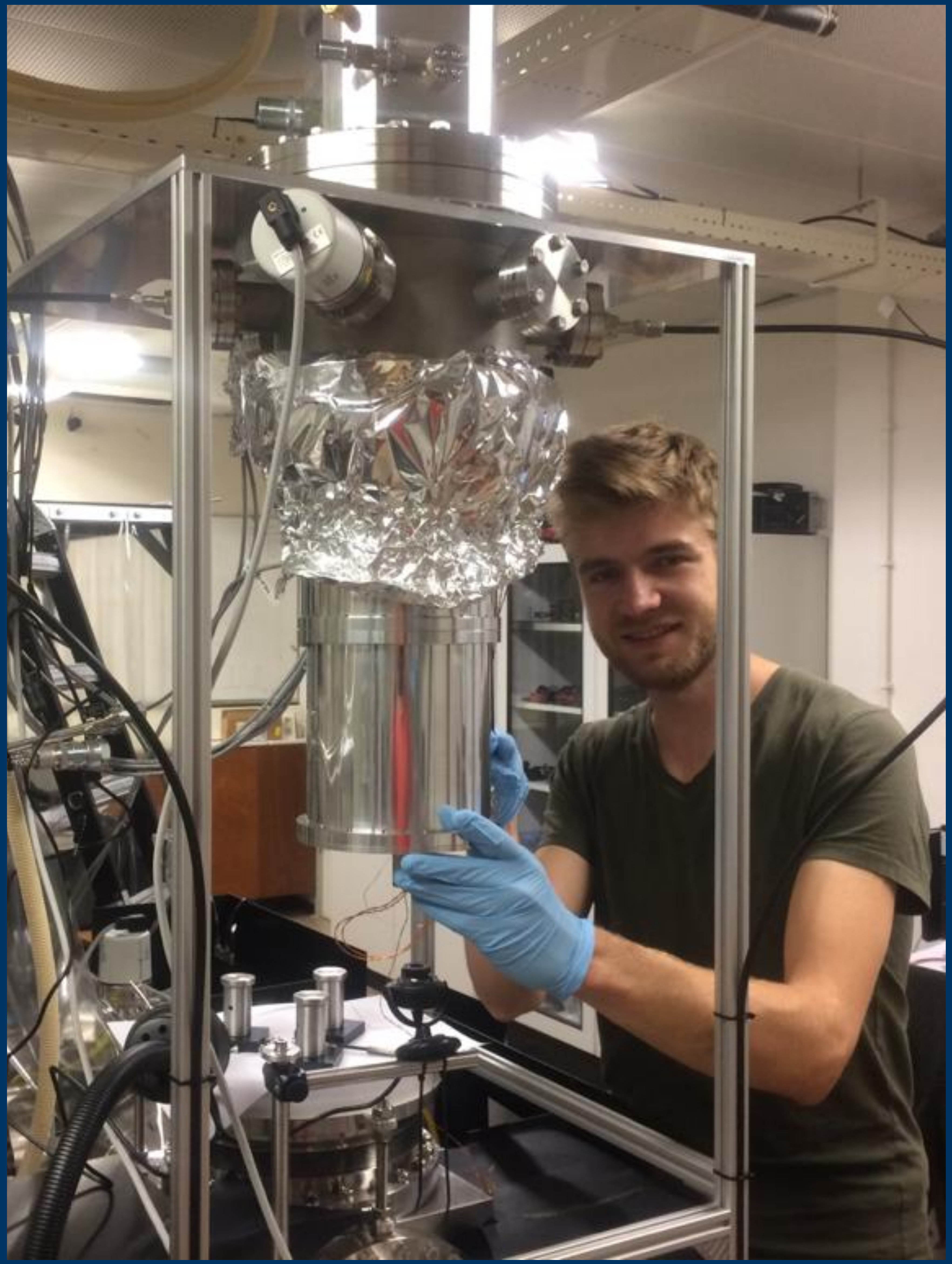
Cryogenic beam

- Evaporating Sr metal target
- Neon carrier gas + SF_6
- Absorption,1 cm from cell
- Fluorescence, 30 cm from cell
- Translational Temp ~10 K
- Velocity 150-200 m/s

Based on design from Stefan Truppe, Mike Tarbutt @ Imperial

Goal: make the most intense slow source of BaF molecules

1 in Groningen (BaF, production)
1 in A'dam (BaF, optimisation)
1 in Groningen (BaOH, exploration)

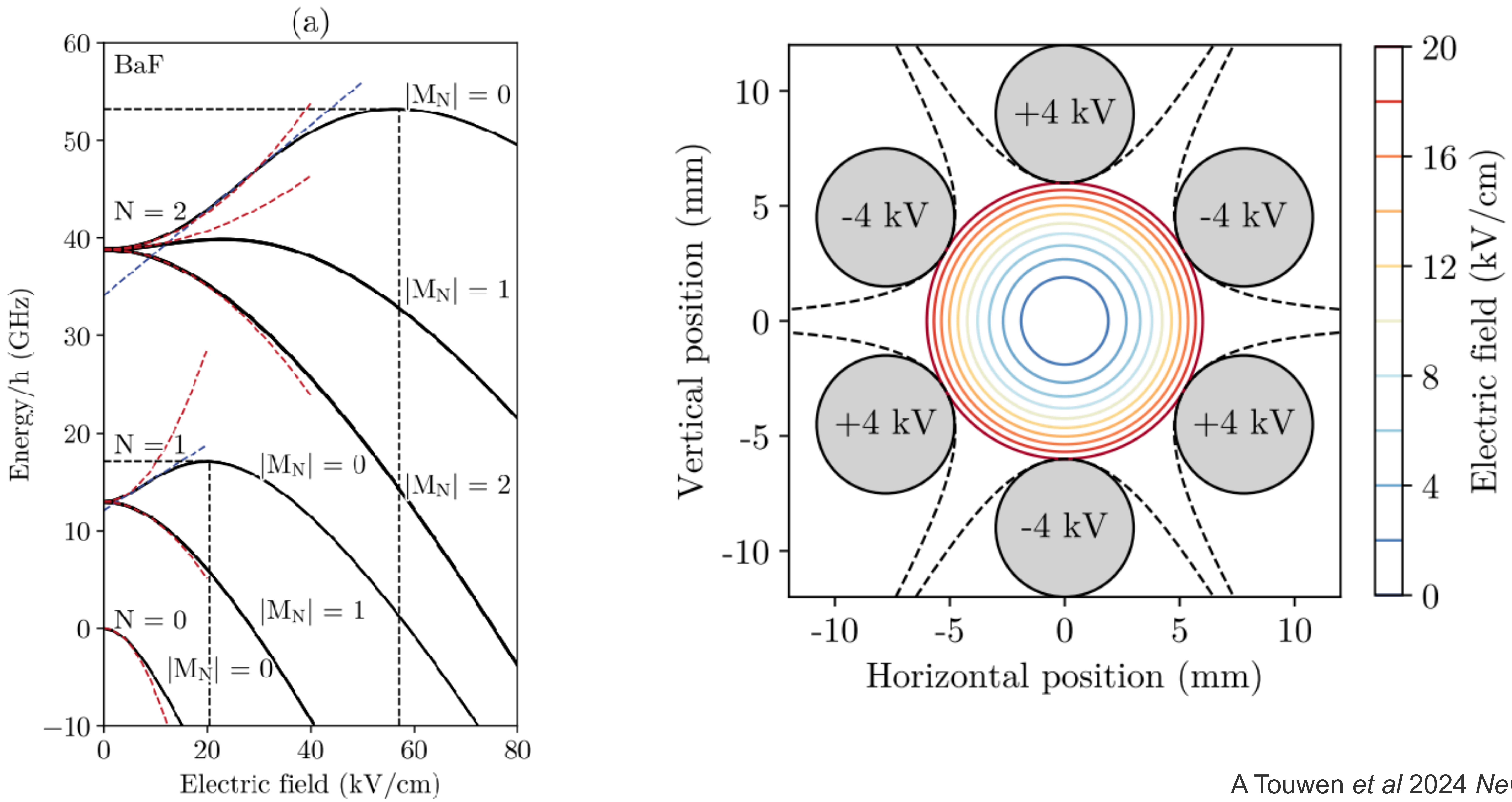


Maarten Mooij, Rick Bethlem @ VU Amsterdam

Mooij et al, New Journal of Physics, vol. 26, 2024, p. 053009
Mooij et al, J. Phys. B: At. Mol. Opt. Phys., vol. 58, 2025, p. 015303

Making a cryogenic molecular beam

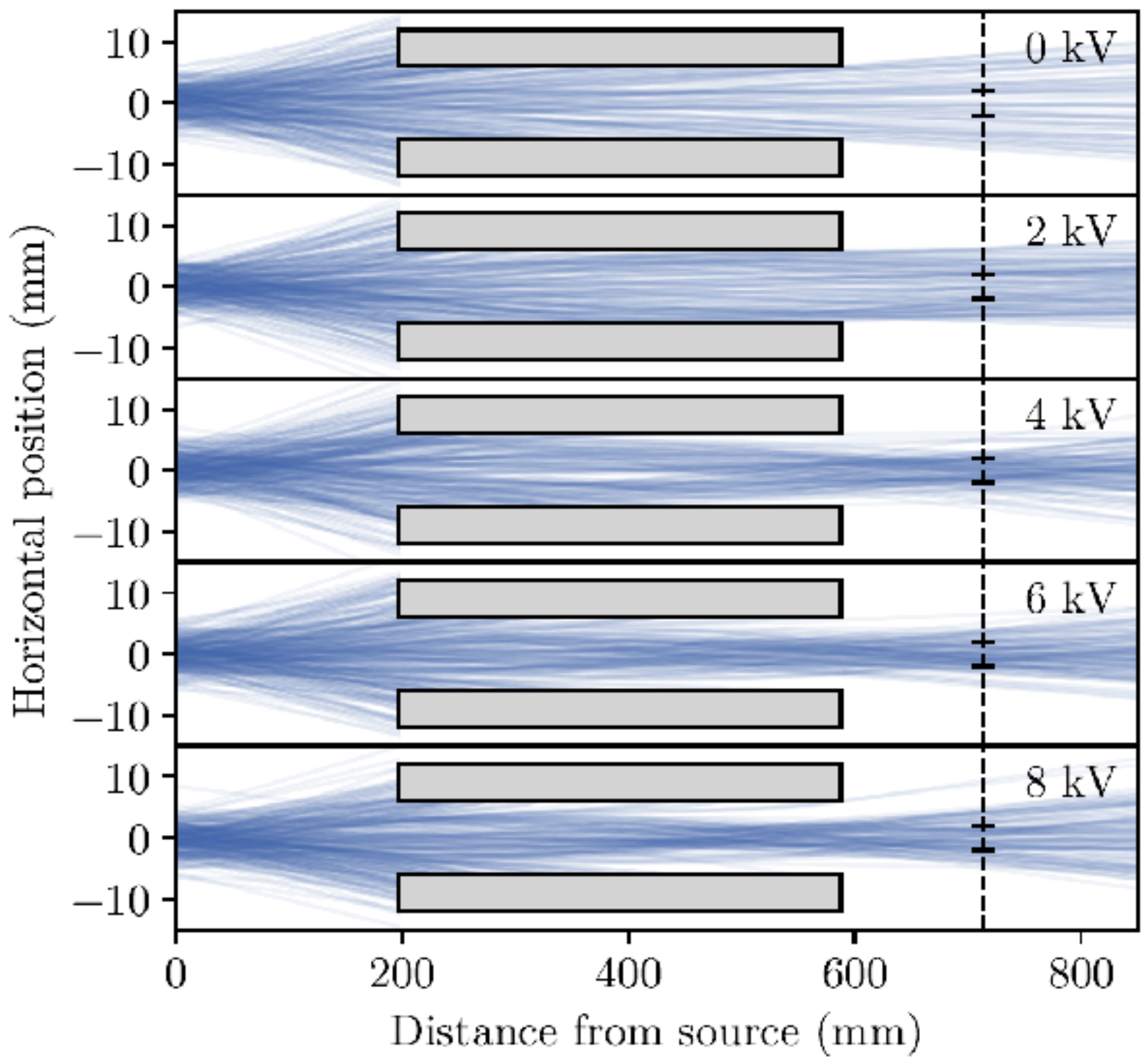
BaF in electric fields: the hexapole lens



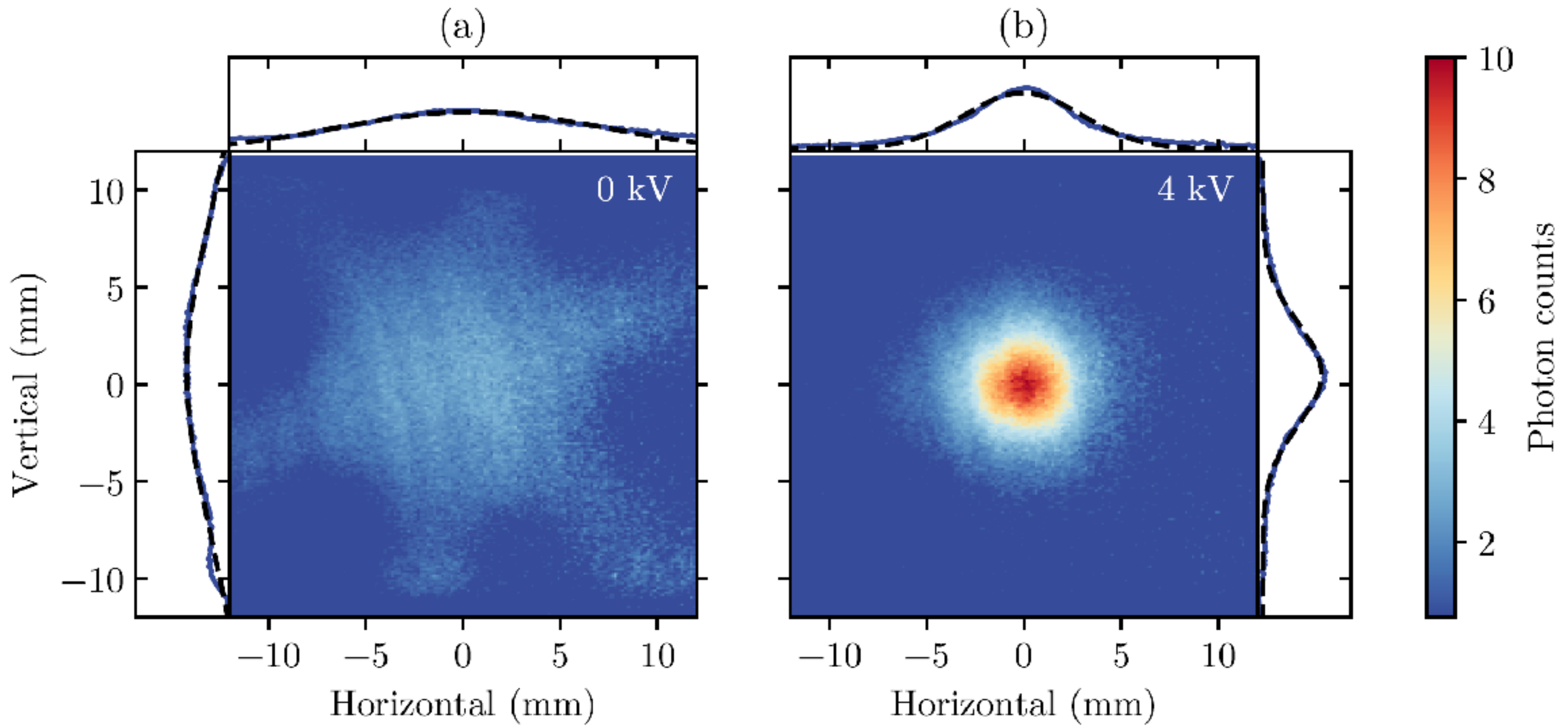
A Touwen *et al* 2024 *New J. Phys.* **26** 073054

BaF in electric fields: the hexapole lens

Simulation



Measurement

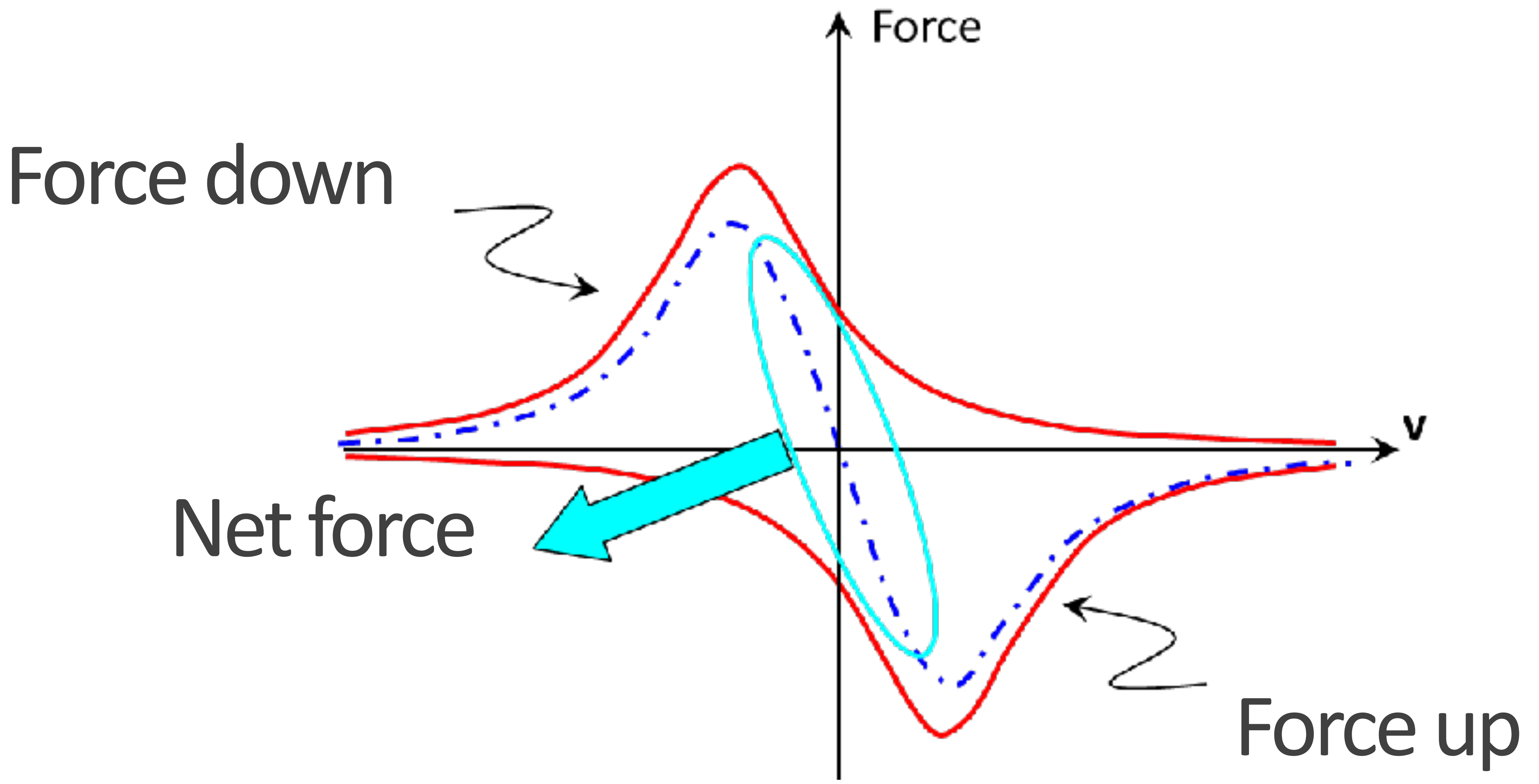


Focus molecules with +-5 m/s into a relatively small area, and then…

A Touwen *et al* 2024 *New J. Phys.* **26** 073054

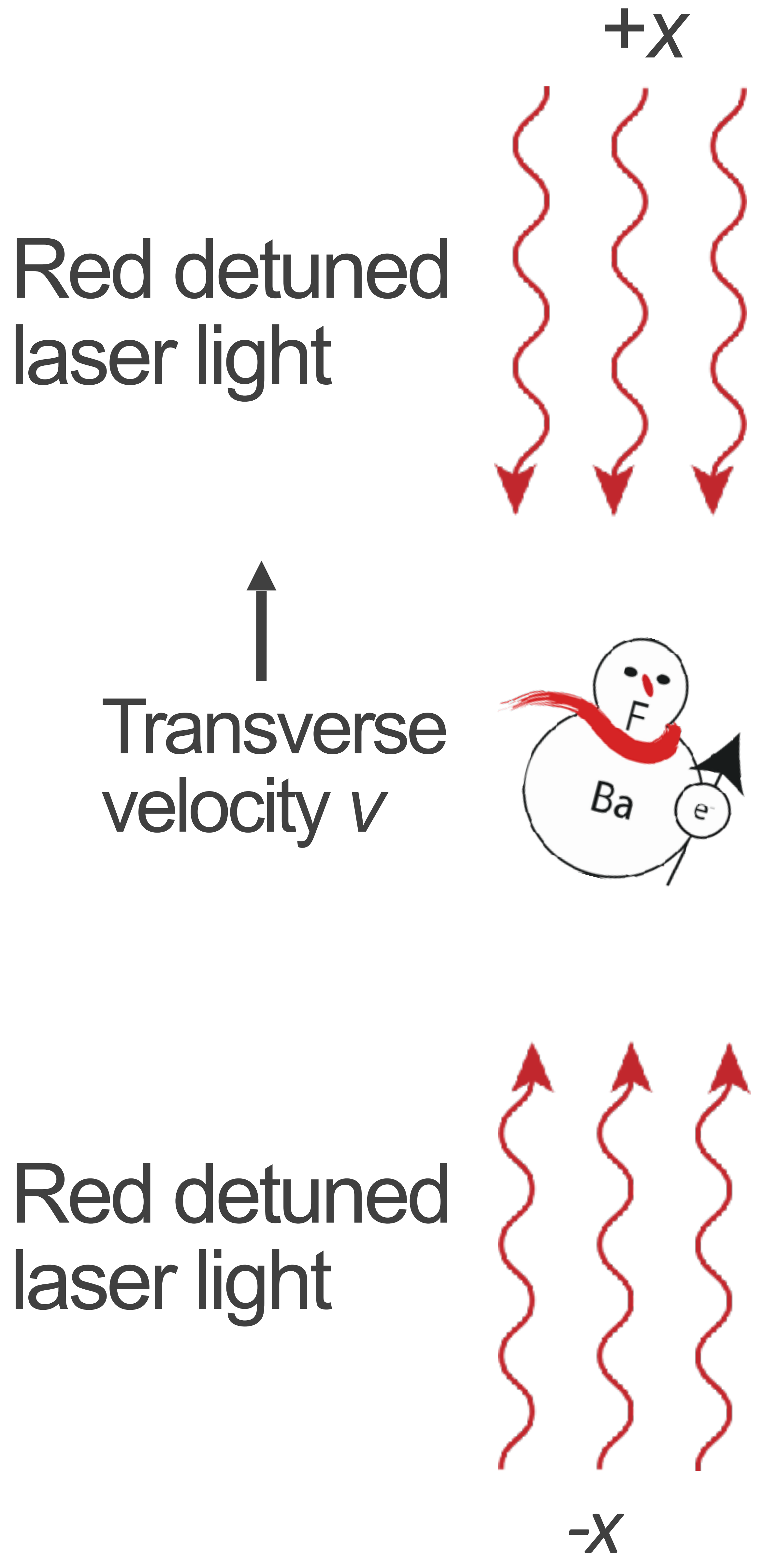
Transverse laser cooling

Beam collimation using Doppler laser cooling



Consequence: force opposing molecule´s transverse velocity.

Lets apply it to our BaF molecule!



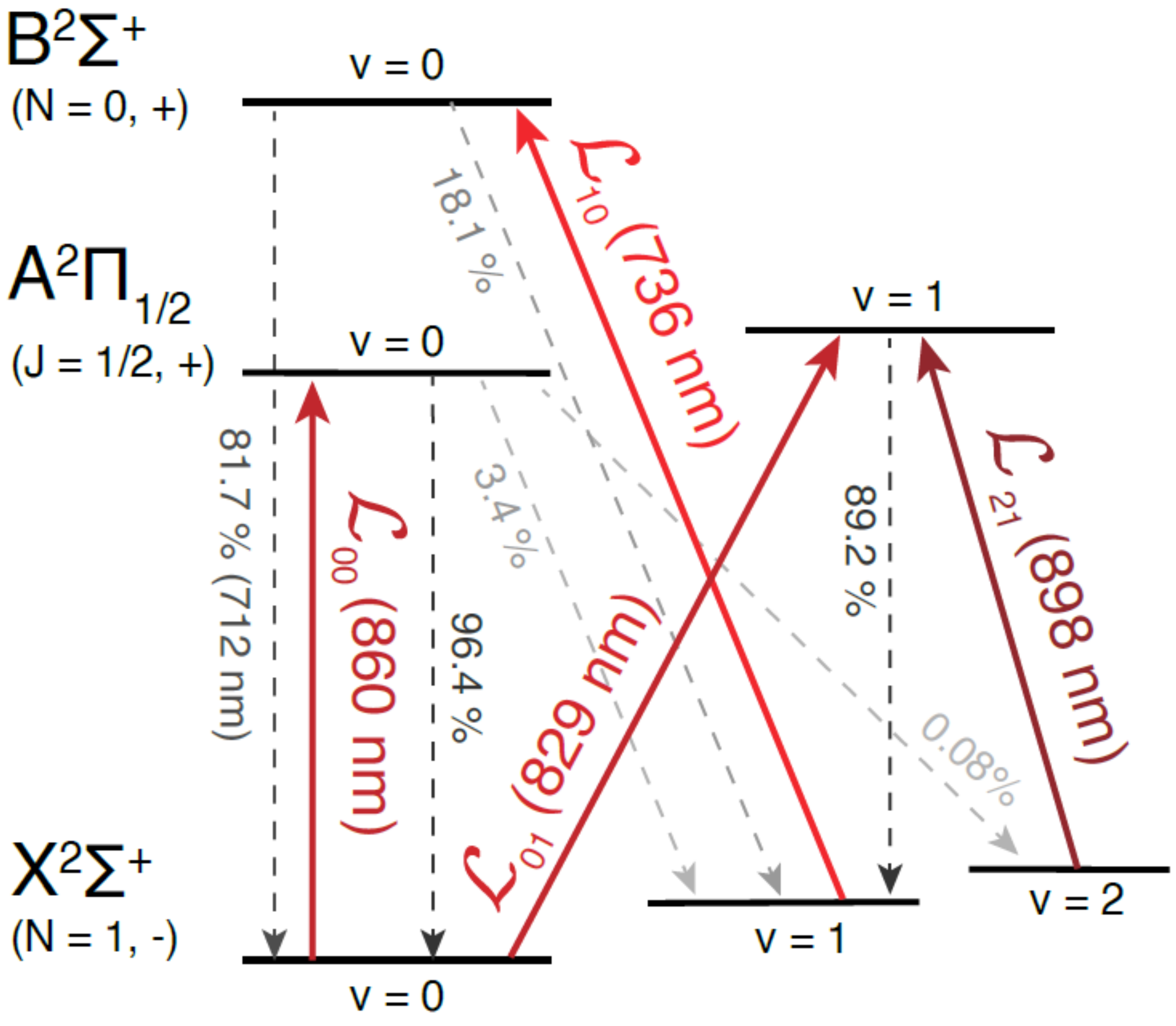
Complexity of laser cooling BaF

Vibrational branching,

hyperfine structure,

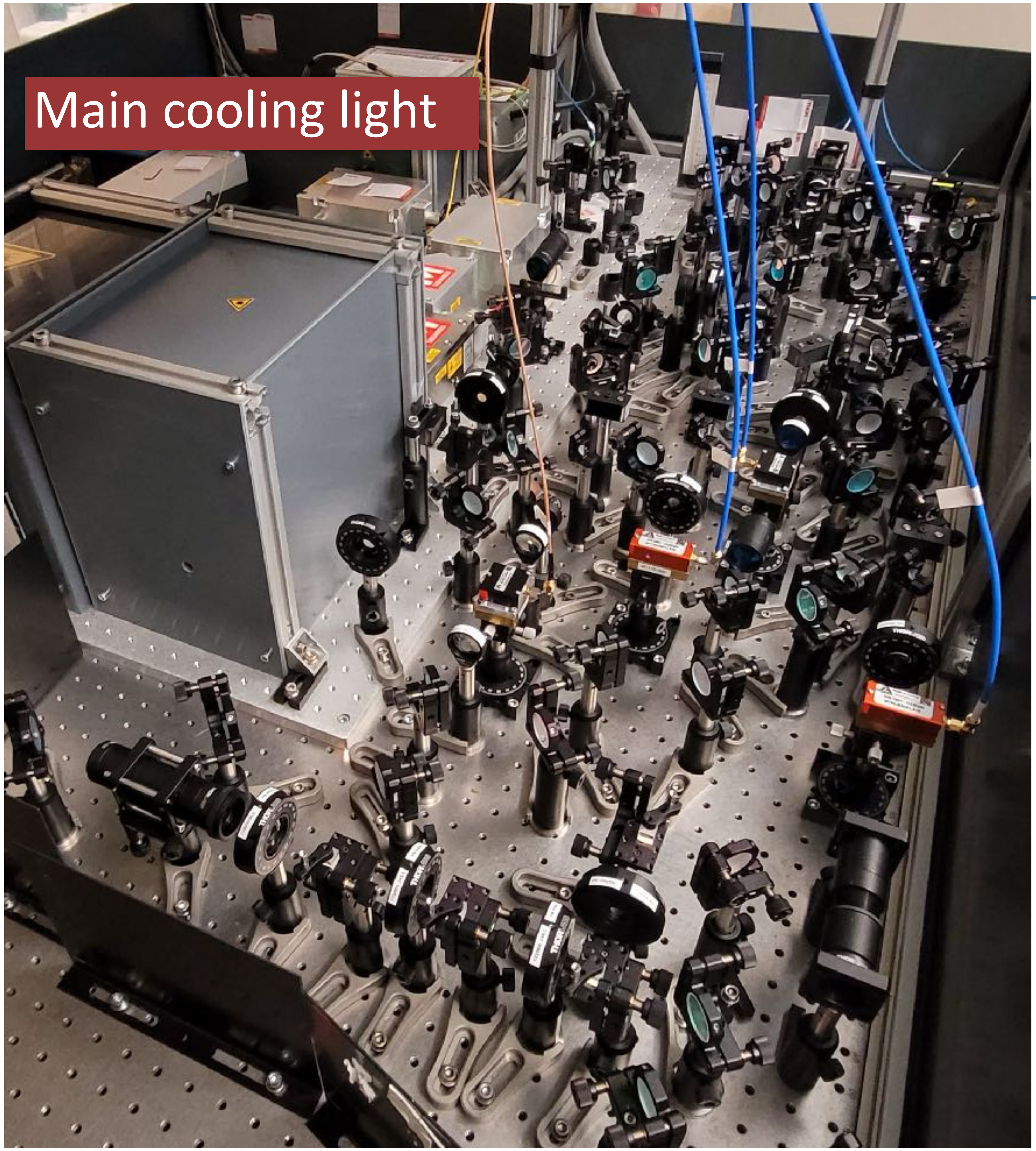
dark Zeeman state remixing,…

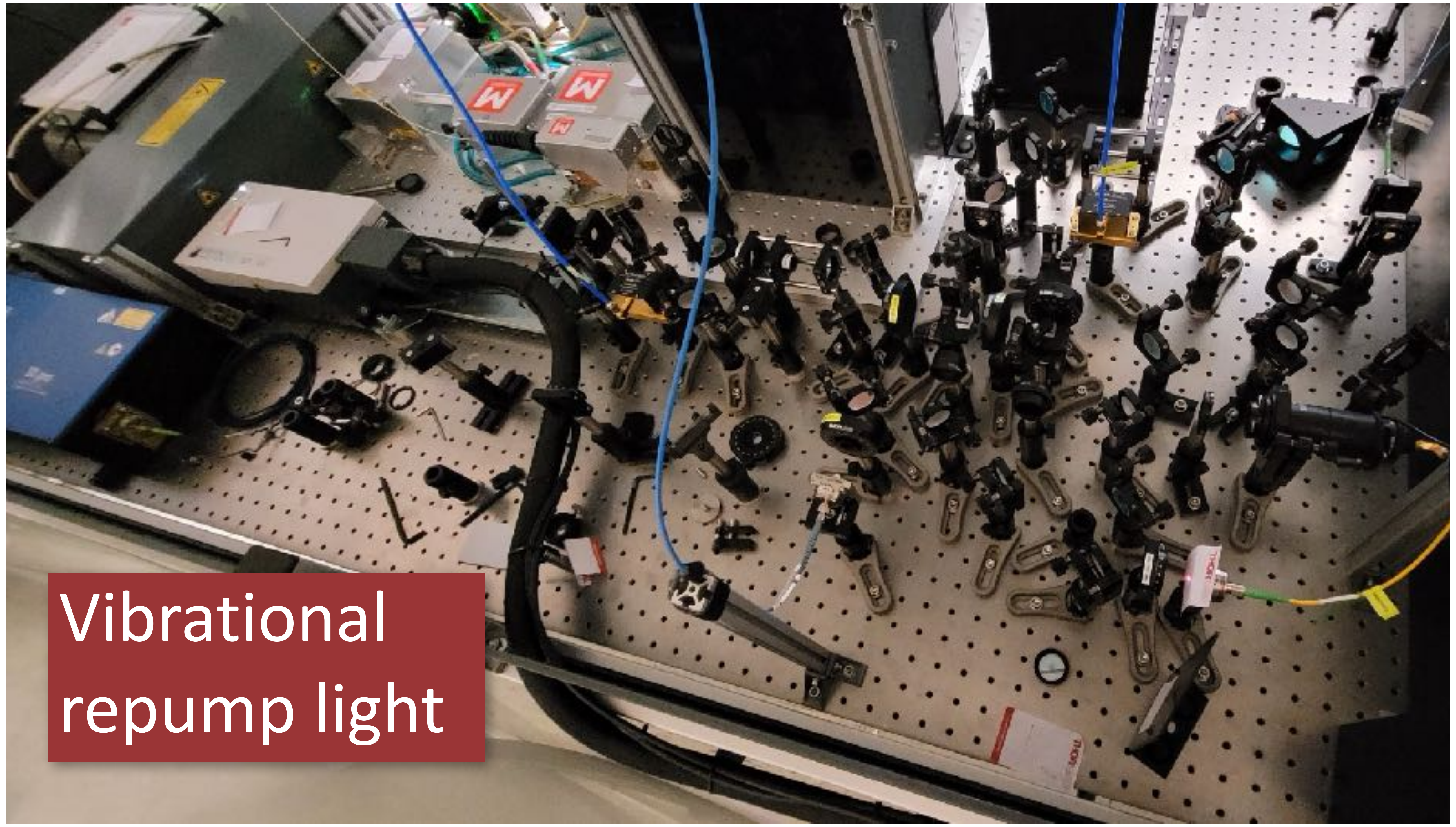
Complicate closing the cycle



Hao et al, *J. Chem. Phys.* 151, 034302 (2019)

Generating all the necessary laser light requires some optics



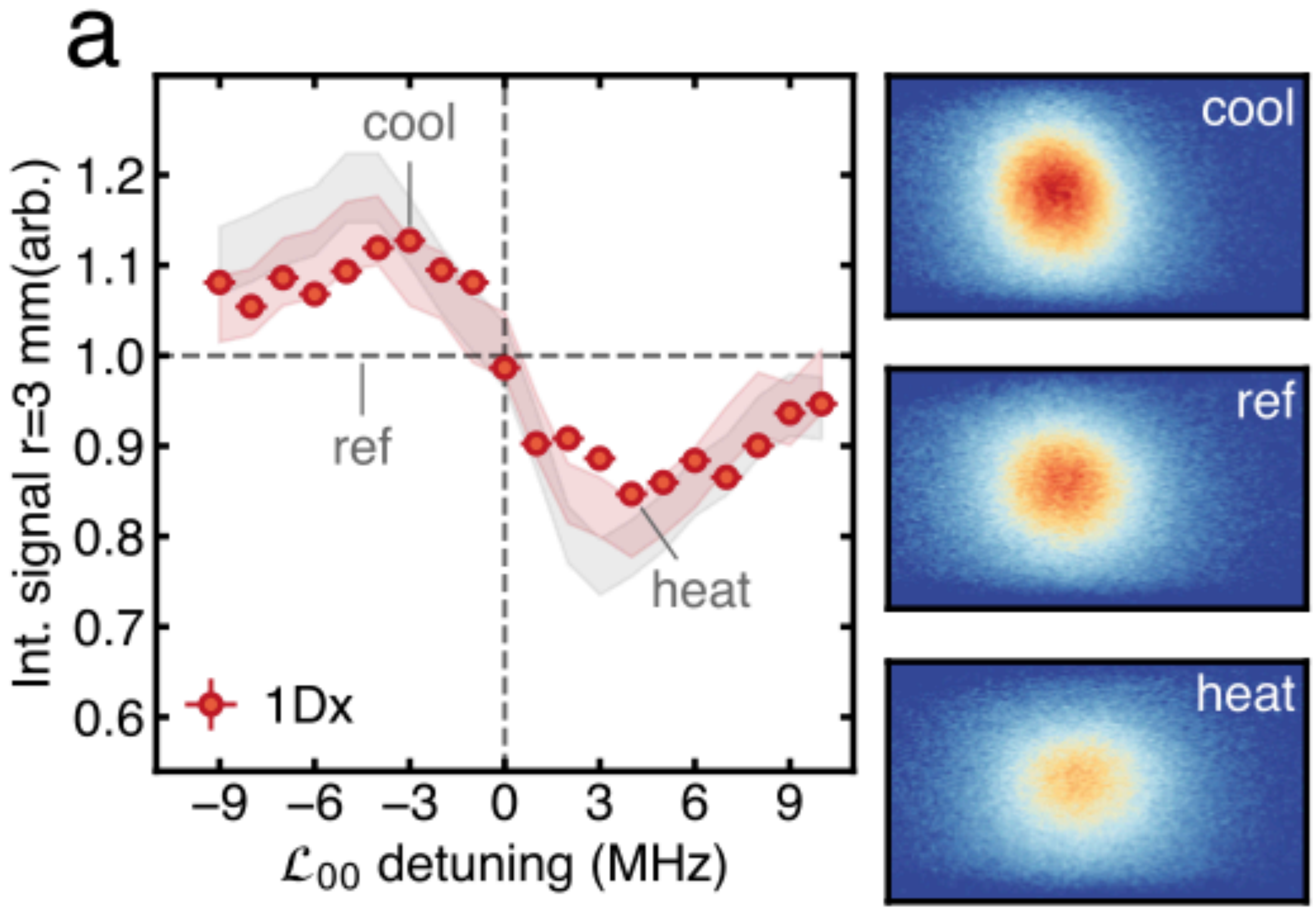


2D transverse laser cooling of a hexapole focused beam of cold BaF molecules

J.W.F. van Hofslot,[1,2] I.E. Thompson,[1] A. Touwen,[1,2,3] N. Balasubramanian,[1,2] R. Bause,[1,2,*] H.L. Bethlem,[1,3] A. Borschevsky,[1,2] T.H. Fikkers,[1,2] S. Hoekstra,[1,2,†] S.A. Jones,[1,2] J.E.J. Levenga,[1,2] M.C. Mooij,[2,3] H. Mulder,[1,2] B.A. Nijman,[1,2] E.H. Prinsen,[1,2] B.J. Schellenberg,[1,2] L. van Sloten,[1,2] R.G.E. Timmermans,[1,2] W. Ubachs,[3] J. de Vries,[1,4] and L. Willmann,[1,2] for the NL-*e*EDM collaboration

[1] *Van Swinderen Institute for Particle Physics and Gravity, University of Groningen, The Netherlands*
[2] *Nikhef, National Institute for Subatomic Physics, Amsterdam, The Netherlands*
[3] *Department of Physics and Astronomy, and LaserLaB, Vrije Universiteit Amsterdam, The Netherlands*
[4] *Institute of Physics and Delta Institute for Theoretical Physics, University of Amsterdam, The Netherlands*

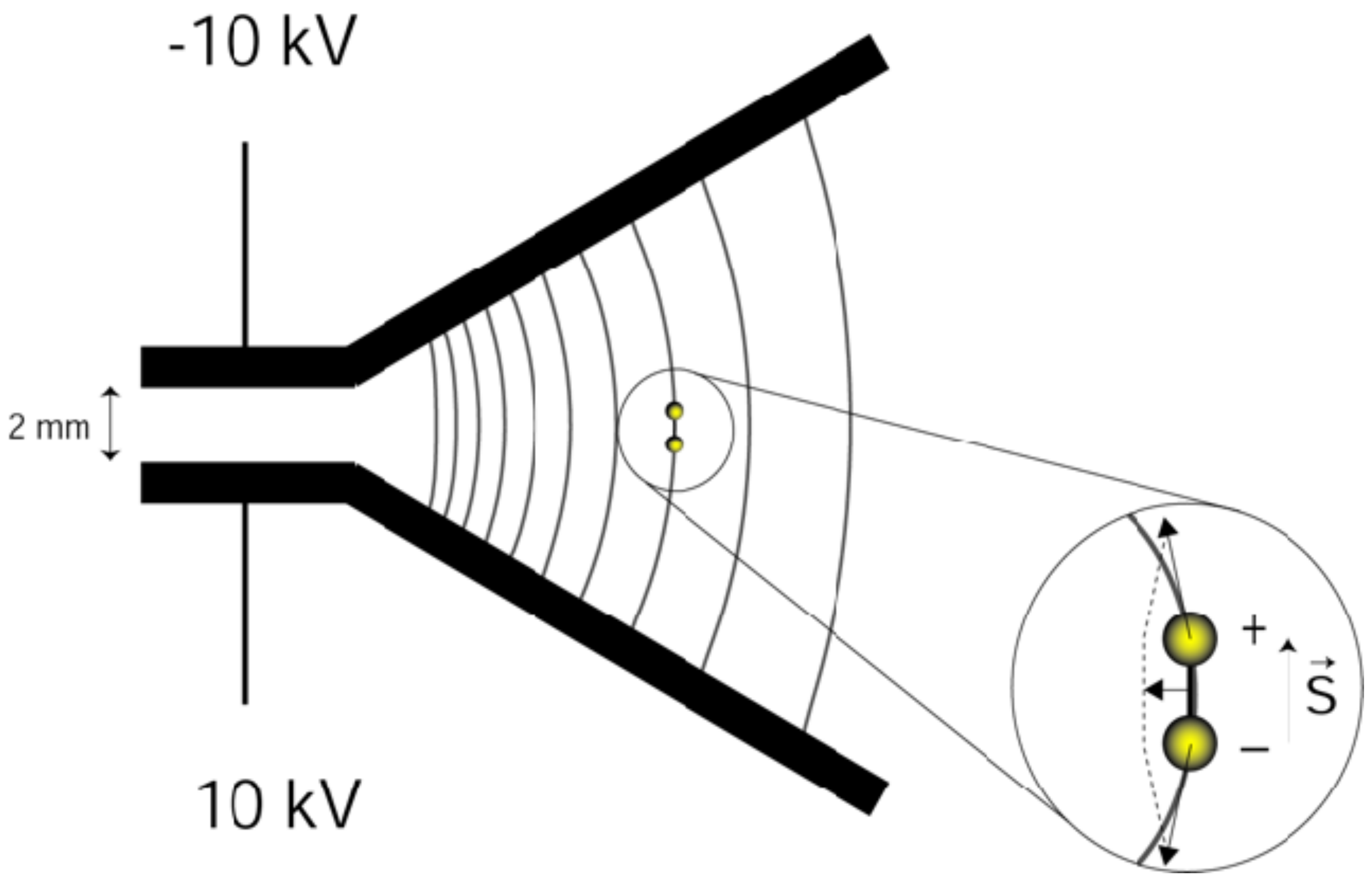
(Dated: June 25, 2025)



Recent results!
arXiv:2506.19069v1

Molecule decelerator

Uniform field leads to torque -> rotation
What about a non-uniform field?



Polar molecule aligned along electric field lines, but still a force on the molecule!

Depending on orientation, force is towards or away from electric field maximum

Basis of Stark deceleration technique, breakthrough in ‘**cold molecule**’ research

$$\vec{F} = \vec{F}_+ + \vec{F}_- = q(\vec{E}_+ - \vec{E}_-) = q(\Delta\vec{E})$$

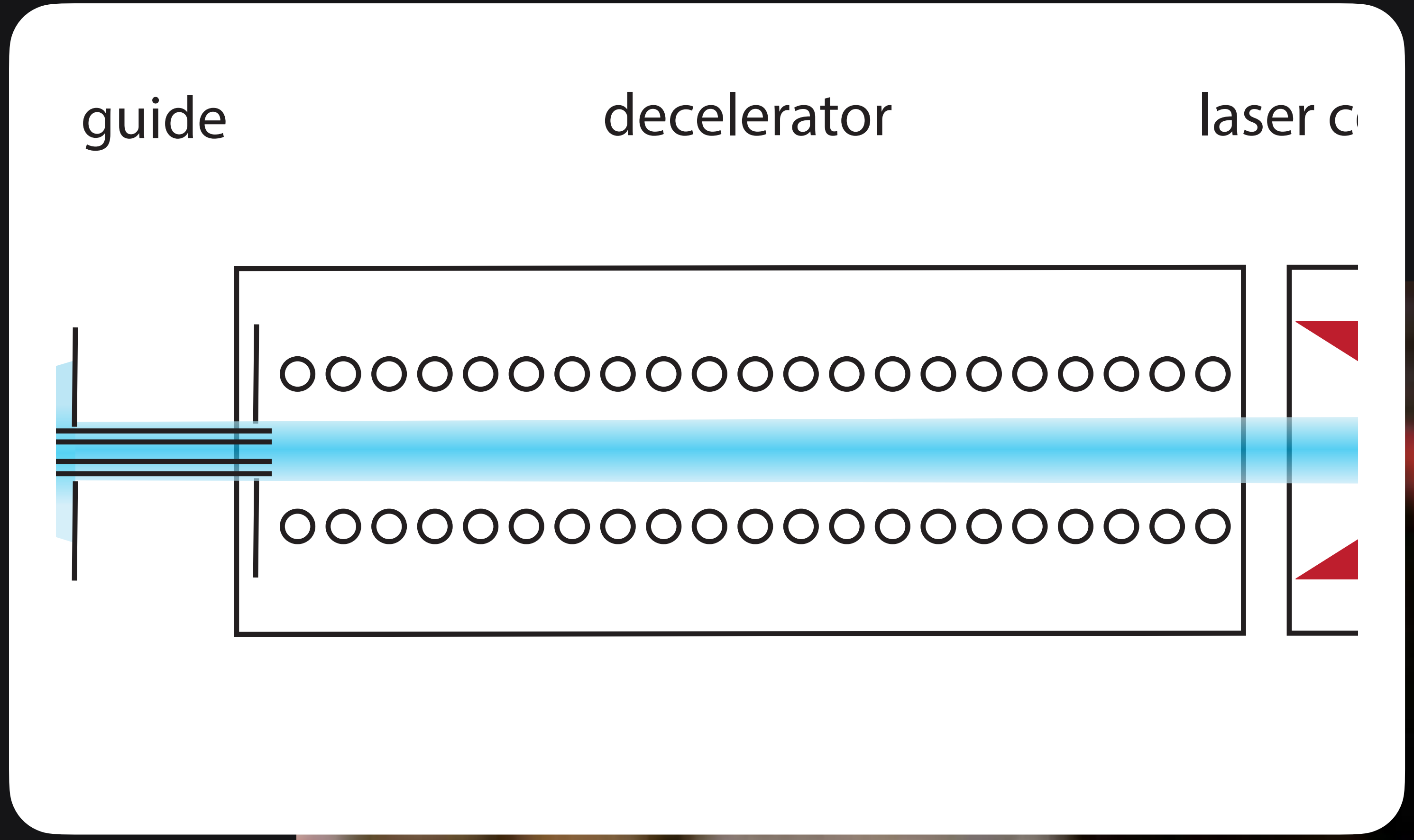
$$\Delta\vec{E} = (\vec{d}\cdot\vec{\nabla})\vec{E} \quad \text{so } \vec{F} = q(\vec{d}\cdot\vec{\nabla})\vec{E} = (\vec{p}\cdot\vec{\nabla})\vec{E}$$

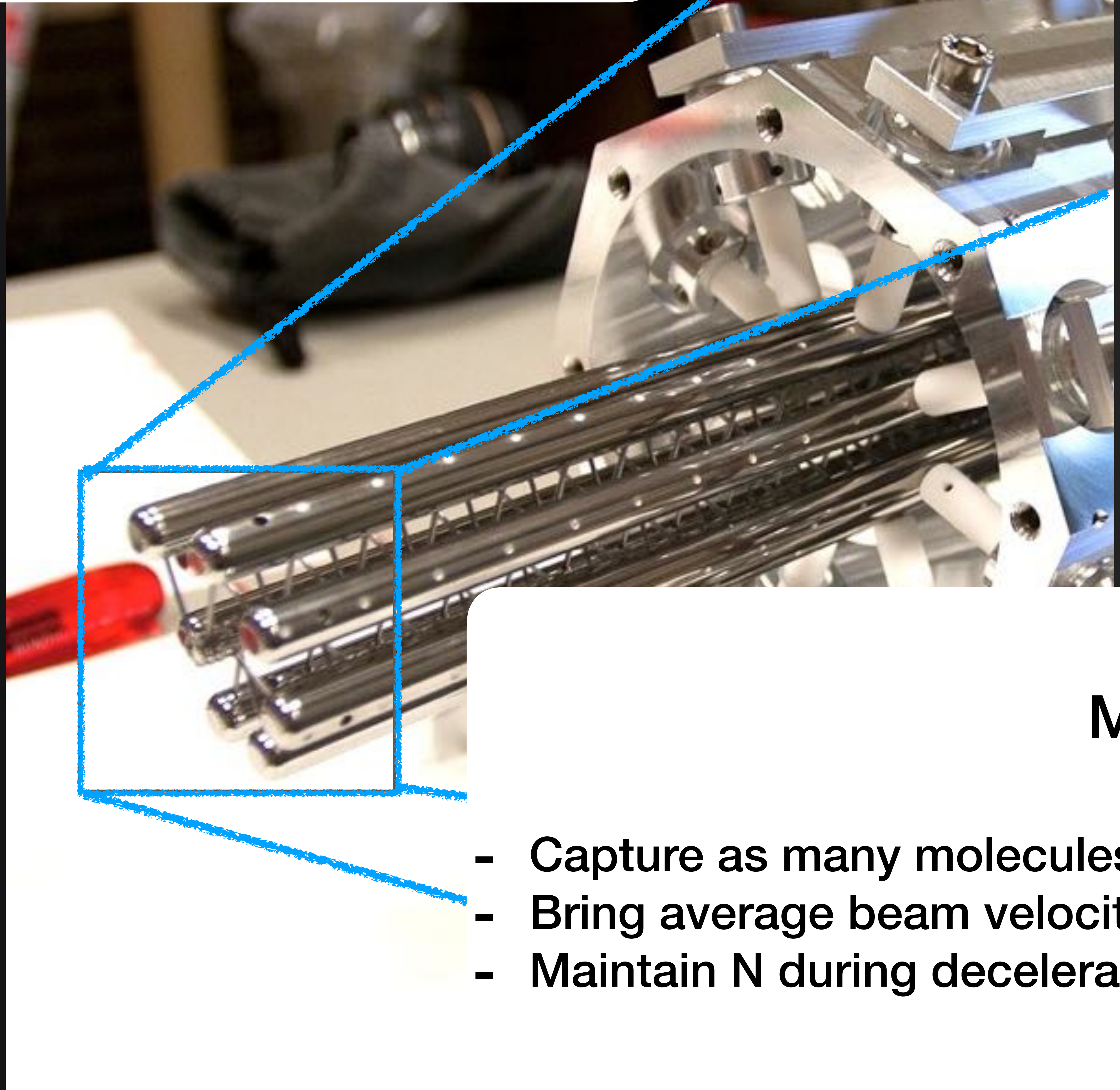
The force on a dipole is proportional to the gradient of the electric field

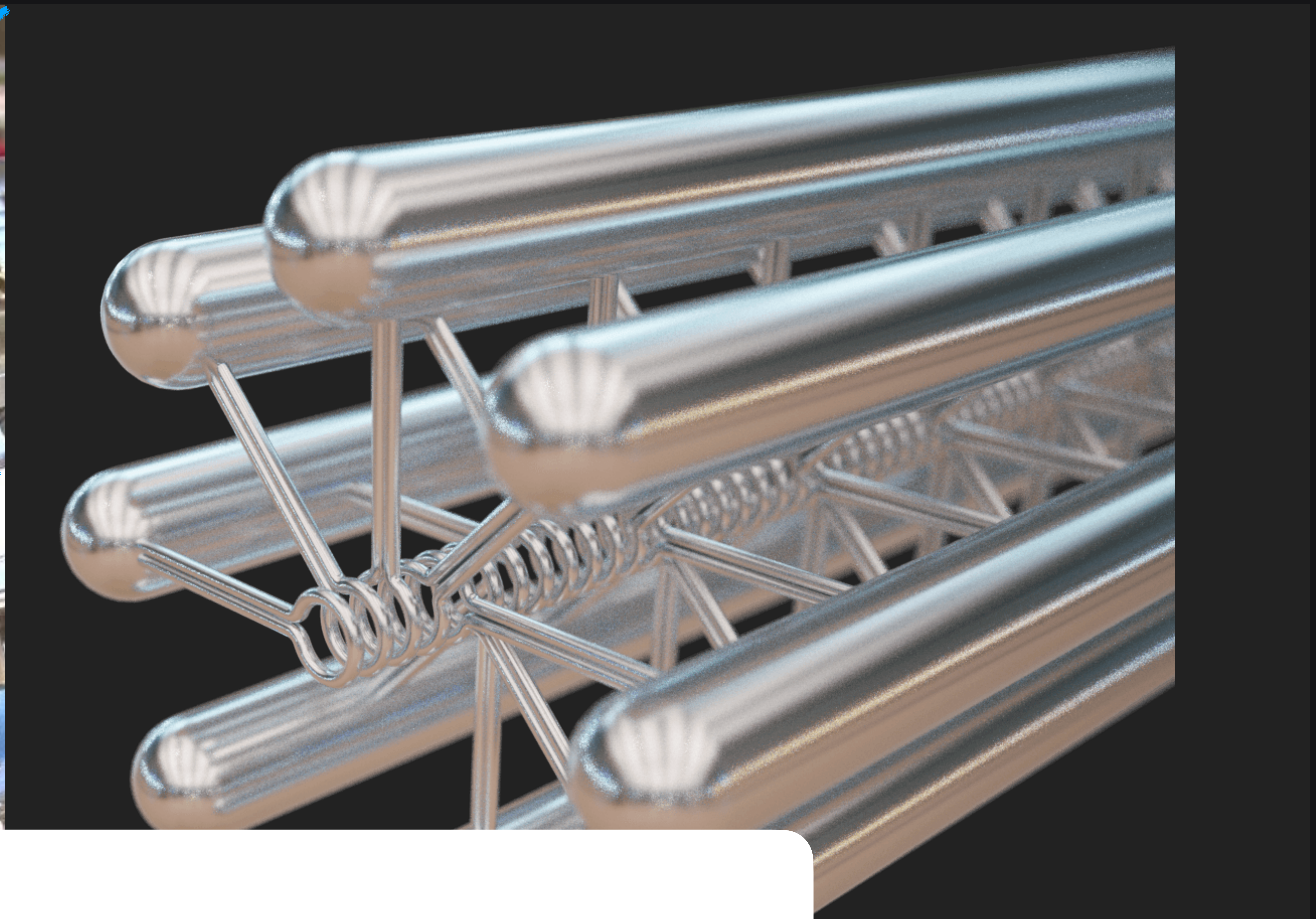
A slow beam of molecules

A traveling-wave with a tunable velocity

Traveling-wave decelerator



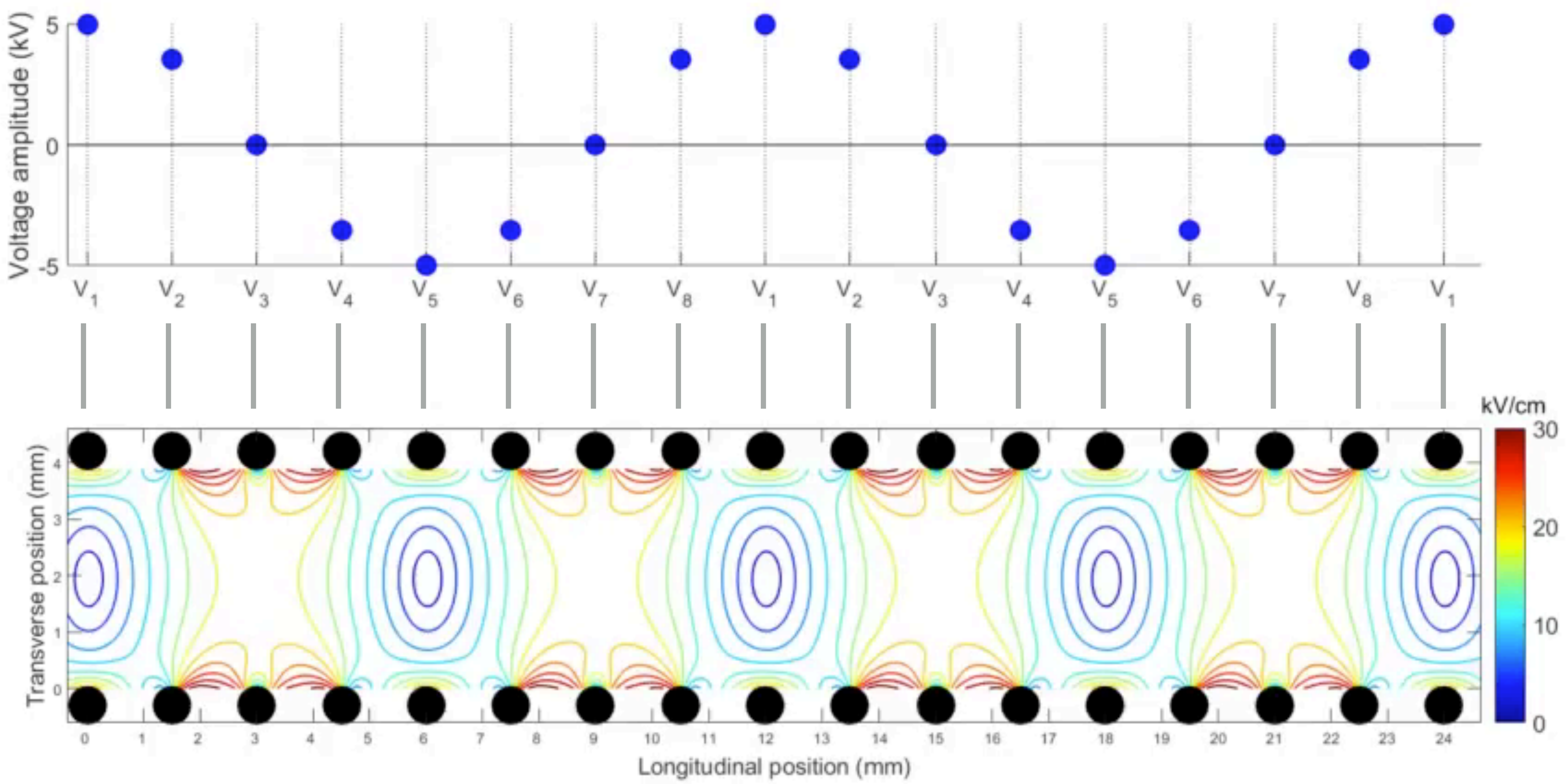




Main aims:

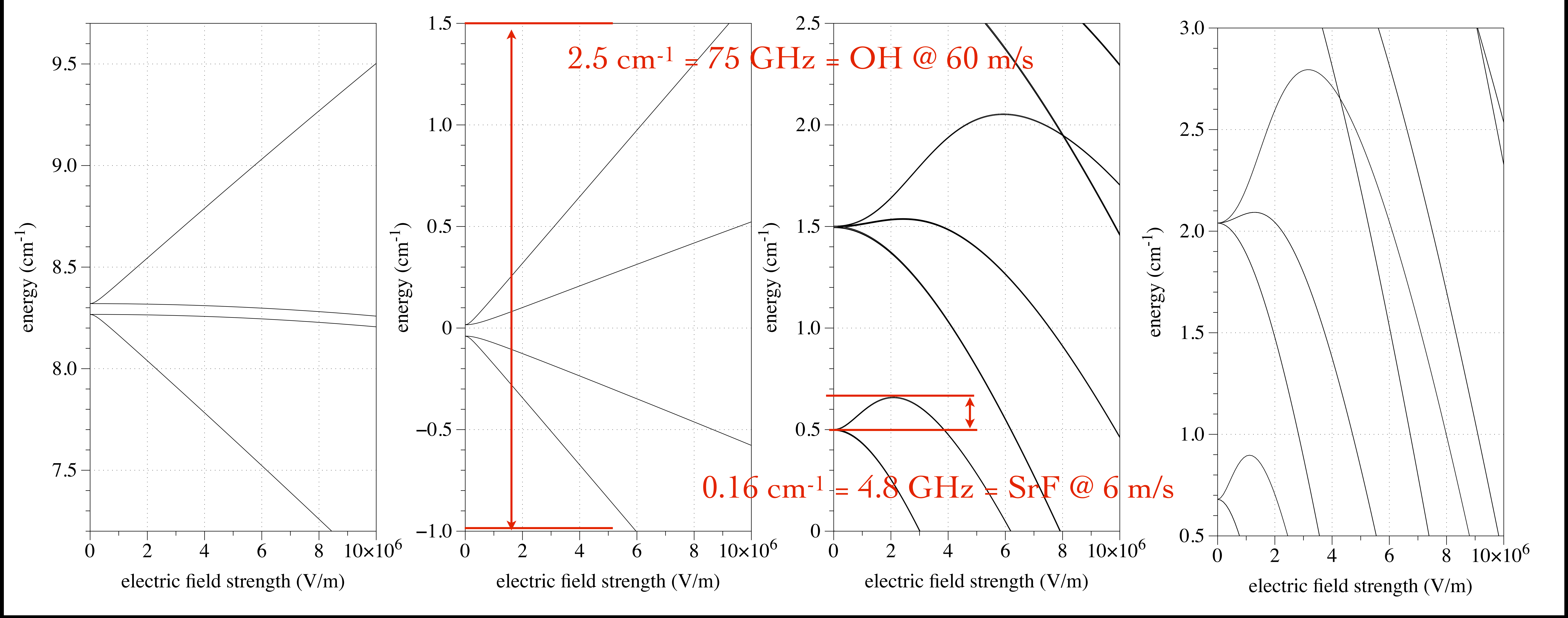
- Capture as many molecules as possible from molecular beam
- Bring average beam velocity from ~190 to ~30 m/s
- Maintain N during deceleration

Traveling-wave decelerator



Challenge: Stark curves of heavy diatomic molecules

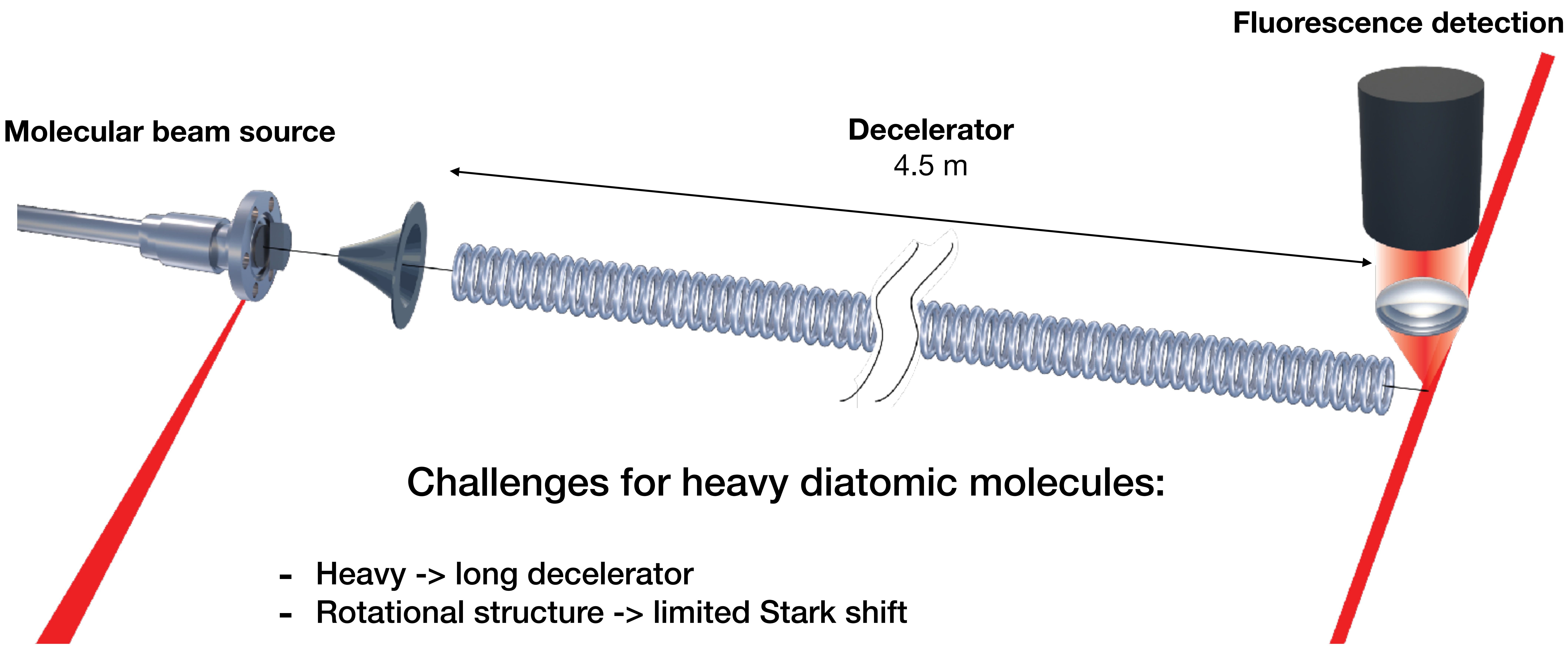
Limited force, because only low fields can be used.
At higher fields, the trajectories in the decelerator become unstable.



ND_3 | OH $^2\Pi_{3/2}$ | SrF $^2\Sigma$ 3.5 Debye | SrO $^1\Sigma$

Limited force: -> a long decelerator

Traveling-wave decelerator



Challenges for heavy diatomic molecules:

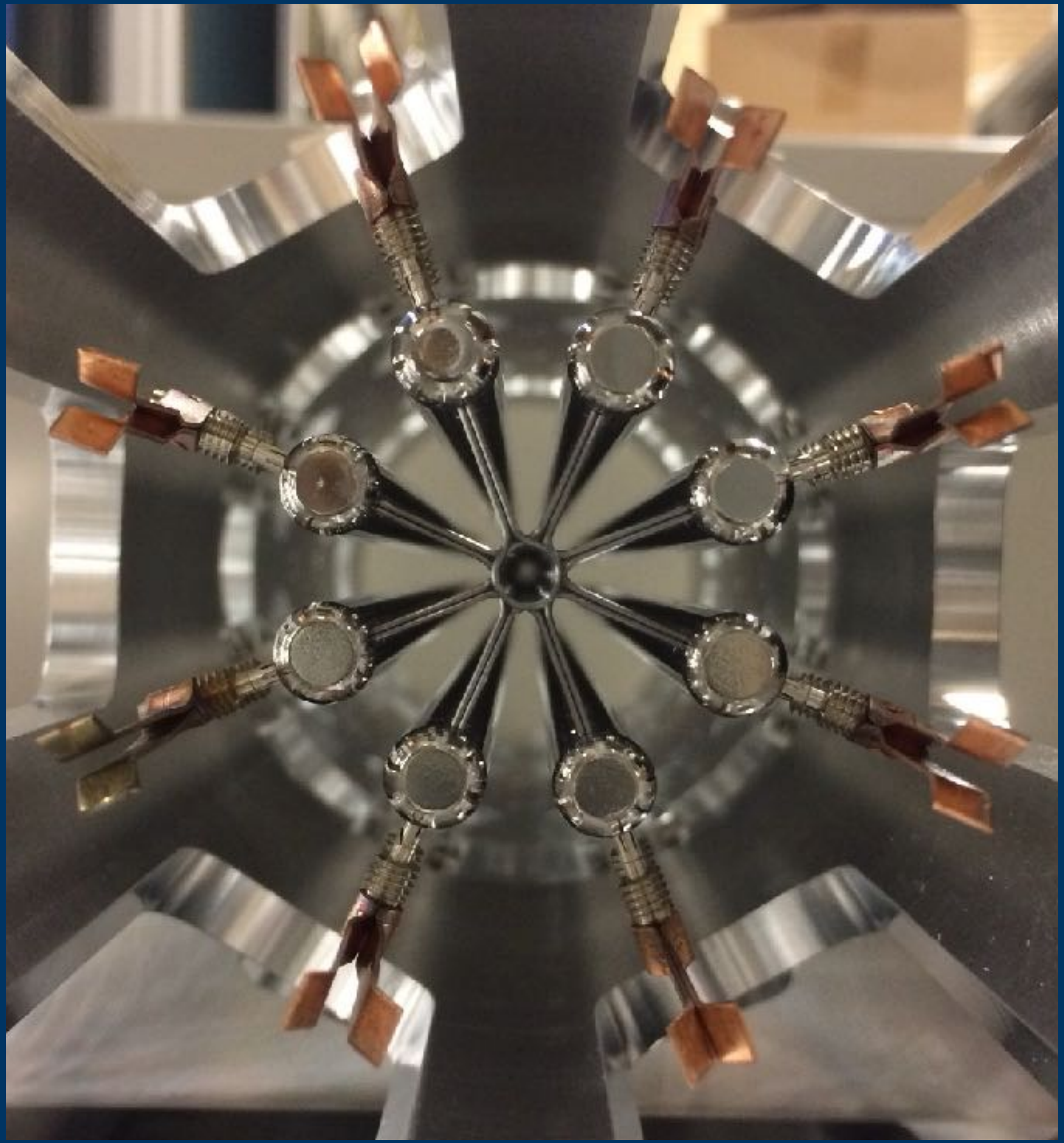
- Heavy -> long decelerator
- Rotational structure -> limited Stark shift

Deceleration, trapping, collision studies, lifetime measurements

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

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

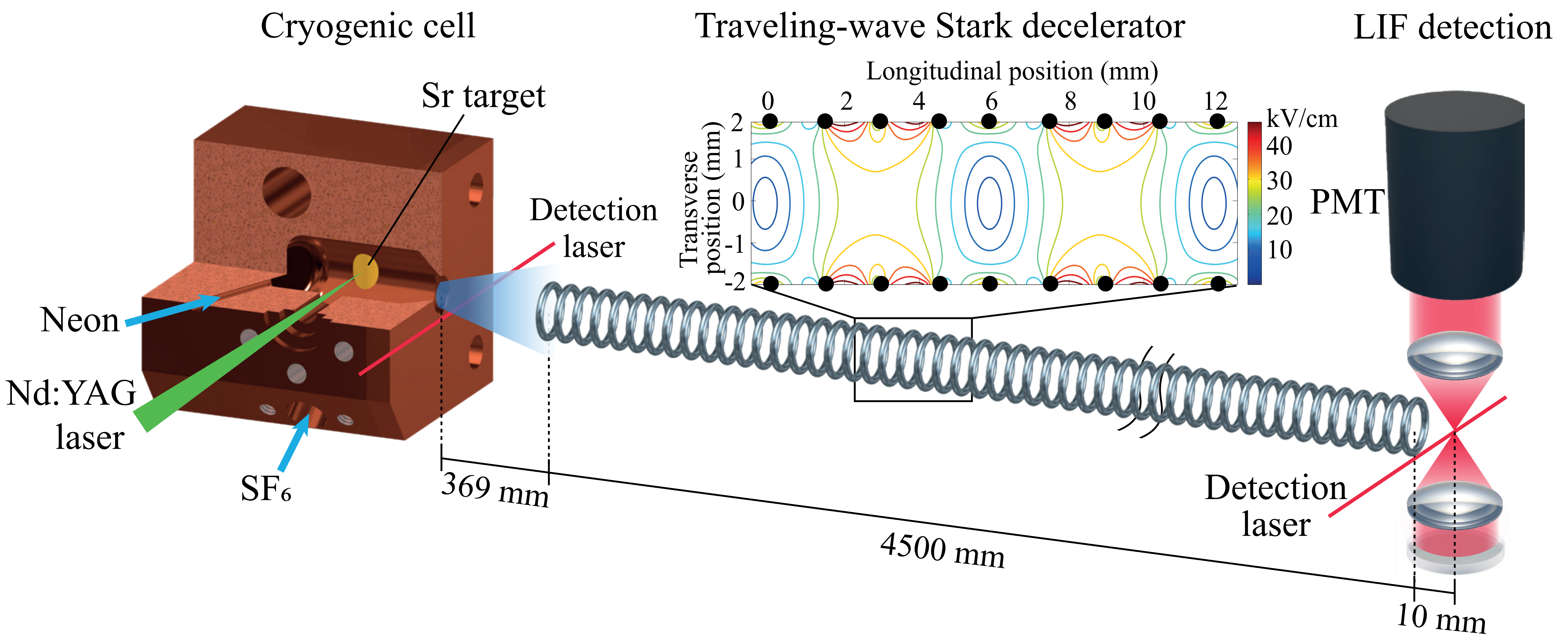
Modular traveling-wave decelerator

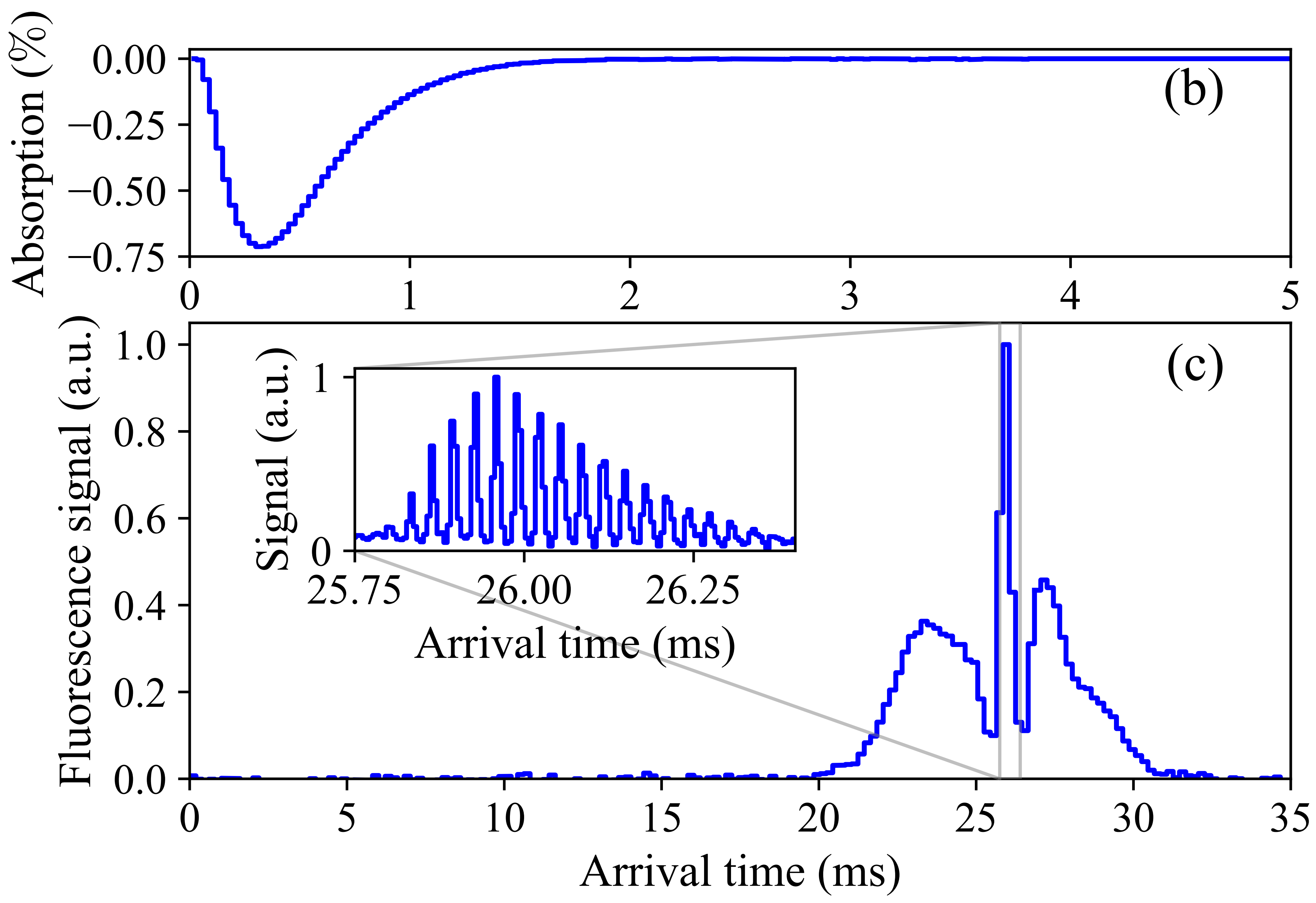


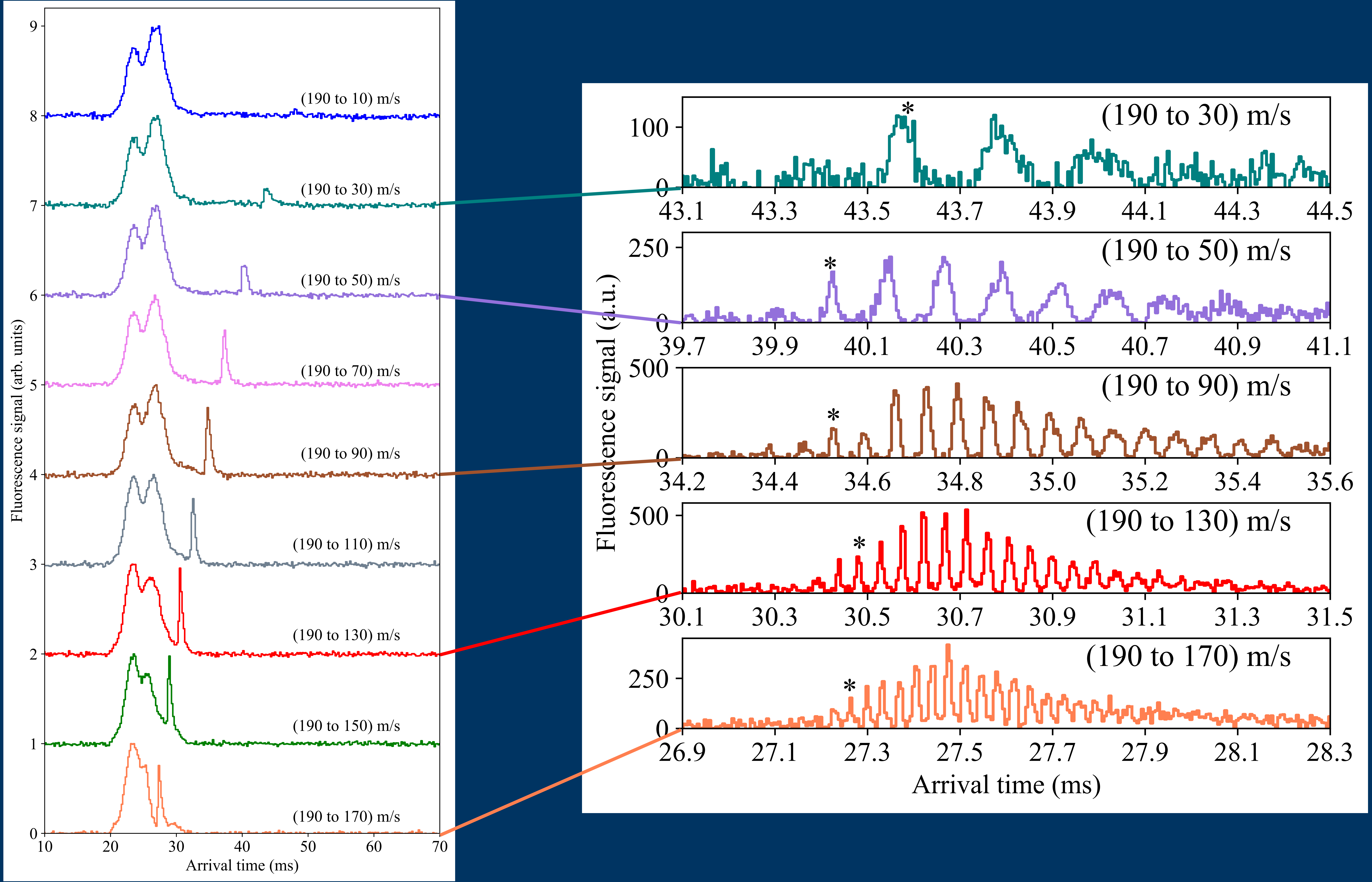
Traveling-wave decelerator

A slow beam of molecules

SrF: First combination of deceleration and cryogenic source





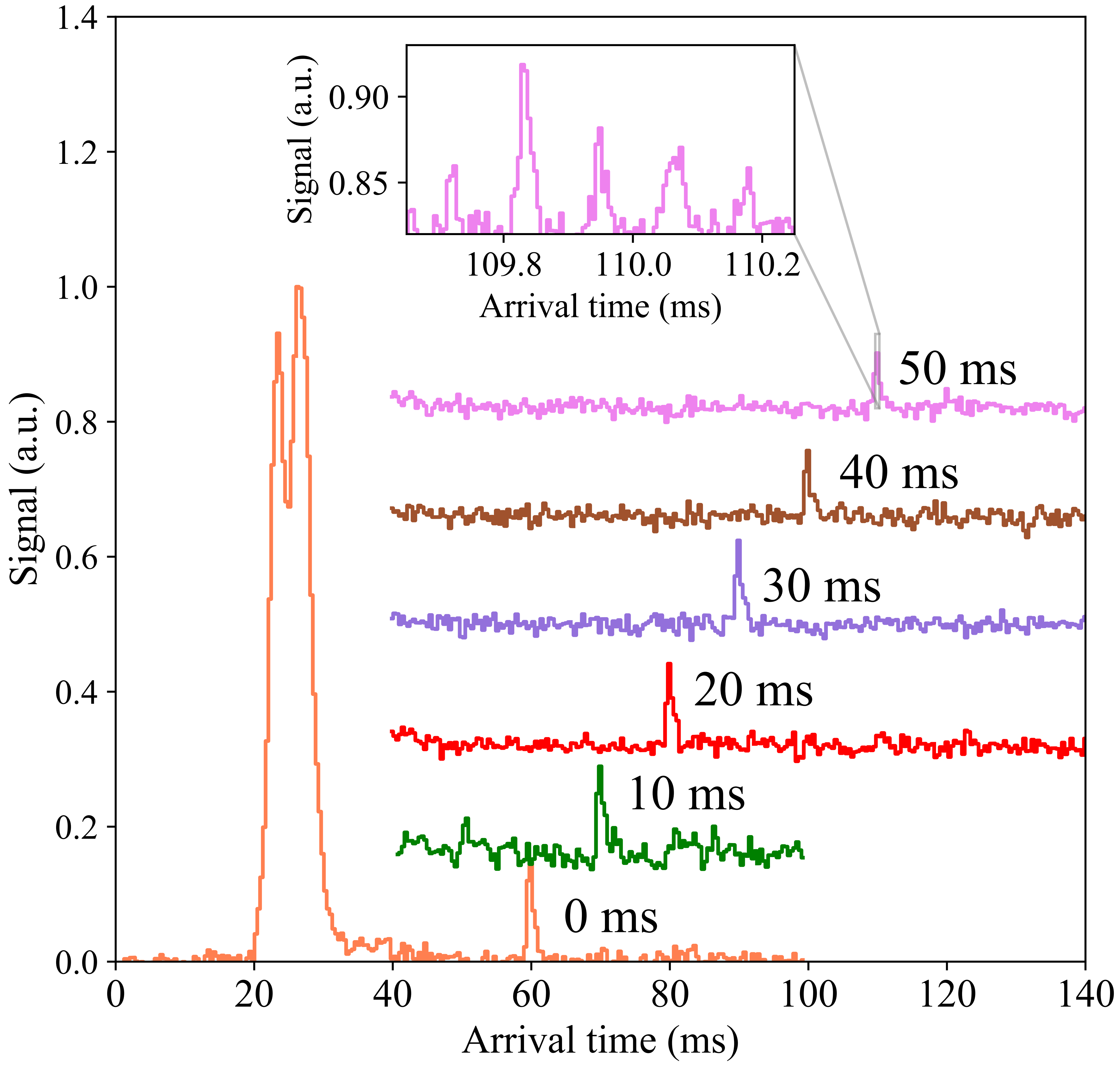


(190 to 10) m/s
(190 to 30) m/s
(190 to 50) m/s
(190 to 70) m/s
(190 to 90) m/s
(190 to 110) m/s
(190 to 130) m/s
(190 to 150) m/s
(190 to 170) m/s
Fluorescence signal (arb. units)
Arrival time (ms)
Fluorescence signal (a.u.)
Arrival time (ms)

A slow beam of molecules

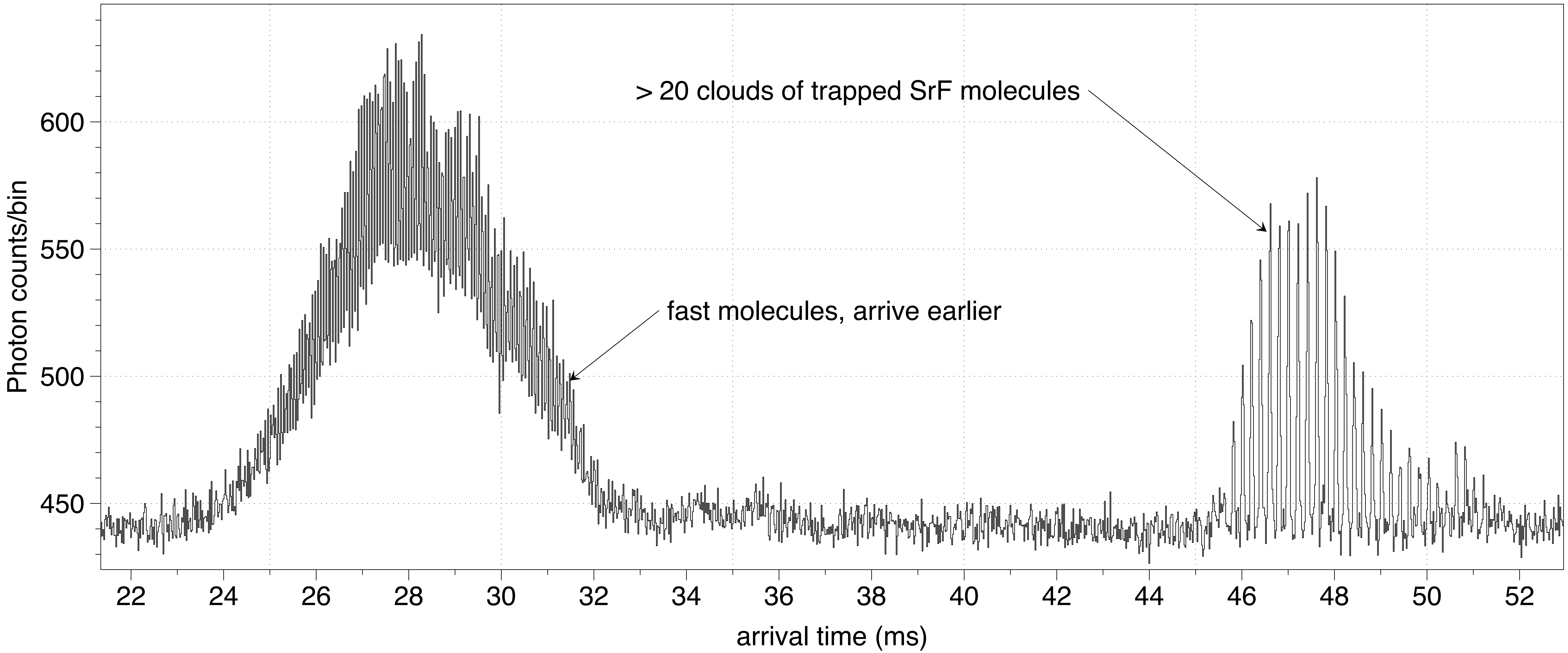
Deceleration to standstill

Deceleration to standstill in 4.2 m, hold there for some time, accelerate out again to 50 m/s to detect



Deceleration and trapping of SrF molecules
Parul Aggarwal, Yanning Yin et al (NL-eEDM),
PRL **127** 173201 (2021)

Stark deceleration



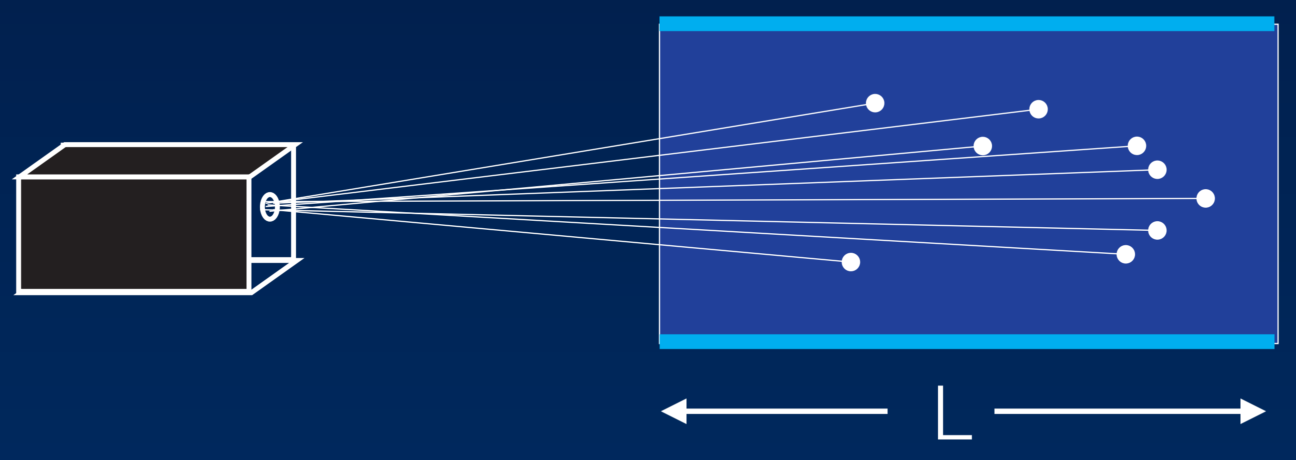
Towards longer coherent interaction times

fast beam

τ ~ 1-2 ms

L ~ 0.5 m

v ~ 250-500 m/s

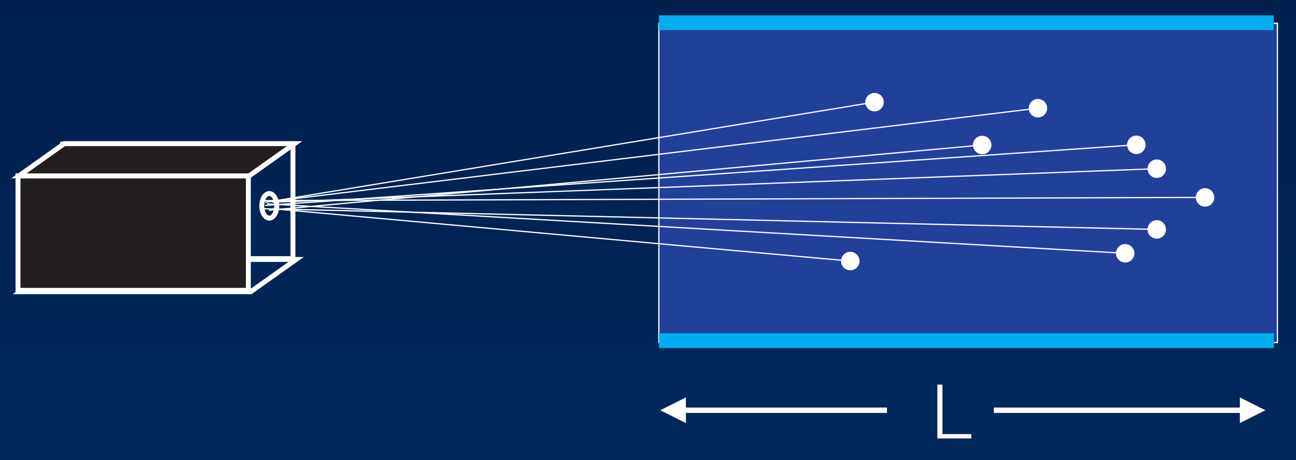


slow beam

τ ~ 15 ms

L ~ 0.5 m

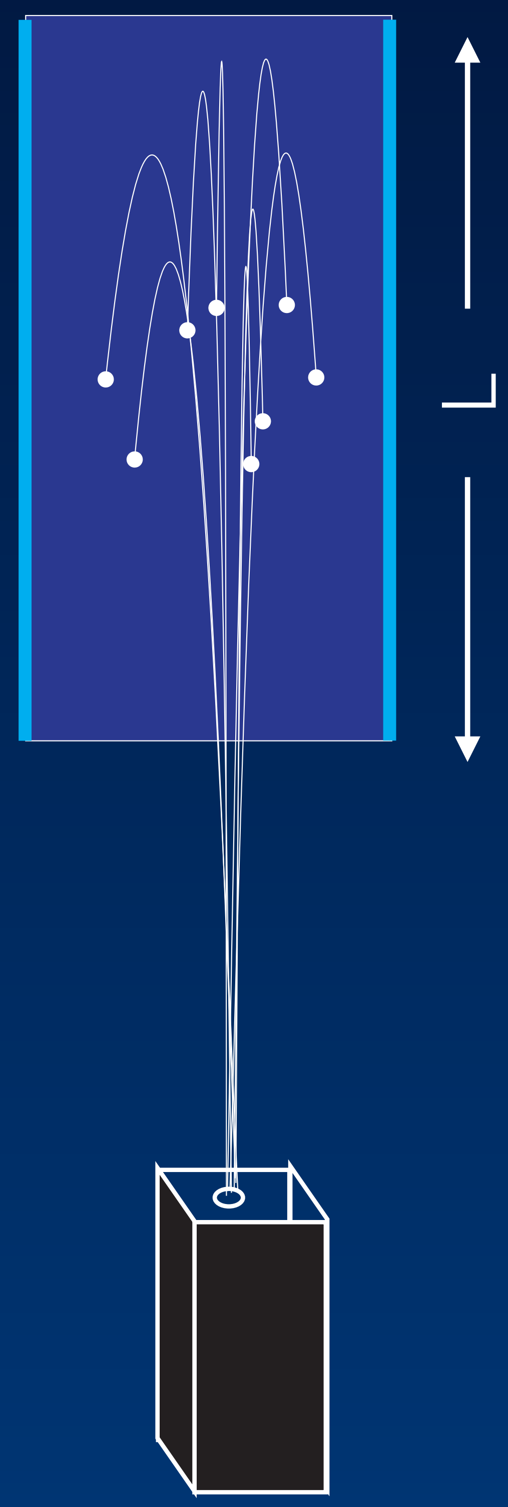
v ~ 30 m/s



fountain

τ ~ 100 ms

L ~ 0.5 m

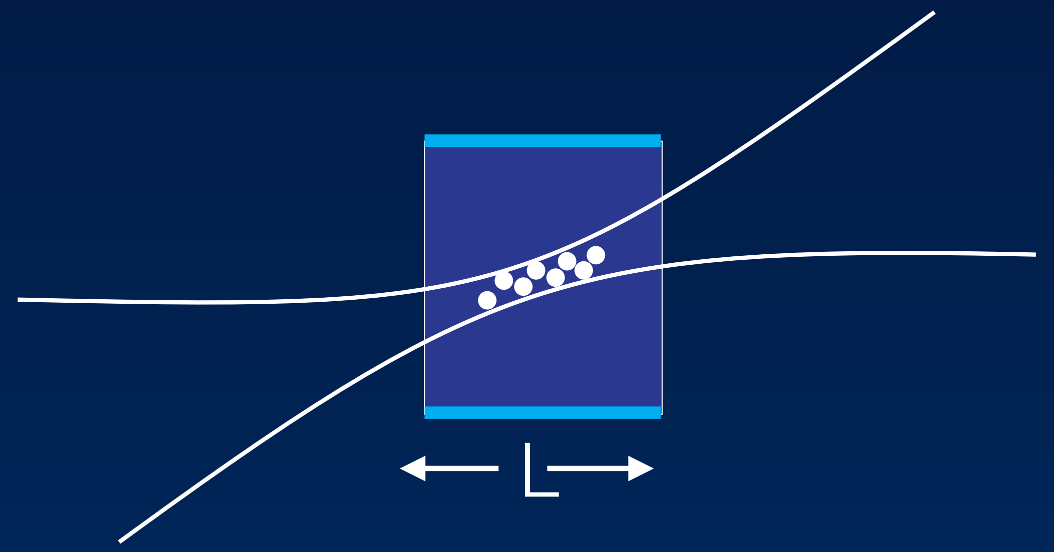


slow vertical beam

trap

τ ~ 1-10 s

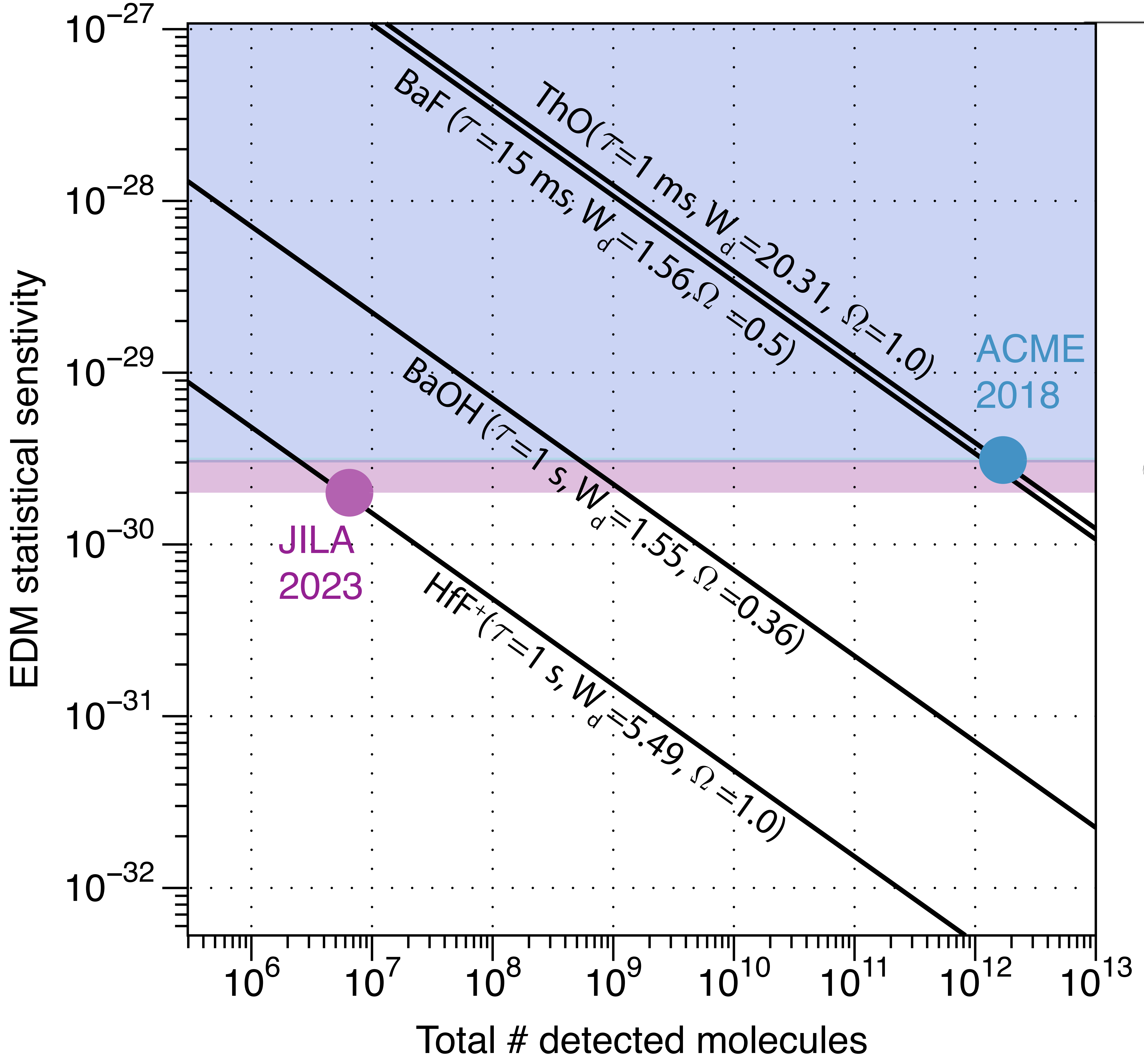
L ~ 0.5 mm



molecules trapped in laser focus

Main challenge:
how to maintain N while increasing t

Strongly connected to choice of molecule!



PHYSICAL REVIEW A **111**, 062815 (2025)

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

Roman Bause,[1,2,*] Nithesh Balasubramanian,[1,2] Ties Fikkers,[1,2] Eifion H. Prinsen,[1,2] Kees Steinebach,[3] Arian Jadbabaie,[4] Nicholas R. Hutzler,[5] I. Agustín Aucar,[1,2,6] Lukáš F. Pašteka,[1,2,7] Anastasia Borschevsky,[1,2] and Steven Hoekstra[1,2,†]

[1]*Van Swinderen Institute for Particle Physics and Gravity, University of Groningen, The Netherlands*
[2]*Nikhef, National Institute for Subatomic Physics, Amsterdam, The Netherlands*
[3]*LaserLaB, Vrije Universiteit Amsterdam, The Netherlands*
[4]*Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA*
[5]*Division of Physics, Mathematics, and Astronomy, California Institute of Technology, Pasadena, California 91125, USA*
[6]*Instituto de Modelado e Innovación Tecnológica (UNNE-CONICET), Facultad de Ciencias Exactas y Naturales y Agrimensura, Universidad Nacional del Nordeste, Corrientes, Argentina*
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(Received 1 November 2024; accepted 3 June 2025; published 17 June 2025)

We present the conceptual design of an experiment to measure the electron's electric dipole moment (eEDM) using ^{138}BaOH molecules in an optical lattice. The BaOH molecule is laser-coolable and highly sensitive to the eEDM, making it an attractive candidate for such a precision measurement, and capturing it in an optical lattice offers potentially very long coherence times. We study possibilities and limitations of this approach, identify the most crucial limiting factors and ways to overcome them. The proposed apparatus can reach a statistical error of 10^{-30} e cm by measuring spin precession on a total number of 5×10^9 molecules over a span of 120 days.

DOI: 10.1103/8ltl-7wsb

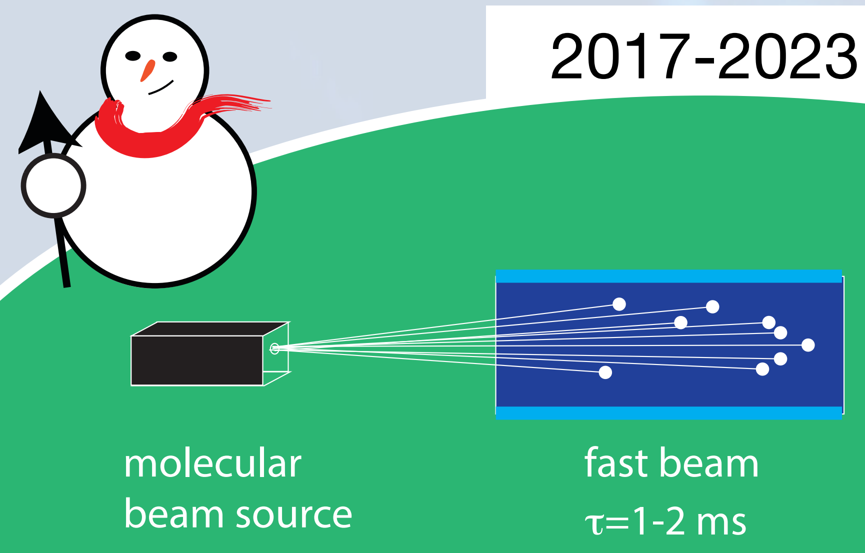
Table-top precision tests

New experimental approaches to study fundamental physics

NL-eEDM: Hendrick Bethlem, Anastasia Borschevsky, Steven Hoekstra, Steven Jones, Rob Timmermans, Wim Ubachs, Jordy de Vries, Lorenz Willmann

Measure the electron's electric dipole moment using cold molecules

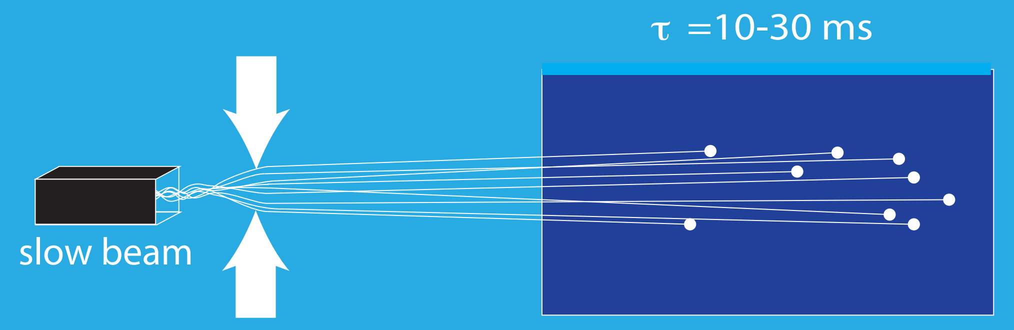
2017-2023



Phase 1
Fast beam of BaF molecules

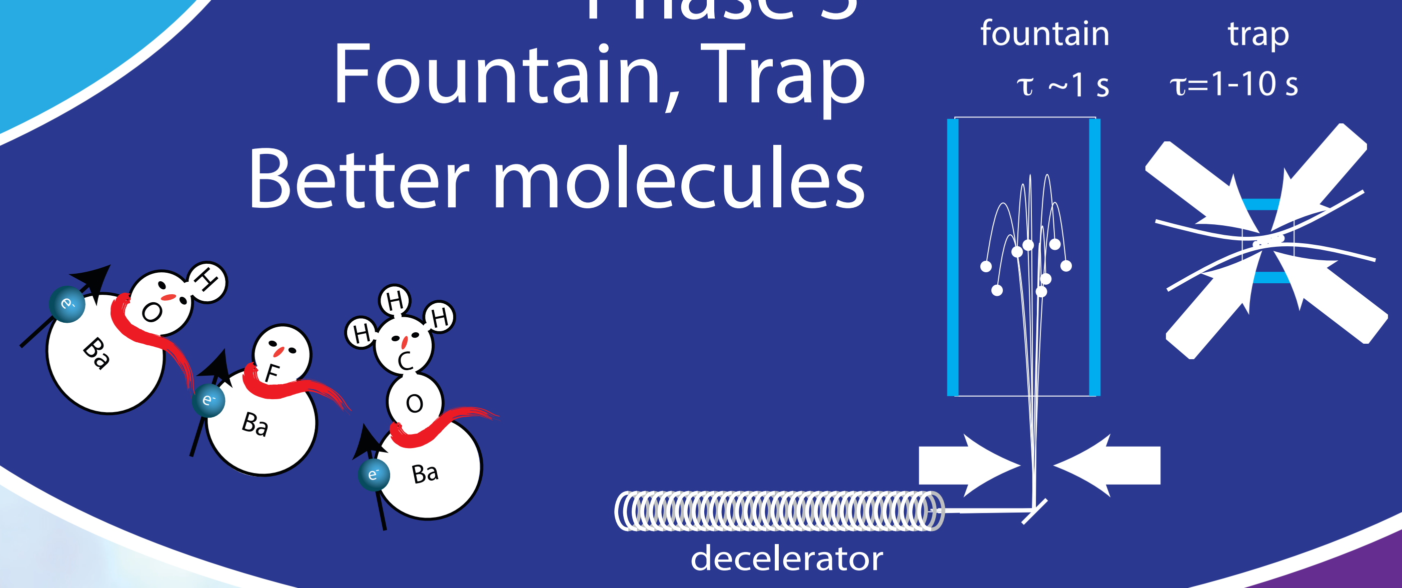
2023-2028

Phase 2
Slow and cold molecular beam



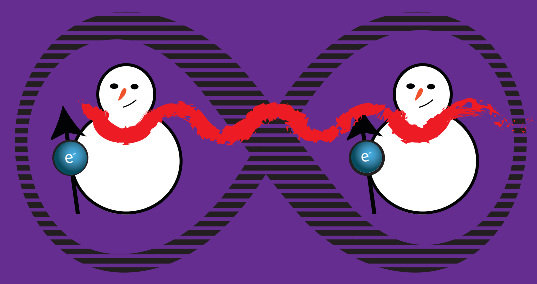
2023-2028
ENW-XL, M2 and VICI

Phase 3
Fountain, Trap
Better molecules



2025-2035

Phase 4
Beyond the quantum limit



Minimize statistical uncertainty

$$S \propto \frac{1}{E\tau\sqrt{N}}$$

Heavy polar molecules give a high sensitivity to new physics

Slow and cold beams provide a long interaction time

Using entanglement enables linear scaling with the number of molecules

Demonstrate an all-optical superposition state creation

Slow beams push magnetic field senstivity to the limit

Polyatomic molecules provide co-magnetometer states

Theory framework: connect eEDM experiments to high-energy particle physics

Control systematic effects