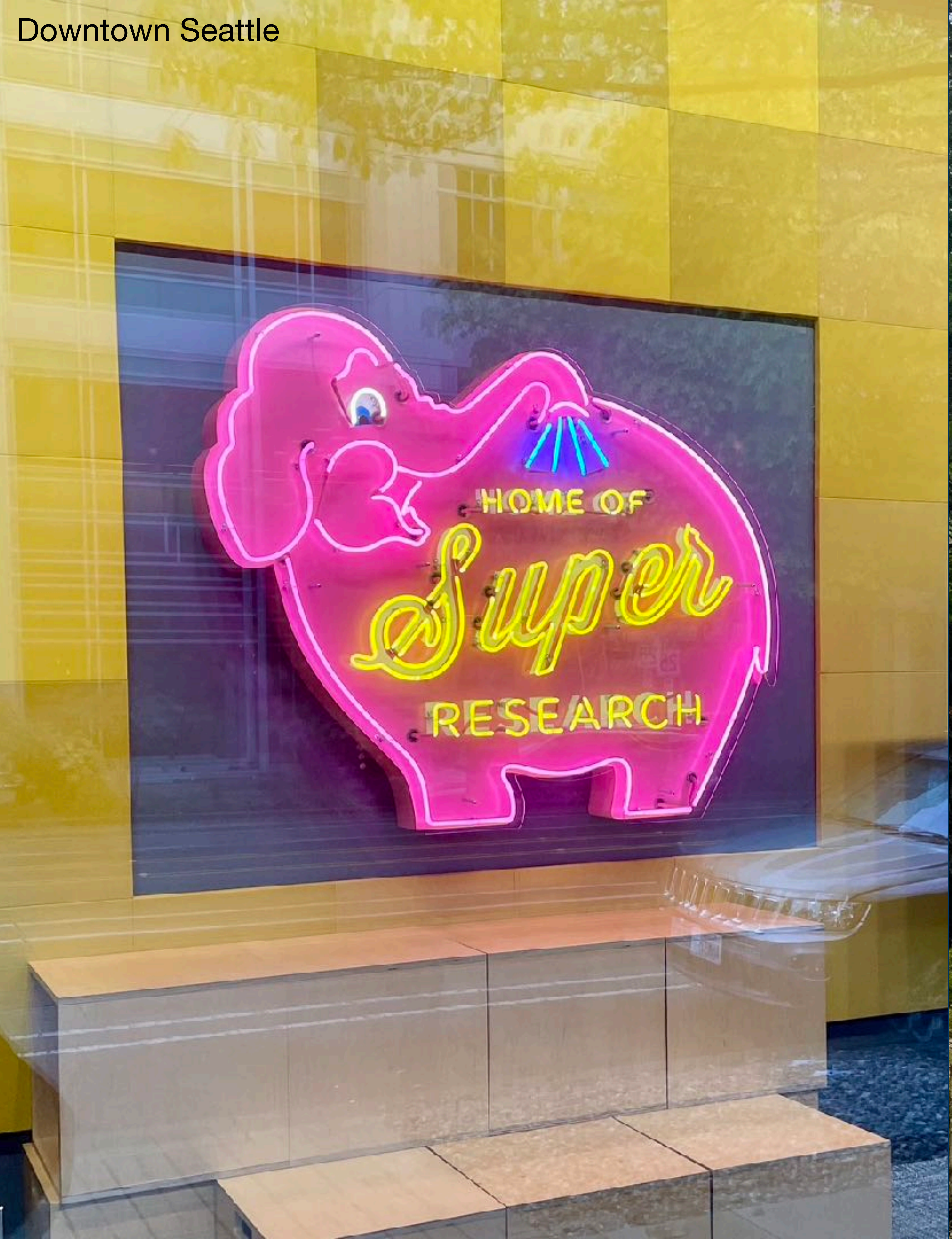
Downtown Seattle



Tongue point, Olympic peninsula

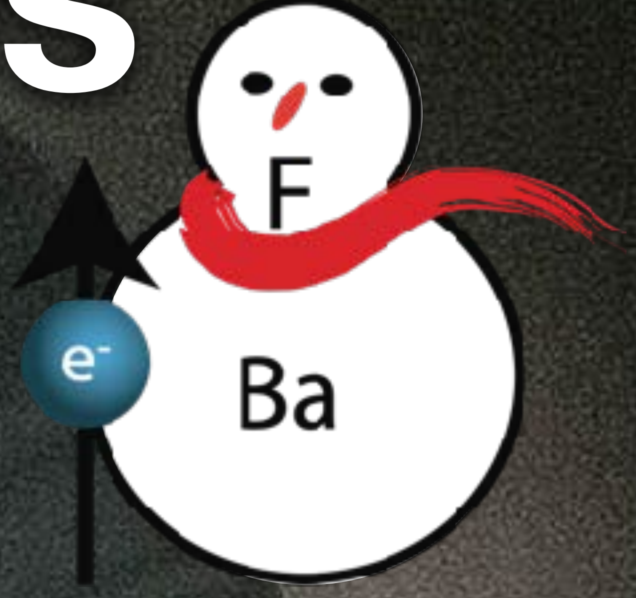
Slow beams and trapped molecules

to probe the electron's EDM

Steven Hoekstra, University of Groningen and Nikhef, The Netherlands

Slow beams and trapped molecules

to probe the electron's EDM



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

Steven Hoekstra, University of Groningen and Nikhef, The Netherlands

Increasing the eEDM sensitivity

Measure shift of molecular energy level that correlates with electric field direction reversal

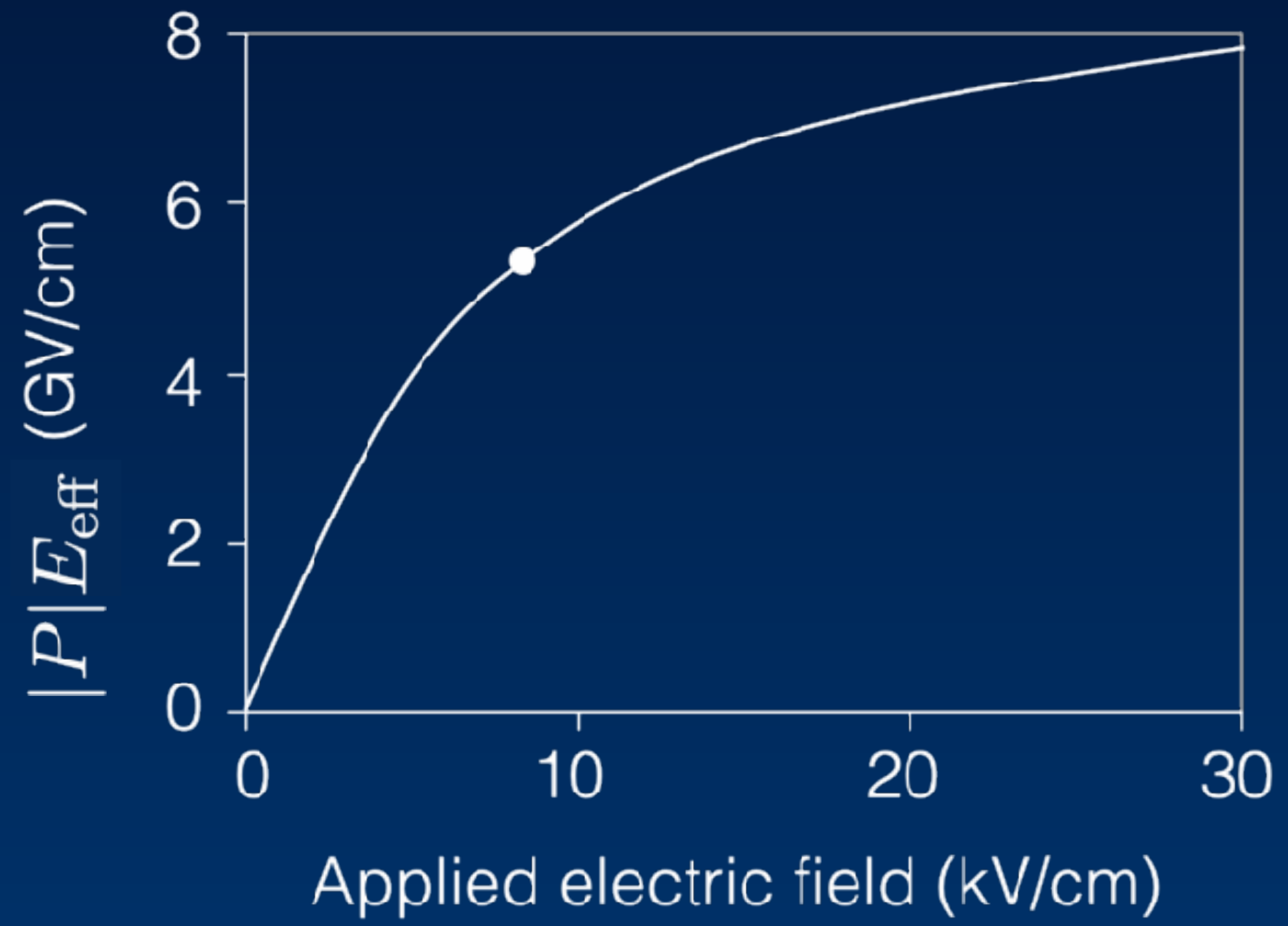
statistical error: $$\sigma_d = \frac{\hbar}{e} \frac{1}{2|P|E_{\mathrm{eff}}\tau\sqrt{\dot{N}T}}$$

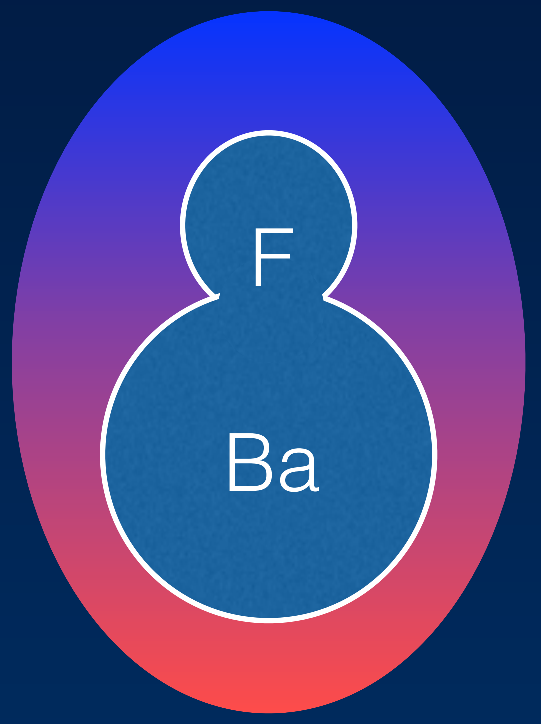
Cold Molecules

Number of detected molecules

Coherent interaction time

Effective electric field

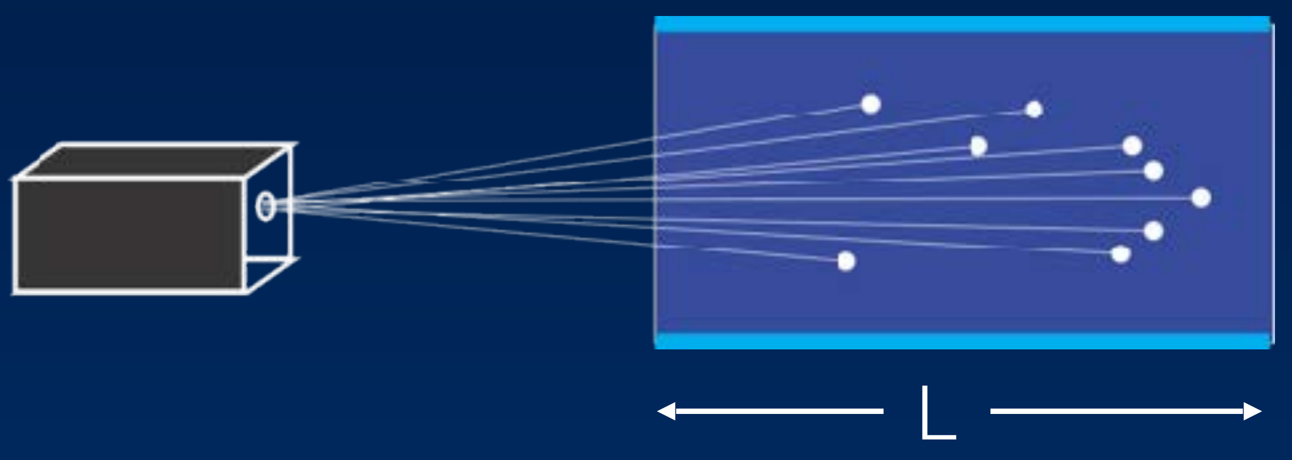




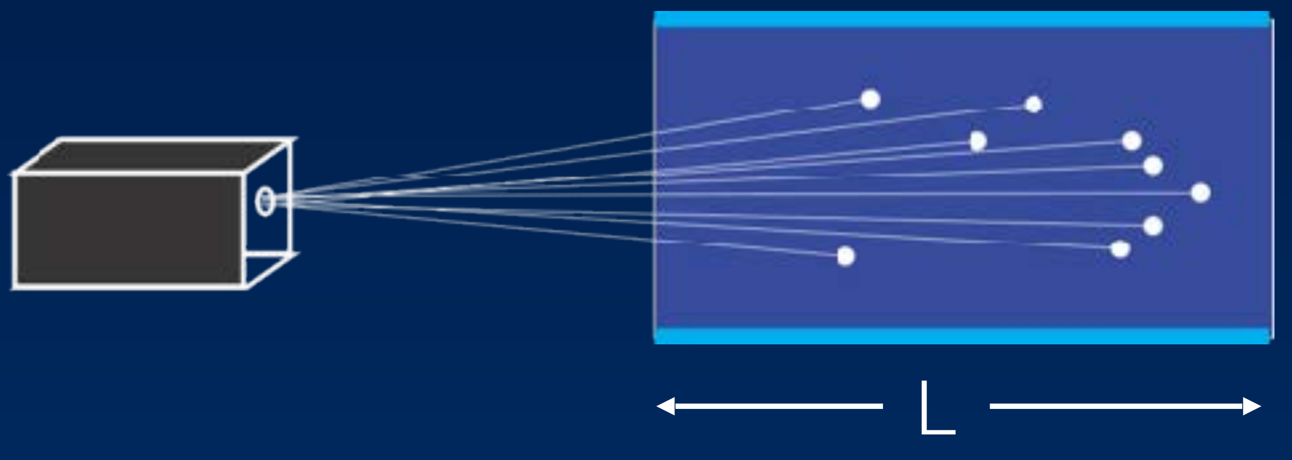
polar molecule

Towards longer coherent interaction times

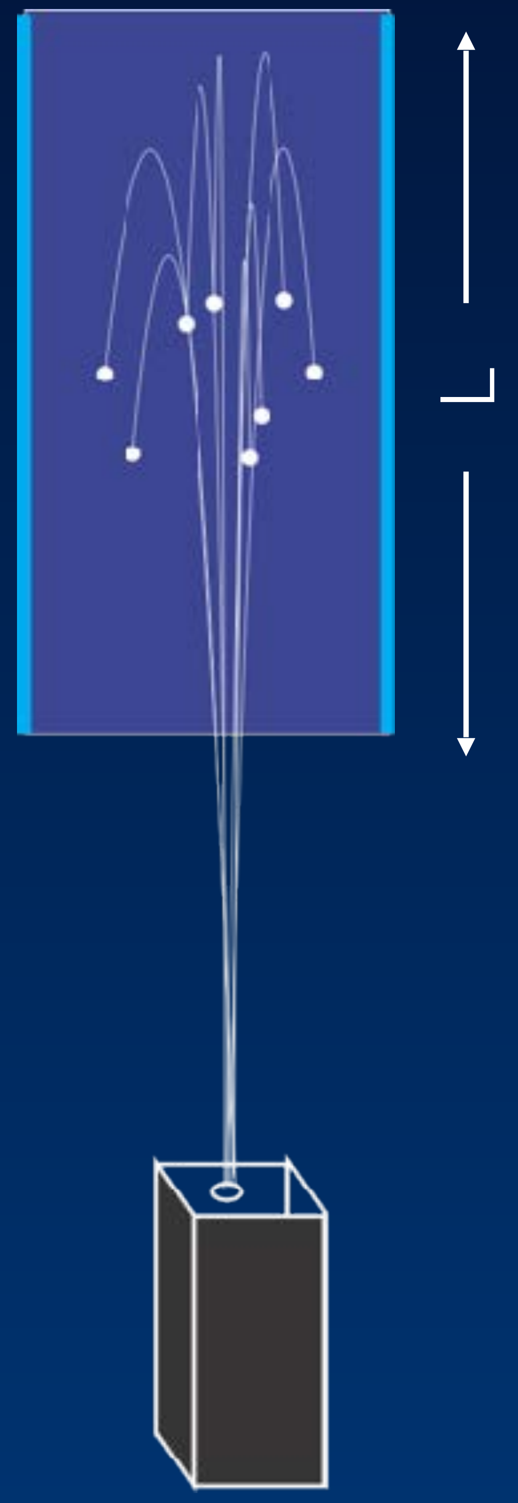
fast beam



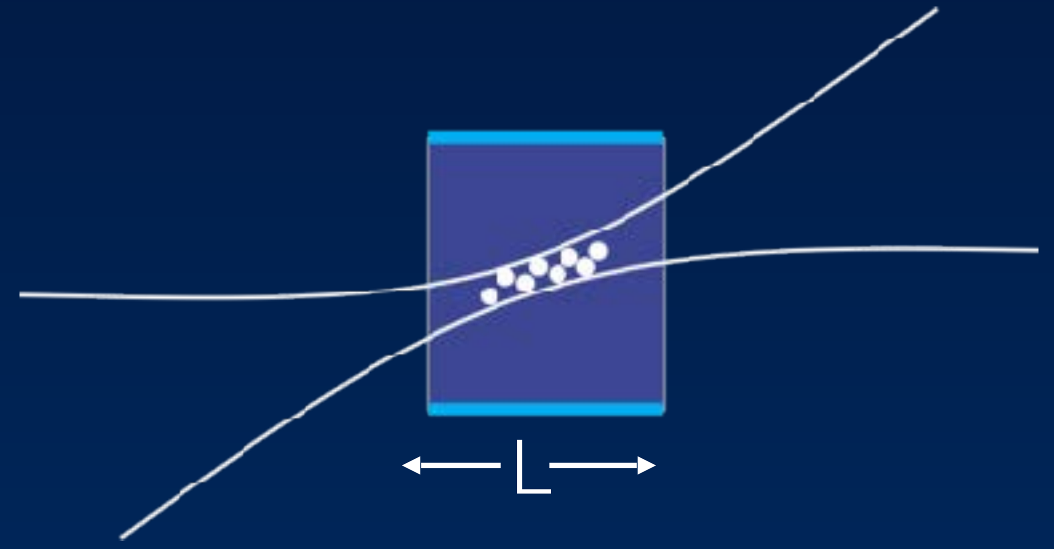
slow beam



fountain



trap



Main challenge:
how to maintain N while increasing t

Strongly connected to choice of molecule!

Longer interaction times
Lower temperatures
Fewer molecules
1000 K
10^10-10^12
ablation
1000 K
10^6-10^12
ion beam
1. Molecular beams
YbF
400-600 m/s
1 K
1 ms
~10^6
supersonic beam
4. Neutralisation and capture
ThO
150-200 m/s
5-20 K
3 ms
~10^8
cryogenic beam
cold matrix
BaF
2. Deceleration and cooling
0-30 m/s
30 ms
~10^6
decelerated beam single state
BaF
0.1-1 mK
10^2-10^5
MOT
1-100 mK
0.1-10 s
10^2-10^5
electric/magnetic trap
1-10 μK
1-10 s
10^2-10^4
optical trap
3. Further cooling and trapping
1-10 s
10^2-10^4
ion trap
HfF+
Ra, Cs, Yb, YbX, RaX, BaX, …

Comparing different approaches

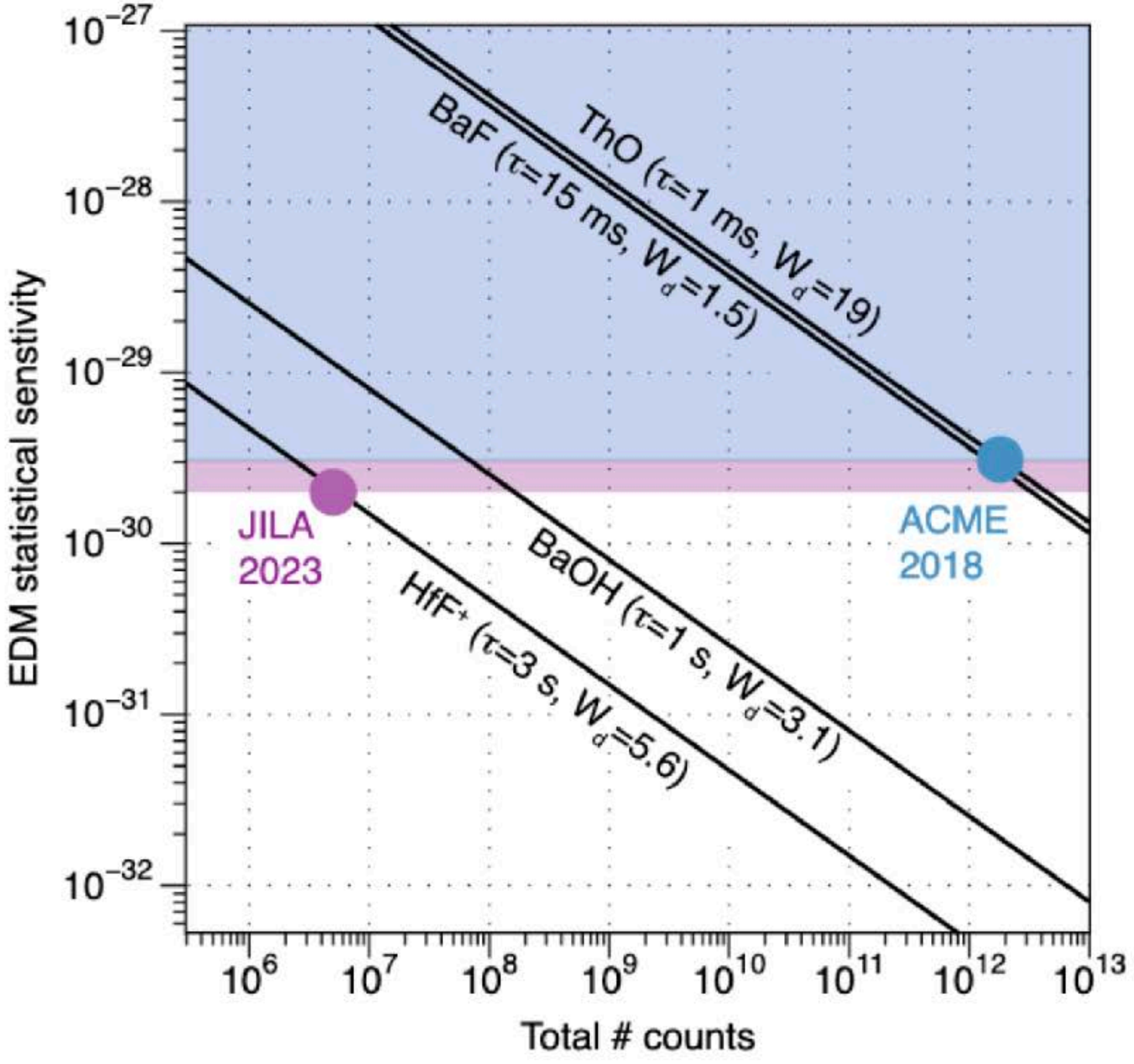


FIG. 1. Overview of statistical sensitivity of different experimental platforms. Each line corresponds to a combination of τ and $|P| \cdot W_d$ [in units of 10^{24} h Hz/(e cm)] which is typical for a given molecule species. The dots represent the two most recent experimental results [6, 7], taking into account only statistical uncertainty. The blue and purple shaded regions are excluded by the ThO and HfF^+ experiments, respectively. It can be seen that our target sensitivity of 10^{-30} e cm can be reached with $N = 6 \times 10^8$ BaOH molecules at the shot noise limit.

Roman Bause et al, manuscript almost completed :-)

Building up towards the ultimate experiment:

university of groningen
van swinderen institute for particle physics and gravity

Nikhef
Dutch National Institute for (astro)Particle Physics

VU VRIJE UNIVERSITEIT AMSTERDAM

UvA

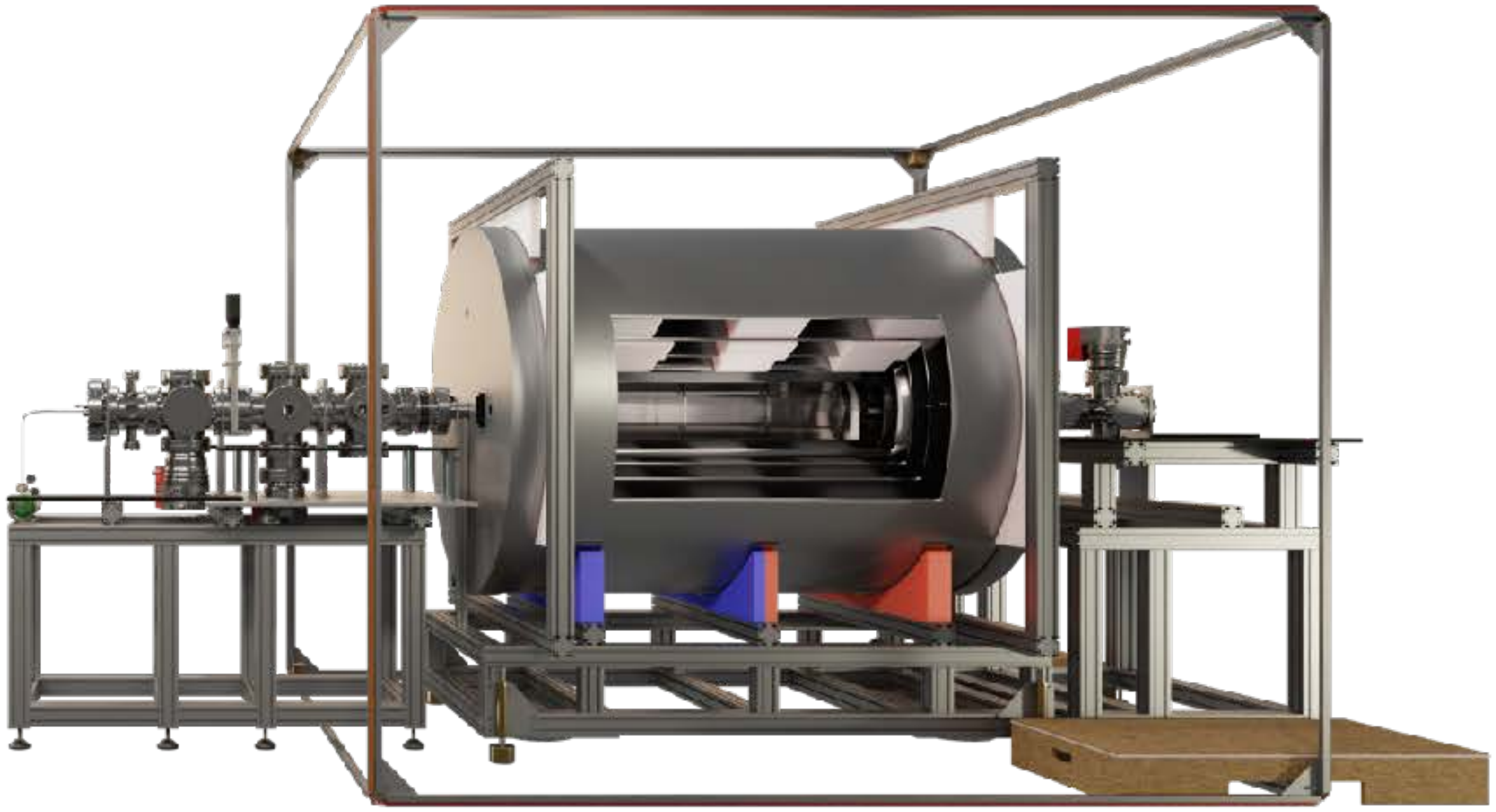
Phase 1: Fast beam

Supersonic beam (600 m/s)

Controlled field environment

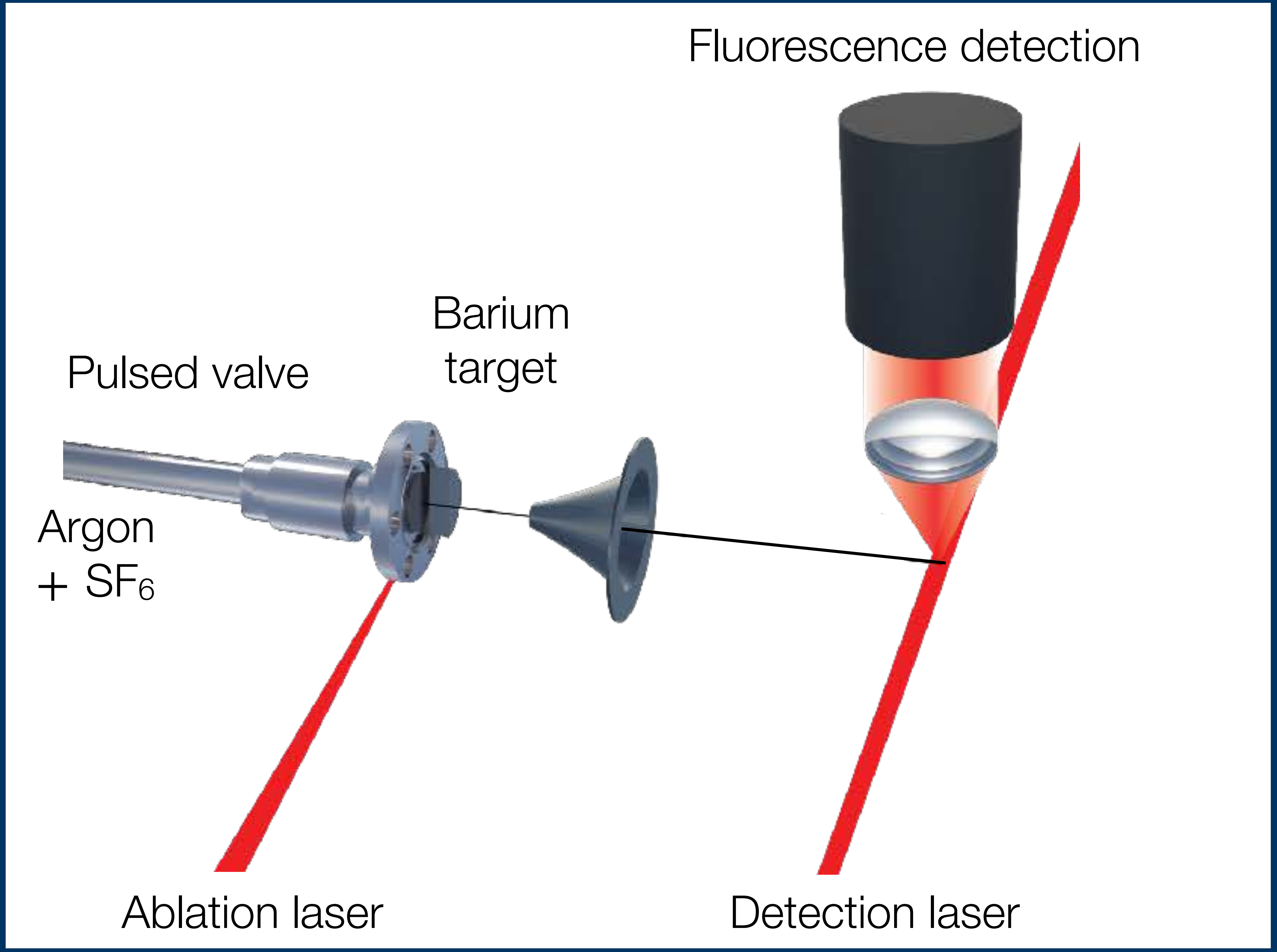
Explore molecular structure

Spin interferometer measurement



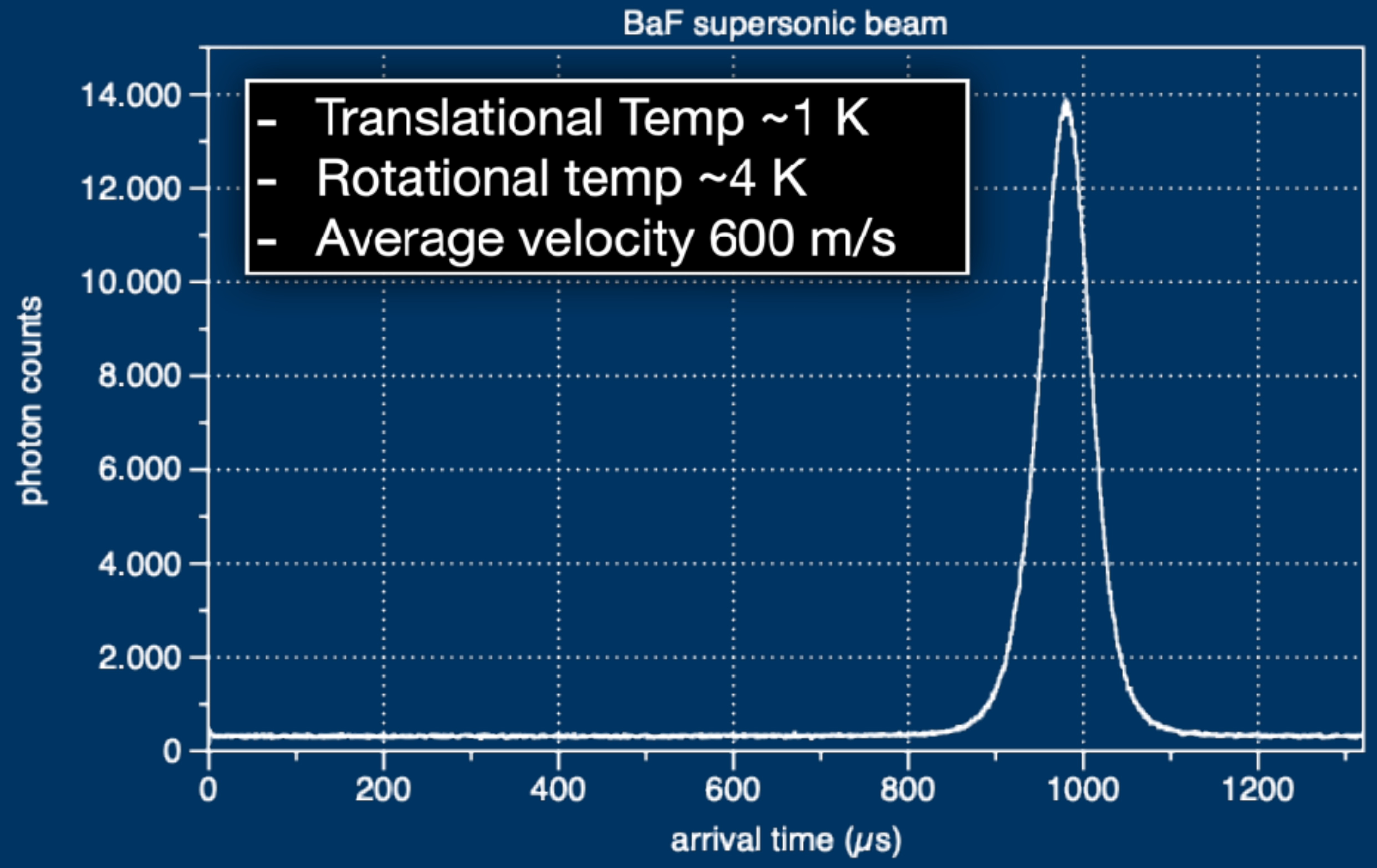
Understand systematics

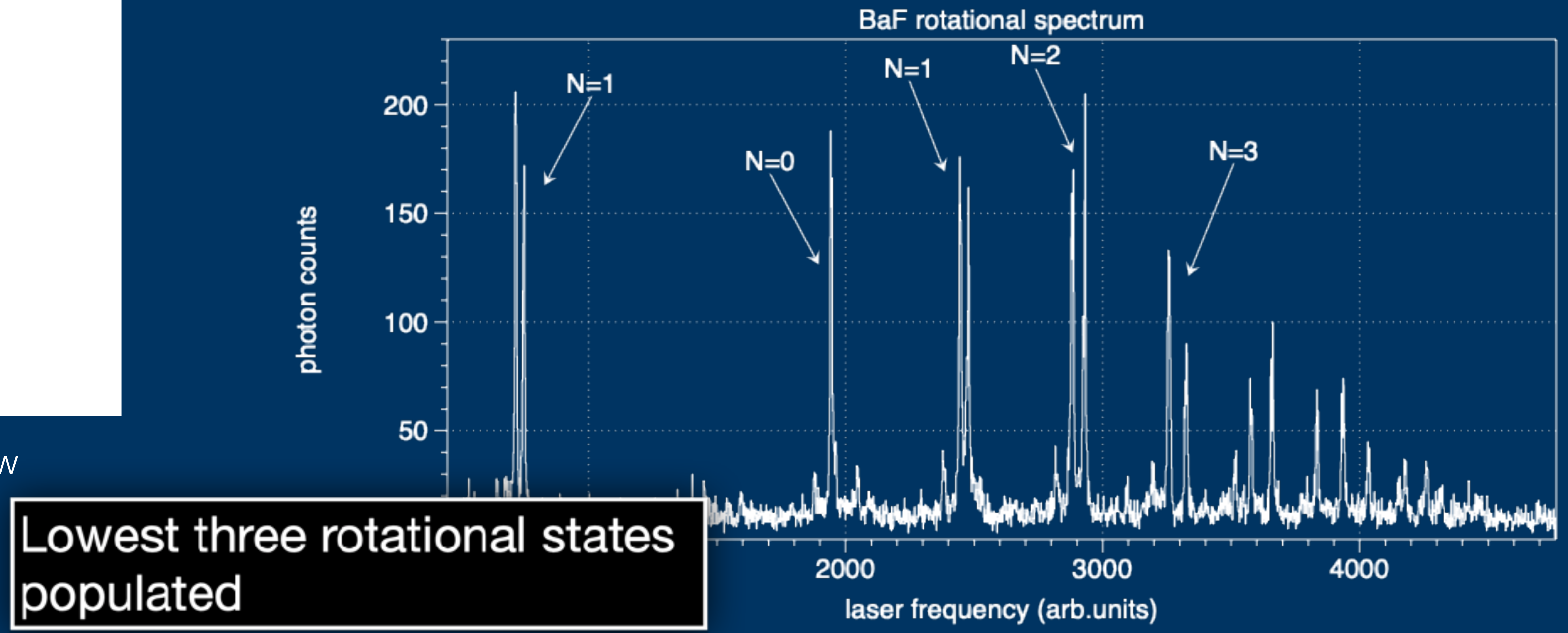
Supersonic beams of SrF and BaF molecules



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).

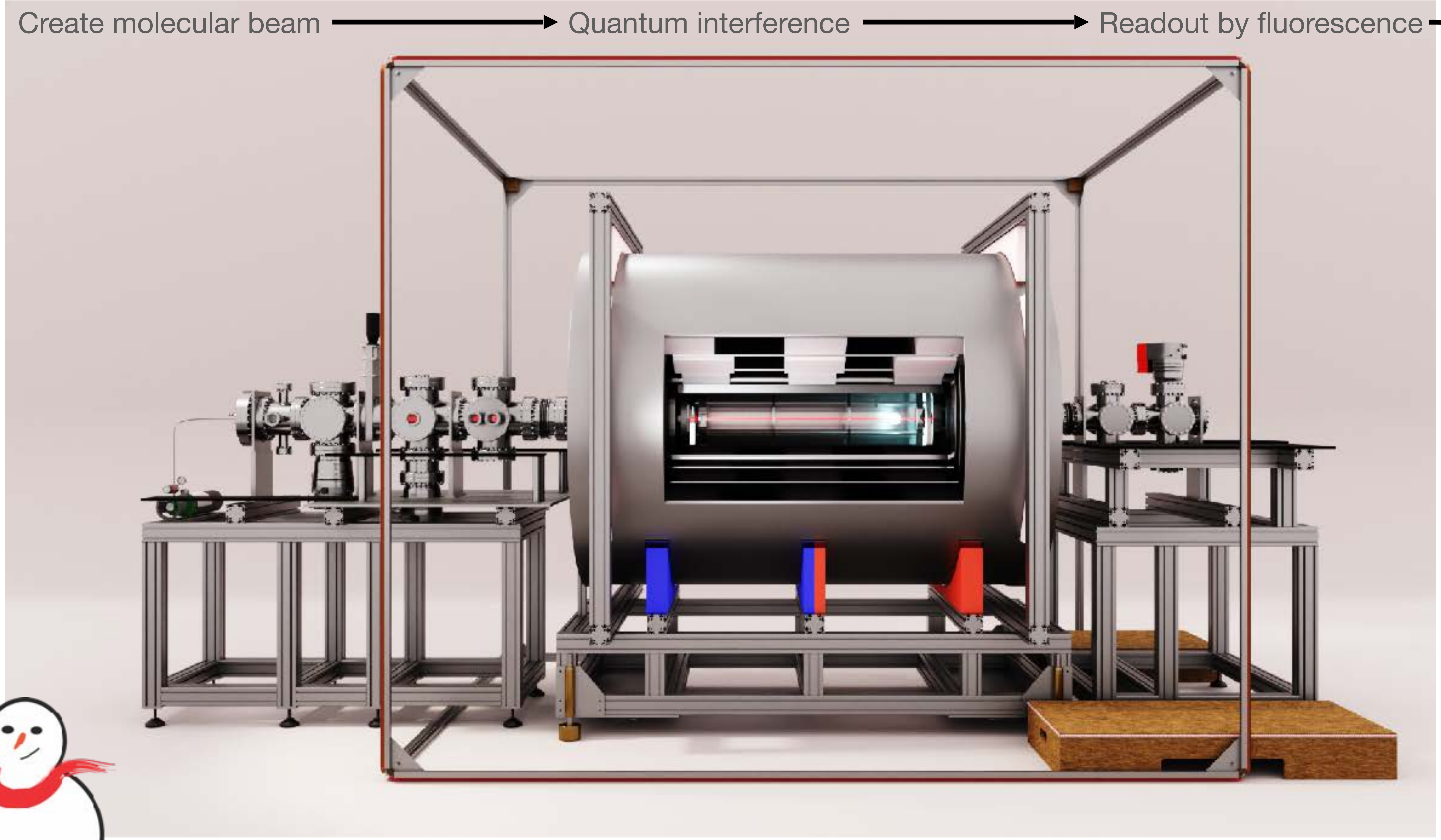




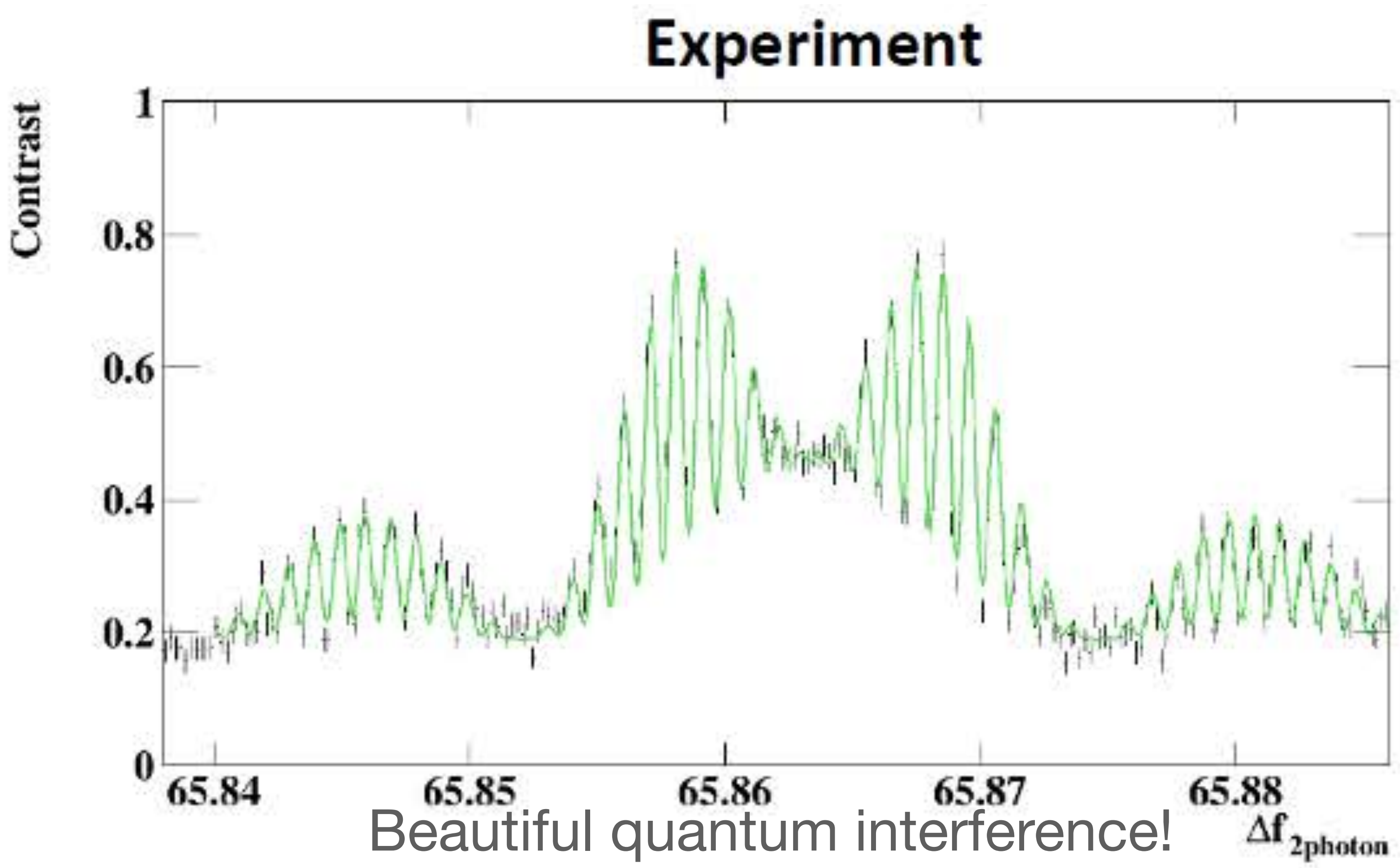
How to read out small energy shifts: spin interferometer

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)



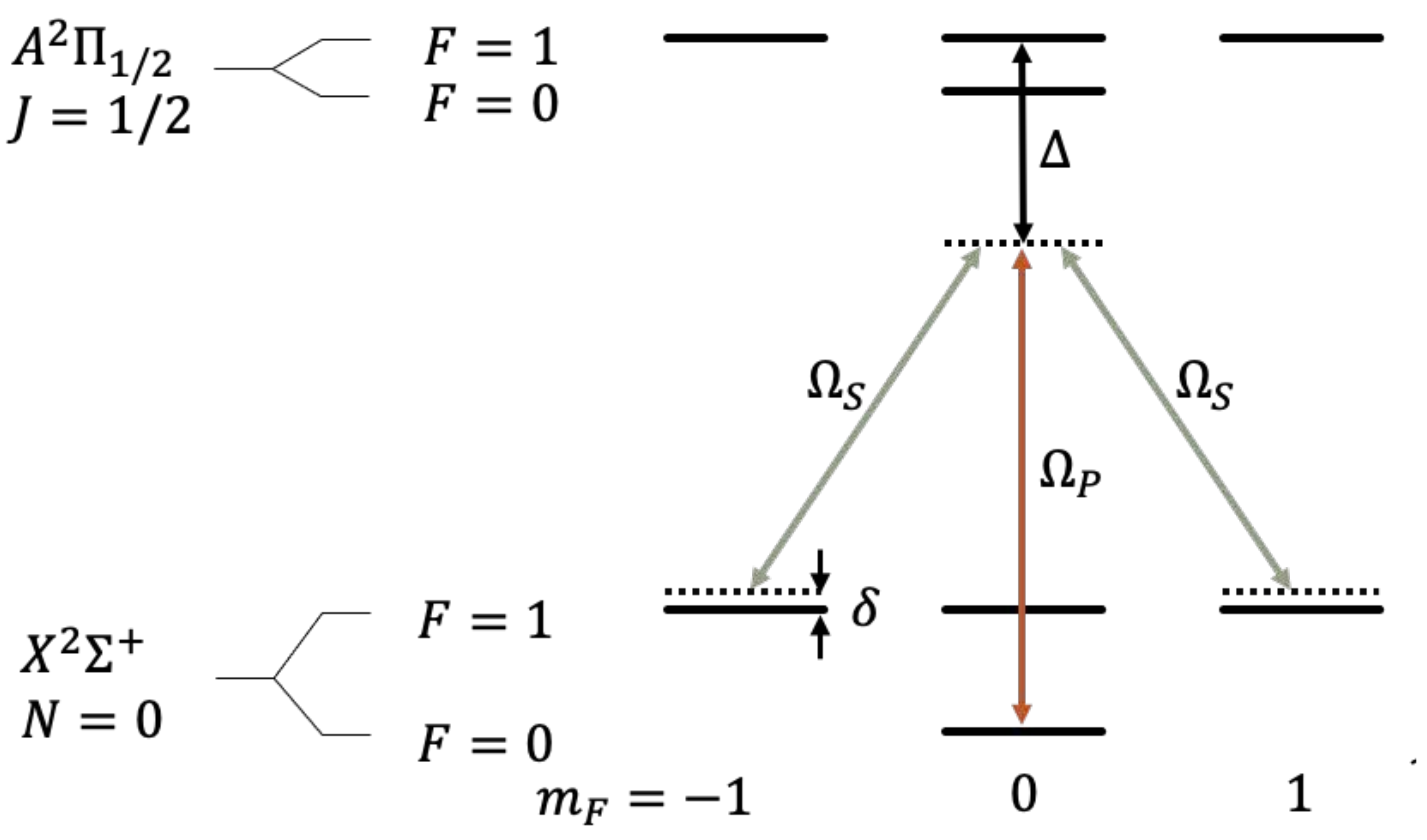
Beautiful quantum interference!

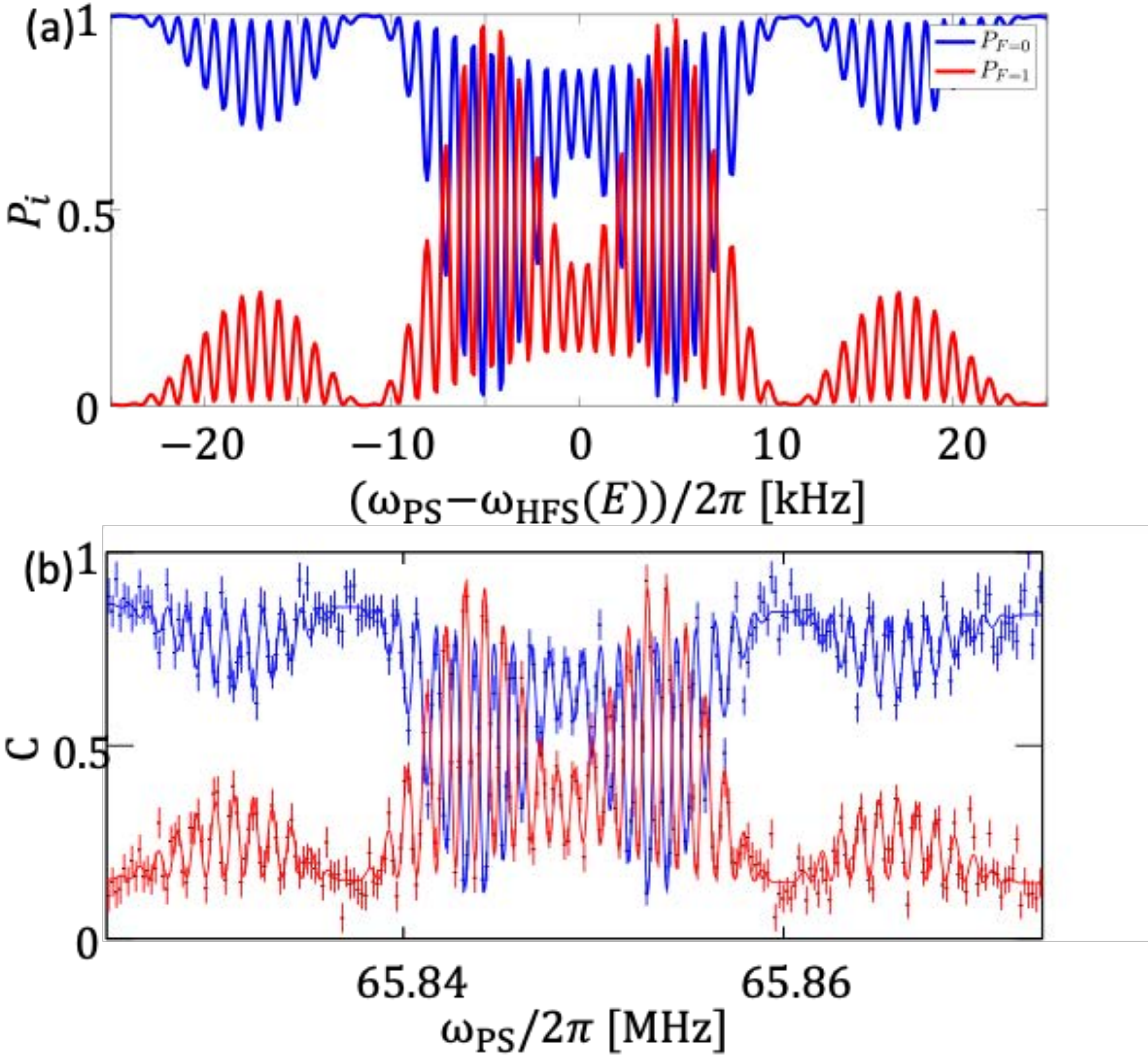
Contains all relevant experimental parameters
Crucial for reduction of systematic effects
(A.Boeschoten et al, NL-eEDM collaboration, arXiv:2303.06402v1)

NL-eEDM

Experiment and theory

Optical Bloch equations

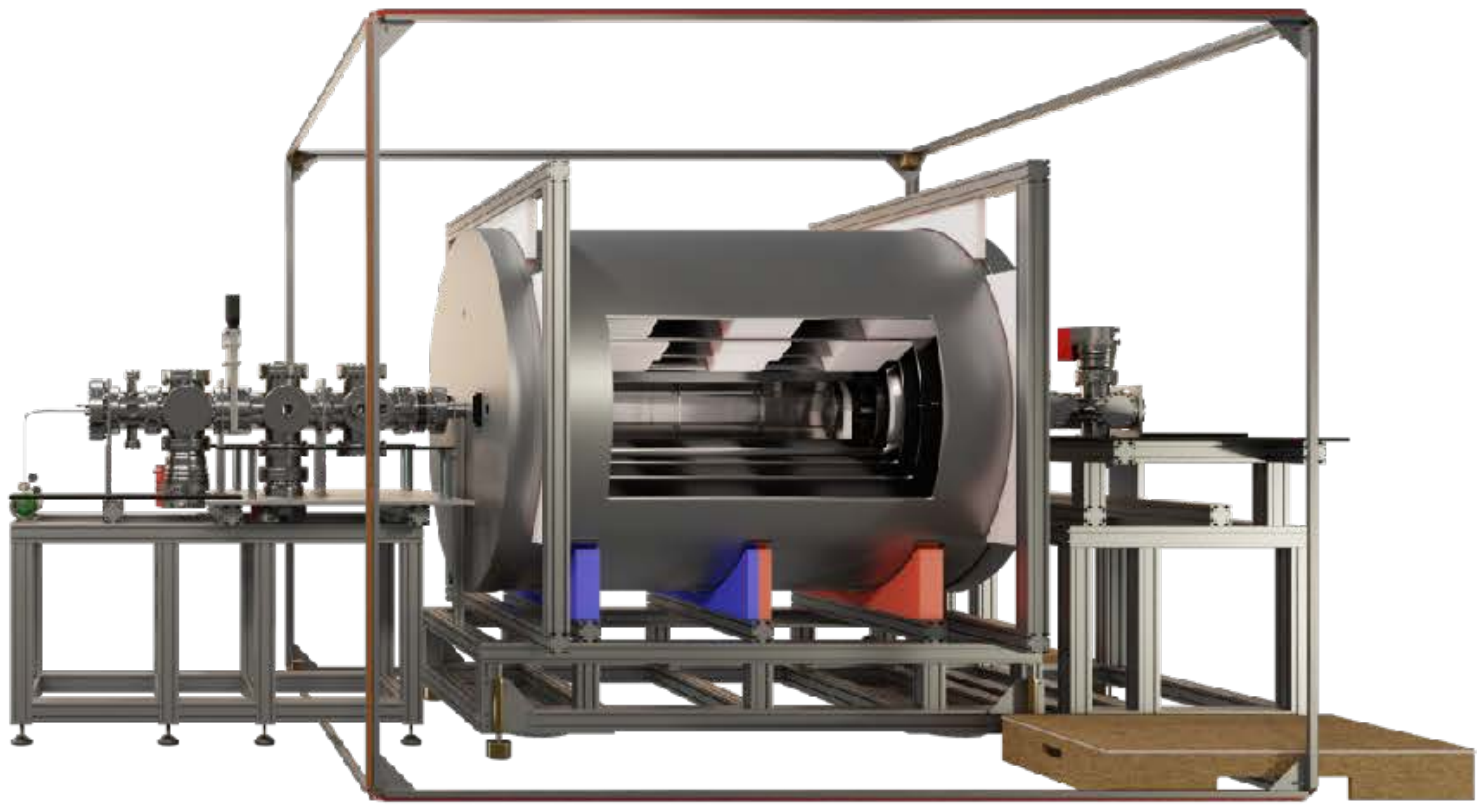




Current status

Phase 1: Fast beam

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



Current status

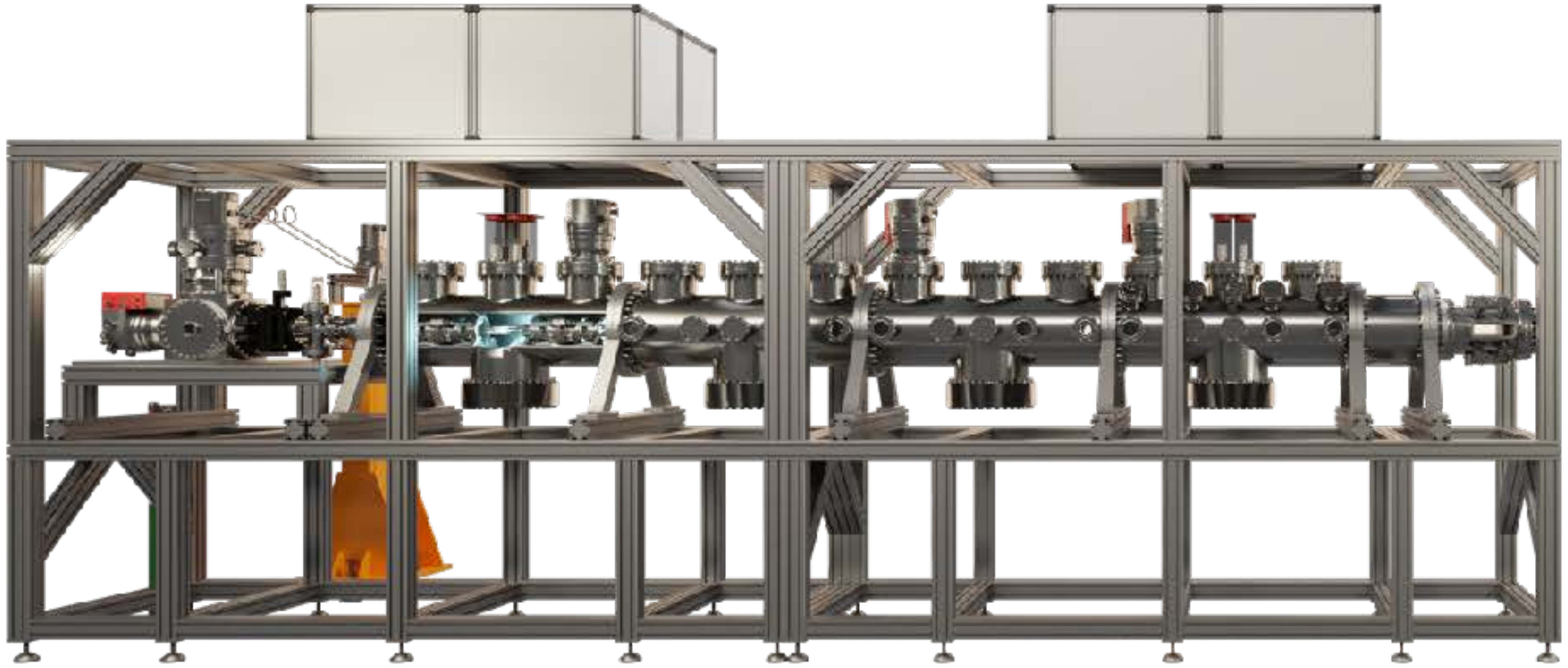
Moving the magnetic shield
to new lab a few weeks ago

Phase 2: Slow beam

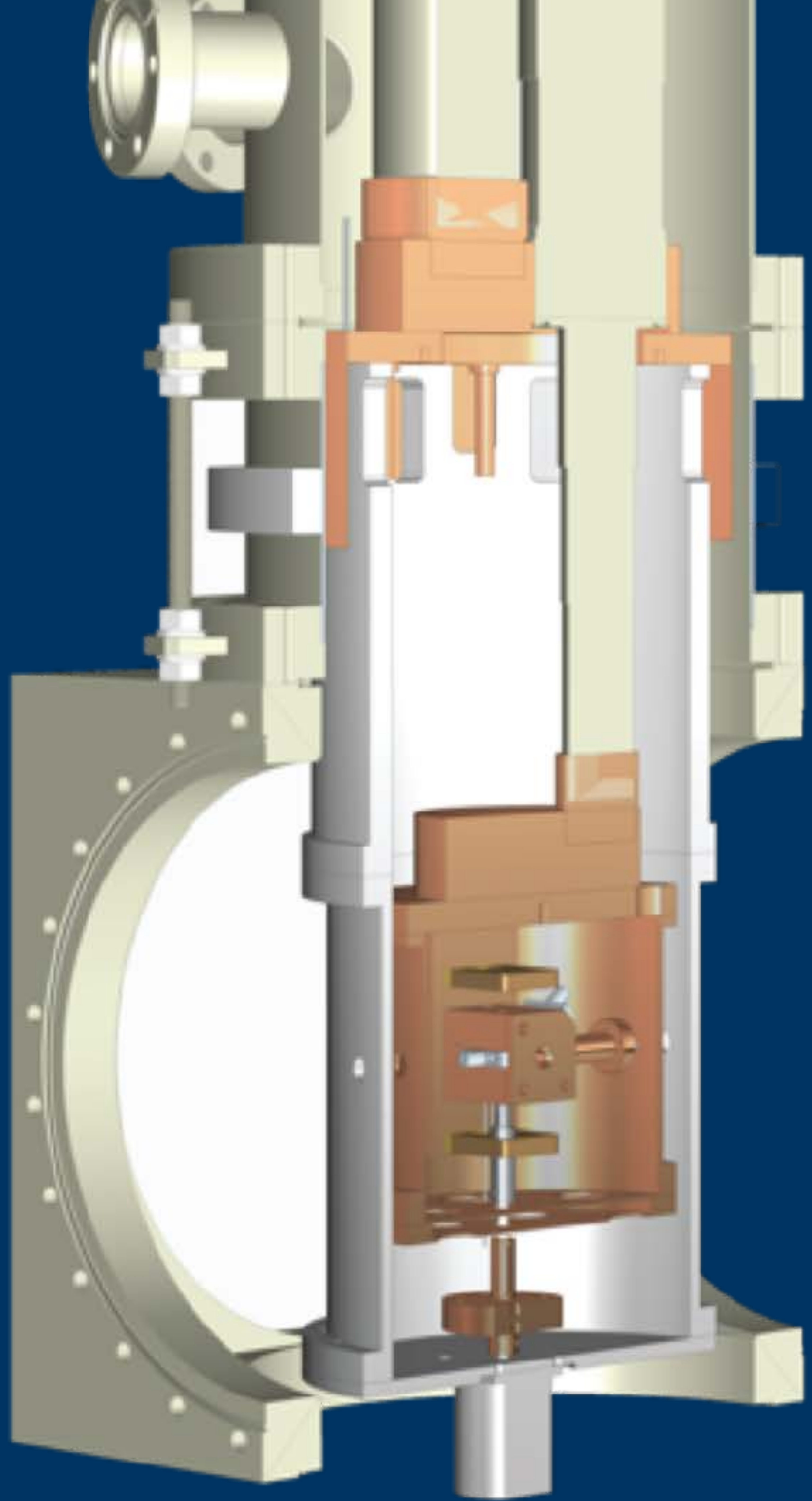
Cryogenic beam (200 m/s)

Transverse laser cooling

Stark deceleration



Increase statistics



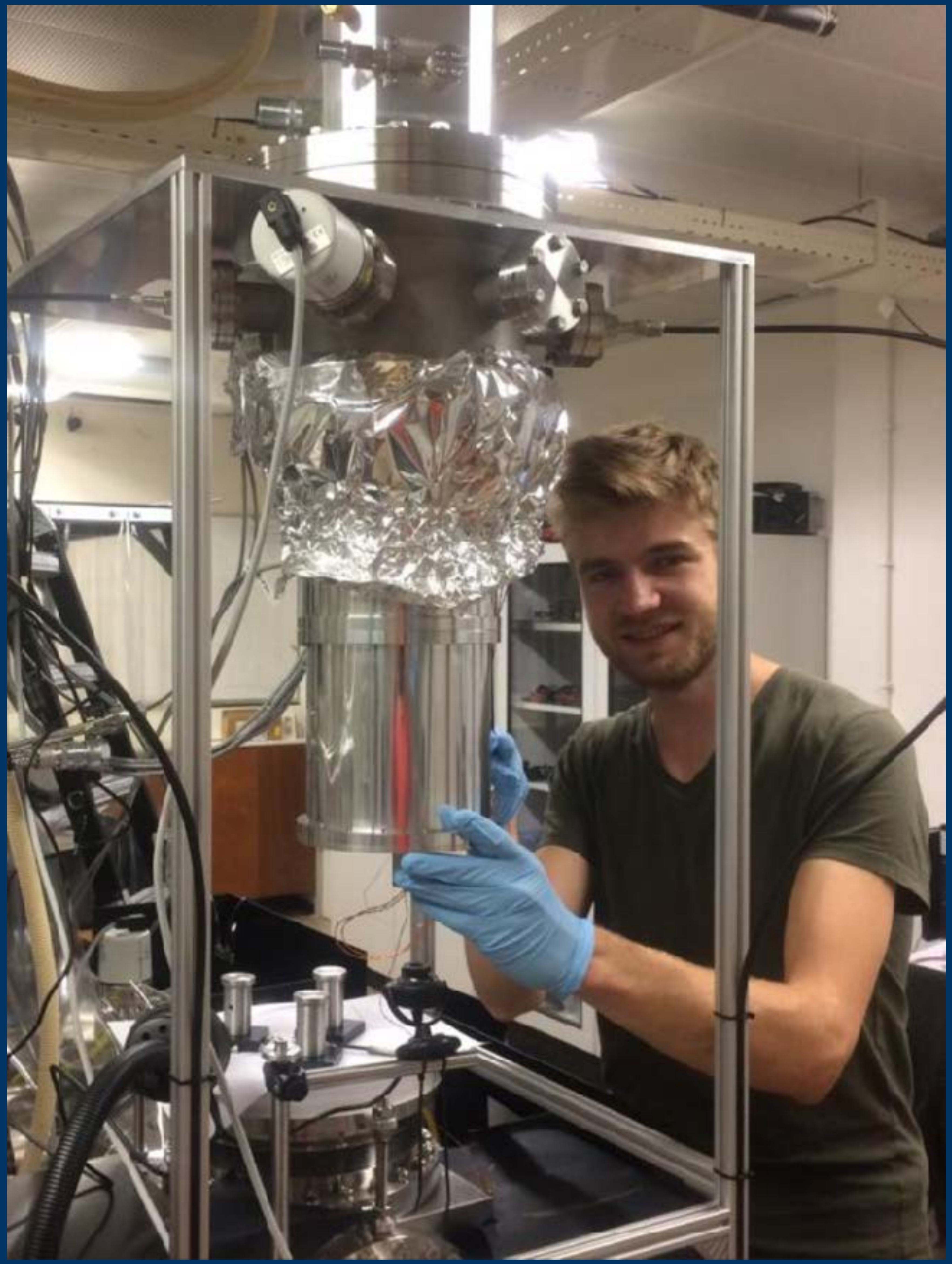
Cryogenic beam

- 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 under construction in Groningen (BaOH and other molecules)

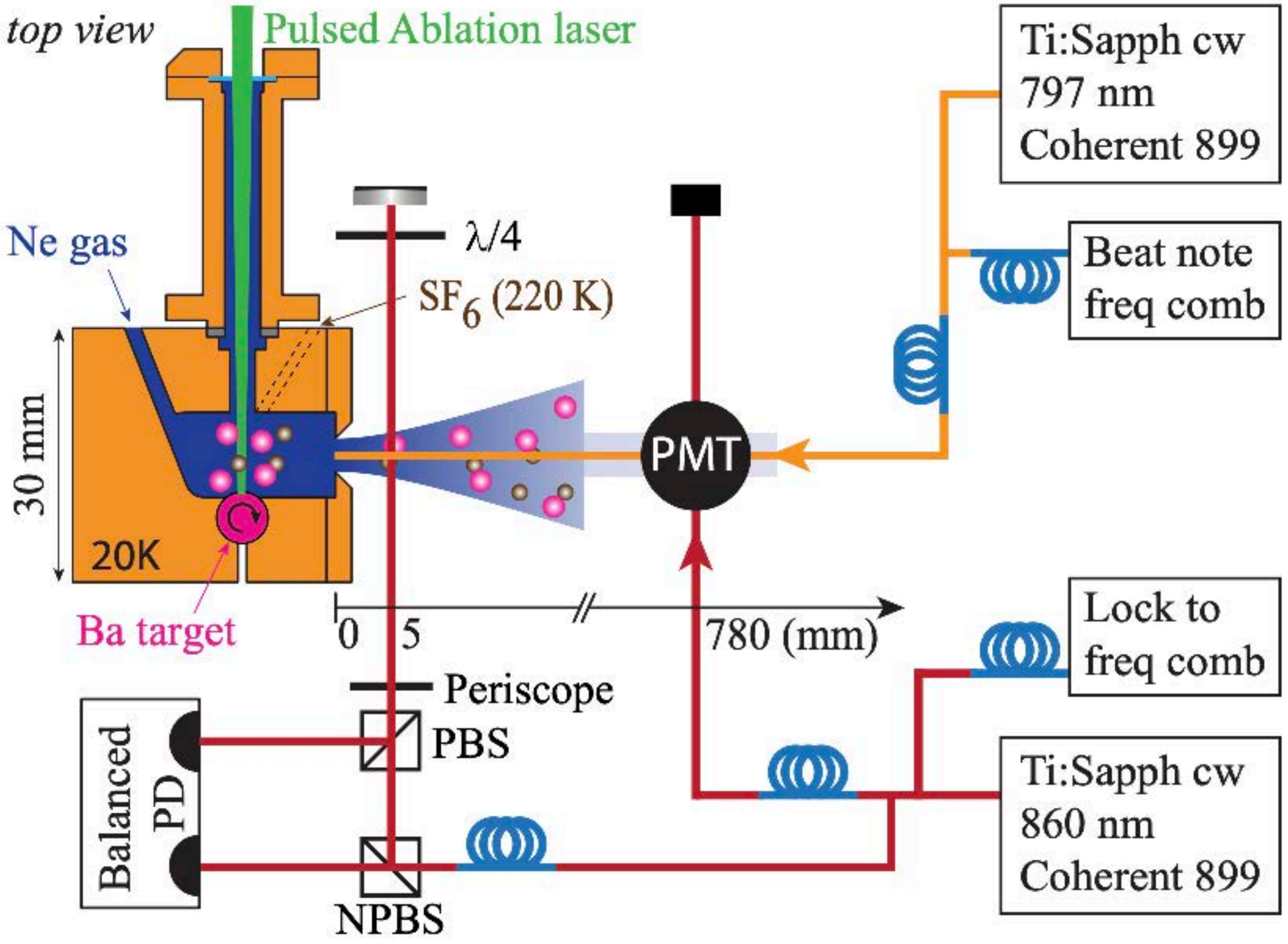
Truppe, S. et al. A buffer gas beam source for short, intense and slow molecular pulses. Journal Of Modern Optics 65, 246–254 (2018).

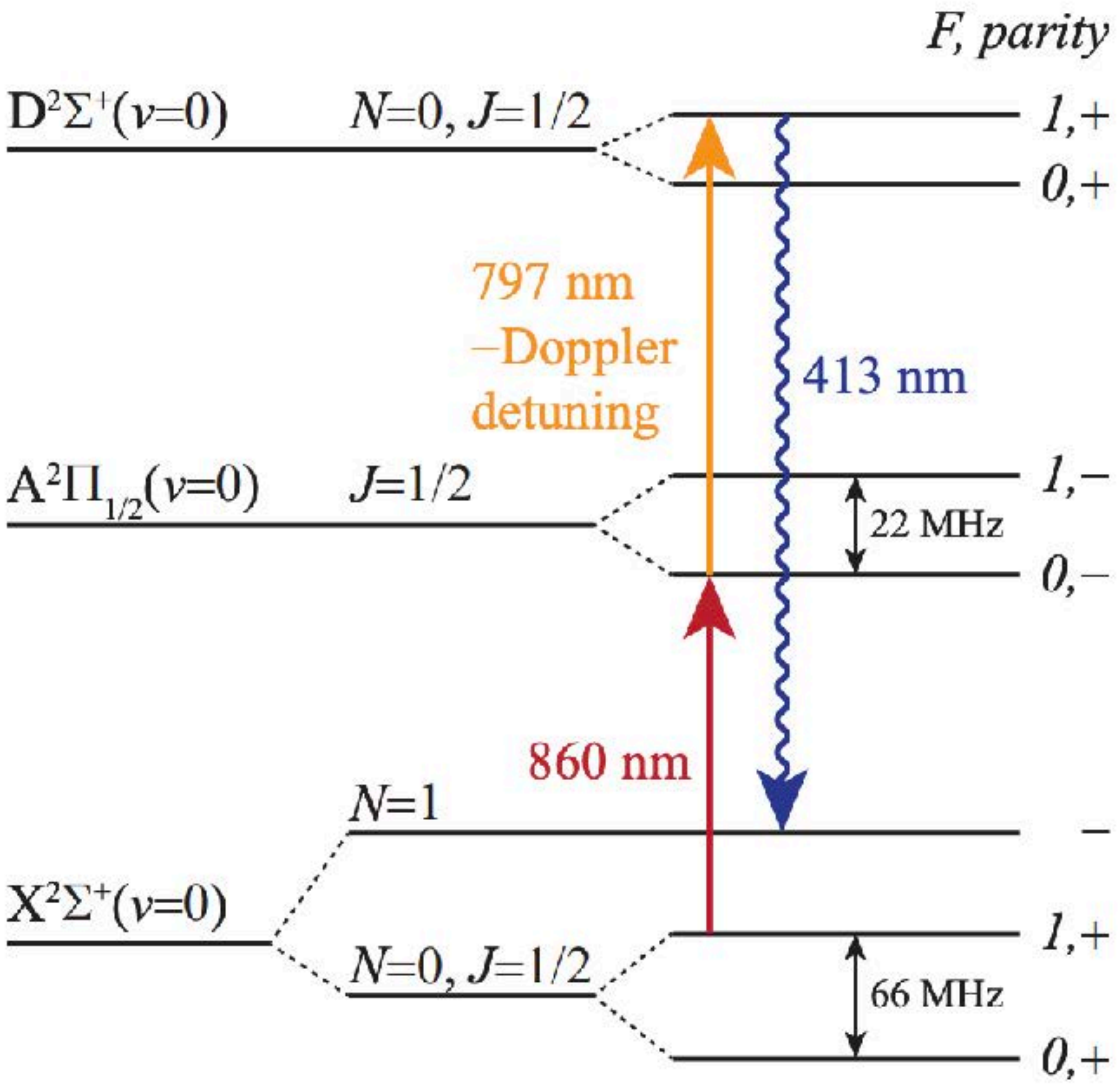


Maarten Mooij, Rick Bethlem @ VU Amsterdam

Optimising the molecular beam source

Mooij et al, arXiv:2401.16588 and arXiv:2401.16590





Combination of arrival time and velocity information gives complete picture

Example: beam cell length

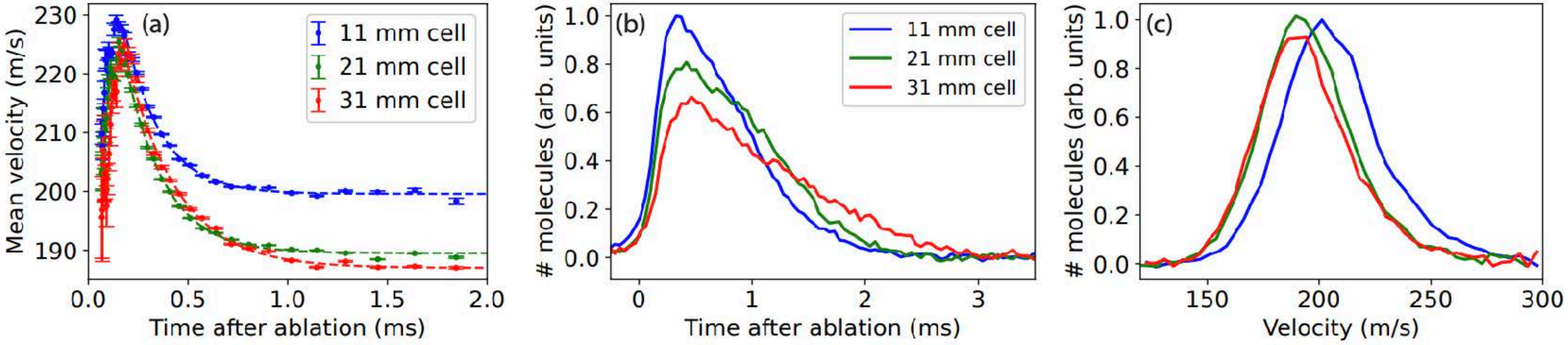
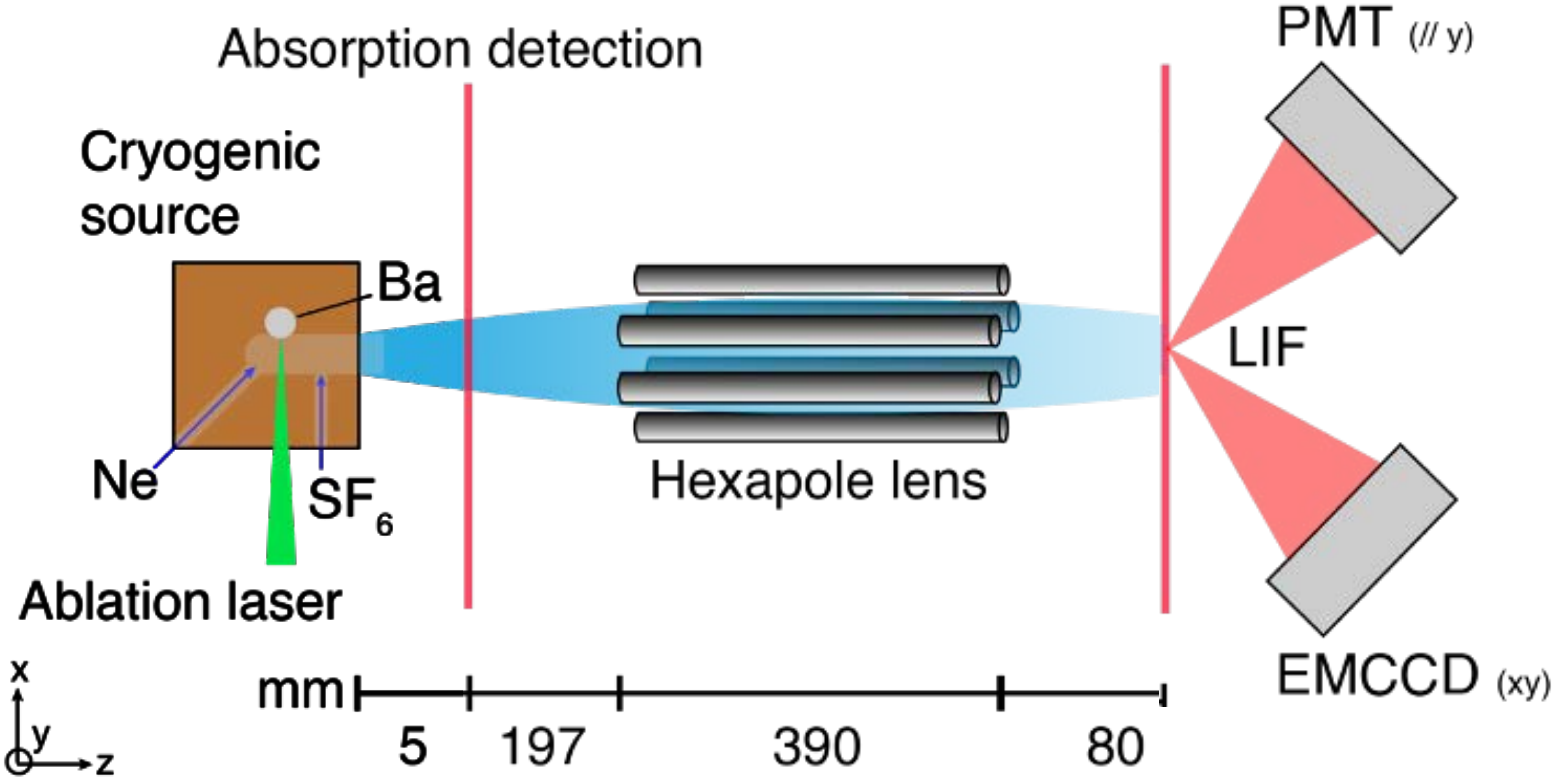


Figure 7. (a) Mean velocity as a function of time, (b) time-of-flight and (c) velocity distribution for three different cell lengths. The velocity in the tail of the molecular pulse is seen to decrease significantly, while the intensity is comparable.

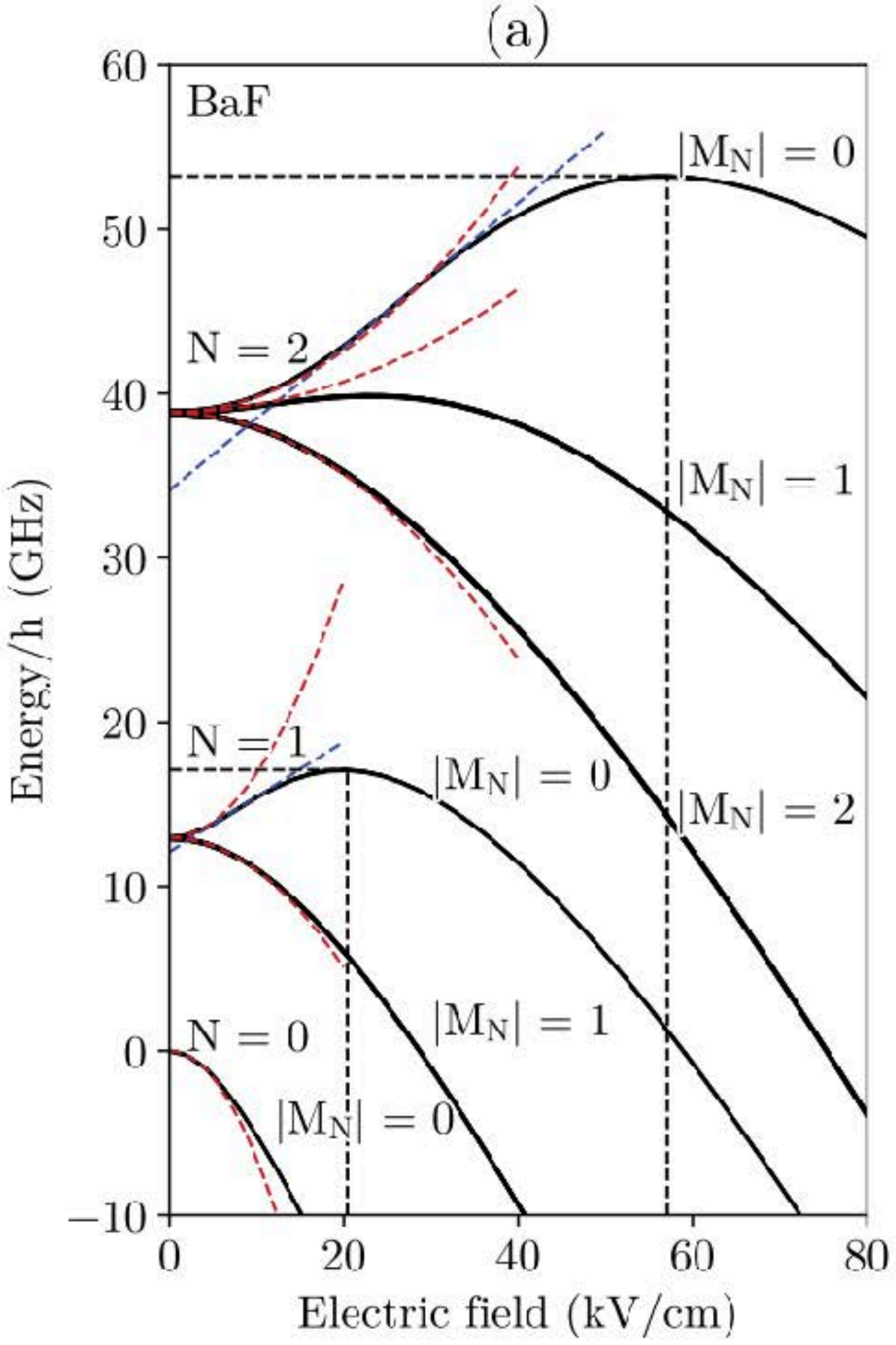
A slow beam diverges

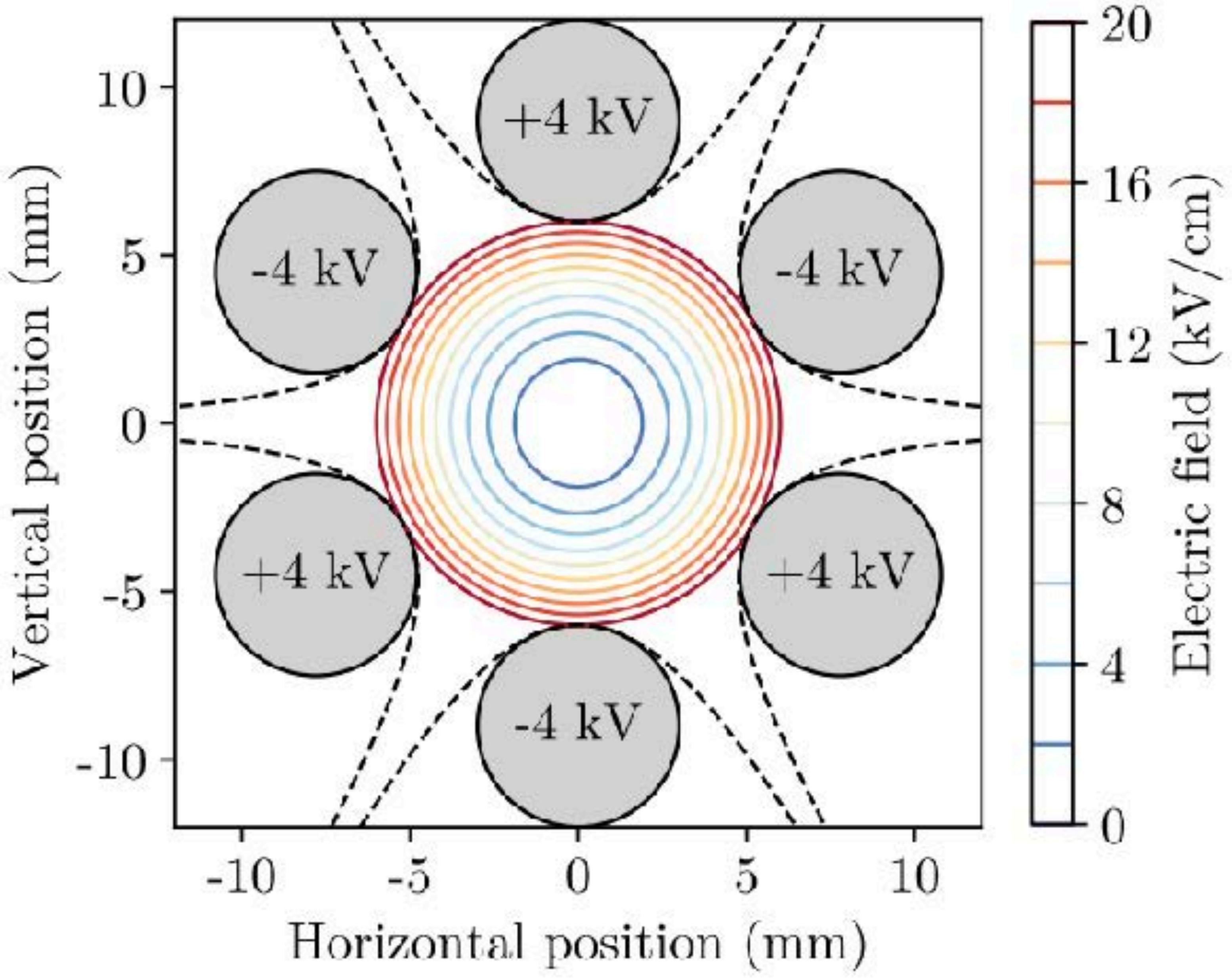
Hexapole focussing

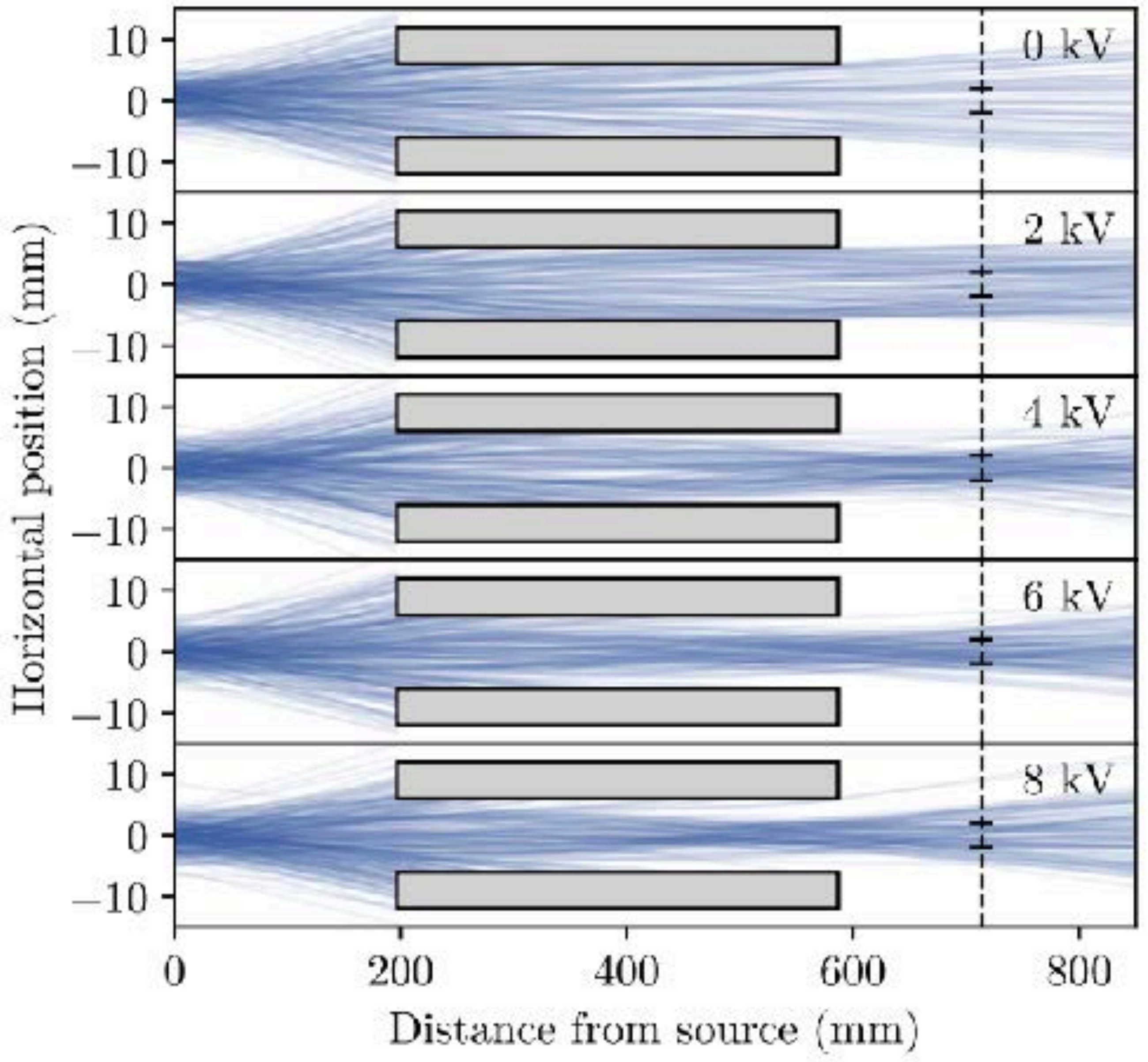


BaF in electric fields

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

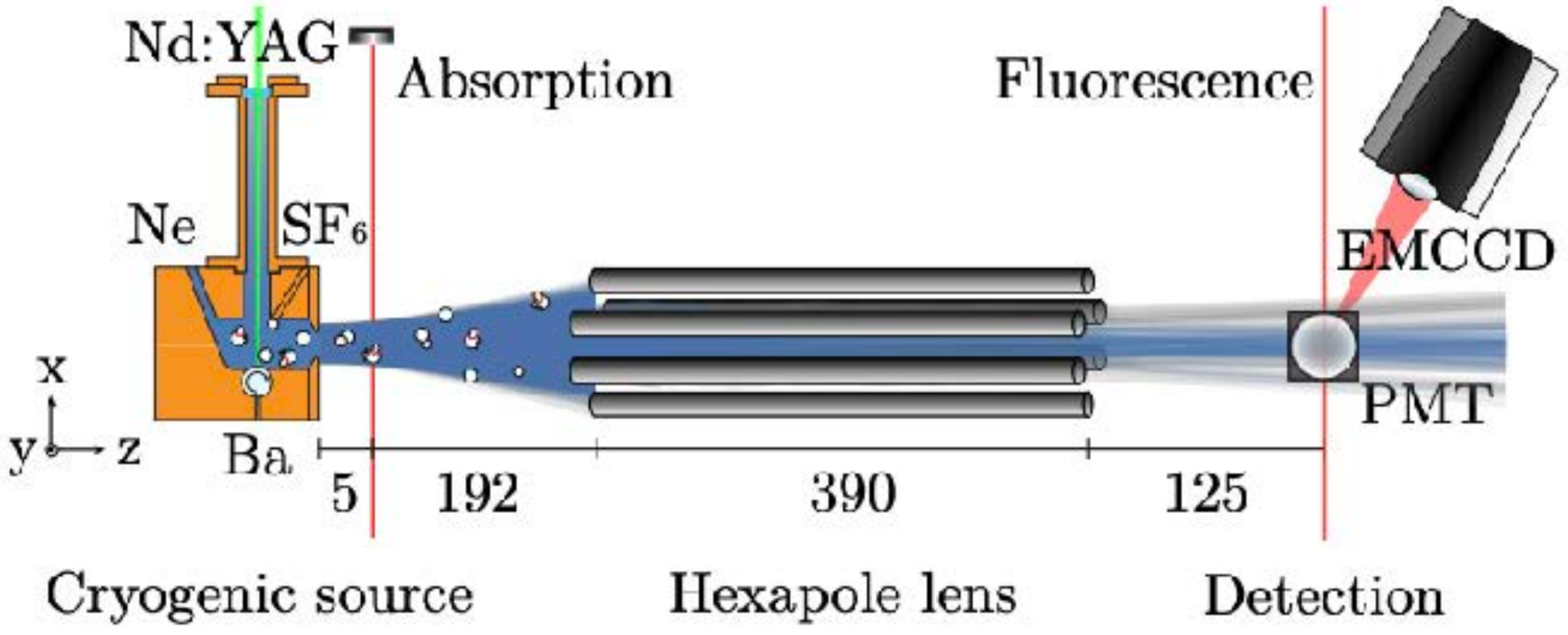


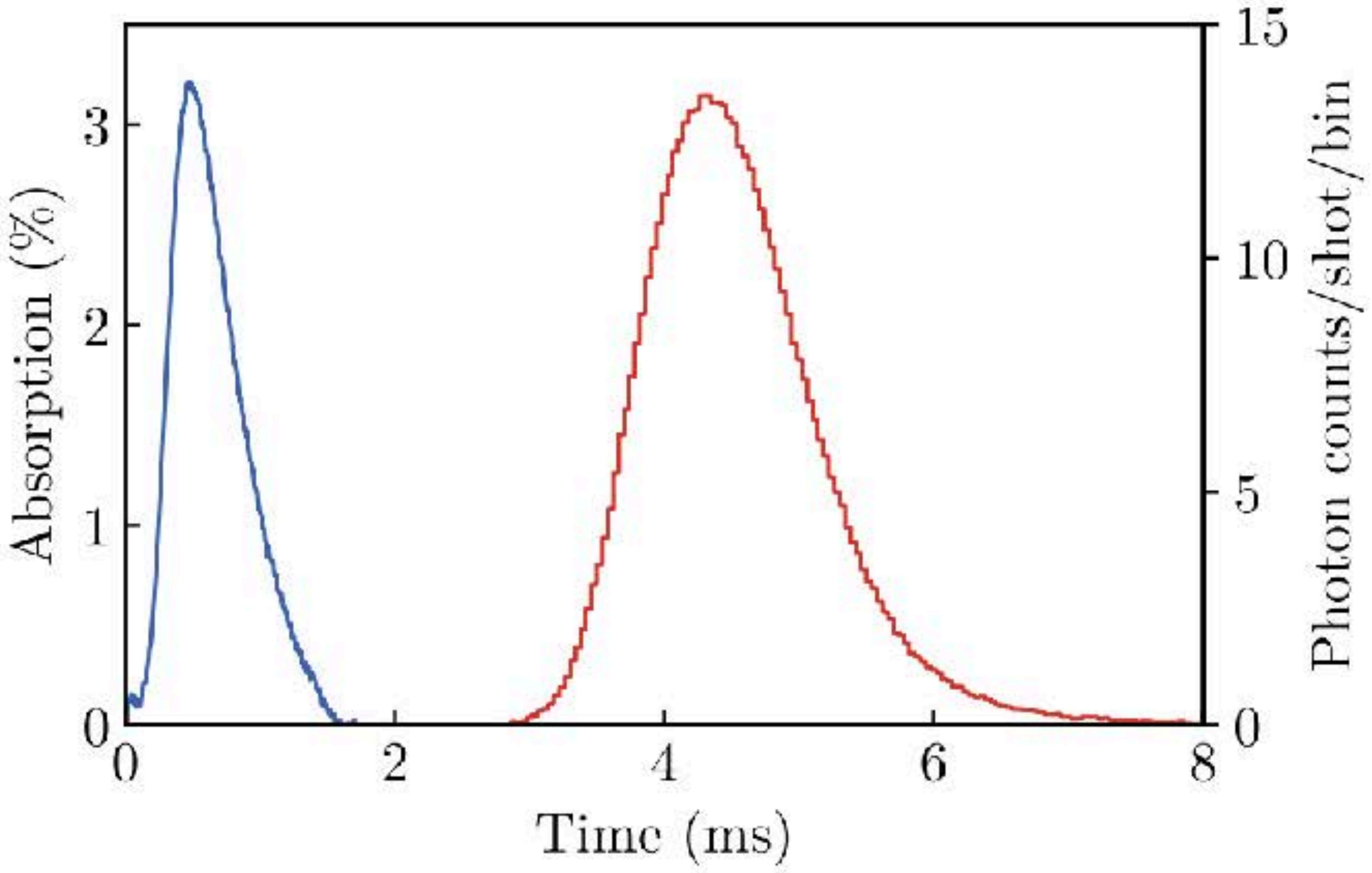


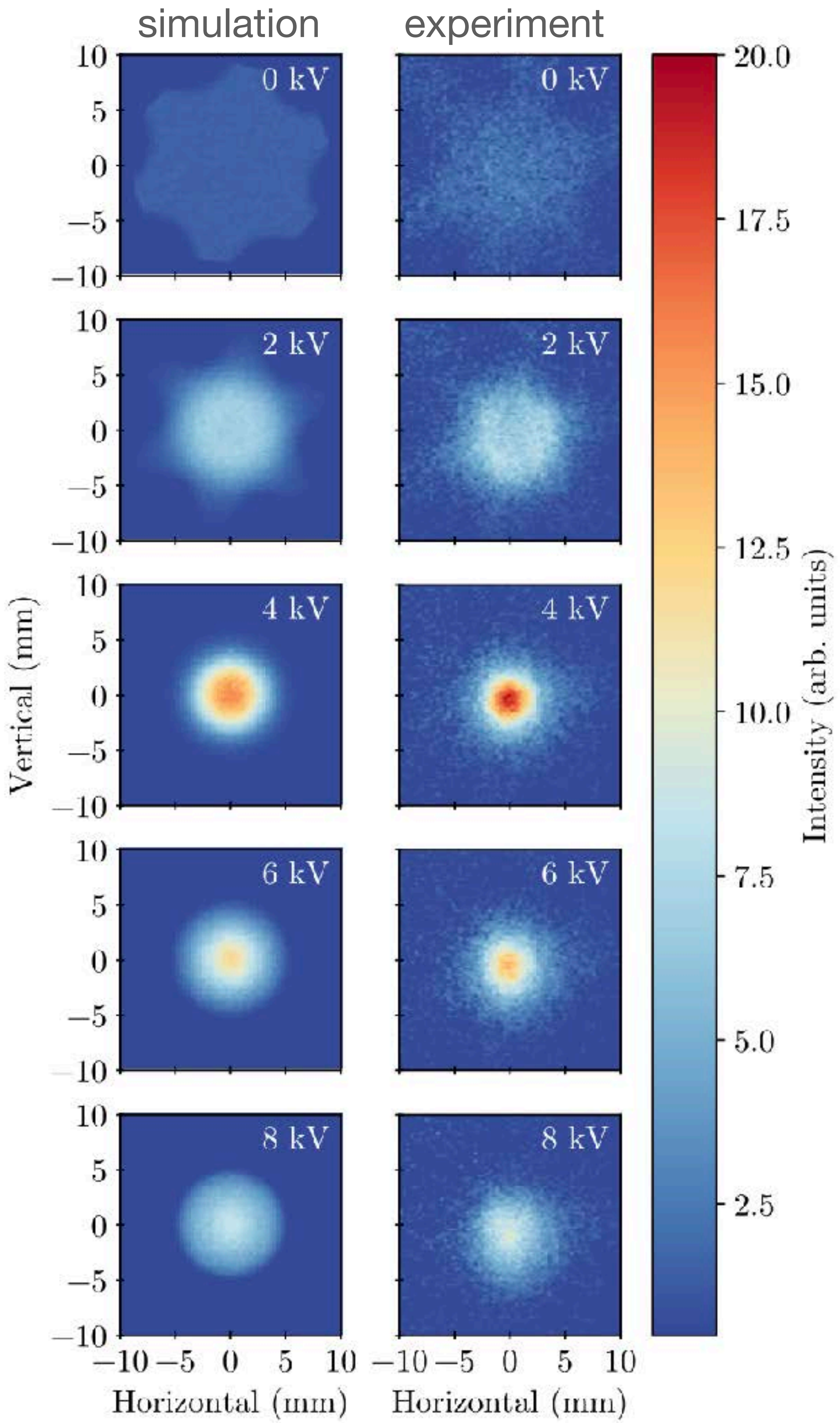


Hexapole focussing

Anno Touwen et al, arXiv:2402.09300

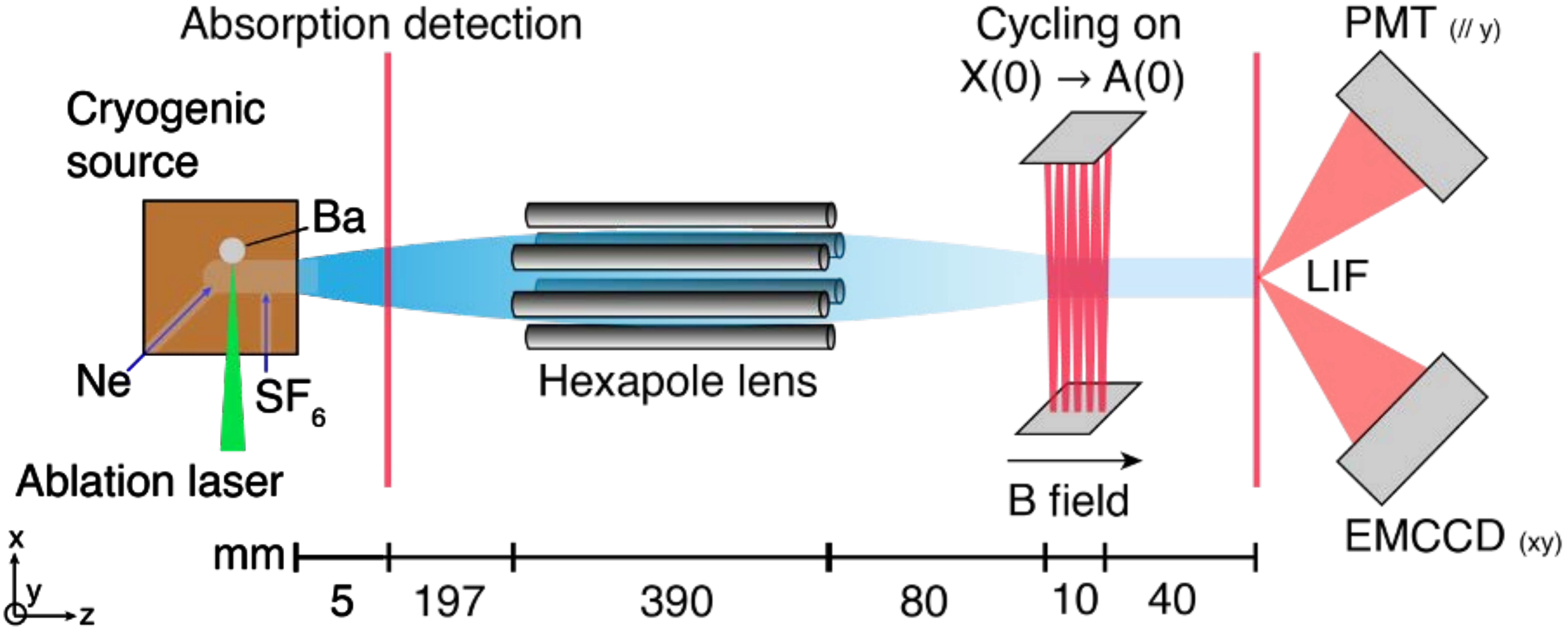






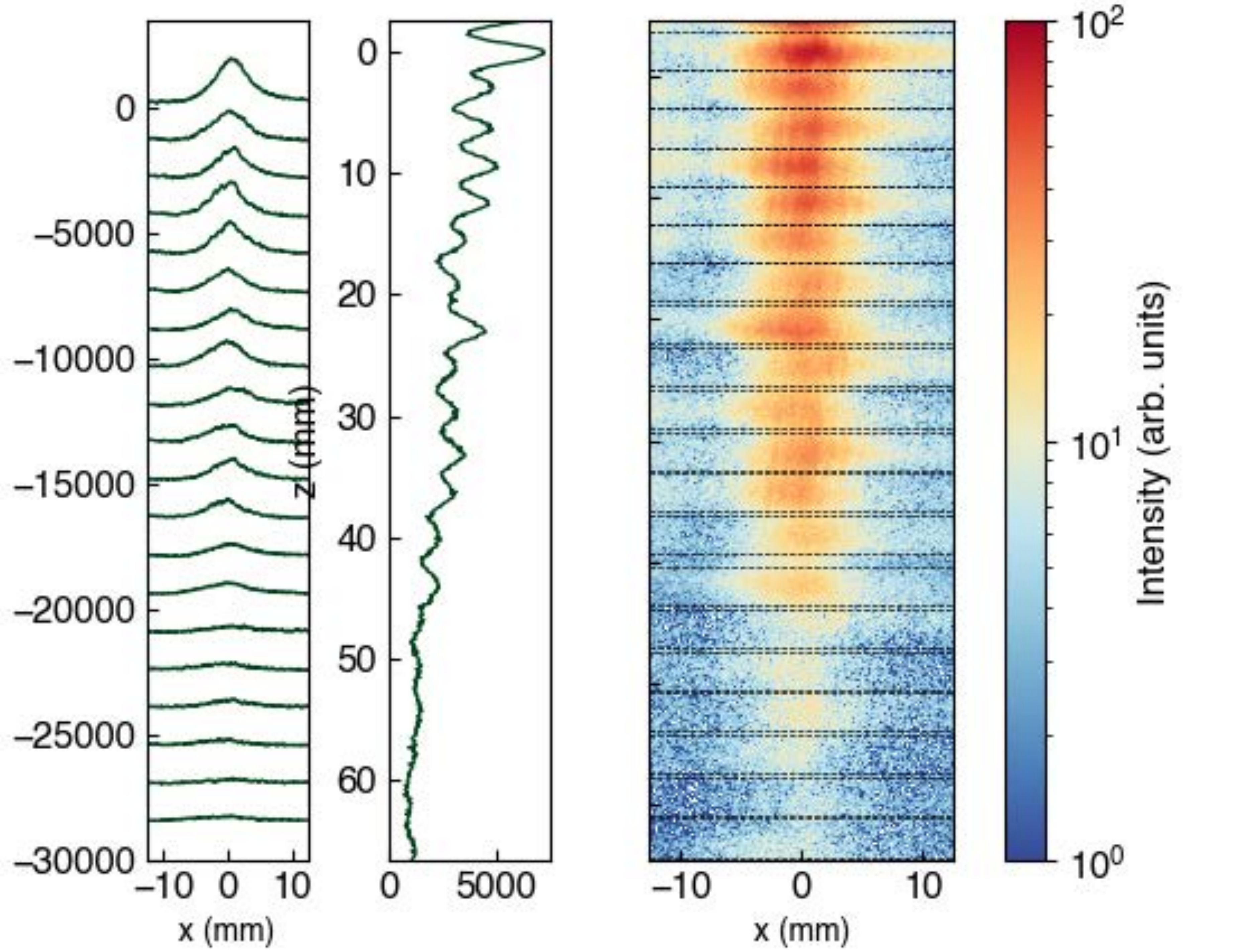
A slow beam diverges

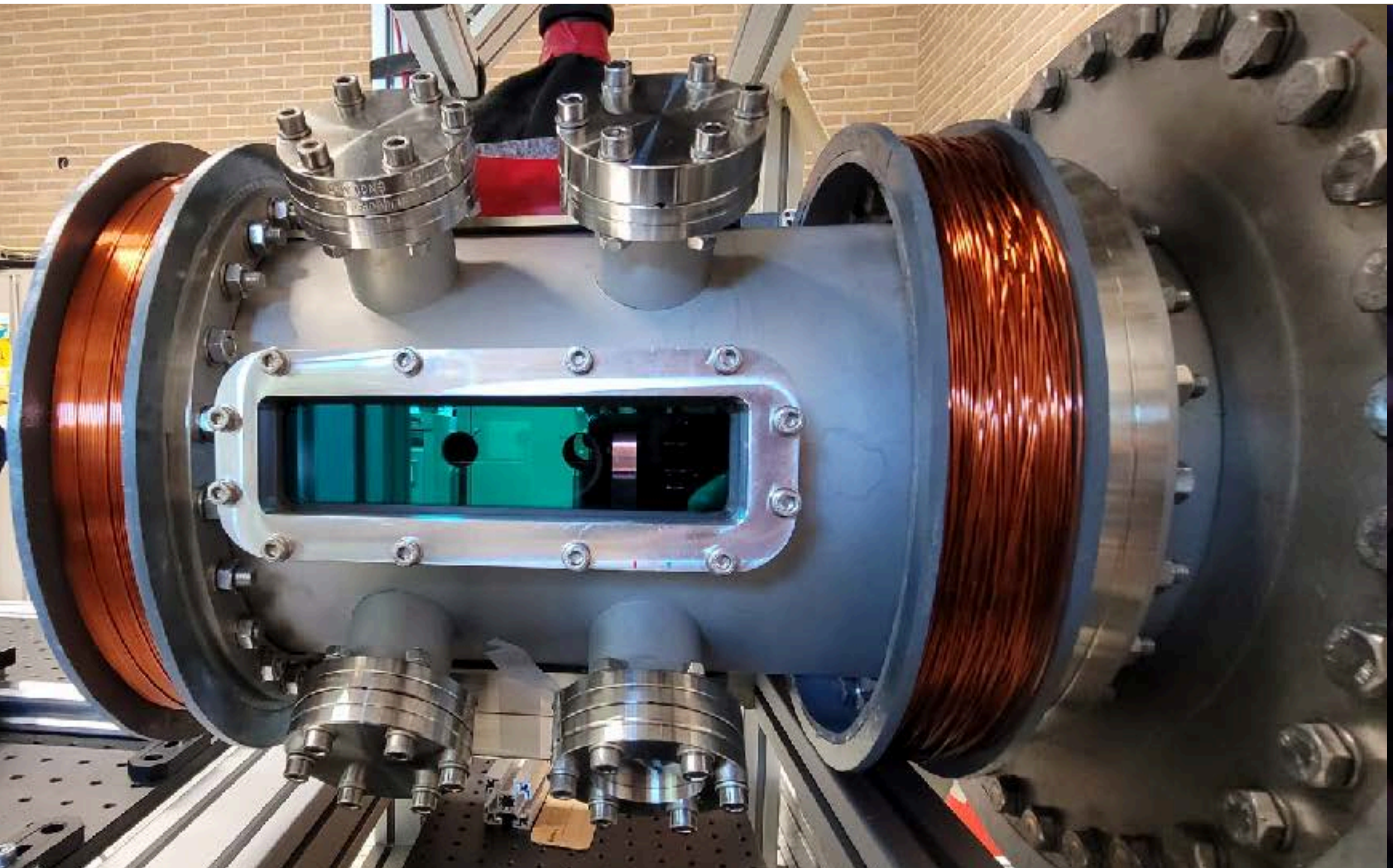
Laser cooling

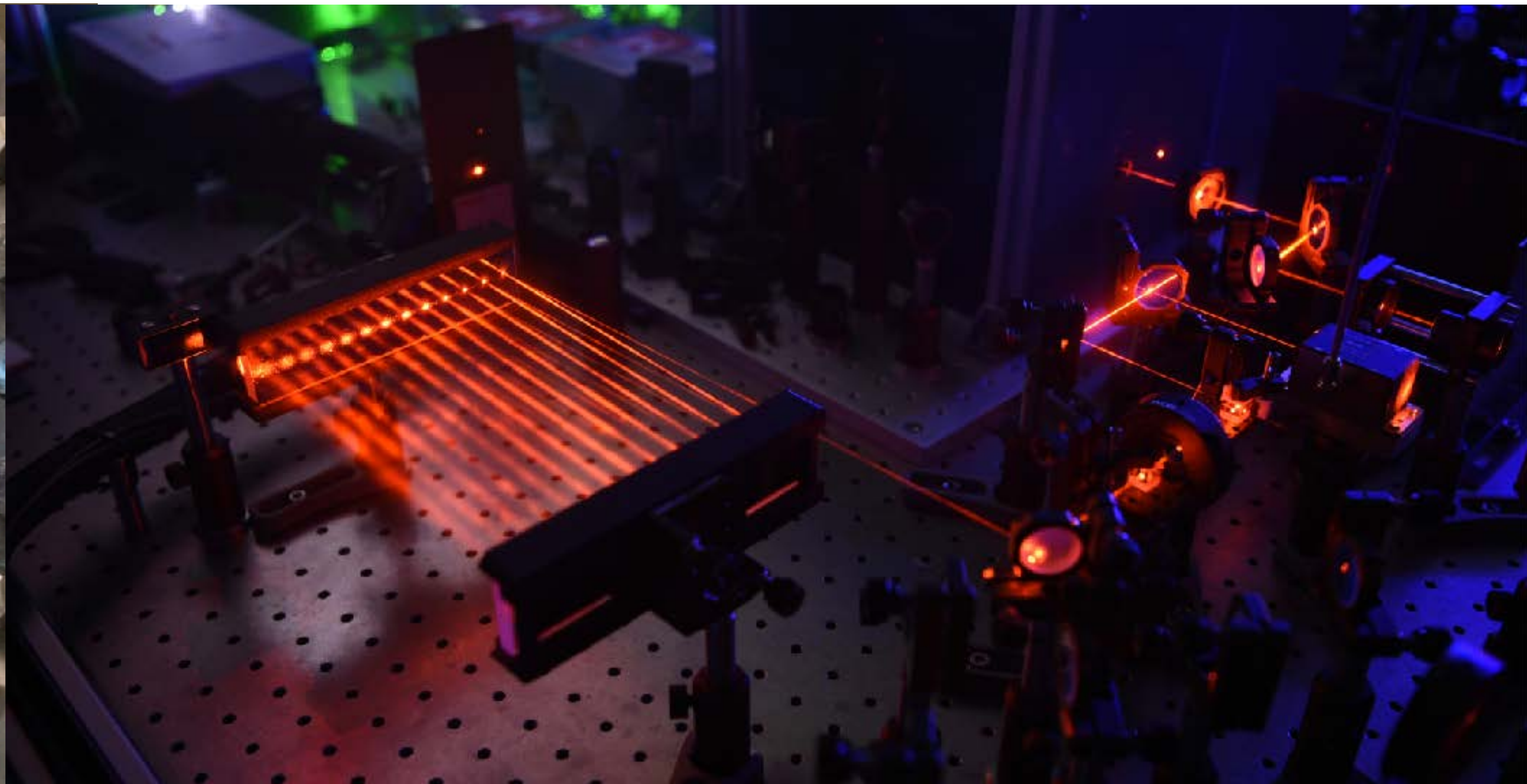


Transverse laser cooling

CCD camera images







A few words on laser cooling

'molecule X can be lasercooled'

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

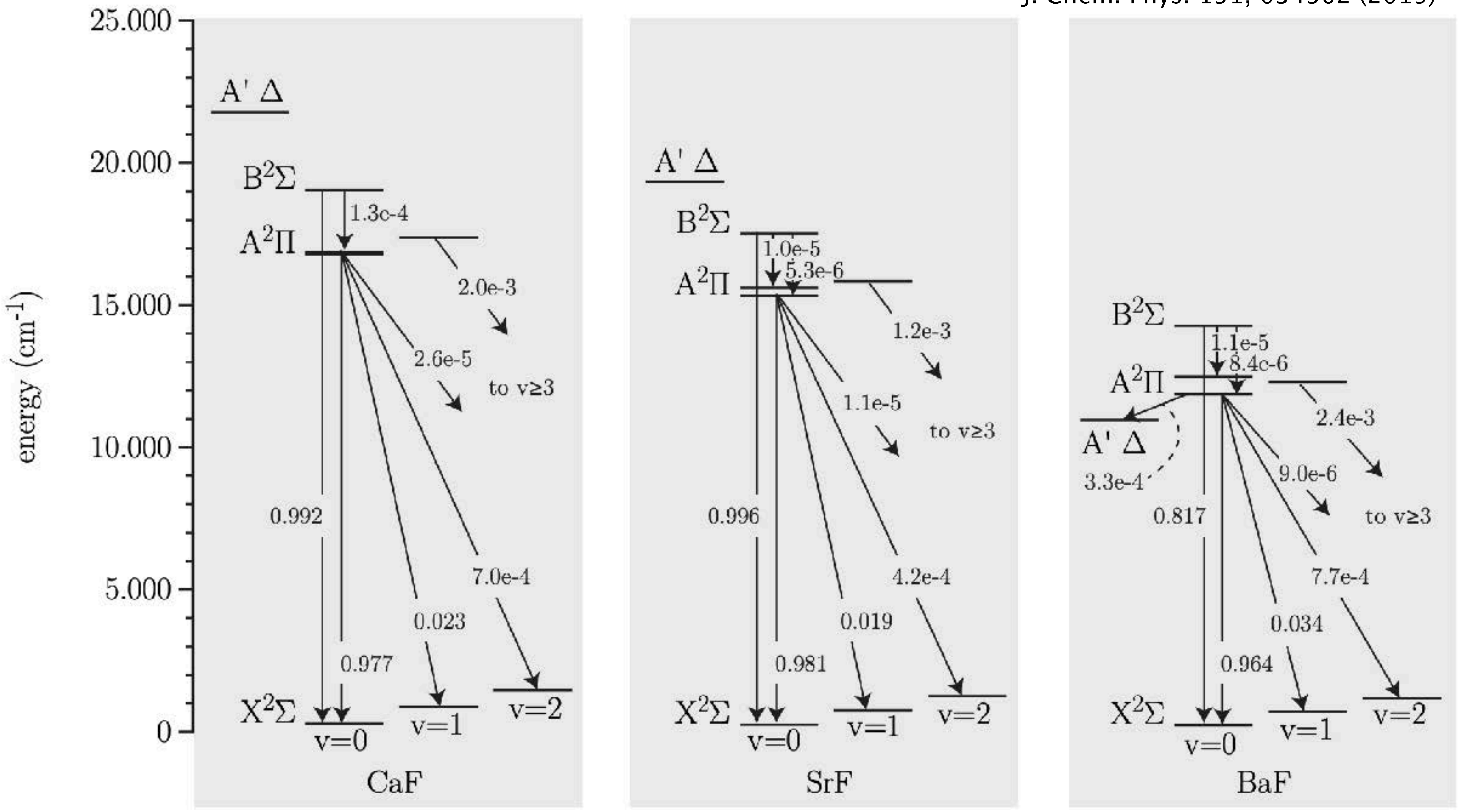
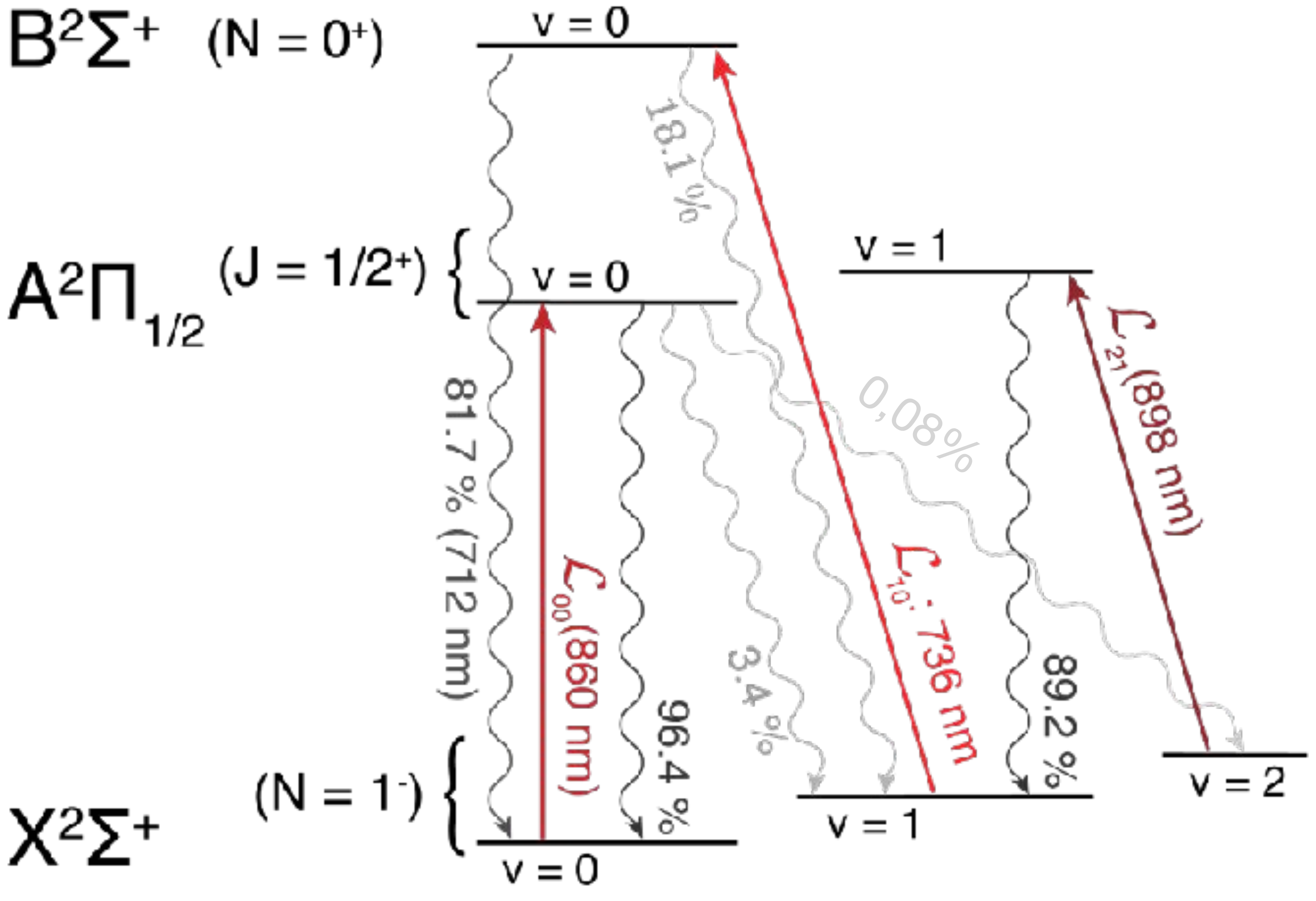
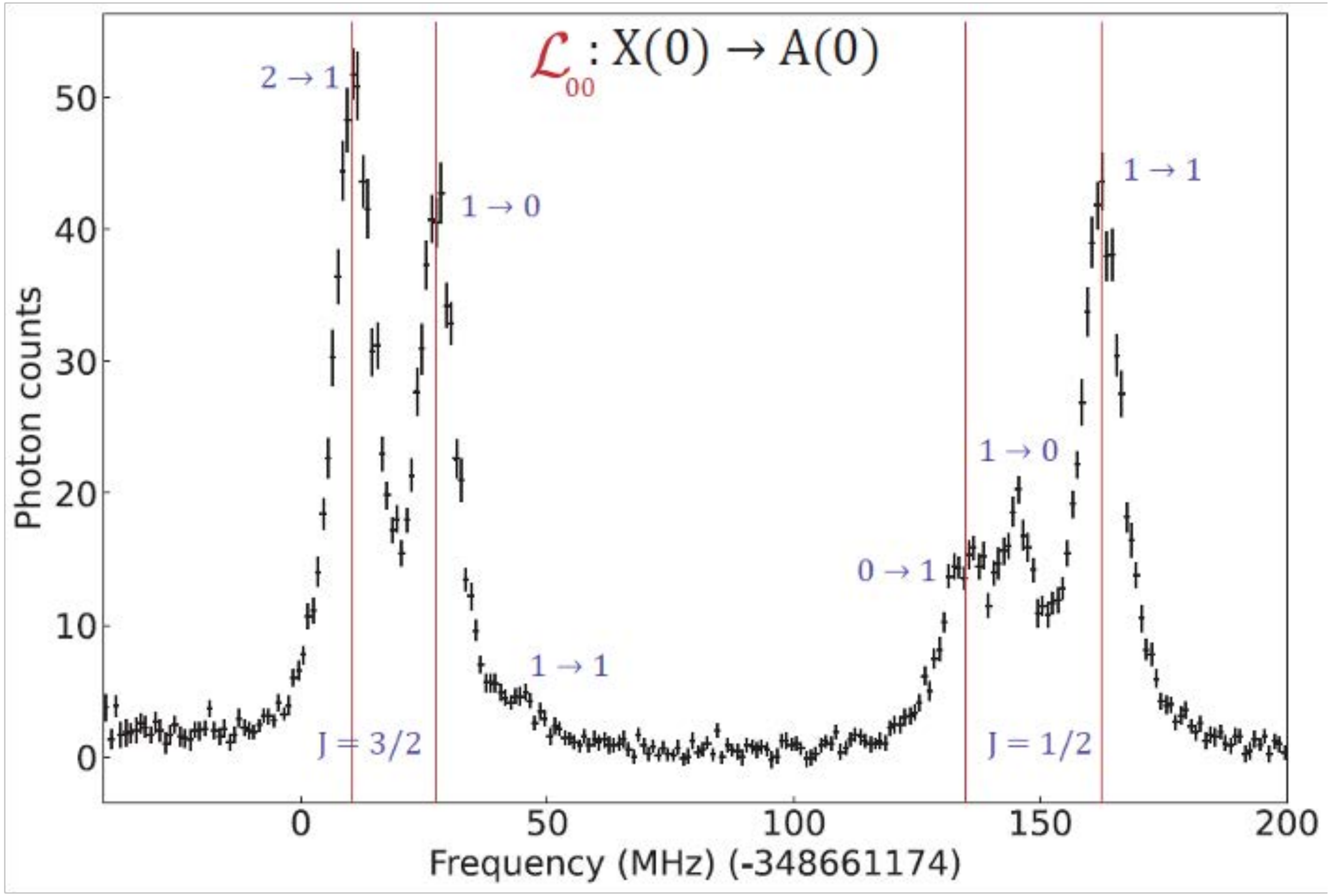


FIG. 5: The most important energy levels for laser-cooling and the calculated relative decay fractions for CaF, SrF, and BaF.

Transitions for laser cooling

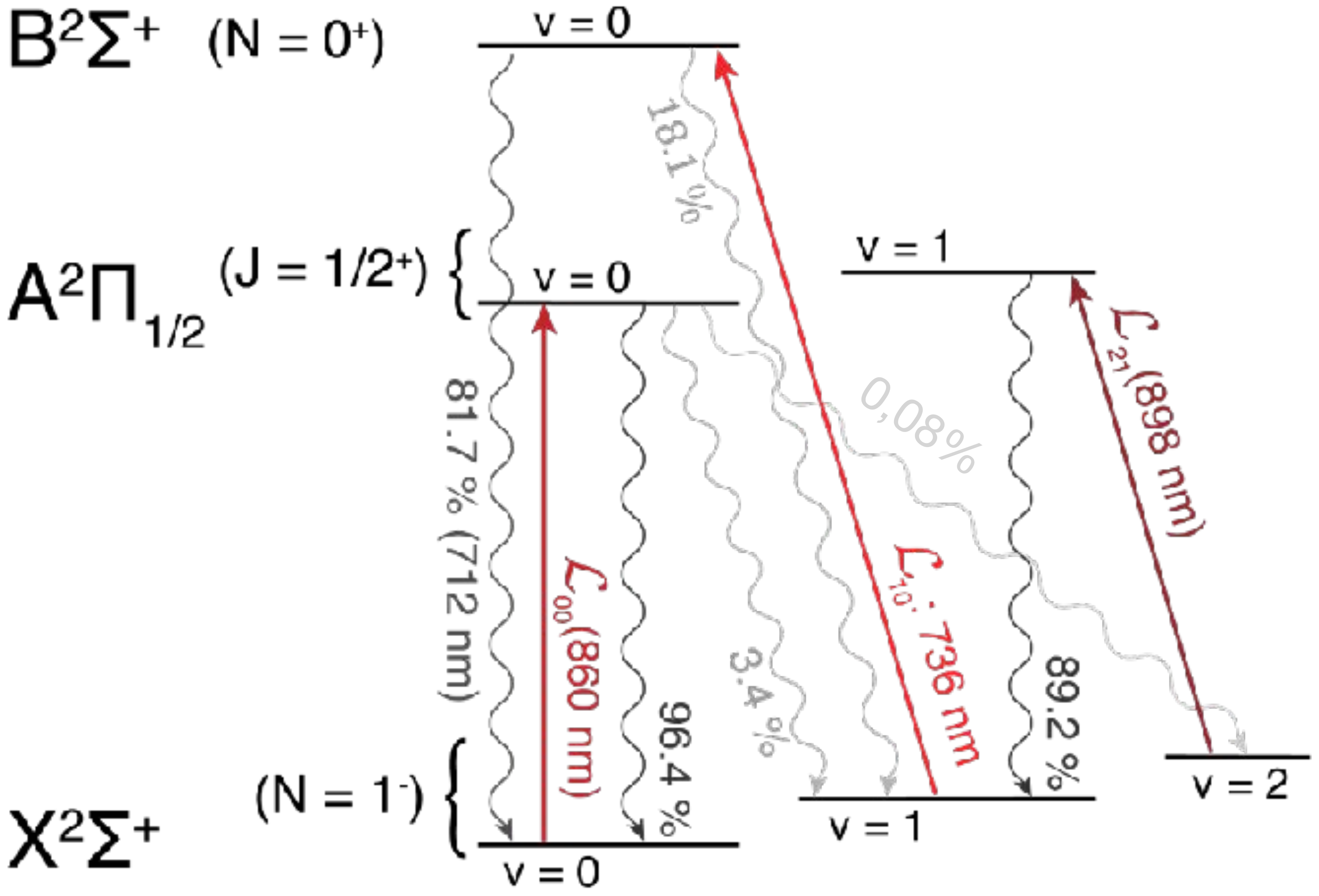
In the presence of hyperfine structure

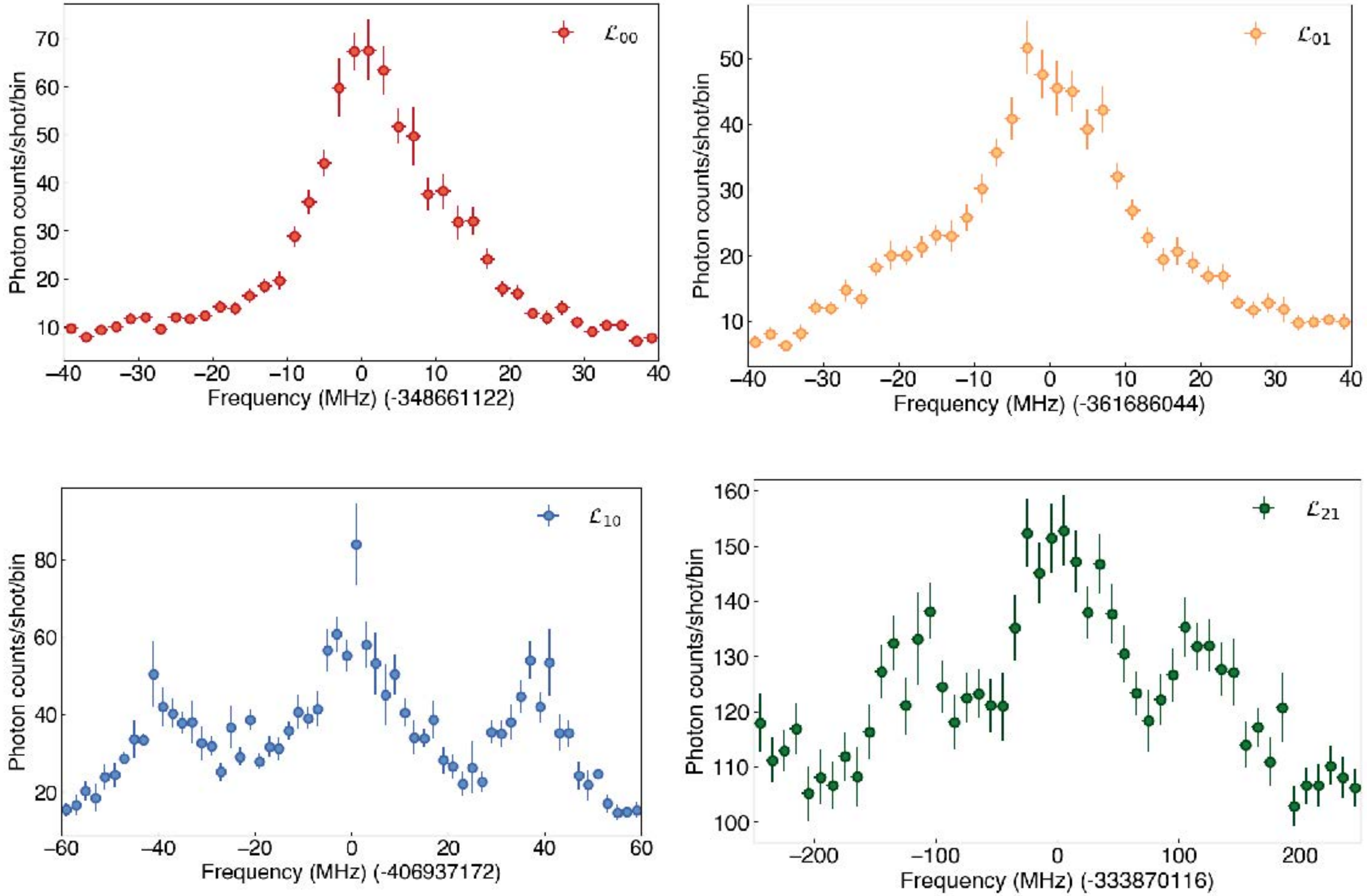




Accumulation in v=1 and v=2

Observation @ 860 nm

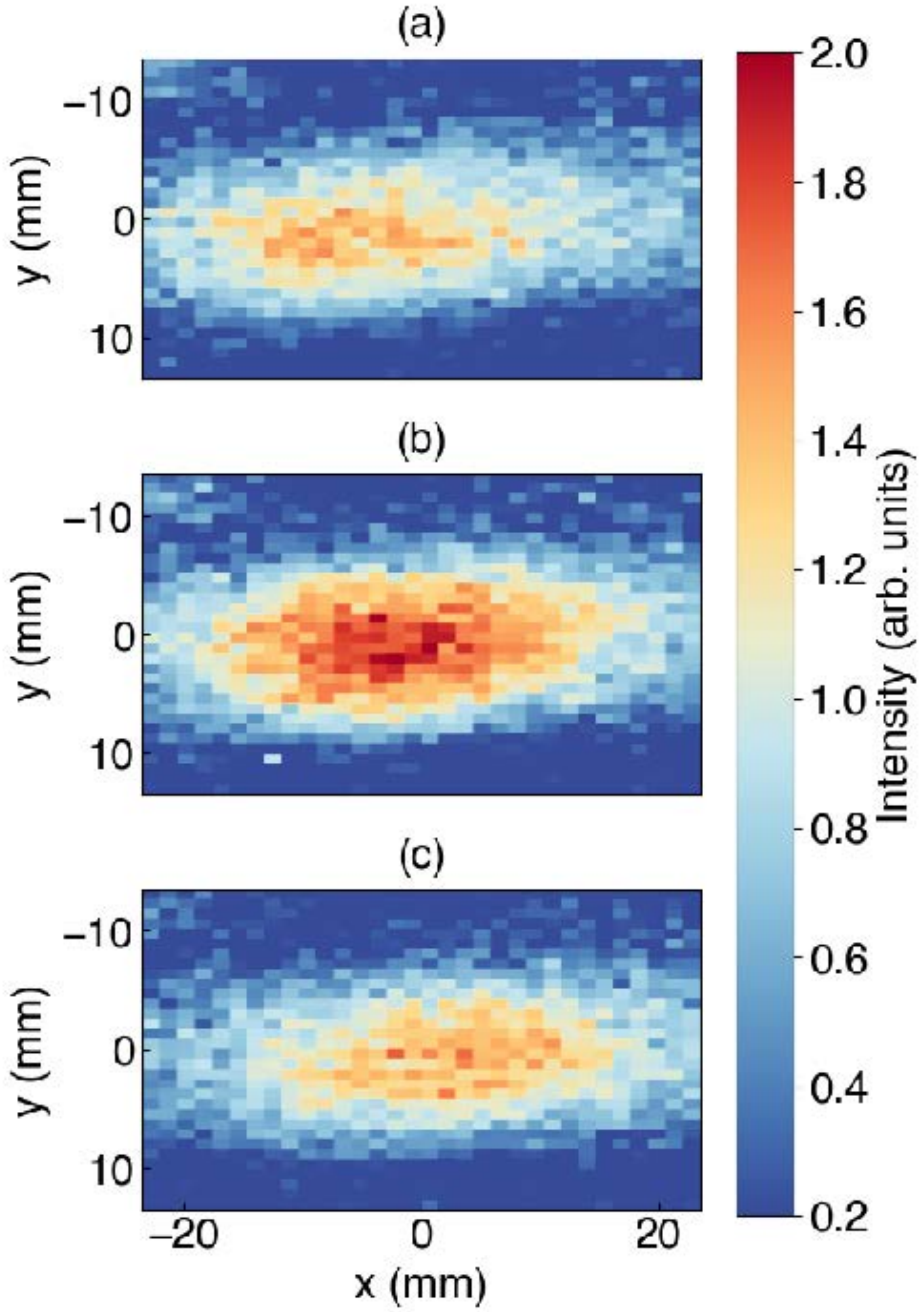


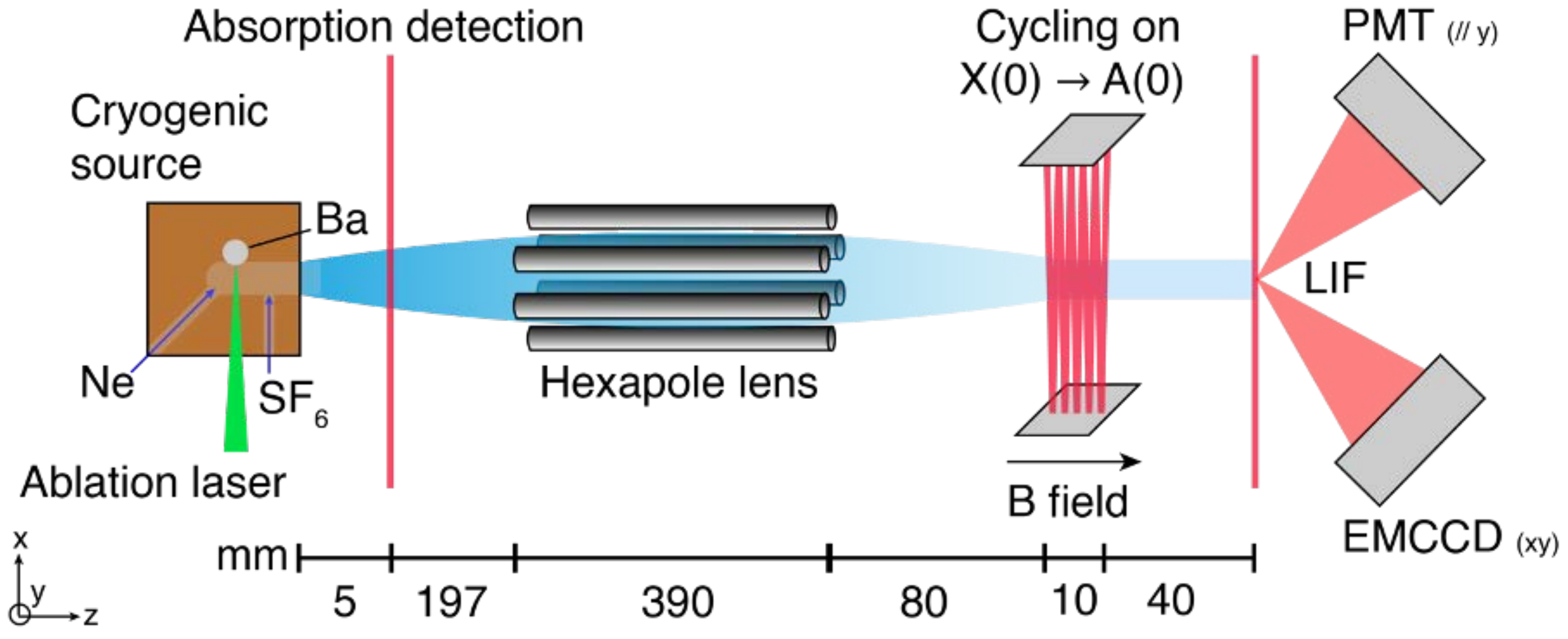


Current status

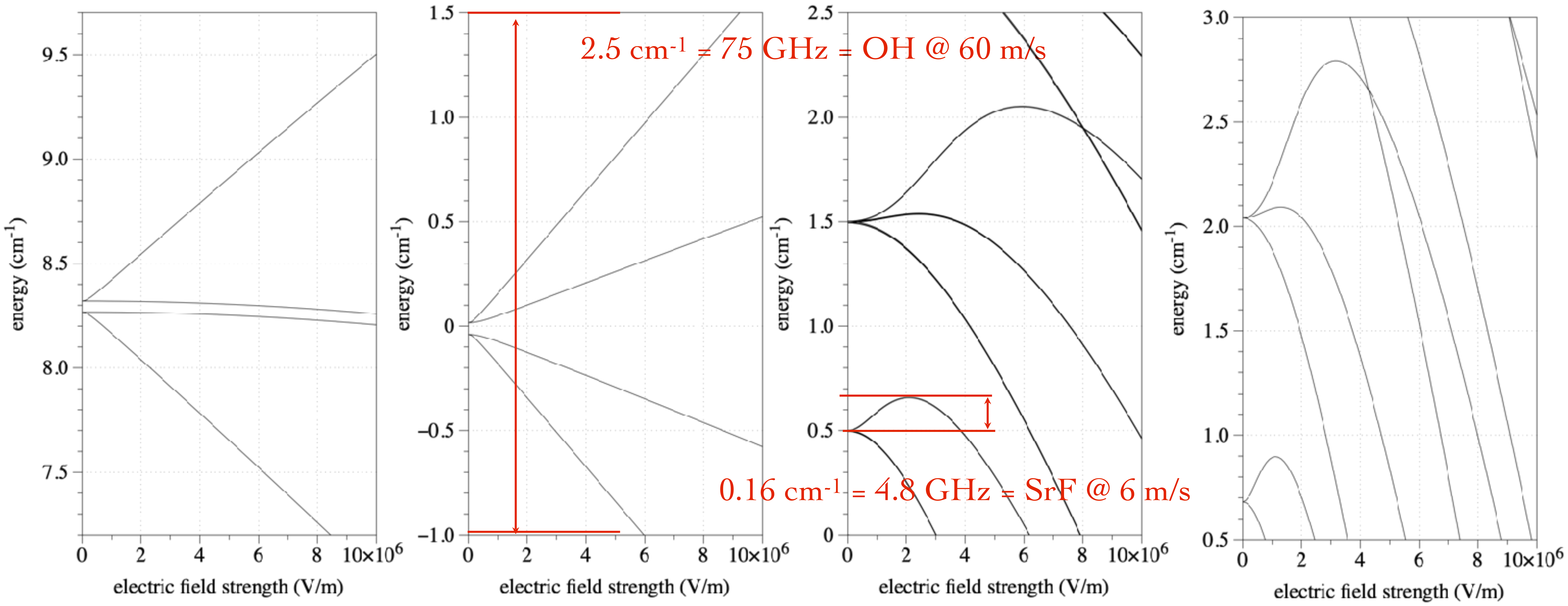
Phase 2a: 200 m/s beam

- Cryogenic beams optimised, ~10^{10} molecules / shot
- Hexapole implemented, gain factor ~5
- Laser cooling setup completed
- Currently extending 1D cooling to 2D transverse cooling
- Combine with interaction zone this year





Use Stark shift to decelerate



ND3 OH SrF, BaF SrO

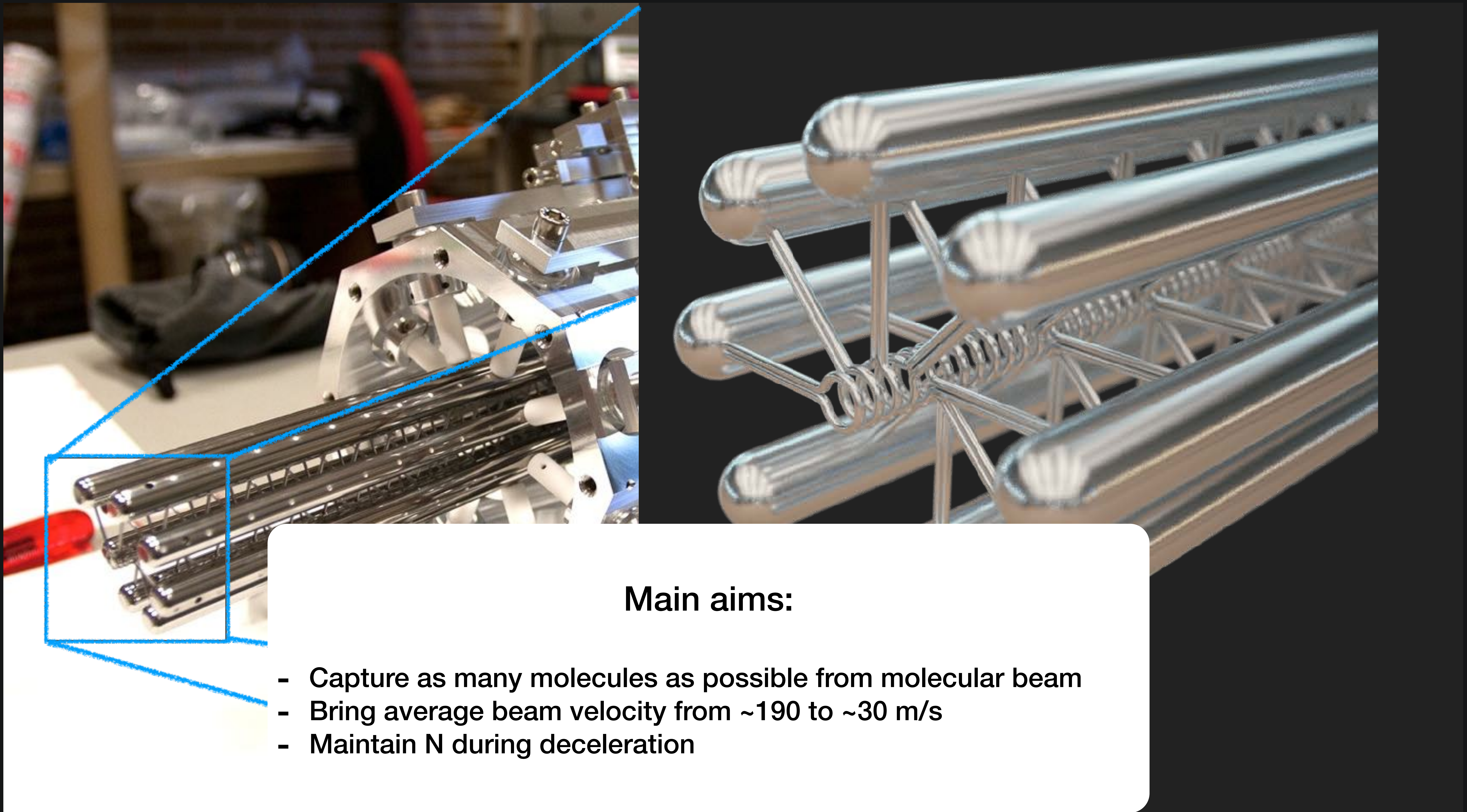
Challenge: extend this technique to heavier species

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)

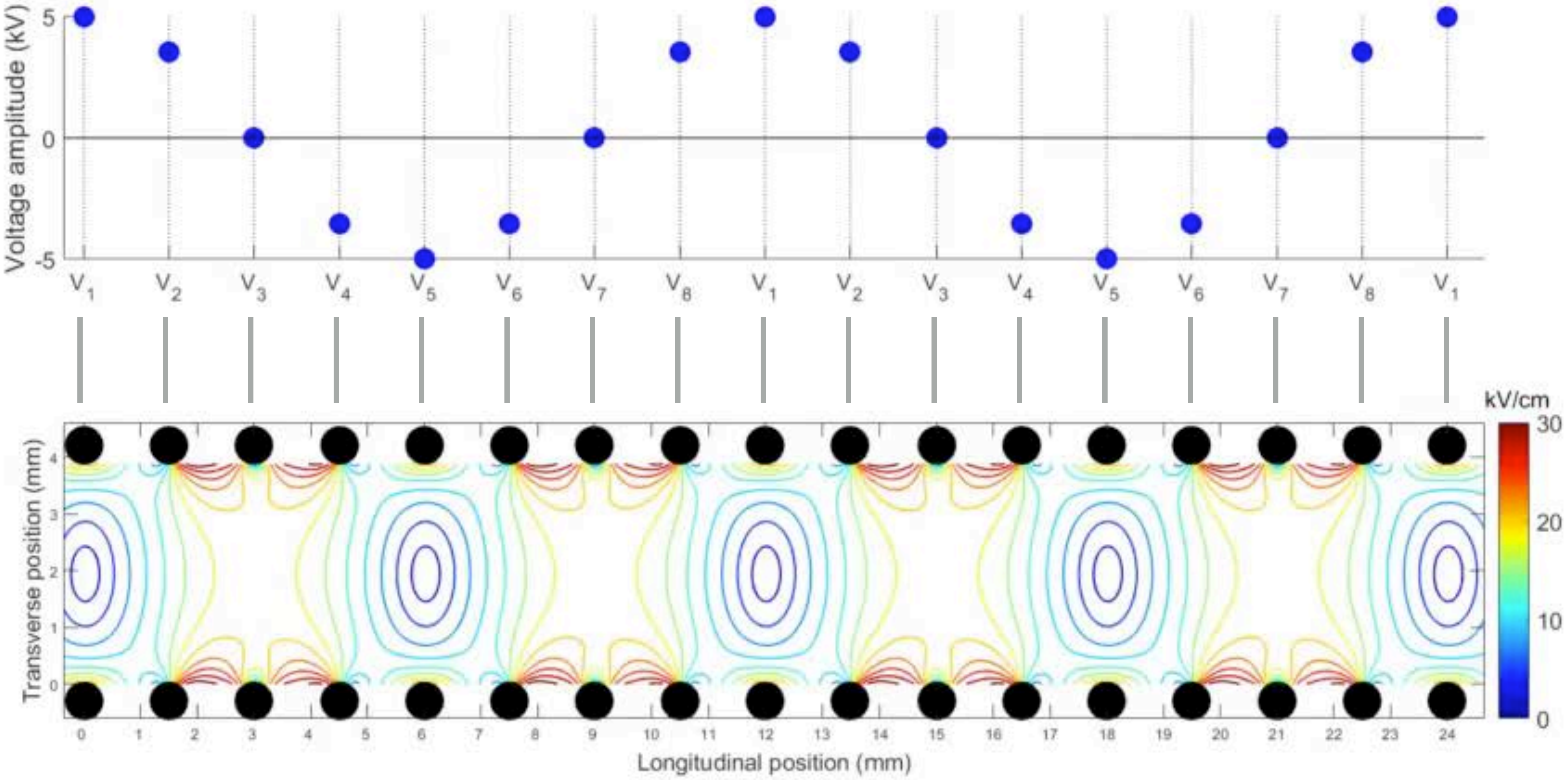
Traveling-wave decelerator: aim for factor 10 increase in interaction time



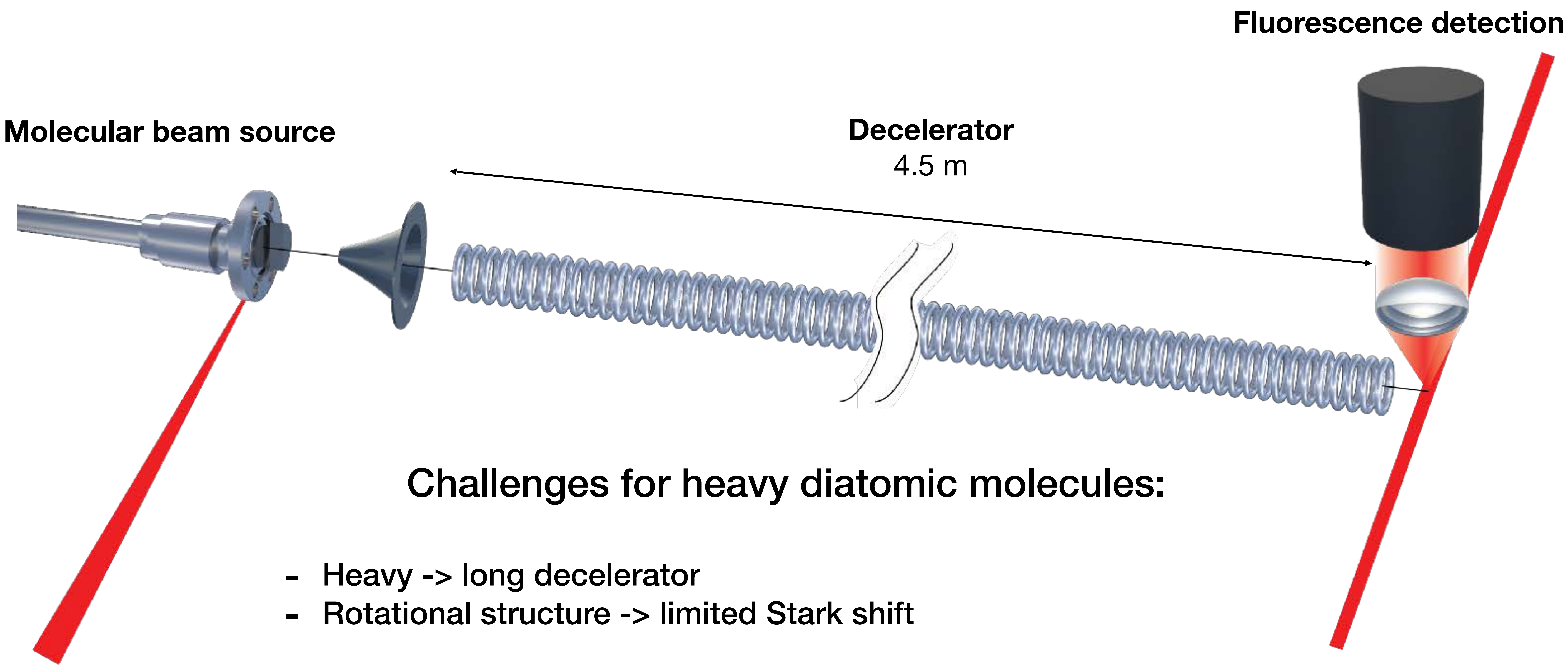
Main aims:

- 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



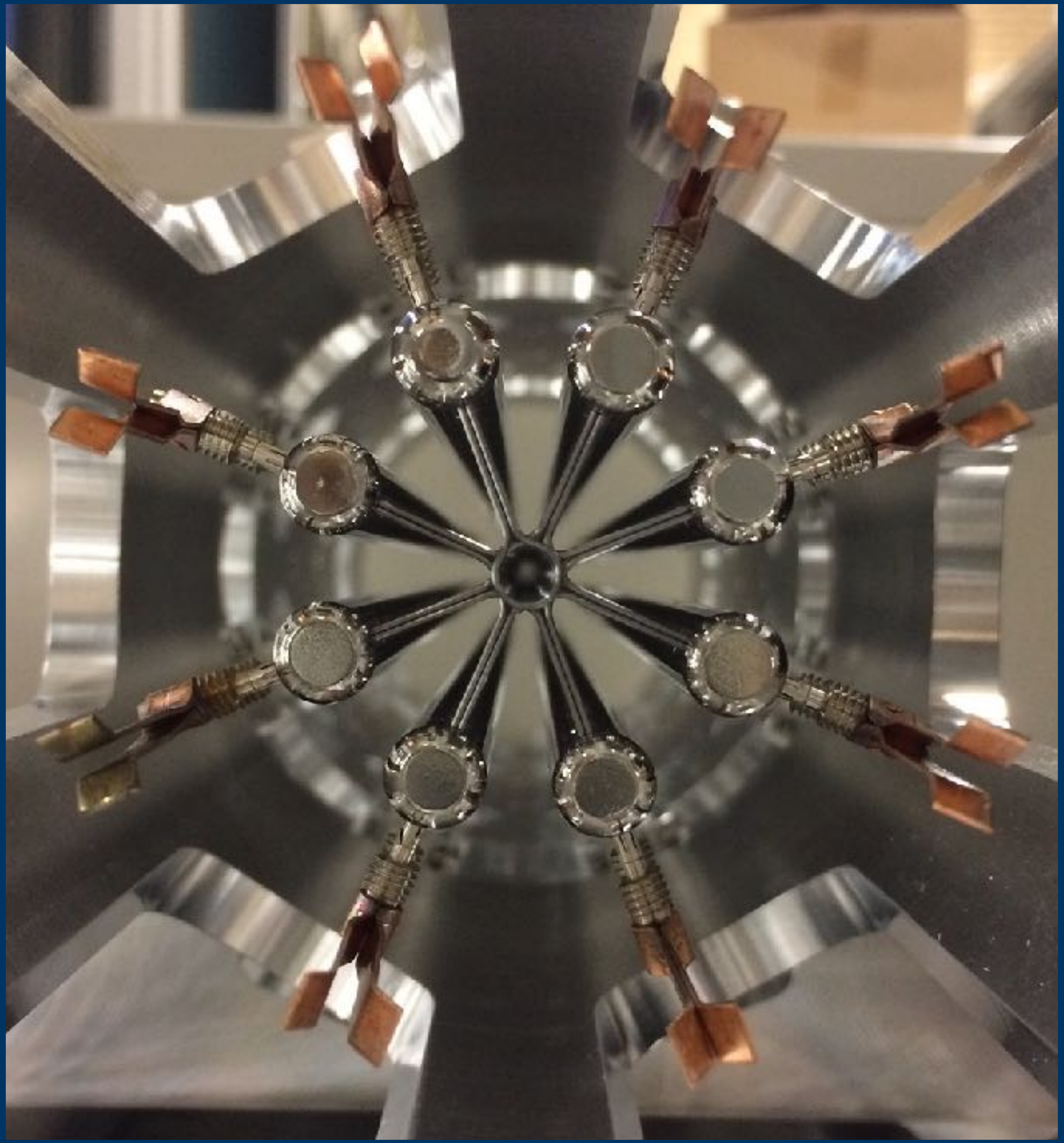
Traveling-wave decelerator



Challenges for heavy diatomic molecules:

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

Modular traveling-wave decelerator

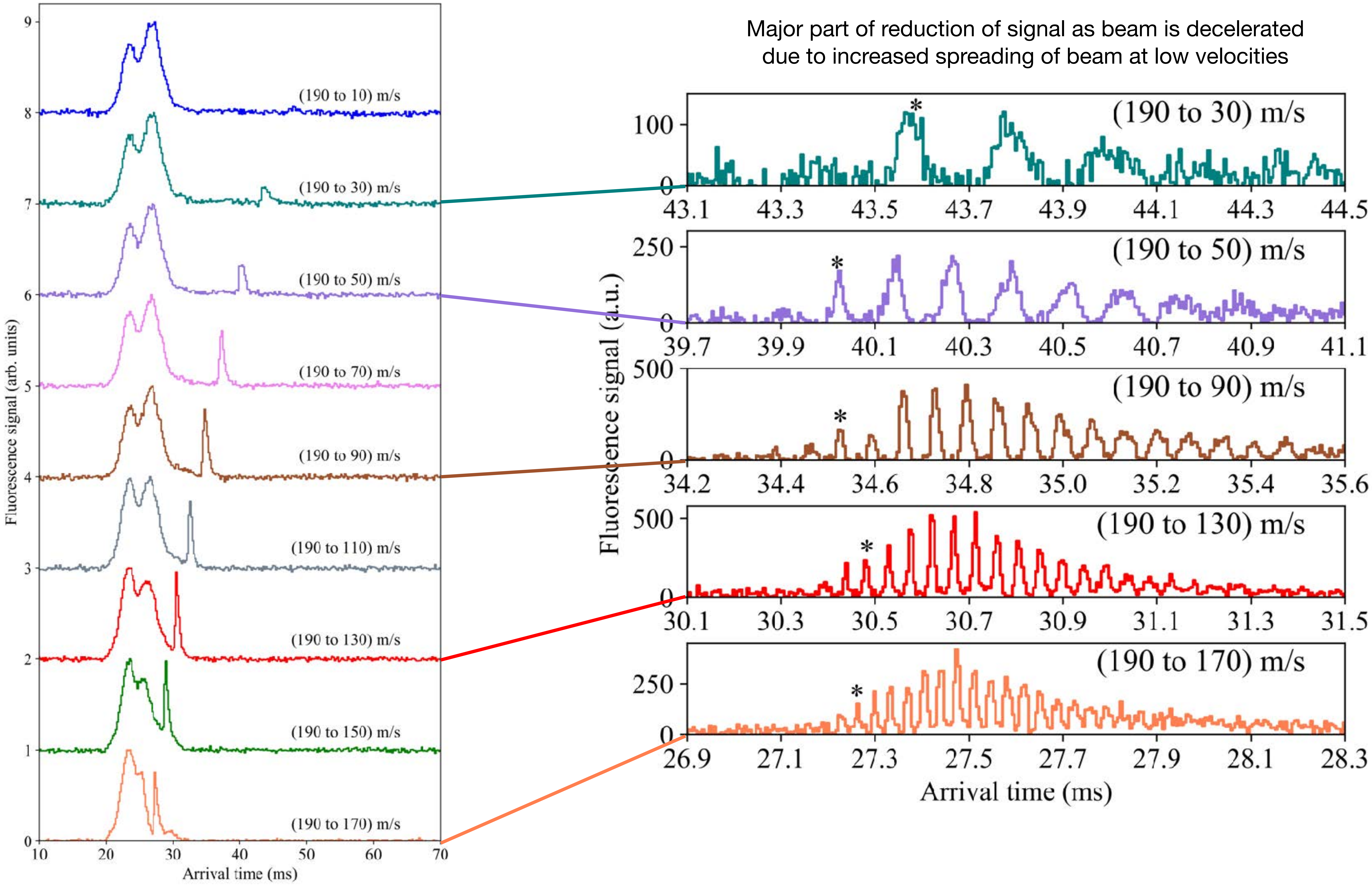


Traveling-wave decelerator

A slow beam of molecules

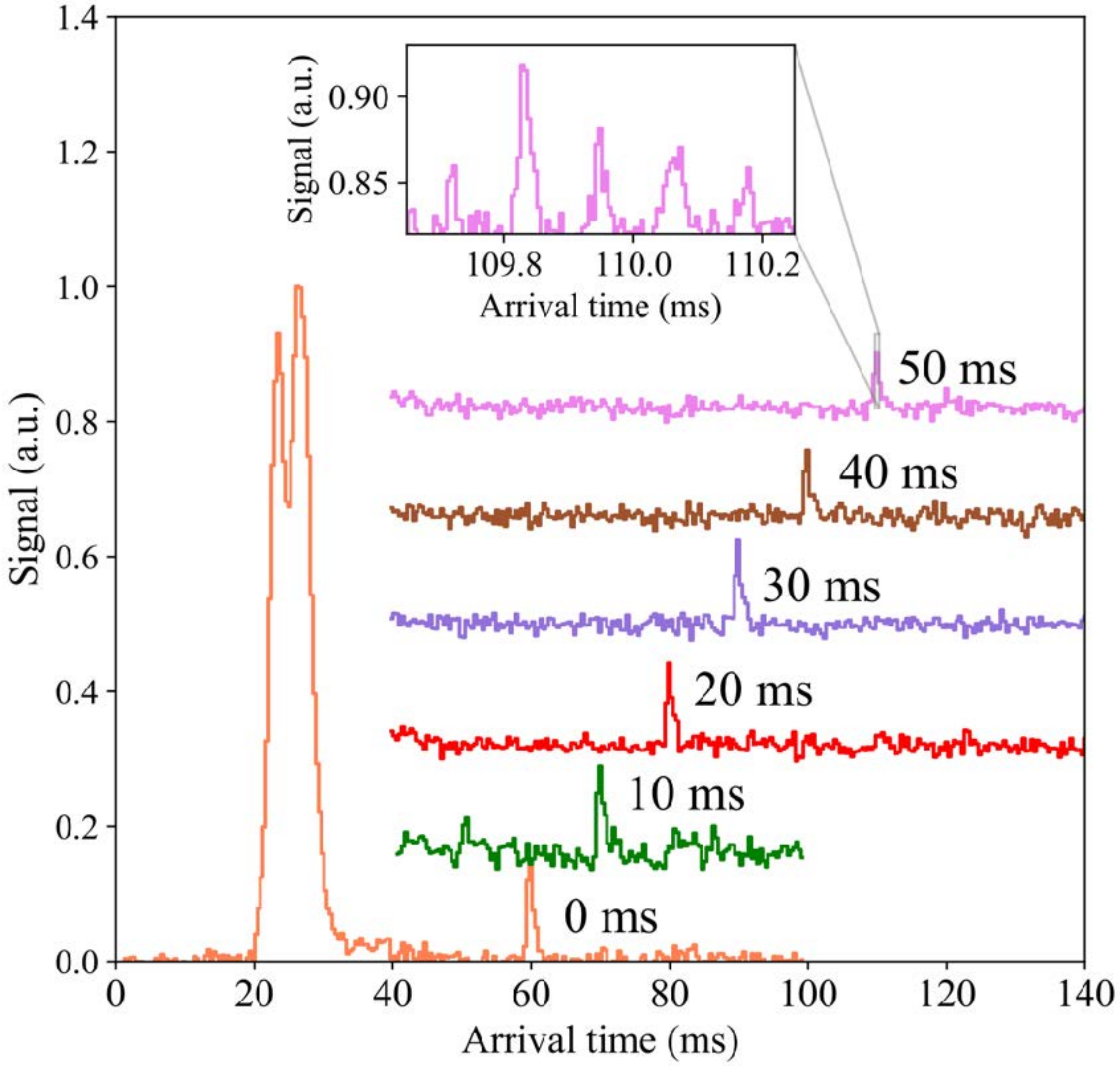
SrF: First combination of deceleration and cryogenic source

Major part of reduction of signal as beam is decelerated due to increased spreading of beam at low velocities



Deceleration to standstill

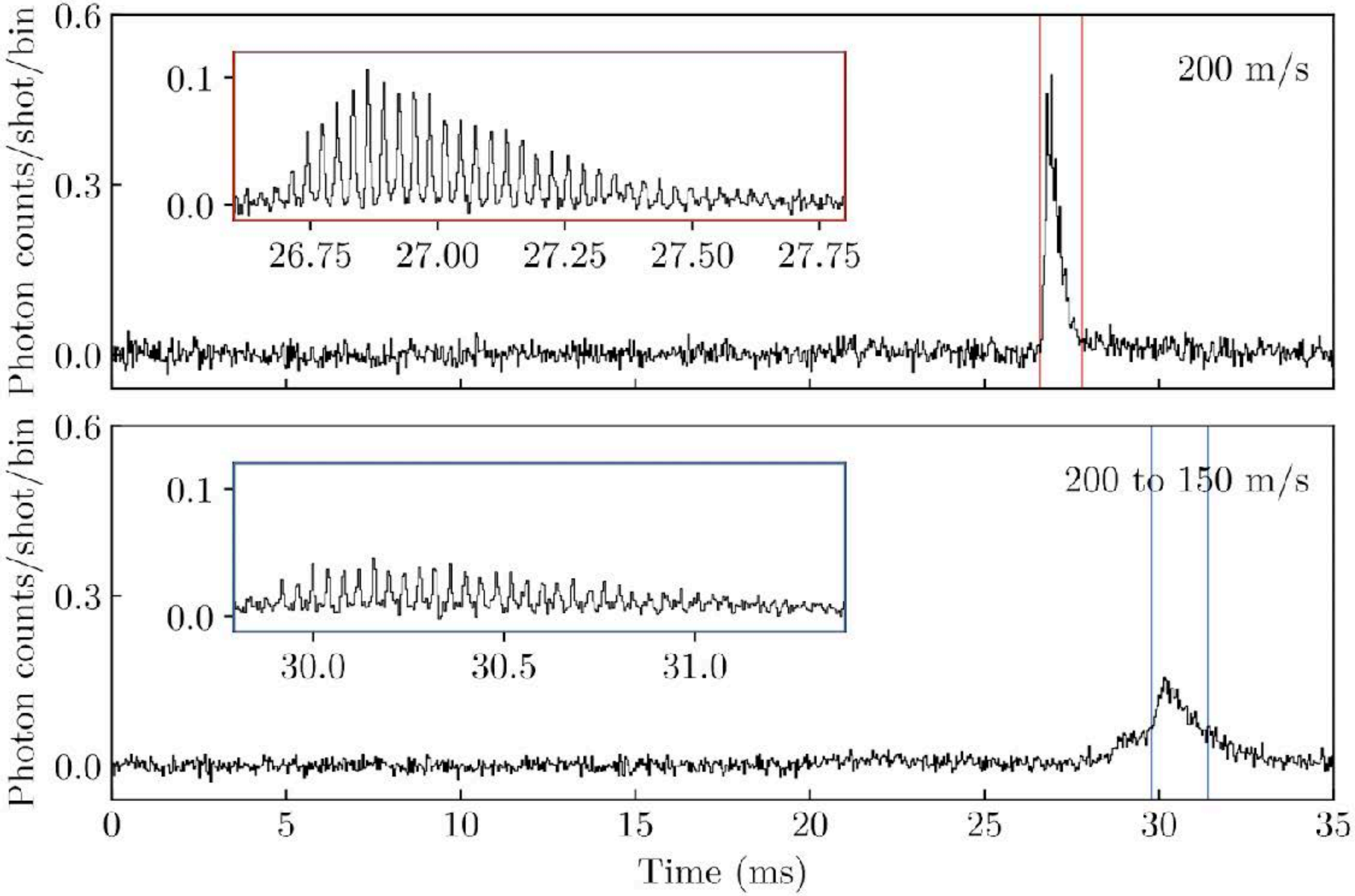
Deceleration of SrF to standstill in 4.2 m, hold there for some time, accelerate out again to 50 m/s to detect



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

Deceleration of BaF

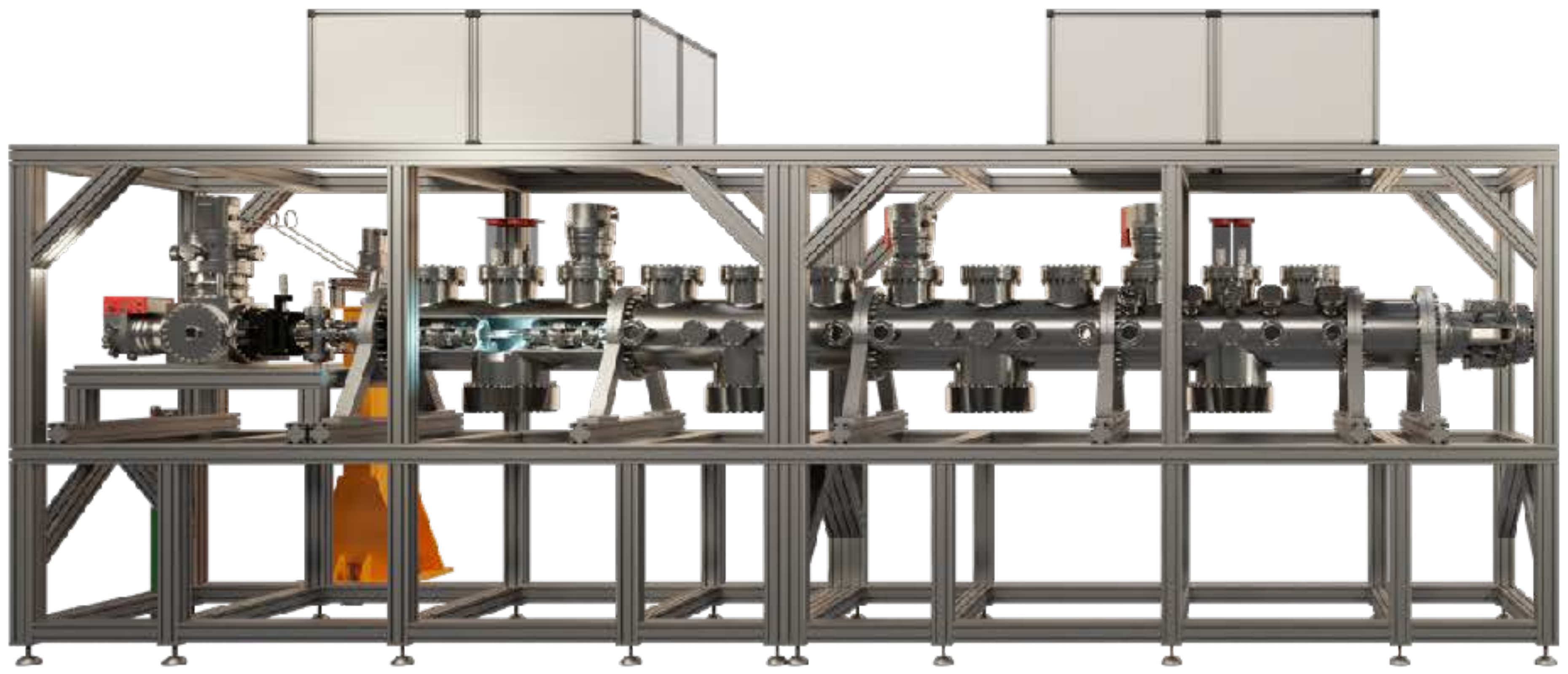
Currently upgrading electronics



Current status

Phase 2b: Slow beam

- Demonstrated first combination of cryogenic source and decelerator
- Deceleration and trapping of SrF, deceleration of BaF
- Upgrading decelerator electronics to capture more molecules from beam



Outlook: even longer interaction times

Phase 3: using trapped molecules

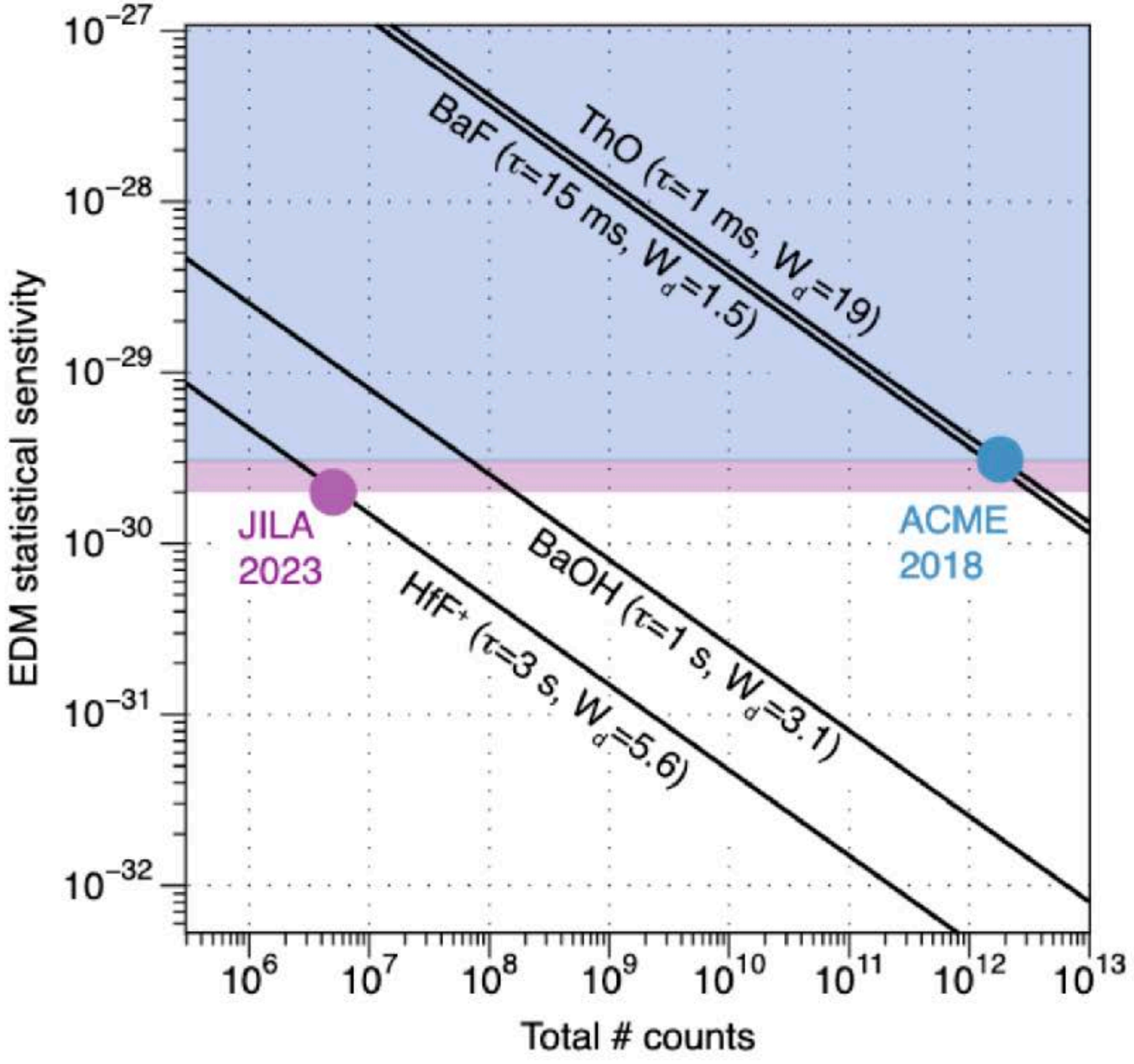
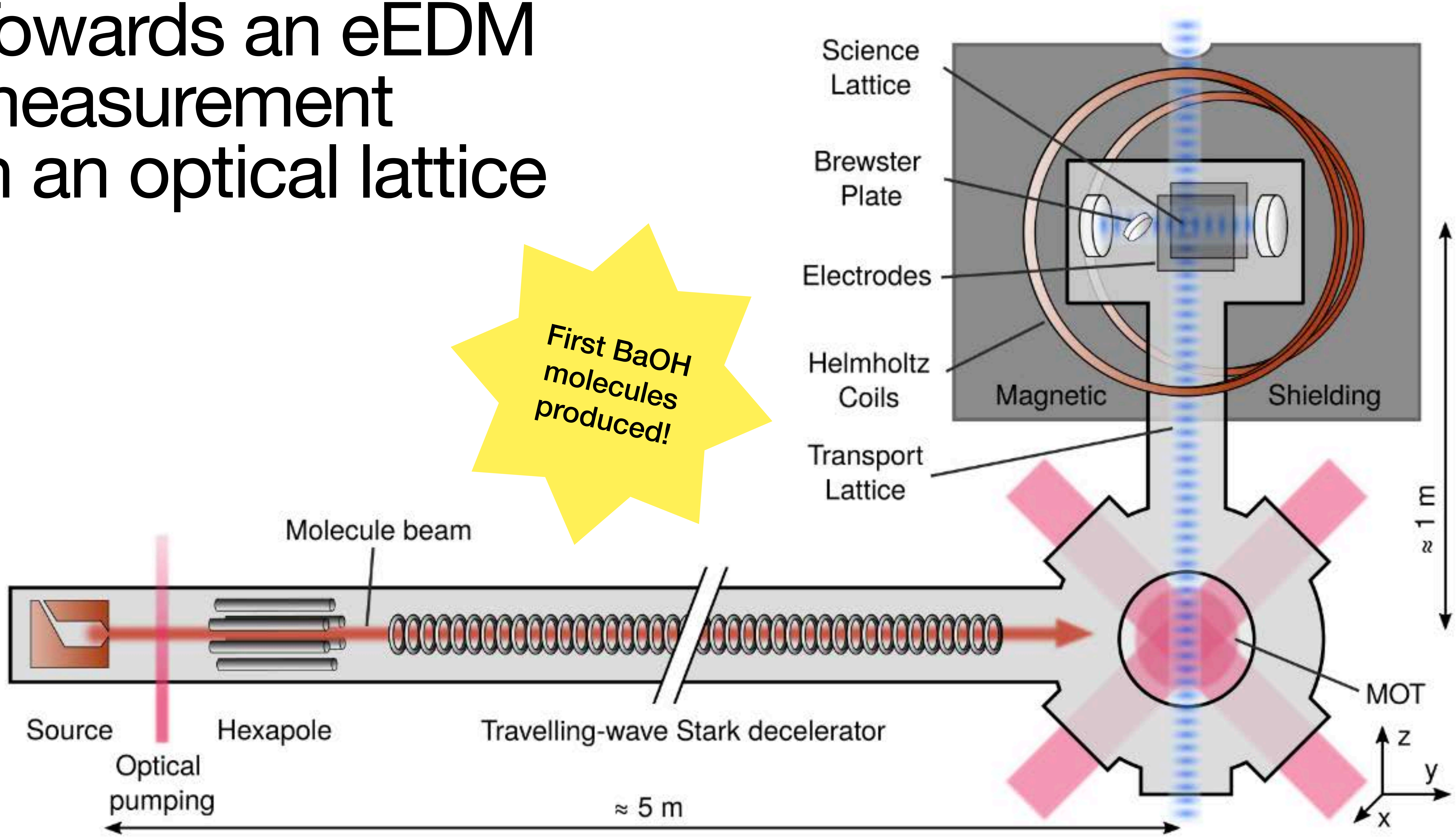


FIG. 1. Overview of statistical sensitivity of different experimental platforms. Each line corresponds to a combination of τ and $|P| \cdot W_d$ [in units of 10^{24} h Hz/(e cm)] which is typical for a given molecule species. The dots represent the two most recent experimental results [6, 7], taking into account only statistical uncertainty. The blue and purple shaded regions are excluded by the ThO and HfF^+ experiments, respectively. It can be seen that our target sensitivity of 10^{-30} e cm can be reached with $N = 6 \times 10^8$ BaOH molecules at the shot noise limit.

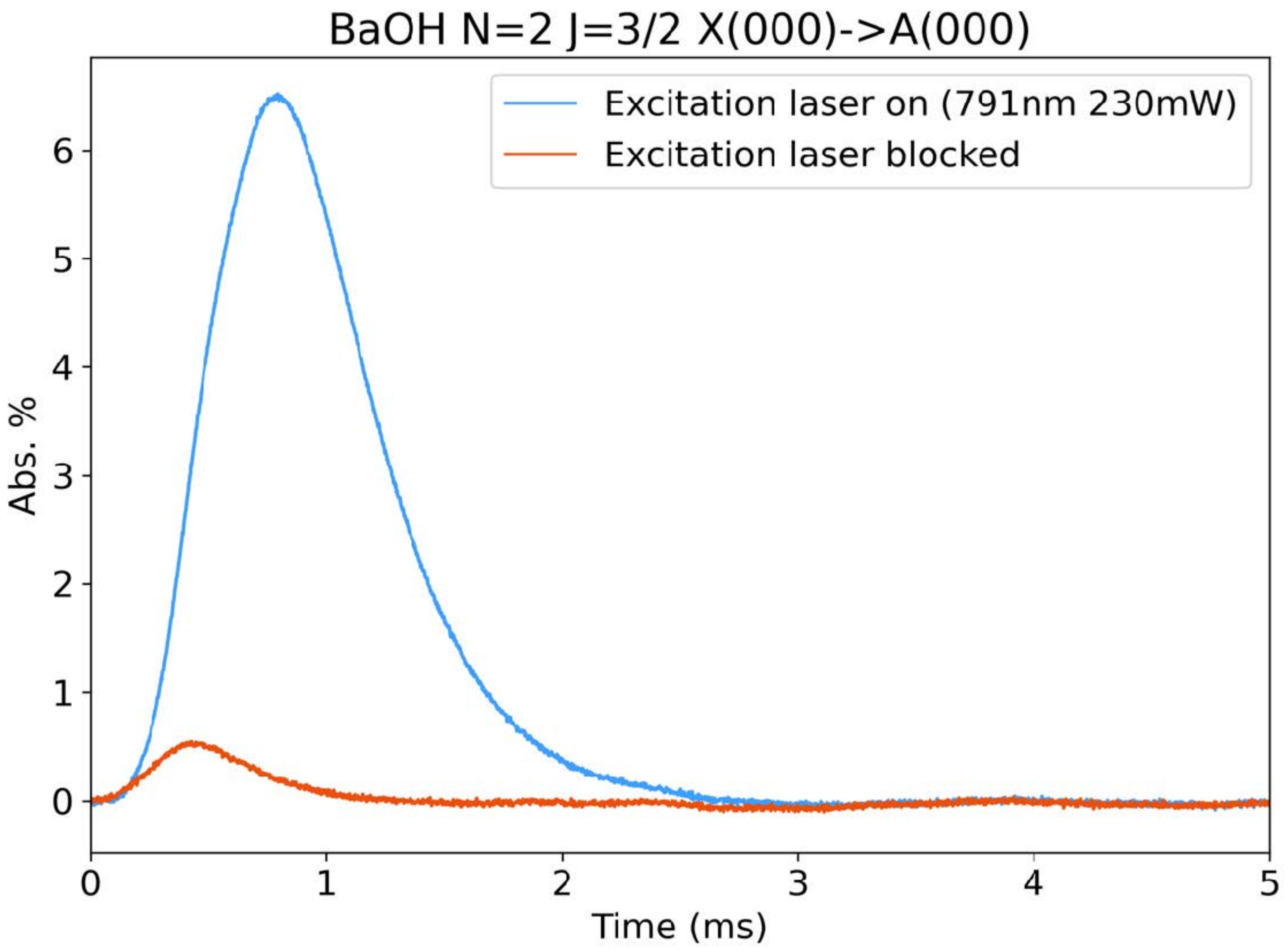
Towards an eEDM measurement in an optical lattice

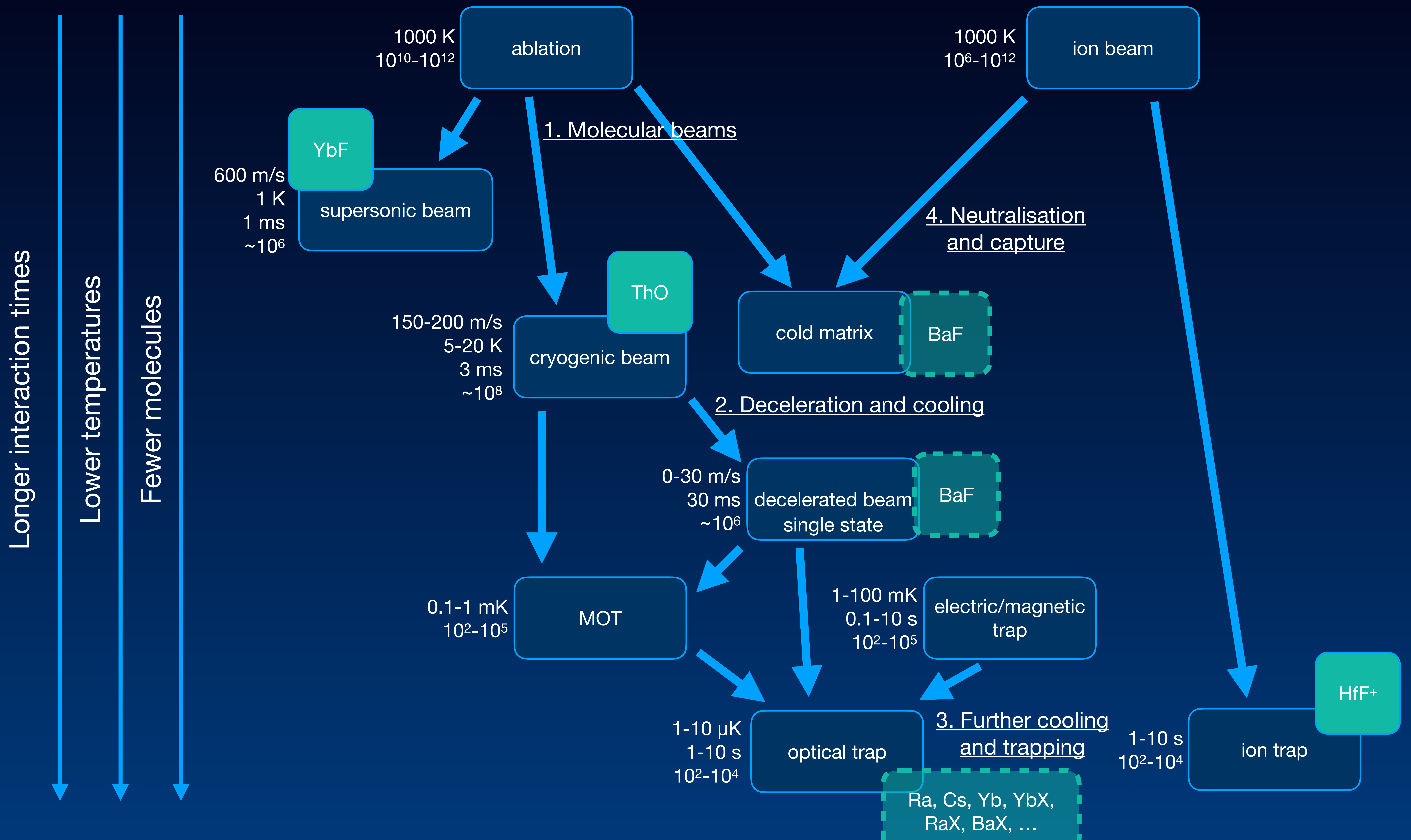
First BaOH molecules produced!



enhanced BaOH production

fresh data!



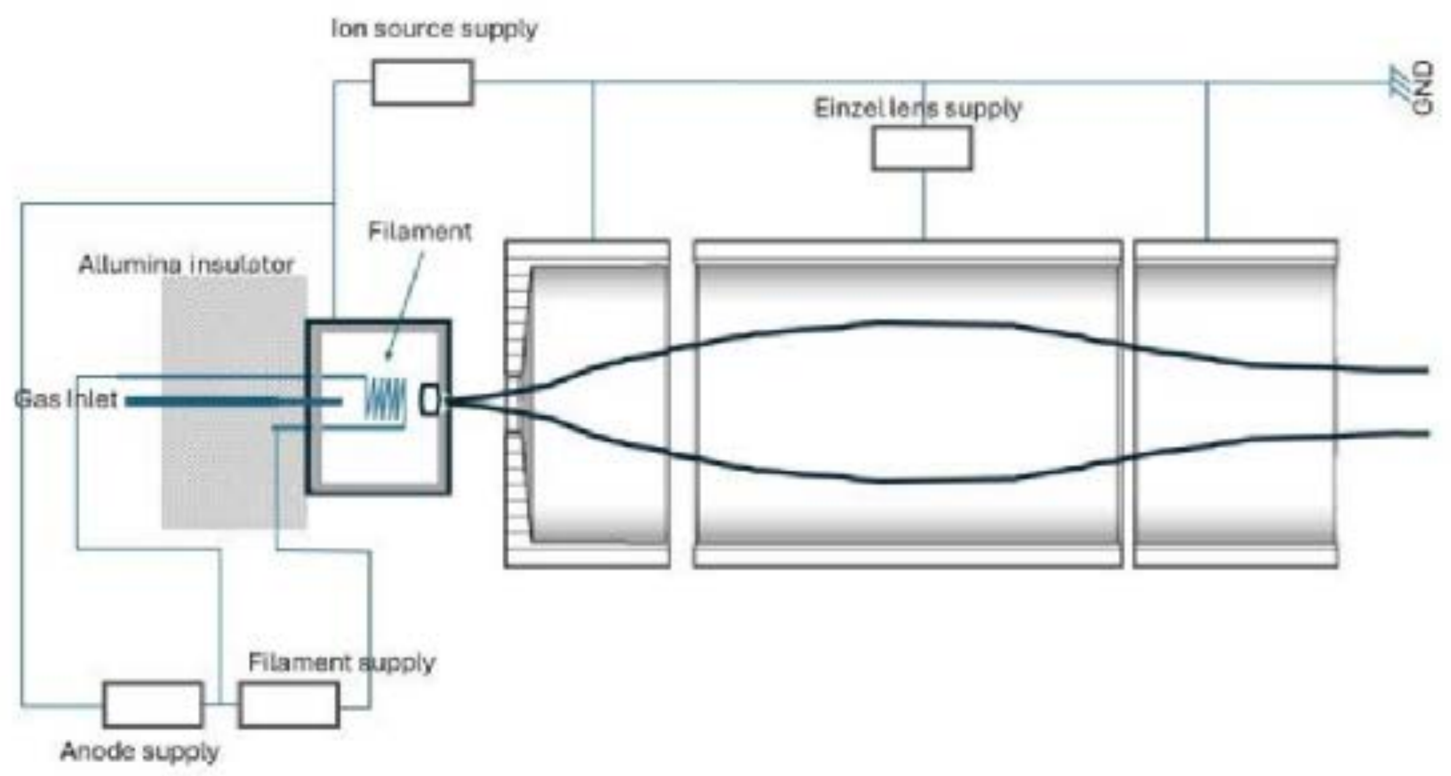


Longer interaction times
Lower temperatures
Fewer molecules
1000 K
10^{10}-10^{12}
ablation
1000 K
10^{6}-10^{12}
ion beam
1. Molecular beams
YbF
600 m/s
1 K
1 ms
~10^{6}
supersonic beam
4. Neutralisation and capture
ThO
150-200 m/s
5-20 K
3 ms
~10^{8}
cryogenic beam
cold matrix
BaF
2. Deceleration and cooling
0-30 m/s
30 ms
~10^{6}
decelerated beam single state
BaF
0.1-1 mK
10^{2}-10^{5}
MOT
1-100 mK
0.1-10 s
10^{2}-10^{5}
electric/magnetic trap
1-10 μK
1-10 s
10^{2}-10^{4}
optical trap
3. Further cooling and trapping
Ra, Cs, Yb, YbX, RaX, BaX, …
HfF+
1-10 s
10^{2}-10^{4}
ion trap

Versatile cold matrix source

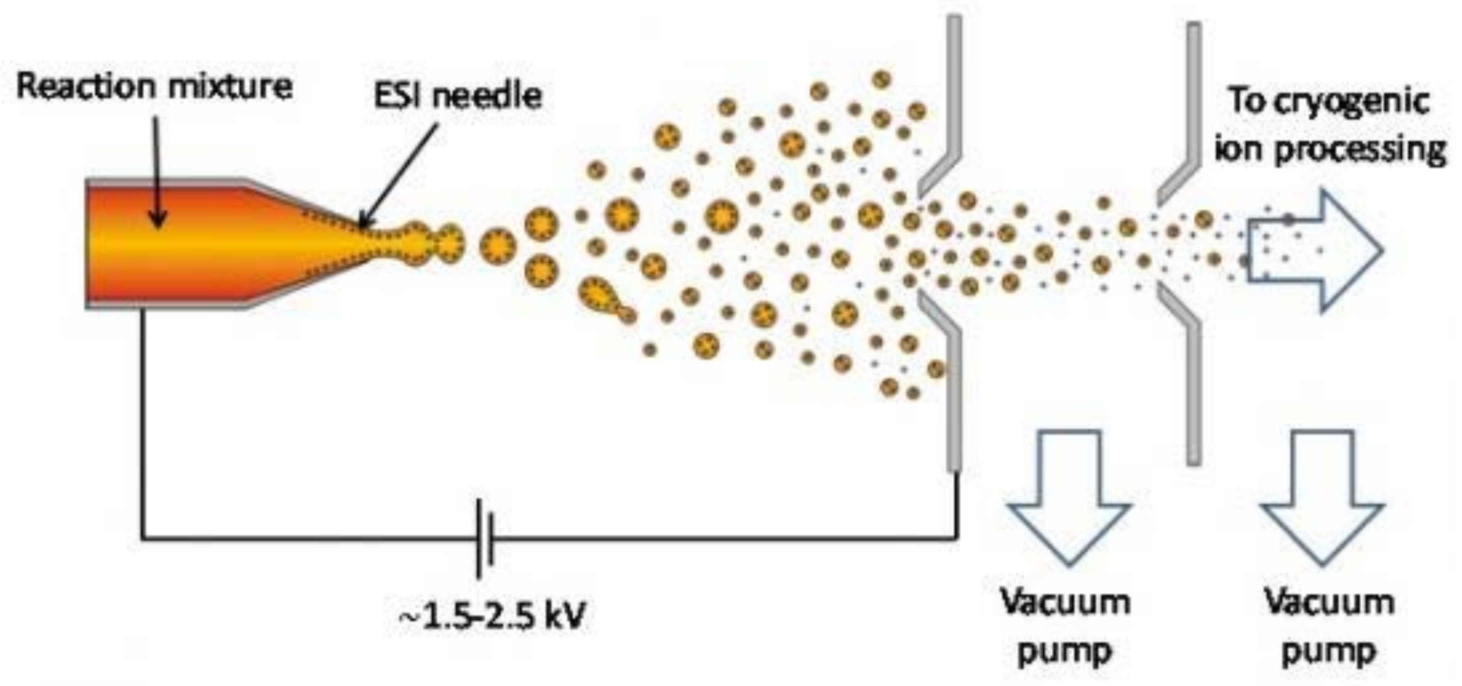
developing with Giovanni Carugno, Daniel Comparat, Hans Lignier

glow discharge,
Intense BaF+ ion beam
few μA @ 1 keV
20 nA @ 5 eV



or

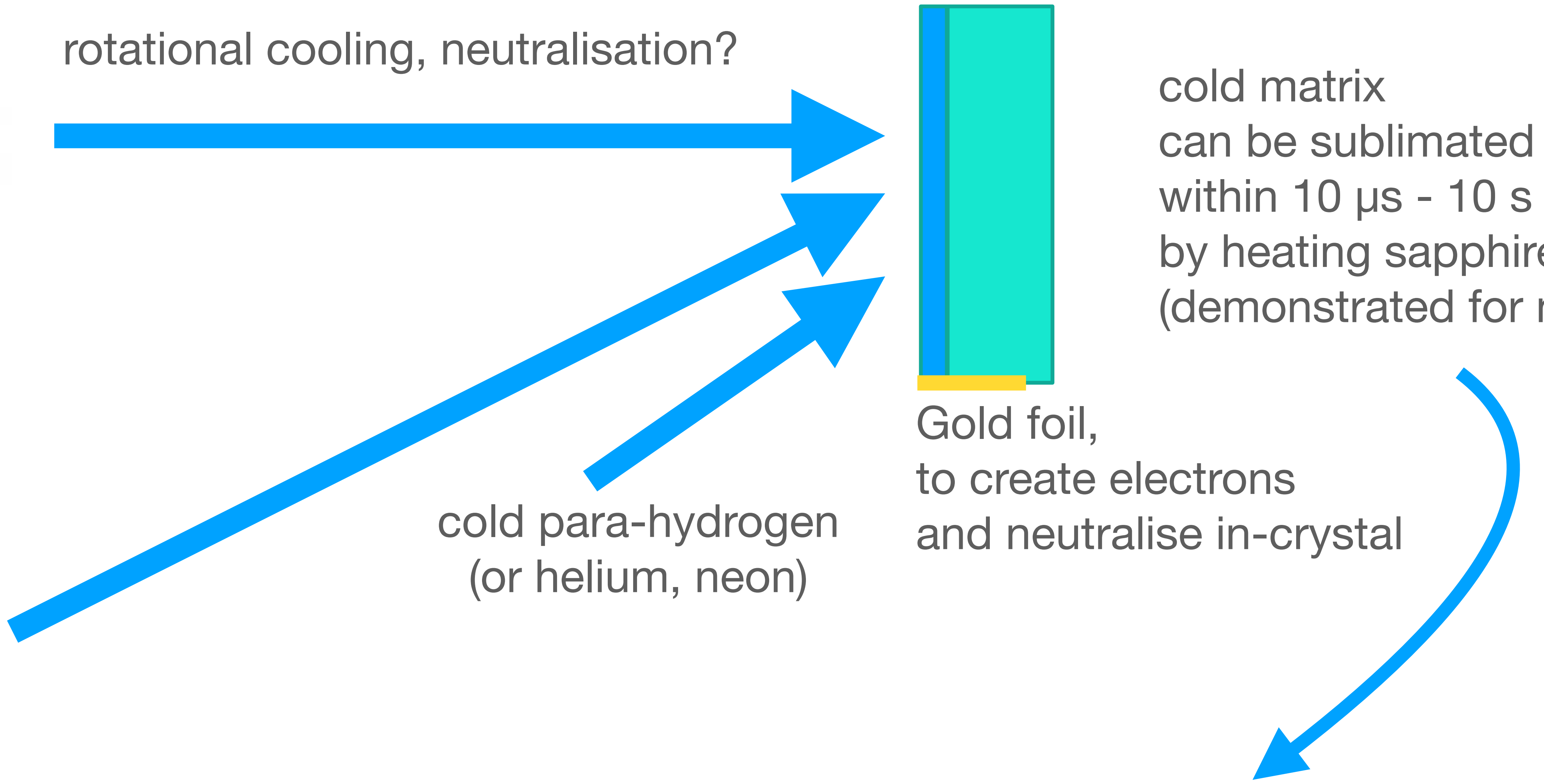
Electrospray
for radioactive
molecules?



(image: Johnson lab, Yale)

or

Neutral molecule beam?

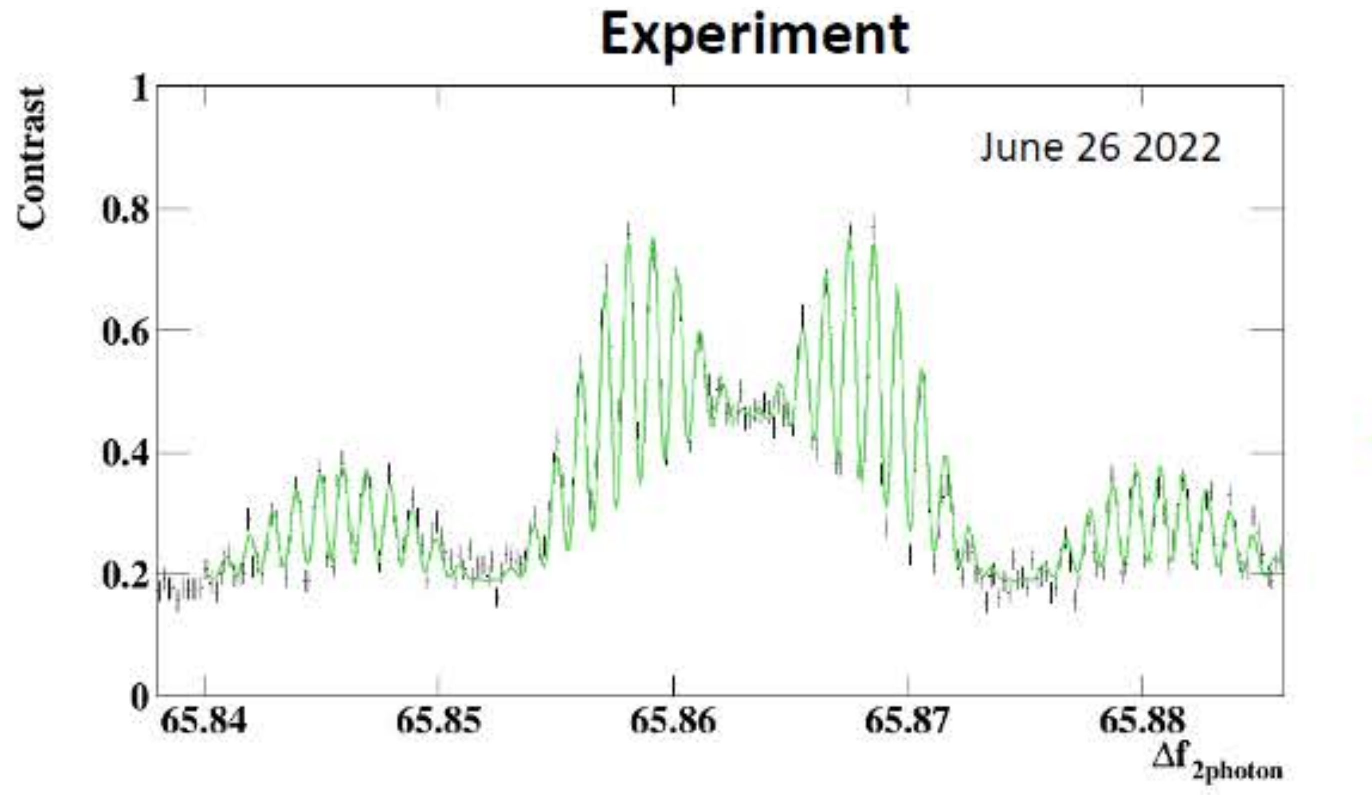


cold matrix
can be sublimated
within 10 μs - 10 s
by heating sapphire crystal
(demonstrated for neon)

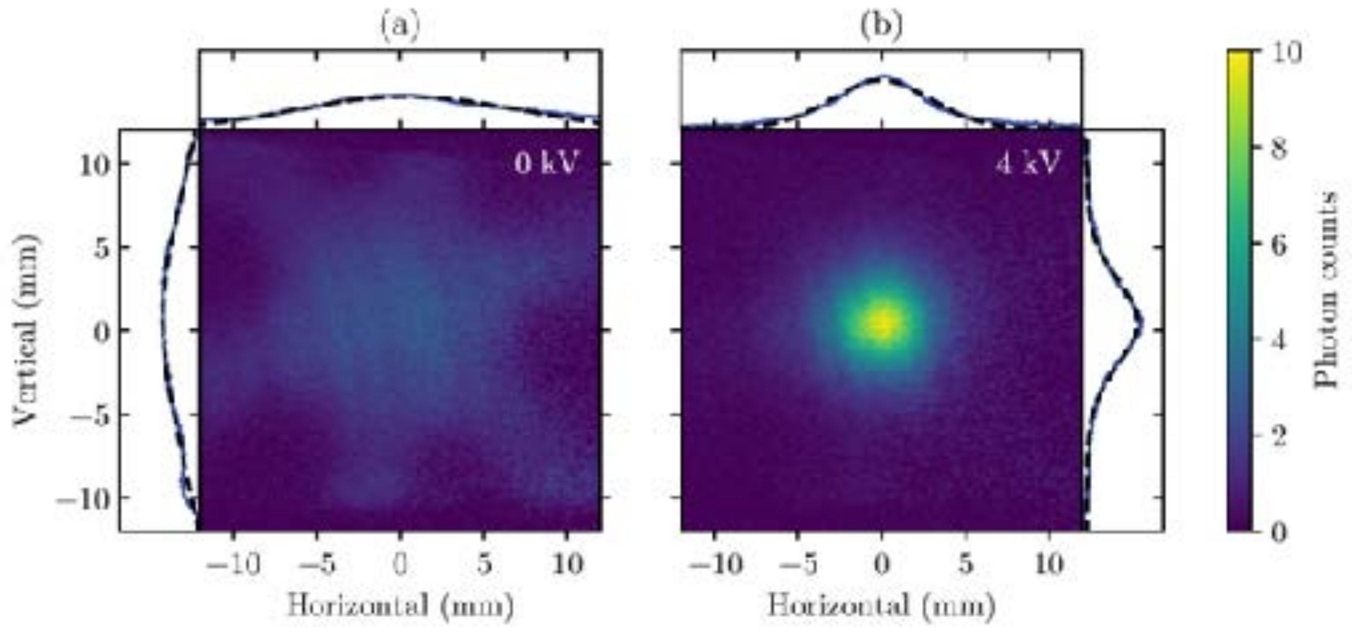
Ideal source for intense low temperature samples
Good starting point for cooling and trapping

Borghesani, A. F., Carugno, G., Messineo, G. & Pazzini, J. Electron thermalization length in solid para-hydrogen at low-temperature. J. Chem. Phys. 159, 104501 (2023).
Courageux, T., Cournol, A., Comparat, D., Lesegno, B. V. de & Lignier, H. Efficient rotational cooling of a cold beam of barium monofluoride. New J Phys 24, 025007 (2022).
Sacramento, R. L. et al. Matrix isolation sublimation: An apparatus for producing cryogenic beams of atoms and molecules. Rev. Sci. Instrum. 86, 073109 (2015).

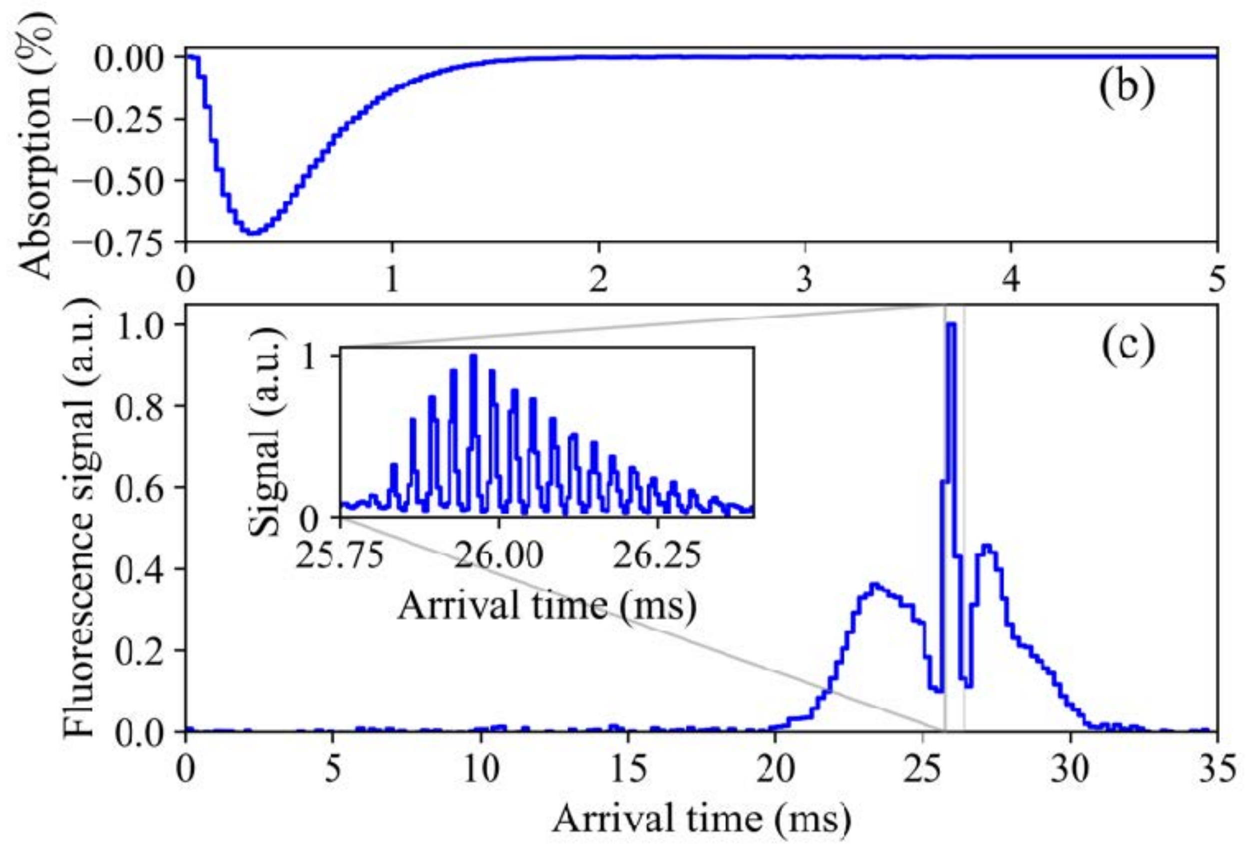
Summary



Spin interference demonstrated and understood



Intense and bright slow beam



Deceleration demonstrated

(part of) the team

positions available

university of groningen
van swinderen institute for particle physics and gravity

Nikhef
Dutch National Institute for (astro)Particle Physics

VU VRIJE UNIVERSITEIT AMSTERDAM

UvA