Towards quantum logic spectroscopy of polyatomic molecular ions

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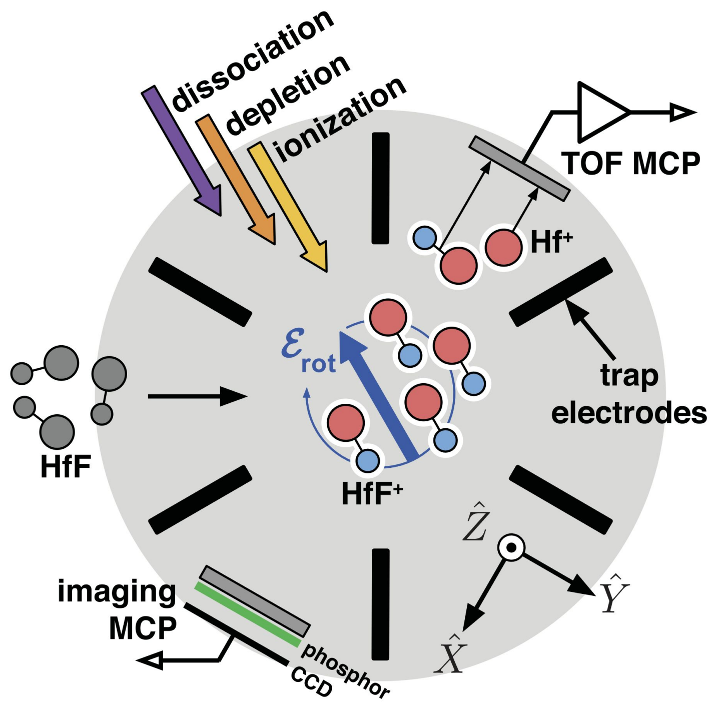
Ultracold molecular ions

Cold molecules have numerous attractive applications:

- Molecular clocks
- Molecular qubits [1], qudits
- Tests of fundamental theories [2]
  - search of electron EDM
  - parity violation theories
  - drift of fundamental constants
- Verification of ab-initio calculations of molecular energy level structures
- Ultracold chemistry [3]



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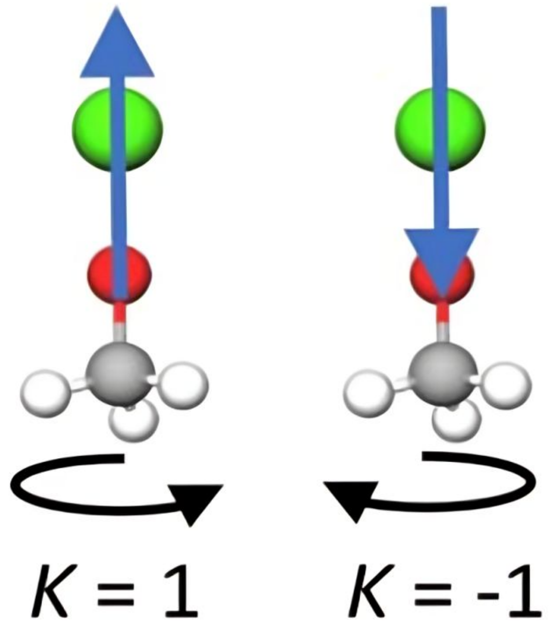
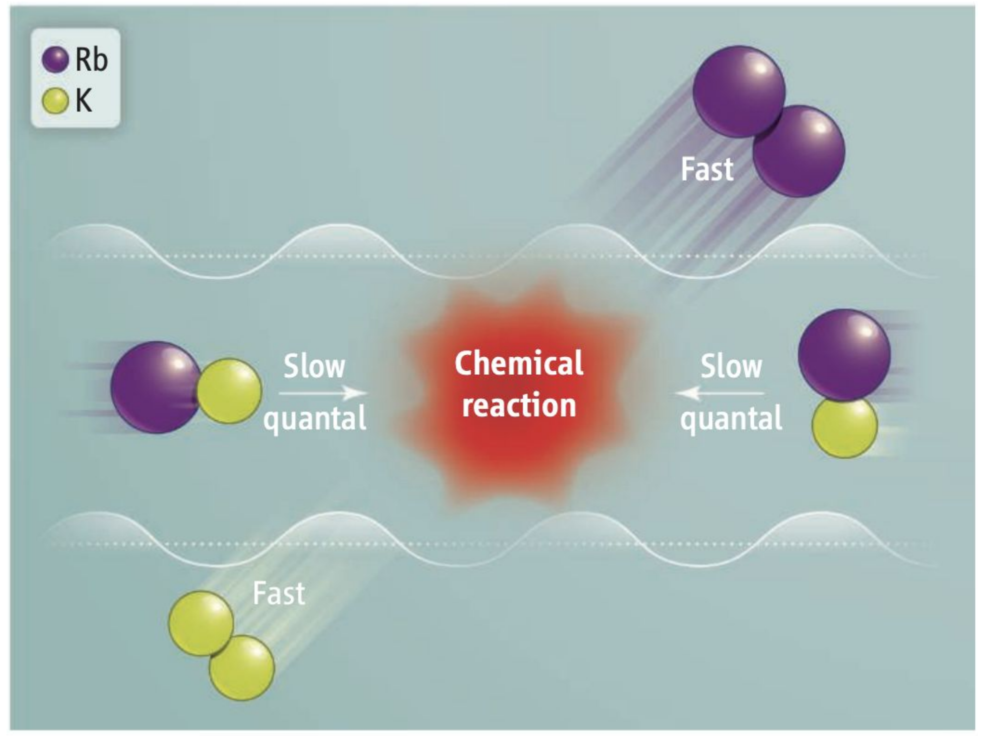


[1]



[3]

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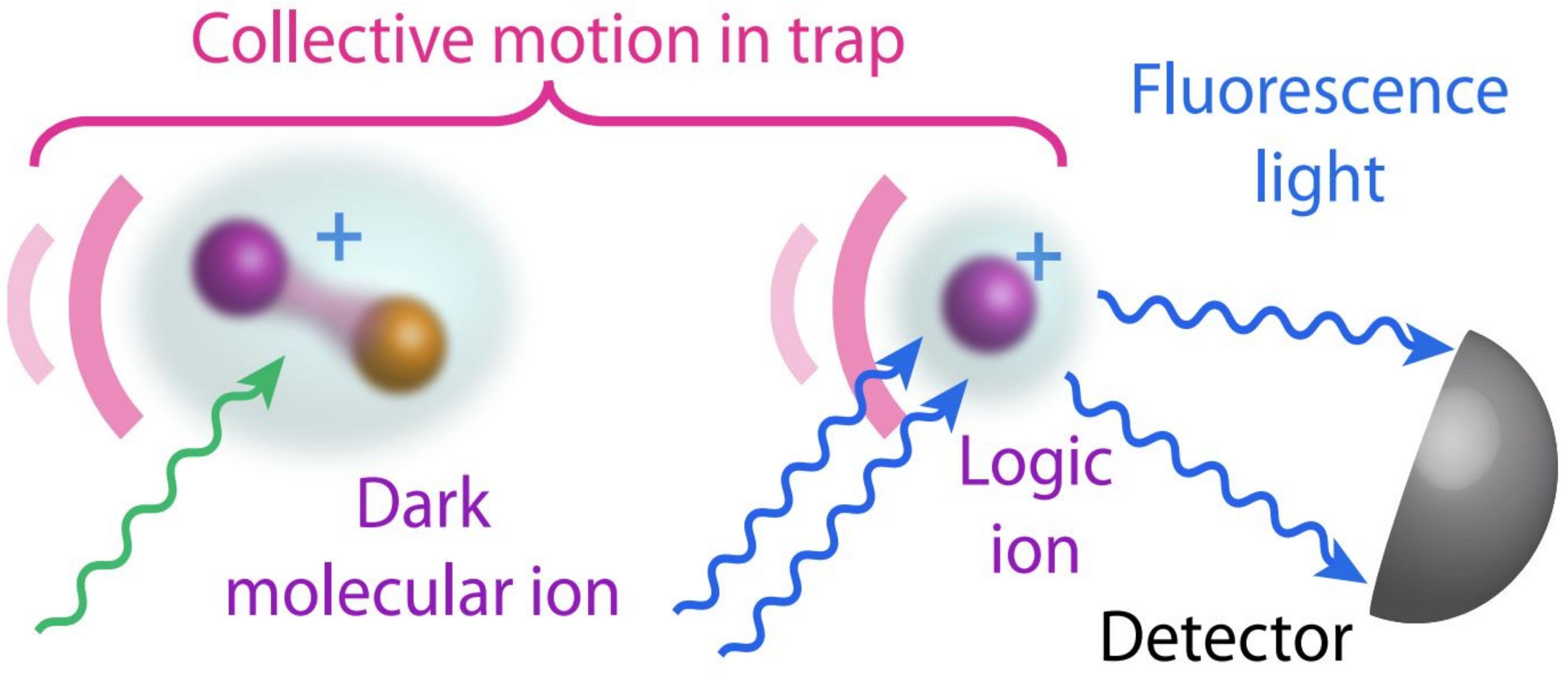
[1] Yu, Phelan, et al. "A scalable quantum computing platform using symmetric-top molecules." *New Journal of Physics* 21.9 (2019): 093049.
[2] Cairncross, William B., et al. "Precision measurement of the electron's electric dipole moment using trapped molecular ions." *Physical review letters* 119.15 (2017): 153001.
[3] Hutson, Jeremy M. "Ultracold chemistry." *Science* 327.5967 (2010): 788-789.

Quantum logic spectroscopy

Control of molecular quantum state possess a serious challenge due to lack of closed optical cycling transitions.

The problem of molecular state control and detection can be solved with a quantum logic spectroscopy (QLS)[1, 2]:

- Molecular (*spectroscopic*) ion's state is mapped to its motional state
- Co-trapped atomic (*logic*) ion is used to read out joint motional state and therefore, state of the molecule.



[3]

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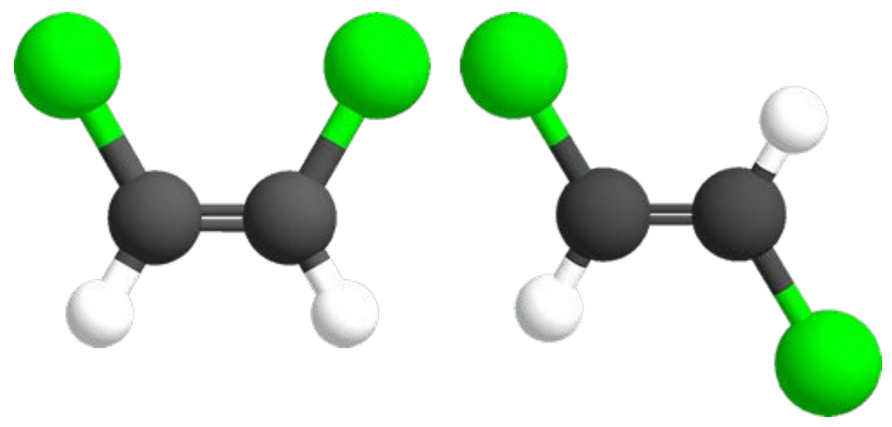
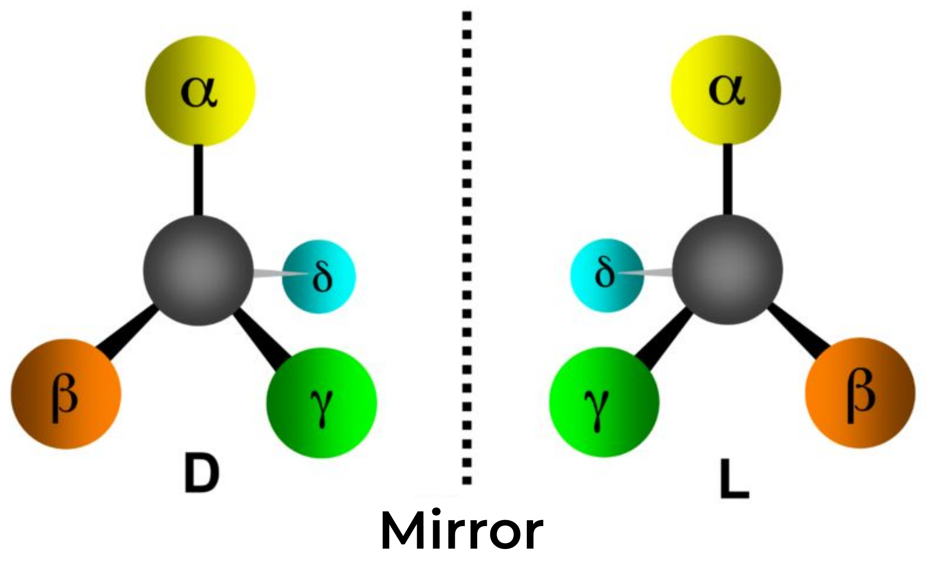
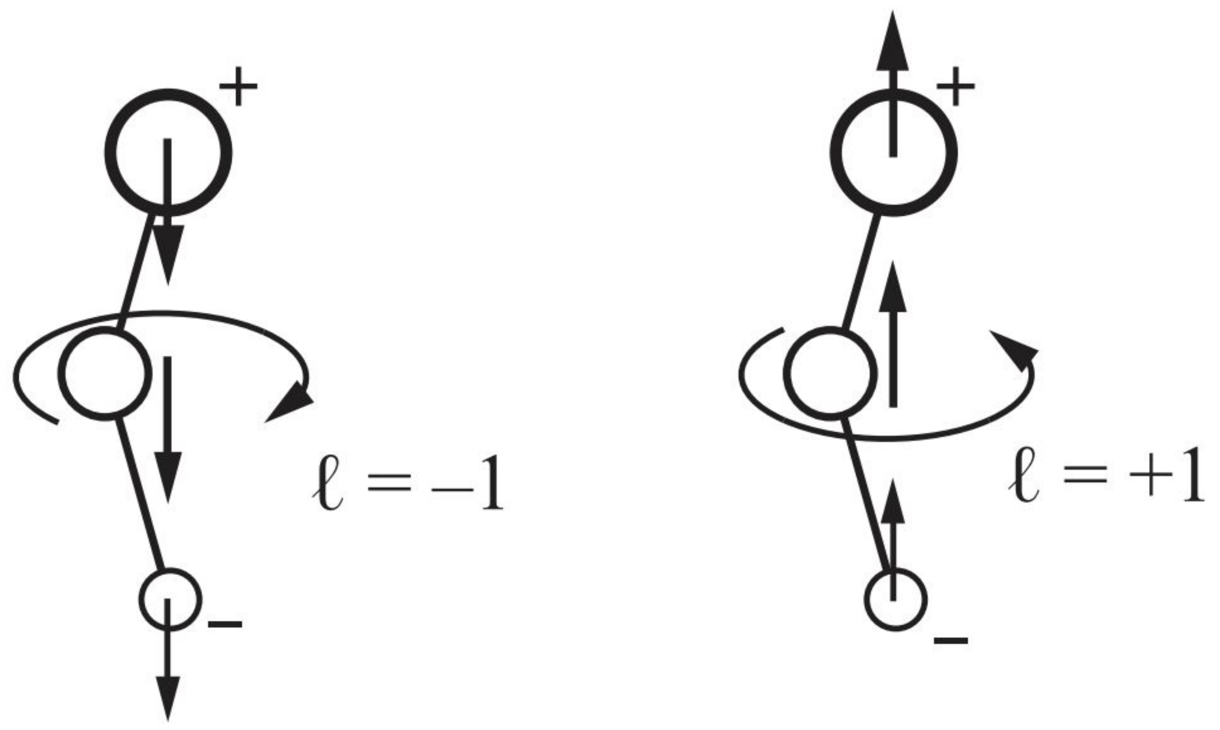
[1] Schmidt, P. Oetal, et al. "Spectroscopy using quantum logic." *Science* 309.5735 (2005): 749-752.
[2] Sinhal, Mudit, and Stefan Willitsch. "Molecular-Ion Quantum Technologies." *Photonic Quantum Technologies: Science and Applications* 1 (2023): 305-332.
[3] Deiß, Markus, Stefan Willitsch, and Johannes Hecker Denschlag. "Cold trapped molecular ions and hybrid platforms for ions and neutral particles." *Nature Physics* (2024): 1-9.

Polyatomic molecules







So far QLS techniques were demonstrated only for diatomic molecules.

Polyatomic molecules offer a new set of properties:

- isomerism
- chirality
- parity doublet states

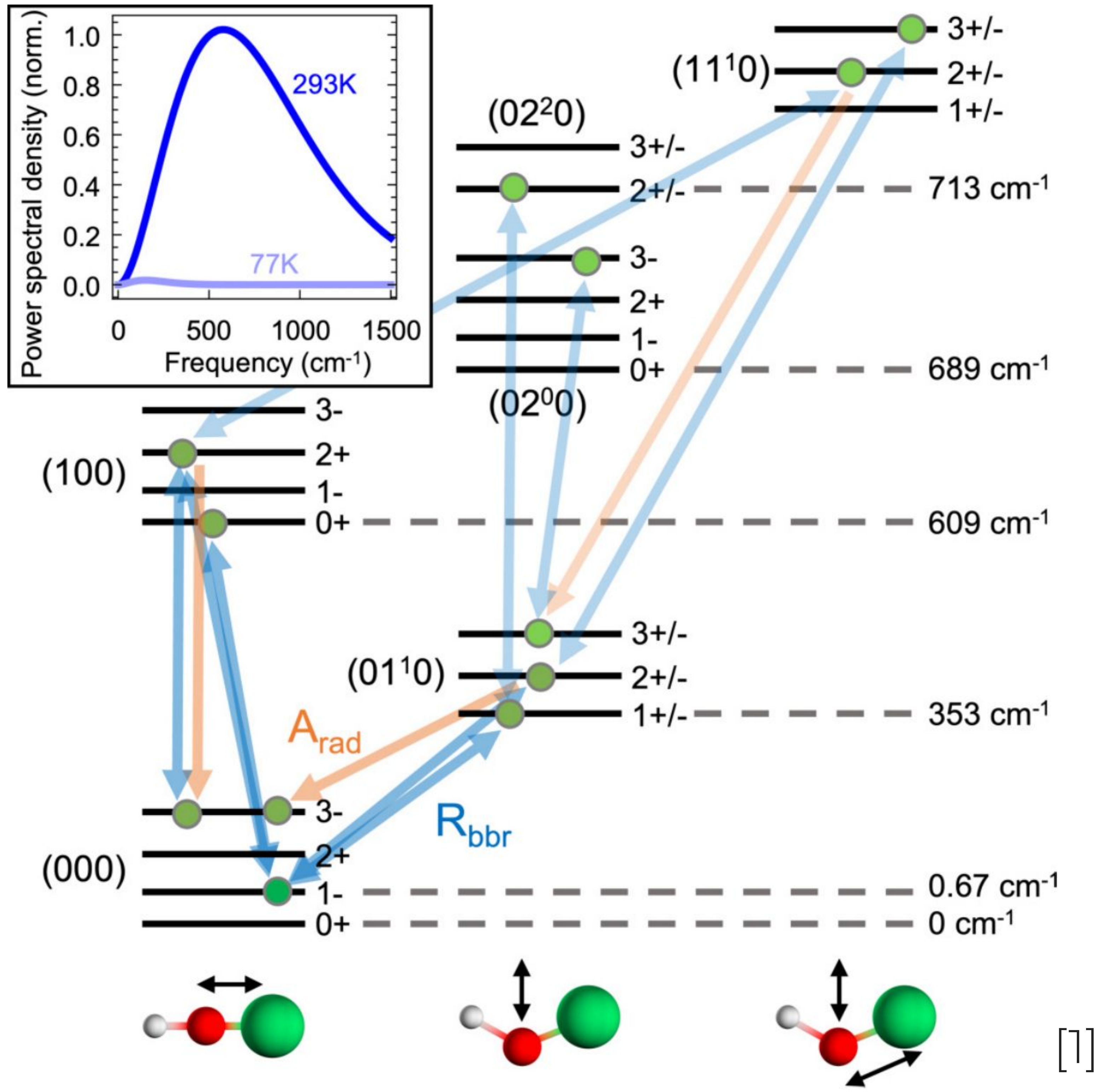
Polyatomic molecules: challenges

Moving to polyatomic molecules possess additional challenges:

- Complex and dense energy level spectrum
- Excitation of rovibrational states by a black body radiation on a timescale of few seconds[1]
- Polyatomic molecular ions are highly reactive

A cryogenic environment is required to preserve prepared rovibrational state during experimentally relevant times.

Rates of collisions with background gas are strongly reduced due to cryopumping.



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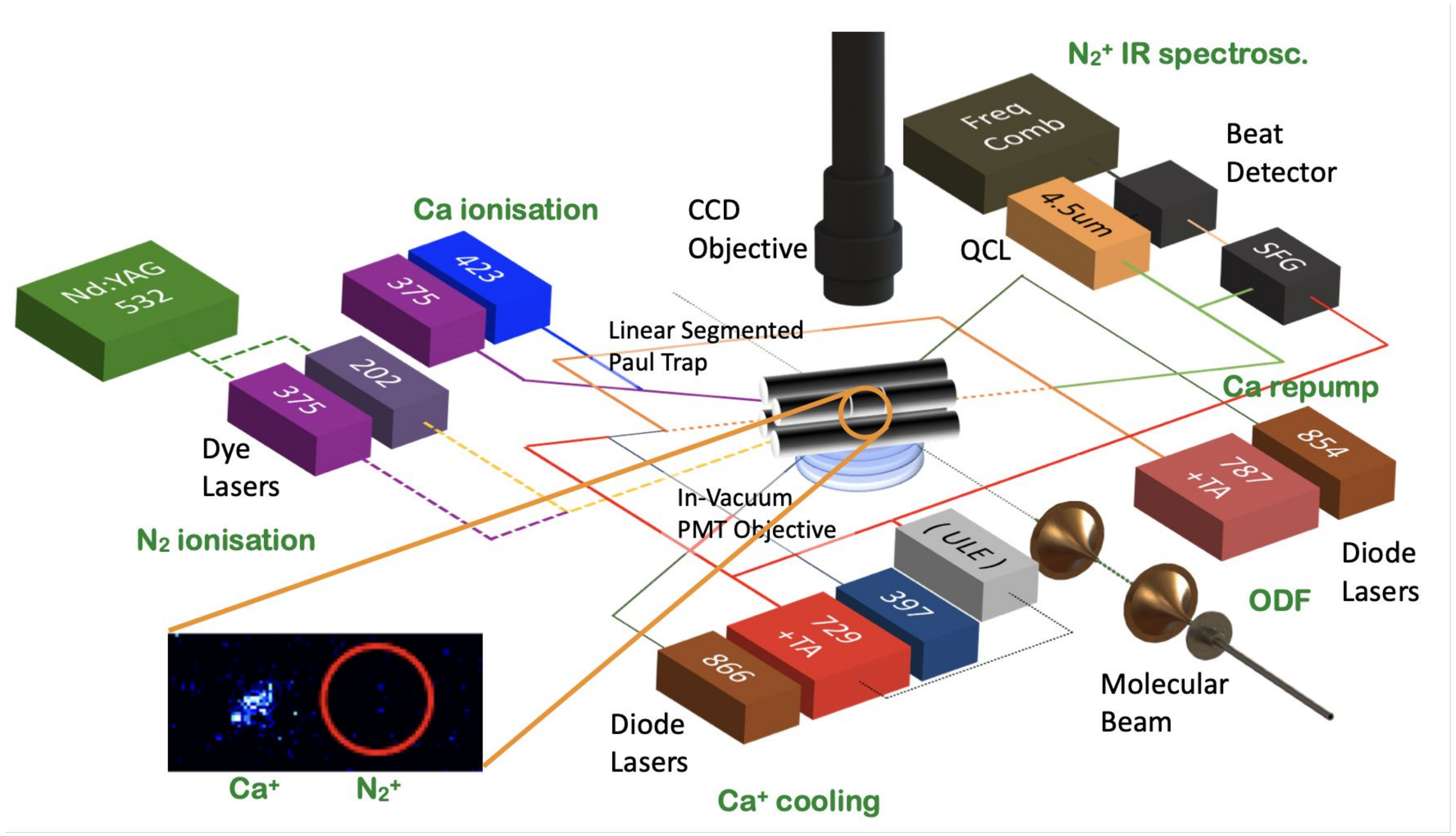
[1] Vilas, Nathaniel B., et al. "Blackbody thermalization and vibrational lifetimes of trapped polyatomic molecules." *Physical Review A* 107.6 (2023): 062802.

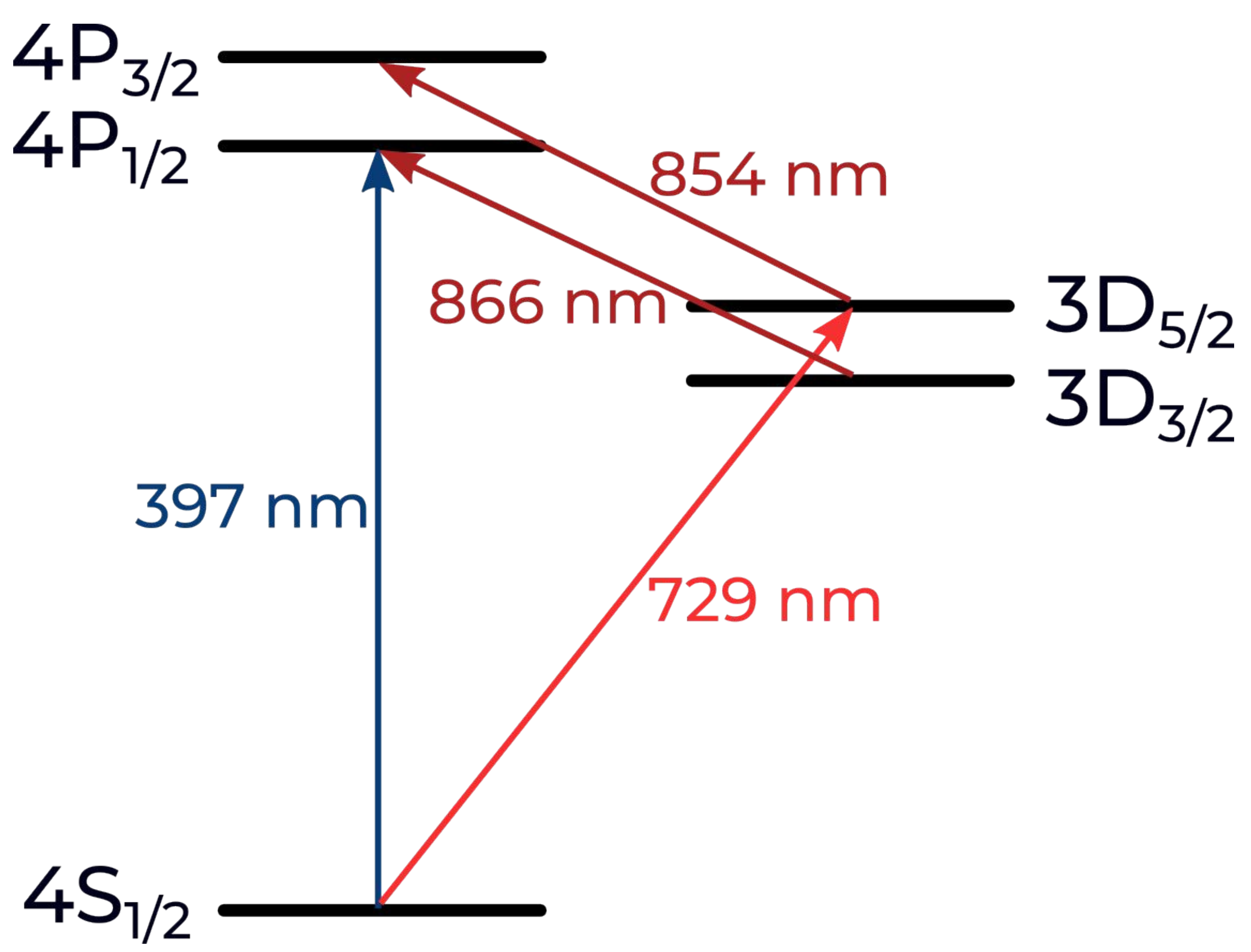
Experimental sequence

Experiment overview

Schematic representation of the previous generation experiment (N_2^+)

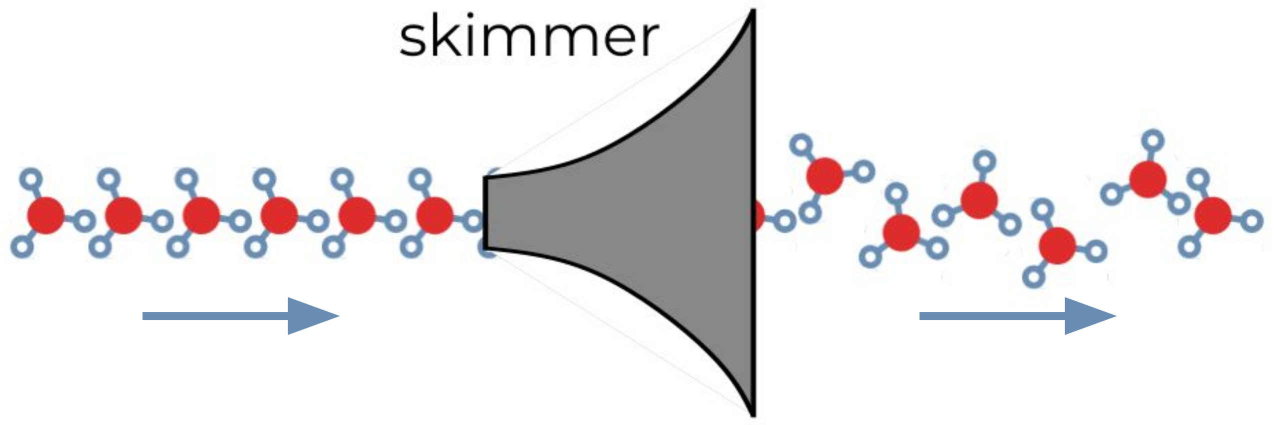


$^{40}Ca^+$ used as an axillary ion for sympathetic cooling of N_2^+ and molecular state readout

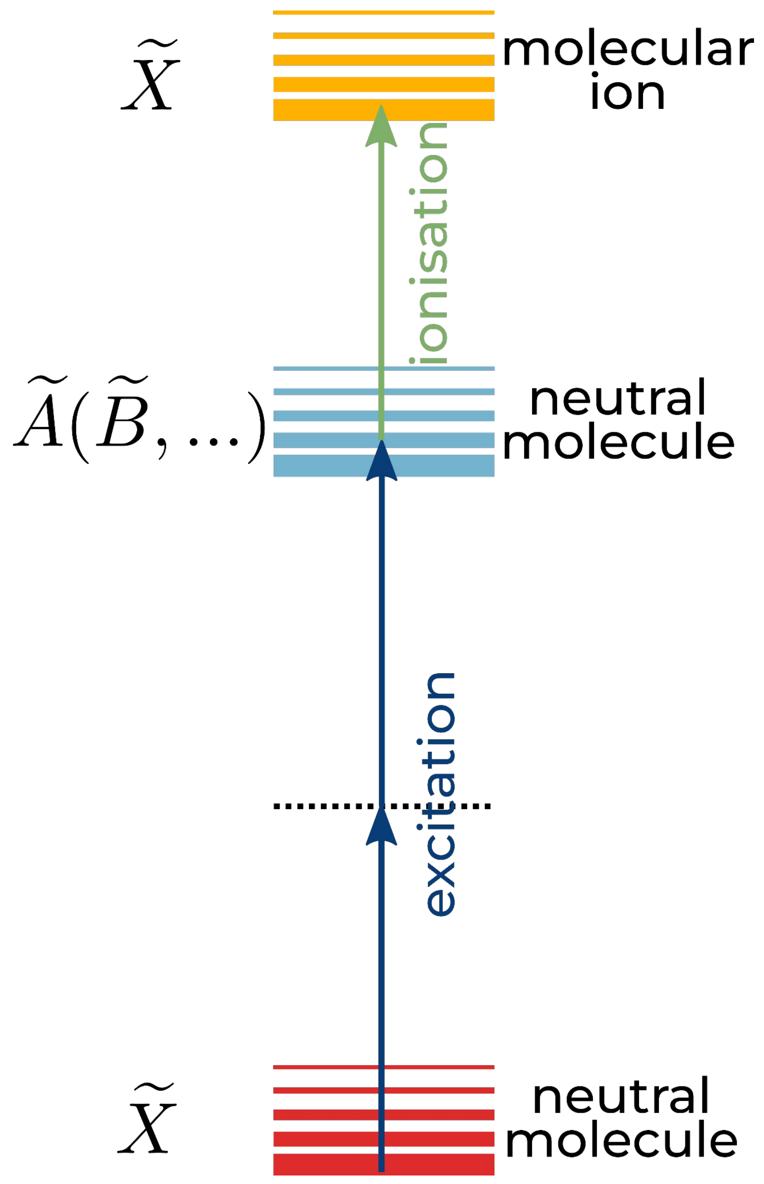


Molecular ion state preparation

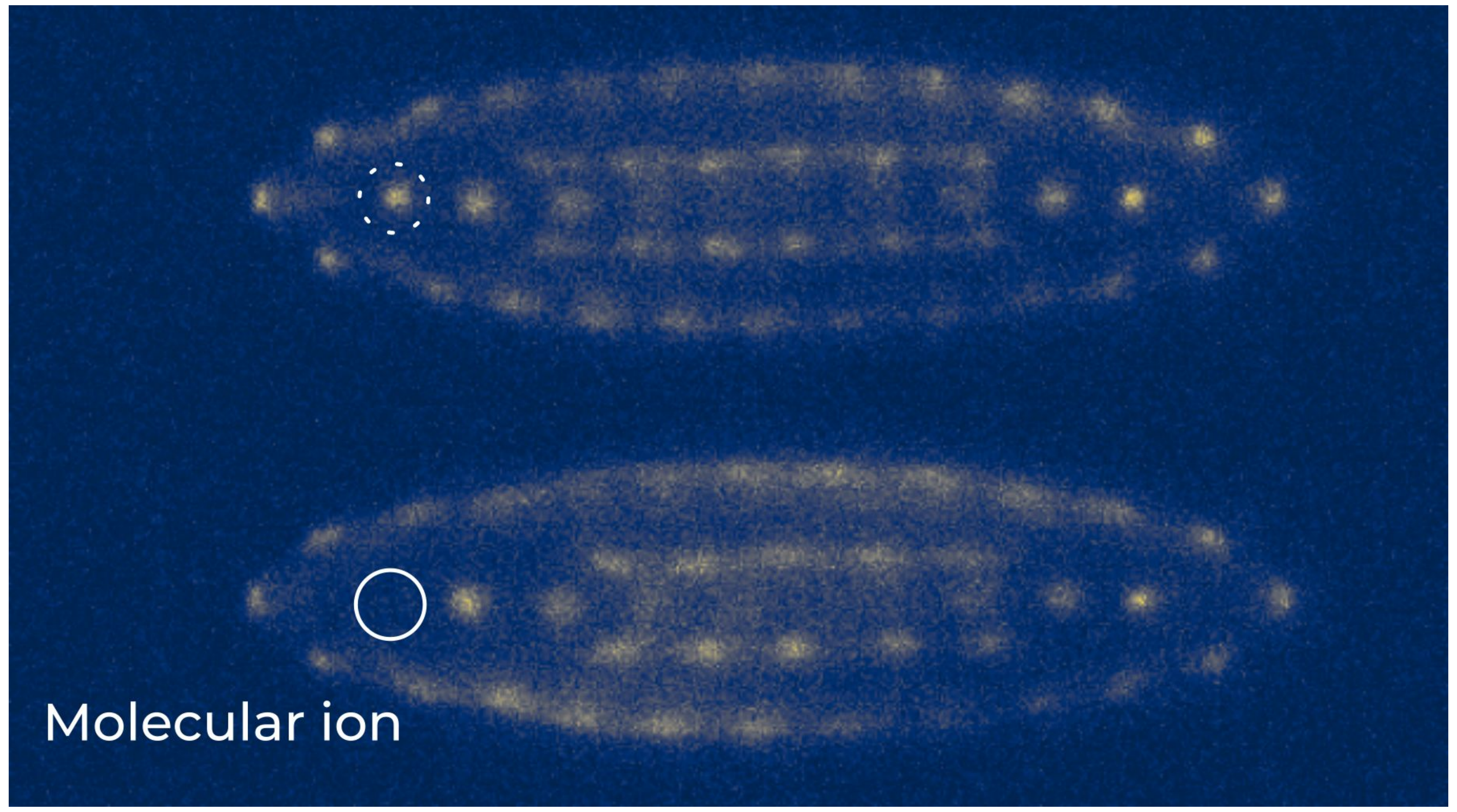
1 Molecular ions are produced in a chosen state by a resonance enhanced multiphoton photoionisation[1] (REMPI).



2 A cold beam of molecules (~10K) is resonantly excited through an intermediate electronic state to the desired state of molecular ion with photons of two colors.



3 Molecular ions are trapped inside a large Ca^+ Coulomb crystal (~10s ions) and sympathetically Doppler cooled



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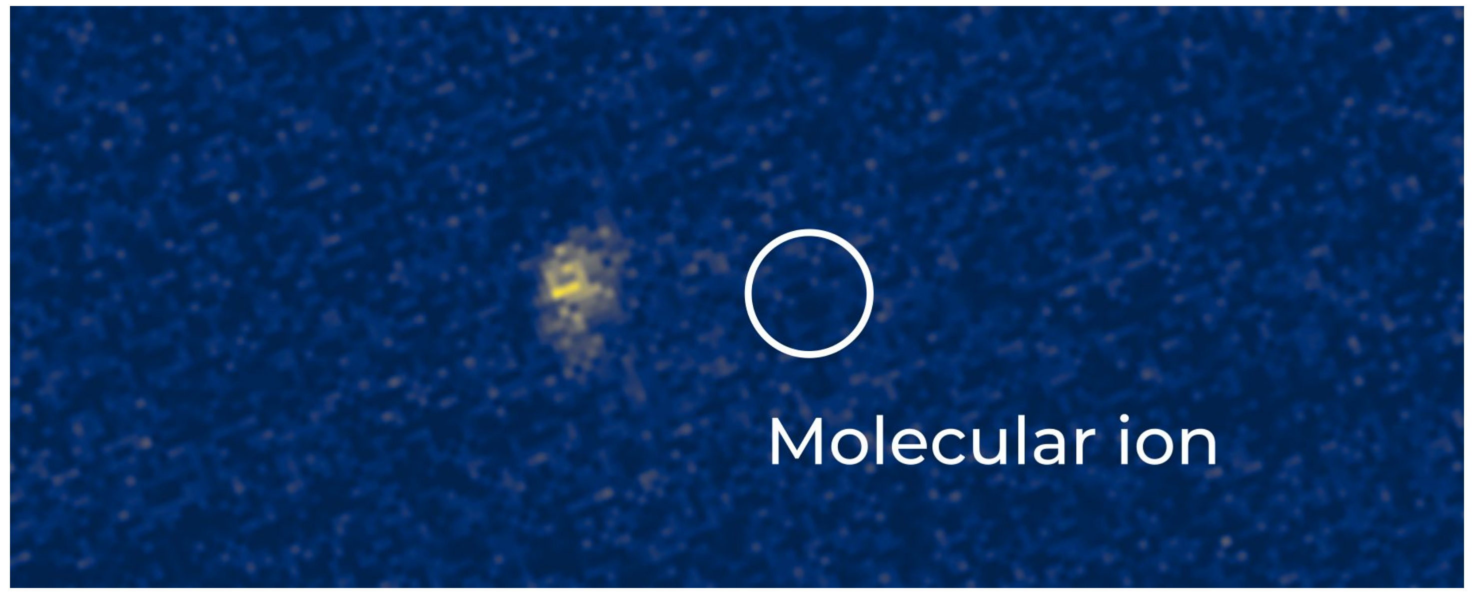
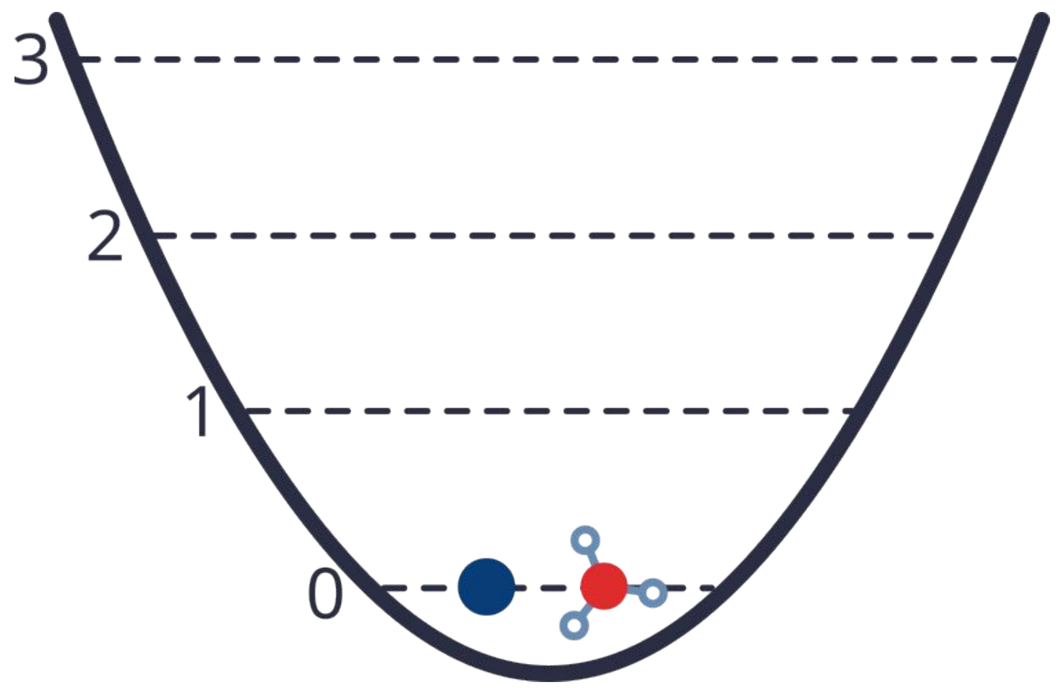
[1] Shlykov, Aleksandr, Mikolaj Roguski, and Stefan Willitsch. "Optimized Strategies for the Quantum-State Preparation of Single Trapped Nitrogen Molecular Ions." *Advanced Quantum Technologies* (2023): 2300268.

Ground-state cooling

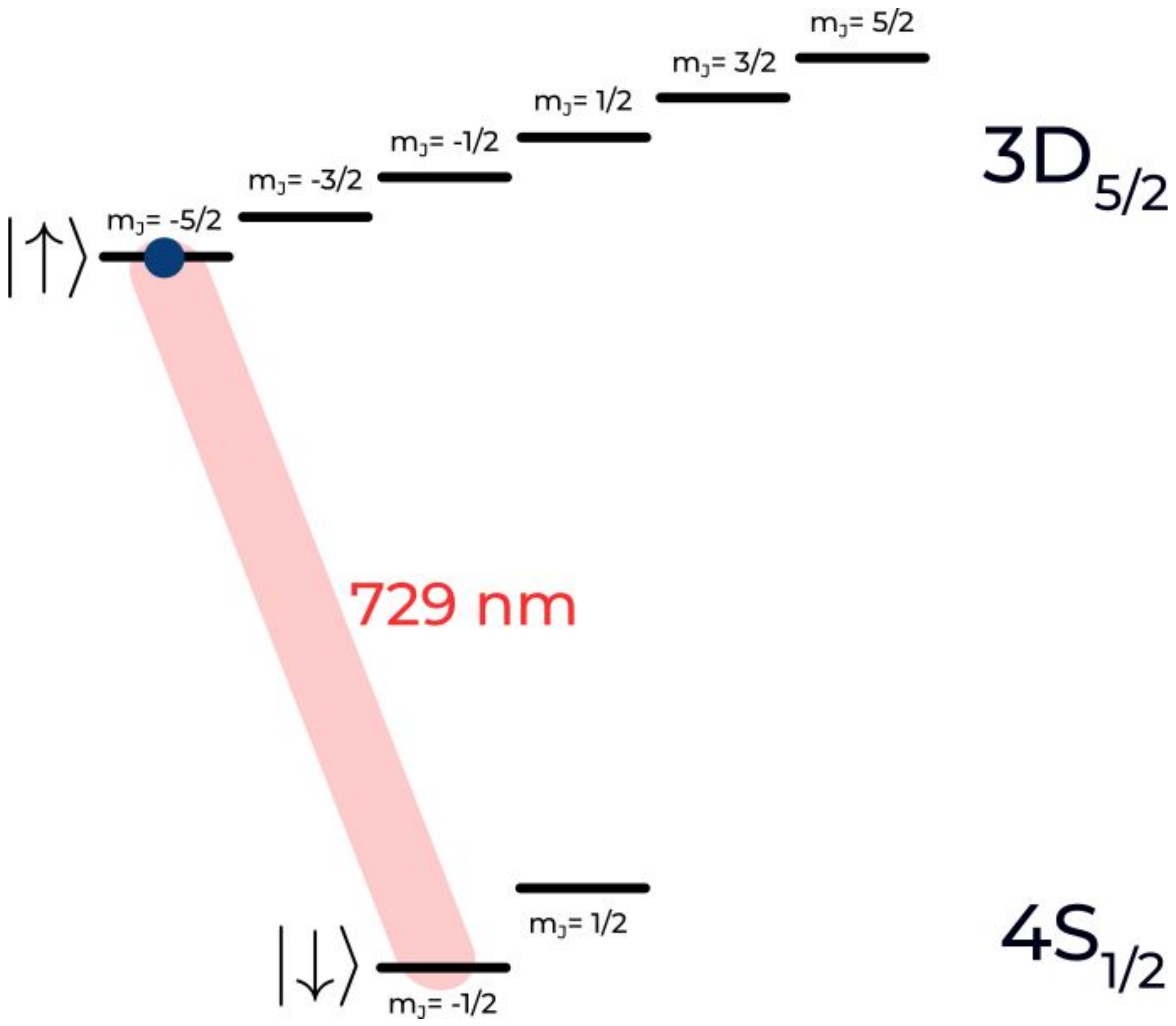
1 The ion crystal is reduced to two ions by lowering trap depth



2 The molecule is sympathetically cooled to the ground state by a resolved sideband cooling on Ca^+

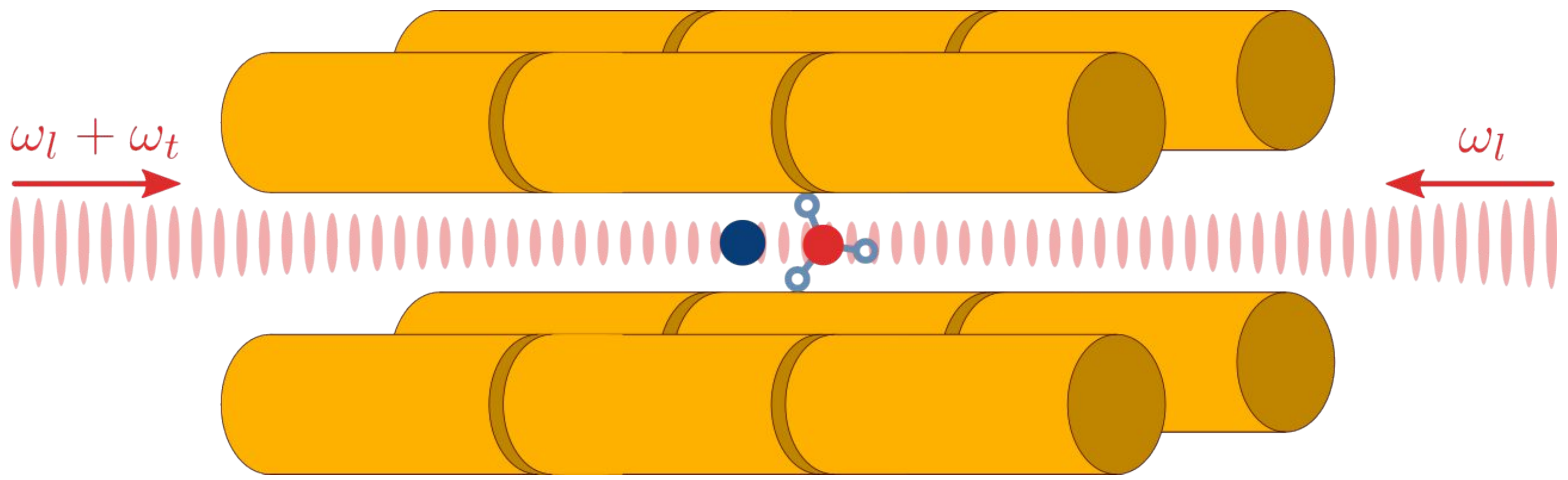


3 Ca^+ ion is shelved into **$3D_{5/2}$** with **m_J= -5/2** state, which is less polarizable by the lattice laser.



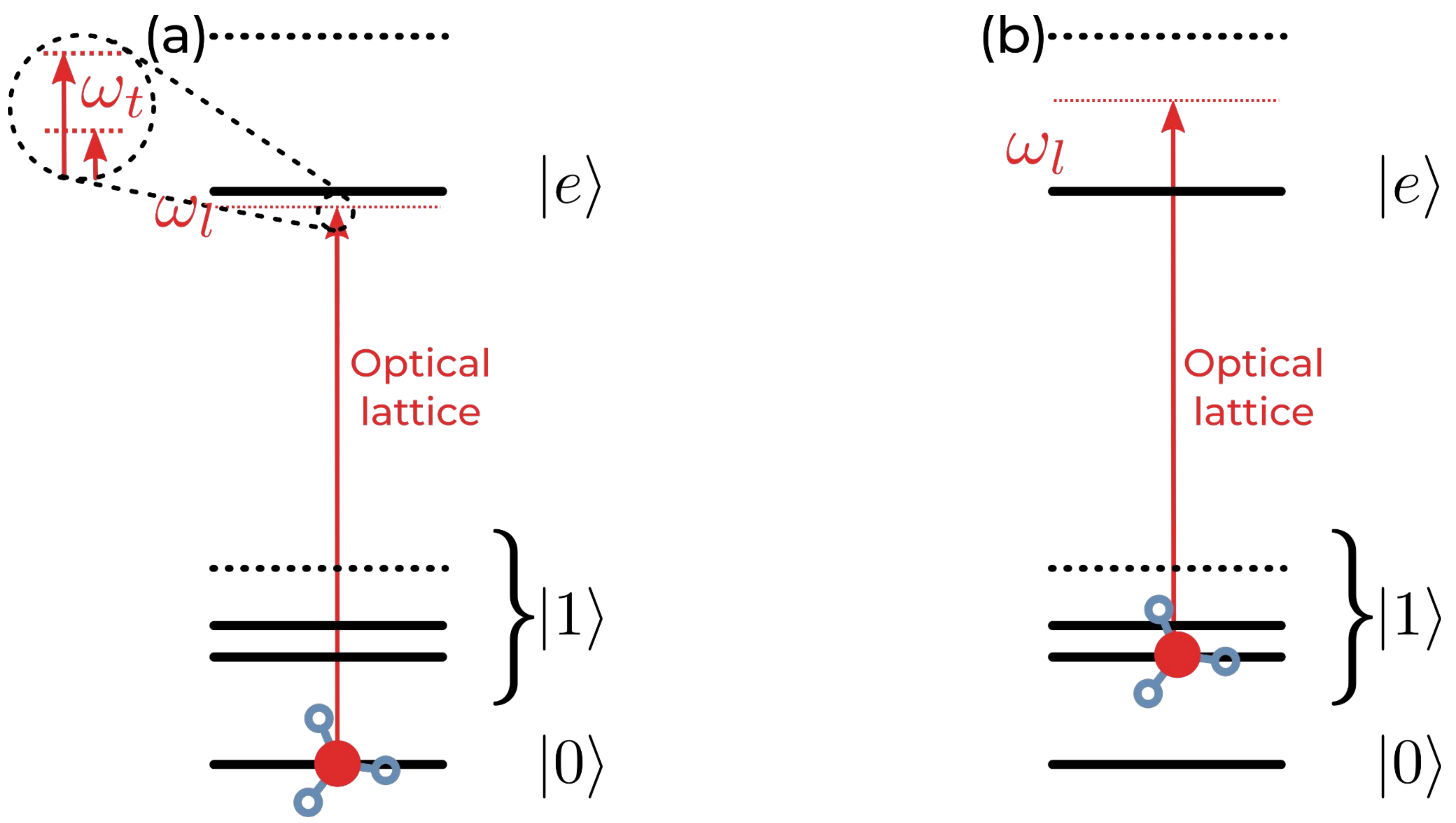
Quantum logic spectroscopy protocol

- A running 1D optical lattice is created along the trap axis



- The lattice exerts an optical dipole force (ODT) on the molecular ion

$$\vec{F}_{ODT} = -\vec{\nabla}(\Delta E_{AC})$$



- The lattice is closely detuned from one of a dipole-allowed optical transitions starting from a rovibrational state of interest (a)
- The lattice is further detuned from all other transitions from the electronic ground state of the molecular ion (b)

Quantum logic spectroscopy protocol

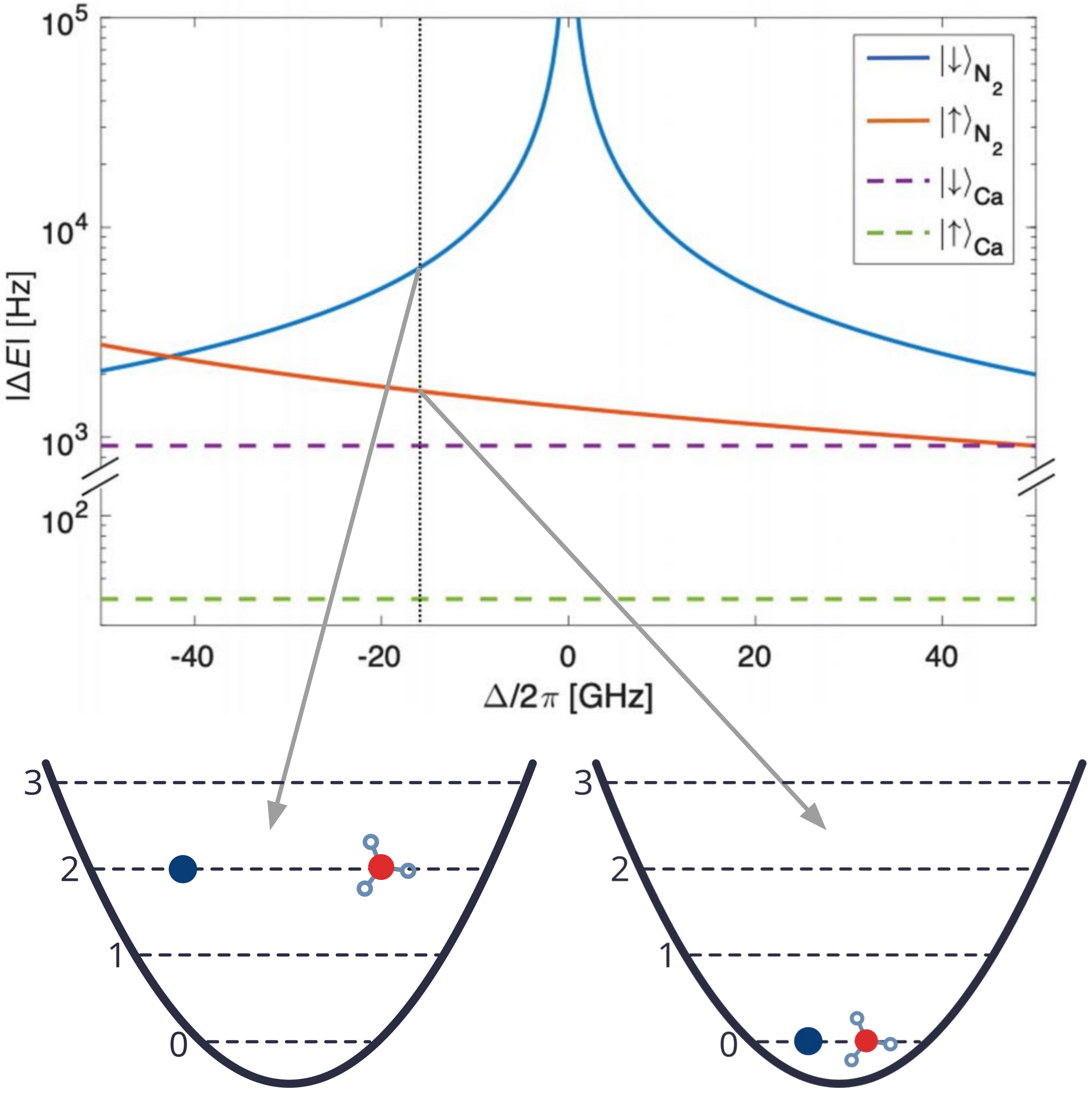
- Lattice beams have a frequency mismatch equal to a normal mode frequency ω_t of the ion string.

- A motion of ions is coherently excited by ODF.

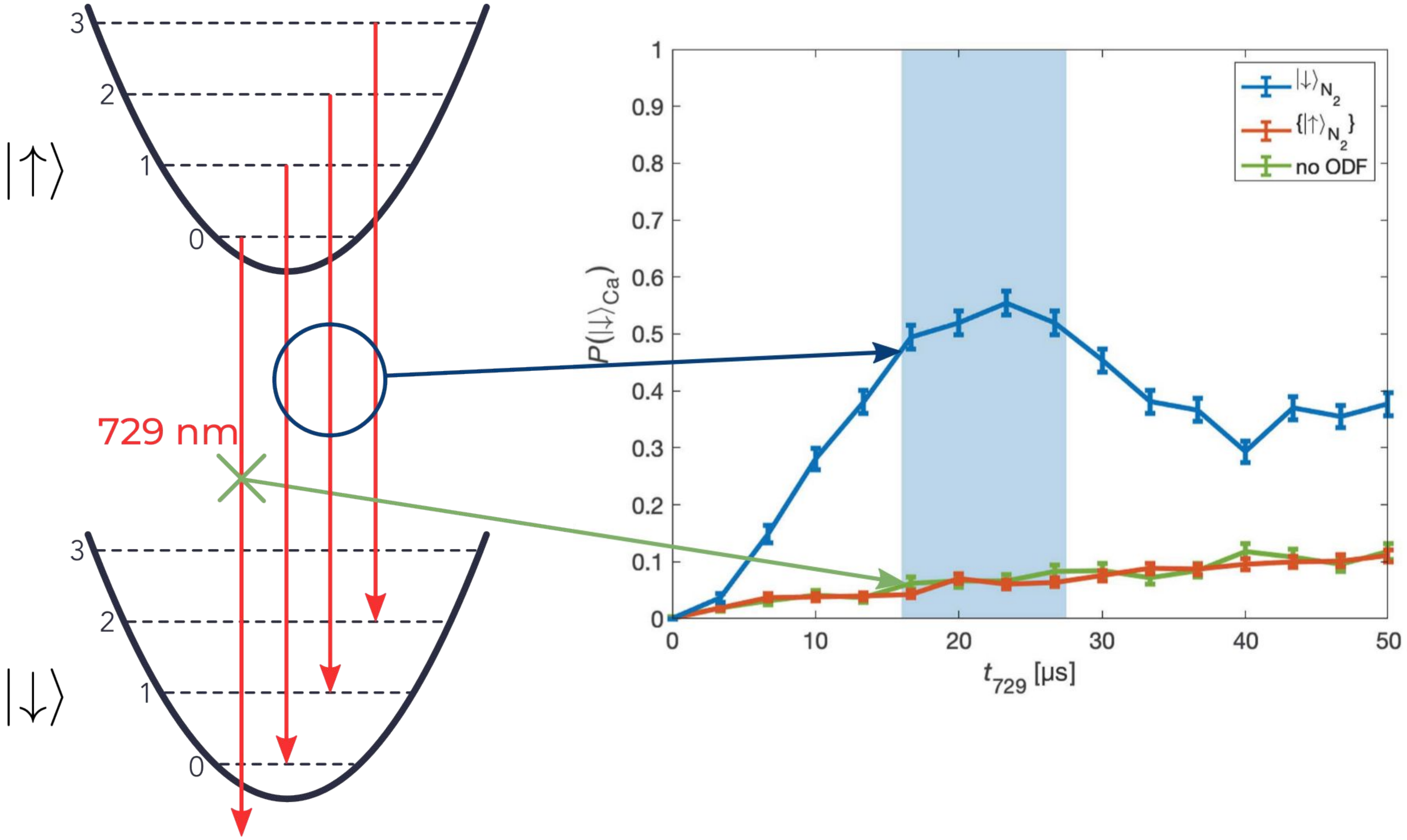
- An amplitude of the motional excitation is proportional to AC Stark shift induced by lattice beams and therefore is state dependent:

$$\alpha \sim \Delta E_{AC}(\omega_l)$$

- Since, lattice is non-resonant with molecular transitions, scattering, changing molecular state, can be minimized



Motional state readout



- Motional state detection is achieved by driving a blue sideband (BSB) on Ca^+ clock transition.
- The transition can only occur, if ions were in an excited motional state (the molecule was in a state of interest).
- The state detection technique is general and can be applied to a wide range of molecules, including polyatomic ones.
- The technique doesn’t alter state of the molecule during detection, which allows for in situ tracking of molecular collision and reaction dynamics.

Experimental setup

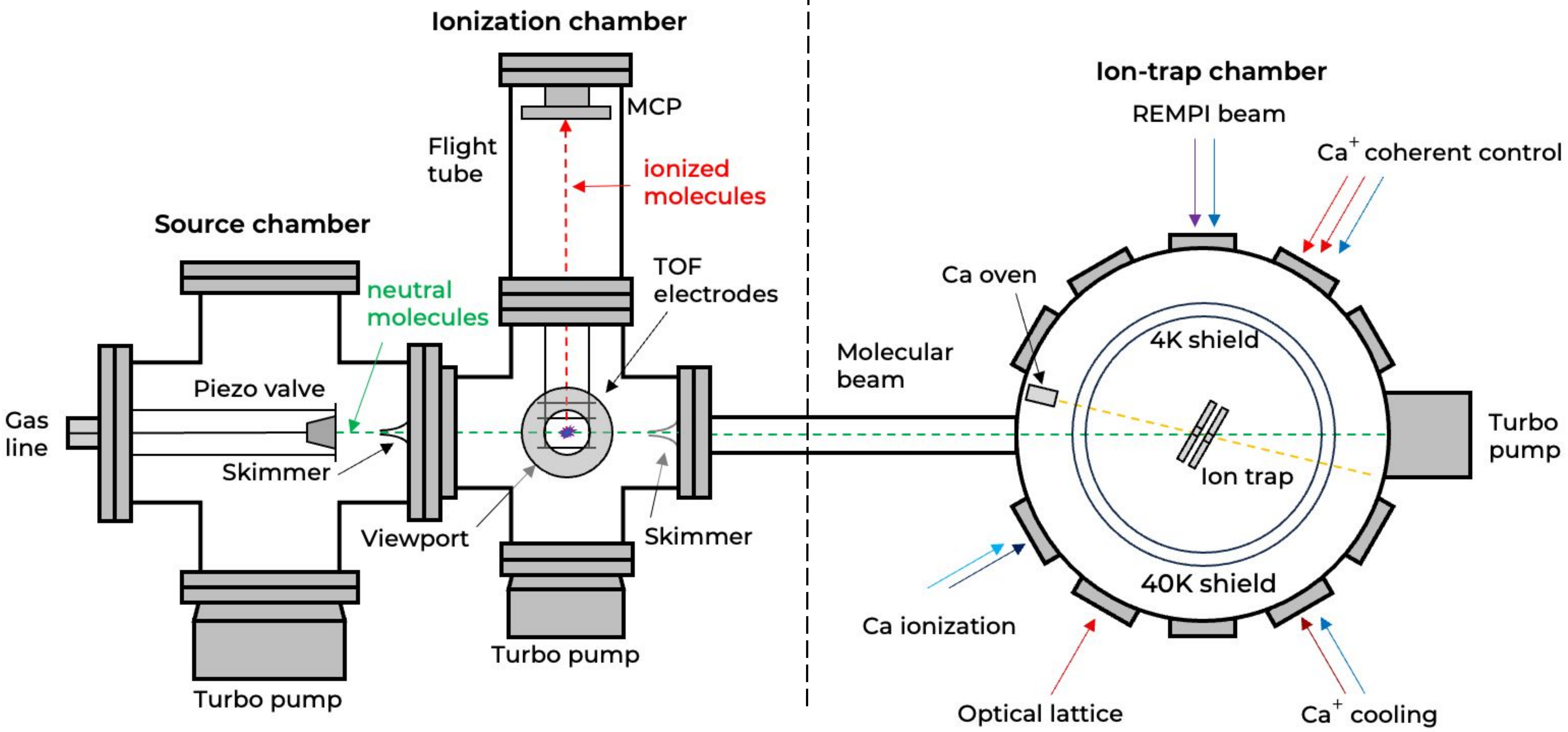
Experimental setup

Molecular beam machine

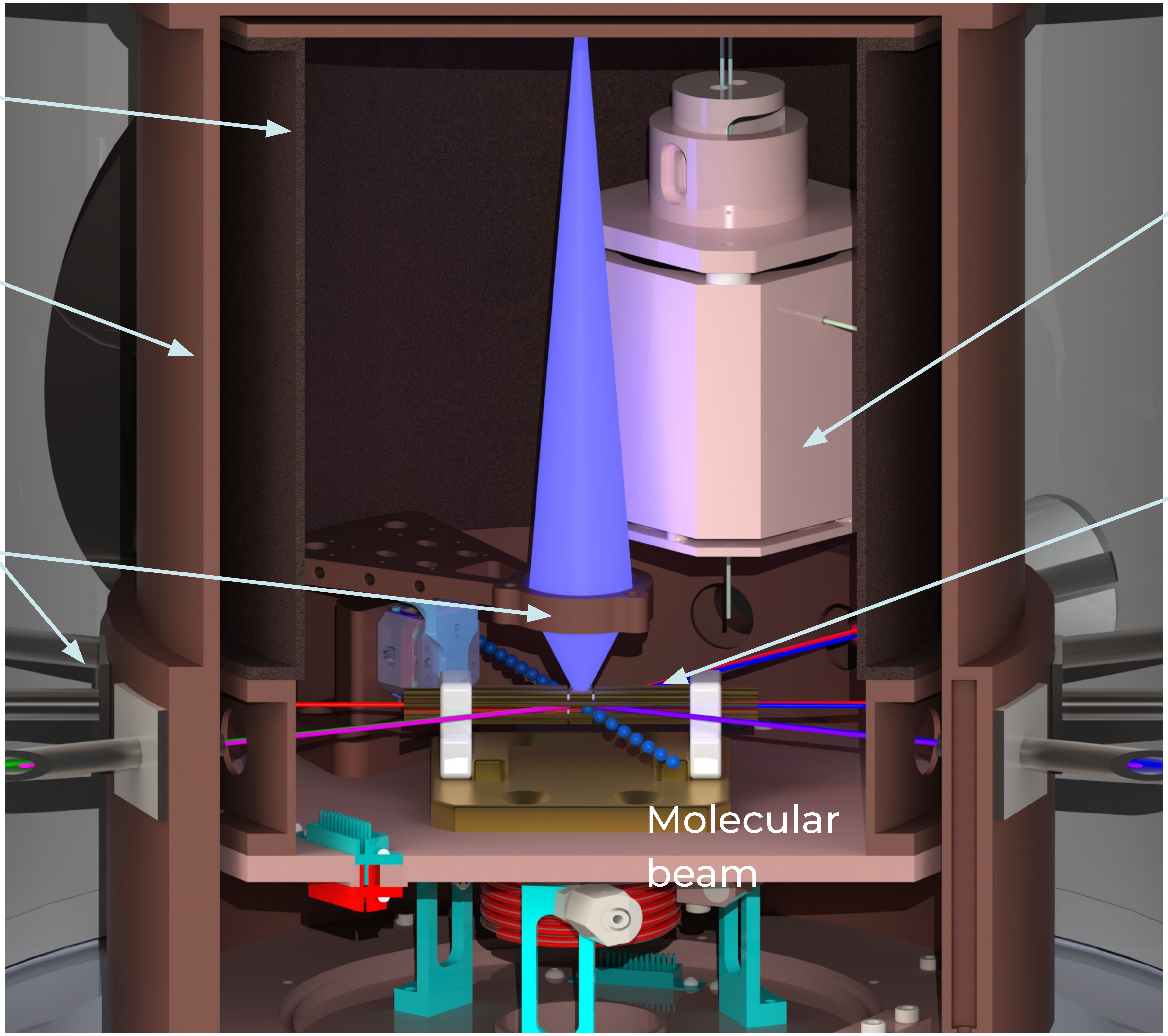
- Creation of molecular beam
- Spectroscopy of molecules in a beam

Science chamber

- Creation and cooling of molecular ions
- QLS
- Collision studies



Experimental setup: science chamber



4K radiation shield

40K radiation shield

light tubes

In-vacuum imaging lens

Optical lattice
Ca+ coherent control
REMPI

Helical resonator is designed to provide 1 MHz radial motional frequency

Gold coated cryogenic ion trap with segmented electrodes

Doppler cooling

Optical lattice

Ca ionisation

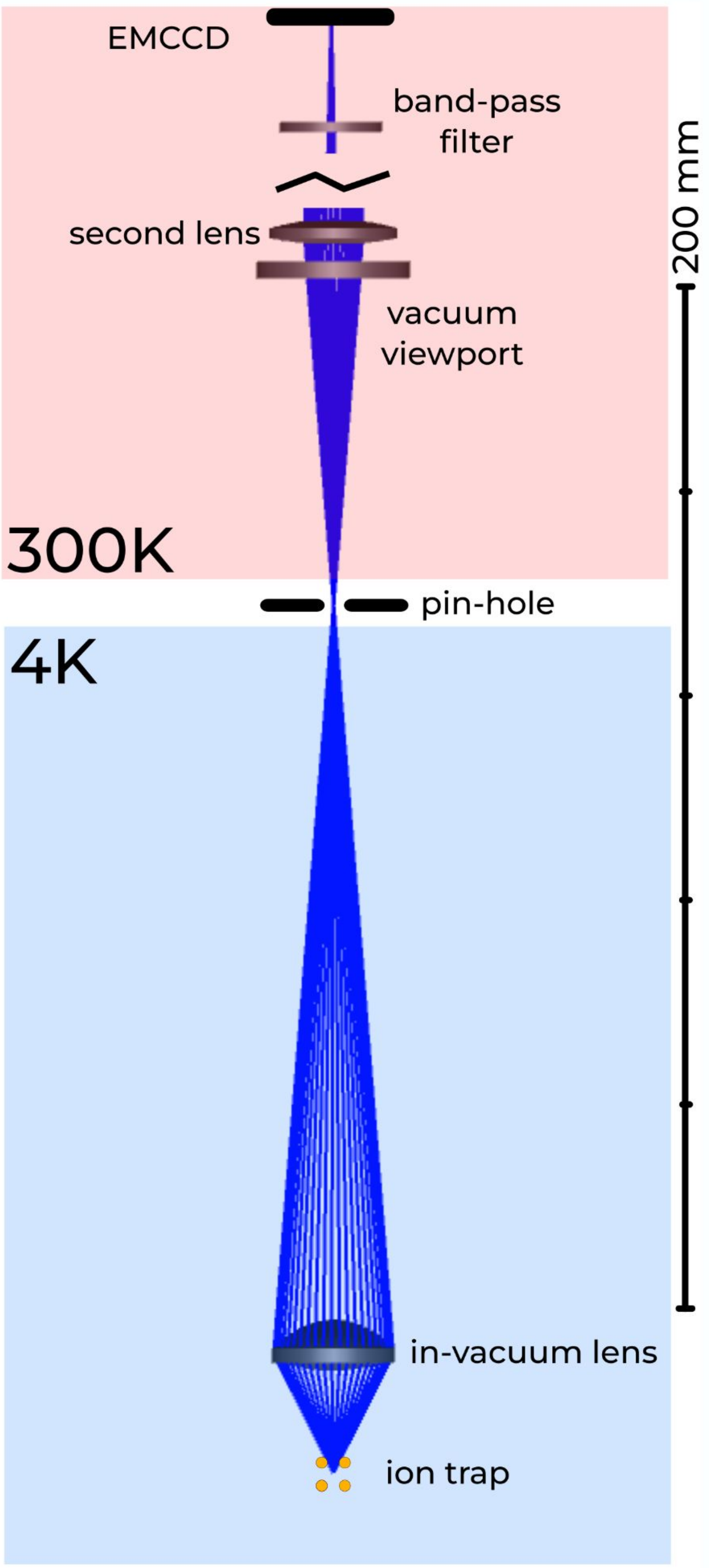
Experimental setup: imaging system

The imaging system consisting of two custom aspheric lenses was implemented.

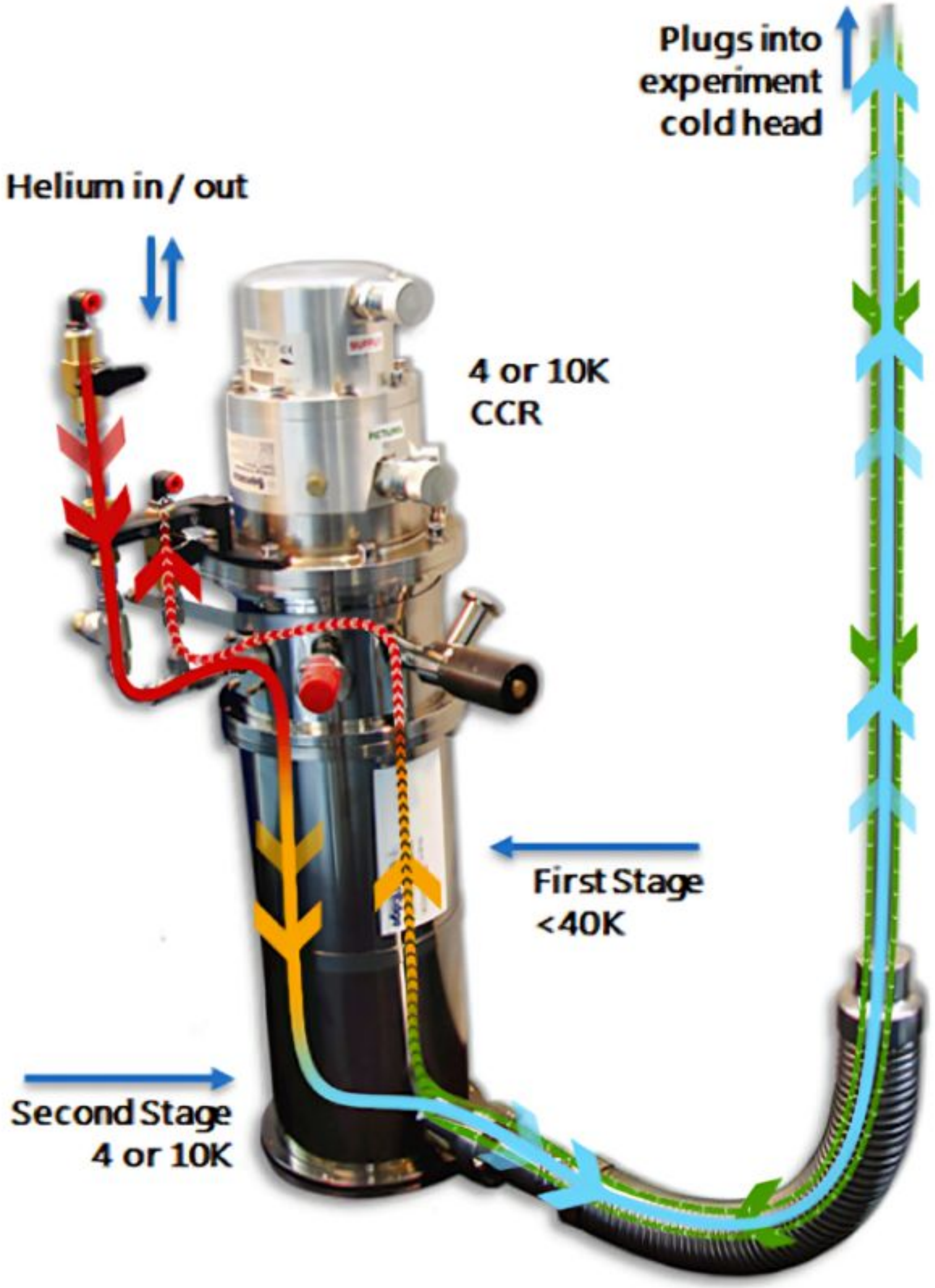
The in-vacuum lens is movable by a piezo-positioner.

A pin-hole in an intermediate focus between 4K and 40K radiation shields limits room temperature BBR from outside.

Parameter	**Value**
Magnification	17.6
Collection NA	0.46
Total collection efficiency	~2%
Field of view (near-diffraction limited)	0.5 mm
Working distance	20 mm



Cryostat



Cold environment is created by a 3 stage **ColdEdge Stinger** cryostat:

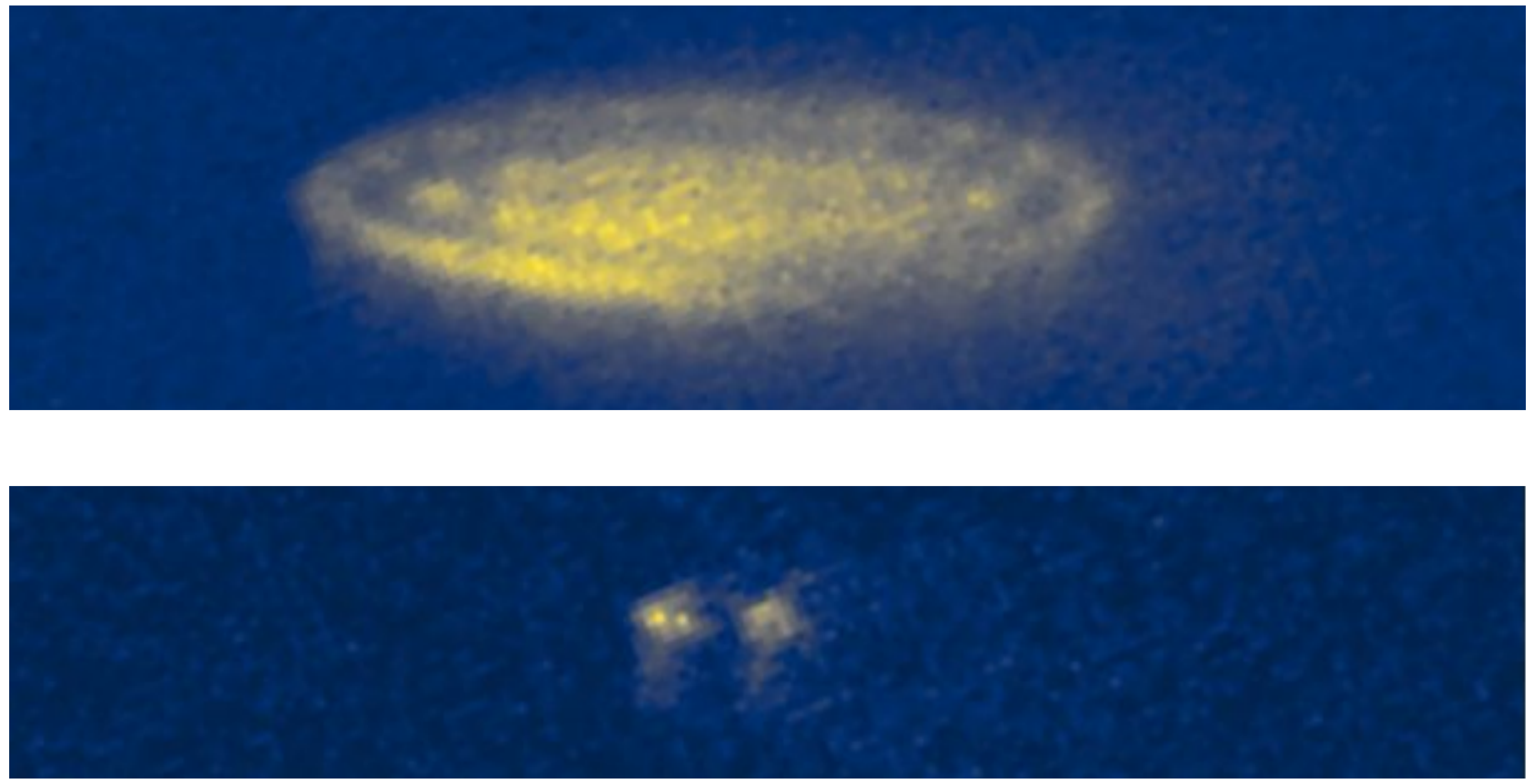
- 2 stage GM cryocooler
- Third stage is closed loop He flow line cooled by a second stage

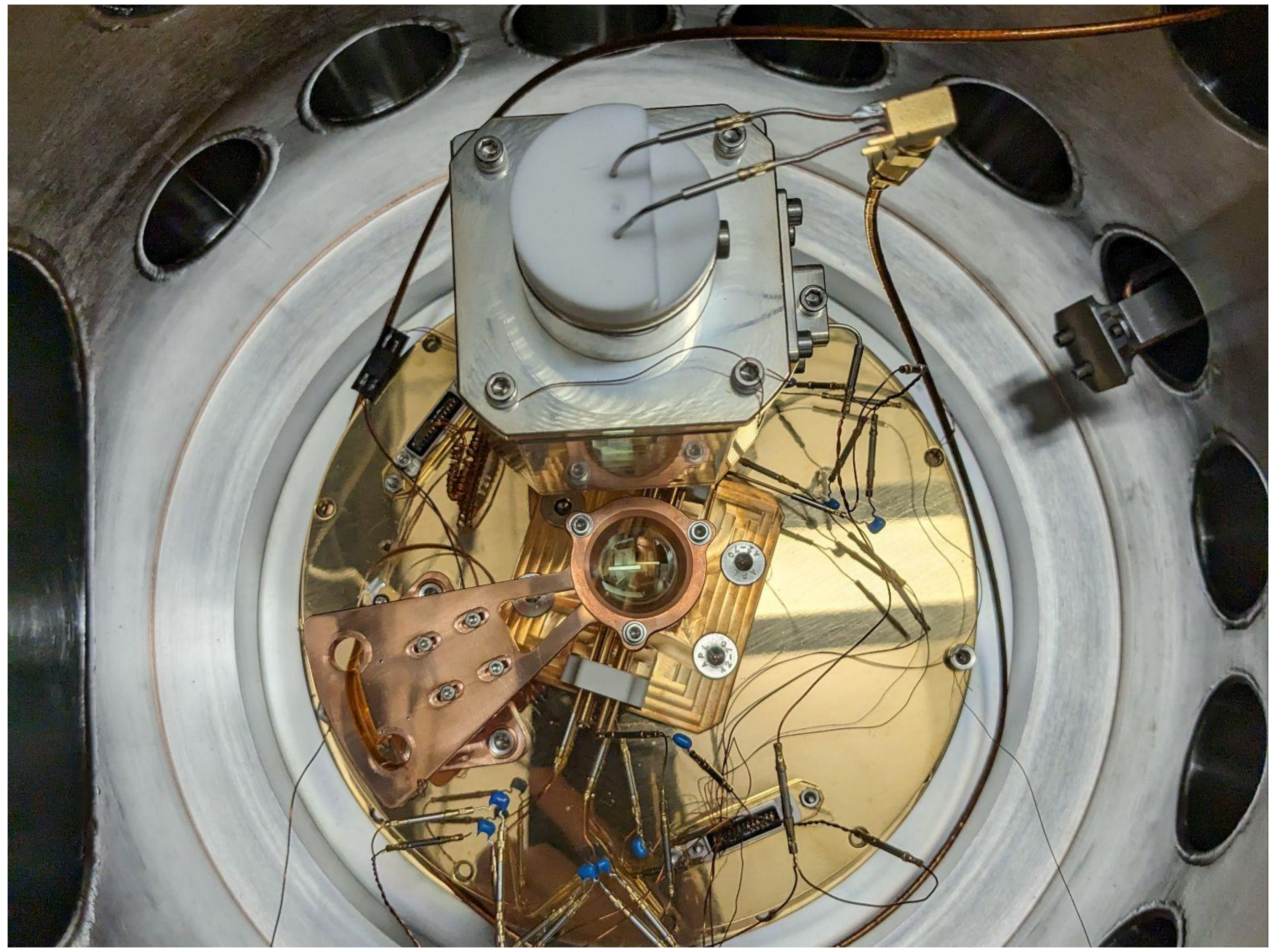
Cryocooler induced vibrations < 10 nm - comparable with flow cryostats.

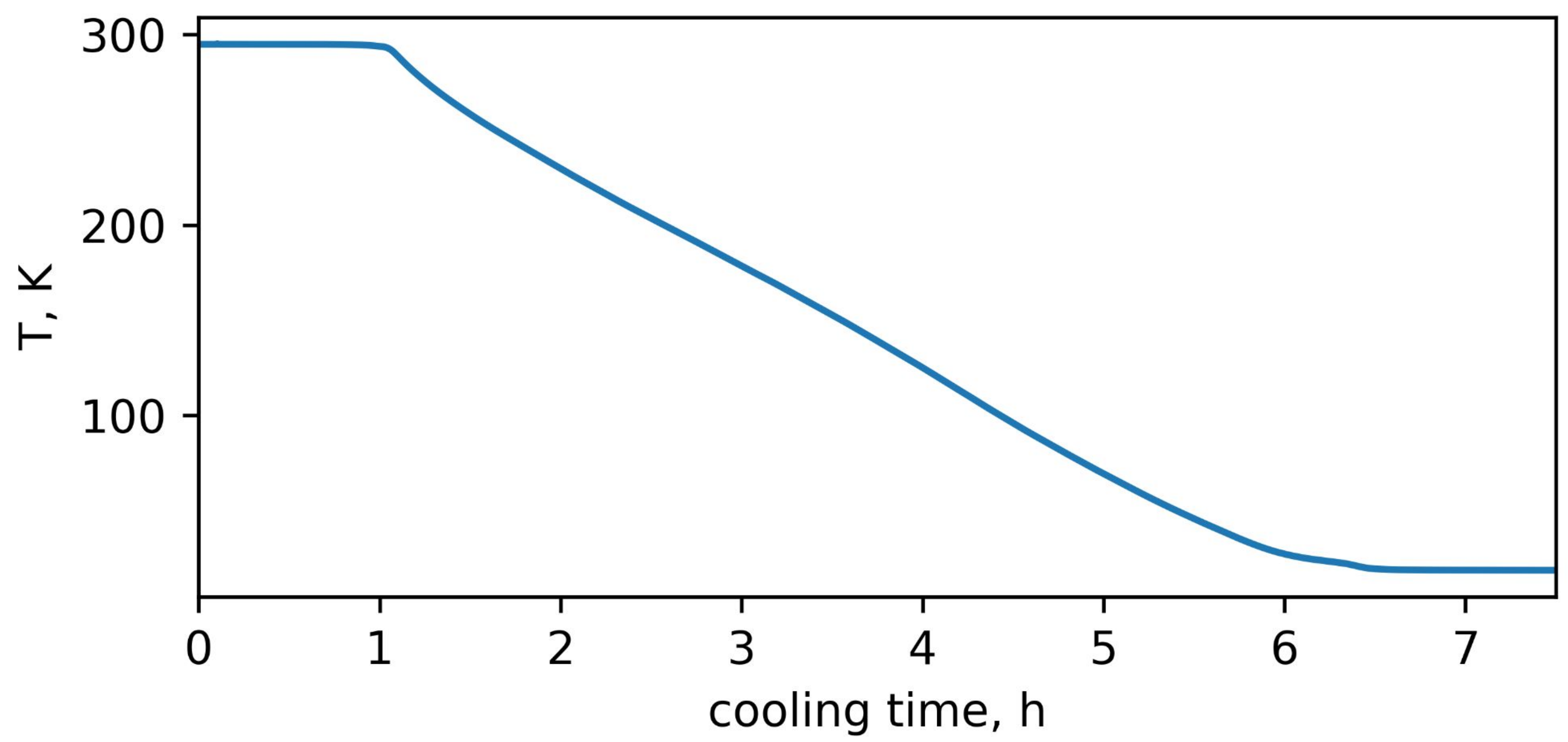
0.9W of cooling power at 4K stage.

Experimental setup: current progress

- Ion trapping setup is commissioned
- No radiation shields at the moment
- Minimum temperature achieved is 16K (expected below 10K with cryoshields)
- Cooling down time is about 6.5 hours
- First Ca^+ ion crystals are trapped and Doppler cooled







Summary

- Quantum logic spectroscopy techniques can be extended to polyatomic molecules.

- Cryogenic environment is required to preserve a state of polyatomic molecules and their chemical identity on experimentally relevant timescales.

- A new cryogenic ion trapping setup for polar and polyatomic molecular ions is currently under development.

Acknowledgements

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Grischa Martin

Electronic workshop

Georg Holderied

Laser engineer

Anatoly Johnson

Thank you for attention!