Cold anions and prospects for antiprotonic molecules

Matthias Germann

18.12.2024

PART I

Antiprotonic molecules and a putative dipole moment of the antiproton

PART II

Cold anions – Borealis experiment and C_2^-@AEgIS

PART I

Antiprotonic molecules and a putative dipole moment of the antiproton

High up in the sky…

PART II

Cold anions – Borealis experiment and C_2^-@AEgIS

…down on Earth.

Antiprotonic molecules and a putative dipole moment of the antiproton

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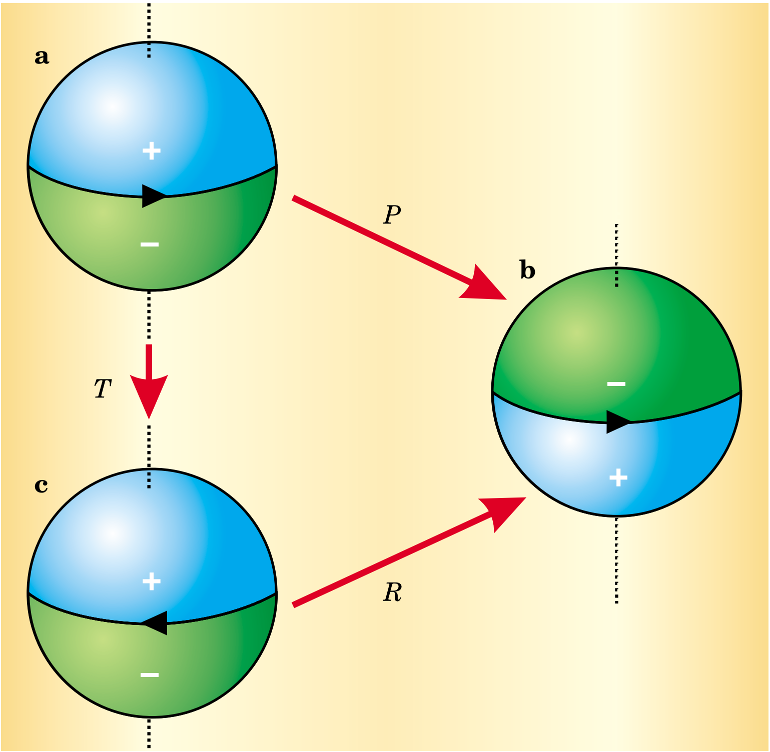
Electric dipole moments of particles



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Assuming CPT, thus also CP symmetry.

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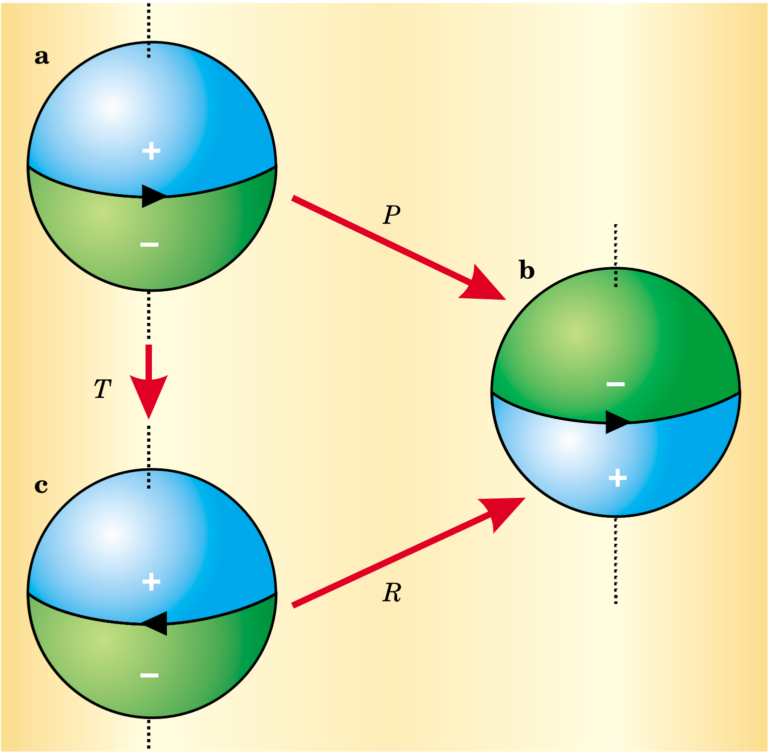

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e^--EDM < 10^{-38} e·cm [1], p^--EDM < 10^{-31} e·cm [2]

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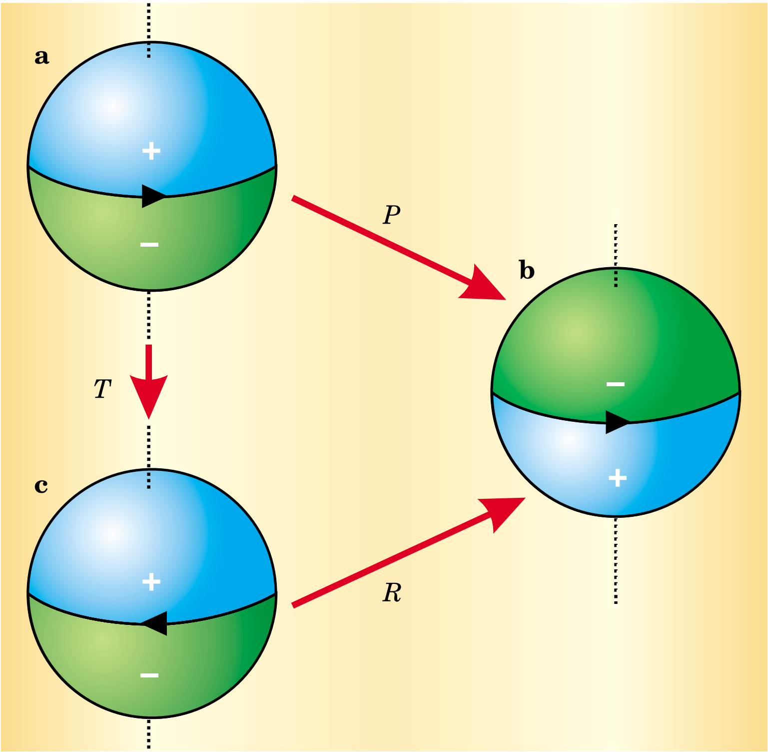

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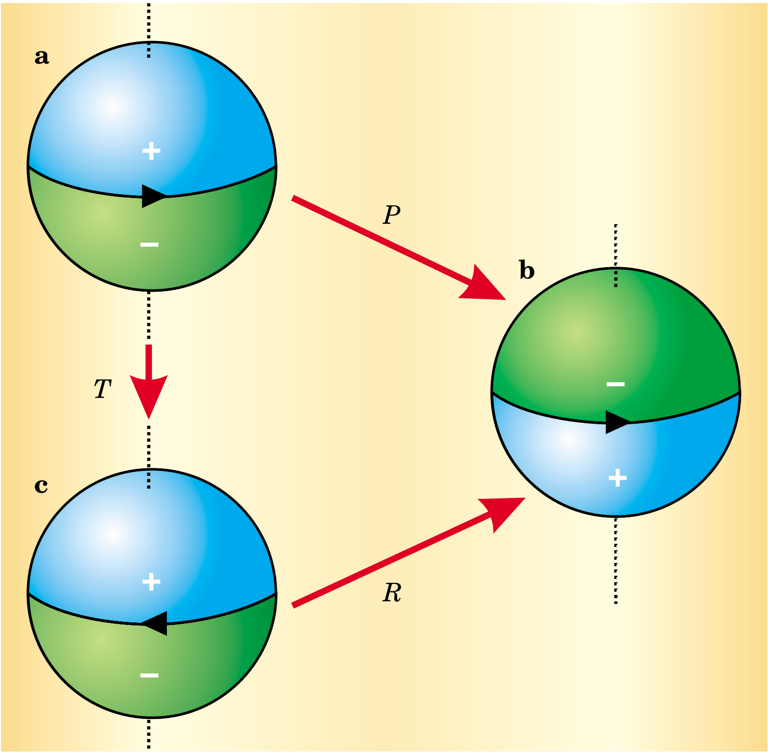

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➔ Constraining EDMs, i.e., establishing upper limits, is promising route to search for beyond-SM physics.

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[2] CPEDM Collab., arXiv:1912.07881v3 (2021).

Current constraints

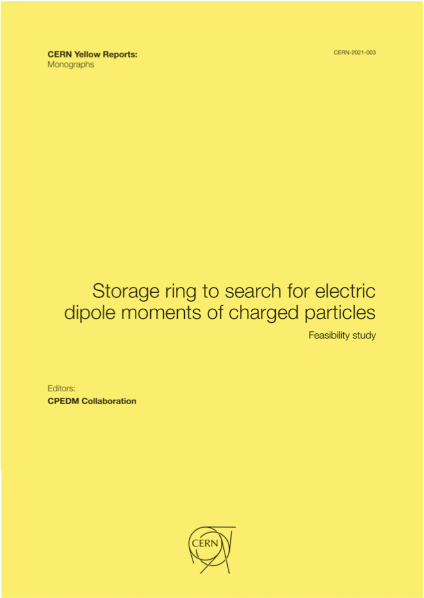
Upper limits for the magnitude of the electric dipole moment for some common subatomic particles [*].

Neutron	**< 1.8 * 10^{-26} e cm**
Electron	**< 1.1 * 10^{-29} e cm**
Proton	**< 2.1 * 10^{-25} e cm**

[*]: R.L. Workman et al. (Particle Data Group), Prog. Theor. Exp. Phys. 2022, 083C01

Proton electric dipole moment

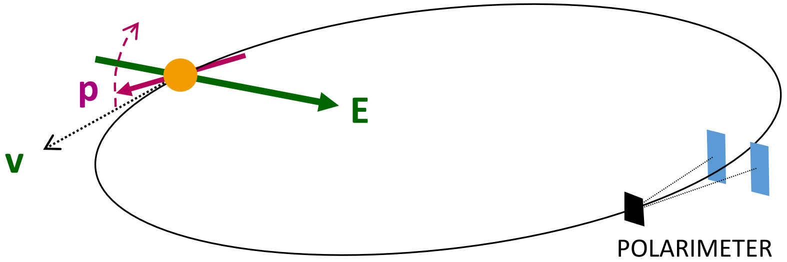
Among the constituents of normal matter (p, n, e^-), the proton EDM has the least constraining limit.



Current efforts [*]:
Storage rings with spin-polarized beams

- US: storage ring EDM collaboration at BNL
- Europe: JEDI (Jülich Electric Dipole moment Investigations)

➔ Promising, but expensive!



[*], see, e.g., R. Alarcon et al. arXiv:2203.08103v2 (2022)

Alternative: $\bar{p}$ in a bound state?

Can we implement an alternative measurement using

- **a $\bar{p}$ in a bound state**
- **spectroscopic methods**
- **quantum technologies**

to derive a constraint on the $\bar{p}$-EDM,

and hence – assuming CPT-symmetry – also one on the p-EDM?

Key: polar, antiprotonic molecule

C.f.: electron EDM measurements

e ELECTRIC DIPOLE MOMENT (d)

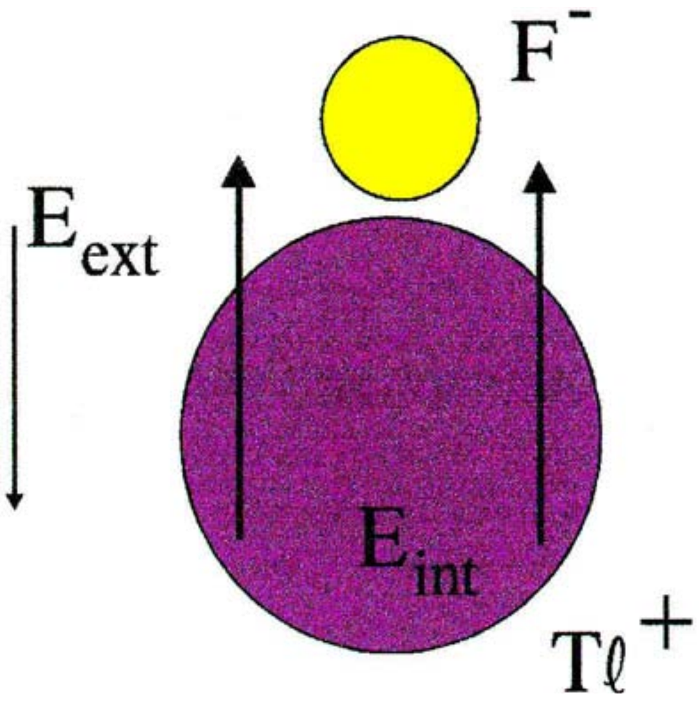
A nonzero value is forbidden by both *T* invariance and *P* invariance.

VALUE (10^{-28} e cm)	*CL%*	*DOCUMENT ID*		*TECN*	*COMMENT*
< 0.11	90	[1] ANDREEV	18	CNTR	ThO molecules
• • • We do not use the following data for averages, fits, limits, etc. • • •					
< 1.3	90	[2] CAIRNCROSS	17	ESR	$^{180}Hf^{19}F$ molecules
−5570 ± 7980 ±120		KIM	15	CNTR	$Gd_3Ga_5O_{12}$ molecules
< 0.87	90	[3] BARON	14	CNTR	ThO molecules
< 6050	90	[4] ECKEL	12	CNTR	$Eu_{0.5}Ba_{0.5}TiO_3$ molecules
< 10.5	90	[5] HUDSON	11	NMR	YbF molecules
6.9 ± 7.4		REGAN	02	MRS	^{205}Tl beams
18 ± 12 ± 10		[6] COMMINS	94	MRS	^{205}Tl beams
−27 ± 83		[6] ABDULLAH	90	MRS	^{205}Tl beams
−1400 ± 2400		CHO	89	NMR	TlF molecules
−150 ± 550 ±150		MURTHY	89		Cs, no *B* field

Most advanced measurements all make use of electrons in polar molecules.

R.L. Workman et al. (Particle Data Group), Prog. Theor. Exp. Phys. 2022, 083C01

Key: Enhancement of electric field by polarization

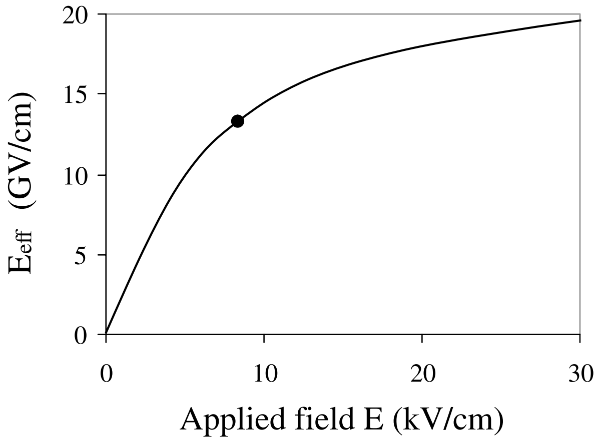


P. G. H. Sandars, Contemp. Phys. 42, 97 (2001)

E_{ext}: external electric field, generated by macroscopic electrodes and lab power supplies.

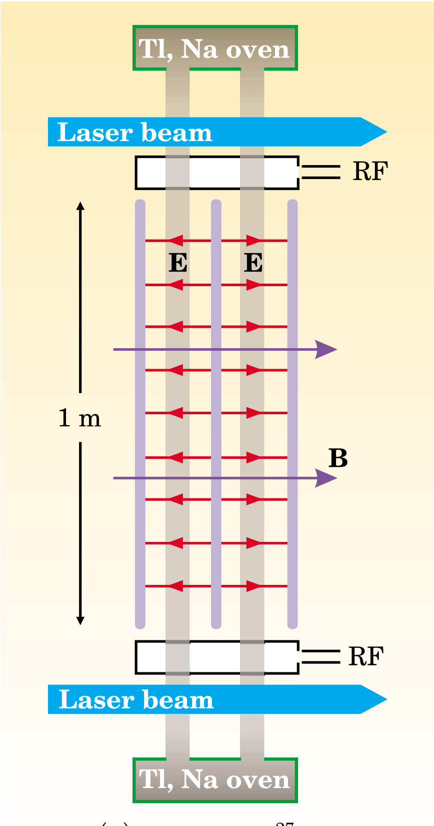
E_{int}: internal electric field, generated by polarization of the microscopic charges in the polar molecule.
→ Field which is "felt" by electron.

➔ $|E_{int}| \gg |E_{ext}|$



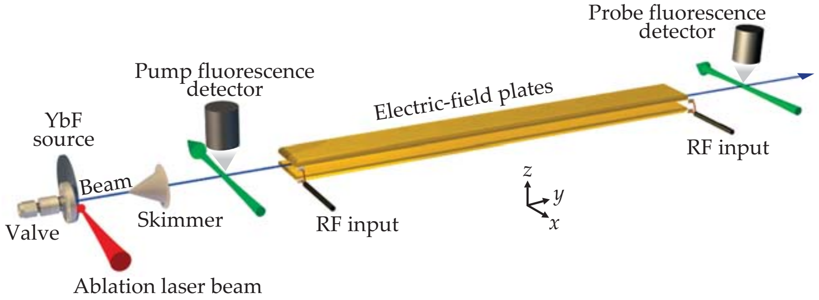
J. J. Hudson et al. Phys. Rev. Lett. 89, 023003 (2002)

Examples of e^--EDM experiments

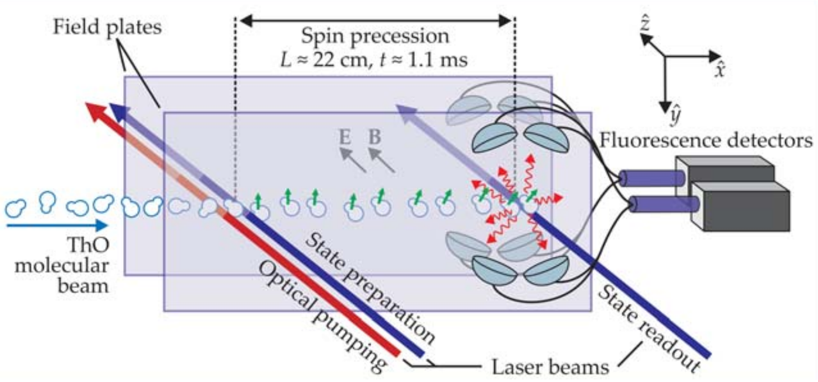


$$d(\mathrm{e}) < 1.6 \times 10^{-27} e\ \mathrm{cm}$$

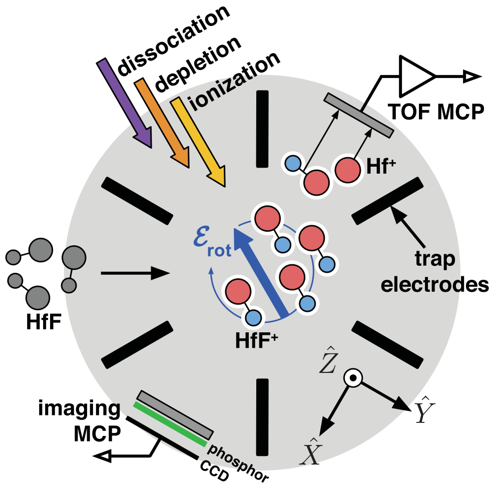
Thallium experiment,
University of California, Berkeley,
Lawrence Berkeley National
Laboratory



YbF experiment,
Imperial College London



ThO experiment,
Harvard and Yale

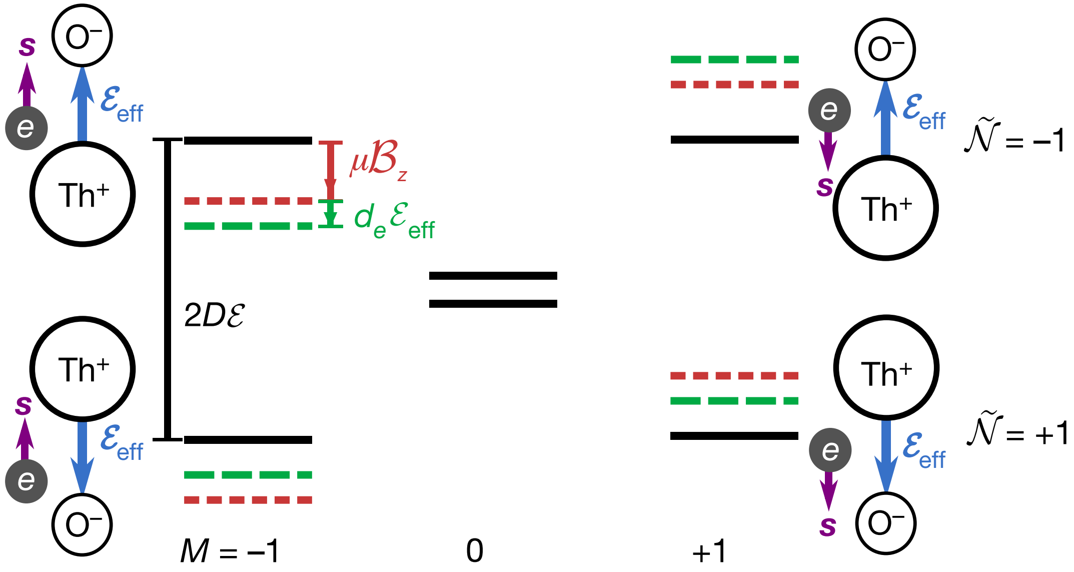


HfH^+ experiment,
JILA, NIST and University of Colorado, Boulder

[1] N. Fortson et al. Physics Today 56, 33 (2003)
[2] B. M. Schwarzschild, Physics Today 64, 12 (2011)
[3] B. M. Schwarzschild, Physics Today 67, 15 (2014)
[4] W. B. Cairncross et al. Phys. Rev. Lett. 119, 153001 (2017)

e^--EDM measurement method – Concept

- **Orientation of a polar molecule in an external electric field splits energy levels.**
- **Putative dipole moment of the electron results in an additional shift, with its sign depending on the external field direction.**
- **Inversion of the external field (and/or the initial quantum state) reverses this shift.**
  → distinguishable from Zeeman shift



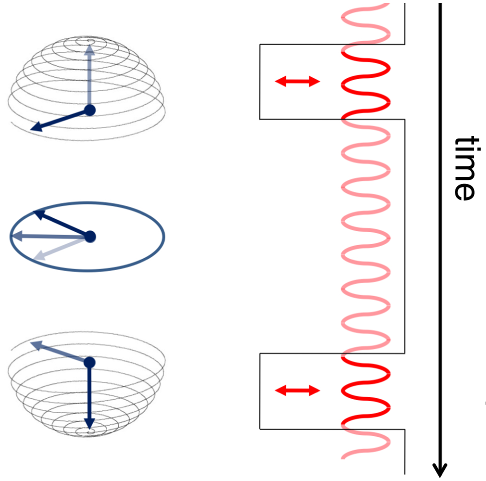
V. Andreev et al. (ACME collaboration), Nature 562, 355 (2018).

e^--EDM measurement method – Implementation

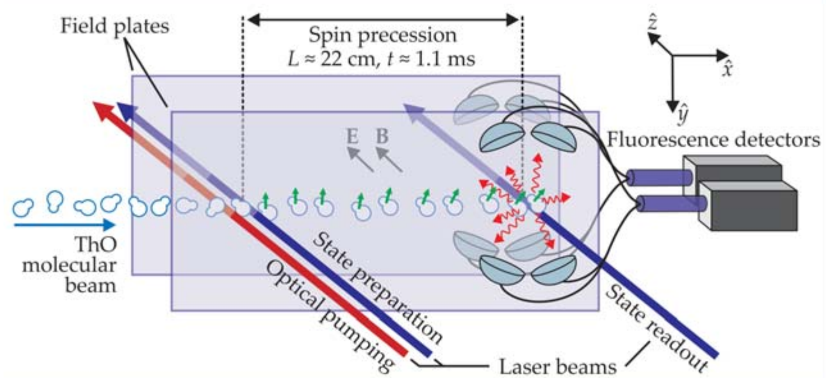
Electron spin resonance:

- **Initialization in superposition of the two states of interest by π/2 pulse.**
- **Phase accumulation of superposition state: rotation on equator of Bloch sphere (free precession).**
- **Second π/2 pulse: transfer phase difference into population difference.**
- **Read out: probing population in the original state.**

(Ramsey separate oscillatory field method.)



T. E. Chupp et al., Rev. Mod. Phys. 91, 015001 (2019)



J. Baron et al. (ACME collab.), Science 343, 269 (2014).

Antiprotonic molecules – Swap e^- with $\bar{p}$

Can we replace the valence electron, which has been used to constrain the electron EDM, with a $\bar{p}$ to constrain the $\bar{p}$-EDM?

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Overall idea

- Produce a polar antiprotonic Rydberg molecule:
  - Polar molecule (for enhanced internal electric field)
  - $\bar{p}$ in Rydberg state (for extended lifetime)
- Implement experimental protocol to measure level shift of $\bar{p}$ due to putative $\bar{p}$ EDM.
- Derive upper limit for $\bar{p}$ EDM based on statistical and systematic uncertainties of the measurement. (Assuming, we will get a null-result – everything else would be a big surprise.)

Production of antiprotonic molecules

Precursor

- Neutral molecule AB (e.g., from gas jet target)
- Molecular ion
  - anion AB^- (co-trapped with $\bar{p}$)
  - cation AB^+ (nested trap)

Formation

- Overlap precursor molecule (or molecular ion) with $\bar{p}$.
- Form $\bar{p}AB$ by $\bar{p}$ capture or charge exchange.
- Excite system to Rydberg state $\bar{p}AB^*$ before or after formation

Neutral precursor: (1.) $\bar{p} + AB \rightarrow \bar{p}AB + e^-$ (2.) $\bar{p}AB + \gamma \rightarrow \bar{p}AB^*$

Cationic precursor: (1.) $\bar{p} + \bar{p} + AB^+ \rightarrow \bar{p} + \bar{p}AB$ (2.) $\bar{p}AB + \gamma \rightarrow \bar{p}AB^*$

Anionic precursor: (1.) $AB^- + \gamma \rightarrow AB^{-*}$ (2.) $\bar{p} + AB^{-*} \rightarrow \bar{p}AB^{-*} + e^-$

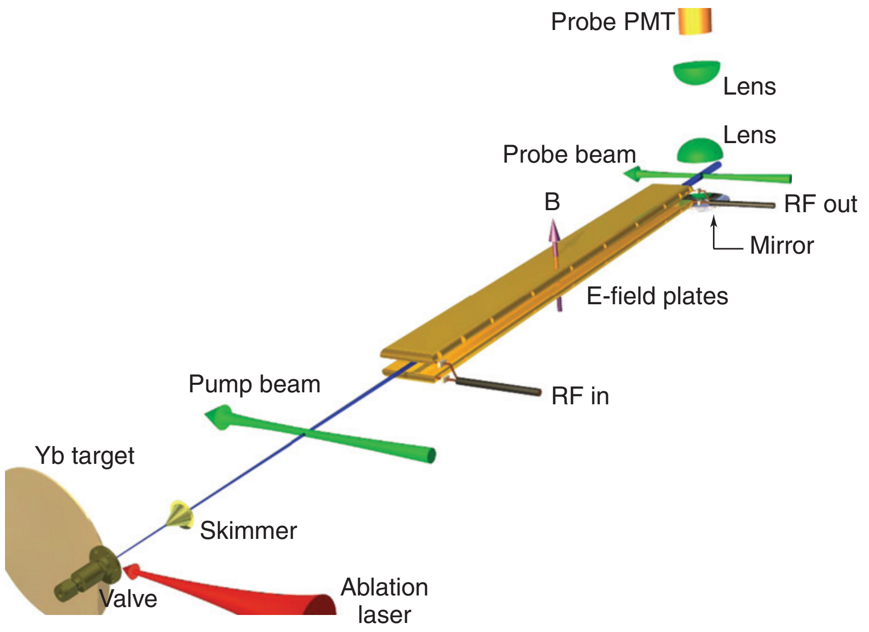
Implementation – Experimental schemes

in Penning trap

- measurement in same trap as formation of $\bar{p}$AB*
- manipulate quantum state in time (laser, mw pulses)
- (-) issue: strong magnetic field
- (-) accurate and precise co-magnetometer needed

in flight

- eject $\bar{p}$AB* (similar as for $\bar{H}$ in gravity measurement)
- manipulate quantum state in space
  (separated field regions, state-selective magnetic deflection)
- similar to classical Ramsey separated-field approach
- (+) magnetic shielding possible
- (+) better access for manipulations
- (-) losses due to transport

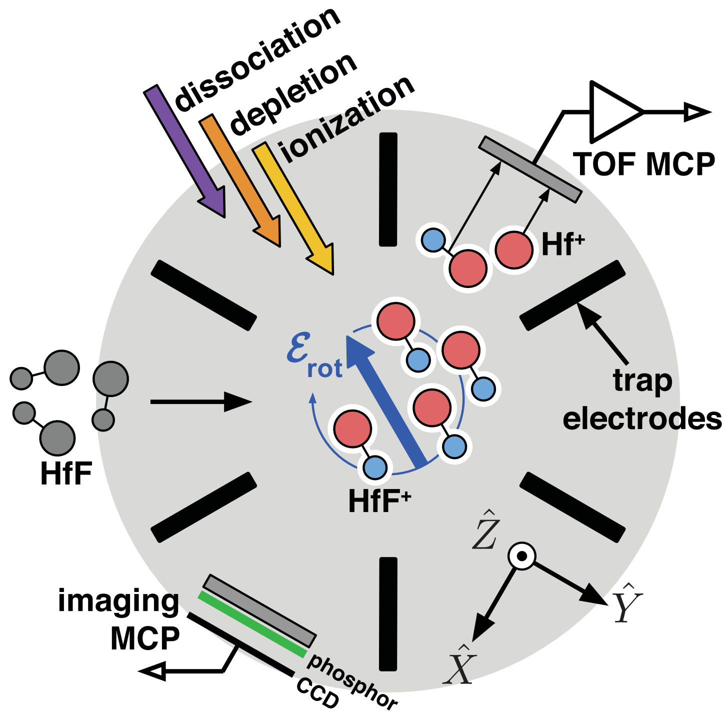


T. E. Chupp et al., Rev. Mod. Phys. 91, 015001 (2019)

Implementation – Experimental schemes

in dedicated RF trap

- eject ionic p̅AB-*
- re-trap in RF trap,
- which has superimposed rotating electric field to polarize molecule
- analogous to HfH^+ e^--EDM measurement at JILA/NIST in Bolder (Jun Ye and Eric Cornell)
- (+) magnetic shielding possible
- (+) long interrogation time (only limited by lifetime)
- (-) most complex



W. B. Cairncross et al. Phys. Rev. Lett. 119, 153001 (2017)

How to proceed?

Theory studies

- Calculate typical achievable energy shift for realistic external electric field.
- Identify suitable molecule AB for the core of the Rydberg-$\bar{p}$-AB system.

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- Possible approaches:
  - Frozen core approximation: calculate molecular core with a Hartree-Fock self-consistent field approach,
    calculate Rydberg $\bar{p}$ state as single-particle problem in the field of the core
    (has been applied successfully for muonic molecules [1])

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→ Establish collaboration with theoretical molecular physicist (for assistance in calculations).

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How to proceed?

Implementation studies

- work out implementation in more detail
- estimate systematic uncertainties
- estimate measurement time to reach a given statistical uncertainty

  → derive achievable constraint for $\bar{p}$-EDM

➔ Assess feasibility of project!

Conclusions

- **Electric dipole moments of subatomic particles open up a route to study beyond-Standard-Model physics.**

- **Proton-EDM: least constrained of p, n and e^-.**

- **Storage-ring experiments for new p-EDM measurements are currently emerging.**

- **Experiments with**
  bound-state $\bar{p}$
  utilizing spectroscopic and
  quantum technologies
  might offer and alternative route to constrain the p-EDM (assuming CPT symmetry) or, alternatively, to a CPT test.

Cold anions – Borealis experiment and C_2^-@AEgIS

Matthias Germann

18.12.2024

Outline

- Goal of anion cooling experiments
- Current issues with the Borealis setup
- Experimental work during the last months
- Plans for C_2^-@AEgIS: evaporative cooling in a Penning trap
- Technical feasibility for evaporative cooling at AEgIS
- Resources-related feasibility for evaporative cooling at AEgIS
- Conclusions and discussion

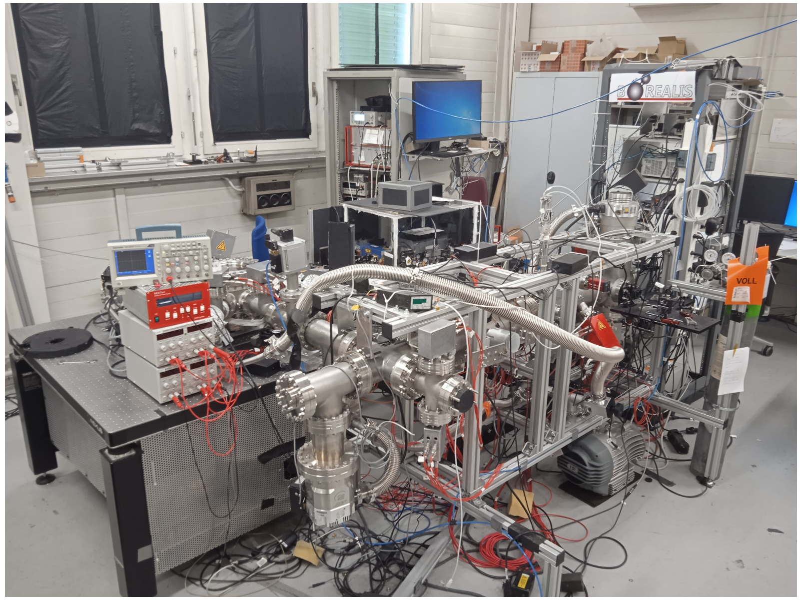
Goal of anion cooling project

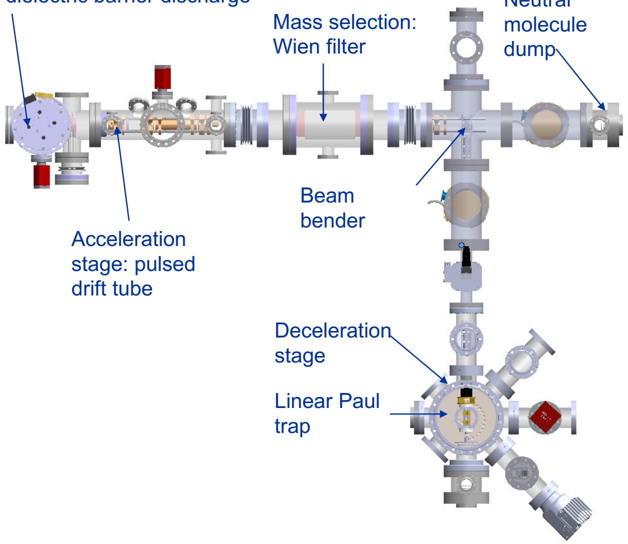
In a nut shell:

- **Laser cool anions (i.e., C_2^-) to $T \leq$ o(100mK).**
- **Co-trap these C_2^- ions with $\bar{p}$.**
- **Cool $\bar{p}$ through exchange of thermal energy via Coulomb interaction: sympathetic $\bar{p}$ cooling.**
- **Use cold $\bar{p}$ to synthesize cold $\bar{H}$**
- **Achieve thus**
  - a smaller transverse $\bar{H}$ emittance and lower axial velocity spread for gravity measurement
  - higher $\bar{H}$ density and lower Doppler broadenings for synthesis and observation of bound antiprotonic systems.

Borealis setup

Separate R&D project within the AEgIS collaboration to achieve laser cooling of anions.

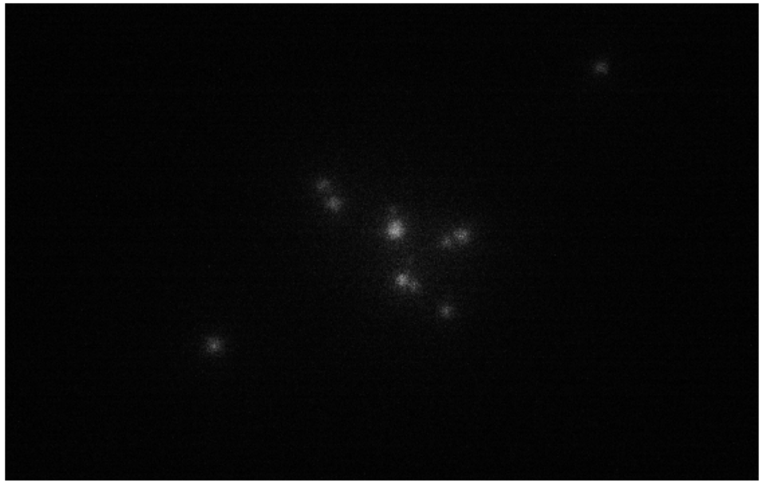


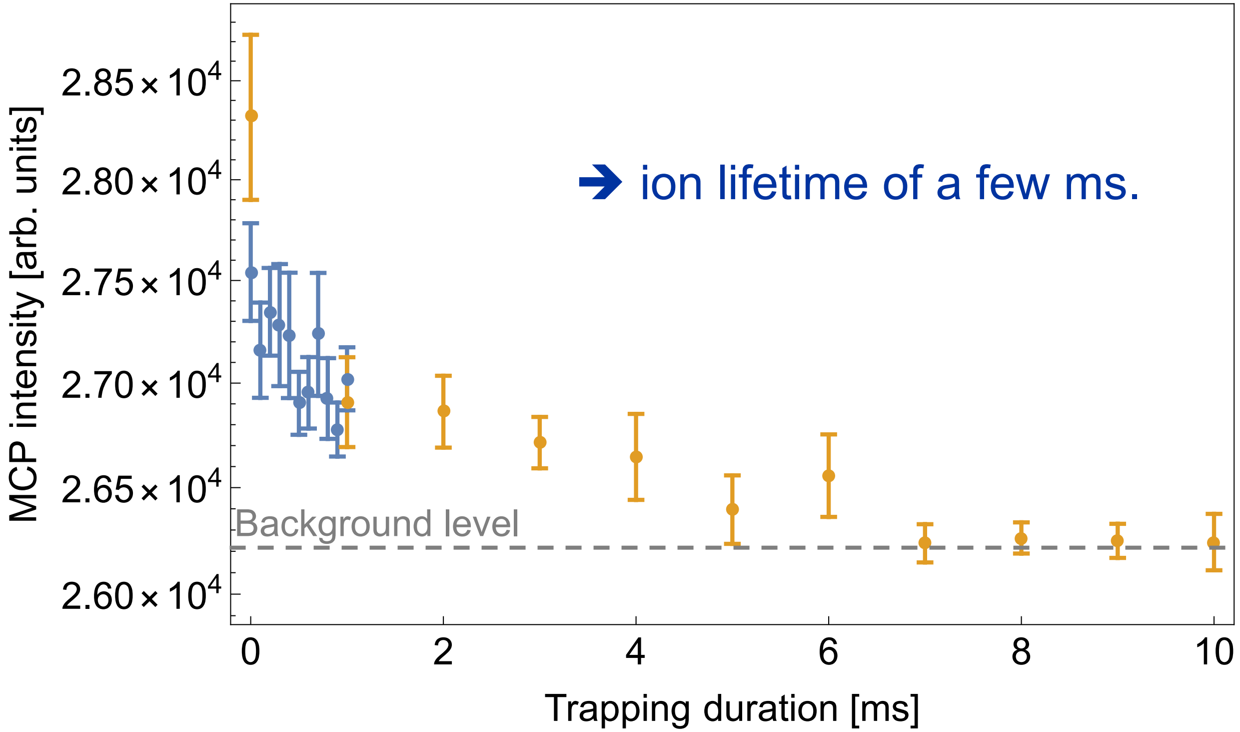


Difficulties with Borealis setup

Number of trapped C_2^- ions is very low: a handful at best

Lifetime of trapped ions is very short: o(ms)





Attempts for improvements

Ion number

- Fixing of ion optical elements
- Beam transport optimization

With help of Frederik Zielke
(BSc thesis TU Dortmund)

Ion lifetime

- Optimize ion catching efficiency of trap
- Optimize compensation of trap patch potentials
- Study influence of vacuum on ion lifetime:
  Improve vacuum in trap a chamber by ~1 order of magnitude

With help of Carla Scullard
(CERN Summer Student)

Results of improvements

Findings

- Beam transport has been moderately improved.
- Number of ions trapped remains very low: a few at best
- Ion lifetime is not limited by vacuum condition and remained in the range of ms.

Conclusions

- To achieve reasonable numbers of trapped ions (hundreds) and reasonable ion lifetimes (≥ seconds), a substantial re-design, re-development and re-construction of the Borealis setup would be needed.
- Beam line: systematic re-design from the source onwards.
- Decelerator: re-design with very systematic treatment of emittance growth.
- Trapping: higher multipole trap (e.g., octopole) to match trap acceptance to beam emittance.
  efficient and systematic compensation of patch potentials and excess RF micromotion

Alternative – Evaporative cooling at AEgIS

Michael and me discussed these problems a while ago.

Conclusions from this discussion:

- The necessary re-build of Borealis, and
- successful demonstration of Doppler cooling of C_2^-

cannot realistically be achieved in the time remaining of my contract.

Suggestion:

- drop Doppler cooling plans
- focus instead exclusively on (simpler) evaporative cooling
- make use of the existing, time-tested AEgIS setup

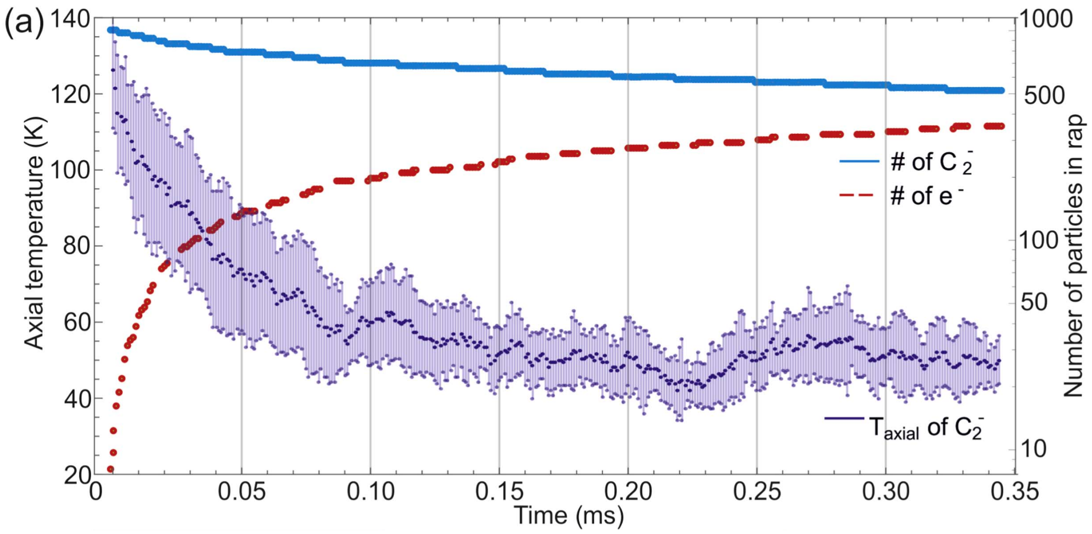
➔ This is a gambit – hopefully a successful one!

Forced evaporative cooling of C_2^- at AEgIS

General idea

- Load C_2^- plasma into the AEgIS 5T trap
- Pre-cool it by electron cooling
- Use velocity-selective laser-induced photodetachement to neutralize the fastest C_2^- ions.
- The fastest (i.e., hottest) C_2^- ions will be lost from the trap.
- The remaining ion ensemble is left in a non-thermal kinetic energy distribution.
- This ensemble will quickly re-equilibrate to a thermal distribution at a lower temperature

➔ We get a colder ion plasma at the cost of a lower number of ions.



Simulation of forced evaporative cooling
[S. Gerber, J. Fesel, M. Doser and D. Comparat, New J. Phys. 20, 023024 (2018)]

Feasibility

Due to the limited time available, we need to be sure about the feasibility of this project before starting it.

Feasibility study:

- **Technical feasibility**
- **Resource-related feasibility**

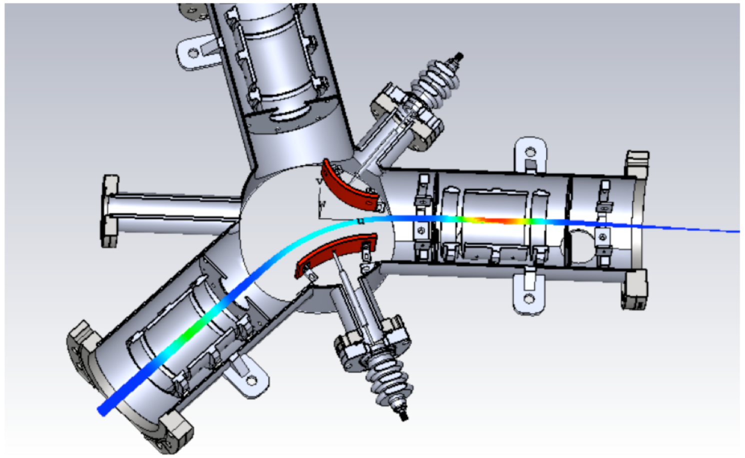
Technical feasibility

Points looked into

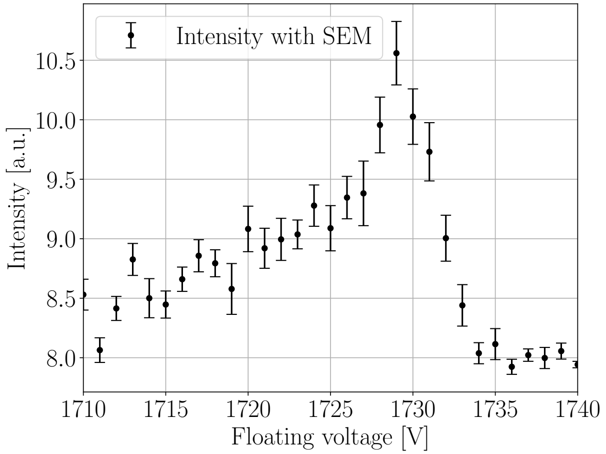
- **Source installation, beam transport, beam energy spread, mass selection, trapping**
- **Laser sources: intensity, linewidth**
- **Detection of ion population**

Source and beam

- Attach source at Starship port.
- Enough space for (bare) source.
- Beam can be transported to trapping region with ~90% efficiency.
- Beam energy spread: o(eV)
  (from previous measurements at Borealis)
  → no (limited) collisional detachment.
- Deceleration and trapping:
  pulsed drift tube decelerator,
  magnetic field: radial confinement
- Mass selection:
  TOF impossible (flight time >> bunch length)
  Wien filter: not enough space, limited resolution and transmittance
  → alternative needed
  → suggestions?

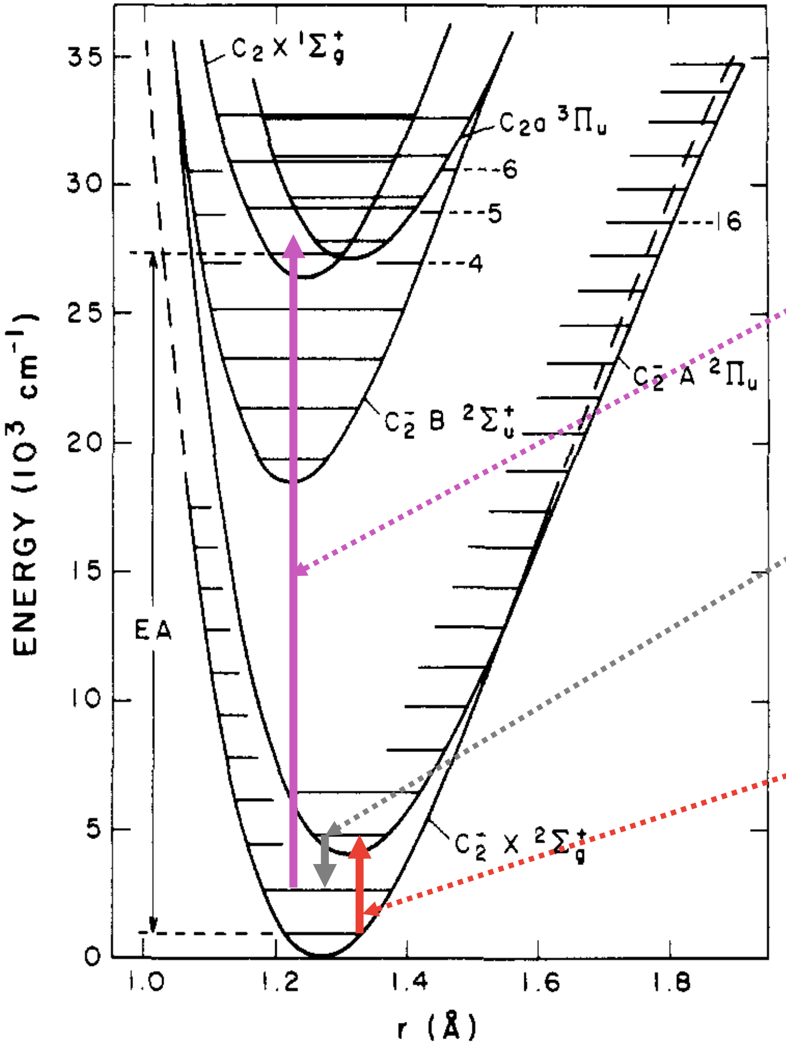


Beam transport simulation
(Thank you Volodymyr for looking into this!)



Trapping efficiency at Borealis
→ energy spread o(eV)

Evaporative cooling at AEgIS



Laser-induced photodetachement
Non-resonant bound-free transition.
380 nm to 405 nm

Radiative decay
from (A, *v*=0) to (X, *v*=1)

Laser excitation from (X, *v*=0) to (A, *v*=0) at 2.53 μm.
Resonant bound-bound → velocity selective

Level scheme from:
K. M. Ervin and W. C. Lineberger
J. Phys. Chem. 95, 1167 (1991)

Thank you to Giovanni for pointing to the possibility to allow for decay to (X, *v*=1)!

Laser sources and wavelength meter

Two cw diode lasers (Toptica DL100) available from Borealis setup:

2.53 µm: 5 mW

400 nm: 130 mW

Linewidth ≤ 3 MHz

Wavelength meter

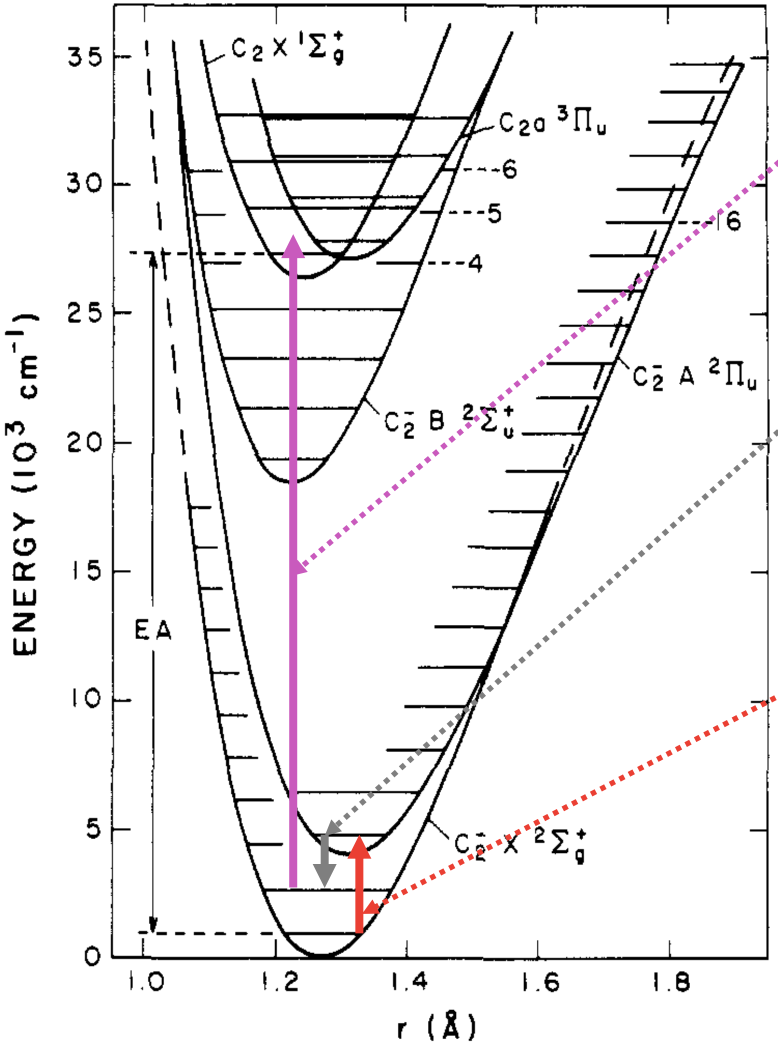
Burleigh WA-1500 IR

Range: 1.5 – 4 µm

Absolute accuracy: ±0.3 ppm (36 MHz at 2.53 µm)

Display Resolution: 10 MHz

Laser intensity and photodetachment rate



Laser-induced photodetachement
$\Gamma_{pd} \approx 1\ s^{-1}$

Radiative decay
$A = 1.4*10^3\ s^{-1}$

Saturation intensity: I_{sat} = o(mW/m^2)
Laser intensity: I_{laser} = o(kW/m^2)
→ Transition saturated.
→ Half of the population in upper level
(actually: ¼ due to spin-rot. splitting)

➔ Overall PD rate is limited by bound-free transition.
➔ Rate is ≈ 1 s^{-1} ion^{-1}

Doppler width

Doppler width of the resonant transition at 2.53 µm

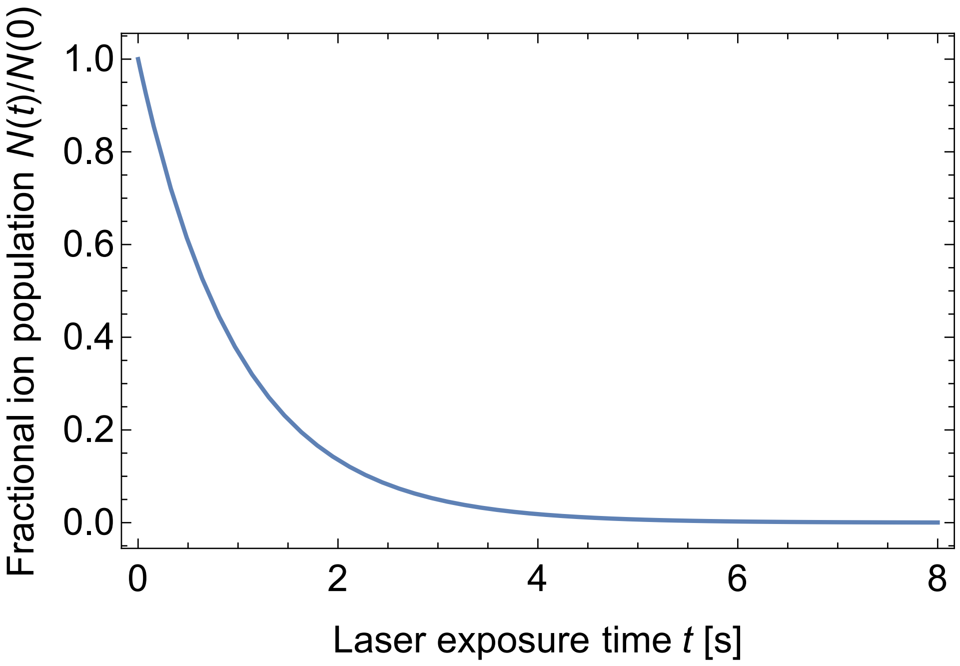
Ion temperature	**FWHM width**	**Comment**
100 K	170 MHz	e^- cooling, upper limit
10 K	55 MHz	e^- cooling, lower limit
1 K	17 MHz	after evaporative cooling (?)

➔ Wavelength stabilization by Burleigh WA-1500 IR wavemeter seems sufficient for a first demonstration.

Detection

Destructive detection of C_2^- ions

- by ejection on axial MCP after end of experiment
- half life of C_2^- when exposed to both laser beams: o(1s)
- decrease in ion number should be detectable after few seconds
  (assuming an ion lifetime without laser interaction ≫ 1s)
- uncertainties should be reasonably low, assuming o(100) ions and Poissonian statistics.



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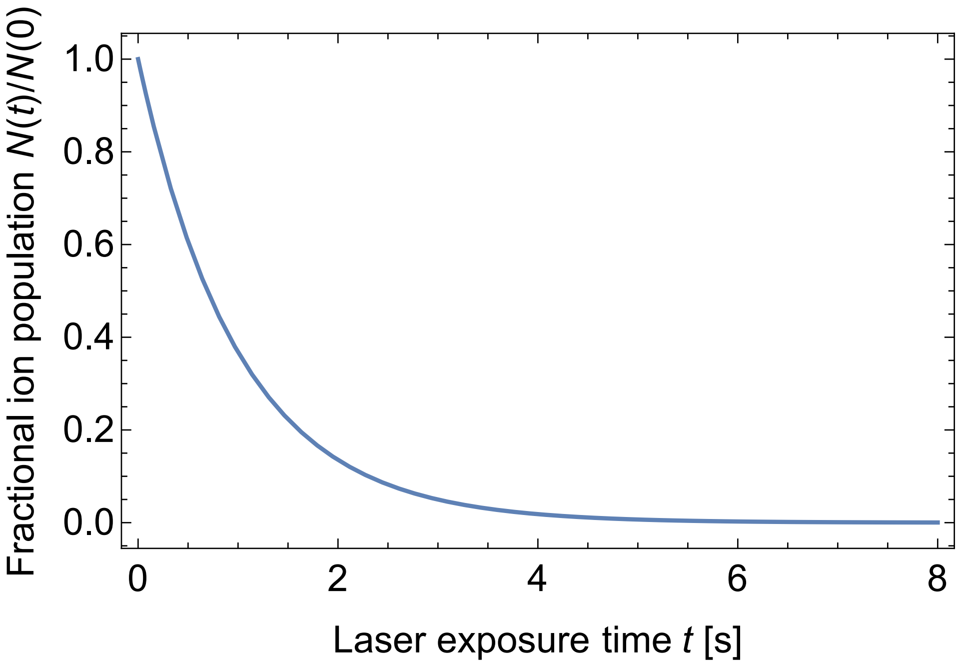

➔ Conclusion from technical feasibility study:
Project should be technically feasible. Mass selection remains a concern.

Resource-related feasibility

List of work packages and milestones

WP 1 – Source operation and beam formation

WP 2 – Anion trapping

WP 3 – Electron cooling

WP 4 – Spectroscopy and thermometry

WP 5 – Forced evaporative cooling

WP 1 – Source operation and beam formation

Tasks
- Source installation (mounting, vacuum interfacing, electronics)
- Source operation
- Beam formation

Materials
- Mounting frame
- Vacuum parts (bellows, nipples, etc.)
- HV supply
- Trigger signals (pulse/delay generator)

Challenges
- Mass selection: ion bunch length too long for TOF mass separation,
- insufficient space to install Wien filter

Milestone

➔ Beam of pure C_2^- is formed and transported to the trap region.

WP 2 – Anion trapping

Tasks

- Beam deceleration: decelerate C_2^- beam from 2 keV to ~1 eV with pulsed drift tubes.
- Trap loading: loading of C_2^- ions into the 5T trap.
- Ejection and detection: eject ions on MCP, quantify number of trapped ions

Materials

- Anything?

Challenges

- Deceleration: emittance growth, losses
- Trapping: matching of phase-space volume of beam to trap (→ ion losses)

Milestone

➔ Reproducible and reliable procedure for loading C_2^- ions into 5T Penning trap developed.
Number of trapped ions and ion lifetime quantified.

WP 3 – Electron cooling

Tasks

- Load electrons into 5T trap
- Mix e^- and C_2^- plasmas
- Rough ion thermometry through escape-barrier potential scan

Materials

- Anything?

Challenges

- Efficient mixing of two-component plasma (simple? complex?)
- Collisional detachment of C_2^-

Milestone

➔ C_2^- ions cooled to o(100 K) by electron cooling.
Ion temperature (roughly) quantified by escape-barrier potential scan.

WP 4 – Spectroscopy and thermometry

Tasks

- Install diode lasers from Borealis setup at AEgIS Lighthouse
- Transport laser light to AEgIS through fibers
- Insert laser beams into AEgIS vacuum chamber
- Lock laser frequency with PID feedback to wavemeters
- Demonstrate photodetachment (*hν* » threshold)
- Record photodetachment spectrum (*hν* ≈ threshold)
- Derive ion temperature from Doppler profile

Materials

- Optics (fibers, couplers, collimators, mirrors, lenses)
- UV wavemeter (only low resolution needed)
- PID regulator

Challenges

- Very limited space close to vacuum vessel (for optics)
- Collimation of IR laser beam
- Stable locking of IR laser frequency
- Efficient detection of photodetachment events

Milestone

➔ Ion temperature determined from Doppler thermometry.

WP 5 – Forced evaporative cooling

Tasks

- Increase of laser power or laser exposure time for velocity selective photodetachment.

Materials

- Likely nothing in addition to material for WP 4

Challenges

- Purity of ion plasma (impurity ions are not cooled)
- Exchange of kinetic energy within plasma
- Coupling between normal modes of plasma

Milestone

➔ C_2^- ions cooled significant below electron cooling temperature by evaporative cooling through velocity selective photodetachment.

Requirements

- **The AEgIS setup is complex.**
- **Implementing anion cooling at AEgIS cannot be realized by one person alone: team effort needed**

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Support needed:

- Construction: integration of C_2^- source into AEgIS (CAD drawings, machining, vacuum setup, etc.)
- Operation: adaption of control system, development of scripts, etc.
- Laser optics: setup of lasers, optics and fiber links, stabilization of laser frequency
- Simulation: C_2^- beam transport, deceleration and loading of C_2^- into trap
- Time: impossible to realize all WPs before $\bar{p}$-run
  → access to AEgIS needs to be coordinated with other experiments

Borealis 2.0?

- I am still very much interested in continuing to pursue the Doppler cooling attempts of C_2^- in a dedicated setup.
- My curiosity/stubbornness/perseverance let's me not to simply give up this effort.
- The difficulties encountered at the Borealis setup are of technical nature.
- They can be overcome with sufficient time and resources.

➔ I am thinking about a redesign of the Borealis setup, as a longer-time project.

Conclusions

- Borealis setup shows a number of issues that can only be solved with a systematic, long-term re-design, re-development and re-construction of the setup.
- Those need to be addressed before anion Doppler cooling at Borealis can be attempted.
- Alternative: attempt only evaporative cooling (less complex),
  make use of time-tested AEgIS setup
- This project is more promising than Doppler cooling at Borealis setup, but still very ambitious.
- The project seems technically feasible.
- It might be feasible w.r.t. reasources if the necessary time and support is provided: Needs a team effort.
- Know-how acquired is beneficial for future anion activities (iodine ion source).

Zeeman structure (at 1T)

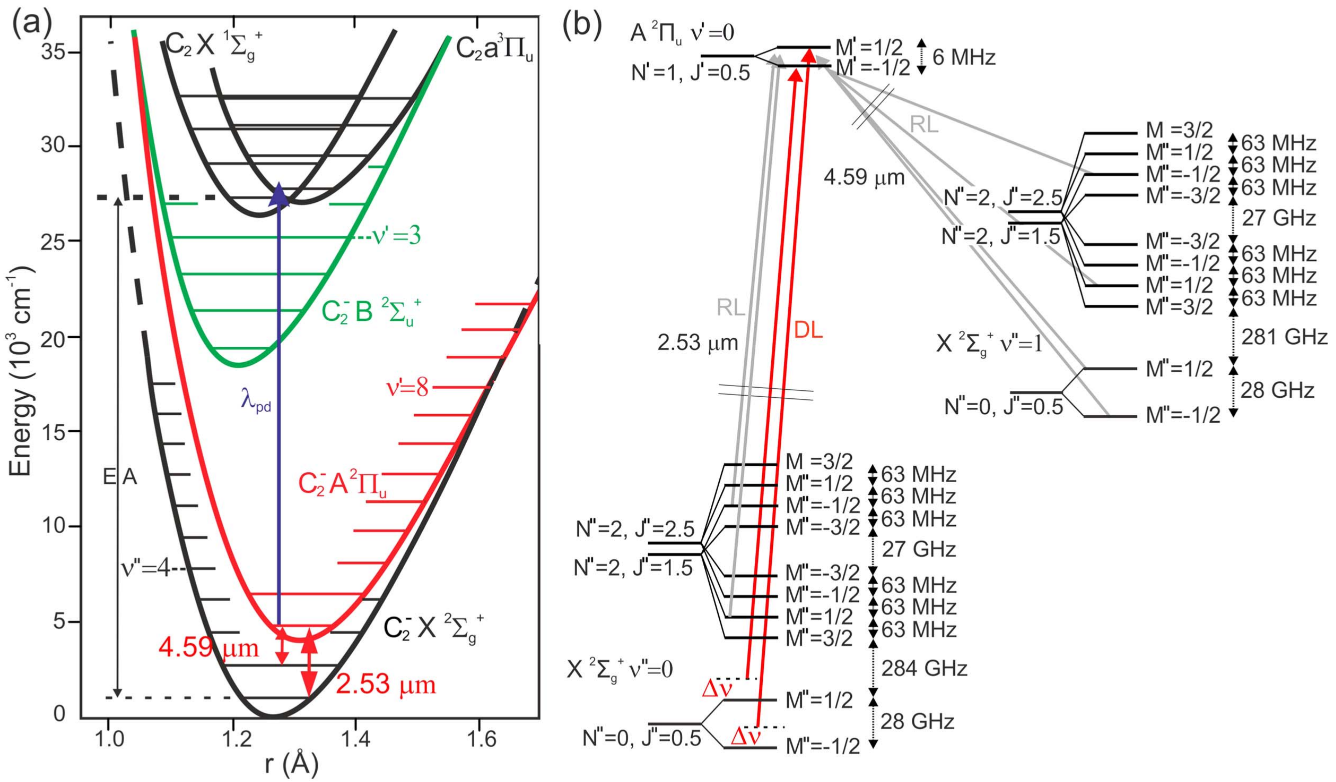

Figure 1. (a) Molecular potential energy of C_2^- versus internuclear separation with the electronic and vibrational levels including two neutral C_2 ($X^1\Sigma_g^+$, $a^3\Pi_u$) curves [37]. The *X*–*A* (red) cooling transitions and the photodetachment (blue, λ_{pd}) transition are indicated with arrows. (b) Zeeman splitted vib–rot sublevels in a 1 T field showing the laser for the Doppler cooling scheme. The electron spin $\frac{1}{2}$ is coupled to the rotational quantum number *N* to form the full angular momentum *J* and its projection *M* on the magnetic field axis. The $\nu'' = 0$ and $\nu'' = 1$ manifold of the *X* state and the excited *A* states are shown (not to scale). The two Doppler cooling lasers (DL, red) addressing the ground states at 2.54 μm are depicted with their detunings $\Delta\nu$. The six repumping lasers are sketched (RL, gray) at 2.54 μm and 4.59 μm, respectively. Reproduced from [25]. CC BY 4.0.

Thermal energy in eV

Ion temperature	$k_B T$	Comment
100 K	8.6 meV	e^- cooling, upper limit
10 K	0.86 meV	e^- cooling, lower limit
1 K	0.086 meV	after evaporative cooling (?)

Sympathetic Cooling of Trapped Negative Ions by Self-Cooled Electrons in a Fourier Transform Ion Cyclotron Resonance Mass Spectrometer

Guo-Zhong Li, Shenheng Guan, and Alan G. Marshall
Center for Interdisciplinary Magnetic Resonance, National High Magnetic Field Laboratory, Florida State University, Tallahassee, Florida, USA

Hot electrons confined in a Penning trap at 3 tesla self-cool to near room temperature in a few seconds by emission of cyclotron radiation. Here, we show that such cold electrons can "sympathetically" cool, in ~10 s, laser desorbed/ionized translationally hot Au^- or C_{70}^- ions confined simultaneously in the same Penning trap. Unlike "buffer gas" cooling by collisions between ions and neutral gas molecules, sympathetic cooling by electrons is mediated by the mutual long-range Coulomb interaction between electrons and ions, so that translationally hot ions can be cooled without internal excitation and fragmentation. It is proposed that electrosprayed multiply charged macromolecular ions can be cooled sympathetically, in the absence of ion-neutral collisions, by self-cooled electrons in a Penning trap. (J Am Soc Mass Spectrom 1997, 8, 793–800) © 1997 American Society for Mass Spectrometry

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