Sympathetic cooling of rotationally and vibrationally state-selected molecular ions

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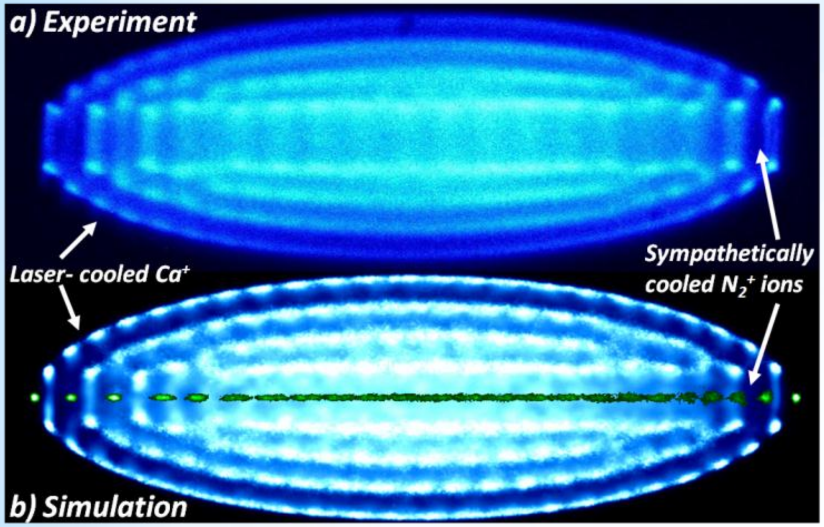
Motivation

Localized (translationally cooled) molecular-ions in well-defined internal states are of broad interest for physics and chemistry

- State-selected ultracold collision studies
- Precision spectroscopy on single molecular ions
- Quantum computing/molecular-ions qubits
- Molecular internal state control experiments
- Ultracold molecular plasmas
- ……

Sympathetic Cooling and bi-component Coulomb Crystal



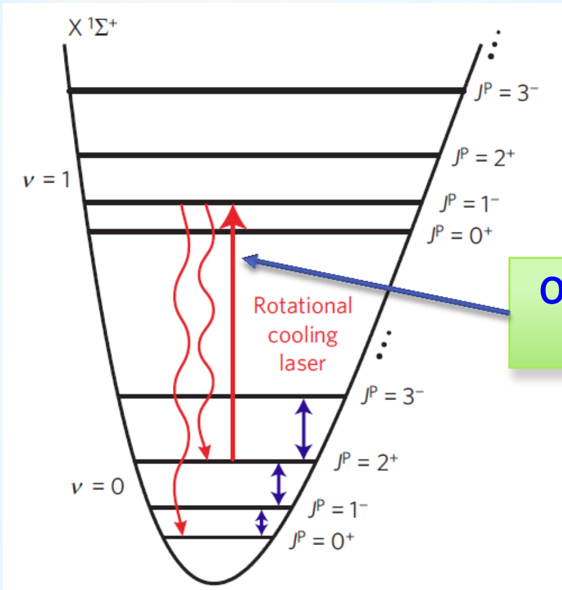
Fluorescence images of a Ca^+/N_2^+ bi-component Coulomb Crystal.

N_2^+ loading movie

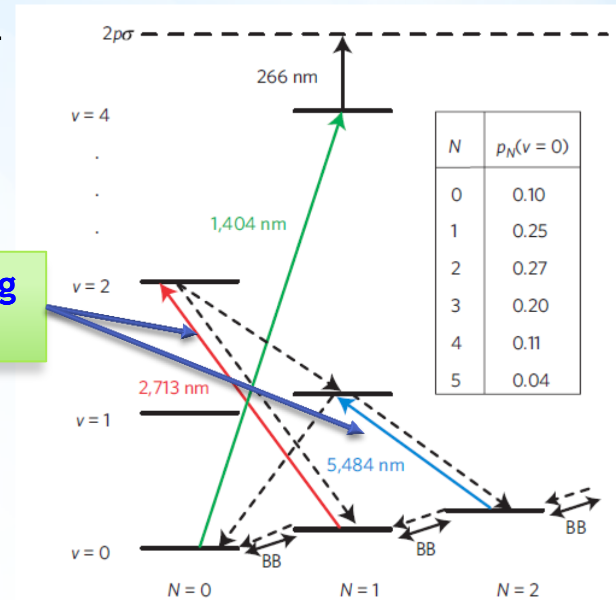
- The N_2^+ ions are only indirectly visible as a dark core.
- Image contains 925±25 Ca^+ and 24±1 N_2^+ ions with averaged thermal energies of 8±1 mK for Ca^+; 11±1 mK for N_2^+ determined by Molecular Dynamics simulation.
- Ions sympathetically cooled from a "warm" sample are translationally cold, but not quantum-state selected.

First approach: Optical pumping of translantionally cold, internally warm molecular ions

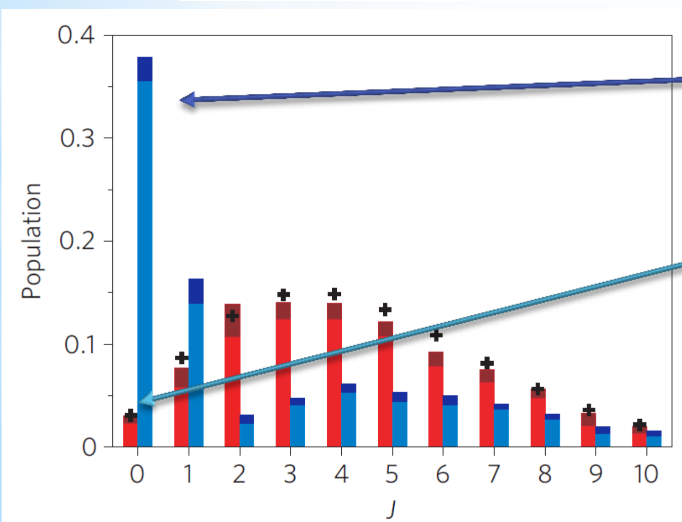
MgH⁺



HD⁺

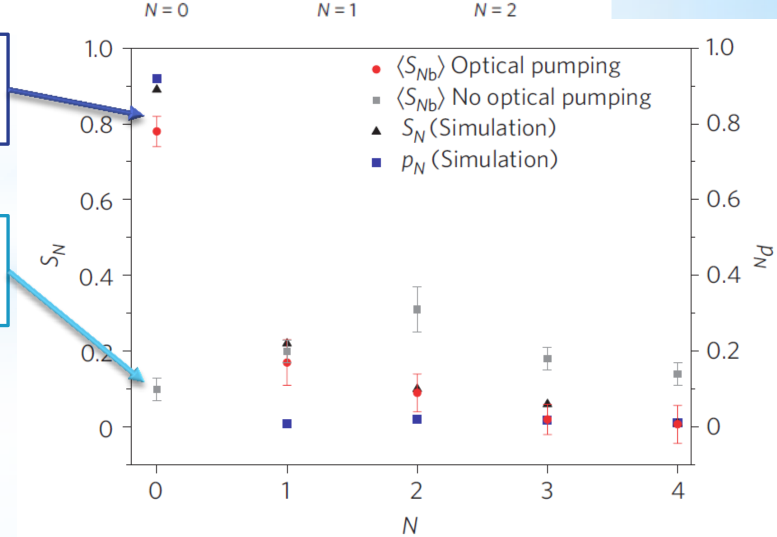


Optical Pumping Laser(s)



After Optical Pumping

Before Optical Pumping



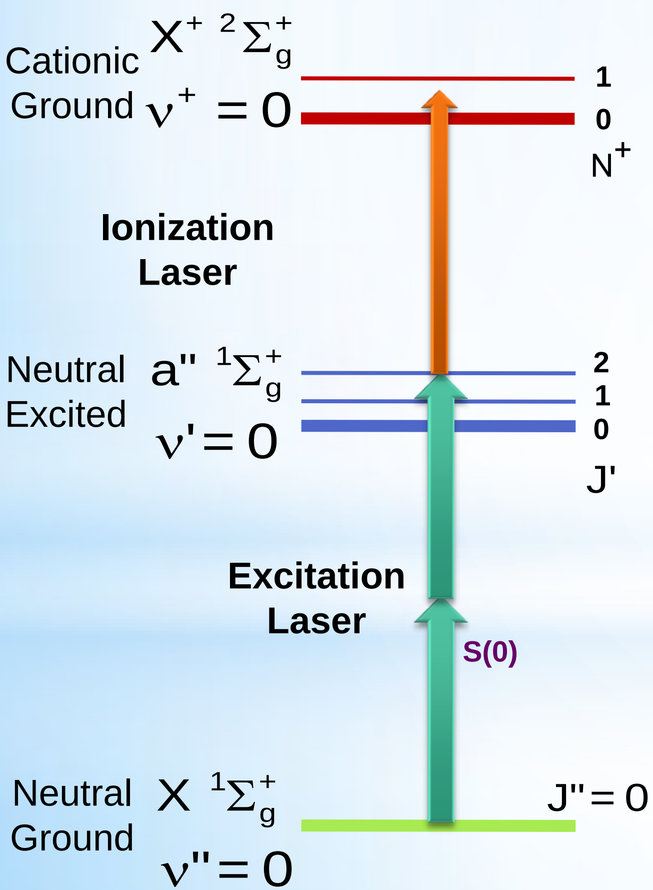
Staanum *et al.*, Nature Physics **6**, 271-274 (2010).

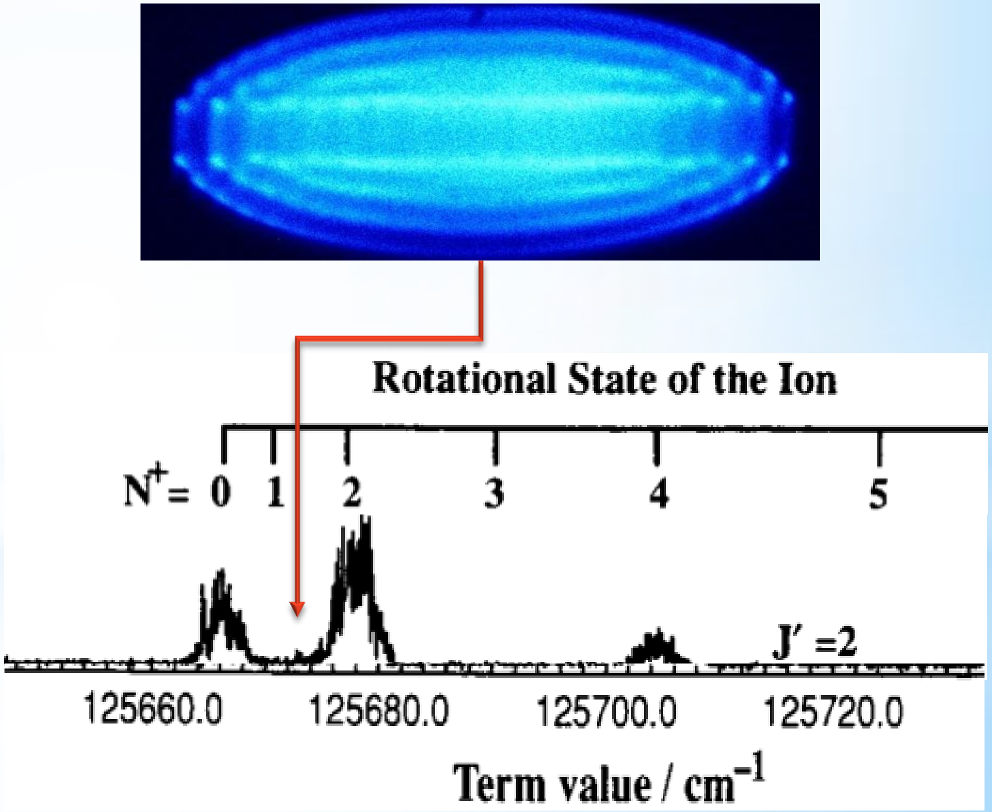
Schneider *et al.*, Nature Physics **6**, 275-278 (2010).

Second (our) approach:
Sympathetic cooling of state-selected Ions (example: N_2^+)

Rovibrational state selection in the cation was achieved by
resonance-enhanced [2+1′] threshold photoionization.

- Rotational propensity rule: $\Delta N = N^+ - J' = 0, \pm 2$.
- Photoionization slightly above the desired ionization threshold.

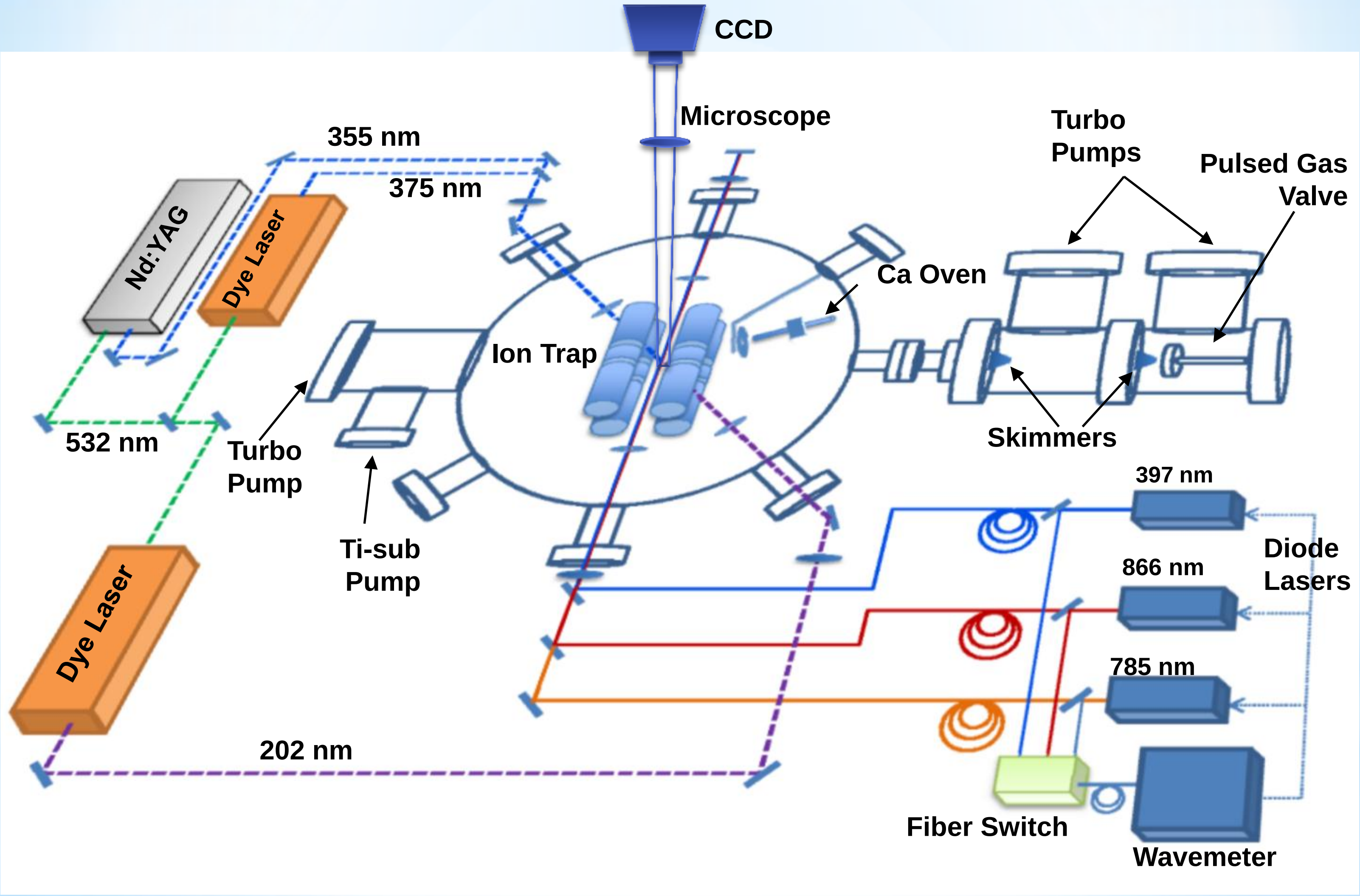




S. Willitsch *et al.*, Int. J. Mass Spectrom. **245**, 14-25 (2005).

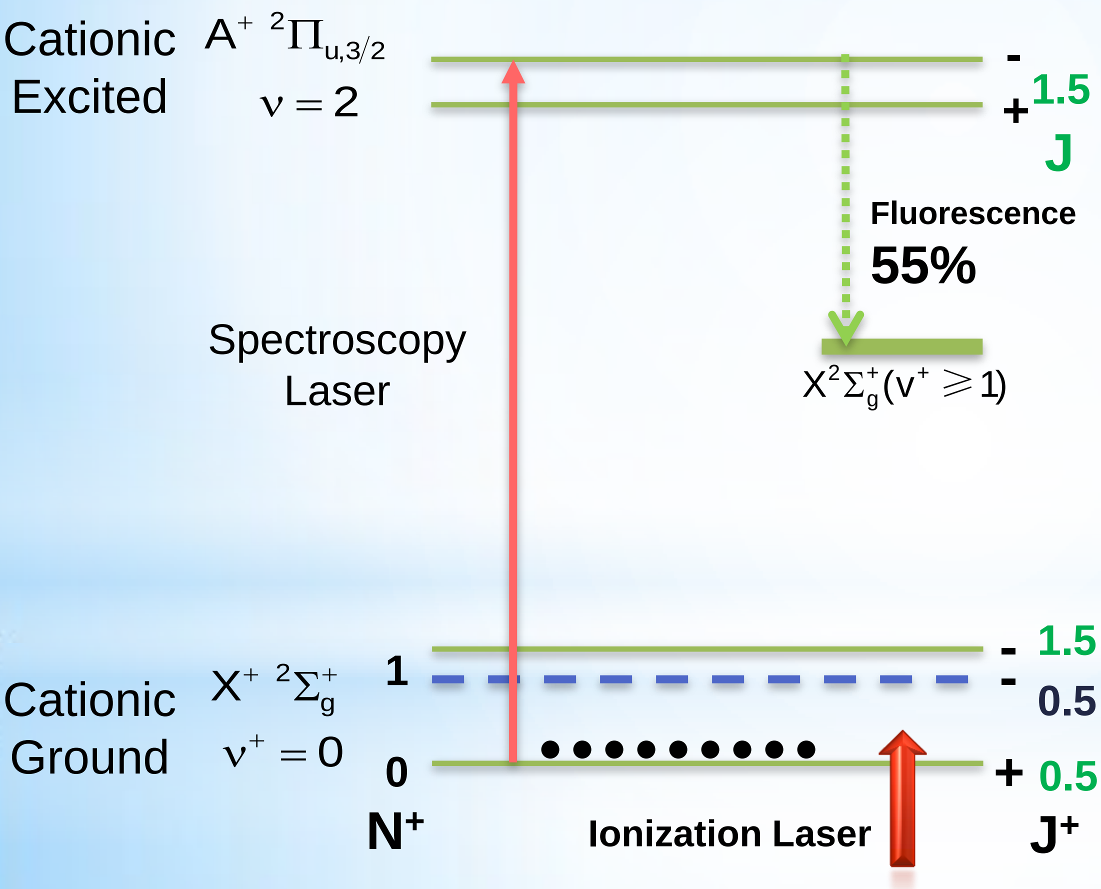
S.R. Mackenzie *et al.*, Mol. Phys. **86**, 1283-1297 (1995).

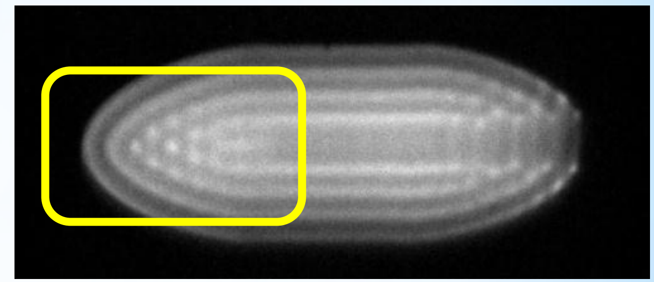
Experimental Setup



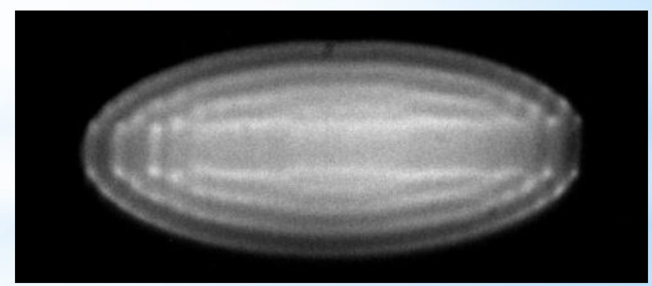
Quantum-State Diagnostics of the sympathetically cooled N_2^+ ions

Rotational-state population was probed using **laser-induced charge-transfer (LICT)** with Ar atoms.





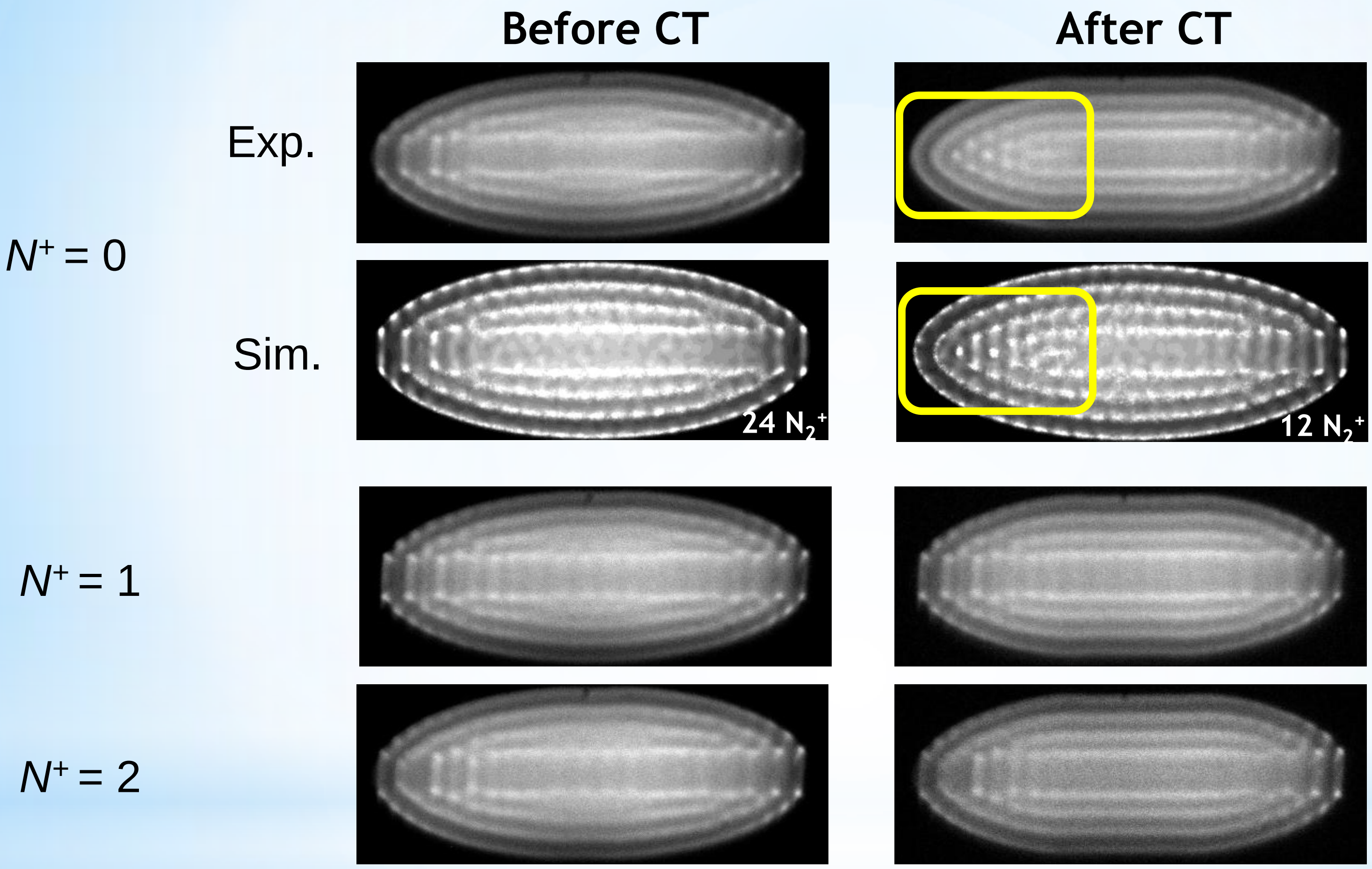
$$N_2^+ + Ar \xrightarrow{k} N_2 + Ar^+ \quad (k = 4\times10^{-10} cm^3 s^{-1})$$



$$N_2^+ + Ar \xrightarrow{k} N_2 + Ar^+ \quad (k < 1\times10^{-12} cm^3 s^{-1})$$

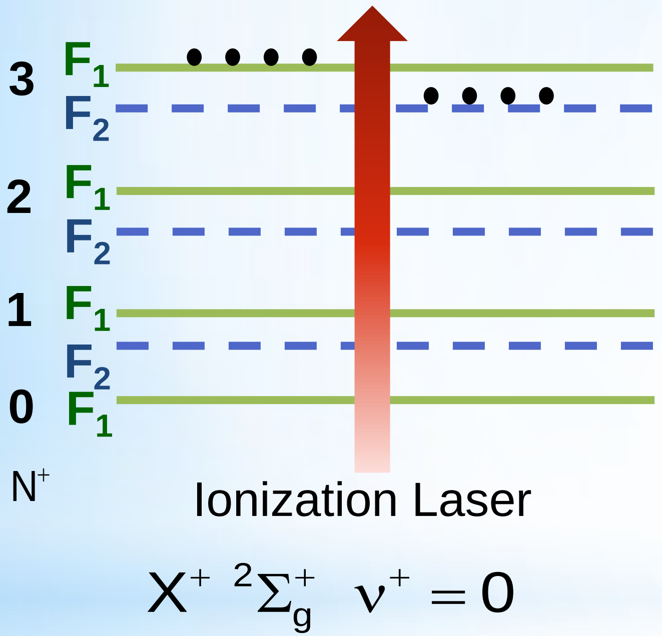
S. Schlemmer, *Int. J. Mass Spectrom.* **185-187**, 589-602 (1999).

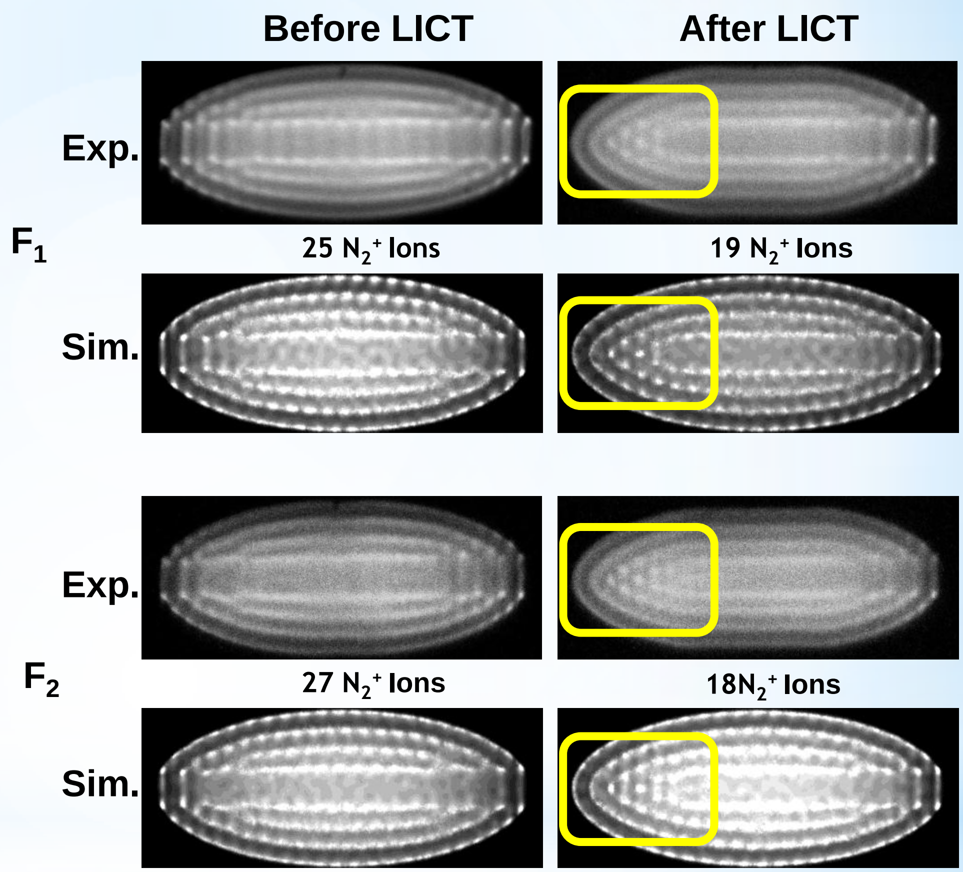
Quantum-State Diagnostics



- LICT efficiency: 51±6% (averaged over five experiments). Max.: 55%
- N_2^+ ions are fully state selective (ground-state population 93±11%).
- No evidence of population observed in $N^+ = 1$ and 2 rotational states.
- The population is preserved in $N^+ = 0$ during the sympathetic cooling process.

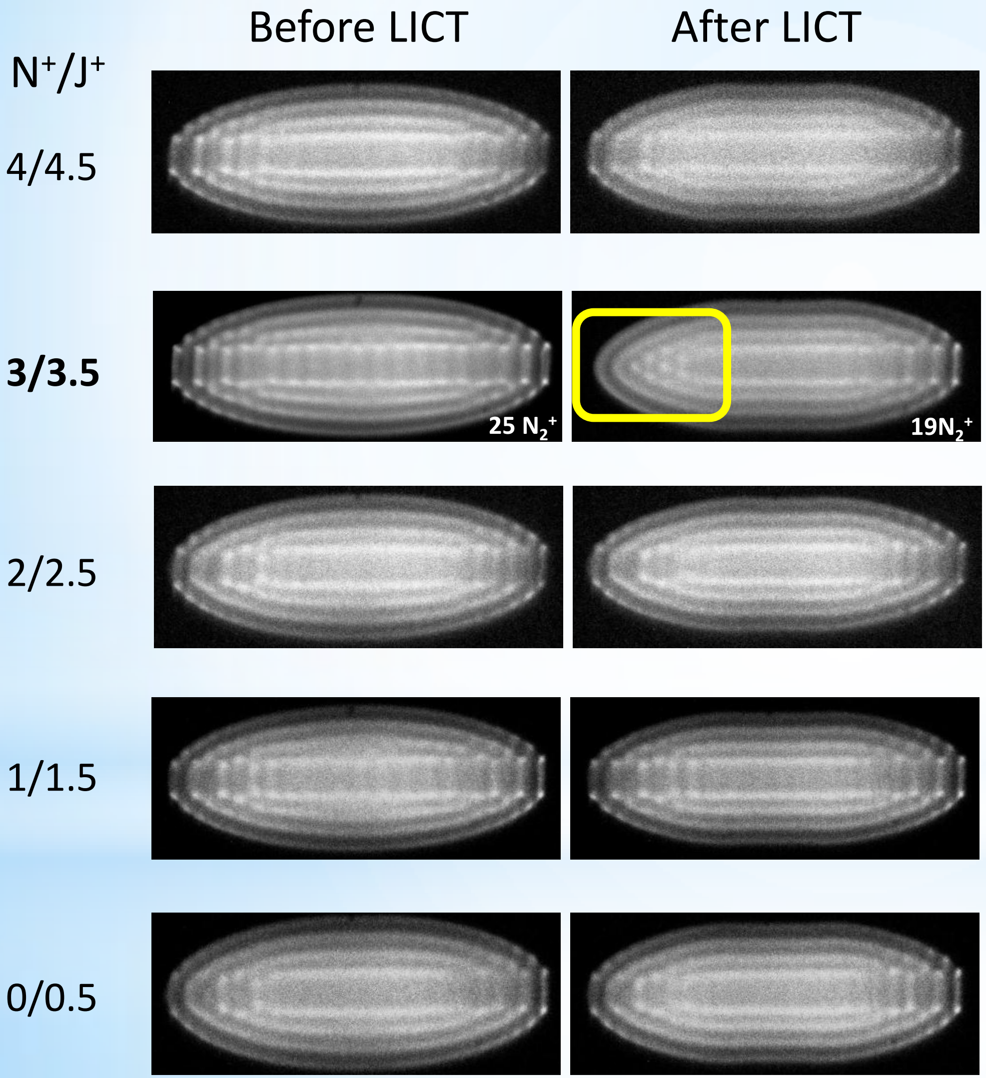
Generation of Rotationally Excited N_2^+ Ions





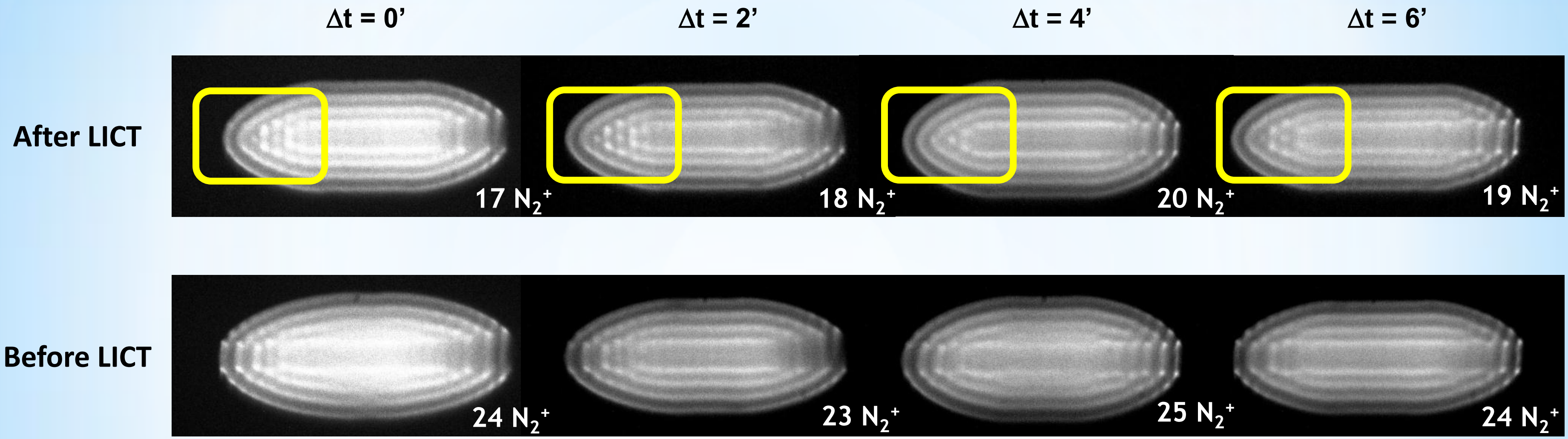
- The LICT efficiencies: 27±7% for F_1 and 28±7% for F_2 (five experiments).
- Both components are produced with equal probability.
- The total LICT efficiency out of $N^+ = 3$ amounts to 55±9%. (Maximum: 55%)

Quantum-State Diagnostics

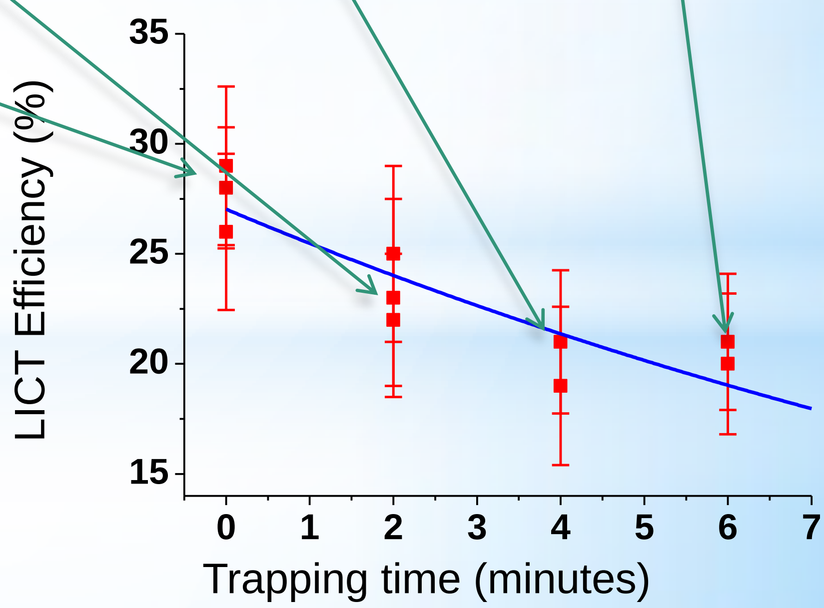


- There is no evidence of population observed in $N^+ = 0, 1, 2$ and 4 rotational states.
- The population is preserved in $N^+ = 3$ during the sympathetic cooling process.

State Lifetimes



- Probing a variable time delay between loading the crystal with N_2^+ ions and LICT.
- The state lifetime is determined to be $\boldsymbol{\tau = 947 \pm 176\ \mathrm{s}}$.
- The redistribution of population is attributed to inelastic and reactive collisions with background gas molecules
  [e.g. $\mathrm{N}_2^+(1) + \mathrm{N}_2(2) \rightarrow \mathrm{N}_2(1) + \mathrm{N}_2^+(2)$]



Conclusion

- Generation of quantum state-selected and translationally cold molecular ions.
- The population is preserved during the sympathetic cooling process.
- States lifetime is on order of 15 minutes limited by collisions with background gas.

X. Tong, A. H. Winney, and S. Willitsch, *Phys. Rev. Lett.* In print (2010). (arXiv 1006.5642)

Outlook

- Molecular ions in selected spin-rotation levels.
- Sympathetic cooling of state-selected polar molecular ions in a cryogenic environment (possible issues: blackbody radiation, dipole collisions)
- State-selected cold chemistry.
- Many others ……

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- Group members

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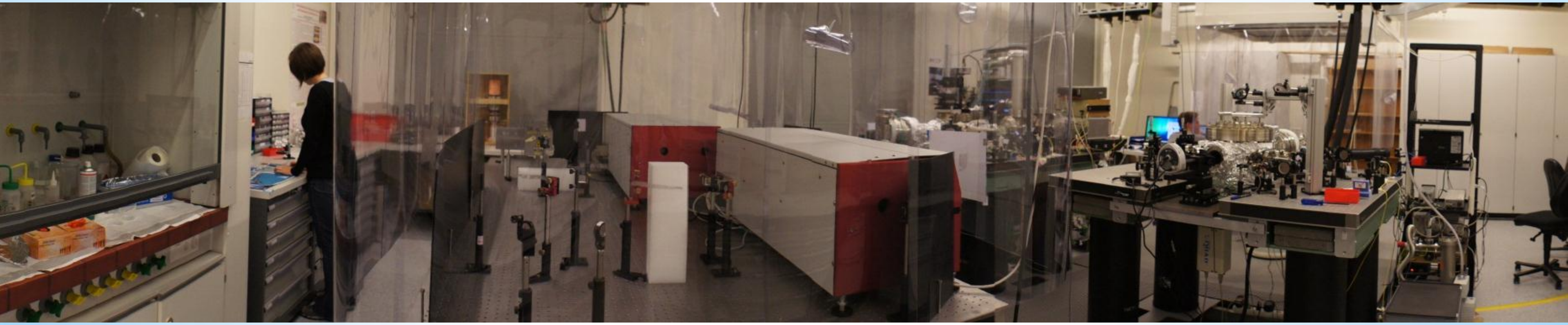

Thank You All