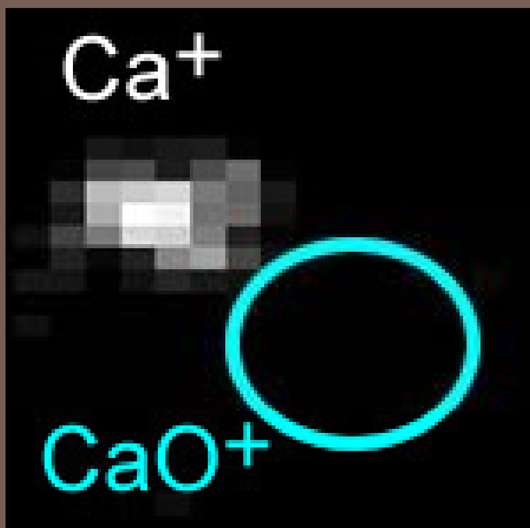
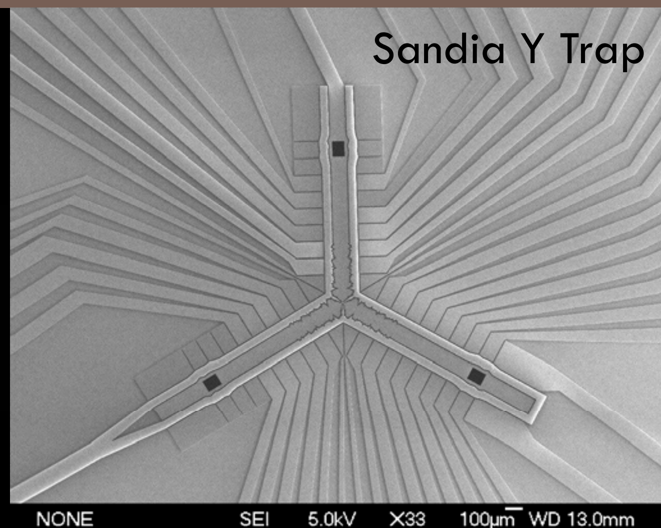
CHALLENGES OF LASER COOLING MOLECULAR IONS

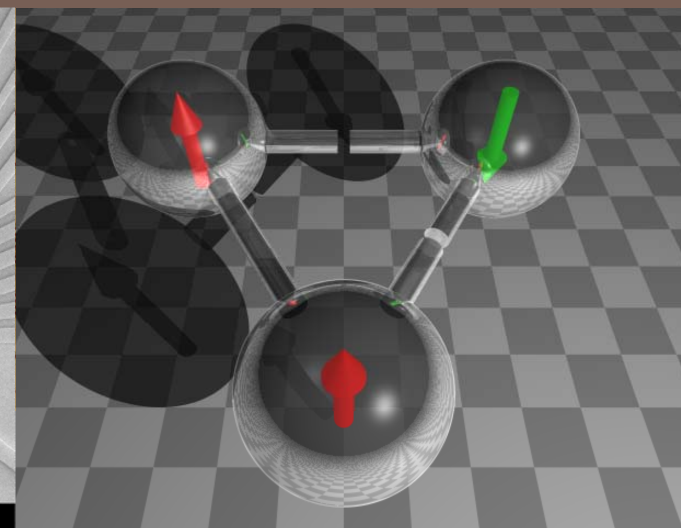
Kenneth Brown, Georgia Tech, Feb. 17, 2011

Cold Molecular Ions

Quantum Information







QIQC
QUANTUM INFORMATION FOR QUANTUM CHEMISTRY

NSF

MUSIQC

IARPA

http://j.mp/brownlab

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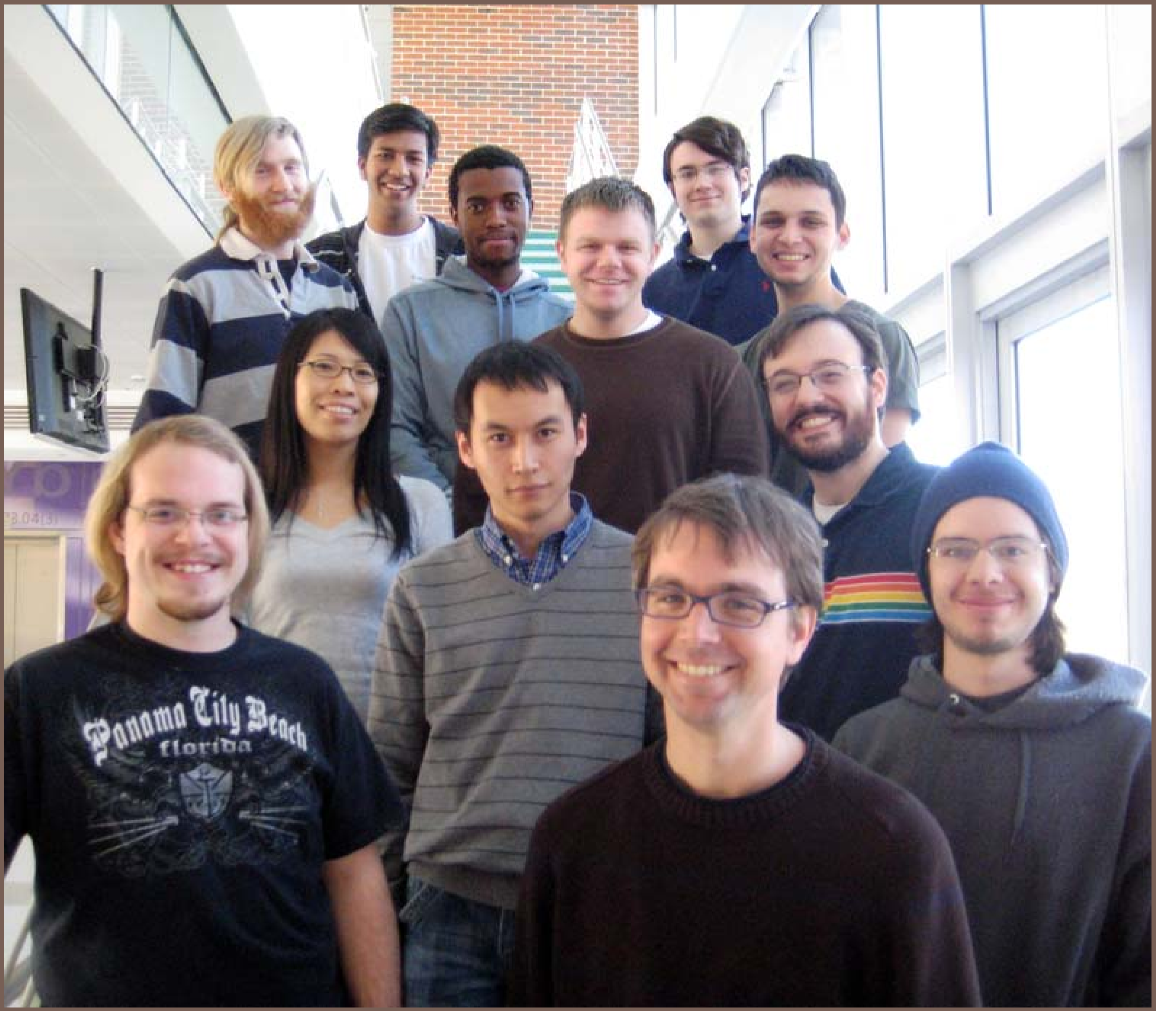

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→Entanglement Technologies

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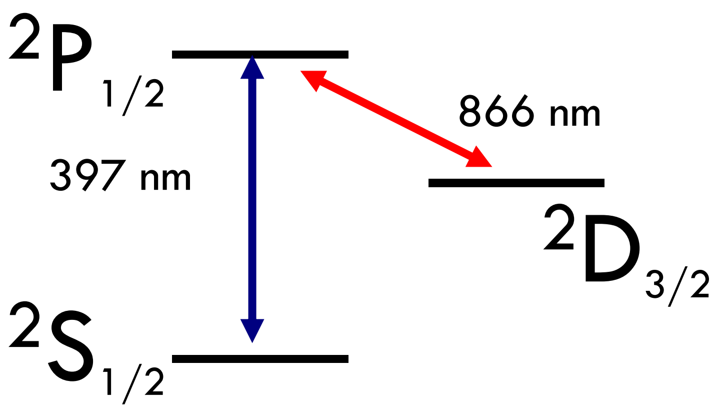
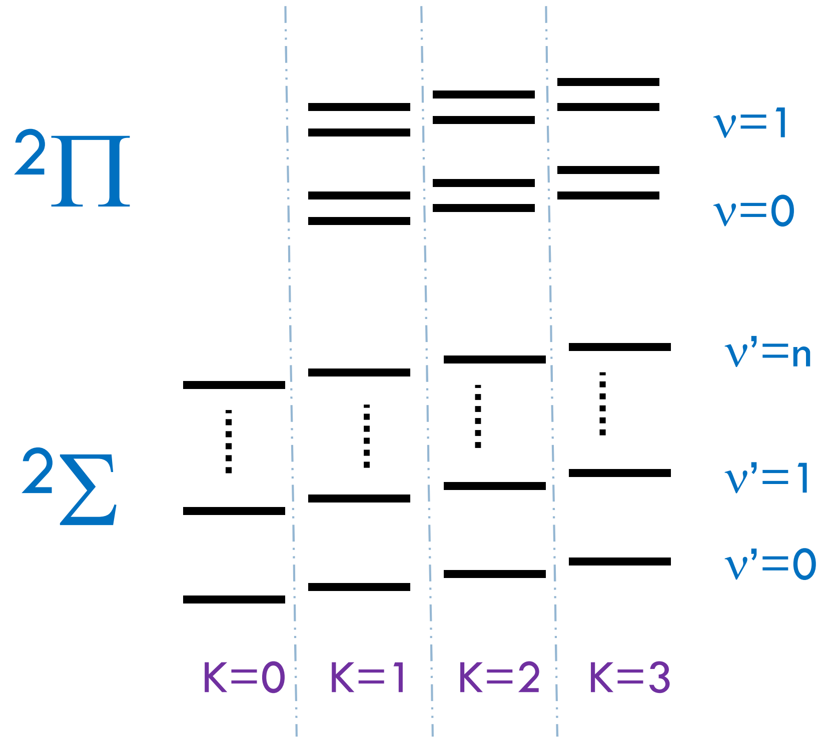
Dr. Jason Nguyen
Prof. David Sherrill
Ed Hohenstein

Challenges of laser cooling molecular ions, arXiv:1102.3368

What challenges?

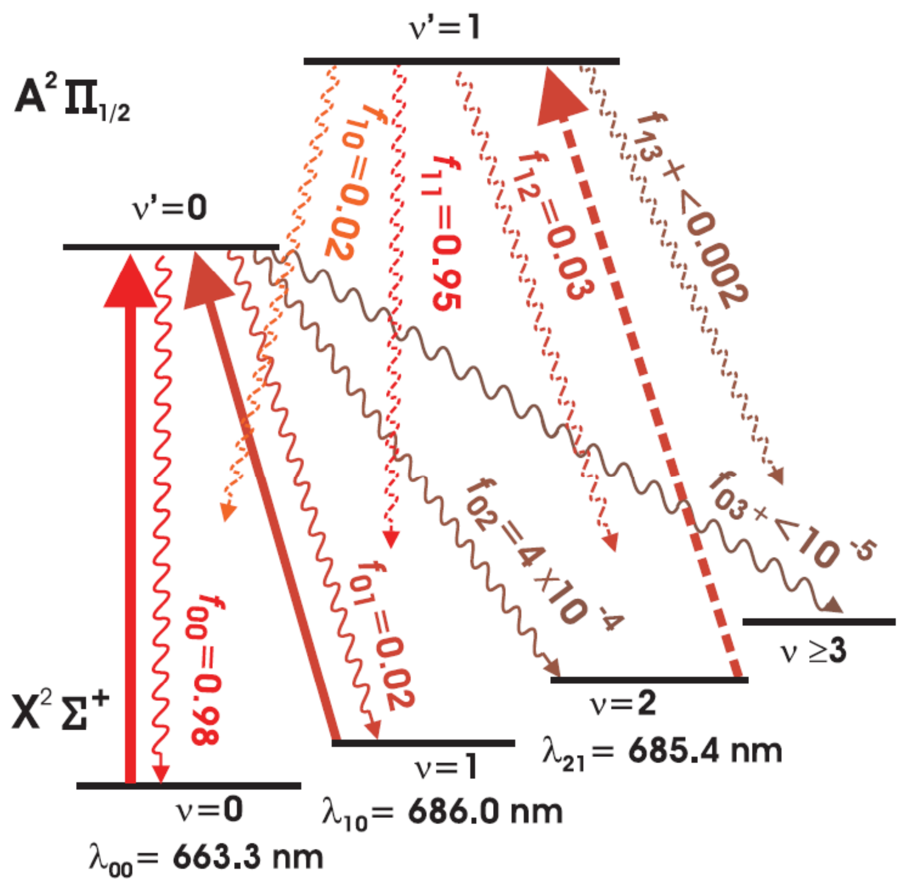


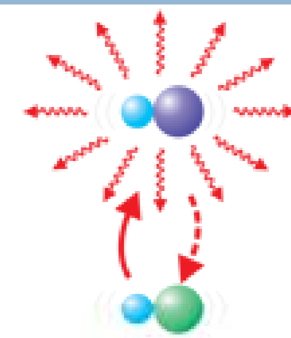
$^{40}Ca^+$: 1 metastable state
two lasers



Molecular ion: Strong dipole allowed transition
….but vibrational degree of freedom leads to n metastable states with no selection rules
…. and each vibrational wavefunction has an infinite ladder of rotational states

SrF, exception that proves the rule

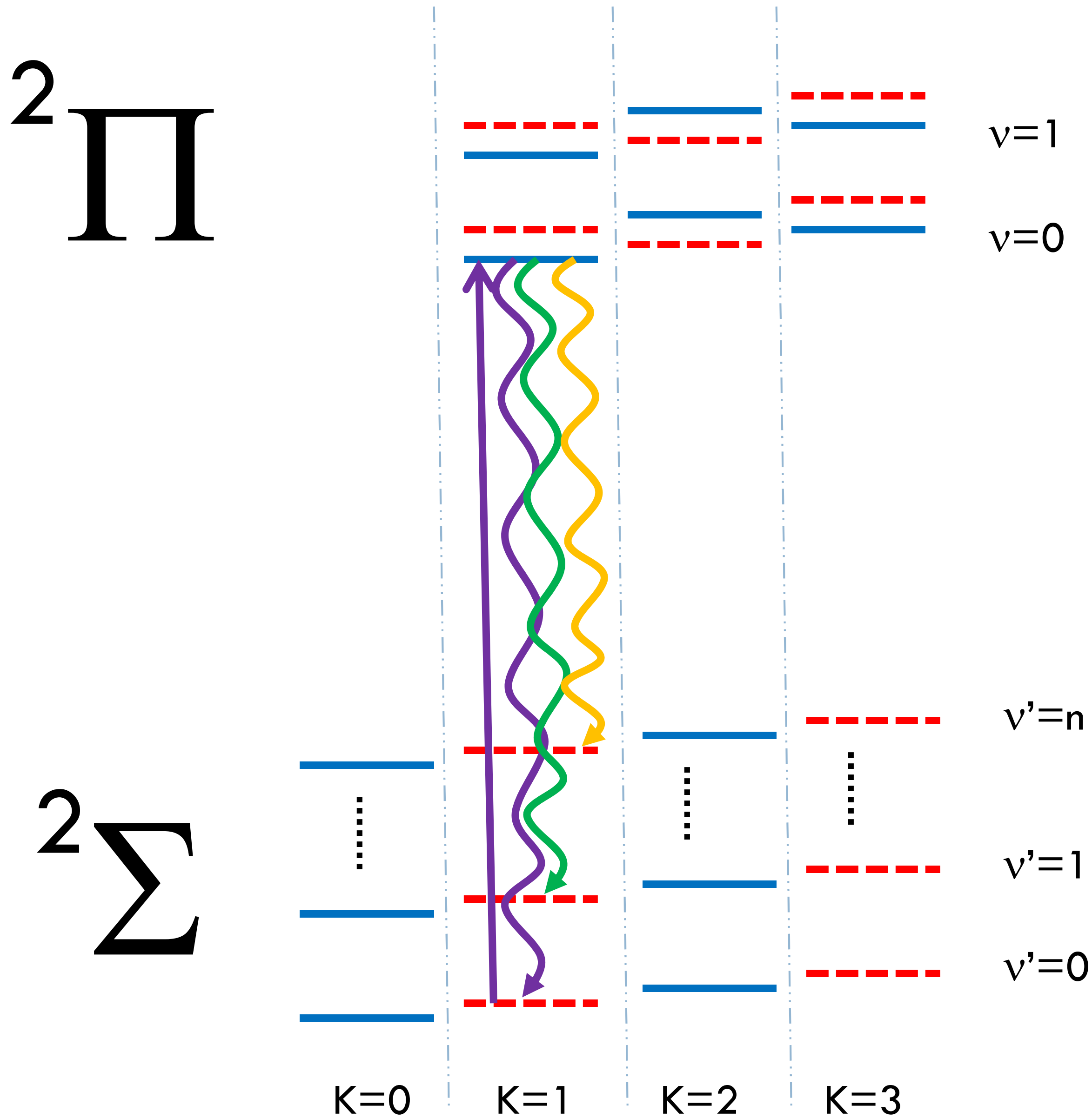




- Ground and excited states have very similar equilibrium bond lengths
- Franck-Condon overlap

E. S. Shuman, J. F. Barry, D. R. Glenn, and D. DeMille, Phys. Rev. Lett. **103**, 223001 (2009)
E.S. Shuman, J.F. Barry, and D. DeMille, Nature **467**, 820 (2010)

Ideal Level Structure for Direct Laser Cooling

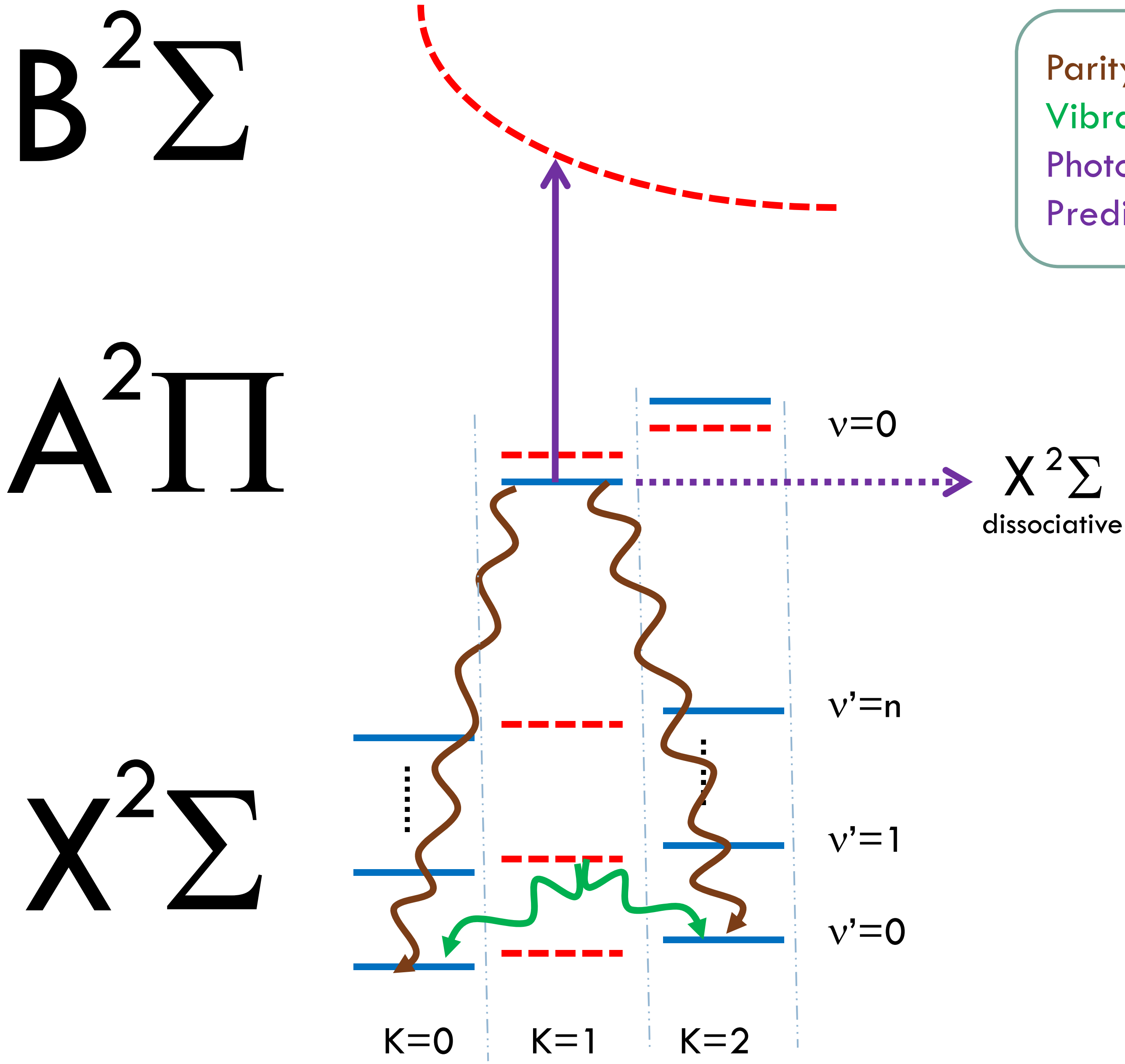


Franck-Condon overlap practically limits n.

A combination of parity and angular momentum selection rules to minimizes accessible rotational states.

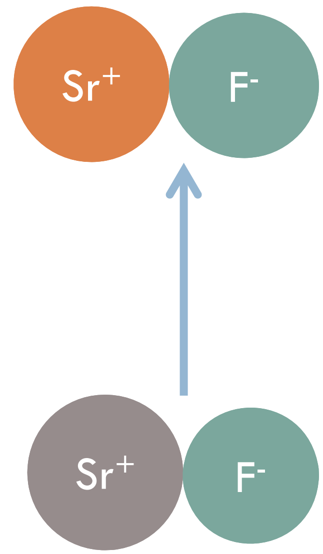
Complications

Parity violation
Vibrational Decay
Photodissociation
Predissociation



Picking a molecular ion

- Franck-Condon factors
- Use chemical intuition
  - SrF $\rightarrow AlF^+$ (no good)
  - Group III Hydrides
  - Transitions well described by a single electron in non-bonding orbital
- Two-level ions: AlH^+ and BH^+
- Three-level ion: SiO^+



M. D. Di Rosa, Eur. Phys. J. D **31**, 395 (2004)
T. A. Isaev, S. Hoekstra, and R. Berger, Phys. Rev. A **82**, 052521 (2010)
Three-level molecular ion: J.H.V. Nguyen and B. Odom, arXiv:1012.3696

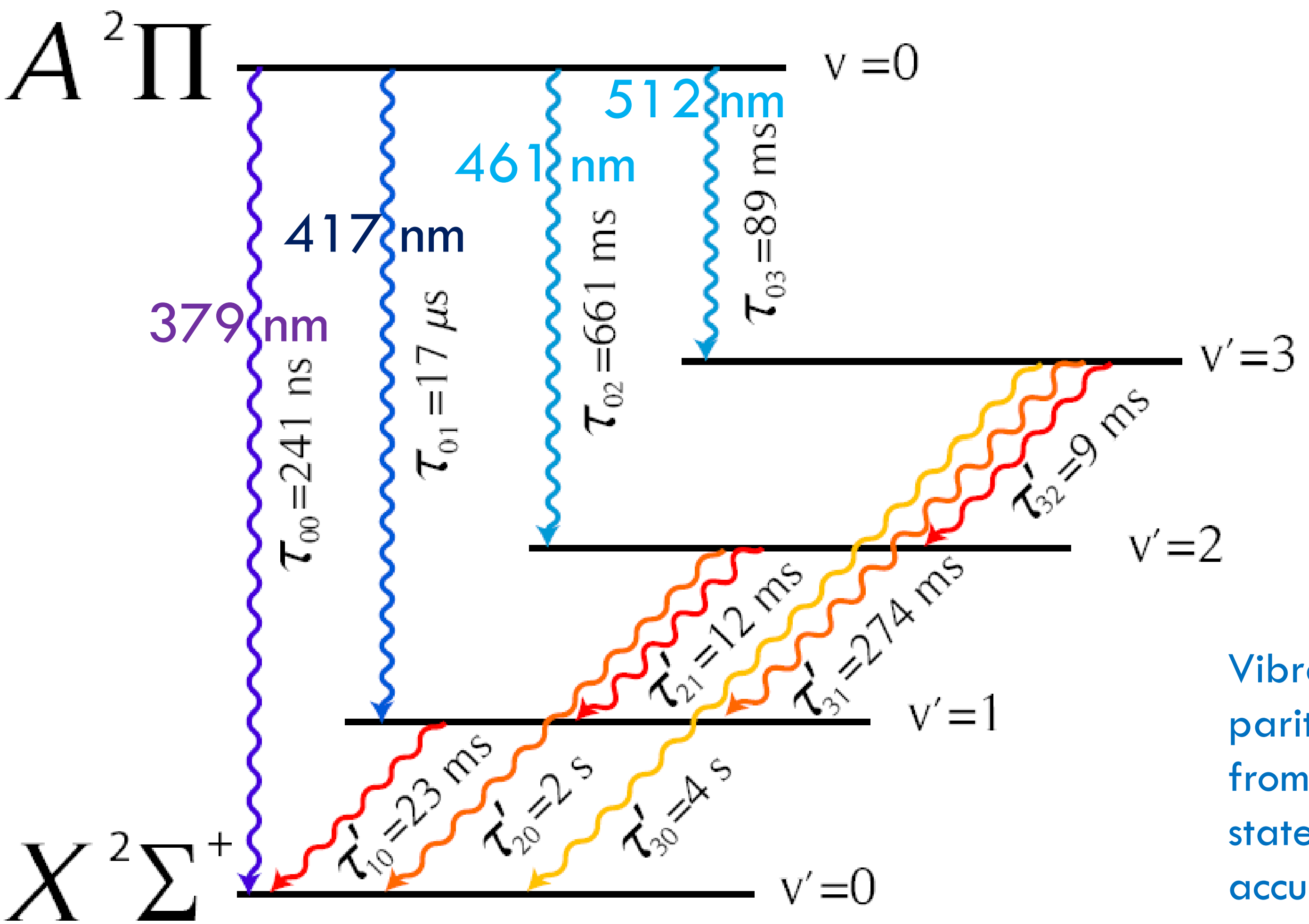
BH^+

	A $^2\Pi$	X $^2\Sigma^+$
***Re* [A]**	1.244	1.203
T_e [cm^{-1}]	26497.7	0
ω_e [cm^{-1}]	2152.8	2396.3
$\omega_e x_e$ [cm^{-1}]	35	56

v'/v	0	1	2	3	4
0	0.9577	0.0406	0.0017	0.0000	0.0000
1	0.0420	0.8913	0.0624	0.0041	0.0000
2	0.0002	0.0677	0.8562	0.0692	0.0065
3	0.0001	0.0002	0.0792	0.8475	0.0642
4	0.0000	0.0002	0.0000	0.0780	0.8607

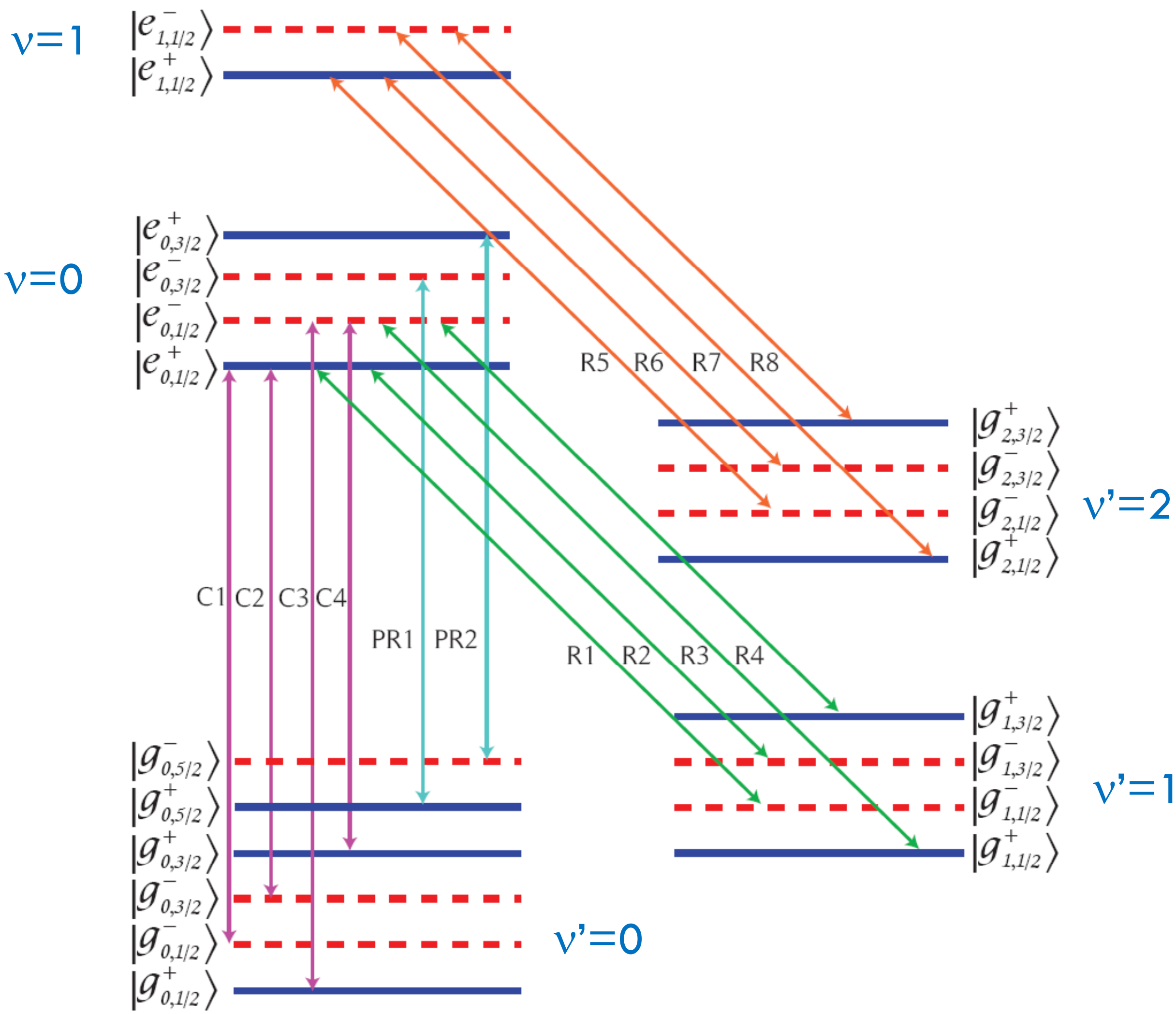
D. A. Ramsay and P. J. Sarre, J. Chem. Soc., Faraday Trans. 2, **78**, 1331 (1982)
R. Klein, P. Rosumus, and H.J. Werner, J. Chem. Phys., **77**, 3559 (1982)

BH^+ rates



Vibrational decay changes parity but clears population from higher vibrational states faster than it accumulates.

How many lasers?



Cooling lasers:

v=0←v'=0, ΔJ=0,-1

C1-C2 (one laser plus EOM)

C3, C4 (two lasers)

Repump lasers:

v=0←v'=1, ΔJ=0,-1

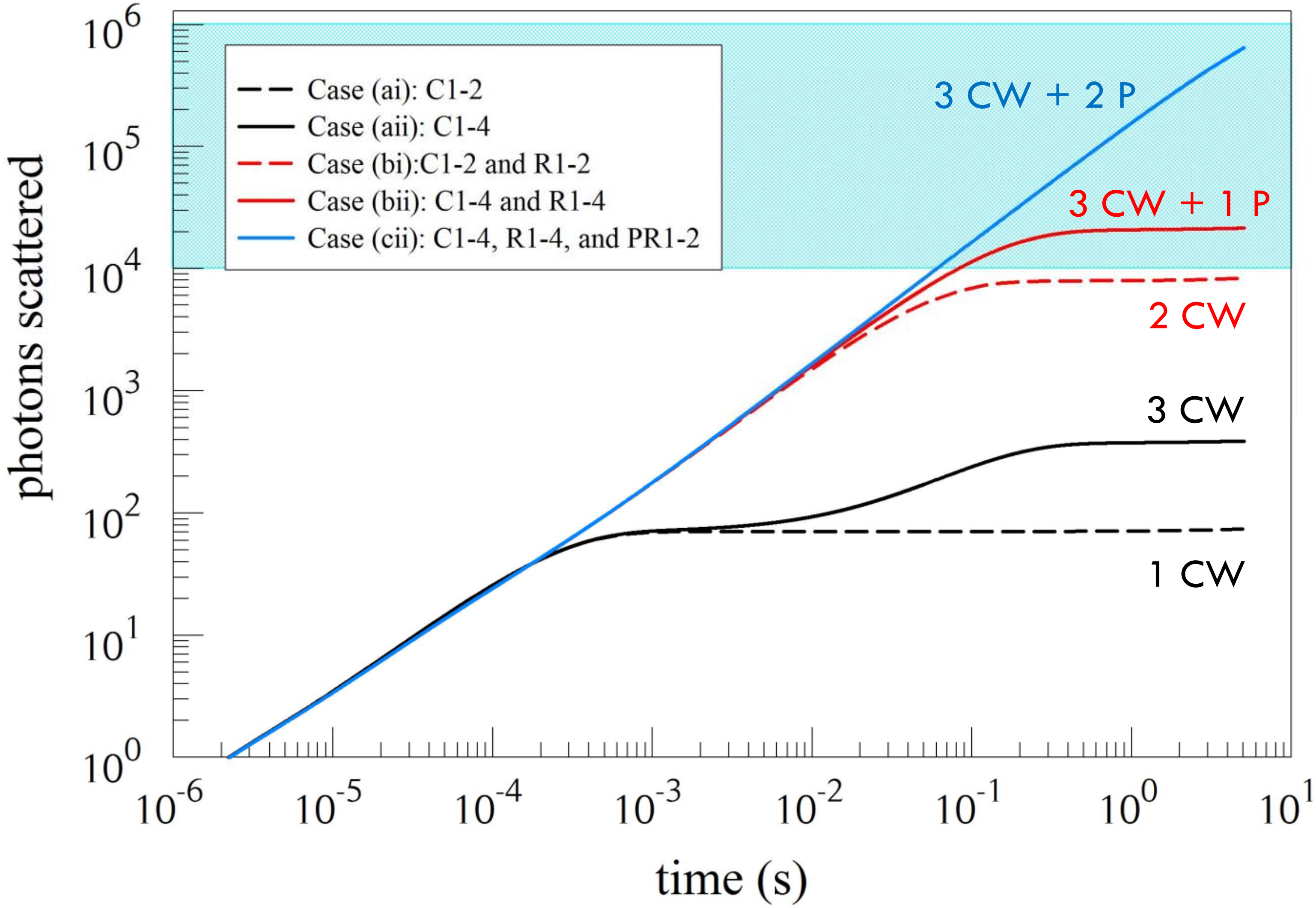
R1-R2 (one laser plus EOM)

R1-R4 (one pulsed laser)

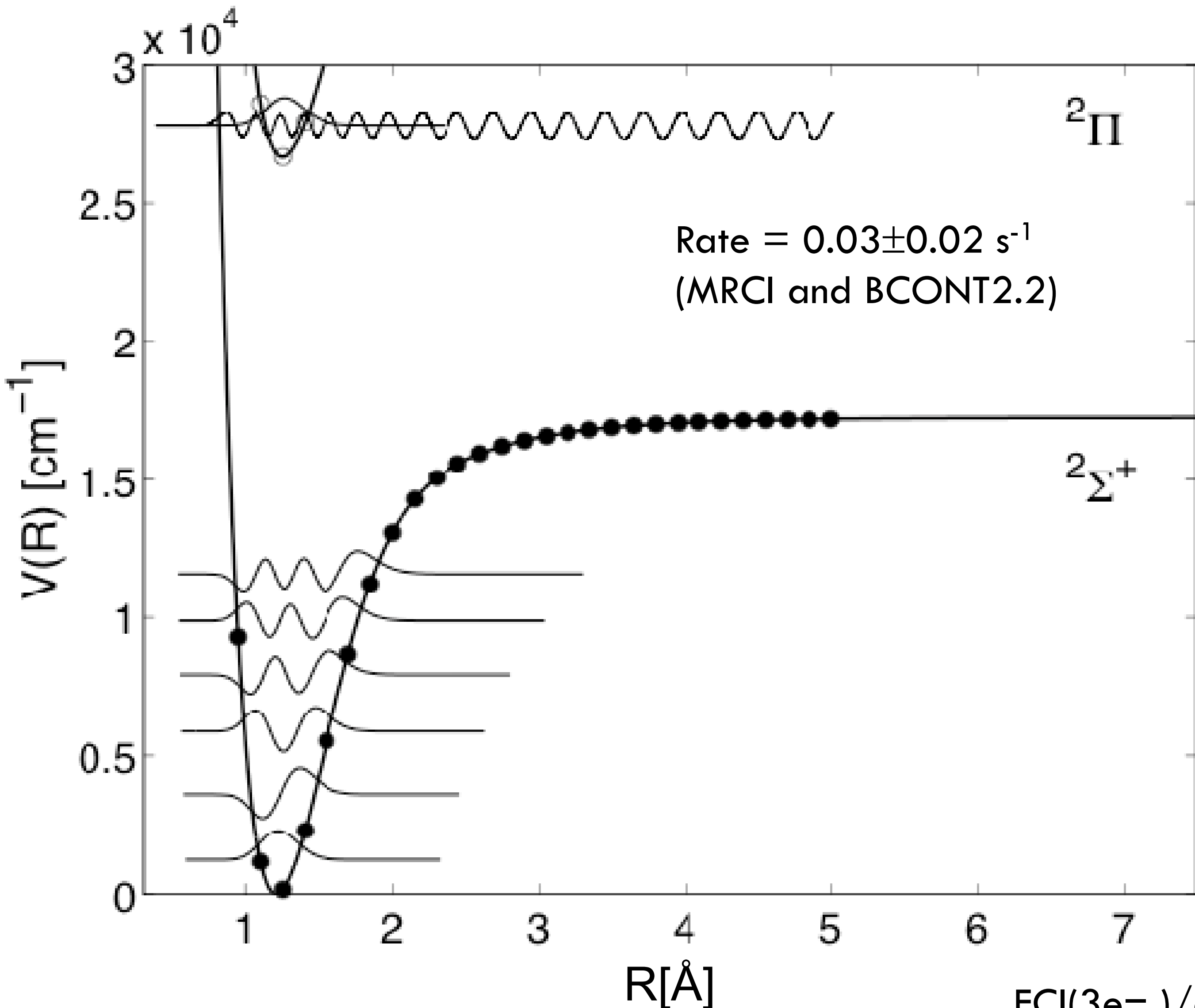
v=0←v'=0, ΔJ=-1

PR1-PR2 (one pulsed laser)

Rate calculations for BH^+

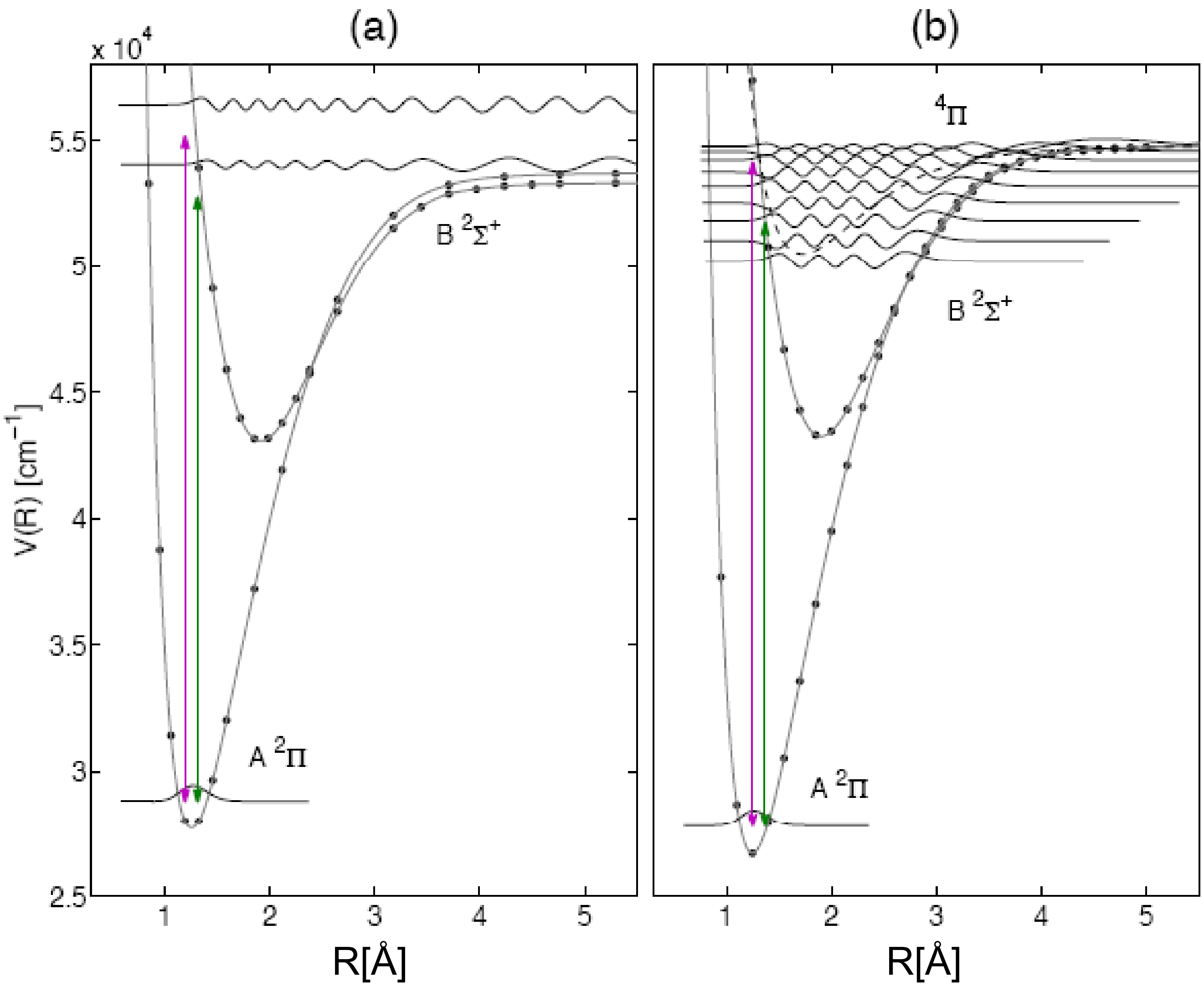


BH^+ : Rare pre-dissociation



FCI(3e−)/aug-cc-pV5Z

Two-photon processes



a) MC-SCF

Dissociative state.

Franck-Condon limits rate.

b) FCI/3e-

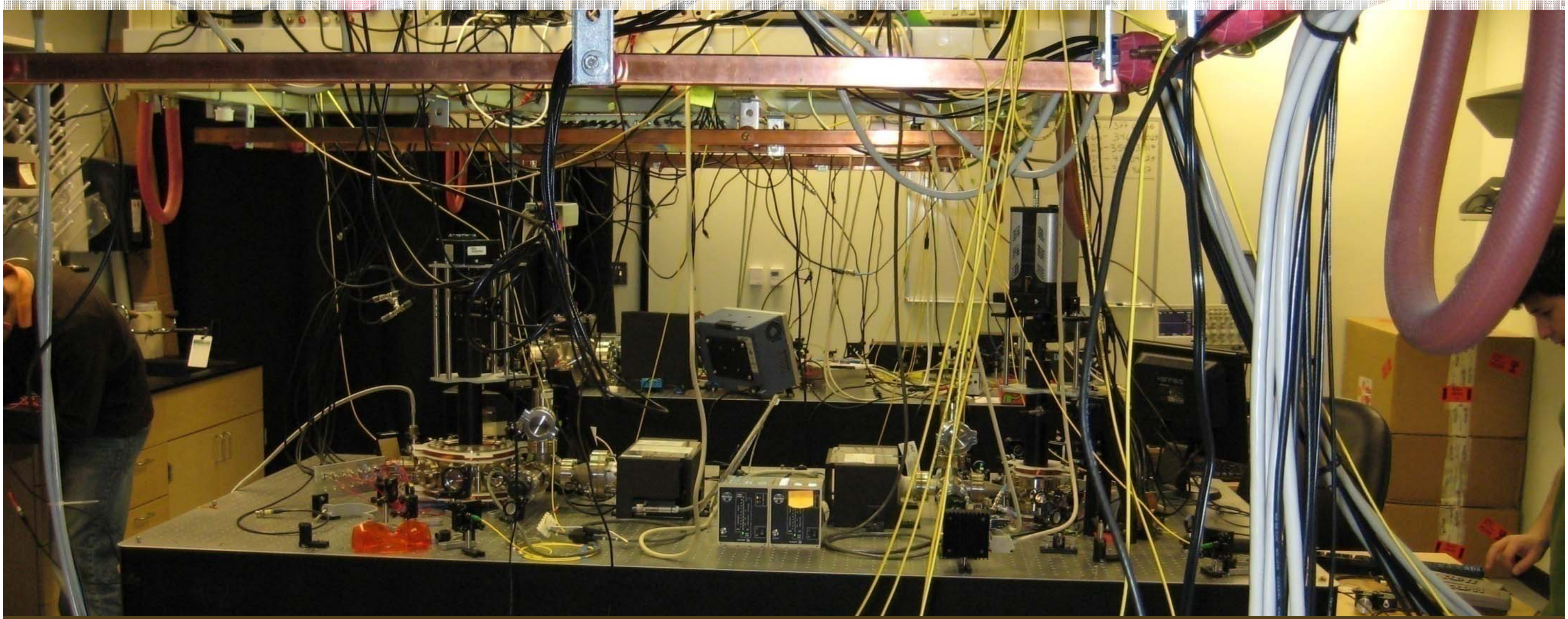
Bound state.

Only a problem if very unlucky.

Prospects for laser cooling

- Five lasers would allow for the direct laser-cooling of BH^+ from room temperature to mK temperatures.
- Two lasers for pre-cooled ions.
- Accurate measurement of the predissociation rate would serve as a benchmark for quantum chemical dynamics simulation.
- J.H.V. Nguyen, C.R. Viteri, E.G. Hohenstein, C.D. Sherrill, K.R. Brown, B. Odom arXiv:1102.3368

Proposal to Experiment



- Generate and load BH^+
- Use atomic ions to sympathetically cool BH^+ and acquire high precision BH^+ spectra
- Laser cool

Boron Chemistry

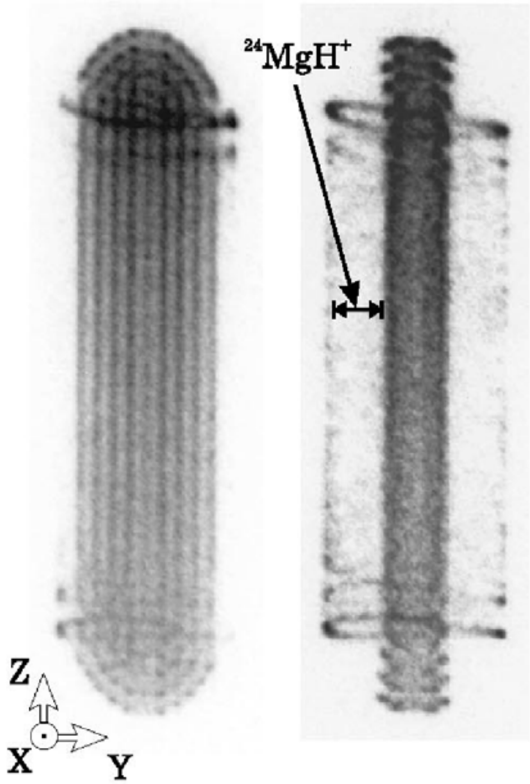
- Synthesize diborane
  - Highly reactive
- Photolyze diborane
  - Excimer laser
- Ionize BH

Isolated core excitation of BH
C.R. Viteri, A.T. Gilkison, S.J. Rixon, and E.R. Grant
Phys. Rev. A **75**, 013410 (2007)

Sympathetic Cooling of Molecular Ions

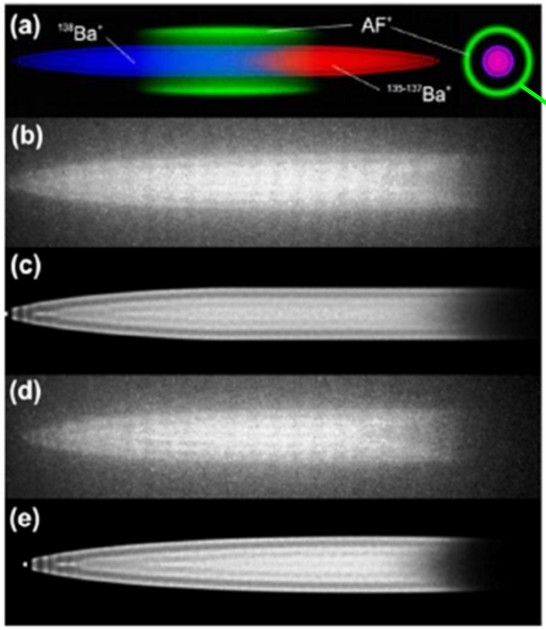
Laser-cooled MS: T. Baba and I. Waki, *Jpn. J. Appl. Phys.*, 35, L1134025 (1996)

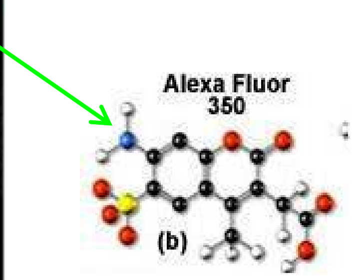
Laser cooled Mg^+ cools MgH^+



K. Mølhave and M. Drewsen,
Phys. Rev. A. **62**, 011401 (2000)

Laser cooled Ba^+ with $AlexaFluor^+$





(115 mK)

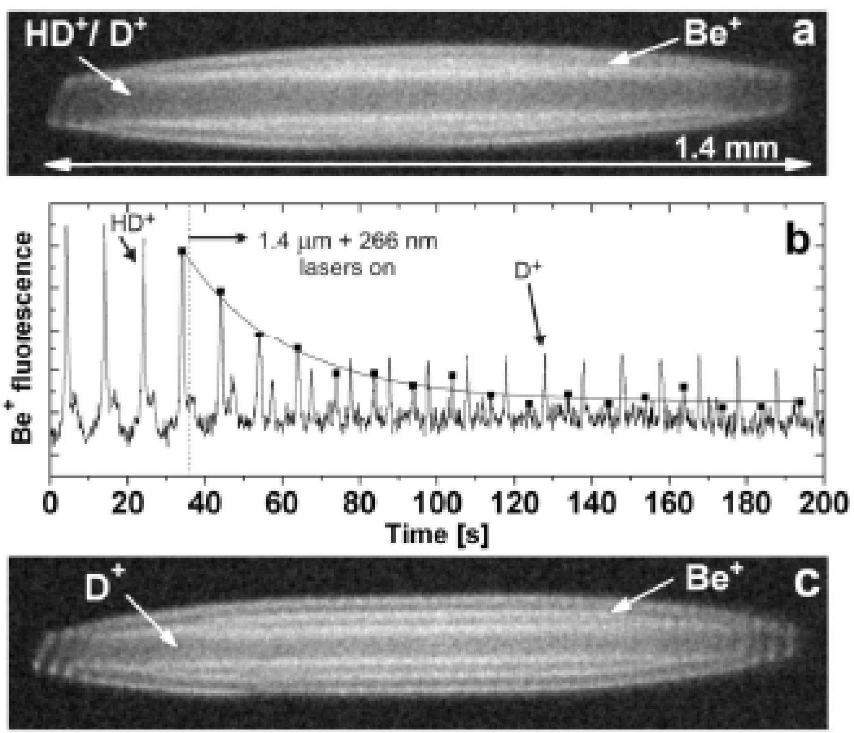
A. Ostendorff et al.
Phys. Rev. Lett. **97**, 243005 (2006)

Fullerene:
V. L. Ryjkov, X.-Z. Zhao, and H. A. Schuessler,
Phys. Rev. A **74**, 023401 2006

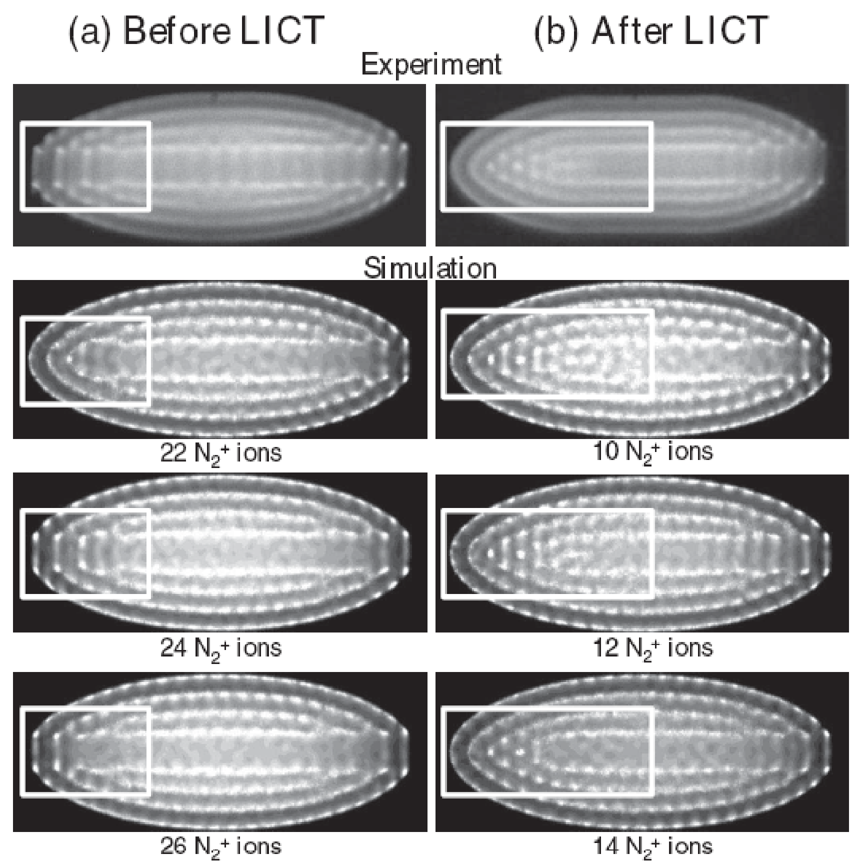
Action Spectra

$N_2^+ + Ar + h\nu \rightarrow N_2 + Ar^+$

REMPD

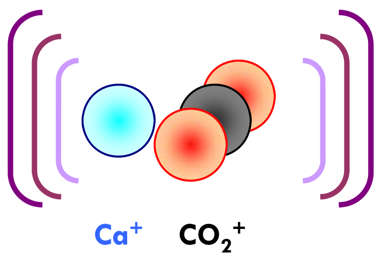


S. Schiller and co-workers,
Phys. Rev. A **74**, 040501R (2006),
Phys. Rev. Lett. **98** 173002 (2007)

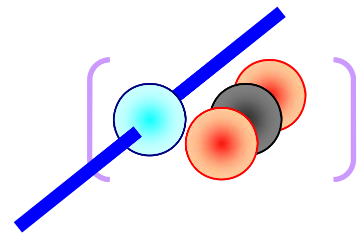


X. Tong, A.H. Winney, and S. Willitsch
Phys. Rev. Lett. **105** 143001 (2010)

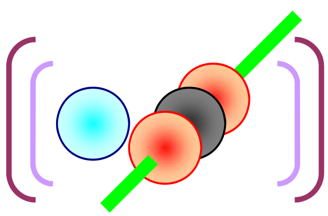
Molecular Ion Spectroscopy



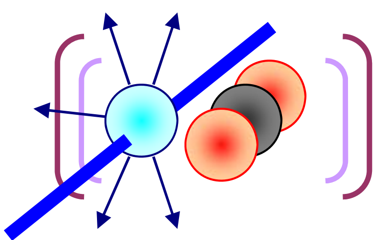
1. Trap atomic and molecular ions



2. Laser cool ion crystal

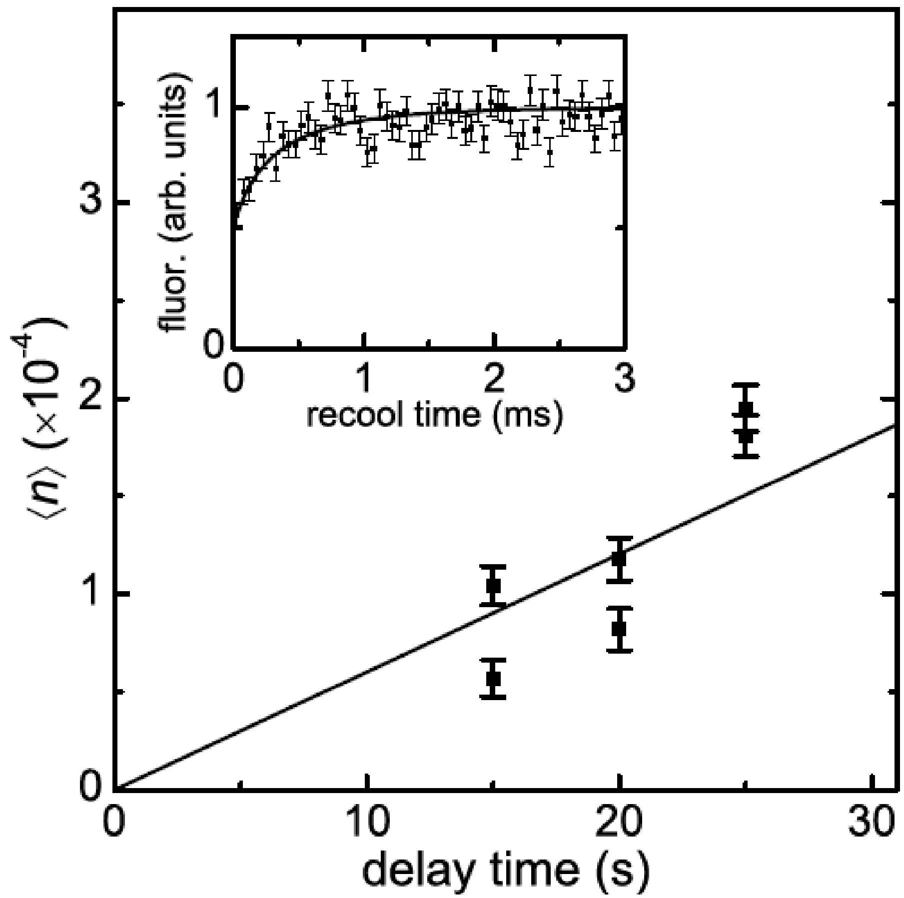


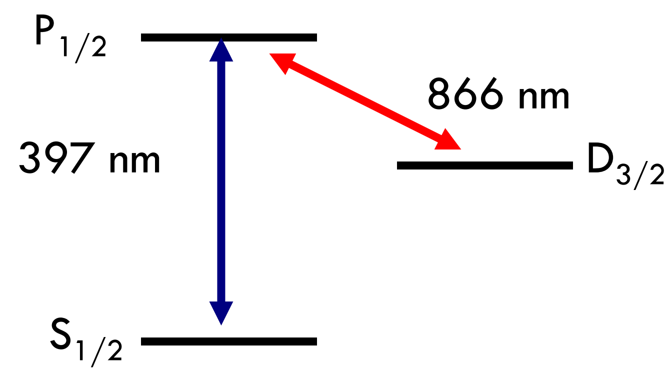
3. Heat ion crystal by exciting the molecular ion.



4. Measure temperature change by laser-induced atomic fluorescence

Doppler Recooling





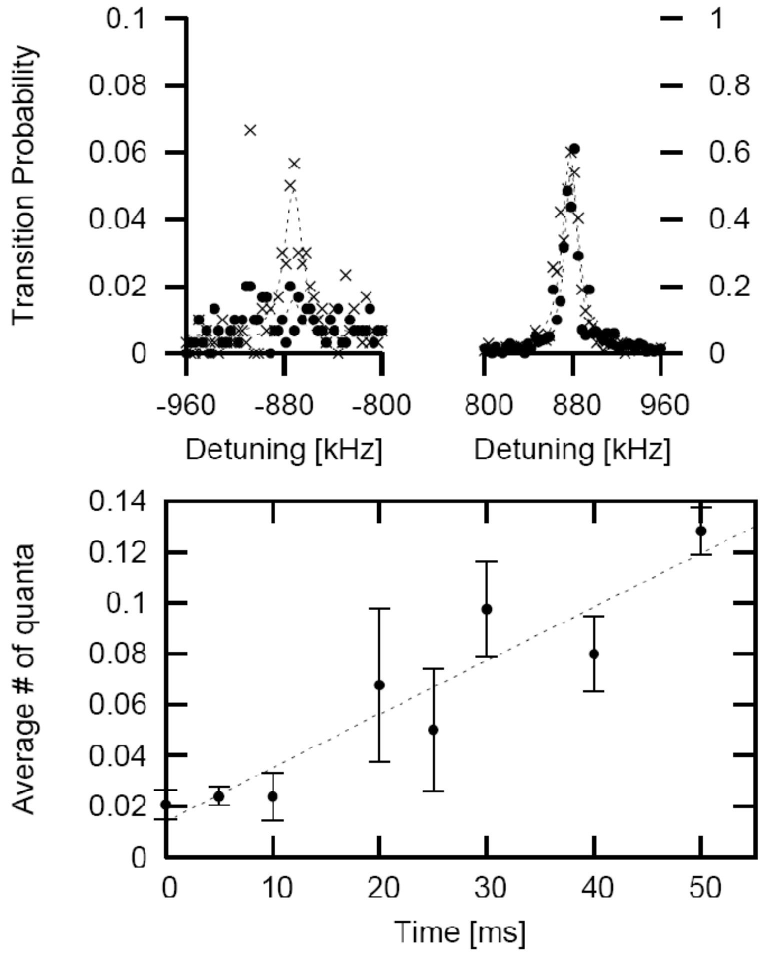
Simple experimental setup.

Difficult for low heating rates.

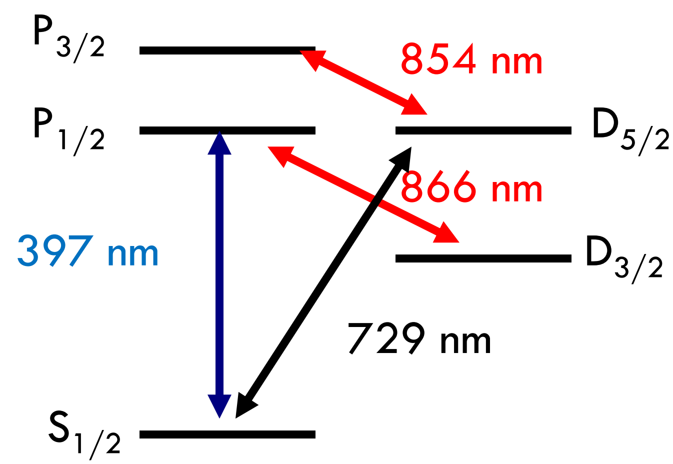
transition linewidth > trap frequency.

R.J. Epstein, et al., Phys. Rev. A **76** 033411 (2007)

Sideband Measurement



J. Labaziewicz, et al.
Phys. Rev. Lett. **100**, 013001 (2008)



Temperature measurement conceptually simpler.
RSB=k<n> BSB=k<n+1>

Transition linewidth < trap frequency .

QLS, QHS, and SHS

- **Quantum Logic Spectroscopy**
  - Detect single phonon excitation
  - Coherent excitation of spectroscopy ion
    - P. O. Schmidt, et al., *Science* **309**, 749 (2005)
- Quantum Heating Spectroscopy
  - Detect heating rate by phonon
  - Incoherent excitation of spectroscopy ion
- Sympathetic Heating Spectroscopy
  - Detect heating rate by Doppler recooling
  - Incoherent excitation of the spectroscopy ion
    - C.R. Clark, J.E. Goeders, Y. Dodia, C.R. Viteri, and KRB, PRA, 81, 043428 (2010)

Conclusions

- AlH^+ and BH^+ are candidates for demonstrating direct laser cooling of molecular ions
  - arXiv:1102.3368
- Mixtures of atomic ions and molecular ions are ideal for cold molecular ion spectroscopy and molecular ion reactions