Internal State Cooling of Molecular Ions

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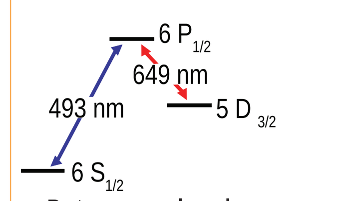
Introduction

Why molecules?

Precisely because of their rich internal structure, molecules are much more difficult to control than atoms. If on-demand control of molecular vibrations and rotations could be achieved, molecular complexity could be turned into an advantage, with a number of applications becoming possible for the first time.

Why barium?



Ba+ energy levels.

1. Heavy atomic species makes it easier to simultaneously trap molecules of comparable mass.
2. All cooling transitions in the optical range; Lasers commercially available.
3. ^{138}Ba has zero nuclear spin.

Paths to precision molecular spectroscopy

1a) Sympathetic cooling of molecules by trapped ions
or
1b) Direct translational cooling of molecules

2) Internal state prepartion of the molecules

3a) Direct spectroscopy of the crystal
or
3b) molecular Quantum Logic Spectroscopy

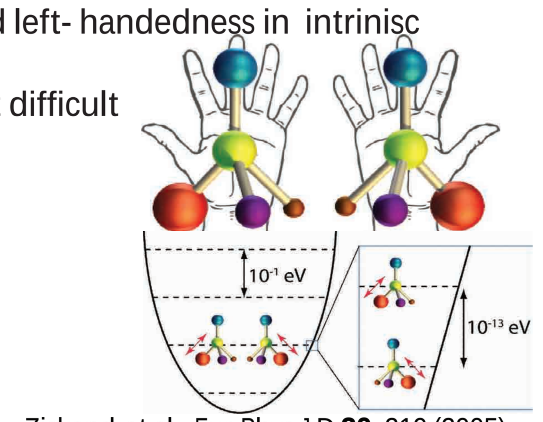
Motivation

Are Mirror-Image Molecules Identical?

Chiral molecules with right- and left-handed configurations are normally assumed to be mirror images of one another. However, the weak nuclear force induces a small parity-violating effect which should cause right- and left-handed molecules to vibrate at slightly different fundamental frequencies This would constitute a first demonstration of asymmetrybetween right- and left- handedness in intrinsic sturcture of stable matter. .

$\Delta\nu^{vib} \sim Z^5$, so heavy constituents are desirable, but difficult

e.g.
CHFClBr
lighter, enatiomeric enhancement achieved
expected $\Delta\nu^{vib}$ =1.7mHz on 10um = 6 x 10^{-17}
best spectroscopy to date (in cell) is 3 x 10^{-13}
BiHFBr
heavier, enatiomeric enhancement not achieved
expected $\Delta\nu^{vib}$ =20Hz = 7 x 10^{-14}



Ziskand, et al., Eur. Phys J D **20,** 219 (2005)
Laerdahl et al., Phys Rev A **60,** 4439 (1999)

Are Fundamental "Constants" Really Constant?

Time-variation of fundamental "constants" is generally expected in many extensions of the Standard Model. There is currently one claim of evidence for variation of the electron-proton mass ratio on cosmic time scales, making improved laboratory searches quite interesting.

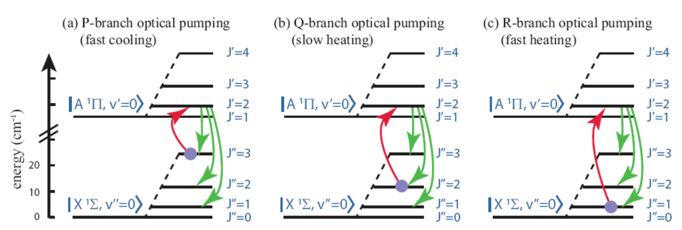
e.g.
Astrophyiscs
Measurement of quasar absorption lines $\quad \frac{\dot{\mu}}{\mu} \leq 10^{-15}/yr$

Laboratory
SF_6 $\quad \frac{\dot{\mu}}{\mu} < 65 \times 10^{-15}/yr$

Flambaum and Berengut, Int J Mod Phys A **24,** 3342 (2009)

Internal State Cooling P-Branch Optical Pumping (PBOP)

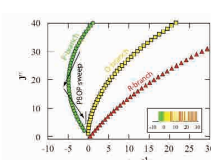
A challenge with molecules remains to achieve good state initialization, specifically to access the ro-vibrational ground state.
Optical pumping of P-branch transitions (*J* ⟶ *J - 1*) results in rotational cooling. [1]

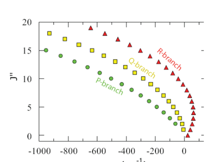


Pictorial representation of different types of optical pumping. In optical pumping, dipole allowed absorption (red arrow) from an initial state (purple dot) is followed by dipole-allowed spontaneous emission (green arrows).

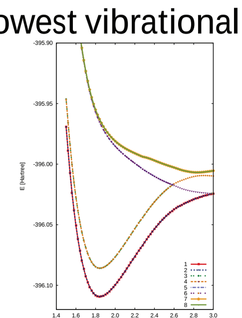
Requirements for PBOP

1. Appropriate A-X band structure.
The P-branch cannot overlap significantly with the Q- and R- branches.


Example of a good molecule choice.


Example of a bad molecule choice.

2. Diagonal Franck-Condon factors.
The lowest vibrational level of the A state must decay preferentially to the lowest vibrational level of the X-state.



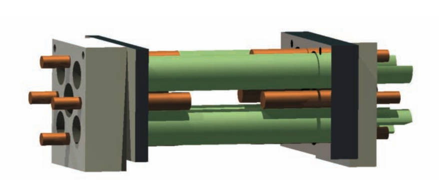
Initial investigations suggest that molecules which exhibit spin orbit splitting in the ground state (e.g. IF^+) may be good candidates.

[1]Bahns *et al*, J. Chem Phys. **104** 9689 (1996)

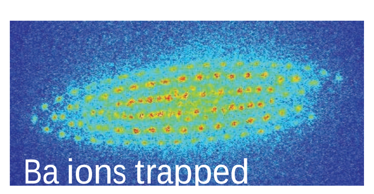
Precision Spectroscopy on a Large Crystal

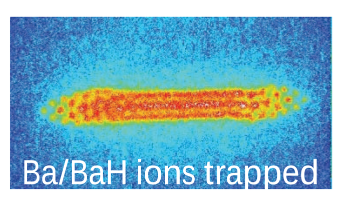
Sympathetic cooling will be used initially for translational cooling of the molecular ions. Barium ions are doppler cooled and co-trapped molecular ions lose kinetic energy by their coulomb interaction.

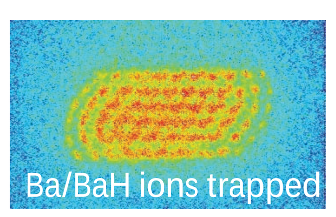
rf Paul Trap parameters
r_0= 4.6 mm
z_0= 9 mm
$\Omega = 2\pi\times3$ MHz



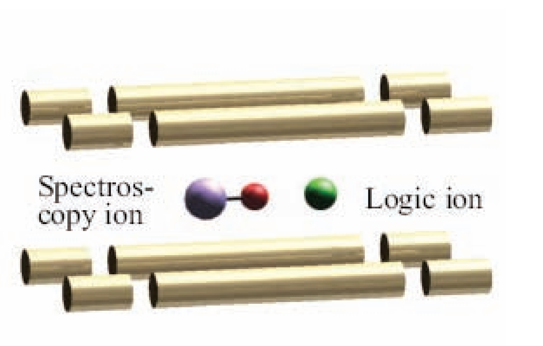
1. Ions confined axial by end caps (brown).
2. Ions confined radial by rf fields applied to two rf rods (green).


Ba ions trapped


Ba/BaH ions trapped


Ba/BaH ions trapped

Molecular Quantum Logic Spectroscopy (mQLS)



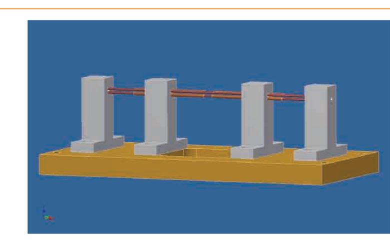
Requirements for Spectroscopy
1. A good transition: narrow & immune to environment;
2. Cooling to minimize doppler broadening;
3. Reliable initial state preparation;
4. Efficient state detection.

Quantum Logic Spectroscopy [2] is a technique for doing spectroscopy on a system that does not possess the above properties by transiently entangling it with a system that does via their shared motional modes in an ion trap.

[2] Schmidt, et al., Science **309,** 749 (2005)

A requirement for QLS is large motional mode sideband spacing which is obtained by large secular frequencies. This is easier to achieve with a smalller trap volume. Our new trap is under devlopment.

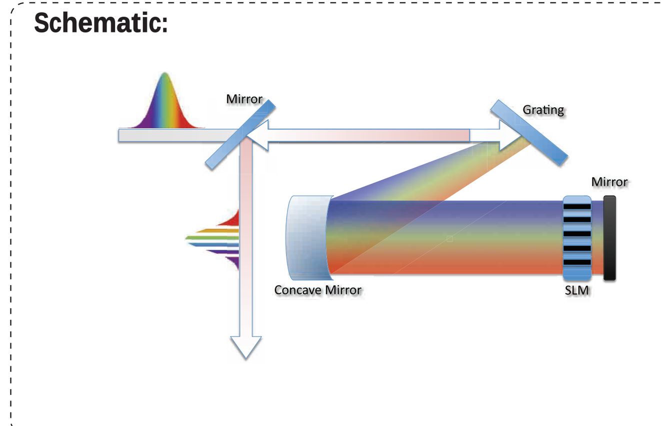
mQLS Trap Parameters
r_0= 0.46 mm
z_0= 1.5 mm
$\Omega = 2\pi\times24$ MHz


Schemtatic of the mQLS trap.

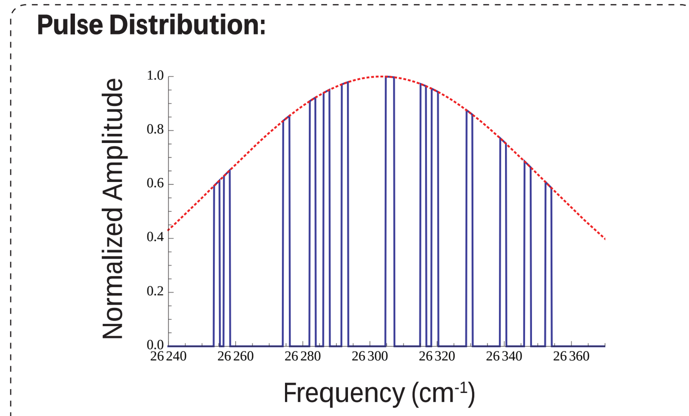
Pulse-Shaped Optical Pumping

Manipulation of the internal states of molecular ions is an important tool for spectroscopy, state-readout schemes, detection schemes as well as direct cooling of molecules. A pulse-shaped optical pumping scheme similar to Viteau *et al.* [1] can be employed for a hydride (such as BH^+) with sufficient resolution to address different rotational states.

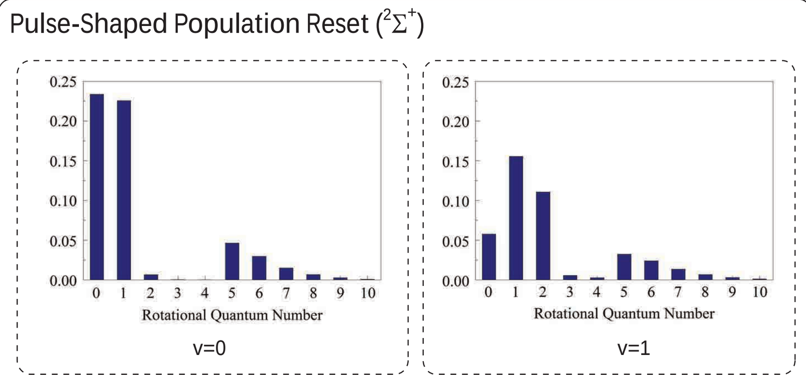
Schematic:



Pulse Distribution:



Pulse-Shaped Population Reset ($^2\Sigma^+$)



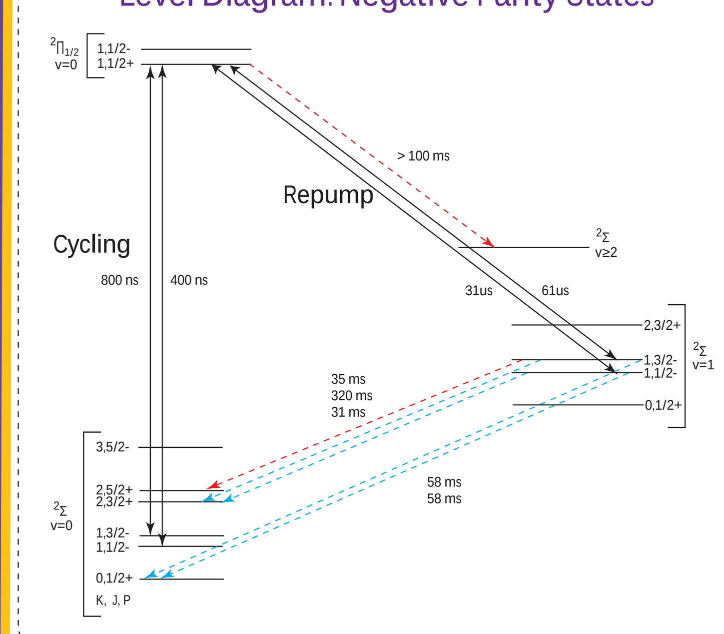
- application of a series of shaped-femtosecond pulses for 20 μs, resulting in depopulation of the higher rotational states
- residual population remains in higher rotational states
- population of v=1 level also occurs (spontaneous emission from excited state)
- additional pulse-shaped can depopulate v=1
- can be used as a population reset for direct cooling of BH^+

[1] M. Viteau *et al.*, Science, **321**, 232 (2008)

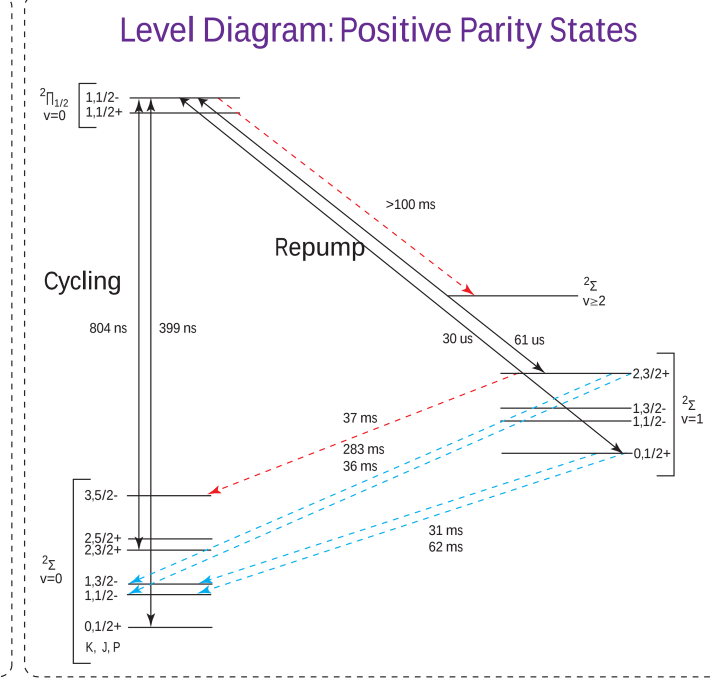
Translational Cooling

Direct translational cooling of molecules is difficult due to the rich internal structure of molecules. Vibrational leak due to non-ideal FCF's results in population of dark states that prevent forming a continuous cycling transition. BH+ is an example of a possible candidate molecule (due to highly diagonal FCF's), although it still suffers from leak into the other vibrational states. Additionally, predissociation occurs on a timescale commensurate with the time required for cooling.

Level Diagram: Negative Parity States

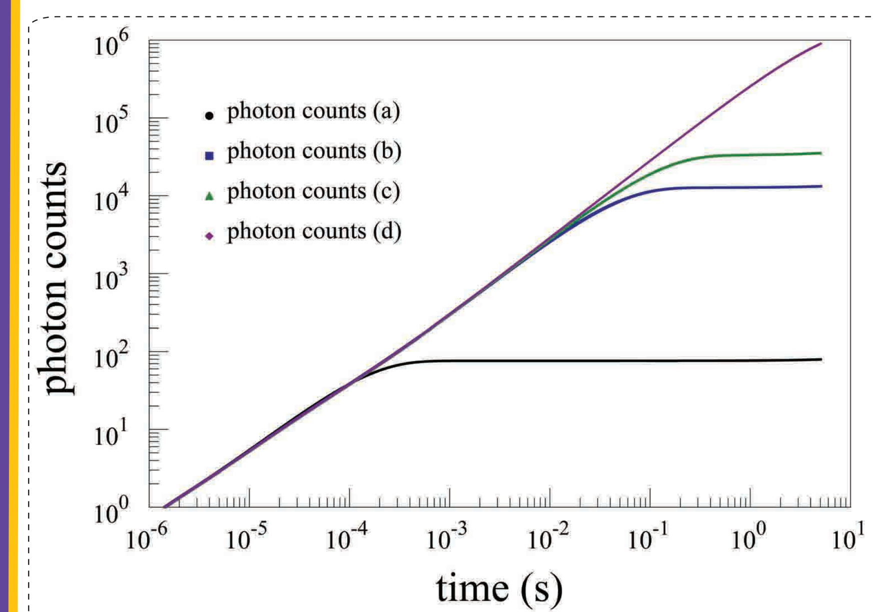


Level Diagram: Positive Parity States



Vibrational leaks populate v≥1 levels, resulting in ground-state parity flips as well as population of higher angular momentum states (due to vibrational leak into the ground state). By adding repump lasers, we increase the time spent in the cycling transition.

Using a rate-equation approach to simulate the population dynamics we obtained the probability of occupying the cooling/repump populations and determined the average total number of photons scattered as the number of lasers is increased:



Case (a): Two Frequencies
Cycling on negative parity transitions only. Population quickly pumps into v=1 level.

Case (b): Four Frequencies
Cycling on negative parity transitions with v=1 repump. Population pumps into other parity and J=5/2 level.

Case (c): Eight Frequencies
Cycling on both parity transitions with v=1 repumps. Population pumps into J≥5/2 levels.

Case (d): Ten Frequencies
Cycling on both parity transitions with v=1 repump and J=5/2 (both parities) repumps. Population pumps into higher J>5/2 levels.

Obtaining continuous cycling is difficult due to leaks in ro-vibrational dark states. By adding appropriate repumps we increase cycling time. For BH+ we calculate that the dominant leak, after v=1 repump, is into higher rotational levels of the ground state. This is because the leak into higher vibrational levels is counteracted by leaks out of those levels back into the vibrational ground state.

A broadband source, such as the scheme discussed in pulse-shaped optical pumping would be suitable for repumping out of the higher-lying dark states since the rotational spacing is small compared to the bandwidth of a shaped-femtosecond laser.