Towards ACME III: Optical Cycling Tests in Beam Box II

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Daniel Ang

Towards ACME III

Main uncertainty equation:

$$\delta d_e = \frac{\hbar}{2C\tau\mathcal{E}_{eff}\sqrt{\dot{n}T}} \tag{1}$$

- Maxed out contrast C: ~ 0.95
- Maxed out precession time τ: $\sim 1\ \mathrm{ms}$ while H-state lifetime is 1.3 ms
- Maxed out E_{eff}: fully polarized at applied $\mathcal{E} \approx 10\ \mathrm{V/cm}$
- Two major ways left to improve ACME:
  - Reduce sources of systematic error
  - Average more (increase T)
  - **Increase molecule flux $\dot{n}$**

Towards ACME III

Main uncertainty equation:

$$\delta d_e = \frac{\hbar}{2C\tau\mathcal{E}_{eff}\sqrt{\dot{n}T}} \tag{1}$$

- Two ways to improve N:
  - Increase initial molecular flux (improve beam source production of useful molecules in X)
  - **Increase efficiency in using molecules produced by beam source**

Molecule Usage Efficiency in ACME II

- Ablation beam source:
  - $\sim 10^{11}$ molecules per pulse, 90 % in $|X, J = 0, 1, 2, 3\rangle$
  - $\sim 0.1\%$ reach detection region 1.1 m away (due to beam divergence)
- Rotational cooling $(X - C)$: $\sim 50\%$ of initial molecules in X ultimately end up in $J = 0$
- STIRAP transfers $\sim 75\%$ of $|X, J = 0\rangle$ to H
- Spin precession occurs in interaction region
- H-state molecules excited to I by 703 nm and $\sim 90\%$ decay to X
- **Only 5% of photons emitted by molecules are detected!**
- Final figure: $\sim 10^6$ photons per pulse (actual: $\sim 300,000$)

Detection Efficiency in ACME II

- Current measurement scheme: detect 512 nm photons emitted by spin-precessed molecules decaying from $I \rightarrow X$
- ACME II detection inefficiencies:
  - Geometric collection efficiency: $\sim 20\%$
  - Quantum efficiency of PDs: $\sim 25\%$
  - Total: $\sim 5\%$ of decaying molecules detected
- Goal: **become molecule shot noise limited by detecting one photon per molecule in experimental state**

How to Maximize Detection Efficiency

Three elements to improve:

- Improve solid angle of collection optics - detection outside of interaction region with optimized reflector
- Improve quantum efficiency of detectors - upgrading to cooled SiPMs ($\approx$ 50% QE at 512 nm)
- **Increase number of photons per molecule - optical cycling**

How to Maximize Detection Efficiency

Three elements to improve:

- Improve solid angle of collection optics - detection outside of interaction region with optimized reflector
- Improve quantum efficiency of detectors - upgrading to cooled SiPMs ($\approx$ 50% QE at 512 nm)
- **Increase number of photons per molecule - optical cycling**
  - At end of precession, project molecules into orthogonal long-lived states (**shelving**)
  - Outside of interaction region, stimulate with light tuned to transition with **high branching ratio**
  - Switch rapidly in cycling between two orthogonal states (parity-selective switching)

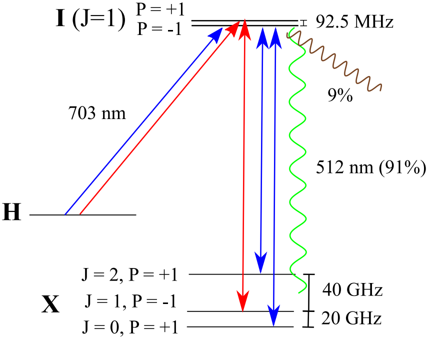
Determining the Cycling Scheme

Questions to answer:

1. **What is the branching ratio of the transition? (**$X-I$**,** $X-C$**, other?)**
2. **How many lasers will we need? (Repumps?)**
3. Will we have enough laser power? (Transition dipole moment)
4. How will the angular distributions of the decay affect the gain? (Will be limited by worst-detected decay.)

Optical Cycling Schemes

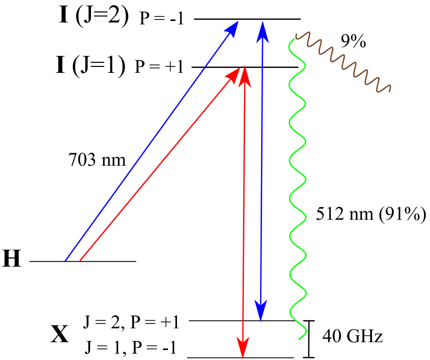
Assuming no repumps. Scheme 1:



Needs one 703 nm, three 512 nm lasers for basic cycling.

Optical Cycling Schemes

Assuming no repumps. Scheme 2:



Needs two 703 nm, two 512 nm lasers for basic cycling.

Determining Branching Ratio

- Branching ratio of $X - C$: $\sim 75\%$.
- Branching ratio of $X - I$: measured (roughly) by Steimle (2014) to be $\sim 91\%$

TABLE III. The transition wave numbers (cm^{-1}), energies (cm^{-1}), branching ratios (%), and transition dipoles (Debye, D) of the dispersed fluorescence spectra resulting from exciting near the Q-branch band head (19 538 cm^{-1}) of the $I(|\Omega| = 1) - X^1\Sigma^+(0,0)$ band.

	Laser excited state: $I(\|\Omega\| = 1)(v = 0)$					
Band[a]	Assignment	Trans.[b]	Energy[c]	Diff.[d]	%[e]	Td[f]
A	$I(\|\Omega\| = 1)(v = 0) \rightarrow X^1\Sigma^+(v = 0)$	19 538	0	0	91	1.84
B	$I(\|\Omega\| = 1)(v = 0) \rightarrow X^1\Sigma^+(v = 1)$	18 648	890	890	1	0.21
C	$E(\|\Omega\| = 0^+)(v = 0) \rightarrow X^1\Sigma^+(v = 0)$	16 313	0	0	<1	
D	$I(\|\Omega\| = 1)(v = 0) \rightarrow H\,^3\Delta_1(v = 0)$	14 224	5314	5314	5	0.69
E	$I(\|\Omega\| = 1)(v = 0) \rightarrow Q\,^3\Delta_2(v = 0)$	13 420	6118	6118	3	0.59

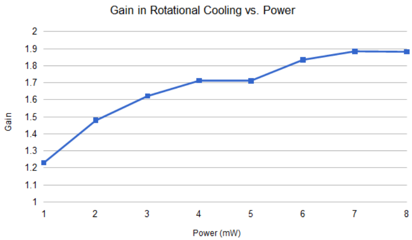
Note: assumes no branches $I \rightarrow A\,^3\Pi_{0^+}$ (1119 nm), $I \rightarrow {}^3\Pi_{0^-}$ (unseen state), $I \rightarrow B^3\Pi_1$ (1163 nm).

Determining Branching Ratio

- Considerations for choosing right transition:
  - $X - I$ (512 nm) has 2x transition dipole moment cf. $X - C$ (690 nm)
  - TAs, DL Pro available for 690 nm (power up to hundreds of mW)
  - No TAs for 512 nm - DL Pro HP available up to $\sim$ 20 mW, need to double above that
  - If we need 1-2 repumps anyway, then $X - C$ might be easier
  - Depending on divergence of beam, might need more 1-2x more power. **Need to consider possible magnetic lens to compress beam.**

Question 1: Experimental Evidence so Far

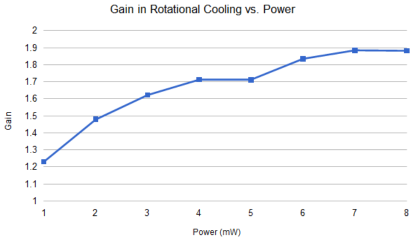
Rotational cooling in ACME II with $X - I$ (May 2017):



- Normal gain using $X - C$: ~ 1.8 at 16 mW
- Expect $X - I$ saturation at 5-10 mW, but windows have only 60% transmission at 512 nm

Question 1: Experimental Evidence so Far

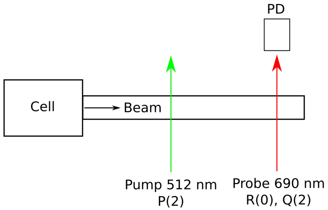
Rotational cooling in ACME II with $X - I$ (May 2017):



Conclusion: $X - I$ **branching ratio at least equal to** $X - C$**, but need more specialized measurement.**

Question 1: Planned Tests in Beam Box II

Test 1: Pump-probe measurement of branching ratio



Three configurations for probe laser absorption (690 nm):

1. R(0) without pump
2. R(0) with pump
3. Q(2) without pump

Question 1: Planned Tests in Beam Box II

- Ratio of absorption cross sections (configs. 1 & 2):

$$\frac{\sigma_{R(0)}^{pump}}{\sigma_{R(0)}^{0}} = \frac{P_0^0 + \frac{3}{5}\frac{(2/3)\mathcal{A}}{1-(1/3)\mathcal{A}}\ P_2^0}{P_0^0} = 1 + \frac{3}{5}\frac{(2/3)\mathcal{A}}{1-(1/3)\mathcal{A}}\frac{P_2^0}{P_0^0} \qquad (2)$$

  Taken from rotational cooling write-ups (Zack, Brendon).

  $\mathcal{A}$ = branching ratio.

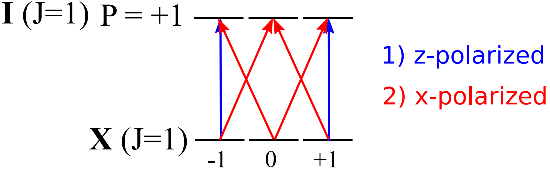
- Ratio of absorption on Q(2) and R(0):

$$\frac{P_2^0}{P_0^0} = \frac{1}{6}\frac{A_{Q(2)}}{A_{R(0)}} \qquad (3)$$

$$\Longrightarrow \mathcal{A} = 3 - \frac{3A_{Q(2)}^{0}}{A_{Q(2)}^{0} - 5A_{R(0)}^{0} + 5A_{R(0)}^{pump}}, \qquad (4)$$

Question 1: Planned Tests in Beam Box II

Test 2: Simple cycling



- Address orthogonal magnetic sublevels by switching between three configurations: 1) x-polarized, 2) z-polarized, 3) rapidly switching both
- Compare gain in signal 3) compared to sum 1) + 2)
- E. Kirilov found a gain of ~ 2 with $X - C$
- Auxiliary question: can we rotationally cool outside of the cell?

Question 2: Do we need repumps?

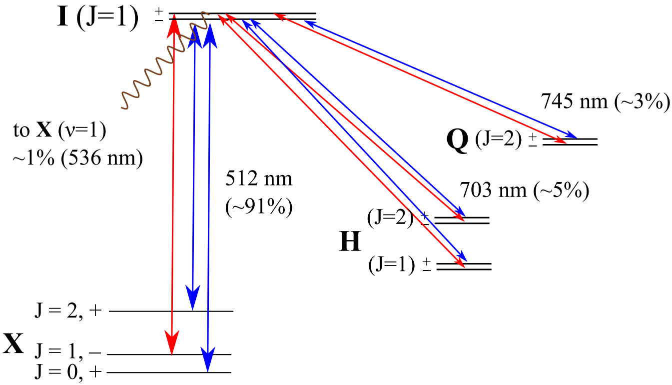
- Current detection effiency of 5 % - maximum gain of 20.
- However, from Zack's paper on cycling phase noise:
  - Overall noise comes from combination of phase projection, photon cycling noise, detection noise
  - Results in gain of $20/F$, defined by

$$F = 2 + \frac{q}{p} - 2q \tag{5}$$

    where $1/q \propto \#$ of photons cycled, $p =$ detection probability. $F \to 2$ as $q \to 0 \implies$ max gain of ~ 10.
  - **Caveat**: photons also emitted by Poisson process (if not in a dark state!)
  - With more photons cycled, can still increase gain to ~ 15 at ~ 100 photons cycled.
  - Conclusion for now: might need anywhere from 10-100 photons, better detection (larger p) helps

Question 2: Repumping options

$X - I$ as main transition:



Question 2: Repumping options

Number of lasers needed for each repump transition (assuming scheme 1):

- Main ($\sim 91\% = 11$ photons): 3 x 512 nm
- $+ H - I$ ($\sim 96\% = 25$ photons): add 2 x 703 nm (total: 5 lasers)
- $+ H - Q$ ($\sim 99\% = 100$ photons): add 2 x 745 nm (total: 7 lasers)

Note: small uncertainties can have drastic effect on # of photons!
Also possible is repumping through C-state (1090 nm, 1196 nm):

- More accessible wavelengths (TAs available)
- Less effective: $+H - C$ gives $\sim 94\%$, $+Q - C$ gives $\sim 96\%$

Summary: Path forward towards Optical Cycling

- Measure $X - I$ branching ratio using pump-probe absorption/fluorescence measurement
- Perform simple cycling in BB II using current beam line
- Construct test collection optics setup:
  - Optimize collection efficiency
  - Reduce potential scatter
  - Integrate with SiPM (?)
- Experiment with repumps: $H - C$ (1090 nm), $Q - C$ (1196 nm) to decide on final cycling scheme
  Note usefulness of Q-C laser - both for cycling and magnetic lens STIRAP!