Improved Optical Cycling of TlF

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Introduction

We investigate the $B^3\Pi_1(v_e = 0) \leftarrow X^1\Sigma^+(v_g = 0)$ transition in thallium fluoride by imaging the UV fluorescence from the laser excitation of a cryogenic molecular beam. The investigation is motivated by the promise TlF holds for a measurement of the nuclear electric-dipole moment

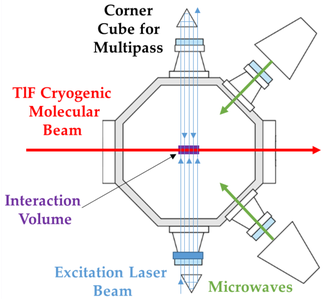

Figure 1. Experimental apparatus.

Type of Angular Momentum	Symbol
Total Less Nuclear Spin	J
Thallium Nuclear Spin	I_1
Total Less Fluorine Nuclear Spin	$F_1 = J + I_1$
Fluorine Nuclear Spin	I_2
Total Angular Momentum	$F = F_1 + I_2$

Table 1. Quantum numbers.

If cycling losses can be limited to only vibrational losses, our branching fraction measurements suggest that we can scatter ~100 photons from each molecule with laser light at 271.7 nm ($v_g = 0$) [1]. The addition of a 278.8 nm ($v_g = 2$) repump laser could allow cycling of up to 1000 photons if other loss mechanisms can be suppressed.

TlF Level Structure and Dark States

Two rotationally closed transitions have been identified, but polarization and hyperfine dark states of the $X(J = 1)$ ground state dramatically reduce their photon cycling rates compared with those of a two-level system (Fig. 2). Natural state evolution produces only a slow evolution of the hyperfine dark states into bright states. However, the rate of cycling out of these dark states can be changed by rapidly switching the excitation laser's polarization and by exciting these two transitions simultaneously.

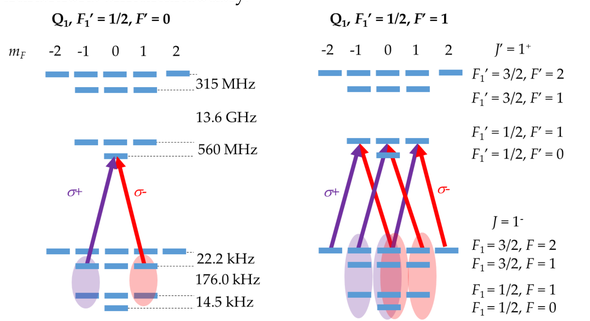

Figure 2. Cycling transitions driven by right- and left-handed circularly polarized light. Due to the unresolved ground state a single polarization couples a single linear combination of ground states to one excited state. The corresponding orthogonal linear combination of ground states are not excited by the laser and pose a serious challenge to cycling.

Imaging

The molecular beam passes through the resonantly-tuned UV laser multipass (Fig. 1). An exotic multipass design which interlaces the multipasses of two pairs of corner prisms increases the interaction time with the molecular beam and helps homogenize the intensity over the interaction region. Molecular fluorescence is then imaged with a UV sensitive CCD camera.

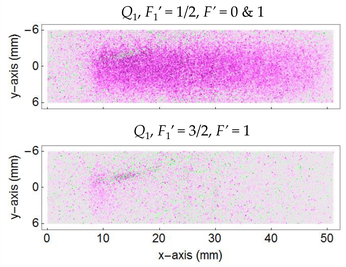

Figure 3. Camera images in which the molecular beam enters from the left side of the image moving in the +x-direction.

Standard Candle

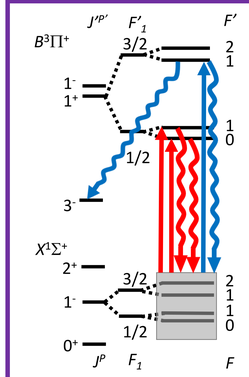

Figure 4. Level diagram.

To determine how many times transitions of interest cycle (red) we compare their fluorescence to the fluorescence of the Q_1, $F_1' = 3/2$, $F' = 1$ calibration transition (blue).

Strong hyperfine mixing allows the $J' = 1$, $F_1' = 3/2$, $F' = 1$ level to decay to the $J = 3$ level [2]. This rotational branching limits the cycling of this calibration transition to about 9 photons on average, indicated by the lack of fluorescence two-fifths of the way through the interaction region (Fig. 5, blue).

Simultaneous Excitation

The fluorescence of the simultaneous excitation of Q_1, $F_1' = 1/2$, $F' = 0$ & 1 remains relatively stable throughout the interaction region, suggesting that the molecules are still cycling as they traverse the final laser pass (red).

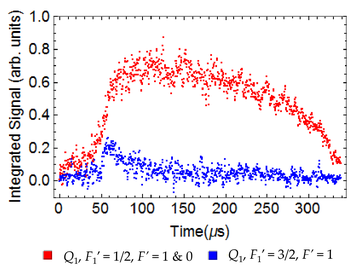

Figure 5. Characteristic decay plots made by taking Fig 3. and summing the intensity over a vertical strip for each value of x. Here the x-axis is converted to units of time.

We find that >80 photons are cycled for the simultaneous excitation scheme (red). This is more photons than either transition alone because simultaneous excitation excites different linear superpositions and consequently destabilizes more dark states, increasing cycling.

Cycle Time

The main limitation on how many photons cycle is the slower than expected cycle time. Although we anticipated that the measured B state lifetime of 99(9) ns would correspond to a minimum cycle time of about 0.5 μs, we find a cycle time of ~3 μs. Empirically we see that the cycle time increases to a point of saturation with increasing power of the exciting lasers.

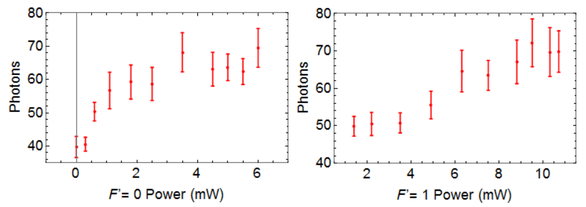

Figure 6. For the simultaneous excitation scheme we fix the initial laser pass power of one exciting laser, and vary the power of the other to guarantee we have achieved saturation.

Resonant Microwaves

In hopes of destabilizing dark states, resonant microwaves were used to drive transitions between $J = 0 \rightarrow 1$ and $1 \rightarrow 2$ rotational ground states (see Fig. 4). Resonant microwaves can both transfer population (Table 2) and destabilize dark states. Consequently they can have significant effects on the characteristic decay, most dramatically for $F' = 0$.

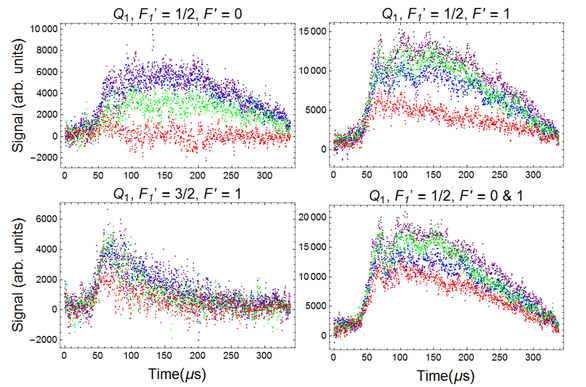

Figure 7. Characteristic decay plots.

With no microwaves, decay to inaccessible states can occur causing a reliance on slow natural state evolution. With the microwaves on, you sample all the states, but you do so slowly, presumably due to the inclusion of additional states in the cycling scheme. Generally we find that microwaves tend to harm cycling for the simultaneous excitation scheme (Table 3).

	J=0⁺	J=2⁺	J=0⁺ & 2⁺
F_1'=1/2, F'=0	>10	>10	>10
F_1'=1/2, F'=1	2.1	1.8	2.4
F_1'=1/2, F'=0 & 1	1.5	1.3	1.7
F_1'=3/2, F'=1	1.8	2.3	2.5

Table 2: Intensity Enhancement Factors

	# Photons
None	83(4)
J=0⁺	70(5)
J=2⁺	51(4)
J=0⁺ & J=2⁺	53(6)

Table 3: Cycled Photons

Vibrational Repumping

To investigate vibrational repumping, optical shutters were used to alternate between blocking and allowing the $v = 2$ repump laser to interact with the molecules. This was done while simultaneously cycling on the Q_1, $F_1' = 1/2$, $F' = 1$ transition. Successful repumping would appear as a difference between the repump on and off signals (Fig. 8). A 50% increase in the repumping fluorescence was expected following directly from the closing of the 1% vibrational branch. No such increase was seen over the ±12 GHz region scanned (Fig. 9).

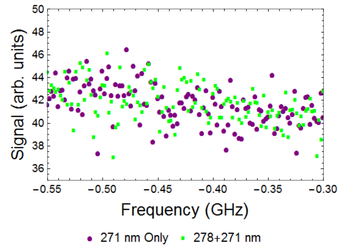

Figure 8. Alternating repump on and off while the repump laser frequency was scanned

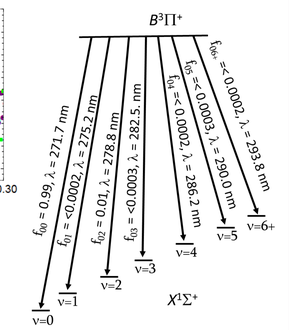

Figure 10. The possible decay paths of the $v_e = 0$ level of the $B^3\Pi_1$ state. The measured branching fractions f_{0v} to the various vibrational levels of the $X^1\Sigma^+$ state are shown.

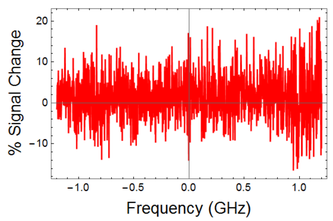

Figure 9. Search for vibrational repumping. Zero frequency corresponds to the expected frequency of the repumping based on well-known Dunham coefficients [3].

Conclusion

We have succeeded in increasing optical cycling on the $v = 0$, X - B Q_1 transition in TlF from about 50 photons to 80 photons by simultaneously exciting the two $F_1' = 1/2$ levels. Vibrational repumping has thus far been unsuccessful. It is possible that the inclusion of the additional states into the cycling scheme effectively slows cycling as we have observed with the microwaves.

Moving forward, Monte Carlo simulations will be performed to help determine why less optical cycling is seen than simple calculations would suggest.

Reference and Acknowledgements

[1] L. R. Hunter et al. Phys. Rev. A **85**, 012511 (2012).

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