Optical Cycling of TlF

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Introduction

We study the optical cycling of the $X^1\Sigma^+(v=0, J=1) \rightarrow B^3\Pi_1(v=0, J'=1)$ transition in TlF by imaging the UV fluorescence from the laser excitation of a cryogenic molecular beam (Fig. 1). Both $Q(J=1)$ transitions which excite to the $F_1' = 1/2$ hyperfine levels have favorable rovibrational branching ratios which should allow for nearly 100 optical cycles when excited by a single UV laser [1]. Such optical cycling would allow for near unit-efficiency internal state detection for the first generation CeNTREX experiment which seeks to precisely measure the time-reversal violating nuclear Schiff moment of thallium.

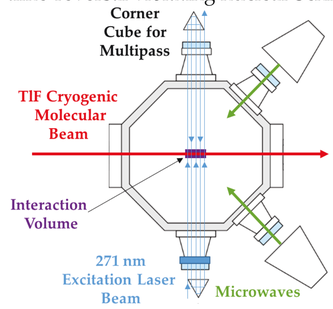

Figure 1. *Experimental apparatus.*

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.*

TlF Level Structure and Dark States

Polarization and hyperfine dark states of the $X(J=1)$ ground state dramatically reduced the 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 increased by rapidly switching the exciting laser's polarization, by driving the microwave transition between the $J = 0$ and $J = 1$ rotational ground states, and by simultaneously exciting more than one of the hyperfine transitions.

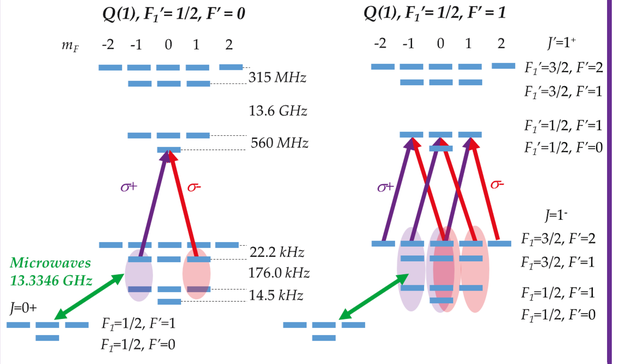

Figure 2. *Cycling transitions driven by right- and left-handed circularly polarized light. (Left) This transition possess multiple dark Zeeman states. (Right) This transition has no dark Zeeman states, but does have hyperfine dark states. Two linear combinations of the three states with m_F = -1,+1 are dark. Two linear combination of the four states with $m_F = 0$ are also dark*

Exotic Multipass

The molecular beam passes through this resonantly-tuned UV laser multipass. A UV sensitive CCD camera allows us to spatially resolve the molecular fluorescence. A new multipass design increases the interaction time with molecular beam and helps homogenize the intensity of the interaction region.

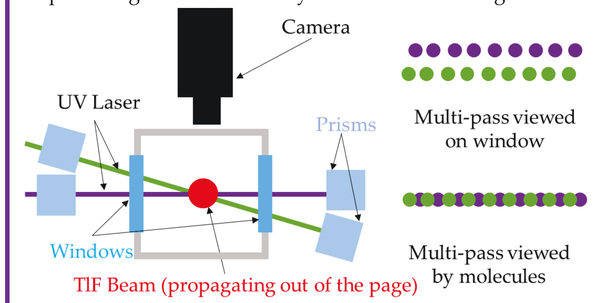

Figure 3. *Exotic Multipass and beam projections.*

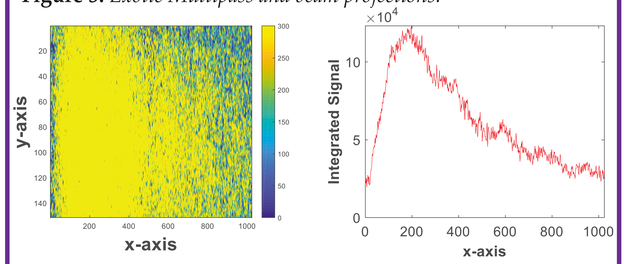

Figure 4. (Left) *Camera image of the Q(1), F_1' = 1/2, F = 1 transition with microwaves and polarization modulation on. The molecular beam enters the left side of the image moving in the +x-direction. Each unit along the x and y-axes corresponds to one pixel. The total viewing area is ~1.5" by 0.2". (Right) This graph is made by taking the image on the left and summing the intensity over a vertical strip for each value of x. This data has been background subtracted and corrected for the optics system collection efficiency.*

Standard Candle

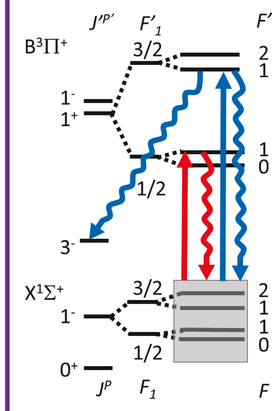

Figure 5. Level diagram.

To determine how many times the $Q(1)$, $F_1' = 1/2$, $F' = 1$ transition cycles (C^*), we compare its integrated signal (P^*) to the integrated (P_{SC}) of another transition. Borrowing language from astronomy, we call this other transition a standard candle because we know how many times it should cycle (C_{SC}) from rotational branching fractions. For the $Q(1)$, $F_1' = 3/2$, $F' = 1$ standard candle we have that:

$$C_* = \frac{P_*}{P_{SC}} C_{SC}$$

Laser Power

Strong hyperfine mixing allows the $J' = 1$, $F_1' = 3/2$, $F' = 1$ level to decay to the $J = 3$ level. This rotational branching limits the cycling of the $Q(1)$, $F' = 3/2$, $F' = 1$ transition to about 7 photons on average. The lack of fluorescence two-fifths of the way through the interaction region indicates that the molecules have mostly decayed to the $J = 3$ dark state at that point (Fig. 6, Left).

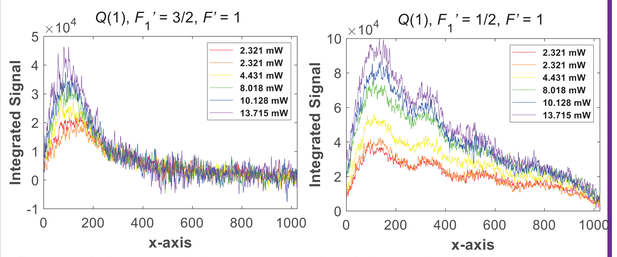

Figure 6. *Integrated fluorescence signal for various laser powers.*

The $Q(1)$, $F_1' = 1/2$, $F' = 1$ transition's fluorescence, unlike the former transition, remains relatively stable throughout the interaction region, suggesting that the molecules are still cycling as they traverse the final pass of the laser (Fig. 6, Right).

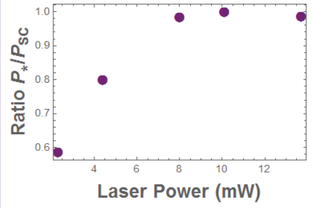

Figure. 7. *The ratio of the integrated fluorescence intensities shown in Figure 6. The ratio plateaus above 8 mW although the individual signal continues to increase presumably because more molecules are participating.*

Microwave Power

Resonant microwaves for the $Q(1)$, $F' = 3/2$, $F' = 1$ transition transfer significant population (1.7x) but do not change the characteristic shape of the decay (Fig. 8, Left). This transition naturally decays to a inaccessible state quickly, the microwave do not significantly effect the rate of the decay.

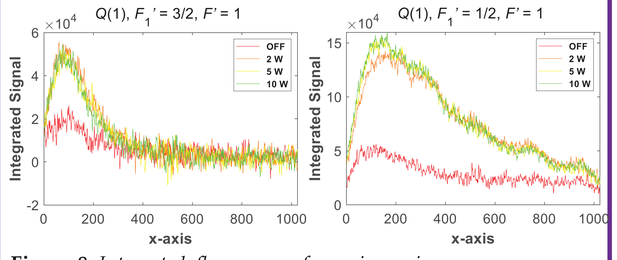

Figure 8. *Integrated fluorescence for various microwave powers*

For the $Q(1)$, $F' = 1/2$, $F' = 1$ transition resonant microwaves transfer significant population (2.6x) and change the characteristic shape of the decay (Fig. 8, Right). With no microwaves, you rely on natural state evolution which is slow and reaches a semi-steady state. With the microwaves on you sample all the states, but you do so slowly.

Resonant vs Non-resonant Microwaves

Resonant microwaves (<40 MHz detuned) cause the fluorescence decay of the $Q(1)$, $F_1' = 1/2$, $F' = 1$ transition to occur rapidly (Fig. 9, Left). Whereas microwaves far off-resonance (>80 MHz detuned) have a characteristic plateauing of their decay (Fig. 9, Right).

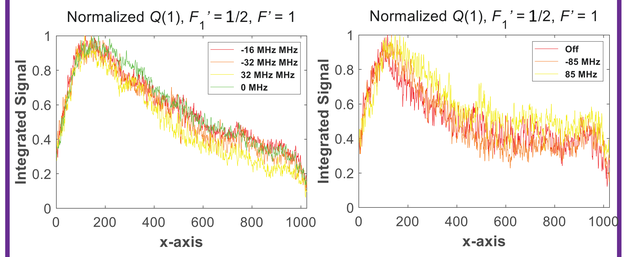

Figure 9. *Normalized fluorescence for various microwave frequencies.*

Population transfer resulting from resonant microwave mixing with the $X(J=0)$ level dramatically affects the fluorescence decay rate by allowing the molecules to sample all the states. Although off-resonance microwaves are expected to perturb energy levels and increase cycling, we see no such evidence.

Multiple Transitions

We use two different lasers to excite both the $Q(1)$, $F' = 1/2$, $F' = 0$ and $Q(1)$, $F' = 1/2$, $F' = 1$ transitions simultaneously. These two lasers excite different linear superpositions and consequently destabilize more dark states, increasing cycling.

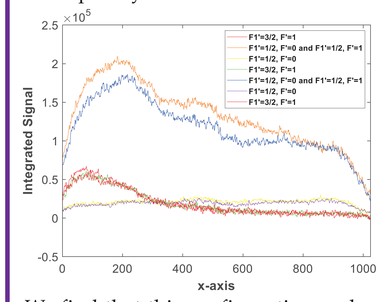

Figure 10. *The fluorescence signal for various Q(1) transitions and combinations.*

We find that this configuration cycles ~50 photons on average. 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 cycle time of about 1 μs, we find that the cycle time is actually about ~4 μs.

Reference and acknowledgements

[1] E. B. Norrgard, E. R. Edwards, D. J. McCarron, M. H. Steinecker, D. DeMille, Shah Saad Alam, S. K. Peck, N. S. Wadia, and L. R. Hunter, Phys. Rev. A **95**, 062506 (2017).

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