Supplementary Material for 'Magnetic Trapping of Molecules via Optical Loading and Magnetic Slowing'

Hsin-I Lu,[1,2] Ivan Kozyryev,[3,2] Boerge Hemmerling,[3,2] Julia Piskorski,[3,2] and John M. Doyle[3,2]

[1]*School of Engineering and Applied Sciences, Harvard University, Cambridge, MA 02138, USA*
[2]*Harvard-MIT Center for Ultracold Atoms, Cambridge, Massachusetts 02138, USA*
[3]*Department of Physics, Harvard University, Cambridge, Massachusetts 02138, USA*

In this supplementary material, we show the experimental results to demonstrate the state transfer of CaF ($N = 0$) during the two-stage optical pumping processes. The Zeeman levels of CaF relevant to the optical pumping scheme are shown as well.

STATE MANIPULATION DURING OPTICAL PUMPING PROCESS

The state transfer of CaF ($N = 0$) during the 1st optical pumping stage with the trap operated at 2.17 T is shown in Fig. 1(a). CaF ($N = 0$) is detected at the trap midplane via laser induced fluorescence excited on the $X^2\Sigma^+(v = 0, N = 0) \rightarrow B^2\Sigma^+(v' = 0, N' = 1)$ transition at 531 nm. We are able to spectroscopically resolve the LFS and HFS in the magnetic field, which are plotted as black and gray traces in Fig. 1(a). The magnetic lens focuses the LFS while defocusing the HFS, resulting in different signal heights. A typical LFS beam (without OPLs applied) arriving at the trap midplane has 10^8 molecules/pulse. When the 1st OPL is applied between 0 and 10 ms, a dip between $10 - 11$ ms shown as blue trace in Fig. 1(a) represents the depletion of the LFS. After the pumping laser is turned off at the saddle, slow molecules take $1 - 2$ ms to reach the trap midplane, causing the LFS signal to recover after 11 ms. Additional HFS molecules (red trace in Fig. 1(a)) appear between $10 - 11$ ms, demonstrating the state transfer.

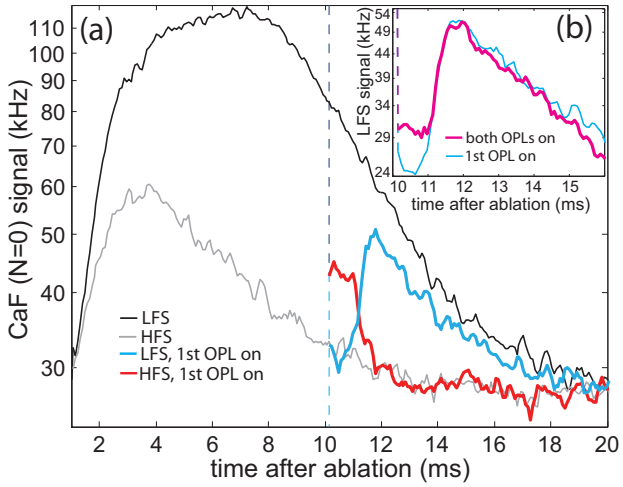

FIG. 1: (a) The LFS (HFS) of CaF ($N = 0$) detected at the trap midplane are shown in black (gray) without the 1st OPL. When the 1st OPL is turned on between $0 - 10$ ms, the PMT is overwhelmed with scattered photons. Blue and red traces represent the LFS and HFS signals when the 1st OPL is switched off (vertical dash line), demonstrating the state transfer from the LFS to HFS. (b) Switching the 2nd OPL on ($9 - 10$ ms) in addition to the 1st OPL ($0 - 10$ ms) pumps the HFS back and recovers the LFS signal between $10 - 11$ ms (pink trace). As a comparison, the blue trace depicts the LFS with only the 1st OPL on.

The full state manipulation necessary for the trap loading is established with the addition of the 2nd OPL, as illustrated in Fig. 1(b). The pink trace represents the LFS signal when the 2nd OPL pumps the HFS (between $9 - 10$ ms) at the saddle in addition to the 1st OPL laser. The replenishment of the LFS owing to the 2nd OPL is observed, yielding an efficiency of 15% for transferring LFS $\rightarrow$ HFS $\rightarrow$ LFS.

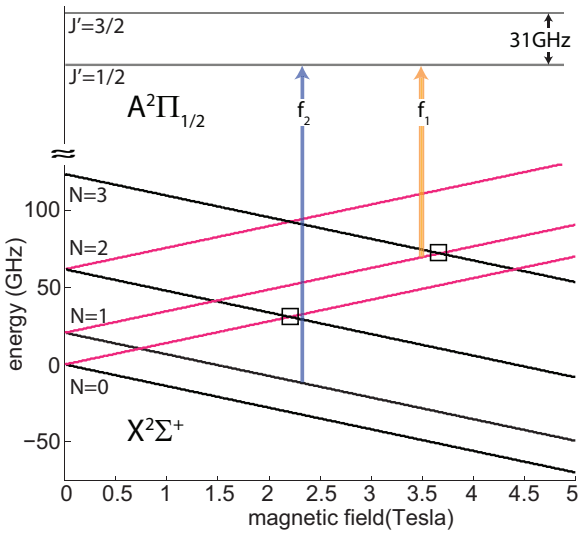

FIG. 2: Zeeman levels of CaF in $X^2\Sigma^+$ ($v = 0, N = 0 - 3$) and $A^2\Pi_{1/2}$ ($v' = 0, J' = 1/2, 3/2$). The low-field seeking and high-field seeking levels of $X^2\Sigma^+$ are plotted in pink and black and the Zeeman levels of $A^2\Pi_{1/2}$ are depicted in gray. Black squares indicate where the avoided crossings occur for the LFS of $N = 0$ and $N = 1$. The orange and blue arrows indicate the optical pumping fields for CaF ($N = 1$) during trap loading.

ZEEMAN LEVELS OF CAF

We plot the Zeeman levels of CaF ($X^2\Sigma^+$) in pink (black) lines for LFS (HFS) up to 5 T in Fig. 2. Note that the low-field seeking state of the rotational level N crosses the high-field seeking state of the $N+2$ state. The anisotropic hyperfine interaction causes these level crossings to become avoided crossings (inside the black squares in Fig. 2) at a field of $B_{\text{avoided}} = (E(N+2) - E(N))/2\mu_B$, where $E(N)$ is the rotational energy. By scanning the frequency of the 1st OPL at different field strengths and measuring the LFS and HFS populations at the trap midplane, we map out the Zeeman levels of CaF and confirm the existence of the avoided crossing for $N = 0$ at $B_{\text{avoided}} = 2.2$ T. When the CaF beam passes through B_{avoided} on the way to the saddle, the LFS adiabatically turn into the HFS, inhibiting the operation of the trap at its full strength. On the other hand, the LFS of $N = 1$ have an increased B_{avoided}=3.67 T due to a larger rotational energy difference between $N = 1$ and $N = 3$, yielding a higher capture velocity than $N = 0$. Therefore, we decide to perform the trapping experiment on $N = 1$ at a field strength of 3.5 T.

Additionally, as pointed out by Ref. [1], the rotational leakage channel can be suppressed by driving a $N = 1 \rightarrow J' = 1/2$ transition (orange and blue arrows in Fig. 2), leading to a more efficient state transfer than $N = 0$. Since the 2nd OPL at frequency f_2 spatially overlaps with the CaF beam, it may excite the LFS to $A^2\Pi_{1/2}(v' = 0, J' = 3/2)$ at certain fields B'. This occurs when $f_2 = f(N = 1 \rightarrow J' = 1/2) + \mu_B B_2 = f(N = 1 \rightarrow J' = 3/2) - \mu_B B'$, where $f(N \rightarrow J')$ represents the transition frequencies at zero field. Given the rotational splitting of the $A^2\Pi_{1/2}$ state, pumping the HFS at $B_2 > 2.2$ T prevents the undesirable removal of the LFS. To load CaF ($N = 1$) into the trap, we choose (B_1, B_2)=(3.5, 2.27) T, yielding $v_c = 29.8 - 33.5$ m/s and a trap depth of $E_D \sim 826$ mK.

TIME DECAY SIGNAL WITH SHUTTER INCORPORATED

After the cryogenic shutter is closed, the trap loss of molecules results from the collisions with the background ^{3}He gas. To observe this process, we use laser spectroscopy to monitor the trap decay of the molecules. Fig. 3 shows the time decay signal of trapped CaF ($N = 1$) when the shutter is incorporated, where the buffer-gas flow is 0.5 sccm during trap loading and the detection laser is switched on at $t_D = 1$ s. The detection process itself is destructive due to decaying to the dark hyperfine states after spontaneous emissions and contributes a decay time constant of $\tau_{pumping}$ to the trapped sample. Therefore, the observed decay time constant of $\tau_{loss} = 251 \pm 28$ ms can be expressed as $\tau_{loss} = (1/\tau_{pumping} + 1/\tau_{BG})^{-1}$. In addition, the time decay signal can be written as $Sig(t') = Sig(0) \times e^{-t_D/\tau_{BG}} \times e^{-t'/\tau_{loss}}$, where $Sig(0)$ is the signal size immediately after the loading process and t' is the delay time relative to t_D. By integrating the time decay signal over a duration of 200 ms (or $t' = 0 - 200$ ms), we can extract τ_{BG} by plotting the integrated signal versus t_D, as shown in Fig. 3 of the main text.

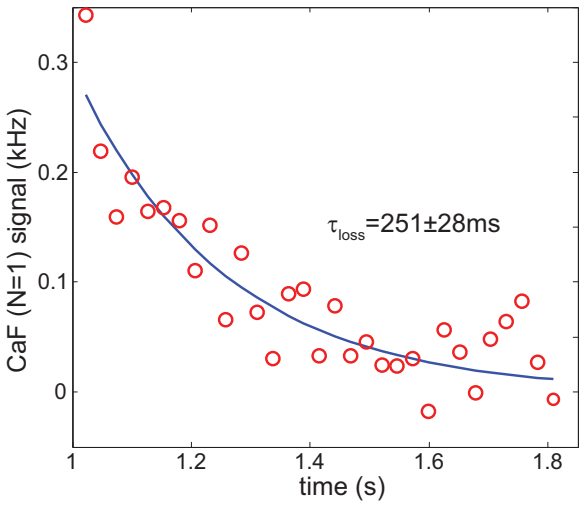

FIG. 3: Time decay signal of CaF ($N = 1$), $Sig(t')$, when the shutter is incorporated. The detection laser is switched on at $t_D = 1$ s.

[1] E. S. Shuman, J. F. Barry, D. R. Glenn, and D. DeMille, Phys. Rev. Lett. **103**, 223001 (2009).