Coherent optical association of single molecules

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We report coherent association of a single NaCs molecule in an optical tweezer through an optical Raman transition. By selecting a deeply bound intermediate state, we suppress the scattering loss during the transfer process. Starting from atoms in their relative motional ground state, we achieve optical transfer efficiency of 50% . The molecule we create have a zero-field binding energy of 770MHz and lifetime up to 1ms . We demonstrate that coherent creation of ground state single molecule is possible, even without Feshbach resonance or narrow optical transition.

Trapped neutral molecules, assembled in an array of optical tweezers, are a promising platform to study quantum infoqrmation and quantum simulations.

In this Letter, we demostrate that a single ground electronic state molecule can be formed from two single optical tweezers by driving an optical Raman transition

Our experiment begins by loading a single ^{23}Na atom and a single ^{133}Cs atom into an optical tweezer from a dual-species MOT into separate optical tweezers. The atoms are imaged to distinguish between loading of two atoms, one atom (Na or Cs), or no atom during post selection. We then perform simutanious Raman sideband cooling (RSC) to cool both atoms into a single 3-dimentional motional ground state in the tweezers. After RSC, the Na tweezer is moved by sweeping the frequency on an acustical optical beam deflector (AOBD) to overlap with the Cs tweezer before smoothly ramping off so that the Na and Cs atoms are merged into the same tweezer . The spin states for the Na and Cs atoms after RSC and during the merge process are $|F = 2, m_F = 2\rangle$ and $|F = 4, m_F = 4\rangle$ respectively. This states combination has a low scattering length of $...a_0$ which allows the two atoms to be merged into the same tweezer with minimum pertubation on each other and remains in their motional ground state after the merge.

After preparing the Na and Cs atoms in the same tweezer in a single quantum state, we perform interaction shift spectroscopy using a Cs Raman transition to drive the Cs into the $|F = 3, m_F = 3\rangle$ state. The new spin state combination has a larger scattering length of $...a_0$ which generates a interaction shift of $...kHz$ in the tweezer. This interaction shift is larger than the differential axial trapping frequency between Na and Cs atoms, which decouples the relative and center of mass motional state and improves the robustness of our preparation of relative motional ground state. The stronger interaction in this spin state also enhances the atomic wavefunction at short range and increases its overlap with the intermediate molecular state used for our Raman transfer.

The optical transfer scheme we used is shown in figure . We use an optical Raman transition to drive the sys-tem from the atomic initial state to a ground electronic molecular state. In order to reduce the size mismatch between the atomic and molecular states, we selected the first bound state for the 3322 spin state as our final state. . For similar reason, the natural choice for the intermediate excited molecular state is one with highly excited motional level. However, from our calculation (and experiment?), the smaller level space for high vibrational state and the smaller detuning from the atomic threshold increases the the scattering rate of the molecular state which causes a reduced Raman Rabi frequency to decoherence rate ratio and a lower transfer efficiency. Therefore, in our experiment, we selected the v=0 state as the intermediate state for our Raman transition.

The pulse sequence for the experiment can be seen in figure (). Instead of adding another beam to drive the Raman transition on the atoms in the tweezer, we use the tweezer itself to achieve this goal. After the total tweezer power is set to the desired value, we smoothly ramp down the power of one frequency in the tweezer while simutaniously ramping up the power of a different frequency so that the total tweezer power remains unchanged. Both frequencies are kept on for a variable length of time before the process is reversed and we return to having a single frequency in the tweezer. The dual use of the tweezer beam ensures that there is not any undesired laser frequency that can interfere with the Raman transition. Using the tweezer beam for the Raman transition also allow us to maximize the intensity of the Raman beams and minimize the transfer time.

Figure () shows the geometry of the experiment setup. A 8.8G B field was used to defines the quantization axis in the experiment and is applied perpendicular to the tweezer axis. The tweezer beam (both frequencies for the Raman transition) has a π polarization relative to the quantization axis, which allows us to selectively drive the Raman transition to a final state with the same total m_F quantum number as our initial state.

This excited state used in the Raman transition was measured in our previous experiment using photoassociation to be at $288...GHz$ from our atomic state. The
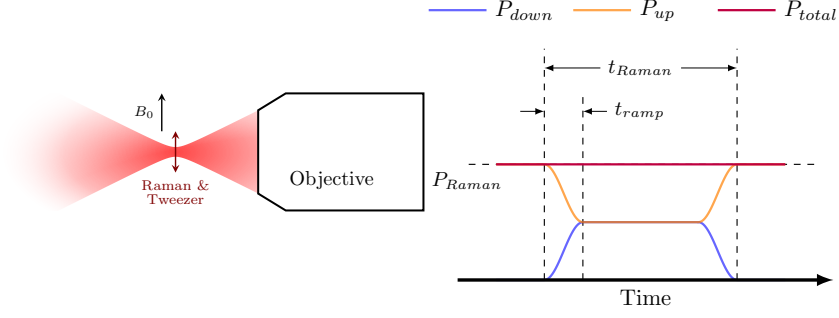
FIG. 1. Optical creation of single molecule from single atoms in tweezer. (A) Schematics of the Raman transition. (B) Geometry and polarization of trap and Raman beam relative to the bias magnetic field. (C) Molecule formation pulse sequence. The tweezer initially consists of only up leg power. This power is smoothly ramped down and the down leg power ramped up over $10\mu s$ while maintaining the total power of the tweezer.
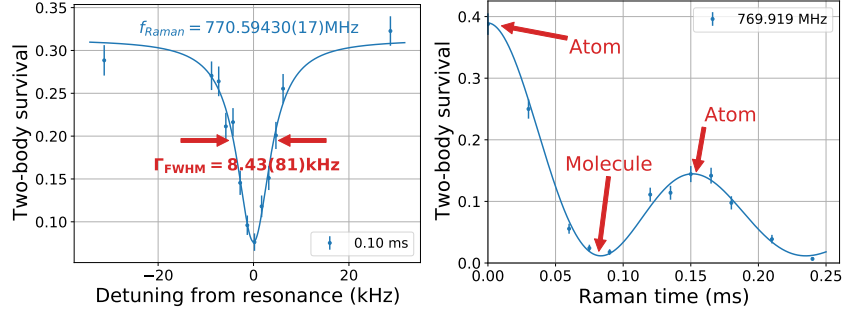

FIG. 2. (A) Raman resonance from atomic state to molecular state, showing Fourier limited linewidth. (B) Rabi oscillation on resonance

ground molecular state has not been observed previously in experimentally. Based on our measurement of FB resonance, interaction shift and the binding energy of the 4422 bound state. Theory prediction was at $770...MHz$.

We locate the Raman resonance for the atom to molecule transition at $770...MHz$ (figure) with a $15mW$ tweezer at $288...GHz$ which corresponds to a $...GHz$ single photon detuning. The background level of $...\%$ corresponds to the propability of preparing the two atoms in the relative motional ground state using interaction shift spectroscopy. When the atoms are transfered into molecule by the Raman transition, there is a decrease in the two body survival since the resulting molecule is not directly detected by our imagine step.

B field dependency $...kHz/G$ which agrees with theory prediction of $...kHz/G$. Dependency on tweezer power $...kHz/mW$, extrapulated to obtain the bare resonance at 0 tweezer power to be at $...MHz$.

After confirming the creating of the molecule via optical Raman transition, we proceed to verify the coherence of the process. This is done by fixing the Raman frequency on the resonance and scanned the pulse time. figure ... shows the observed Rabi oscillation between the atomic and molecular states. Fitting the data with a decaying Rabi oscillation suggests that $..\%$ of initial ground state atoms are transfered into the molecular state.

Combining the frequency and time scan allow us to accurately fit the parameters for the Raman transition. Raman Rabi frequency Molecular state lifetime. The result shows that the decoherence of the Rabi oscillation is caused by finite lifetime of the molecule in the tweezer. We can directly measure this by (figure ...)...

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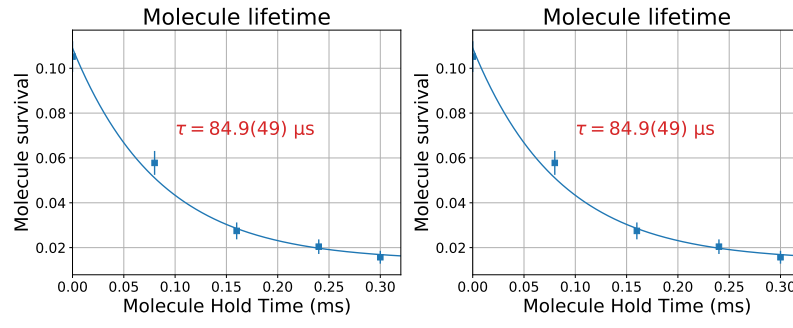† ni@chemistry.harvard.edu

FIG. 3.   (A) Molecule lifetime in 15 mW of trap depth   (B) Two-body atom lifetime in 15 mW of trap depth