

Optical trapping of SrOH molecules for dark matter and T -violation searches

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We report an optical dipole trap of strontium monohydroxide (SrOH) with 1400(300) trapped molecules. Through optical pumping, we access vibrational states that are proposed for improved probes of the electron's electric dipole moment (eEDM) and ultralight dark matter (UDM). For each of these states, the lifetime of trapped molecules is measured and found to be consistent with spontaneous radiative decay and blackbody excitation limits, making this platform viable for these eEDM and UDM searches.

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I. INTRODUCTION

Molecules, due to their rich internal structure, are useful for many applications in quantum science, including quantum information [1–7], quantum chemistry and collisions [8], and precision searches of physics beyond the standard model (BSM) [9–12]. To take full advantage of these applications, trapped ultracold molecules and realization of long internal quantum state coherence times are of key importance [13–16]. Toward these ends, recent laser cooling and optical pumping experiments have yielded several species of diatomic and polyatomic molecules prepared with single quantum state control in the ultracold regime [17–25].

The rotational degree of freedom that is present in all molecules is a resource for quantum science. Generically, polyatomic molecules have an additional useful property, the projection of angular momentum along the internuclear axis. This degree of freedom is a powerful tool for precision measurement [9–11,26] and has potential for quantum simulation [6,27,28]. Linear combinations of these projection states give rise to long-lived parity doublets (PDs). PD states can be fully polarized using only very modest external electric fields, resulting in optically addressable oppositely oriented molecules. PDs also have specific structural features that aid in suppression of systematic errors, as used in the current leading electron electric dipole moment (eEDM) searches [29,30].

A certain class of heavy polyatomic molecules, which includes strontium monohydroxide (SrOH), presents important

opportunities because they are amenable to both laser cooling and optical trapping, while also having long-lived PD states. The $\tilde{X}^2\Sigma^+(010)$ [31] vibrational bending mode in SrOH has such PD states. This makes it a promising candidate for future eEDM measurements that can enjoy the very long coherence times available with optically trapped molecules [9,16]. SrOH can also be used for sensing variations in fundamental constants, another frontier of BSM physics searches. In well-motivated models of dark matter, oscillations of the ultralight dark matter (UDM) field can couple to fundamental constants, such as the proton-to-electron mass ratio $\mu \equiv m_p/m_e$ [32–39]. The gap between pairs of states within a group of closely spaced heterogeneous vibrational levels in SrOH—between a rotational state in the $\tilde{X}^2\Sigma^+(200)$ manifold and a rotational state in the $\tilde{X}^2\Sigma^+(03^10)$ manifold—varies sensitively with changes of μ due to the differing anharmonicities of these vibrational modes. The transitions arising from these $\tilde{X}^2\Sigma^+(200)$ – $\tilde{X}^2\Sigma^+(03^10)$ pairs have the technical advantage of being in the microwave regime. Due to the many rotational states in each vibrational manifold, multiple combinations of UDM sensitive states can be easily studied for systematic error rejection [10].

In this article, we report optical trapping of SrOH. We achieve this through sub-Doppler cooling [40–43], spatial compression with a conveyor-belt (CB) magneto-optical trapping (MOT) [44–47], and single-frequency (SF) cooling to load an optical dipole trap (ODT) [43,48]. We conduct molecular lifetime measurements of selected vibrational states in the ODT. The selected states are sensitive to the eEDM and to variations of μ . These results demonstrate trapped ultracold SrOH for improved searches for time-reversal violating physics (T violation) in the >10 TeV mass range [9] and UDM searches in the 10^{-22} to 10^{-14} eV mass range [10].

II. TRAPPING AND SUB-DOPPLER COOLING

Magneto-optical trapping is a workhorse technique used to reach the ultracold regime. We previously demonstrated

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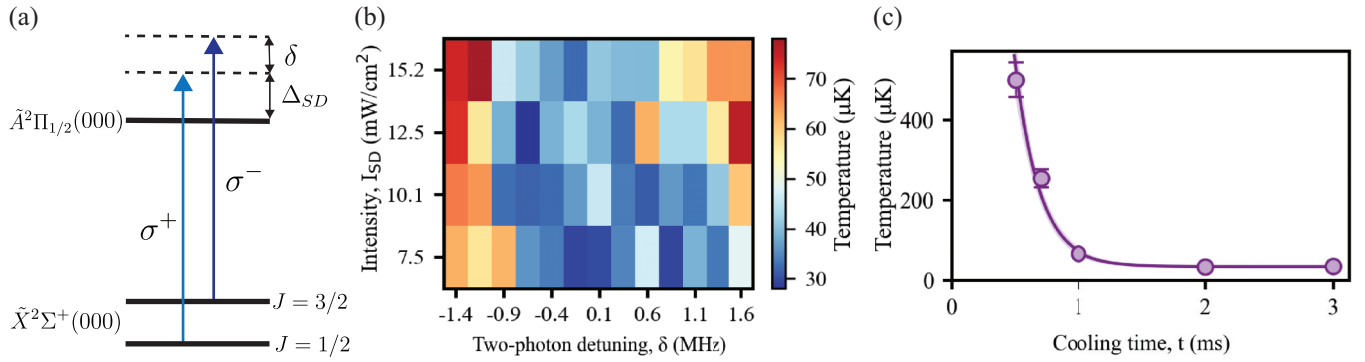


FIG. 1. Sub-Doppler cooling of SrOH. (a) Level diagram showing transitions involved in Λ cooling. For SF cooling, only the frequency addressing the $J = 1/2$ ground state (with σ^+ polarization) is present. (b) Temperature (T) of the SrOH molecules depending on two-photon detuning δ and total cooling light intensity I_{SD} . (c) T vs cooling time (the time over which cooling light is present) under optimal Λ -cooling parameters, where an exponential decay function (e^{-t/τ_Λ}) is fit, yielding a characteristic time of $\tau_\Lambda = 0.19(1)$ ms. For later times, error bars are smaller than the points.

a radio-frequency MOT (rf MOT) of SrOH, using (typical) red detuning from the main transition, $\tilde{X}^2\Sigma^+(000) \rightarrow \tilde{A}^2\Pi_{1/2}(000)$. Details of the laser cooling scheme, slowing technique, and rf MOT can be found in Ref. [24]. In our current work, following initial capture of molecules in the rf MOT, the intensity of the rf MOT light beams is lowered from 3.7 to 0.5 mW/cm^2 , and the magnetic field gradient is increased from 12 to 16 G/cm, over 15 ms. This spatially compresses the rf MOT cloud of molecules from an initial root-mean-squared diameter of $\sigma = 1.3$ mm down to $\sigma = 650$ μm , with a molecular temperature of ~ 1 mK.

Efficient loading of an ODT requires molecules that are much colder than the ODT trap depth, in our case ~ 750 μK . We achieve this with sub-Doppler cooling using blue-detuned optical molasses methods [23,40]. To preserve rotational closure in molecules, “type-II” laser cooling schemes are used, where the angular momentum in the ground manifold exceeds that of the excited manifold. As such, dark states are present in the ground manifold, leading to polarization gradient forces that provide sub-Doppler cooling (heating) for blue (red) detuning relative to the main transition [49–51].

Two sub-Doppler configurations are used in our work, “ Λ cooling” and “SF cooling,” as described below. Experimentally, sub-Doppler cooling is achieved in the following way. First, the rf MOT magnetic field and light polarization switching are both turned off. Then, to form a blue detuned molasses, the detuning of the trapping laser is rapidly changed from ~ 1 Γ (7 MHz) red of the main transition to a blue detuning Δ_{SD} . The realized molasses consists of two frequency components addressing the spin-rotation components $J = 1/2$ and $J = 3/2$ of the ground state $\tilde{X}^2\Sigma^+(000; N = 1)$, with σ^+ and σ^- polarizations, respectively, up to the excited state $\tilde{A}^2\Pi_{1/2}(000; J = 1/2)$. Appropriately tuning the two-photon detuning δ [see Fig. 1(a)] creates zero-velocity dark states and provides enhanced cooling through velocity-selective coherent population trapping [52]. This configuration is known as Λ -enhanced gray molasses cooling (Λ cooling) [41,53]. We use the high capture velocity of Λ cooling to cool the molecules directly from MOT temperatures of ~ 1 mK. Λ -cooling light is applied for 2 ms, longer than the characteristic cooling time $\tau_\Lambda = 0.19(1)$ ms [see Fig. 1(c)]. The resultant

temperature of the molecular cloud is measured using time-of-flight expansion, as described in the Supplemental Material [54]. A minimum temperature of 34(3) μK is achieved with $\Delta_{SD} = 13.2$ MHz, $\delta = -0.4$ MHz, and total light beam intensity $I_{SD} = 3.7$ mW/cm^2 .

Single-frequency cooling [48] is used to directly load the ODT, as described later, after the section on the conveyor-belt MOT. SF cooling achieves lower temperatures than Λ cooling and is less affected by the ODT trap light shifts. However, because its capture velocity is too low to directly cool from the rf MOT, we must first apply Λ cooling. Our SF cooling employs only the frequency component that addresses the $J = 1/2$ spin-rotation state (i.e., the $J = 3/2$ light is simply turned off). We set $\Delta_{SD} = 96$ MHz and $I_{SD} = 9.5$ mW/cm^2 , for both ODT loading and free space cooling studies. With cooling light applied for 5 ms, a minimum temperature of 16.9(1.3) μK is achieved in free space.

III. CONVEYOR-BELT MOT

Once the molecules are cooled to sub-Doppler temperatures using the Λ cooling described above, a key remaining step toward efficient ODT loading (with SF cooling) is further compression of the molecular cloud. Given that the rf MOT has a final size of $\sigma = 650$ μm compared to the ODT waist diameter of $\sigma_{\text{ODT}} = 50$ μm , direct loading from the rf MOT is inefficient due to the large spatial mode mismatch. The advent of the conveyor-belt MOT [44–46] allows for increased compression, with a resulting ~ 10 -fold decrease in cloud diameter. As described in Ref [44], this arises from a slow-moving lattice with molasses that shuttles the trapped molecules toward the magnetic field center where they eventually settle.

As shown in Fig. 2(a), the CB MOT uses a “(1 + 2)” frequency configuration, where one frequency component addresses the $J = 1/2$ spin-rotation level with σ^+ polarization and two other components address the $J = 3/2$ level, with σ^- and σ^+ polarizations. The CB MOT beams share a common blue-detuning Δ_{CB} , and we refer to their relative two-photon detunings as δ_a and δ_b . Experimentally, the CB MOT immediately follows Λ cooling—a dc MOT magnetic field is

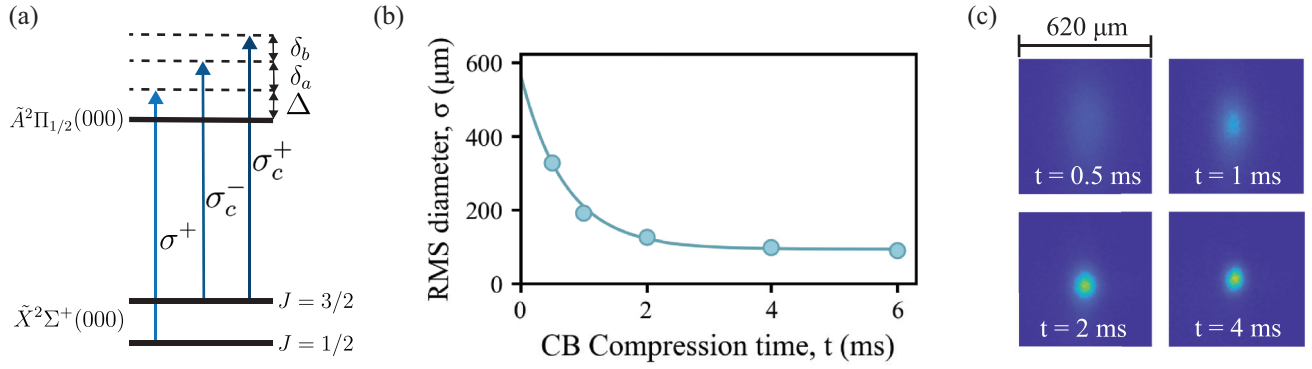


FIG. 2. Conveyor-belt MOT of SrOH. (a) Level diagram showing transitions involved in the CB MOT, where the additional frequency component σ_c^+ is introduced after Λ cooling, forming the conveyor-belt condition. (b) CB MOT spatial rms diameter σ as a function of compression time, where the data are fit to a decaying exponential function ($e^{-t/\tau_{\text{CB}}}$) resulting in $\tau_{\text{CB}} = 0.7(1)$ ms. The 2σ error bars are smaller than the heights of the data points and are not visible. (c) Electron-multiplying charge-coupled device (EMCCD) fluorescence images of molecules compressed in the CB MOT at different compression times t . At $t = 4$ ms, the density of the cloud is increased by a factor of 245.

turned on and the additional light frequency (σ_c^+) is added. To achieve optimal compression, the dc magnetic field gradient, B'_{CB} , is subsequently ramped up and the total CB MOT light intensity, I_{CB} , is ramped down over a time τ_{CB} .

We explore the dependence of the CB MOT size σ on the various detunings, beam intensities, ramp time, and magnitude of the magnetic field gradient. We achieve a temperature of $91(4)$ μK by employing $\Delta_{\text{CB}} = 7.7$ MHz, $\delta_a = -0.9$ MHz, $\delta_b = 0.1$ MHz, and with the light beam intensities ramped from 14.0 to 8.9 mW/cm² and a final magnetic field gradient $B'_{\text{CB}} = 88$ G/cm. The CB MOT is compressed over a time $\tau_{\text{CB}} = 10$ ms, with $\sim 50\%$ of the molecules remaining in the trap. Starting from the rf MOT cloud diameter $\sigma = 650$ μm , CB MOT compression results in a molecular cloud diameter of $\sigma = 83(1)$ μm . This cloud size is well matched to the diameter of our ODT light beam, $\sigma_{\text{ODT}} = 50$ μm .

IV. OPTICAL DIPOLE TRAPPING

After cooling and compression in the CB MOT, SrOH molecules are loaded into an ODT [43] using SF cooling. The ODT is formed by a focused 1064 nm beam with 10.8 W of power, producing a trap depth of ~ 750 μK . To load molecules into the ODT, we turn on SF cooling for 60 ms with the ODT light on. SF cooling is advantageous for this stage as the light shift due to the infrared ODT beam does not appreciably change the optimal cooling detuning. Once the molecules are loaded, the trap is held for a total of 30 ms with all cooling lasers switched off. The molecules are then imaged with Λ -cooling light for 50 ms. Employing an EMCCD camera, we detect the 611 nm wavelength photons emanating from the decay of molecules driven through the $\tilde{B}^2\Sigma^+(000)$ state.

We observe a total number of $N = 1400(300)$ SrOH molecules trapped in the ODT. To measure this quantity, the molecules are recaptured in the rf MOT after being held in the optical trap, and the number of molecules is determined using the procedure detailed in Ref. [24].

V. SCIENCE STATE LIFETIMES IN THE ODT

We measure the lifetimes of molecules in the ODT for the vibrationally excited “science states” $\tilde{X}^2\Sigma^+(010)$, $\tilde{X}^2\Sigma^+(200)$, and $\tilde{X}^2\Sigma^+(03^10)$. The molecules are first prepared in the ODT in the state, after which they are held for varying times (up to $T = 2$ s). The molecule number is measured as a function of T and we fit a decaying exponential to extract the time constant corresponding to the lifetime of the molecules in the trap (see Fig. 3).

We also measure the lifetime of molecules in the ODT without further state preparation after SF cooling, which leaves the population dominantly in the ground state $\tilde{X}^2\Sigma^+(000)$. The lifetime is $\tau = 1.5(0.1)$ s, which is dominated by blackbody radiation (BBR) excitation-driven loss of $\tilde{X}^2\Sigma^+(000)$ molecules [54].

The $\tilde{X}^2\Sigma^+(010)$ state has been proposed for an eEDM measurement [9,16,26] due to its closely spaced parity doublet structure and good sensitivity [55]. Since this state is addressed in our optical cycle, we optically pump into the state by simply turning off the corresponding repump laser that addresses the $\tilde{X}^2\Sigma^+(010) \rightarrow \tilde{B}^2\Sigma^+(000)$ transition. We measure a lifetime of $\tilde{X}^2\Sigma^+(010)$ molecules in the ODT of $\tau_{(010)} = 320(30)$ ms.

The $\tilde{X}^2\Sigma^+(200)$ and $\tilde{X}^2\Sigma^+(03^10)$ states have been proposed for UDM searches [10], where the energy of the $\tilde{X}^2\Sigma^+(03^10)$ state has recently been measured to high precision [56]. The method to prepare the molecules in $\tilde{X}^2\Sigma^+(200)$ is similar to that for $\tilde{X}^2\Sigma^+(010)$, given that it also resides in our optical cycle. We measure a lifetime $\tau_{200} = 135(17)$ ms. For $\tilde{X}^2\Sigma^+(03^10)$, we must incorporate a separate optical pumping step because the state is not present in the optical cycle. This is done by first populating the $\tilde{X}^2\Sigma^+(010)$ state as described above. Next, the $\tilde{X}^2\Sigma^+(010) \rightarrow \tilde{A}(030)\kappa^2\Pi_{1/2}$ transition is driven using ~ 100 mW of laser power. The $\tilde{A}(030)\kappa^2\Pi_{1/2}$ state predominantly decays to the $\tilde{X}^2\Sigma^+(03^10)$ ground state. In order to detect the molecules, they must be transferred back into the optical cycle. This is done by driving the $\tilde{X}^2\Sigma^+(03^10) \rightarrow \tilde{A}(010)\kappa^2\Sigma_{1/2}$ transition using

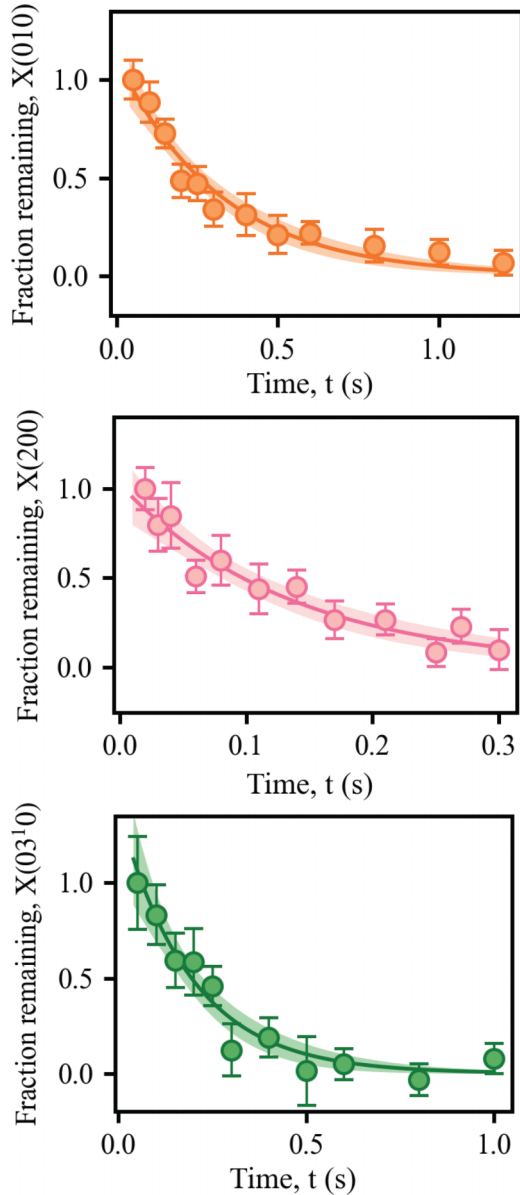


FIG. 3. Science state lifetimes in the ODT. Lifetimes are extracted by measuring the molecular fluorescence over time t and fitting to a decaying exponential function. Error bars are 1σ , and the shaded area around curves corresponds to the 1σ error on the fit. The measured lifetimes are 320(30), 135(17), and 190(30) ms for the $\tilde{X}^2\Sigma^+(010)$, $\tilde{X}^2\Sigma^+(200)$, and $\tilde{X}^2\Sigma^+(03^10)$ states, respectively.

a ~ 100 mW light beam. We report the lifetime $\tau_{(03^10)} = 190(30)$ ms.

All of these science state molecular lifetimes in the ODT are consistent, within reasonable uncertainties, with the theoretically determined decay rates [54], including both spontaneous and BBR-driven loss.

VI. CONCLUSIONS

In summary, we demonstrate an optical dipole trap of SrOH molecules and prepare the trapped molecules in the

$\tilde{X}^2\Sigma^+(010)$, $\tilde{X}^2\Sigma^+(200)$, and $\tilde{X}^2\Sigma^+(03^10)$ science states, which are proposed for searches of BSM physics. The lifetimes of molecules in the ODT are dominantly limited only by spontaneous radiative decay and blackbody radiation excitation loss. The long interrogation times available in this system, on the order of hundreds of milliseconds, make this a promising platform for EDM, UDM, and other future precision measurements. With the demonstrated number of 10^3 trapped SrOH molecules in the ODT, improvement in the search for UDM over the current best limits is available, as proposed in Ref. [10].

We anticipate a future increase in the number of optically trapped molecules through transverse cooling [57,58] of the cryogenic buffer gas cooled molecular beam [59] that feeds the rf MOT. In Ref. [58], calculations show a projected ~ 20 -fold increase in the number of trapped molecules in the rf MOT for SrF molecules by implementing transverse cooling. We expect similar improvements for SrOH. Furthermore, by increasing the ODT diameter to $\sigma > 100$ μm , we expect to achieve $\sim 25\%$ loading efficiency from the CB MOT to the ODT, as demonstrated in Ref. [46], which will result in over a factor of 5 improvement in the loaded number in the ODT. We estimate that these two improvements combined will enable trapping of $\sim 10^5$ molecules in the ODT. With this projected number of molecules, an eEDM measurement with competitive sensitivity to the next projected advanced cold molecule electron EDM (ACME) result should be possible using the $\tilde{X}^2\Sigma^+(010)$ bending mode of SrOH. Methods to conduct such eEDM measurements in polyatomic molecules have been demonstrated in CaOH [16]. In searching for the eEDM, it is important to have experiments with very different possible systematic errors, as is the case with the two best published limits, the ACME and JILA experiments [29,30]. The work here demonstrates the key experimental building block for a viable third experimental approach to eEDM searches and provides a milestone toward trapping of more exotic species, such as RaOH [60,61], which can provide even further improvements in the search for T -violating physics, into the 1000 TeV range.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and

hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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- the vibrational angular momentum associated with the bending mode and is typically omitted when $v_2 = 1$, where $\ell = 1$ is the only allowed value.
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