Submillikelvin Dipolar Molecules in a Radio-Frequency Magneto-Optical Trap

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We demonstrate a scheme for magneto-optically trapping strontium monofluoride (SrF) molecules at temperatures one order of magnitude lower and phase space densities 3 orders of magnitude higher than obtained previously with laser-cooled molecules. In our trap, optical dark states are destabilized by rapidly and synchronously reversing the trapping laser polarizations and the applied magnetic field gradient. The number of molecules and trap lifetime are also significantly improved from previous work by loading the trap with high laser power and then reducing the power for long-term trapping. With this procedure, temperatures as low as 400 μK are achieved.

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Recently, there has been great interest and advancement in producing samples of cold and ultracold (T < 1 mK) polar molecules [1]. The rich internal structure of molecules naturally lends itself to diverse and exciting applications including ultracold chemistry [2], precision measurements [3,4], and quantum simulation [5]. A number of indirect [6–9] and direct [10–13] methods for obtaining ultracold molecules have been demonstrated or are under development. Recently, we have demonstrated use of a magneto-optical trap (MOT) [14,15] to directly cool and trap laser-slowed [16] SrF molecules. A MOT provides simultaneous confinement and cooling, making it the starting point of nearly all experiments with ultracold atoms; the MOT is similarly promising for use with molecules. In this Letter we demonstrate and characterize a scheme for producing a molecular MOT that yields much lower temperature and much higher phase space density than in our previous work.

Most atomic MOTs use a type-I level structure [17] (F → F′ = F + 1, where F is the total angular momentum and the prime indicates the excited state), where all ground states are bright states (here, defined as states addressed by a laser beam that provides a radiative confining force). In molecules, rotational structure requires use of a type-II configuration (F → F′ = F or F → F′ = F − 1), which has ground state sublevels not coupled to a confining laser (dark states) for any fixed polarization. The dark state population must be rapidly returned to bright states to allow significant cooling and confinement. Typically, MOTs based on type-II structures are observed to yield weaker confining forces and higher temperatures than those using a type-I structure [18,19].

In this Letter we demonstrate a molecular MOT which destabilizes dark states using time-varying fields. A 3D confining force is produced by diabatically transferring molecules from dark states to bright states; this is accomplished by rapidly and synchronously reversing the B field and the trapping light polarization [20] [see Fig. 1(a)]. If the switching is done at a frequency \( f_{\text{MOT}} \gtrsim R_{\text{sc}} \), where \( R_{\text{sc}} \) is the molecules’ photon scattering rate, then the molecules spend a reduced amount of time in dark states and should experience a correspondingly larger force. For a two-level system the maximum value of \( R_{\text{sc}} \) is \( \Gamma / 2 \), where the excited state decay rate \( \Gamma \) is typically on the order \( \Gamma \sim 10^7 \text{ s}^{-1} \) [21] (\( \Gamma = 2\pi \times 7 \text{ MHz} \) for SrF). Hence, we refer to the regime \( f_{\text{MOT}} \gtrsim R_{\text{sc}} \) as the radio-frequency (rf) MOT; we distinguish it from the regime \( 0 < f_{\text{MOT}} \lesssim R_{\text{sc}} \), where dark states are destabilized predominantly through other means, which we refer to as the ac MOT. The rf MOT principle was previously used to apply 2D transverse compression on a beam of YO molecules [20], and an ac MOT was used to trap K atoms [22].

In previous work [14,15], we demonstrated MOTs of SrF with static polarizations and B field. Much of this Letter is devoted to a comparison between these MOTs (which we review here briefly) and the rf MOT. The ground state of SrF has resolved spin-rotation and hyperfine (SR/HF) structure; each of these sublevels must be addressed by

![Diagram](image_url)

**FIG. 1.** (a) rf MOT trapping concept. Excitation by a confining laser (the solid arrows) may lead to decay (the dashed arrows) into dark states. The B field and polarization rapidly reverse such that dark states become bright states; this leads to increased confinement and cooling. (b) LIF from trapped molecules vs relative phase \( \phi \) of the B field and the laser polarization. Molecules are only trapped for \( |\phi| \leq 90^\circ \).
a separate optical frequency and polarization to achieve reasonable scattering forces. In our original static molecular MOT [14] (referred to here as the dc MOT), the polarizations of the individual frequency components [23] were chosen so that the states Zeeman shifted closest to resonance were bright states. Surprisingly, this scheme does not maximize the restoring force [24], and the confinement was very weak compared to typical atomic MOTs. In subsequent work [15], the polarizations of the components were chosen so that each, treated independently, provided a restoring force [24,25]. This scheme, referred to as the dc MOT, gave increased confining forces but significantly higher temperatures. In the rf MOT, the polarizations are the same as in the original dc MOT [23].

In this Letter, we show that the rf MOT makes it possible to obtain much higher phase space density than in the dc or dc MOTs: it provides significant confinement and a long lifetime even at low scattering rates, where the lowest temperatures are achieved. Some qualitative explanation of this behavior will be provided by an analytical model of the system.

Much of the experimental scheme has been described elsewhere [14–16,26,27]. Briefly, SrF molecules from a cryogenic beam source [27–29] are slowed by lasers Λ00, Λ10, and Λ21 [16], where Λi denotes a laser tuned to the \[|X^2\Sigma_j\rangle, N = 1, v \rangle \rightarrow |A^2\Pi_j\rangle \text{ transition} \text{ where } P \text{ is the parity}. Slow molecules are captured by the MOT, which contains the additional vibrational repump laser Λ22; due to SrF’s highly diagonal matrix of Franck-Condon factors [21], these four lasers allow each molecule to scatter \(3 \times 10^6 \) photons before decaying to uncoupled vibrational levels \(v \geq 4\). Laser \(\Lambda^{N=3}_0\) addresses decay to \(|v = 0, N = 3\rangle\) caused by off-resonant excitation from the optical cycle to \(J' = 3/2\) excited states [15]. The SR/HF structure is addressed by adding rf sidebands to each of these MOT lasers. Because one SR/HF level requires a different polarization for trapping [14,24], an additional single-frequency trapping laser Λ00 with polarization opposite to Λ00 and tuned closest to the closest sideband of Λ00 to this line allows for a better approximation to the ideal polarization scheme [14,15]. An acousto-optic modulator (AOM) allows the power of main cycling lasers Λ00 and Λ10 to be rapidly changed. Losses from additional optics reduce the maximum Λ00 and Λ10 power to 80 and 30 mW, respectively, compared to 210 and 50 mW in our previous work [14,15]. Trapped molecules are detected by imaging laser-induced fluorescence (LIF) from the main cycling transition onto a CCD camera.

We rapidly switch the polarization of the main cycling lasers using a Pockels cell. The sinusoidally oscillating B-field gradient is produced by a pair of in-vacuum aluminum nitride (AlN) boards with direct bond copper coils on each side. Variable capacitors external to the vacuum chamber form a parallel LC tank circuit with the coils resonant at frequency \(f_{\text{MOT}}\), and impedance match to 50Ω. Adjusting the capacitors allows \(f_{\text{MOT}}\) to be easily varied from dc to 3 MHz (limited by the coils’ self-capacitance) with loaded \(Q \approx 50\) for \(f_{\text{MOT}} > 0.5\) MHz.

A peak current of 5 A is required to produce the optimum rms axial B-field gradient of 9 G/cm. Given our coil inductance of 40 μH, this requires a peak voltage \(V_{pk} = 4\) kV when \(f_{\text{MOT}} = 3\) MHz. We take care to minimize the \(E\) field produced by this voltage drop: the four coils (one on each side of each board) are wired in series, with the coils on the bottom board ordered first and last and the top board coils intermediate, leading to equal average potentials on the two boards. Nevertheless, the residual \(E\) field has observable effects on the trap lifetime (discussed below and in the Supplemental Material [30]).

Defining the time of the laser ablation pulse that initiates a molecular beam pulse as \(t = 0\), we find the maximum population occurs at \(t \approx 70\) ms. Figure 1(b) shows the LIF recorded for a 60 ms exposure starting at \(t = 70\) ms as a function of the relative phase \(\phi\) of the B-field gradient and laser polarization. As expected, the signal is greatest when \(\phi = 0\) and vanishes for \(|\phi| > 90^\circ\). This clearly demonstrates trapping via the rf MOT mechanism. For optimal trap loading, \(\Lambda_{00}\) and \(\Lambda'_{00}\) are detuned from a nominal resonant frequency (the frequency which produces maximum LIF when the lasers are applied orthogonal to the molecular beam in zero B field) by about −9 and −6 MHz, respectively.

The number of trapped molecules is determined from \(R_{sc}\), MOT lifetime \(\tau_{\text{MOT}}\), and integrated LIF intensity from a 60 ms camera exposure as in Ref. [14]. The peak trap population vs \(\Lambda_{00}\) power is plotted in Fig. 2(a) (throughout this Letter, whenever \(\Lambda_{00}\) varies from full power, it is implicit that the \(\Lambda'_{00}\) power is changed by a proportional amount using the AOM). The number of trapped molecules grows monotonically with the laser power. Under optimal conditions, >2000 molecules are loaded into the rf MOT at full power, and there is no clear dependence on \(f_{\text{MOT}}\) in the range 1.23–2.40 MHz. Operation at \(f_{\text{MOT}} = 111\) kHz was much more sensitive to MOT beam alignment than for \(f_{\text{MOT}} > 1\) MHz, so we speculate that the slightly lower number loaded may be attributed to drifting beam pointing.

We find that the rf MOT loads ~3 times more molecules than the dc MOT.

The photon scattering rate \(R_{sc}\) (plotted vs \(\Lambda_{00}\) power in Fig. 2(b)] is determined by comparing the measured decay rate of the LIF signal when \(\Lambda_{32}\) is rapidly shuttered vs when \(\Lambda_{32}\) is present, given the known branching ratio into \(v = 3\) (\(\approx 9.6 \times 10^{-6}\) [33,34]). A simple analytical rate model predicts that the steady-state scattering rate in the MOT has the same form as a two-level system [4]:

\[
R_{sc} = \frac{\Gamma_{\text{eff}}}{2} 1 + \frac{I_{00}}{I_{\text{sat,eff}}} + 4\Delta_{a}^{2}/\Gamma^{2},
\]

Here, \(I_{00}\) is the \(\Lambda_{00}\) intensity, \(I_{\text{sat,eff}}\) is an effective saturation intensity, \(\Gamma_{\text{eff}}/2\) is the maximum scattering rate, and \(\Delta_{a}\) is

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the detuning from resonance. The model is described in more detail in the Supplemental Material [35], where expressions for $\Gamma_{\text{eff}}$ and $I_{\text{sat,eff}}$ in terms of known molecular parameters are also given. These expressions predict $\Gamma_{\text{eff}} = 2\pi \times 1.7 \text{ MHz}$ and $I_{\text{sat,eff}}$ corresponding to saturation power $P_{\text{sat}} = 2.6 \text{ mW}$ for our 7 mm $1/e^2$ radius MOT beams. The solid line in Fig. 2(b) shows a fit of Eq. (1) to the data for $f_{\text{MOT}} = 1.23 \text{ MHz}$. We fix $\Delta = -2\pi \times 9 \text{ MHz}$ and find the best fit parameters to be $\Gamma_{\text{eff}} = 2\pi \times 1.0(1) \text{ MHz}$, about 40% smaller than the predicted value, and $P_{\text{sat}} = 1.9(3) \text{ mW}$, about 25% smaller than predicted. We see that, despite the complexity of the MOT, this simple model predicts the scattering rate quite well. However, we are unable to quantitatively explain the much lower $R_{\text{sc}}$ observed for $f_{\text{MOT}} = 111 \text{ kHz}$, which is well into the ac MOT regime for all but the very lowest $R_{\text{sc}}$ explored here.

In simplifying limits, other well-known equations approximating trap properties, such as the spring constant $k$ and temperature $T$, may be expressed in terms of either $R_{\text{sc}}$ or $I_{00}/I_{\text{sat,eff}}$. Because $R_{\text{sc}}$ may be easily measured in the lab and the expected dependence of the trap properties is simpler when expressed in terms of $R_{\text{sc}}$, we choose to plot the remainder of our data as a function of $R_{\text{sc}}$, rather than power or intensity.

The MOT lifetime $\tau_{\text{MOT}}$ is measured by monitoring LIF as a function of time [see Fig. 2(c)]. Radio-frequency $\mathcal{E}$ fields caused by the large rf voltage applied to the MOT coils mix the excited $|J = 1/2, P = \pm \rangle$ states and lead to unwanted branching to the $N = 0, 2$ ground states. In the absence of a mechanism to couple these states to the main cycling transition, this leads to relatively short trap lifetimes. As shown in Fig. 2(d), we mitigate this loss with a $\sim 5 \text{ mW}$ laser $\mathcal{L}_{N=2}$ (which pumps $N = 2$ into $N = 0$ via $|A^1\Sigma^+_J, J = 1/2, P = -\rangle$) plus microwaves resonant with both $|N = 0, J = 1/2, F = 0, 1\rangle \leftrightarrow |N = 1, J = 1/2, F = 1, 0\rangle$ transitions. For intermediate scattering rates [$R_{\text{sc}} \sim (2-8) \times 10^5 \text{ s}^{-1}$] with these additional fields applied, $\tau_{\text{MOT}}$ is independent of $f_{\text{MOT}}$ and is especially long ($\sim 500 \text{ ms}$). Despite decreasing the maximum $R_{\text{sc}}$ by $\sim 40\%$ roughly the expected amount due to additional levels coupled to the cycling transition [35]; see Fig. 2(b)], microwaves are found to not dramatically affect other trap properties. For the highest scattering rates, the trap loss rate $R_{\text{loss}} = 1/\tau_{\text{MOT}}$ is proportional to $R_{\text{sc}}$, indicative of a scattering-related loss mechanism whose origin is not currently understood. In contrast to the rf MOT, the dc$^+$ MOT lifetime decreases sharply for lower scattering rates, rendering it incapable of trapping at $R_{\text{sc}} \lesssim 10^6 \text{ s}^{-1}$. The dc MOT was found to be even more short lived at any reduced power; the single points in Figs. 2 and 3 were taken from Ref. [14].

While the MOT loads the most molecules at full laser power, we observe that other trap properties improve as the power of the main cycling lasers is reduced. We therefore employ a loading stage at full laser power to capture the largest possible sample, then reduce the $\mathcal{L}_{00}$ and $\mathcal{L}_{00}$ powers. By linearly decreasing the power from its full initial value to a lower final value over a time $t_{\text{ramp}} > 30 \text{ ms}$, the fraction of trapped molecules remaining is near unity, even when the power is reduced by 99.5%. For all data in Fig. 3, we use $t_{\text{ramp}} = 50 \text{ ms}$.
FIG. 3. Filled symbols, microwaves applied; open symbols, without; red line, analytical model fit to $f_{\text{MOT}} = 1.23$ MHz data. Measured (a) spring constant $k$ and (b) temperature $T$ vs scattering rate $R_{sc}$. The maximum $k$ for the dc⁺ MOT is ~2 times that of the rf MOT, but it decreases much more rapidly for lower $R_{sc}$. The analytical model, with $\Gamma_{\text{eff}}$ set to the fit value from $R_{sc}$ vs $I_{00}$, and B-field gradient $\partial B_z/\partial z$ and detuning $\Delta$ set to experimental values, has $\mu_{\text{eff}}$ as the only free parameter. $T$ decreases with falling $R_{sc}$, but at a slower rate than predicted by the analytical model with no free parameters. $T$ is lower for $f_{\text{MOT}} > 1$ MHz than for $f_{\text{MOT}} = 111$ kHz. The model correctly predicts $T \sim T_D$ for a low $R_{sc}$. The spread in values for a given MOT configuration is indicative of variations between measurements made on the same day, while the representative error bars are standard errors of measurements taken on multiple days under similar conditions. (c) Phase space density $\rho$ vs $R_{sc}$ after a linear power ramp from full power. Ramping increases $\rho$ by up to 2.5 orders of magnitude in the rf MOT. In the dc⁺ MOT, $\rho$ increases only slightly when $R_{sc}$ is lowered.

The radial and axial temperatures $T_r$ and $T_z$ and spring constants $\kappa_r$ and $\kappa_z$ are measured by observing the free expansion of molecules released from the MOT. The trapping potential is removed by turning off the B field and the main cycling lasers for a variable duration $t_{\text{TOF}}$, during which time the molecules ballistically expand. The main cycling lasers are then restored, and the molecule cloud is imaged for 2 ms.

The geometric mean spring constant $\bar{k} \equiv (k_r/k_z)^{1/3}$ is shown as a function of $R_{sc}$ in Fig. 3(a). While $\bar{k}$ remains significant even for a low $R_{sc}$ in the rf MOT, it falls dramatically with a decreasing $R_{sc}$ in the dc⁺ MOT. The spread in $\bar{k}$ for different $f_{\text{MOT}}$’s at low $R_{sc}$ is typical of day-to-day fluctuations in $\bar{k}$ due to drifting alignment. In the analytical model detailed in the Supplemental Material [35], $\bar{k}$ is related to $R_{sc}$ by

$$\bar{k} = -\frac{8\Delta k R_{sc} (1 - 2 R_{sc}/\Gamma_{\text{eff}}) \mu_{\text{eff}} (\partial B_z/\partial z)}{3 \times 4^{3/2} \Gamma^2 (1 + 4 \Delta^2 / \Gamma^2)}$$

(2)

where $k$ is the wave number, $\partial B_z/\partial z$ is the axial B-field gradient, and $\mu_{\text{eff}}$ is an effective magnetic moment. The solid line in Fig. 3(a) shows a fit of Eq. (2) to the data with $f_{\text{MOT}} = 1.23$ MHz. In this fit, we fix $\Gamma_{\text{eff}}$ to the value found above and use experimental values $\partial B_z/\partial z = 9$ G/cm and $\Delta = -2 \pi \times 9$ MHz, leaving $\mu_{\text{eff}}$ as the only free parameter. The best fit gives $\mu_{\text{eff}} = 0.32(1) \mu_B$. This is not too different from the level-averaged value $\langle |\mu| \rangle = \sum_i |\mu_i|/n = 0.49 \mu_B$, where $\mu_i$ is the magnetic moment in the weak-field limit of Zeeman sublevel $i$, and the sum is over the sublevels of $(v = 0, N = 1)$. We see that the trapping forces in the rf MOT are reasonably well described by this simple model.

The geometric mean temperature $\bar{T} \equiv T_r^{2/3} T_z^{1/3}$ is shown vs $R_{sc}$ in Fig. 3(b). There is little frequency dependence to $\bar{T}$ for $f_{\text{MOT}} > 1$ MHz, while the 111 kHz ac MOT is substantially hotter. The temperature decreases with decreasing scattering rate for all $f_{\text{MOT}}$’s, qualitatively matching the expected behavior for Doppler cooling [35]:

$$T = -\frac{\hbar \Gamma^2}{8 k_B \Delta} \left( 1 + 4 \Delta^2 / \Gamma^2 \right)$$

(3)

The solid line in Fig. 3(b) shows this predicted behavior, with $\Gamma_{\text{eff}}$ and $\Delta$ fixed as above. There are no free parameters. At low $R_{sc}$’s, we measure $\bar{T}$ as low as 400 $\mu$K, fairly close to the 245 $\mu$K predicted by Eq. (3) (for SrF, the minimum Doppler cooling temperature $T_D = 160$ $\mu$K would be expected for $\Delta = -\Gamma/2\pi \sim 2\pi \times 3$ MHz). Cooling to near the temperature predicted by simple Doppler cooling theory at low intensities has been observed in systems where sub-Doppler cooling mechanisms are weak or absent [37,38]. However, we observe that $\bar{T}$ increases far more rapidly with an increasing $R_{sc}$ than suggested by Eq. (3). Similar behavior has been observed in Sr [37] and is not well understood, but this may be due to stimulated-force heating effects [39–41].

Figure 3(c) shows the phase space density $\rho$ vs $R_{sc}$, inferred from $\bar{k}$, $\bar{T}$, and molecule number. At the highest scattering rates, $\rho$ is similar for the dc, dc⁺, and rf MOTs. Lowering $R_{sc}$ increases $\rho$ by ~2.5 orders of magnitude in the rf MOT, with $\rho$ roughly constant over the range $R_{sc} \sim 10^5$–$10^6$ s⁻¹. The hotter $f_{\text{MOT}} = 111$ kHz ac MOT has $\rho \sim 10$ times lower than rf MOTs with $f_{\text{MOT}} > 1$ MHz. In the dc⁺ MOT, $\rho$ increases only modestly at lower $R_{sc}$’s.

The dc and dc⁺ MOTs have polarization schemes intended to repump dark states with orthogonal and
anticonfining MOT laser beams [15], while in the rf MOT the laser polarizations and the $B$ field are reversed before this repumping occurs. The rf MOT was therefore expected to have a larger value of $\kappa$ than the $dc^+$ MOT. However, we find them to be comparable at high values of $R_m$ [see Fig. 3(a)]. Recently, it was shown that the close spacing of the SR/HF levels in SrF leads to a dual-frequency mechanism, where two oppositely polarized frequency components addressing the same transition can deplete dark states [23,25]. A state which is dark to one component is bright to the oppositely polarized component, so molecules can scatter continuously from a confining beam, with a preference for that beam dictated by the Zeeman shift. This mechanism is responsible for the majority of the confinement in the $dc^+$ MOT [25]. In the rf MOT, dark states in all SR/HF levels are automatically converted to bright states by the time-varying fields.

While the rf MOT was not found to provide a larger restoring force than the $dc^+$ MOT, its ability to provide significant confinement even at greatly reduced power is extremely useful. By loading at high power and ramping to low power, we produce a sample of 2000 molecules with 0.5 s lifetime at 400 $\mu$K with density $6 \times 10^4$ cm$^{-3}$ and phase space density $1.5 \times 10^{-14}$. These correspond to factors of 3, 4, 6, 15, and 1000 improvement, respectively, over previous molecular MOTs [14,15]. There are promising ideas for delivering much larger numbers of slow molecules for loading into a rf MOT, such as microwave guiding [42], bichromatic force slowing [43], or (for molecules with a $^1\Sigma$ ground state, where the magnetic moment is negligible) a Zeeman slower [34,44].

The temperatures reported here, $\approx 3T_D$, are sufficiently cold to have a broad impact on the field. For example, molecular fountains could be constructed to offer long interaction times for precision measurements [4]. Additionally, 400 $\mu$K molecules could be magnetically trapped with standard techniques. Cotrapping with an ultracold atom would allow study of atom-molecule collisions and potentially sympathetic cooling to yet lower temperatures [45–47].

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