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Sub-millikelvin 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 three 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.

Recently, there has been great interest and advancement in producing samples of cold and ultracold ($T < 1\text{ mK}$) 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] ($F \rightarrow F' = F + 1$, 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 ($F \rightarrow F' = F$ or $F \rightarrow F' = F - 1$), 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 paper 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] (Fig. 1a). If the switching is done at a frequency $f_{\text{MOT}} \gtrsim R_{\text{sc}}$, where R_{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_{sc} is $\Gamma/2$, where the excited state decay rate Γ 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 paper 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/hyperfine (SR/HF) structure; each of these sublevels must be addressed by 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-

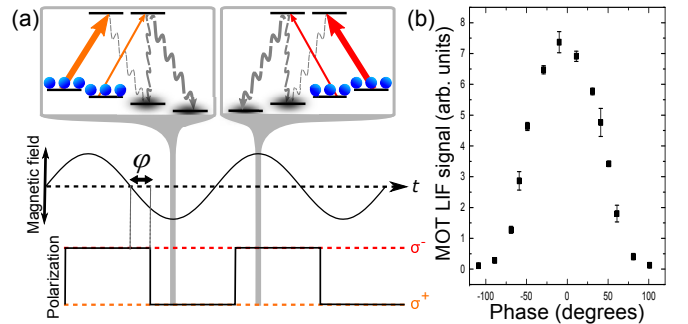


FIG. 1. (Color online) (a) RF MOT trapping concept. Excitation by a confining laser (solid arrows) may lead to decay (dashed arrows) into dark states. The B -field and polarization rapidly reverse such that dark states become bright states; this leads to increased confinement and cooling. (b) LIF from trapped molecules vs relative phase ϕ of B -field and laser polarization. Molecules are only trapped for $|\phi| \lesssim 90^\circ$.

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 work, 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 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 \mathcal{L}_{00} , \mathcal{L}_{10} , and \mathcal{L}_{21} [16], where $\mathcal{L}_{vv'}$ denotes a laser tuned to the $|X^2\Sigma, N=1, v\rangle \rightarrow |A^2\Pi_{1/2}, J=1/2, P=+, v'\rangle$ transition (where P is the parity). Slow molecules are captured by the MOT, which contains the additional vibrational repump laser \mathcal{L}_{32} ; due to SrF’s highly diagonal matrix of Franck-Condon factors [21], these four lasers allow each molecule to scatter $\sim 3 \times 10^6$ photons before decaying to uncoupled vibrational levels ($v \geq 4$). Laser $\mathcal{L}_{00}^{N=3}$ addresses decay to $|v=0, N=3\rangle$ caused by off-resonant excitation from the optical cycle to $J'=3/2$ excited states [15]. 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 \mathcal{L}_{00}^\dagger with polarization opposite to \mathcal{L}_{00} and tuned closer than the closest sideband of \mathcal{L}_{00} 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 \mathcal{L}_{00} and \mathcal{L}_{00}^\dagger to be rapidly changed. Losses from additional optics reduce the maximum \mathcal{L}_{00} and \mathcal{L}_{00}^\dagger power to 80 mW and 30 mW, respectively, compared to 210 mW 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_{MOT} , and impedance match to 50 Ω . Adjusting the capacitors allows f_{MOT} to be easily varied from DC–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 $V_{\text{pk}} = 4$ kV when $f_{\text{MOT}} = 3$ MHz. We take care to minimize the \mathcal{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 top board coils intermediate, leading to equal average potentials on the two boards. Nevertheless the residual \mathcal{E} -field has observable effects on the trap lifetime (discussed below).

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. Fig. 1b shows the LIF recorded for a 60 ms exposure starting at $t=70$ ms as a function of the relative phase ϕ of the B -field gradient and laser polarization. As expected, the signal is greatest when $\phi \approx 0$, and vanishes for $|\phi| \gtrsim 90^\circ$. This clearly demonstrates trapping via the RF MOT mechanism. For optimal trap loading, \mathcal{L}_{00} and \mathcal{L}_{00}^\dagger 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 MHz and -6 MHz, respectively.

The number of trapped molecules is determined from R_{sc} , MOT lifetime τ_{MOT} , and integrated LIF intensity from a 60 ms camera exposure as in [14]. The peak trap population vs \mathcal{L}_{00} power is plotted in Fig. 2a (throughout this work, whenever \mathcal{L}_{00} varies from full power, it is implicit that the \mathcal{L}_{00}^\dagger 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_{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 the slightly lower number loaded may be attributed to drifting beam pointing. We find that the RF MOT loads $\sim 3 \times$ more molecules than the DC* MOT.

The photon scattering rate R_{sc} (plotted vs \mathcal{L}_{00} power in Fig. 2b) is determined by comparing the measured decay rate of the LIF signal when \mathcal{L}_{32} is rapidly shuttered vs when \mathcal{L}_{32} is present, given the known branching ratio into $v=3$ ($\approx 9.6 \times 10^{-6}$ [30, 31]). 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]:

$$R_{\text{sc}} = \frac{\Gamma_{\text{eff}}}{2} \frac{I_{00}/I_{\text{sat,eff}}}{1 + I_{00}/I_{\text{sat,eff}} + 4\Delta^2/\Gamma^2}. \quad (1)$$

Here I_{00} is the \mathcal{L}_{00} intensity, $I_{\text{sat,eff}}$ is an effective saturation intensity, $\Gamma_{\text{eff}}/2$ is the maximum scattering rate, and Δ is the detuning from resonance. The model is discussed in more detail in the Supplemental Materials [32], where expressions for Γ_{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 $7 \text{ mm } 1/e^2$ radius MOT beams. The solid line in Fig. 2b shows a fit of Eq. (1) to the data for $f_{\text{MOT}} = 1.23 \text{ MHz}$. We fix

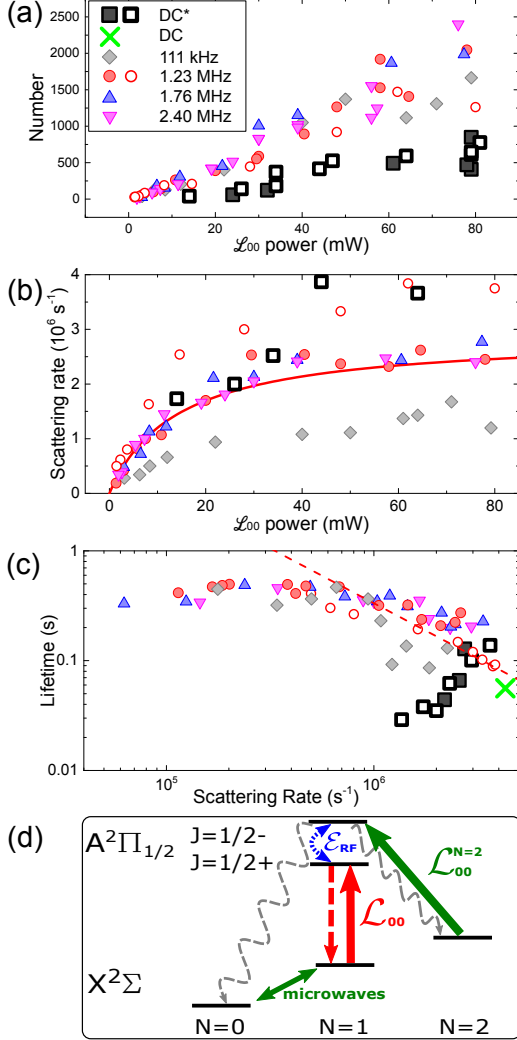


FIG. 2. (Color online. Marker legend in figure. Filled markers with microwaves applied, open markers without.) (a) Number of molecules loaded into the MOT vs \mathcal{L}_{00} power. The number increases with increasing \mathcal{L}_{00} power in all cases. (b) Scattering rate vs \mathcal{L}_{00} power. Solid line is a fit of the model described in the text to $f_{\text{MOT}} = 1.23 \text{ MHz}$ data, with Δ set to $-2\pi \times 9 \text{ MHz}$ and fit values $\Gamma_{\text{eff}} = 2\pi \times 1.0(1) \text{ MHz}$ and $p_{\text{sat}} = 1.9(3) \text{ mW}$. R_{sc} is lower for $f_{\text{MOT}} = 111 \text{ kHz}$ than for $f_{\text{MOT}} > 1 \text{ MHz}$. R_{sc} increases in the absence of repump microwaves by $\sim 40\%$. (c) Trap lifetime τ_{MOT} vs R_{sc} . τ_{MOT} as long as 500 ms are achieved for low R_{sc} in the RF MOT. For higher R_{sc} , $\tau_{\text{MOT}} \propto 1/R_{\text{sc}}$ (dashed line is a $1/R_{\text{sc}}$ trend line), and τ_{MOT} is much shorter in the absence of repump microwaves. For the DC* MOT, τ_{MOT} decreases for lower R_{sc} . (d) Parity mixing (curved blue) due to the coil-induced electric field \mathcal{E}_{RF} causes loss (wiggly grey) from the main cycling transition (vertical red), which is repumped via $\mathcal{L}_{00}^{N=2}$ and resonant microwaves (diagonal green).

$\Delta = -2\pi \times 9 \text{ MHz}$, 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_{sc} observed for $f_{\text{MOT}} = 111 \text{ kHz}$, which is well into the AC MOT regime for all but the very lowest R_{sc} explored here.

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

The MOT lifetime τ_{MOT} is measured by monitoring LIF as a function of time (Fig. 2c). RF \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. 2d, we mitigate this loss with a $\sim 5 \text{ mW}$ laser $\mathcal{L}_{00}^{N=2}$ (which pumps $N = 2$ into $N = 0$ via $|A^2\Pi_{1/2}, J = 1/2, P = -\rangle \leftrightarrow |N = 1, J = 1/2, F = 1, 0\rangle$ transitions. For intermediate scattering rates ($R_{\text{sc}} \sim 2 - 8 \times 10^5 \text{ s}^{-1}$) with these additional fields applied, τ_{MOT} is independent of f_{MOT} and is especially long ($\sim 500 \text{ ms}$). Despite decreasing the maximum R_{sc} by $\sim 40\%$ (roughly the expected amount due to additional levels coupled to the cycling transition [32], see Fig. 2b), 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_{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 [14].

While the MOT loads the most molecules at full laser power, we observe 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}^\dagger 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 $t_{\text{ramp}} = 50 \text{ ms}$.

The radial and axial temperatures T_ρ and T_z and spring constants κ_ρ and κ_z are measured by observing

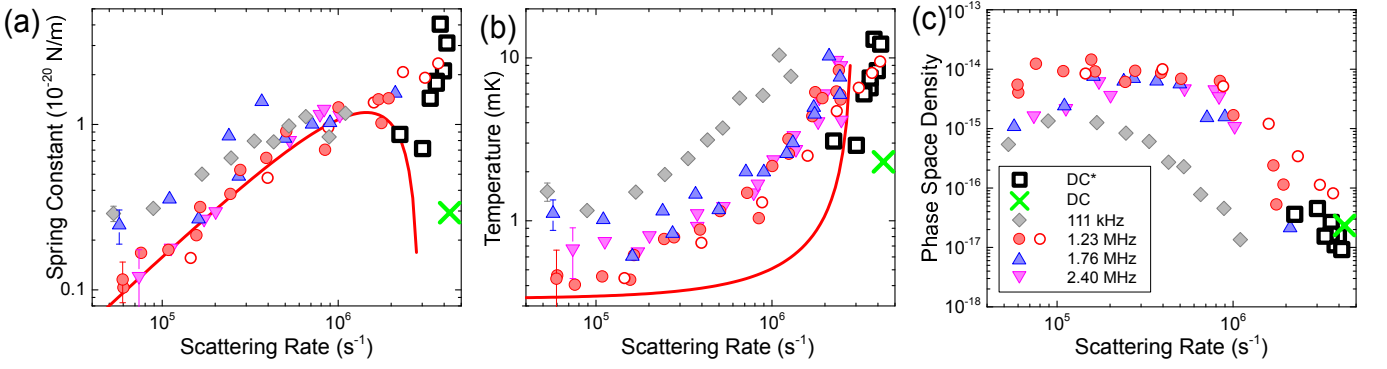


FIG. 3. (Color online. Marker legend in figure. Filled markers with microwaves applied, open markers without. Red line analytical model fit to $f_{MOT} = 1.23$ MHz data.) Measured (a) spring constant $\bar{\kappa}$ and (b) temperature \bar{T} vs scattering rate R_{sc} . The maximum $\bar{\kappa}$ for the DC* MOT is $\sim 2\times$ that of the RF MOT, but decreases much more rapidly for lower R_{sc} . The analytical model, with Γ_{eff} set to the fit value from R_{sc} vs I_{00} , and B -field gradient $\partial B_z/\partial z$ and detuning Δ set to experimental values, has μ_{eff} as the only free parameter. \bar{T} decreases with falling R_{sc} , but at a slower rate than predicted by the analytical model with no free parameters. \bar{T} is lower for $f_{MOT} > 1$ MHz than for $f_{MOT} = 111$ kHz. The model correctly predicts $\bar{T} \sim T_D$ for low R_{sc} . The spread in values for a given MOT configuration are 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 ρ vs R_{sc} after a linear power ramp from full power. Ramping increases ρ by up to 2.5 orders of magnitude in the RF MOT. In the DC* MOT, ρ increases only slightly when R_{sc} is lowered.

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_{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 $\bar{\kappa} \equiv \kappa_\rho^{2/3} \kappa_z^{1/3}$ is shown as a function of R_{sc} in Fig. 3a. While $\bar{\kappa}$ remains significant even for low R_{sc} in the RF MOT, it falls dramatically with decreasing R_{sc} in the DC* MOT. The spread in $\bar{\kappa}$ for different f_{MOT} at low R_{sc} is typical of day-to-day fluctuations in $\bar{\kappa}$ due to drifting alignment. In the analytical model detailed in the Supplemental Material [32], $\bar{\kappa}$ is related to R_{sc} by

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

where k is the wavenumber, $\partial B_z/\partial z$ is the axial B -field gradient, and μ_{eff} is an effective magnetic moment. The solid line in Fig. 3a shows a fit of Eq. (2) to the data with $f_{MOT} = 1.23$ MHz. In this fit, we fix Γ_{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 μ_{eff} as the only free parameter. The best fit gives $\mu_{eff} = 0.32(1) \mu_B$. This is not too different from the level-averaged value $\langle |\mu| \rangle = \sum_i^n |\mu_i|/n = 0.49 \mu_B$, where μ_i is the magnetic moment in the weak-field limit of Zeeman sub-level i , and the sum is over the sub-levels 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 $\bar{T} \equiv T_\rho^{2/3} T_z^{1/3}$ is shown vs R_{sc} in Fig. 3b. There is little frequency dependence to \bar{T} for $f_{MOT} > 1$ MHz, while the 111 kHz AC

MOT is substantially hotter. The temperature decreases with decreasing scattering rate for all f_{MOT} , qualitatively matching the expected behavior for Doppler cooling [32]:

$$T = -\frac{\hbar \Gamma^2}{8k_B \Delta} \frac{(1 + 4\Delta^2/\Gamma^2)}{(1 - 2R_{sc}/\Gamma_{eff})}. \quad (3)$$

The solid line in Fig. 3b shows this predicted behavior, with Γ_{eff} and Δ fixed as above. There are no free parameters. At low R_{sc} , 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 $\Delta = -\Gamma/2 \approx -2\pi \times 3$ 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 [33, 34]. However, we observe \bar{T} increases far more rapidly with increasing R_{sc} than suggested by Eq. (3). Similar behavior has been observed in Sr [33] and is not well understood, but may be due to stimulated-force heating effects [35–37].

Fig. 3c shows the phase space density ρ vs R_{sc} , inferred from $\bar{\kappa}$, \bar{T} , and molecule number. At the highest scattering rates, ρ is similar for the DC, DC*, and RF MOTs. Lowering R_{sc} increases ρ by ~ 2.5 orders of magnitude in the RF MOT, with ρ roughly constant over the range $R_{sc} \sim 10^5$ – $10^6 s^{-1}$. The hotter $f_{MOT} = 111$ kHz AC MOT has $\rho \sim 10\times$ lower than RF MOTs with $f_{MOT} > 1$ MHz. In the DC* MOT, ρ increases only modestly at lower R_{sc} .

The DC and DC* MOTs have polarization schemes intended to repump dark states with orthogonal and anti-confining MOT laser beams [15], while in the RF MOT the laser polarizations and B -field are reversed before this

repumping occurs. The RF MOT was therefore expected to have a larger value of $\bar{\kappa}$ than the DC* MOT. However, we find them to be comparable at high values of R_{sc} (Fig. 3a). 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 μK with density $6 \times 10^4 \text{ cm}^{-3}$ and phase space density 1.5×10^{-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 an RF MOT, such as microwave guiding [38], bichromatic force slowing [39], or (for molecules with a $^1\Sigma$ ground state, where the magnetic moment is negligible) a Zeeman slower [31, 40].

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 μK molecules could be magnetically trapped with standard techniques. Co-trapping with an ultracold atom would allow study of atom-molecule collisions and potentially sympathetic cooling to yet lower temperatures [41–43].

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