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Boerge Hemmerling$^{1,2,7,8}$, Eunmi Chae$^{1,2,8}$, Aakash Ravi$^{1,2}$, Loic Anderegg$^{1,2}$, Garrett K Drayna$^{2,3}$, Nicholas R Hutzler$^{1,2}$, Alejandra L Collopy$^{4,5}$, Jun Ye$^{4,5}$, Wolfgang Ketterle$^{2,6}$ and John M Doyle$^{1,2}$

$^1$Department of Physics, Harvard University, Cambridge, MA 02138, USA
$^2$Harvard-MIT Center for Ultracold Atoms, Cambridge, MA 02138, USA
$^3$Department of Chemistry and Chemical Biology, Harvard University, Cambridge, MA 02138, USA
$^4$JILA, National Institute of Standards and Technology and University of Colorado, Boulder, CO 80309, USA
$^5$Department of Physics, University of Colorado, Boulder, CO 80309, USA
$^6$Department of Physics, Massachusetts Institute of Technology, Cambridge, MA 02139, USA

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Abstract

Laser slowing of CaF molecules down to the capture velocity of a magneto-optical trap for molecules is achieved. Starting from a two-stage buffer gas beam source, we apply frequency-broadened ‘white-light’ slowing and observe approximately $6 \times 10^4$ CaF molecules in a single pulse with velocities $10 \pm 4 \text{ m/s}$. CaF is a candidate for collisional studies in the mK regime. This work represents a significant step towards magneto-optical trapping of CaF.

Keywords: laser slowing of molecules, molecular magneto-optical trap, white-light slowing, cryogenic buffer-gas beam source

(Some figures may appear in colour only in the online journal)

Coherent association of ultracold atoms has been successful in generating ultracold ground state molecules at high phase-space densities [22–26]. However, this approach so far is restricted to atomic species with easily accessible laser cooling transitions. This method is not yet applicable to a large variety of molecules, including free radicals such as calcium monofluoride (CaF). An alternative approach is the direct cooling of molecules. A direct cooling scheme typically starts with slowing of molecules to load a trap, where further cooling can take place to reach ultracold temperatures.

Various approaches have been pursued for trapping molecules, including electrostatic traps [27–31], magnetic traps [18, 32–38], and magneto-optical traps (MOTs) [39–42]. These traps are typically $\lesssim 1$ K deep, and therefore a source of cold molecules is necessary. The highest intensity source of cold and slow molecules is the buffer gas beam [43–46], which utilizes collisions with an inert, cryogenic gas. However, even these slow sources still typically inhibit direct loading of traps like a MOT since the vast majority of molecules have velocities above the trap’s capture velocity.

The creation and control of samples of ultracold atoms enabled many milestones in atomic physics, such as Bose–Einstein condensation [1, 2], quantum simulations of many-body systems [3, 4], development of quantum information systems [5], precision measurements and atomic clocks [6, 7]. Ultracold polar molecules may advance these and other areas of science even further, owing to the molecules’ additional degrees of freedom, large electric dipole moments, and chemical characteristics. These properties are at the core of many proposals and experiments [8], including quantum simulation of strongly correlated systems [9–11], precision measurements and tests of fundamental physics [12, 13], quantum information processing [14–16], studies of ultracold collisions [17, 18], and control of ultracold chemical reactions [19–21]. However, a basic requirement for many proposed experiments is trapped molecules at temperatures around 1 mK or below.

$^7$Present address: Department of Physics, University of California, Berkeley, California 94720, USA.
$^8$These authors contributed equally to this work.
Hence, an initial slowing stage is required to provide a significant fraction of molecules that can be trapped. At present, several groups have managed to slow molecules in various ways [27, 38, 47–49], including laser cooling and radiation pressure slowing [50–52]. Here, we present laser slowing of CaF molecules, originating from a two-stage cryogenic buffer-gas beam (CBGB) source [44, 45], to velocities around 10 m s⁻¹, which is below the expected capture velocity of \( \sim 14 \text{ m s}^{-1} \) of a molecular MOT for CaF using the \( A(v' = i) - X(v'' = f) \) transition and the laser cooling scheme employed [53, 54].

The complex internal structure of molecules renders the use of a Zeeman slower difficult. Instead, ‘white-light’ slowing is used, in which spectrally broadened lasers counterpropagate with respect to the molecular beam to address a range of velocity classes and the internal hyperfine structure of the molecules, as the molecules decelerate [50, 52]. Due to the divergence of the molecular beam, a significant fraction of the molecules do not reach the MOT capture volume. As a result, the total number of molecules inside a molecular MOT has never surpassed 2000 in recent experiments [40–42]. To realize many of the proposed applications of trapped molecules, such as evaporation of the trapped sample in a subsequent (magnetic) trap, higher molecule numbers are required. The most straightforward way to increase the number of molecules that reach the MOT capture volume is to shorten the distance required for slowing, which in turn increases the solid angle of molecules captured from the source. This is achieved here by starting from slower initial forward velocities by using a two-stage CBGB source and by using a low mass molecule (which can be decelerated over a shorter distance), at the cost of lower on-axis beam intensity compared to a single stage source. This two-stage CBGB produces a molecular beam with peak forward velocity as low as 60 m s⁻¹, more than a factor of two slower than a single-stage buffer-gas beam source, and has a flux of \( \sim 10^4 \) molecules/steradian/state/pulse [45].

CaF is a favorable candidate for laser cooling due to its highly diagonal Franck–Condon factors (with a measured \( A(v' = 0) - X(v'' = 0) \) branching ratio of \( f_{00} = 0.987 \) [55, 56]. It is a \( ^2\Sigma^+ \) molecule which has a free electron in its outermost orbital. This electron’s spin degree of freedom makes CaF attractive and distinct from bi-alkali molecules. It also has a large electric dipole moment of 3 Debye. The relevant energy levels of CaF are shown in figure 1. The lowest electronic excited state \( (A^2\Pi_{1/2}) \) has a lifetime of 19.2 ns [56].

We follow the laser cooling scheme reported in [50]. A CaF molecule can scatter about \( 10^5 \) photons with the \( X(v'' = 0) - A(v' = 0) \) (main) laser and the two vibrational repump lasers (figure 1) before decaying into the higher vibrational states. Rotational branching within each vibrational manifold is avoided by driving a \( P(1) \) rotational transition [57]. Due to the interaction of the electron spin \( S = 1/2 \) and the fluorine’s nuclear spin \( I = 1/2 \), the rotational state splits into four hyperfine states. Each of these states need to be addressed with laser radiation to keep the molecules in the optical cycle (figure 1 inset). All slowing lasers are spectrally broadened to cover the hyperfine splittings in the ground states and to compensate the changing Doppler shift as the molecules are decelerated. The maximum starting velocity of a molecule that can be addressed in this configuration is then determined by the total width of the spectra of the slowing lasers and their interaction time with the molecules. While a broad spectrum allows for addressing high velocities, the resulting lower power density limits the scattering rate which, in turn, limits the deceleration rate. An alternative approach that would maintain a higher power density is chirped slowing, which was successfully implemented with YO [52]. This latter approach was not employed by us as frequency chirping of the slowing laser pulses is limited by the finite bandwidth of the laser servos (<100 Hz).

Figure 2 shows the spectrum of the main slowing laser. The spectrum is broadened by two sequential phase-modulating electro-optic modulators (EOMs): one covers the hyperfine splitting of the ground state with a 24.8 MHz modulation frequency with a modulation index of \( \approx 4 \). The second EOM is driven at a frequency of 4.5 MHz with a modulation index of \( \approx 17 \). This drive frequency is chosen to be about half the excited state linewidth of 8.29 MHz to maintain the resonance condition throughout the slowing
The slowing laser is sent in a direction counter-propagating with respect to the molecular beam. Field coils to remix the dark magnetic substates are placed between the cell and the detection region. The slowing laser while keeping the 1st and 2nd repump lasers on for both cases. This removes the effect of optical pumping of the naturally populated excited vibrational states in the molecular beam. Figure 4(a) shows the CaF beam signal with only the repump lasers applied. The arrival time for each velocity agrees well with the time-of-flight hyperbola in the plot. When the slowing lasers are applied, molecules with velocities as low as 10 m s\(^{-1}\) are observed. They arrive ahead of the predicted time from time-of-flight, indicating that they have been slowed down from a higher initial velocity. The time-integrated signal (figure 5(a)) shows that slowing lasers modify the velocity distribution of the beam: the number of molecules with speeds >80 m s\(^{-1}\) has decreased and that of molecules with speeds <80 m s\(^{-1}\) has increased. It should be noted that the white-light slowing process mainly shifts the velocity of the molecules and does not bunch them up at a final velocity due to the soft edge of our broadened light spectrum. This together with the initial velocity distribution and transverse spreading determines the final velocity distribution.

The signal from the slowest molecules is shown in figure 5(b). Molecules with velocities between 10 ± 4 m s\(^{-1}\) (which is near the expected capture velocity of a MOT for CaF) are observed. They arrive ~10 ms after they leave the cell at 2 ms, indicating that their initial longitudinal velocity was at least 50 m s\(^{-1}\). To within a factor of two, using the laser-induced fluorescence signal with and without the main slowing laser while keeping the 1st and 2nd repump lasers on for both cases. This removes the effect of optical pumping of the naturally populated excited vibrational states in the molecular beam. Figure 4(a) shows the CaF beam signal with only the repump lasers applied. The arrival time for each velocity agrees well with the time-of-flight hyperbola in the plot. When the slowing lasers are applied, molecules with velocities as low as 10 m s\(^{-1}\) are observed (figure 5(b)). They arrive ahead of the predicted time from time-of-flight, indicating that they have been slowed down from a higher initial velocity. The time-integrated signal (figure 5(a)) shows that slowing lasers modify the velocity distribution of the beam: the number of molecules with speeds >80 m s\(^{-1}\) has decreased and that of molecules with speeds <80 m s\(^{-1}\) has increased. It should be noted that the white-light slowing process mainly shifts the velocity of the molecules and does not bunch them up at a final velocity due to the soft edge of our broadened light spectrum. This together with the initial velocity distribution and transverse spreading determines the final velocity distribution.

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slowing laser stays resonant while the molecules are being slowed, similar to [51, 59].

In summary, we have demonstrated laser slowing of CaF molecules from a two-stage buffer-gas beam. The slow initial velocity of our source allows slowing over a shorter distance \( \approx 20 \) cm; this, in turn, could lead to an overall increase in the number of molecules captured by a MOT. Once the molecular MOT is achieved, we plan to co-trap an atomic species to study atom-molecule collisions and the possibility of sympathetic cooling of CaF. A promising coolant atom for this endeavor is Li, where the ratio of elastic-to-inelastic scattering rates is predicted to be favorable [60, 61]. We expect the co-loading of an atomic species to be straightforward since it has been demonstrated that an atomic MOT can be loaded directly from our buffer-gas source without additional laser slowing [62]. Generalization of such methods to molecules may pave the way to using ultracold molecules for probing new physics, such as the study of exotic phases of matter using the spin degree of freedom and long-range dipole–dipole interactions of polar molecules [11].

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