Bright, continuous beams of cold free radicals

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We demonstrate a cryogenic buffer gas-cooled molecular beam source capable of producing bright, continuous beams of cold and slow free radicals via laser ablation over durations of up to 60 seconds. The source design uses a closed liquid helium reservoir as a large thermal mass to minimize heating and ensure reproducible beam properties during operation. Under typical conditions, the source produces beams of our test species SrF, containing \(5 \times 10^{12}\) molecules per steradian per second in the \(X\Sigma^+(v = 0, N = 1)\) state with a rotational temperature of \(1.0(2)\) K and a forward velocity of \(140\) m/s. The beam properties are robust and unchanged for multiple cell geometries but depend critically on the helium buffer gas flow rate, which must be \(\geq 10\) standard cubic centimeters per minute to produce bright, continuous beams of molecules for an ablation repetition rate of 55 Hz.

Beams of cold and slow molecules from cryogenic buffer gas sources have played a central role in recent improved precision measurements [1, 2], high-resolution spectroscopy [3, 4] and the direct laser cooling and trapping of molecules at ultracold temperatures [5, 6]. Direct cooling methods for molecules have the potential to produce a chemically diverse range of diatomic and polyatomic species at ultracold temperatures which are well-suited for proposed applications including tests of fundamental physics [7], and controlled chemistry [8].

Cryogenic buffer gas beam sources rely on a flow of cold inert gas, usually helium or neon, to sympathetically cool the molecular species of interest and collapse the occupied rovibrational state distribution [2, 12]. This thermalization takes place inside an enclosed cell as the species of interest becomes entrained within the inert gas flow and exits the cell through a small hole to form a beam. These molecular beams have forward velocities between \(\sim 50\) and \(200\) m/s and advances in slowing techniques using radiation pressure [13, 14] have enabled molecules below \(\sim 10\) m/s to be captured and cooled by magneto-optical traps (MOTs) [15]. Today’s molecular MOTs provide confining and damping forces comparable to those in atomic MOTs but can only capture \(10^4 - 10^5\) molecules at densities up to \(\sim 10^7\) cm\(^{-3}\) due to the low trappable flux and short loading times (\(\sim 20\) ms) attainable when loading single pulses of molecules. While the first interactions between laser-cooled molecules were recently observed [16], many proposed applications require larger trapped samples at higher density and increasing the trappable flux remains a key challenge. More efficient slowing techniques are currently being pursued by multiple groups [17-21], and the production of brighter, slower molecular beams remains an active and complementary area of research [22, 23].

This Rapid Communication presents a cryogenic source capable of producing bright, continuous beams of cold free radicals via laser ablation, thereby realizing the first step towards longer MOT loading times and the continuous accumulation of conservatively trapped dark-state molecules using an intermediate MOT stage. Our source uses helium buffer gas at 2.6 K and produces \(20\) ms duration pulses of molecules at repetition rates up to \(55\) Hz, limited by our ablation laser. To the best of our knowledge, these are the brightest time-averaged beams of free radicals reported from a helium buffer gas source and mark the first demonstration of a continuous beam of cold free radicals.

The heart of our source is a two-stage pulse-tube refrigerator (Cryomech PT420) paired with a closed liquid helium reservoir [27] between the refrigerator’s second-stage and the cooled copper cell (fig. 1a). When cold, the reservoir contains \(\sim 7\) g (1.7 moles) of helium which acts as a large thermal mass (heat capacity \(\sim 16\) J/K) to both dampen temperature oscillations from the refrigerator and allow the source to absorb high thermal loads from the ablation laser with limited heating. A constant source temperature is desirable to ensure reproducible molecular beam properties including flux, forward velocity and rotational state distribution. We note that an equivalent thermal mass using copper alone at 2.6 K is impractical and would require cooling \(\sim 400\) kg of material. However, rare-earth alloy plates have been successfully used to dampen thermal oscillations in a similar manner [28]. While beneficial once cold, the closed helium reservoir leads to longer source cool-down and warm-up times. To counter this increase, our design limits the additional thermal mass to 6 kg of machined aluminum and copper parts while largely replicating the source geometry of ref. [10] (fig. 1b). Our design cools from 295 K to 2.5 K in \(\sim 2\) hours and can warm-up to 280 K in \(\sim 4\) hours, allowing rapid prototyping (fig. 1c). To help reduce warm-up times, we typically apply \(0.5\) W of 808 nm laser light to the cell to increase the liquid helium evaporation rate. At base temperature, the cell

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and refrigerator second-stage are stable to ±5 mK and ±60 mK respectively (fig. 1d). For reference, without the helium reservoir the second-stage temperature stability is typically ±200 mK as the refrigerator pulses [29]. These larger oscillations have been reported to correlate with a ∼ 25 % peak to peak variation in molecular beam flux [10], forcing several experiments to synchronize their repetition rates to the period of the pulse-tube refrigerator to recover reproducible pulses of molecules [2, 13].

This work uses SrF molecules to characterize the source performance by ablating a SrF$_2$ target, mounted inside the cell at 30° relative to the molecular beam axis, using 15 mJ pulses of 532 nm light with 6 ns duration from a Nd:YAG laser. This light is tightly focused onto the surface of the target using a 200 mm focal length lens outside the vacuum chamber and the pulse energy is stable to within 1 %. Cold helium buffer gas enters the cell through a fill line at the rear and exits through a conical face with a 40° half-angle and a 3 mm diameter aperture (fig. 1b). The typical helium buffer gas flow rate is 15 standard cubic centimeters per minute (sccm), equivalent to an in-cell steady-state helium density of 10$^{16}$ cm$^{-3}$ and a Reynolds number of ≈ 60. At this flow rate the vacuum inside the cryogenic source chamber is 10$^{-7}$ Torr, maintained by ∼700 cm$^2$ of cold charcoal cryopump.

Properties of the molecular beam are typically probed on the X$^2\Sigma(v = 0, N = 1)$ to A$^2\Pi_{1/2}(v’ = 0, J’ = 1/2)$ transition at 663 nm using either absorption 20 mm downstream of the cell exit aperture or fluorescence detection 940 mm downstream. The molecular beam forward velocity is ≈ 140 m/s with a FWHM of ≈ 50 m/s. This was measured through the Doppler shift between two fluorescence profiles recorded using probe lasers transverse and counter-propagating to the molecular beam. The measured FWHM transverse velocity spread is 80 m/s, corresponding to a FWHM angular spread of 30°. The rotational temperature of the molecular beam is 1.0(2) K, measured by extracting the relative populations in X$^2\Sigma(v = 0, N = 0 – 4)$ using fluorescence signals from the X$^2\Sigma(v = 0, N = 0 – 4)$ to A$^2\Pi_{1/2}(v’ = 0, J’ = 1/2 – 9/2)$ transitions and calculated branching ratios to account for the varying line strengths [30]. Here molecules cool rotationally to below the cell temperature due to isentropic cooling near the cell aperture [31] and at our rotational temperature ≈ 50 % of the molecules populate the X$^2\Sigma(v = 0, N = 1)$ state. All of these parameters are in good agreement with measurements performed on a source with similar geometry [10].

To extract the number of molecules exiting the source in the X$^2\Sigma(v = 0, N = 1)$ state we use the time-integral of the resonant absorption signal, Doppler broadened absorption cross section [32], and assume a uniform density over the cross sectional area of the molecular beam [10]. At ablation repetition rates of 1–2 Hz, where other helium buffer gas sources typically operate [10, 33, 34], the source produces 10$^{11}$ molecules per steradian per pulse with negligible heating. We note that all reported numbers can vary by ≈ ±50 % depending on the spot ablated on the

FIG. 2. Absorption and cell temperature traces measured over 1.5 seconds of source operation for ablation rates of (a) 10 Hz (blue), (b) 20 Hz (red) (c) and 55 Hz (black). During these measurements the source temperature (d) increased by 40, 80 and 200 mK respectively. The time needed to cool back to 2.64 K was 1 s, 30 s and 70 s for 10, 20 and 55 Hz operation respectively.
target. At ablation repetition rates of 10 and 20 Hz the pulses of molecules are unchanged and the source produces $10^{12}$ and $2 \times 10^{12}$ time-averaged molecules/sr/sec respectively (figs. 2a and 2b). At 55 Hz we consistently observe a $\sim 10\%$ decrease in brightness ($\times$ the time-integrated absorption signal) over the first 2-5 pulses, with negligible change in rotational temperature, and typically produce $5 \times 10^{12}$ molecules/sr/sec (fig. 2c). In-cell absorption measurements show that this initial decrease in brightness is correlated with decreasing in-cell molecular density and the extraction efficiency from the cell remains unchanged at $\sim 50\%$. This decrease in yield is temporary and a 100 ms pause in ablation pulses is sufficient to recover the original yield from the next pulse using the same ablation spot. This behavior is presumably due to heating within the cell, which is measured to increase by 40, 80 and 200 mK over 1.5 seconds of operation at 10, 20 and 55 Hz respectively (fig. 2d). Once the cell reaches thermal equilibrium, we typically measure a temperature increase of $\approx 1\text{ mK/mW}$ of incident power, corresponding to a steady-state cell temperature of 3.5 K at 55 Hz.

The successful production of bright, continuous beams of cold free radicals via ablation at 55 Hz depends critically on the helium buffer gas flow rate through the cell (fig. 3). At flow rates of 2 and 5 sccm, the initial yield per ablation pulse is reduced by factors of 10 and 5 respectively relative to our standard 15 sccm flow rate. We attribute this decrease to insufficient buffer gas density for complete thermalization. At 2 sccm, few molecules are detected in the beam after 250 ms of operation while a flow of 5 sccm is sufficient to consistently produce pulses of molecules and realize $10^{12}$ molecules/sr/sec. Note that the sporadic spikes in fig. 3 visible at flows of 2 and 5 sccm are $\sim 100\mu$s bursts of $10^8-10^9$ molecules and are not optical pickup due to the ablation laser. At flows of 10 sccm we begin to continuously detect molecules exiting the cell with a brightness of $3 \times 10^{12}$ molecules/sr/sec, increasing to $7 \times 10^{12}$ molecules/sr/sec at 20 sccm. At 15 sccm the beam brightness is typically modulated at 55 Hz by $\sim 80\%$ 20 mm downstream of the cell. We project that this modulation will decrease to $\sim 60\%$ a further 1.5 m downstream by convolving the measured molecular pulse temporal and velocity distributions.

The source performance is robust at 55 Hz for helium buffer gas flow rates $\geq 10$ sccm while a flow rate of $\geq 5$ sccm is sufficient to sustain continuous operation at 10 and 20 Hz. Other groups using helium-based cryogenic sources typically use lower buffer gas flow rates, between 1-5 sccm, and several have reported erratic source behavior for ablation repetition rates $> 5$ Hz. We assume these observations are due to insufficient helium flow and, other than the ablation pulse energy, we have found no other parameter that strongly effects beam brightness. Our source performance does not depend critically on the cell geometry; molecular beams with similar brightness were realized when the conical cell exit was replaced with a flat 0.5 mm thick copper plate containing a 3 mm diameter aperture. This lack of dependence on cell geometry is in contrast to observations for a capillary fed cryogenic source. Similar source performance was also realized for an elevated cell temperature of 4 K, after accounting for an increased rotational temperature of $2.0(3)$ K, which reduced the number of molecules in the $X^2\Sigma (v = 0, N = 1)$ state by $\approx 30\%$. This factor of 2 increase in rotational temperature highlights the importance of limiting heating during source operation to ensure reproducible beam properties. This source design has also proven to be straightforward to replicate and a second unit is now operational in our group and produces similar continuous beams of molecules.

As a demonstration of the stable and continuous nature of these molecular beams we produce uninterrupted pulses of SrF molecules at 55 Hz over a 60 second duration using a buffer gas flow rate of 15 sccm (fig. 4). During this time the ablation spot was moved every $\sim 10$ s to restore the decaying ablation yield and we measure a mean brightness of $\sim 3 \times 10^{12}$ molecules/sr/sec along-side a cell temperature increase of 0.6 K. Currently the main limit on beam brightness during continuous operation of our source is the durability of the ablated target. Free radical production methods that ablate metals in the presence a reactant gas (e.g. SF$_6$) have been shown to produce brighter, more reproducible beams, and could potentially work well with our source design at high

![FIG. 3. Absorption versus helium buffer gas flow at a 55 Hz ablation repetition rate. From top to bottom, the helium flow rates were 2, 5, 10, 15 and 20 sccm. At 2 and 5 sccm some ablation pulses produce $\sim 100\mu$s bursts of molecules, these are not detected at higher flow rates. Molecules are continuously detected leaving the source for flow rates $\geq 10$ sccm. These data were recorded in a random order using the same ablation spot on the target and highlight the temporary nature of the decrease in brightness measured over the first 2-5 pulses.](image-url)
The mean brightness measured during this minute was showing (a) absorption and (b) the cell temperature. The brightness of up to 7 cold free radicals via laser ablation with a time-averaged gas source capable of producing continuous beams of with substantially reduced cooling power [37, 38].

1 K and produce colder, slower molecular beams, but also pump on this reservoir to cool the source towards cool-down and warm-up times. In principle, one could source temperature stability at the expense of longer increasing the helium mass would further improve the liquid helium fills ~ 25 % of the closed reservoir and increasing the helium mass would further improve the source temperature stability at the expense of longer cool-down and warm-up times. In principle, one could also pump on this reservoir to cool the source towards 1 K and produce colder, slower molecular beams, but with substantially reduced cooling power [37, 38].

In summary, we have realized a robust cryogenic buffer gas source capable of producing continuous beams of cold free radicals via laser ablation with a time-averaged brightness of up to 7 × 10^{12} molecules/sr/sec in a single rovibrational state. Crucially, the performance of our source does not depend critically on the cell geometry or temperature. This suggests that other groups currently using helium-based sources could immediately adopt our method to realize brighter molecular beams, provided that there is sufficient buffer gas flow and operation times are short to limit heating. These molecular beams represent the first step towards longer MOT loading times to trap larger samples at higher density and are well-suited for continuous beam slowing and cooling techniques such as centrifugal deceleration [39, 40], Zeeman slowing [21], and Zeeman-Sisyphus deceleration [18]. For reference, today’s molecular MOTs load single pulses of molecules over ~ 20 ms. If similar loading times were used for atomic MOTs only ~ 10^5 ~ 10^6 atoms would be trapped [41, 42]. Given that typical molecular MOT lifetimes are short, ~ 100 ms [5], the continuous accumulation of conservatively trapped molecules via an intermediate MOT stage is a particularly promising approach, a method which has been successfully demonstrated with chromium atoms [43]. These advances have the potential to increase the numbers and densities realized in laser-cooled samples of molecules by orders-of-magnitude and provide routine access to the molecule-molecule interactions required for many proposed applications.

Drawings of the machined parts needed to replicate this cryogenic source design and assembly instructions can be provided by contacting the corresponding author.

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