Proposal for designing a two-stage buffer gas cooled beam source

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A novel two-stage helium buffer gas cooled beam source is introduced. The properties of the molecular beams produced from this source are investigated theoretically using the CaF as a test molecule. The gas phase molecules are first produced inside a 3 K helium buffer gas cell by laser ablation and subsequently cooled down to 3 K by collisions with buffer gas atoms. The precooled molecules are then extracted into the 0.5 K helium buffer gas cell where they are cooled further down to 0.5 K by collisions with cold helium atoms. Finally, the cold molecules are extracted out into the high vacuum through the 0.5 K cell exit aperture and form a molecular beam. The mean forward velocity and the flux of the molecular beams are calculated to be 45 m/s and $8 \times 10^{12}$ molecules per pulse respectively, when both cells are operated in the so-called hydrodynamic entrainment regime. The calculations further indicate that the mean forward velocity of the beam can be reduced below 25 m/s whilst maintaining the beam flux over $10^{16}$ molecules per pulse by operating the helium buffer gas cell in the so-called diffusive regime. These calculations are well supported by helium flow-field simulations. Combining this source with moving trap decelerators paves the way for an efficient magneto-optical trapping of the molecules and performing ultrahigh precision measurements.

I. INTRODUCTION

The collisions of the cold buffer gas atoms with warm molecules cool their translational and rotational temperatures [4,3]. Thus, helium buffer gas cooled beams are prerequisite for the laser cooling of the molecules [4,5]. Further, the number of photon absorption and emission cycles required for the laser cooling of atoms and molecules decrease with their forward velocity and hence the laser cooling efficiency of the molecules increases with decrease in their forward velocity [4,8]. In addition to this, the slower beams are also beneficial for an efficient deceleration of the atoms and molecules by moving trap decelerators [3,11]. This is because the reduced initial trap velocity facilitates in producing deeper traps. This results in increased velocity acceptance and hence the overall efficiency of the decelerator. Furthermore, the intense and slow beams of buffer gas cooled molecules increase the product $\sqrt{N\tau}$, where $N$ is the total number of molecules that participate in the experiments and $\tau$ is the measurement time. Consequently, they enhance the statistical sensitivity of the ultrahigh precision measurements [12-15].

To extract the maximum number of molecules from the buffer gas cell into the molecular beam, the cell is operated in the so-called hydrodynamic entrainment regime [2,17]. In this regime, the collisions of the buffer gas atoms at the cell exit boost the mean velocity of the molecules as high as the mean forward velocity ($v_f = \sqrt{\frac{2k_BT}{m}}$) of the helium atoms. This boosting can however be suppressed partially by collisions of the molecules with helium atoms inside a low helium density slowing cell attached to the main helium buffer gas cell [4,18]. But due to the accelerated diffusion losses inside the slowing cell, the fraction of the molecules extracted from the cell into the molecular beam is reduced by several orders of magnitude [17,18].

This paper introduces a new approach for reducing the forward velocity of the buffer cooled beams without compromising the flux of molecules. The approach allows to operate the beam source at 0.5 K temperature with repetition rates over 20 Hz. At this temperature, the helium atoms acquire a mean forward velocity of 45 m/s. Thus, allowing to reduce the mean forward velocity of the buffer gas cooled molecular beams below 45 m/s even though the cell is operated in hydrodynamic entrainment regime. Using the $^4$He as buffer gas, a density of $10^{15}$ cm$^{-3}$ can be achieved at 0.5 K cell temperature [19]. While this density is sufficient for the buffer gas cooling of the atoms and molecules, the heat load on the buffer gas cell from the vaporization techniques such as laser ablation usually exceeds the cooling power of the evaporation refrigerators [3,4]. To implement this new approach, avoiding the heat load on the 0.5 K stage of the helium buffer gas cell from the laser ablation is therefore prerequisite. This can be achieved by using a novel two stage helium buffer gas cooling technique introduced here.

The first stage helium buffer gas cell is attached to the second stage of the cryogenic refrigerator and cooled below 3 K [3,20]. On the other hand, the second stage buffer gas cell is attached to the $^3$He bath and cooled down to 0.5 K by evaporative cooling [3,4]. Due to the significantly higher vapor pressure of the $^3$He compared to $^4$He at low temperatures [21], a higher cooling power can be achieved by pumping on a $^3$He bath at 0.5 K [22]. The molecules are first vaporized and precooled down to 3 K inside the first stage helium buffer gas cell [23]. The precooled molecules are then extracted into the second stage where they are further cooled down to 0.5 K by collisions with buffer gas atoms. The heat load on the 0.5 K helium buffer gas cell is therefore reduced well.
below the cooling power of the $^3$He cryogenic refrigerator at 0.5 K [21].

The technical details of this novel two stage buffer gas cooled beam source are presented. The properties of the molecular beams produced from this source are investigated theoretically using the CaF as a test molecule. The relevant equations are simplified and can be utilized straightforwardly for investigating the properties of buffer gas cooled beams of any diatomic or small polyatomic molecule [24, 25, 26]. The calculations demonstrated that a two stage buffer gas cooled beam source produces two times slower molecular beams with several orders of magnitude higher flux compared to the current state of art single stage buffer gas cooled beam sources [18]. These calculations are well supported by helium flow-field simulations.

Combining this source with the moving trap decelerators will allow for an efficient magneto-optical trapping of the molecules [4, 7]. Thus, making it feasible to laser cool a large number ($N > 10^6$) of the CaF molecules below 100 µK and paves the way for quantum simulations of lattice spin models [26]. Further, the two stage buffer gas cooled beams can also improve the current limit of the measurement of the electric dipole moment (EDM) of the electron [12, 13]. This is because at 0.5 K temperatures only ground and first exited rotational states of the ThO or YbF molecules are populated. Using resonant microwave fields and optical pumping, these molecules can be efficiently transferred to state in which the EDM measurement of the CaF molecules below 3 K can be estimated from [14, 15].

The initial temperature of the CaF is assumed to be 1000 K. Using the hard sphere collision model, the number of collisions required to thermalize the CaF molecules with 3 K $^4$He buffer gas can be estimated from Equation 1 [2].

$$T_N = T + (T_1 - T) \exp \left( \frac{-N}{\kappa} \right)$$

(1)

Where $T_N$ is the temperature of the molecules after $N$ collisions with buffer gas atoms, $T$ is the buffer gas temperature, $T_1$ is the initial temperature of the molecules, $\kappa = \frac{(M+m)^2}{2Mm}$, and $M$ and $m$ are the masses of the molecule and buffer gas atoms respectively. For thermalizing the CaF molecules with 3 K $^4$He buffer gas atoms only 100 collisions are sufficient.

The length ($l$) required to thermalize the molecules with the buffer gas is related to the number of collisions ($N$) and the mean free path ($\lambda = 1/(\sqrt{2\pi}\sigma n)$), by the Equation 2.

$$l = N\lambda$$

$$= N/(\sqrt{2\pi}\sigma n)$$

Where $\sigma$ is the collision cross-section of molecules with the buffer gas atoms and $n$ is the buffer gas density.

In steady state, the buffer gas density inside the cell is related to the buffer gas flow rate $f$ by the Equation 3 [2].

$$n \approx \frac{4f}{Av}$$

(3)

Where $A$ is the area of cell exit aperture. The $\bar{v}$ is the mean thermal velocity of the buffer gas atoms and is given by the following equation.

$$\bar{v} = \sqrt{\frac{8k_B T}{\pi m}}$$

(4)

The $k_B$ is the Boltzmann constant. Using Equation 3 and Equation 4, the $^4$He buffer gas density $n_{He}$ inside the first stage of the buffer gas can be estimated from Equation 5.

$$n_{He} \approx 9 \times 10^{14} \left( \frac{f}{1 \text{SCCM}} \right) \left( \frac{16 \text{mm}^2}{A} \right) \left( \frac{3 \text{K}}{T} \right)^{1/2} \left( \frac{m}{6.6 \times 10^{-27} \text{Kg}} \right)^{1/2} \text{cm}^{-3}$$

(5)

Where SCCM stands for standard cubic centimeter per minute. For the buffer gas flow of 10 SCCM and the cell with a square shape exit aperture of 4 mm diameter, the calculated buffer gas density inside the cell is $\approx 9 \times 10^{13} \text{ cm}^{-3}$. Using Equation 2 and Equation 5, the thermalization length of the molecules with the $^4$He buffer gas can be given by the Equation 6.

$$l \approx 8 \times \left( \frac{N}{100} \right)^{10^{-14} \text{ cm}^2 \text{mm}^2} \left( \frac{1 \text{SCCM}}{f} \right) \left( \frac{A}{16 \text{mm}^2} \right) \left( \frac{3 \text{K}}{T} \right)^{1/2} \left( \frac{6.6 \times 10^{-27} \text{Kg}}{m} \right)^{1/2} \text{cm}^{-3}$$

(6)
The molecules cooled down to 0.5 K by collisions with helium atoms through the 8 mm diameter square aperture and form a molecular beam. The helium density inside the 0.5 K buffer gas cell can be controlled by helium vent lines. A pressure of 10−6 mbar can be maintained inside the vacuum chamber using a charcoal cryogenic sorption pump [20].

Here \( \sigma_{\text{CaF-He}} \) is assumed to be \( 10^{-14} \text{ cm}^2 \) [2]. As expected the thermalization length decreases with increase in flow rate. For the helium flow rate of 12 SCCM, the length \( l \) required to thermalize the molecules with the 3 K helium buffer gas is 0.7 cm. Thus, a few cm long helium buffer gas cell should be sufficient for the first stage buffer gas cooling of molecules. The number of the molecules extracted from the buffer gas cell into the molecular beam are characterized by the cell extraction parameter \( \gamma_e = \tau_{\text{diff}}/\tau_{\text{pump}} \), where \( \tau_{\text{diff}} \) is the diffusion time of the molecules to the cold cell walls and \( \tau_{\text{pump}} \) is the typical time spent by the molecules inside the buffer gas cell. For the given cell and buffer gas parameters, the \( \gamma_e \) can be estimated from the Equation 7 [2].

\[
\gamma_e = \frac{\sigma f}{Lv} \approx 1.1 \times \left( \frac{\sigma}{10^{-14} \text{ cm}^2} \right) \left( \frac{f}{12 \text{ SCCM}} \right) \times \left( \frac{0.8 \text{ mm}}{L} \right) \left( \frac{3 \text{ K}}{T} \right)^{1/2} \left( \frac{m}{6.6 \times 10^{-27} \text{ Kg}} \right)^{1/2}
\]

(7)

For the first stage helium buffer gas cell length \( L \) of 4 cm, and buffer gas flow rate of 12 SCCM, \( \gamma_e \approx 1.1 \).

The number of the CaF molecules as high as \( 5 \times 10^{13} \) can be produced in each ablation pulse [23]. With optimized buffer gas flow and cell geometry [20], the diffusion loss of the molecules inside the buffer gas cell can be minimized and hence over 40% of the buffer gas cooled molecules from the buffer gas cell can be extracted from the cell into the molecular beam [17]. With this extraction efficiency, the number of the buffer gas cooled molecules transmitted through the 3 K buffer gas cell exit aperture are \( \approx 2 \times 10^{13} \) per pulse.

Owing to their larger mass, the mean thermal velocity of the molecules inside the buffer gas cell is smaller than that of the buffer gas atoms. At the cell exit, the collisions of the buffer gas atoms boost the mean forward velocity of the molecules. The number of collision between the molecules and buffer gas atoms can be estimated from Equation 8 [2].

\[
\frac{d}{L} \approx \frac{R_e}{2}
\]

(8)

Where \( d \) is the diameter of cell exit aperture and \( R_e \) is the Reynolds number. Using Equation 2, Equation 3, and Equation 8, the Reynolds number can be given by the following equation.

\[
R_e \approx \frac{10 \sigma f d}{A \nu} \approx 8.6 \times \left( \frac{\sigma}{10^{-14} \text{ cm}^2} \right) \left( \frac{f}{1 \text{ SCCM}} \right) \left( 4 \text{ mm} \right) \left( \frac{3 \text{ K}}{T} \right)^{1/2} \left( \frac{m}{6.6 \times 10^{-27} \text{ Kg}} \right)^{1/2}
\]

(9)

The Reynolds number increases with buffer gas flow rate. For the helium flow rate of 12 SCCM, the Reynolds number is about 100. At the cell exit, the number of collisions of molecules with buffer gas are \( \approx 50 \). In each collision with buffer gas atom, the change in the forward momentum of the molecules is \( \approx \mu \). Thus, the corresponding change in the forward velocity \( (R_e \mu / 2M) \) of the molecules can be estimated from Equation 10.

\[
\Delta v_{\text{mol, f}} \approx 40 \times \left( \frac{\sigma}{10^{-14} \text{ cm}^2} \right) \left( \frac{f}{1 \text{ SCCM}} \right) \left( \frac{4 \text{ mm}}{d} \right) \left( \frac{m}{4 \text{ amu}} \right) \left( \frac{59 \text{ amu}}{M} \right) \text{ m/s}
\]

(10)

The number of collisions between the molecules and the buffer gas atoms increase with buffer gas flow rate. At 12 SCCM helium rate, the forward velocity of the molecules is expected to be boosted to the forward velocity \( (\sqrt{2 \mu n T} \text{ m/s}) \) of the buffer gas atoms, i.e. 110 m/s.

Further, the transverse velocity of the buffer gas inside the cell \( (v_{\text{cell}}) \) can be approximated by Equation 11 [20].

\[
v_{\text{cell}} \approx \bar{v} \times \left( \frac{d}{d_{\text{cell}}} \right)^2 \approx 0.8 \times \left( \frac{50 \text{ mm}}{d_{\text{cell}}} \right)^2 \left( \frac{m}{6.6 \times 10^{-27} \text{ Kg}} \right)^{1/2} \text{ m/s}
\]

(11)

Where \( d_{\text{cell}} \) is the cell diameter. For the helium buffer gas cell with 50 nm diameter, \( v_{\text{cell}} \approx 0.8 \text{ m/s} \). The change in the transverse velocity \( (R_e \mu v_{\text{cell}} / 2M) \) of the molecules due to the collisions with buffer gas atoms at the cell exit.

FIG. 1. A schematic diagram of the two stage helium buffer gas cooled molecular beam source as discussed in the text. The molecules are produced by laser ablation and subsequently cooled down to 3 K in a 4 cm long and 5 cm diameter first stage helium buffer gas cell. The molecules then extracted out through a square exit aperture of 5 mm diameter and enter into a 3 cm long and 5 cm diameter 0.5 K helium buffer gas cell through a square shape entrance aperture of 5 mm diameter. The molecules cooled down to 0.5 K by collisions with helium atoms. The cold molecules then extracted out into the high vacuum through the 8 mm diameter square aperture and form a molecular beam. The helium density inside the 0.5 K buffer gas cell can be controlled by helium vent lines.
can be estimated from Equation 12
\[ \Delta v_{\text{mol}, \perp} \approx 0.2 \times \left( \frac{1 \text{ SCCM}}{10^{-15} \text{ cm}^2} \right) \left( \frac{f}{1 \text{ mol}} \right) \left( \frac{d}{4 \text{ mm}} \right) \times \left( \frac{50 \text{ mm}}{d_{\text{cell}}} \right)^2 \left( \frac{m}{4 \text{ amu}} \right) \left( \frac{59 \text{ amu}}{M} \right) \text{ m/s} \] (12)

The change in the transverse velocity of the molecules also increases with increase in the buffer gas flow rate. At 12 SCCM, the change in transverse velocity is around 2.4 m/s. Assuming the initial beam spread of 4 mm×4 mm (size of the exit aperture), the calculated transverse molecular beam spread 1 cm away from the first stage buffer gas cell exit is 4.4 mm×4.4 mm [50]. Thus, using an entrance aperture of 5 mm×5 mm, all the precooled CaF molecules transmitted from the exit aperture of the first stage buffer gas cell can be extracted into the second stage buffer gas cell.

III. SECOND STAGE BUFFER GAS COOLING OF THE MOLECULES

From Equation 1, the number of collisions of the CaF molecules with helium buffer gas atoms required to cool them from 3 K down to 0.5 K are 50. For a helium flow rate of 5 SCCM and a square shape cell exit aperture of 8 mm diameter, the thermalization length of the molecules with the 0.5 K helium buffer gas calculated using Equation 6 is 1.5 cm. For this flow, the helium buffer gas density estimated from Equation 5 is \( \approx 10^{15} \text{ cm}^{-3} \).

With a helium buffer gas length of 3 cm, the calculated extraction efficiency of \( \epsilon \approx 1.5 \). At this extraction efficiency over 40% of the buffer gas cooled molecules out of 2×10^{13} can be extracted from the cell into the beam [17]. Thus, the flux of produced CaF molecules per pulse is \( \approx 8 \times 10^{12} \).

In addition to the cell temperature, the mean forward velocity of the molecules in the beam will also depend upon the number of the collisions of molecules with the buffer gas atoms at the cell exit aperture. For the second stage helium buffer gas cell parameters, the estimated Reynolds number from Equation 9 is 50. The number of collisions of molecules with buffer gas are \( \approx 25 \). From the Equation 10, it is clear that these collisions are sufficient to boost the forward velocity of the molecular beam to the forward velocity (\( v_f = \sqrt{\frac{2k_B T}{m}} \)) of the buffer gas atoms. At 0.5 K, the mean forward velocity of the CaF after the exit aperture of the second stage cell is expected to be 45 m/s.

Thus, the two-stage buffer gas cooled beam source introduced here is theoretically demonstrated to produce much slower beams with 8×10^{4} times higher molecular beam flux compared to the current state of art buffer gas cooled beam source [15]. Further slowing of these beams with broad band lasers or moving trap decelerators is beneficial for more efficient magneto-optical trapping and laser cooling of the molecules [3] [17]. Furthermore, the cold, intense, and slow beams produced from the two stage buffer gas cooled beams source can also increase the statistical sensitivity (\( \sqrt{N_T} \)) of the precision measurements by a factor of 10 [12] [13], as discussed before.

The forward velocity of the molecular beam can be further reduced by minimizing the number of collisions between the molecules and helium atoms at the cell exit. This can be achieved by decreasing the helium flow rate into the 0.5 K buffer gas cell. With a helium flow rate of 0.7 SCCM, the required cell length to cool the CaF from 3 K down to 0.5 K is 9.5 cm. At this helium flow rate, the cell extraction parameter \( \epsilon \approx 0.2 \). At this flow, the cell will operate in the diffusive regime [17]. The mean forward velocity (\( v_{\text{fm}} \)) of the molecules can be estimated from Equation 13 [18].

\[ v_{\text{fm}} = \sqrt{\frac{2k_B T}{M}} \] (13)

For the CaF molecules cooled down to 0.5 K, \( v_{\text{fm}} = 10 \text{ m/s} \). Also, considering the collisions of buffer gas atom with molecules at the cell exit, the change in the mean velocity of the molecules calculated at a helium flow rate of 0.7 SCCM using Equation 10 is 14 m/s. Thus, the mean velocity of the molecules in this regime is \( \approx 25 \text{ m/s} \).

Further, the fraction of the molecules extracted from the helium buffer gas cell into the molecular beam can be estimated from the Equation 14 [17].

\[ f_{\text{ext}} \approx \frac{A_{\text{aperture}}}{\pi L^2} \] (14)

For \( L = 9.5 \text{ cm} \), \( f_{\text{ext}} \approx 2 \times 10^{-3} \). The number of the molecules extracted from the cell into the beam are \( \approx 10^{16} \) per pulse. This shows that changing the cell operating regime from hydrodynamic entrainment to diffusive, the beam forward velocity is decreased only by a factor of 2. On the other hand, the flux of the molecules reduced by two orders of magnitude. Thus, operating the both cells in hydrodynamic entrainment regime is advantageous for many applications including ultrahigh precision measurements.

IV. SIMULATION OF HELIUM BEHAVIOR INSIDE THE BUFFER GAS CELLS

To further support the calculations performed in Section II and Section III the helium flow-fields inside the buffer-gas cells are simulated using a finite-element solver, COMSOL Multiphysics with laminar flow interface [31], treating helium as an ideal gas in the temperature range 3-0.5 K. For 12 SCCM of helium flow and a pressure of 10^{-6} mbar outside the cell exit, the resulting flow fields are shown in Figure 2 as slices through the cylindrically-symmetric cell volume, i.e., in \( r, z \) space. From the simulations, the helium pressures inside the cells and the velocity of helium at the exit of the 0.5 K cell are extracted. These extracted values are shown in Table 1 and are in a good qualitative agreement with the calculations.
gas cell suppresses the heat load on the second cooling stage and hence facilitates the operation of the second stage buffer gas cell at 0.5 K with the repetition rates over 20 Hz. At this temperature, the mean forward velocity and the flux of the CaF beam are calculated to be 45 m/s and 8×10¹² molecules per pulse respectively, when both buffer gas cells are operated in hydrodynamic entrainment regime. The calculations further indicate that the mean forward velocity of the molecular beam can be decreased below 25 m/s while maintaining a flux of 10¹⁰ molecules per pulse by operating the helium buffer gas cell in the diffusive regime. The cold, intense, and slow beams produced from the two stage buffer gas cooled beam source can be brought near standstill by moving trap decelerators. The decelerated molecules can be utilized for efficient magneto-optical trapping of the molecules and ultrahigh precision measurements.

VI. APPENDIX

This section calculates the heat load on the first and second stages of the cryogenic refrigerator and ³He bath. As shown below, the heat load on the each stage of the refrigerator is well below the cooling power of the corresponding stage. Thus, making it feasible to design a two stage buffer gas cooled beam source with the parameters considered here.

A. Calculated mass flow rate for 10 mW cooling power of the helium bath

At 0.5 K, the ³He vapor pressure = 0.2 mbar [21]. The latent heat of vaporization of the ³He is \( L_v = 8.5 \text{ J/gm} \) at 3.2 K temperature and 1 bar pressure. One gram of ³He contains 2 × 10²³ atoms. Thus, the latent of vaporization \( L_v = 4.25 \times 10^{-23} \text{ J/atom} \).

The number of atoms \( N \) contained by 1 L of ³He at \( T = 0.5 \text{ K} \) and \( P = 0.2 \text{ mbar} \) can be calculated from the ideal gas law \( PV = Nk_B T \). \( N = (0.2 \text{ mbar})(1 \text{ L})/((1.38 \times 10^{-23} \text{ J/K})(0.5 \text{ K})) = (20 \text{ Pa})(0.001 \text{ m}^3)/((1.38 \times 10^{-23} \text{ J/K})(0.5 \text{ K})) = 30 \times 10^{20}. \)

Thus, at \( T = 0.5 \text{ K} \) and \( P = 0.2 \text{ mbar} \), \( L_v = (4.25 \times 10^{-23} \text{ J/atom})(30 \times 10^{20} \text{ atom/L}) = 0.13 \text{ J/L}. \)

To achieve a cooling power of 10 mW, the boil off rate = (10 mW/0.13 J/L) = 0.078 L/s. To operate the refrigerator continuously, the liquid ³He flow into the helium bath must be equal to the boil off rate. Thus,
the required flow through the helium condenser (3 K, 1 bar) calculated using ideal gas law = (0.2 mbar/1 bar) (3 K/0.5 K) (0.078 L/s) = 9.4 × 10^-5 L/s.

At \( T = 3 \text{ K} \) and \( P = 1 \text{ bar} \), this corresponds to a gas flow of \( N = PV/kT = (100000 \text{ Pa})(9.4 \times 10^{-5} \text{ L/s})(0.001 \text{ m}^3/\text{L})/((1.38 \times 10^{-23} \text{ J/K}) (3 \text{ K})) = 2.3 \times 10^{20} \text{ atom/s} = (2.3 \times 10^{20} \text{ atom/s})(1 \text{ gm}/2 \times 10^{23} \text{ atom}) = 1.2 \text{ mg/s}.

B. Calculated heat load on the first stage of the refrigerator

At the first stage both \(^3\text{He}\) (evaporated from the helium bath) and \(^4\text{He}\) (used for helium buffer gas cooling) are required to be cooled down from 300 K to 30 K. The \(^3\text{He}\) mass flow rate = 1.2 mg/s. The \(^4\text{He}\) flow rate = 20 SCCM (over estimated) = (20 × 4.5 × 10^17)(4 gm/s)/(6.023 × 10^23) = 0.06 mg/s.

The total heat load on the first stage of the cryogenic refrigerator = ((1.2 + 0.06) mg/s)(5.2 J/(gm K)) (270 K) = 1.8 W. This load is much smaller compared to the targeted cooling power (10 mW) at the second stage.

C. Calculated heat load on the second stage of the refrigerator

At the second stage, the \(^3\text{He}\) required to cool from 30 to 3 K and convert from gas to liquid, the \(^4\text{He}\) needs to be cooled from 30 to 3 K, and molecules are vaporized by 10 mJ laser pulse with 20 Hz repetition rate by laser ablation in the first stage of the buffer gas cell attached to the second stage.

The total load on the second stage = \((1.2 \text{ mg/s})(5.2 \text{ J/(gm K)})(27 \text{ K}) + (1.2 \text{ mg/s})(8.5 \text{ J/gm}) + (0.06 \text{ mg/s})(5.2 \text{ J/(gm K)})(27 \text{ K}) + (10 \text{ mJ})(20 \text{ pulse/s}) = 0.4 \text{ W}\). This heat load is significantly smaller compared to the second stage cooling power power (2 W at 4.2 K) of the cryogenic refrigerator.

D. Calculated heat load on the 0.5 K stage of the buffer gas cell

The 20 SCCM flow from the first stage of the buffer gas cell required to be cooled down from 3 K to 0.5 K on the second stage of the buffer gas cell attached to the helium bath. Thus, the load on the 0.5 K helium bath = (0.06 mg/s)(5.2 J/(gm K))(2.5 K) = 0.8 mW.

Further, the heat load from cooling of the 2 \times 10^{13} CaF molecules from 3 K to 0.5 K with 20 Hz repetition rate is below 0.1 mW [32].

Thus, the total heat load on the 0.5 K stage is much smaller compared to the targeted cooling power (10 mW) of the helium bath.

[1] Jonathan D. Weinstein, Robert deCarvalho, Thierry Guillet, Bretislav Friedrich, and John M. Doyle, “Magnetic trapping of calcium monohydride molecules at millikelvin temperatures,” Nature 395, 148–150 (1998).

[2] N. R. Hutzler, H.-I. Lu, and J. M. Doyle, “The buffer gas beam: An intense, cold, and slow source for atoms and molecules,” Chem. Rev. 112, 4803 (2012).

[3] Vijay Singh, Kyle S. Hardman, Naïma TARIG, Mei-Ju Lu, Aja Ellis, Muir J. Morrison, and Jonathan D. Weinstein, “Chemical reactions of atomic lithium and molecular calcium monohydride at 1 K,” Phys. Rev. Lett. 108, 203201 (2012).

[4] Loïc Anderegg, Benjamin L. Augenbraun, Eunmi Chae, Boerge Hemmerling, Nicholas R. Hutzler, Aakash Ravi, Alejandra Collopy, Jun Ye, Wolfgang Ketterle, and John M. Doyle, “Radio frequency magneto-optical trapping of CaF with high density,” Phys. Rev. Lett. 119, 103201 (2017).

[5] E. S. Shim, J. F. Barry, and D. DeMille, “Laser cooling of a diatomic molecule,” Nature 467, 820–823 (2010).

[6] Ivan Kozyr’ev, Louis Baum, Kyle Matsuda, Benjamin L. Augenbraun, Loïc Anderegg, Alexander P. Sedlack, and John M. Doyle, “Sisyphus laser cooling of a polyatomic molecule,” Phys. Rev. Lett. 118, 173201 (2017).

[7] H. J. Williams, L. Caldwell, N. J. Fitch, S. Truppe, J. Rodewald, E. A. Hinds, B. E. Sauer, and M. R. Tarbutt, “Magnetic trapping and coherent control of laser-cooled molecules,” Phys. Rev. Lett. 120, 163201 (2018).

[8] V. Zhelyazkova, A. Courniol, T. E. Wall, A. Matsushima, J. J. Hudson, E. A. Hinds, M. R. Tarbutt, and B. E. Sauer, “Laser cooling and slowing of CaF molecules,” Phys. Rev. A 89, 053416 (2014).

[9] Etay Lavert-Ofir, Sasha Gersten, Alon B Henson, Itamar Shani, Liron David, Julia Narevicius, and Edvardas Narevicius, “A moving magnetic trap decelerator: a new source of cold atoms and molecules,” New J. Phys. 13, 103030 (2011).

[10] Lewis A. McDard, Arin Mizouri, Paul A. Walker, Vijay Singh, Ulrich Krohn, Ed A. Hinds, and David Carty, “A moving-trap zeeman decelerator,” submitted (2018).

[11] Marina Quintero-Pérez, Paul Jansen, Thomas E. Wall, Joost E. van den Berg, Steven Hoekstra, and Hendrick L. Bethlem, “Static trapping of polar molecules in a traveling wave decelerator,” Phys. Rev. Lett. 110, 133003 (2013).

[12] J. J. Hudson, D. M. Kara, I. J. Smallman, B. E. Sauer, M. R. Tarbutt, and E. A. Hinds, “Improved measurement of the shape of the electron,” Nature 473, 493–496 (2011).

[13] ACME Collaboration, J. Baron, W. C. Campbell, D. DeMille, J. M. Doyle, G. Gabrielse, Y. V. Gurevich, P. W.
Hess, N. R. Hutzler, E. Kirilov, I. Kozyryev, B. R. O’Leary, C. D. Panda, E. S. Petrik, B. Spaun, A. C. Vutha, and A. D. West, “Order of magnitude smaller limit on the electric dipole moment of the electron,” Science 343, 269–272 (2014).

[14] Martin Quack, “On the measurement of the parity violating energy difference between enantiomers,” Chem. Phys. Lett. 132, 147 – 153 (1986).

[15] S K Tokunaga, R J Hendricks, M R Tarbutt, and B Darquié, “High-resolution mid-infrared spectroscopy of buffer-gas-cooled methyltrioxorhenium molecules,” New J. Phys. 19, 053006 (2017).

[16] Ivan Kozyryev and Nicholas R. Hutzler, “Precision measurement of time-reversal symmetry violation with laser-cooled polyatomic molecules,” Phys. Rev. Lett. 119, 133002 (2017).

[17] David Patterson and John M. Doyle, “Bright, guided molecular beam with hydrodynamic enhancement,” J. Chem. Phys. 126, 154307 (2007).

[18] Hsin-I Lu, Julia Rasmussen, Matthew J. Wright, Dave Patterson, and John M. Doyle, “A cold and slow molecular beam,” Phys. Chem. Chem. Phys. 13, 18986–18990 (2011).

[19] Bretislav Friedrich, Robert deCarvalho, Jinha Kim, David Patterson, Jonathan D. Weinstein, and John M. Doyle, “Towards magnetic trapping of molecules,” J. Chem. Soc., Faraday Trans. 94, 1783–1791 (1998).

[20] Vijay Singh, Amit K. Samanta, Niels Roth, Daniel Gusa, Tim Ossenbrüggen, Igor Rubinsky, Daniel A. Horke, and Jochen Küpper, “Optimized cell geometry for buffer-gas-cooled molecular-beam sources,” Phys. Rev. A 97, 032704 (2018).

[21] Y.H. Huang and G.B. Chen, “A practical vapor pressure equation for helium-3 from 0.01k to the critical point,” Cryogenics 46, 833 – 839 (2006).

[22] From the relation, \( \frac{dP}{dT} = \frac{LP}{RT^2} \), the cooling power of a evaporation refrigerator is proportional to the vapor pressure (P). To achieve higher cooling power at low temperatures, a \(^3\)He evaporation refrigerator is preferred over that \(^4\)He. Further, the evaporation refrigerators are operated in closed cycles and hence there is no \(^4\)He consumption. This brings down the operational cost of a \(^3\)He refrigerator similar to that of a \(^4\)He refrigerator. Currently, \(^4\)He refrigerators are operating at the Harvard University and the University of the Nevada Reno (UNR). The technical challenges of developing such refrigerators has already been overcome. The author played a leading role in development of the \(^4\)He refrigerator at the UNR [3].

[23] The second stage of the cryogenic refrigerator (CRYOMECH:PT420) has a cooling power of 2 W at 4.2 K (http://www.cryomech.com/cryorefrigerators/pulse-tube/). As shown in the Section VI, the total heat load on the second stage of the cryogenic refrigerator is much smaller than this cooling power.

[24] As shown in the Section VI, the calculated heat load on the \(^3\)He cryogenic refrigerator stays below 1 mW, when the beam source is operated with a repetition rate of 20 Hz.

[25] Julia Piskorski, David Patterson, Sandra Eibenberger, and John M. Doyle, “Cooling, spectroscopy and non-sticking of trans-stilbene and nile red,” ChemPhysChem 15, 3800–3804 (2014).

[26] A. Micheli, G. K. Brennen, and P. Zoller, “A toolbox for lattice-spin models with polar molecules,” Nat. Phys. 2, 341–347 (2006).

[27] M R Tarbutt, B E Sauer, J J Hudson, and E A Hinds, “Design for a fountain of YbF molecules to measure the electron’s electric dipole moment,” New J. Phys. 15, 053034 (2013).

[28] Kenneth Maussang, Dima Egorov, Joel S. Helton, Scott V. Nguyen, and John M. Doyle, “Zeeman relaxation of CaF in low-temperature collisions with helium,” Phys. Rev. Lett. 94, 123002 (2005).

[29] Nicholas R. Hutzler, Maxwell W. Parsons, Yulia V. Gurevich, Paul W. Hess, Elizabeth Petrik, Ben Spaun, Amar C. Vutha, David DeMille, Gerald Gabrielse, and John M. Doyle, “A cryogenic beam of refractory, chemically reactive molecules with expansion cooling,” Phys. Chem. Chem. Phys. 13, 18976–18985 (2011).

[30] The time taken by the beam in traveling 1 cm is 0.09 ms. The transverse distance traveled by the beam during this time is ± 0.2 mm.

[31] COMSOL Multiphysics v. 5.3, http://www.comsol.com. COMSOL AB, Stockholm, Sweden.

[32] This heat load is calculated using the literature value of the CaF\(_2\) specific heat of 854 J Kg\(^{-1}\) K\(^{-1}\) (https://www.crystran.co.uk/optical-materials/calcium-fluoride-caf2).