Testing the validity of supersonic molecular beam modelling in the high throughput range (p_0d up to 2000 mbar · cm) using an Even-Lavie valve

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Supersonic molecular beams are used in several scientific fields ranging from surface science to spectroscopy of molecules, nanoparticles and even viruses. It is therefore of interest to understand the fundamental properties of the expansion and resulting beam. Here we present a study of the change in most probable velocity with reservoir pressure, using a so called Even-Lavie pulsed source with a 200 µm diameter nozzle. Experiments were carried out in the throughput range p_0d = 400-2000 mbar · cm, where p_0 is the reservoir stagnation pressure and d is the nozzle diameter. Experiments were done on both neutral and metastable helium beams. A two-detector measurement setup was used to account for valve opening delay. The measurements are modelled with theory which has hitherto only been applied to a p_0d regime up to 190 mbar · cm, showing good agreement.

I. INTRODUCTION

Supersonic helium beams are used for a wide range of applications. Uses for neutral, ground state helium beams include microscopy[1–3], surface science[4–8], and seeded beam experiments[9,10]. Metastable helium beams are used among others in spectroscopy and material characterisation experiments[11–13], for lithography[14–17], for fundamental atom physics experiments on the lifetime of atomic states[18,19], for Bose-Einstein condensate generation and for other fundamental studies related to beam properties[20,21].

Supersonic beams are made by an adiabatic expansion of the gas from a high pressure reservoir through a nozzle with a diameter larger than the mean free path of the particles in the gas reservoir. This leads to a high intensity beam with a velocity distribution which is significantly narrower than its effusive beam counterpart[22] (where the diameter of the nozzle is smaller than the mean free path). Supersonic molecular beam sources come in continuous or pulsed varieties. A pulsed beam enables an increase in intensity over the duration of the beam pulse compared to a continuous one for a given pumping systems[23–25]. In the experiments presented here, we use a so called Even-Lavie valve[26,27] pulsed source. The Even-Lavie valve is commercially available and is used to generate molecular beams for a wide range of applications including the prevously mentioned fields of study, see also references[28–31]. The behaviour and characteristics of supersonic helium beam expansions have been investigated in a range of experiments and theoretical works.[32–35] A summary is included in a review of neutral helium microscopy by Palau et al.[36]. Of particular interest here is work related to measurements and prediction of the most probable velocity of the helium atoms in the beam. The effect of varying nozzle size and source stagnation pressure has so far to the best of our knowledge only been measured for the p_0d (source stagnation pressure · nozzle diameter) regime up to 190 mbar·cm.[37] Here we present measurements of the most probable velocity of the helium beam from the Even-Lavie valve up to 2000 mbar·cm and compare them to theoretical modelling first presented by Reisinger et al.[38] The theory has been used in several other, for example in investigations of the centre-line beam intensity.[35]

II. EXPERIMENTAL SETUP

Experiments were performed with a room temperature Even-Lavie pulsed valve source purchased from the company *LAMID Multidisciplinary Instruments Development*. The source has a 200 µm diameter trumpet shaped expansion nozzle[33]. Experiments were done using ground state and metastable helium beams. The experiments performed with metastable helium atoms use the Dielectric Barrier Discharge unit (DBD) mounted in the front of the nozzle.[27] The DBD unit is explained in greater detail by Luria et al.[34]. A 3D diagram of the Even-Lavie pulsed valve with the DBD unit as well as a cross section of the DBD unit is shown in figure 1. For the experiments presented here, the original preioniser "electron brush" from the supplier was replaced by an ionising filament supplied with 12 V and a heating current set above the level required for ignition. The filament is mounted in front of the nozzle opening. For the neutral helium measurements, the source was mounted on the molecular beam apparatus at the University of Bergen known familiarly as MAGIE[39]. For the metastable helium measurements, the source was mounted in another setup where the time of flight (TOF) could be measured at two different locations along the beamline. In both setups, the central part of the beam was selected by letting the beam pass through a 3 mm skimmer (Beam Dynamics, model 2), located 492 mm (neutral) and 374 mm (metastable) downstream from the nozzle opening. The MAGIE source chamber has dimensions 492×600×510 mm^3 (V=0.15 m^3) and is pumped by a 3200 l/s turbomolecular pump (Leybold MAG W 3200), backed by a 65 m^3/h rotary vane pump (Leybold TRIVAC D65B) and two single roots blowers of 505 m^3/h (Leybold RUVAC WS501) and 253 m^3/h (Leybold RUVAC WS251). The second setup

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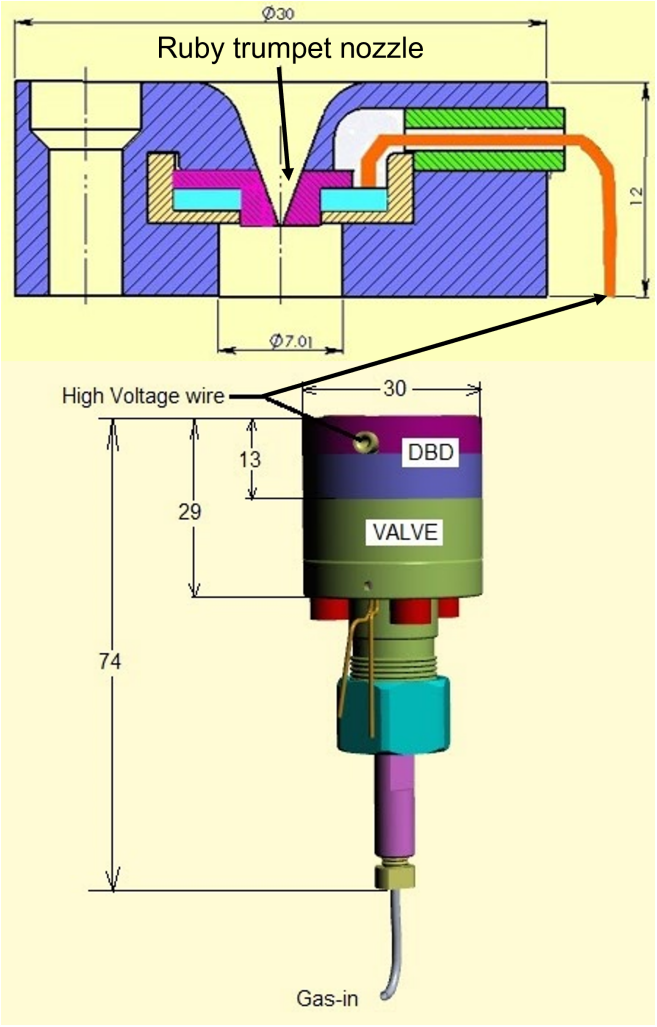

FIG. 1. Schematic cross section of the Even-Lavie pulsed valve (room temperature model) and the DBD (Dielectric Barrier Discharge) unit, responsible for exciting the helium atoms to a metastable state. Trumpet shaped ruby expansion nozzle (200 µm diameter). The numbers show dimensions in mm. Figures from the manufacturer's website[27].

has a source chamber volume of 0.045 m^3 and a total of 3600 l/s pumping capacity from 3 × Leybold MAG W, backed by a 55 m^3/h multi-stage roots pump (Leybold ECODRY 65 Plus).

Figure 2.A shows a diagram of the neutral helium detector setup. Figure 2.B shows a diagram of the metastable helium setup. Please note here the use of two detectors along the beamline. This was used to be able to account for the initially unknown internal, pressure dependent delay in the valve mechanism. For all experiments the source was left running for one hour before measuring to ensure a stable operation temperature, because it had been observed from the beam performance that the nozzle initally heats up during operation. Unfortunately it was not possible to measure the temperature of the nozzle directly in the current setup.

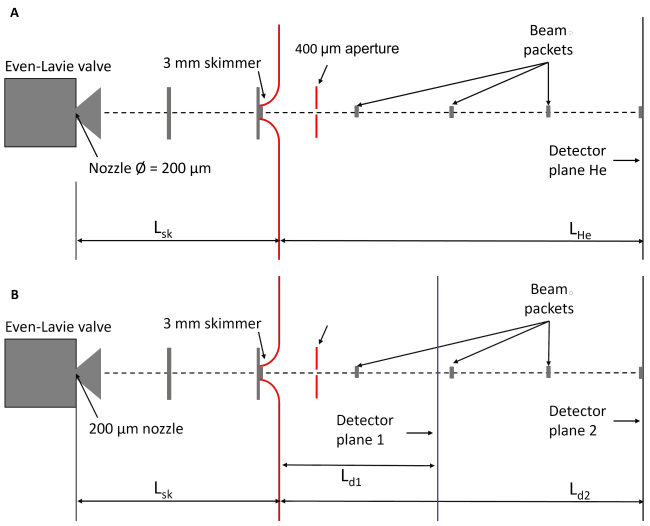

FIG. 2. (A) Schematic of the neutral helium beam setup. The distances L_{sk} (492 mm) and L_d (3132 mm) are the distances from the nozzle to the skimmer and detector, respectively. (B) Schematic of the metastable helium beam setup. Two detectors are used to account for the effects of internal delay in the valve. The distances L_{sk} (374 mm), L_{d1} (604 mm), and L_{d2} (1024 mm) are the distances from the nozzle to the skimmer and the two detectors, respectively.

A. TOF experimental method 1 - Neutral He

The neutral He time-of-flight (TOF) spectra were recorded using an ionisation detector located 3132 mm from the source nozzle opening (2640 mm from the skimmer (L_{He})). The distance from the source to the skimmer (L_{sk}) was 492 mm. The signal sent from the source controller to trigger the Even-Lavie valve was also used as the trigger (t = 0) for the TOF measurement. A 400 µm aperture was placed 960 mm downstream after the skimmer to avoid saturation in the detector. The background pressure never exceeded 1.2×10^{-4} mbar in the source chamber and 3×10^{-7} mbar in the rest of the instrument. Please note that all pressure measurements in the source chamber have been corrected for helium with the correction factor[40] K = 5.9. The source was running at a repetition rate of 100 Hz and TOF spectra were recorded for every 10 bar of source pressure (p_0) from 20-100 bar, in random order. The DBD unit was not operated during these measurements. An example of a TOF spectrum, recorded at p_0 = 100 bar, can be seen in figure 3 (top).

B. TOF experimental method 2 - Metastable He* differential TOF

The second set of TOF spectra were recorded in a setup with two detectors (see figure 2.B). The detectors were located 604 mm and 1024 mm from the nozzle, respectively. The TOF spectra were recorded by collecting the metastable He* atoms on a 4 mm diameter stainless steel plate. When a metastable helium atom hits the detector plate, it decays leading to emis-

sion of an electron. Measuring the current on the plate thus gives a measure of the incident metastable helium signal.[41] Using the same trigger as for the neutral He TOF measurements, the measured current signal from the detector plate also provides a TOF spectrum. Using the peak values (i.e. the most probable TOF) obtained from the two detectors, the true, delay independent, most probable velocity, V was calculated by $V = (L_{d_2} - L_{d_1}/)\Delta t$, where Δt is the difference in measured TOF between the two detectors and L_{d_1} and L_{d_2} are the flight distances (see figure 2.B). From these measurements, we also obtained values for the pressure dependent valve delay of the source, as shown in figure 4. The measured signal from a single pulse is low compared to background noise, therefore the measurements were done by averaging over 8 pulses per recorded spectrum, an example of a measurement can be seen in figure 3 (bottom). The source was running with a repetition rate of 10 Hz and the measurements were again performed two times per set point for source pressures 20-100 bar in 10 bar increments. A total of five sets of two measurement series were performed at different times, due to fluctuating valve delay as shown in figure 4. The chamber pressure was always below 10^{-5} mbar in the source chamber and 10^{-8} mbar in the rest of the instrument.

C. Theoretical Model

The gas expansion is described by using a theoretical model based on the solution of the Boltzmann equation by means of the moment method[42], which has successfully been used in previous studies for lower throughputs as mentioned in the introduction[37]. In this approach, since the expanding gas is not in equilbrium, two different temperatures are assumed for the motion parallel ($T_{||}$) or perpendicular ($T_{\perp}$) to the streamlines with the expansion assumed spherically symmetric[43]. Therefore the real distribution is approximated by the anisotropic ellipsoidal Maxwell velocity distribution

$$f_{aeM}(\vec{v}) = n\left(\frac{m}{2\pi k_B T_{||}}\right)^{\frac{1}{2}}\left(\frac{m}{2\pi k_B T_{\perp}}\right) \exp\left(-\frac{m}{2k_B T_{||}}(v_{||}-V)^2 - \frac{m}{2k_B T_{\perp}}v_{\perp}^2\right)$$

where k_B is the Boltzmann constant, n is the number density, m the He mass, V is the most probable velocity of the expanding gas, $v_{||}$ and $v_{\perp}$ are the parallel and perpendicular components of the velocity $\vec{v}$, respectively.

The collisions, that determine the evolution of the parameters n, V, $T_{||}$ and $T_{\perp}$ in the expanding gas, are described by the collision integral

$$\Omega^{(2,1)}(T_{eff}) = \left(\frac{k_B T_{eff}}{\pi m}\right)^{\frac{1}{2}} \int_0^{\infty} Q^{(2)}(E)\gamma^5 \exp(-\gamma^2) d\gamma \quad (1)$$

where T_{eff} is an effective average temperature varying between $T_{\perp}$ and $T_{||}$, E is the collision energy of two atoms in the center-of-mass system, $\gamma = \sqrt{\frac{E}{k_B T_{eff}}}$, and $Q^{(2)}(E)$ is the viscosity cross section.

The viscosity cross section for Bose-Einstein particles is expressed by

$$Q^{(2)}(E) = \frac{8\pi\hbar^2}{mE} \sum_{0,2,4,\ldots} \frac{(l+1)(l+2)}{2l+3} \sin^2(\eta_{l+2} - \eta_l) \quad (2)$$

where η_l is the phase shift of the partial wave with orbital angular momentum l and the sum is evaluated on even values. For the calculation of phase shifts,the spherically symmetric He-He interaction was assumed to be a Lennard-Jones potential which has proven to provide good results for explaining the properties of He beams generated by a source at room temperature[37]. The Schrödinger equation for the radial wavefunction, apart the Lennard-Jones potential, contains the centrifugal potential which depends on angular momentum and this equation has been solved by a numerical integration based on the Runge-Kutta method for a collision energy E. At sufficiently large distance, the behaviour of the wavefunction allows the estimation of the phase shifts for any orbital angular momentum l. An iterative procedure estimates phase shifts increasing the distance until the difference between phase shifts at two subsequent iterations is less than 0.005. Then eq. 2 is evaluated taking into account more l terms until a new term increases the sum for less than 0.005. This procedure has been repeated for different collision energy E in order to generate a numerical approximation to $Q^{(2)}(E)$ for the interpolation scheme employed in the evaluation of the collision integral eq. 1. Finally, a set of four equations to be solved is derived for the moments of the anisotropic distribution, i.e. gas density, most probable velocity, parallel and perpendicular temperatures as was described in ref.[43].

As a conclusive remark of this theoretical section, we observe that other studies on the supersonic beam expansion performed at low temperatures have shown the importance of including real gas properties even for helium[44,45]. In our case, where the source is operated around room temperature, this effect could be considered less relevant. Therefore the simple assumption of enthalpy independent on the source pressure, especially for helium which is considered an example of a perfect gas, may be taken for granted providing a constant terminal speed $V = \sqrt{\frac{5k_B T_0}{m}}$. On the contrary, the collected data have shown that V indeed depends on the source pressure, therefore the present calculations were carried out including the enthalpy based on the helium equation of state obtained by McCarty[46].

III. RESULTS AND ANALYSIS

Figure 3 shows examples of TOF spectra for the two experimental methods. From the neutral He TOF spectra, the most probable velocity, V, was calculated, from the TOF peak time t and the source-detector distance L as $V = L/t$. The most probable velocity of the metastable He* atoms was calculated by finding the peak time t of the TOF spectra from both detectors (figure 3 (bottom) and using the distance between them to calculate the most probable velocity by $V = (L_{d_2} - L_{d_1}/)\Delta t$. The five sets of metastable measurements (10 measurements

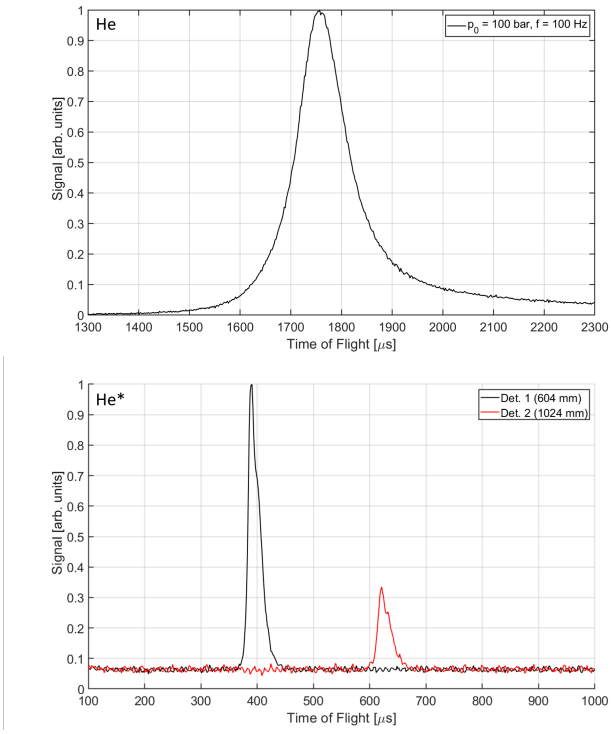

FIG. 3. Raw time-of-flight measurements of neutral helium (top) and metastable helium (bottom). The source pressure is 100 bar and the source repetition rate, f = 100 Hz and 10 Hz respectively. The neutral He detector is located 3132 from the source. Two detectors were used for the metastable helium measurements, located 604 mm and 1024 mm from the source nozzle, respectively.

per p_0d in total) have been averaged and the error bars represent the standard deviation. The measurement error of the neutral He measurements was obtained from an estimation of the accuracy by which we can determine the peak positions and there is an additional error contribution of 2 µs in the time-of-flight, due to the bin size of 4 µs of the neutral He TOF measurement setup. The most probable velocity of the He and He* atoms as a function of p_0d, where p_0 is the source pressure (or reservoir pressure) and d is the nozzle diameter, are shown in figure 5 together with theoretical predictions for temperatures 300 K, 310 K, 315 K, and 320 K, as described in section II.C. The measurements were fitted with a linear fit in accordance with the theoretical prediction. The slope of the fits is displayed in the legend of figure 5, together with the 95% confidence level interval for each fit. All measurements show an increase in the most probable velocity with increasing source pressure as predicted by theory. Considering the slopes of the fits, the agreement between theory and experiments is excellent for the neutral helium beams. This agreement allows us to estimate that the gas stagnation temperature is slightly less than 320 K, around 318.5 K. At this temperature the constant terminal speed value is 1818.8 m/s, which is also shown in figure 5 as a dash-dotted line. Its trend clearly does not represent the measured points. For the metastable helium beams, the slope is slightly higher than predicted by the theory. We suggest that this must be an effect caused by the excitation process. The difference between the most probable velocities of the neutral beams and the metastable beams is likely due to the different source repetition rates. The neutral helium measurements were performed with a source repetition rate of 100 Hz, whereas the metastable helium measurements were performed at a source repetition rate of 10 Hz. It is probable that this leads to an increase in nozzle temperature, increasing the velocity of the exiting gas.

We include also a published velocity measurement from Even et al.[47], obtained by determining the peak position of the TOF measurement in figure 10 in reference[47] and calculating the velocity from the flight distance also provided in the reference. This measurement is obtained with a pressure of 50 bar and a source repetition rate of 10 Hz[48]. It is stated in the paper that the measurement is carried out at 300 K, in good agreement with our theoretical prediction.

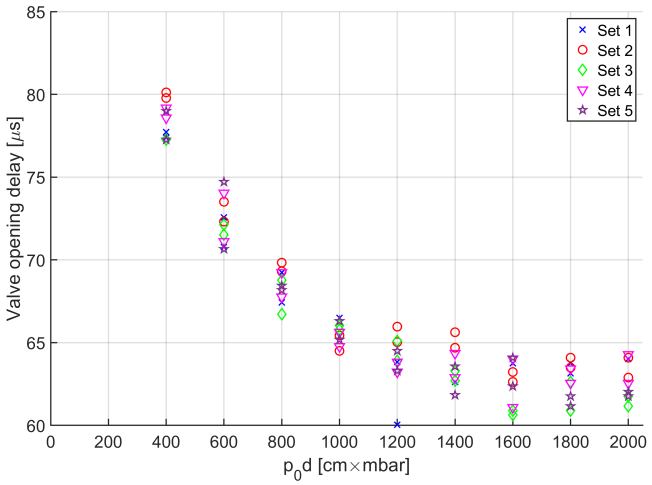

FIG. 4. The valve delay measured as a function of source pressure. The five measurement sets were taken at five different times.

IV. CONCLUSION

We present measurements of the most probable velocity for a supersonic helium expansion close to room temperature in the throughput regime p_0d = 400-2000 mbar·cm. The measurements have been compared to an existing theoretical model that is tested for the first time in this regime. Measurements are presented for neutral and metastable helium beams. For the neutral helium beams the increase in most probable velocity with p_0d is in excellent agreement with theory. For the metastable helium beams it appears that the increase is slightly bigger than predicted. We suggest that this is due to to the excitation mechanism.

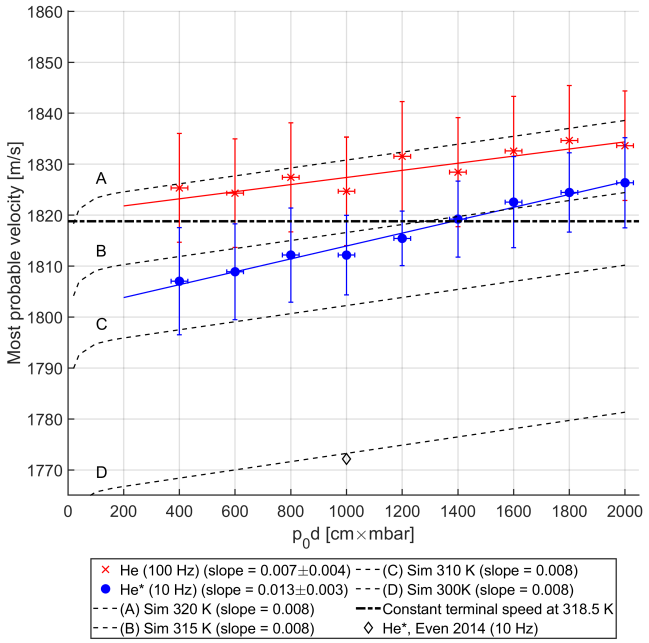

FIG. 5. The measured most probable velocity as a function of the source pressure and nozzle diameter for the two experimental setups, as well as the value measured by Even in 2014[47]. The points in the figure represent the average measured values. Linear fits were made for the measurements and the slope was calculated for each. The error of the slopes represents the 95% confidence level interval. The measurements are compared to simulations using the model described in section IIc, for the temperatures 300 K, 310 K, 315 K, and 320 K. The predicted velocity for an ideal helium gas at 318.5 K is shown as a dash-dotted line. Note also the good agreement with prevous room temperature measurements.

ACKNOWLEDGMENTS

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