Chemical Physics Letters xxx (2018) xxx–xxx

Contents lists available at ScienceDirect

Chemical Physics Letters

journal homepage: www.elsevier.com/locate/cplett

Editor's choice

Pulsed helium droplet beams

Deepak Verma [a], Andrey F. Vilesov [a,b,*]

[a] Department of Chemistry, University of Southern California, Los Angeles, CA 90089, USA
[b] Department of Physics and Astronomy, University of Southern California, Los Angeles, CA 90089, USA

ARTICLE INFO

Article history:
Received 10 December 2017
In final form 17 January 2018
Available online xxxx

ABSTRACT

We report a systematic study of pulsed cryogenic expansion at temperatures ranging from 20 K down to 4.5 K, which enables us to span a large range of helium droplet sizes from $\sim 10^5$ to $\sim 10^{11}$ He atoms, as obtained via titrations of the droplet beam with helium gas at room temperature. The measured peak flux is a factor of one thousand larger than in a continuous beam. Comparison with continuous nozzle results show similar droplet sizes at low T < 6 K, indicating in both cases the droplets are created via the fragmentation of the fluid.

1. Introduction

Over the past two decades, helium (He) droplets have been widely used for laser spectroscopic studies of molecules, molecular and atomic clusters [1–5]. Molecular species isolated in cold ($\sim$0.4 K) and weakly interacting droplets often demonstrate well resolved vibrational spectra from which the structural information could be inferred. On the other hand, resolved rotational spectra have been used to study interaction of molecules with the superfluid environment [1,5–7]. Experiments with simple molecules and small ($N < 10$) clusters [8–10] employ smaller He droplets that are a few nanometer in diameter and contain up to about 10^4 atoms. More recently, helium droplets were used to form large atomic and molecular clusters containing thousands to millions of particles, which were studied via spectroscopy *in situ* as well as by electron microscopy upon surface deposition [11–14]. Due to low evaporation enthalpy of liquid helium of $\sim$7 K per atom, to sustain evaporative losses, the number of helium atoms in the droplets must be a factor of $\sim$1000 larger that the number of the embedded particles. Capture of protein ions also requires a sufficiently large droplet [15]. Recent diffraction experiments with 100 nm sized doped droplets with an X-ray free electron laser (XFEL) demonstrated the emergence of quantized vortices in superfluid helium droplets [16] and enabled study of atomic aggregation mediated by quantum vortices [17–19]. Such studies demand the production of a large droplet with average number of atoms, $\langle N_{He}\rangle > 10^7$.

Helium droplets are produced from cryogenic expansion of pressurized helium in vacuum; larger droplets are formed at lower nozzle temperature. Most of the experiments up to date involve continuous nozzle beam expansion technique, which is well-characterized [20–23] and covers droplet sizes from $\sim$100 to $\sim 10^{12}$ atoms. Pulsed droplet sources [24–27] are advantageous for studies involving pulsed techniques, such as laser-induced fluorescence, photo ionization or X-ray coherent diffraction imaging. Pulsed nozzles, which usually contain an electromagnetic valve, however, are difficult to run at the low temperatures of $T < 6$ K required for production of large droplets due to heat evolved during their operation. Nevertheless, experiments with helium droplets involving pulsed cryogenic nozzles are emerging [15,20,26–32]. Most of the groups employing pulsed valve use so called Even-Lavie valve [24,27,32–34], which is rather expensive and difficult to acquire. Recent study with Even-Lavie valve reports sizes of $\sim 10^{10}$ atoms at the lowest temperature of 5 K [32].

We have continued the development of an alternative pulsed valve technique which is based on a commercially available Parker Series 99 electromagnetic valve with 0.5 or 1 mm diameter [15,20,26,29,31]. Here, we report on the formation of pulses of large sized helium droplets, $\langle N_{He}\rangle = 10^5$–$10^{11}$, which were obtained at stagnation pressures of 5 and 10 bar and temperatures of 4–15 K. The low operation temperature is achieved upon confining the valve within a copper shroud in thermal contact with second stage of the closed cycle refrigerator. The average droplet sizes are obtained by attenuation of the droplet beam with collisional helium gas at room temperature as described by Gomez et al. [20]. Additionally, the pulsed helium droplet beam intensity is measured and compared with the previous results with a continuous source.

2. Experiment

A schematic of the molecular beam setup is shown in Fig. 1. It comprises of a source chamber which hosts a close cycle refriger-

* Corresponding author at: Department of Chemistry, University of Southern California, Los Angeles, CA 90089, USA.
E-mail address: vilesov@usc.edu (A.F. Vilesov).

https://doi.org/10.1016/j.cplett.2018.01.035
0009-2614/© 2018 Published by Elsevier B.V.

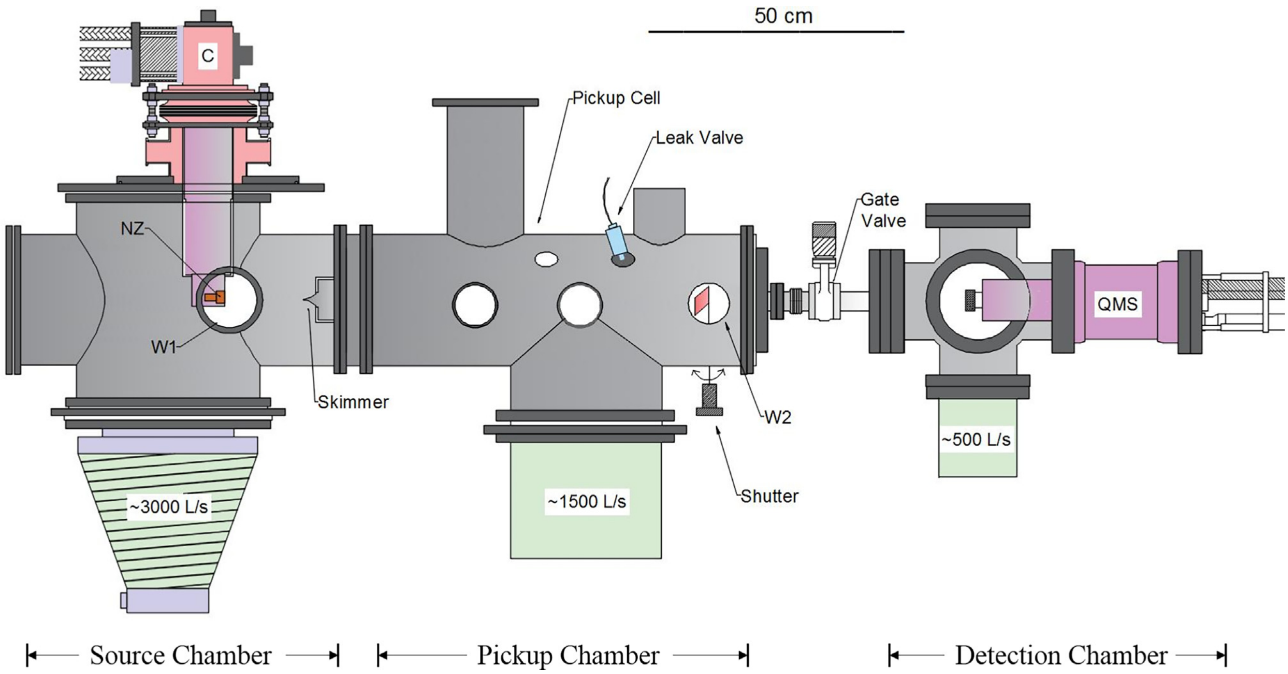

Fig. 1. Schematic of the He droplet setup. The source chamber on the left hosts pulsed valve nozzle (NZ) attached to a cold head (C). It is backed by a 3000 L/s diffusion pump. The droplets pass through a 2 mm skimmer into the pickup chamber where the collisional He gas is introduced through a leak valve. This chamber is backed by a 1500 L/s turbomolecular pump. The pickup chamber also hosts a fast ion gauge (not shown) and a shutter for beam block. The droplets enter the UHV detection chamber through a 5 mm orifice and a gate valve where the helium ions are produced and detected by an axial quadrupole mass spectrometer (QMS). W1 and W2 are optical windows.

ator (Sumitomo, RDK-408D). The droplets are produced by expanding high purity (99.9999%) helium at stagnation pressure P_0 and temperature T_0 into vacuum. We employed a Series 99 Parker (formerly General Valve) valve equipped with coppers gaskets and a Kel-F poppet which is operated by IOTA-ONE (General Valve) external driver. The valve consists of a body and a faceplate. The body accommodates a solenoid which holds an iron cylinder with attached spring and a poppet. The tip of the poppet sits on the orifice of the faceplate. The 1 mm (0.5 mm) orifice has a conical opening with angle of ~90°. Upon application of the electric pulse to solenoid, the poppet moves back, letting helium flow through the orifice, see Ref. [26] for more details. The central part of the beam is isolated by a 2 mm diameter skimmer placed at ~15 cm downstream from the nozzle. After passing through the skimmer, the beam enters the 0.82 m long pickup chamber. Further downstream, the beam passes through a gate valve and enters a UHV detection chamber containing an axial quadrupole mass spectrometer (QMS) (Extranuclear laboratories, 020-1) equipped with an electron impact ionizer. The QMS ionizer resides 130 cm downstream from the nozzle. The default ionization current and energy were set to 5 mA and 100 eV, respectively. The QMS is employed to measure intensities and the time of flight profiles of the droplet beam. The pressures in the vacuum chambers were monitored by hot cathode ion gauges. The pickup chamber is additionally equipped with a fast ion gauge, which is used for the droplet flux measurements as described in the following section.

The average droplet sizes were determined via attenuation of helium droplet beam by collisional He gas, as developed previously in Ref. [20] for continuous beams. During the attenuation experiments, the pickup chamber is flooded with He gas, which enters through a leak valve. The droplets encounter multiple collisions with the helium atoms as they pass through the pickup chamber leading to decrease of the average droplet size and concomitant decrease of the droplet beam intensity, as dictated by the collisional cross-section. The change of the He droplet beam intensity upon increasing of the pressure of the He collision gas in the pickup chamber is monitored by QMS which was tuned to detect mass $m = 8$ due to He_2^+ splitter ions.

A three dimensional rendering of the adapter of valve to the cold head is presented in Fig. 2. The adapter has a copper cylinderical body with a flat rectangular face (Fig. 2(c)). The top of this design attaches to the second stage of the cold head, as shown in Fig. 2(f). The bottom end is covered by a copper lid (Fig. 2(d)). The rectangular face has two circular cavities. The outer cavity ensures proper fitting of the valve face plate while the inner cavity extends further inside the body of the cylinder. This extension provides contact between the valve body and the surrounding copper for efficient heat exchange. Once the valve is secured in place with screws, an auxiliary copper slab (outer cover in Fig. 2(f)) with a conical opening is placed in front of the faceplate and secured with bolts for better heat exchange between the valve face plate and the cold source. The He inlet line runs from the top of the cold head down to the bottom where the copper part is attached. It is coiled around the copper assembly from outside through 3 turns before it enters through a hole and attaches to the valve (Fig. 2(d)). Temperature is monitored by two silicon diode sensors: the first is placed on the second stage of the cryostat (Fig. 2b), whereas the second is attached to the supply line of the pulsed valve (Fig. 2(d)). The nozzle temperatures, T_0, reported in this paper are measured from the second silicon diode which gives the true reading of the gas temperature. The temperature is controlled through resistive heating. The temperature measured with the second sensor is higher than with the first one, due to the finite heat conductivity between the adapter assembly and the cold head.

3. Results

Fig. 3(a) and (b) show the time dependence of the QMS signal at mass 8, I_8, as measured at the two repetition rates of 1 Hz and 20 Hz, respectively, and lowest temperature with heater off. Fig. 3(b) shows two prominent peaks. The peak at the earlier arrival time of

Please cite this article in press as: D. Verma, A.F. Vilesov, Chem. Phys. Lett. (2018), https://doi.org/10.1016/j.cplett.2018.01.035