Cluster size determination using shadowgraphy measurements

Hanna Eick*, Philipp Brandl, Christian Mannweilerl, Sophia Vestrickl, and Alfons Khoukazl

1Westfälische Wilhelms-Universität Münster, Institut für Kernphysik, Wilhelm-Klemm-Straße 9, 48149 Münster, Germany

Abstract. Cluster-jet beams offer a wide variety of possible applications. The advantages of an internal and windowless cluster-jet target make it suitable for many experiments. It is important to know all the properties of the cluster-jet to prepare it for the desired conditions of the respective experiments. An essential property is the size of the clusters and their size distribution. To investigate the size of the clusters shadowgraphy measurements are performed. This article provides an overview of the shadowgraphy method and presents first results of these analyses using hydrogen clusters. These show an average cluster size of a few micrometers, which will also be of high interest, besides the here shown laser cluster interaction, for other installations using cluster beams.

1 Introduction

Cluster-jet targets developed and built by the University Münster are central components for a wide range of experiments. Some of the advantages, which make this type of target highly suitable for the future PANDA experiment at FAIR in Darmstadt, are that the target is windowless and achieves the required thickness in the range of $10^{15}$ atoms/cm² after a distance of more than 2 meters from the nozzle [1, 2]. To optimally adapt the cluster-jet targets to the desired conditions of every experiment, it is necessary to understand all properties of the target. One of the properties is the size of the clusters and their size distribution. This information is also important for other analyses and simulations, which require understanding of cluster-jet target properties like, e.g., the cluster evaporation process.

2 Shadowgraphy measurements at the ARCTURUS TW laser system

For the investigation of the cluster size distributions, shadowgraphy measurements have been performed at the Heinrich-Heine University in Düsseldorf. A cluster-jet target [2, 3, 4], which can be seen in fig. 1, has been mounted there to investigate the laser cluster interaction [4, 5] with the ARCTURUS TW laser system [6] in several measurement campaigns.

The target is constructed in a way, that the process gas, which is in most of the cases hydrogen, is cooled down by a two-staged cryocooler. Due to the specially shaped convergent-divergent nozzle, called Laval nozzle, clustering/atomization of the cooled gas or liquid can take place. The high mass of the clusters allows them to travel a large distance of several meters without an impact of scattering with the residual gas under usual operating conditions and therefore high densities at this distance can be reached. In contrast to the future PANDA experiment, the experiments with the ARCTURUS laser are performed only a few centimeters behind the nozzle, so additional elements for extracting the target beam and separating it from residual gas are not necessary.

![CAD drawing of the cluster-jet target operated in Düsseldorf to study laser cluster interactions.](image)

Fig. 1. CAD drawing of the cluster-jet target operated in Düsseldorf to study laser cluster interactions. The gas is purified with a purification cartridge before it enters the target, where it is cooled at the two stages of a cold head. The gas enters the nozzle at cryogenic temperatures and at high pressures and forms a cluster-jet. Figure generated by D. Bonaventura (WWU).

2.1 Setup of the measurements

The setup used for the shadowgraphy measurements can be seen in fig. 2. The image shows the experimental setup with the cluster-jet target and the ultrashort pulse laser beam. The laser beam with a wavelength of 800 nm and a pulse length of 30 fs enters the chamber, hits the cluster beam and is directed onto an objective and a subsequent camera with an exposure time much longer than the pulse length of the laser. The cluster beam is

* Corresponding author: h_eick03@uni-muenster.de

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generated in the cluster source and directed to the vacuum pumps. Due to the short duration of the laser beam, the clusters can be assumed as stationary in this time window. Shadowgraphy images are taken at different target stagnation conditions with liquid hydrogen in front of the nozzle and horizontal positions in the cluster beam.

![Diagram](image)

**Fig. 2.** Setup of the shadowgraphy measurements with the cluster-jet beam from top to bottom and the laser beam from left to right. The shadowgraphy images are taken with a CCD camera outside of the chamber.

### 2.2 Shadowgraphy image

With the setup shown in fig. 2, a large number of shadowgraphy images have been taken. An example for a background subtracted shadowgraphy image is shown in fig. 3. Therefore, background images are taken at a gaseous target condition, where the clusters are smaller and thus not visible. In the picture one can see three sharp clusters, that are visible as dark, slightly smeared out spots. Some disturbing structures, like e.g., interference rings of clusters, that are too far away from the focal plane, are apparent and some dust on the camera and the optics still remains despite the background subtraction due to the shot-to-shot variation of the laser beam.

![Image](image)

**Fig. 3.** Shadowgraphy image of a cluster beam with three dark, slightly smeared out clusters. Additional structures on the picture are due to other clusters outside of the focal volume or due to dust on the optical components.

### 2.3 Determination of cluster size distribution

To determine the size distribution of the clusters an algorithm has been developed, which finds potential cluster candidates and decides via cluster selection criteria whether it is a cluster or not. After this sorting process all remaining cluster-like structures are fitted with a two-dimensional function and the diameter is extracted. The resulting cluster size distribution can be seen in blue in fig. 4.

![Graph](image)

**Fig. 4.** Measured raw cluster size distribution for a target setting of 24 K and 16 bar and a fixed position in the cluster beam (blue). To correct for imaging effects a deconvolution is performed, which is shown in green.

In the analysis of the data, it has been found that the extracted size of the clusters depends on the individual distance between the cluster to be examined and the focal plane of the microscope objective. A deconvolution is made to account for this optical imaging effect. The real cluster size distribution can be described well by a Gaussian function, which is shown in fig. 4 in green. To approximate the raw data (blue histogram), a convolution of this Gaussian function with a smearing effect (orange histogram), caused by the mentioned out-of-focus plane observation, is fitted to the data in the blue histogram. The convolution describes the measured data sufficiently well. With the shadowgraphy method, clusters are found in the range from 3 to approx. 10 μm. The latter value establishes an upper limit on the maximum cluster size. The course of the measured cluster size distribution (blue curve in fig. 4) shows a lower boundary limited by resolution. This resolution limit can be seen as a steeper trend on the left-hand side of the distribution and is caused by the wavelength of the laser and the numerical aperture of the microscope objective. To get more insight into the distribution of possibly smaller and with this method invisible clusters, different methods have to be tested [7, 8].
2.4 Determination of volume density

Using the evaluated cluster size distributions, it is possible to determine a volume density and to compare what percentage of the gas flowing through the nozzle is contained in the visible clusters analyzed with the shadowgraphy measurements. Fig. 5 shows the measured volume density based on the shadowgraphy results in blue under the usage of the mean volume of a cluster, the number of all clusters at a measured setting, the liquid hydrogen density and the volume of the images. The expected volume density in orange is calculated from the hydrogen flow, which passes the nozzle and the cluster velocity that can be measured via a ToF setup [9].

As the data analysis is not yet completed, no final quantitative conclusion can be drawn from fig. 5 at this stage.

![Graph showing measured and expected volume density](image)

**Fig. 5.** Volume density from the shadowgraphy analysis shown in blue and expected volume density calculated from the gas flow through the nozzle in orange (work in progress).

However, two important results can be highlighted from fig. 5. First, it is evident that not all the gas that went initially through the nozzle is embedded in the large clusters. This result confirms the assumption made earlier that there are still smaller clusters that are below the resolution limit of the shadowgraphy images. Then, spatial structures can be recognized in the measured volume density. This means that the cluster beam is not homogeneously distributed, but that there are regions in the beam that are denser than others. This is in agreement with observations on high-intense hydrogen cluster beams, produced, e.g., at the PANDA cluster-jet target [10]. Further investigations related to this result will be carried out in the future.

When liquid hydrogen passes through a Laval nozzle, atomization takes place and clusters in the range between 3 μm and 10 μm are found to be produced. In comparison, no clusters are seen with gaseous hydrogen entering the nozzle, concluding that the formed clusters are then smaller than 3 μm.

Based on the analysis and the comparison of the integrals of the curves in fig. 5, it is seen that not all of the hydrogen is contained in large clusters, but there must also be smaller clusters, which are not detected on the images. The clusters found are in the order of a few micrometers, and a structure can be detected in the cluster beam. An upper limit of the cluster sizes of 10 μm has been determined.

The results presented here open possibilities for future measurements and give important information for experiments with cluster-jet beams, as well as for their analyses and simulations.

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