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Gas Cluster Ion Formation Under Pulsed Supply of Various Working Gases[1]

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Abstract—Gas cluster formation during gas expansion through a supersonic nozzle with a skimmer is described. The role of the buffer zone between a pulsed valve and a nozzle in the regime of pulsed supply of working gas is shown. Influence of the working gas type on the cluster ion pulse parameters is investigated.

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Gas cluster ion beams are widely used for precise surface smoothing, ultra-shallow implantation, ion-assisted deposition of thin films, as a probe for SIMS [1–5]. In research and industrial accelerators, cluster ions are formed during working gas adiabatic expansion under high pressure into a vacuum chamber through a supersonic nozzle. A large gas flow causes a high load of pumping system. Pulsed supply of the working gas is a way of mitigation the gas load.

The goal of this work consisted in investigating the influence of parameters of working gas pulse as well as the pressure and the type of the gas on the properties of the cluster ion beam being formed. The experiments were carried out using the gas cluster ion accelerator of Lomonosov Moscow State University, described in [5]. Schematic diagram of the experiments is represented in Fig. 1. A working gas was supplied into a conical supersonic nozzle *3* with critical cross-section of 130 μm with a pulsed valve *1*. Gas pressure was set in range from 1 to 7 atm. There was a buffer zone *2* between the valve and the nozzle.

It is known that a gas expanding from a nozzle forms a barrel-like structure, bounded by a normal shock, so called Mach disk. The distance between the nozzle exit and the Mach disk x_M is given by the next expression [6]:

$$\frac{x_M}{d} = 0.67\sqrt{\frac{p}{p_\infty}}, \quad (1)$$

where p and p_∞ are gas pressure at the entrance of the nozzle and in the vacuum chamber, respectively; d is nozzle critical cross-section. Clusters, formed during gas expansion, exist only inside the Mach barrel and collapse when passing through the normal shock. For cluster extraction, a conical skimmer *4* is used, which penetrates through the Mach disc and cuts out a part of the cluster flow, permitting it to pass into the next chamber.

The particles passed through the skimmer were ionized by an electron impact and accelerated by a potential of 2 keV in the ionization and acceleration unit *5*. For mass analysis of the ionized particles beam, an electromagnet *6* was mounted. A Faraday cup *9* was placed at the beam axis, and another Faraday cup was at some distance from the axis. Without magnetic field, the central Faraday cup measured current of both atomic and cluster ions. When the magnetic field was on, only large cluster ions *7* came to the central Faraday cup.

Figure 2 shows oscillograms of argon cluster current to each Faraday cup without magnetic field and with the field, which magnitude corresponds to deflection of the working gas monomers *8* (Fig. 1) to the second Faraday cup. It is well seen that cluster ions are present in the beam.

Under the largest available magnitude of the magnetic field, which was 300 mT, cluster current signal almost did not change, which means that the cluster consist of at least 500 argon atoms per cluster. We should emphasize that the current pulse duration (350 ms) is significantly larger than the duration of pulsed valve open state (20 ms, showed in the Fig. 2). This value of open state duration corresponds to experimental estimation of time, needed for filling the buffer zone with the gas.

Similar experiments were carried out for xenon, nitrogen and neon. The duration of pulsed valve open state was every time set to 20 ms. For xenon and argon clusters are most prominent, for nitrogen cluster cur-

[1] The article was translated by the authors.

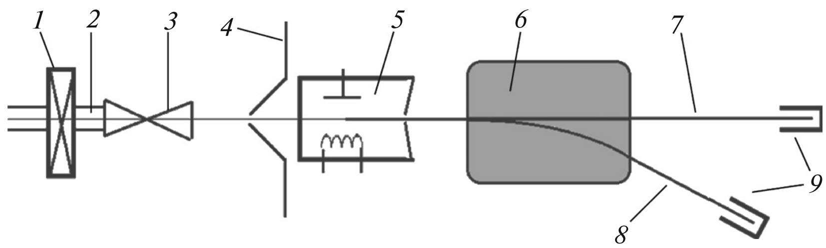

Fig. 1. Schematic diagram of the experiments.

rent is much less then monomer current, and neon clusters were not found in the beam.

This experimental data is in good agreement with calculated values of Hagena parameter Γ*, which is used for estimation of mean cluster size in a nozzle cluster source [7]:

$$\Gamma = \frac{k(d_{ef}[\mu\mathrm{m}])^{0.85} p[\mathrm{mbar}]}{(T_0[\mathrm{K}])^{2.29}}, \quad d_{eq} = 0.74\frac{d}{\tan\alpha}, \tag{2}$$

where k is the condensation parameter, T_0 is the working gas temperature at the nozzle entrance, d_{eq} is the equivalent nozzle diameter, and α is the half-opening angle of the supersonic nozzle. It is known that formation of dimers and trimers starts when Hagena parameters is larger than 200, and effective formation of large clusters—when Γ* > 1000. The value of the parameter is evaluative and can decrease or increase depending on the particular nozzle shape and experimental conditions.

Condensation parameters, given in the table, decrease from xenon to nitrogen. Hagena parameter values, calculated for the gases under experimental conditions, are as well given in the table. As it was mentioned, in the experiments nitrogen cluster formation was suppressed, and neon clusters are practically absent.

The shape of the signals to each Faraday cup can be explained as follows. It was shown that clusters exist only in the silence zone inside Mach barrel, and a skimmer prevents them from collapsing when its cutting edge penetrates into the jet core. When the skimmer cutting edge stops penetrating into the zone of silence, clusters cannot come through the normal shock, so monomers are prevailing in the beam.

Pressure at the entrance of the nozzle is set by the pressure of the gas supplied to the system. When the pulsed valve turns open, gas starts flowing into the buffer zone between the valve and the nozzle. Since valve orifice is significantly larger than nozzle critical cross-section, the gas flows in quite rapidly, and inside the buffer zone pressure becomes equal to the set working gas pressure value. After the valve closes and the gas flow into the buffer zone is eliminated, pressure inside it slowly decreases because of outflow through the nozzle.

When the pressure in the buffer zone decreases so that according to the expression (1) the distance between nozzle exit and normal shock is less than the distance between the exit and the skimmer edge, the skimmer stops penetrating into the Mach barrel. Clus-

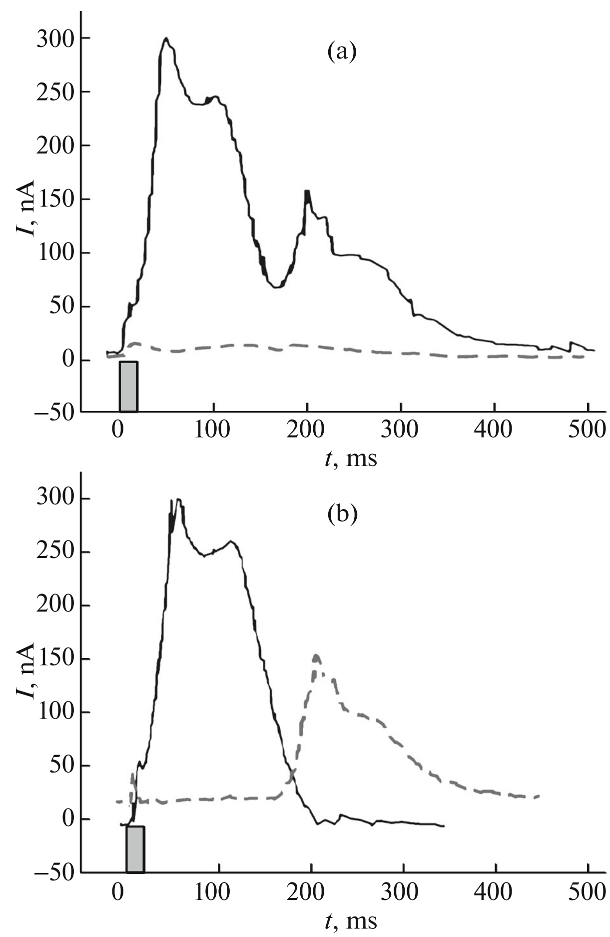

Fig. 2. Oscillograms of (Ar)N cluster ion current (a) without magnetic field and (b) with magnetic field corresponding to deflection of the monomers to the second Faraday cup: (solid line) current on the axis; (dashed line) current to the side Faraday current. Open state duration of the pulsed valve is shown.

Condensation parameters [8] and Hagena parameters for various gases

Parameters	Xe	Ar	N_2	Ne
Condensation k	5500	1650	528	185
Hagena Γ*	17957.5	5387.25	1723.92	604.025

Gas pressure 5 bar, room temperature; nozzle with critical cross-section 130 μm and half-opening angle 6°.

ters collapse passing the normal shock, and the gas comes into the skimmer as separate monomers.

So, the first, or monomer, stage of the current pulse development corresponds to rapid growth of the pressure at the entrance of the nozzle, enlargement of the Mach barrel and beginning of penetration of the skimmer into it. The middle stage corresponds to the period of cluster existence in the ionized beam. The final stage, which is monomer again, corresponds to skimmer going out from the zone where clusters exist.

A formula was suggested for evaluation of outflow time of a gas with molar mass M and heat capacity ratio γ from a cylindrical buffer zone with length l and diameter D, when the pressure inside it drops from p_0 to p, which means duration of cluster pulse [5]:

$$t = \tau\left[\left(\frac{p_0^{\frac{\gamma-1}{2\gamma}}}{p} - 1\right)\right],$$
$$\tau = \frac{2Dl}{d^2(\gamma-1)}\sqrt{\frac{(\gamma+1)M}{2\gamma R T_0}}, \tag{3}$$

where R is the gas constant. The formula was derived assuming the process is isentropic.

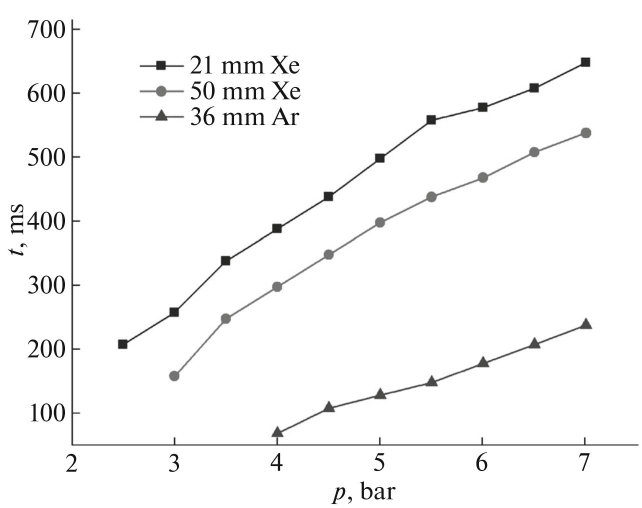

Fig. 3. Duration of cluster pulse for xenon and argon depending on working gas pressure for different distances between the nozzle exit and the skimmer edge.

For argon, τ equals 300 ms. The value of τ is proportional to square root of gas molar mass, i.e., heavier gases outflow from the buffer zone slower. Thus, the pressure inside it decreases slower, and cluster pulse duration increases if the other parameters are set equal. Besides, the duration is influenced by the distance between the nozzle exit and the skimmer edge. Figure 3 shows experimental values of cluster pulse duration for argon and xenon for various pressures and nozzle–skimmer distances. Indeed, the duration increases under increase of stagnation pressure, under decrease of nozzle–skimmer distance and under transition to a heavier working gas.

We should note that expression (1) for the distance to the Mach disk includes value of the pressure inside the cluster formation chamber. In a pulsed regime, this pressure changes with the pulsed valve periodicity. Direct measurement of it is complicated because of finite time of pressure equalization inside the chamber and lag effect of gauges. Nevertheless, it has influence on the value of cluster pulse duration. Therefore, in case of a large duration of valve open state the pressure inside the chamber has time to significantly rise, diminishing the Mach barrel and shortening cluster pulse duration.

Thus, it was shown that the key role in gas cluster pulse formation has a buffer zone between a pulsed valve and a nozzle. Cluster pulse duration is considerably larger than the valve open state duration. At the same time, duration of a cluster pulse is determined by the working gas type and pressure, as well as the distance between the nozzle and the skimmer. In particular, it grows under increasing molar weight of the gas because of increasing time of gas outflow from the buffer zone. The obtained results are in a good agreement with the model developed earlier.

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