

LEMPUS-2: Laboratory vacuum facility for comprehensive study of rarefied flows

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ABSTRACT

The paper provides an up-to-date description of the LEMPUS-2 experimental gas-dynamic facility, which offers an integrated approach to studying rarefied flows. The diagnostic methods used, based on electron-beam fluorescence technique and molecular beam diagnostics, are described and validated. The feasibility of applying these methods in flows with advanced condensation is considered. Results demonstrating the influence of the condensation process on the gas dynamics and molecular kinetics of supersonic rarefied flows are presented.

1. Introduction

Methods and approaches to the study of rarefied flows have been gradually improving since the middle of the last century, in line with the needs of aerospace engineering. Since conducting research in outer space conditions requires enormous financial and material resources, ground-based experimental facilities are actively involved in solving emerging tasks. The simplest solution is to create a vacuum facility that reproduces research objects at full scale [1–7]. However, large complexes are extremely difficult to operate and require colossal investments for equipment, which justifies the search for simpler and more cost-effective solutions. To minimize costs, it is possible to transition from a steady-state gas flow regime from a nozzle to a pulsed one [8–10]. However, as shown in Refs. [11,12], this alternative has a significant limitation: the formation of a quasi-stationary core is observed only when the condition of small pressure disturbance in the surrounding space is met. An important step in reducing the cost of ground-based experiments is the use of numerical simulation methods, which have received wide development in recent decades [13–23]. At the same time, in order to create new spacecraft, develop research on other bodies in the Solar System, discover new gas-dynamic effects, and test the constructed models, it is necessary to continue developing experimental methods for studying rarefied flows that are applicable under various initial conditions. A miniaturization method based on the use of the principles of similarity between model and full-scale objects

was presented in Refs. [1,24,25]. This approach allows for the transfer of research to small laboratory setups that are relatively inexpensive to equip and easy to operate [8,9,26–31]. However, due to the difficulty of reproducing fuel combustion temperatures and working with flows at low stagnation temperatures (approximately room temperature), this approach is associated with the intervention of the condensation process [32,33].

During adiabatic expansion, the internal energy of the gas transforms into the kinetic energy of directed motion, which leads to a temperature decrease (see Appendix A) [34]. If the temperature before expansion was room temperature (about 300 K), then in the flow it can drop to cryogenic values, passing the phase transition point. Upon reaching the critical value where the adiabat intersects with the saturated vapor pressure curve, expansion continues along the “dry” isentrope into the supersaturation region. Subsequently, the collapse of the supersaturated state occurs, the expansion curve shifts from the adiabat to the equilibrium line with decreasing temperature, and progressive cluster formation begins, which leads to changes in the mass distribution in the flow [32]. In the absence of covalent bonds, the interaction between particles is due to weak dispersion forces (specific binding energy <80 meV [35]), formed during the emergence of instantaneous electronic dipoles (and higher multipoles) inducing induced dipoles in neighboring atoms. Due to the low energy capacity of the primary cluster (dimer), the dimerization process occurs through triple collisions in the flow, which serve to remove the released binding energy. In turn,

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during the formation of larger clusters, the binding energy is redistributed in volume, leading to heating of the associate. Thus, during the clusterization process, condensation heat is released, leading to changes in the internal energy of the thermodynamic system, which affects the

gas-dynamic parameters of the flow [36–38].

According to Refs. [37,39], the similarity parameter determining cluster formation in the gas flow behind an axisymmetric nozzle is the correlation complex Γ^* , determined by the pre-expansion gas state:

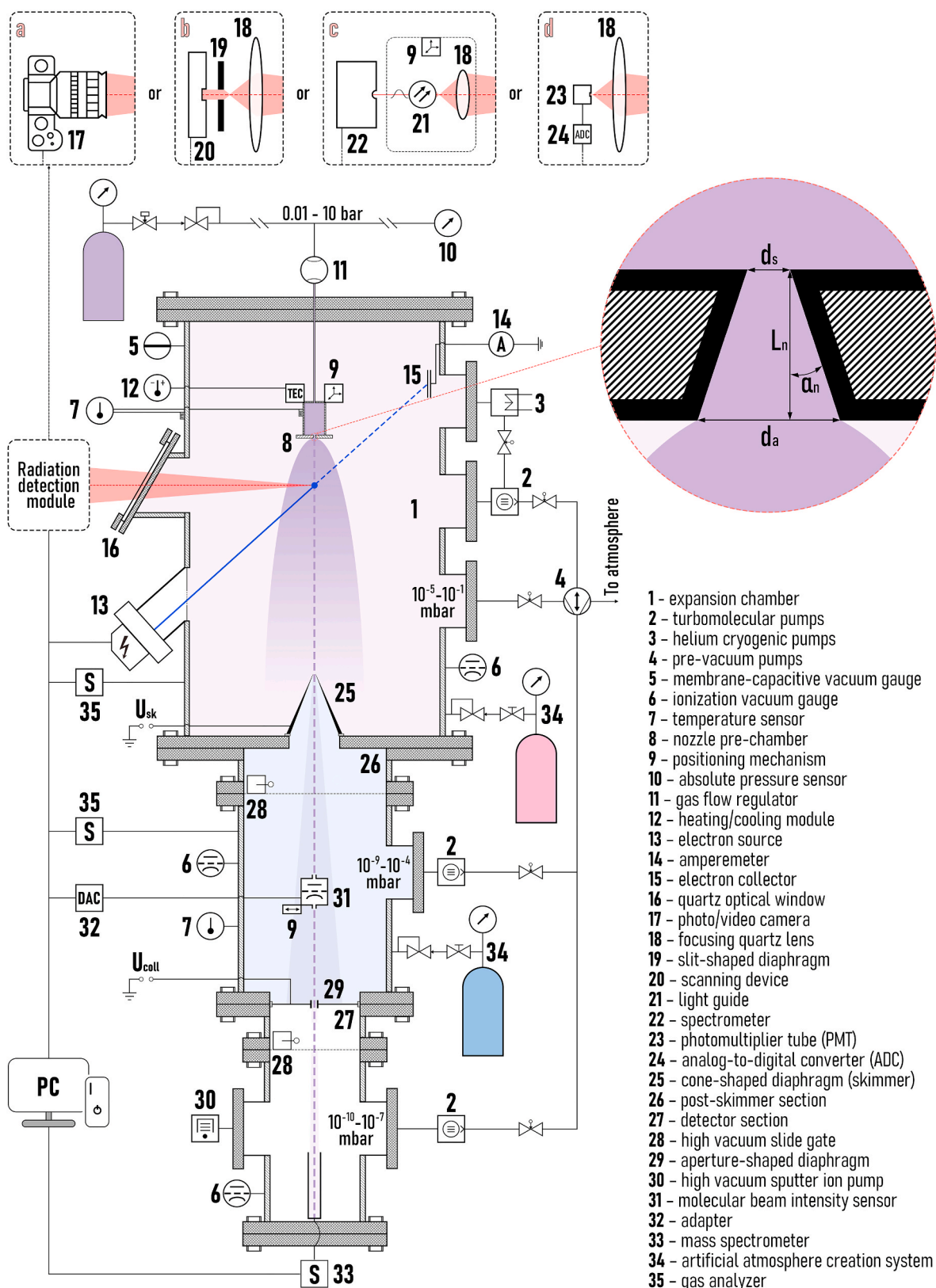


Fig. 1. Schematic representation of the gas-dynamic stand «LEMPUS-2».

$$\Gamma^* = a_1 \cdot P_0 \cdot d_{eq}^{a_2} \cdot T_0^{-a_3}, d_{eq} = a_4 [\gamma] \cdot d_s / \tan \alpha_n, \quad (1)$$

where d_{eq} is the equivalent diameter of the supersonic conical nozzle; $a_1 - a_4$ are empirical constants depending on γ [39]. In Ref. [32] it is indicated that:

- $\Gamma^* < 200$ — no particle condensation is observed;
- $200 < \Gamma^* < 1000$ — transition to flow with cluster formation occurs;
- $\Gamma^* > 1000$ — mass condensation with large cluster formation is observed in the flow.

Research by different authors allowed using Γ^* for estimation of the average size of clusters formed in the flow $\langle S \rangle$:

$\Gamma^* < 350$	$\langle S \rangle = b_0 + b_1 \cdot \Gamma^* + b_2 \cdot (\Gamma^*)^2 + b_3 \cdot (\Gamma^*)^3$ [33],	a	(2)
$\Gamma^* < 1800$	$\langle S \rangle = 38.4 \left(\frac{\Gamma^*}{1000} \right)^{1.64}$ [33],	b	
$\Gamma^* > 1000$	$\langle S \rangle = 33 \left(\frac{\Gamma^*}{1000} \right)^{2.35}$ [37],	c	
$\Gamma^* > 1800$	$\langle S \rangle = \exp \{ c_0 + c_1 (\ln \{ \Gamma^* \})^{0.8} \}$ [33],	d	
$\Gamma^* > 10000$	$\langle S \rangle = 1.64 \cdot 10^{-4} \cdot (\Gamma^*)^{1.8}$ [40],	e	

Where $b_0 = 2.23$; $b_1 = 7.0 \cdot 10^{-3}$; $b_2 = -8.3 \cdot 10^{-5}$ (the work [33] contains an error in the sign of the constant, the correct version has a minus sign); $b_3 = 2.55 \cdot 10^{-7}$; $c_0 = -12.83$; $c_1 = 3.51$ are constant numerical coefficients.

This paper will present a description and validation of the diagnostic methods implemented at the LEMPUS-2 gas-dynamic facility, as well as a consideration of the feasibility of their application in flows with advanced condensation within the context of experimental modeling for aerospace engineering. Furthermore, the results of an investigation into the influence of the condensation process on the gas dynamics and molecular kinetics of supersonic rarefied flows will be demonstrated.

2. Technical equipment of the LEMPUS-2 facility

In the early 2010s, the Department of Applied Physics at Novosibirsk State University (NSU) developed the LEMPUS-2 universal vacuum facility, designed for comprehensive studies of gas flows in vacuum or highly rarefied medium. The initial technical description of the facility was presented in Refs. [27,29]. Previously, research at the LEMPUS-2 facility focused on studying gas-surface interaction processes, the formation of various films, layers, and structures on surfaces, as well as excitation processes and chemical reactions in the gas and solid phases. During subsequent operation, the equipment underwent a significant upgrade, and diagnostic techniques were implemented and further developed, allowing for an expansion of the research scope. The current schematic representation of the experimental setup is shown in Fig. 1.

The expansion chamber (1) is a cylinder with a diameter of 700 mm and length of 1200 mm (approximately 0.46 m^3), equipped with a 500 mm diameter hatch. The pressure in the expansion chamber P_∞ is maintained by turbomolecular pumps (2) Shimadzu TMP-2203LMC (total pumping capacity for N_2 – 4300 l/s) and helium cryogenic pumps (3) Helix Technology Cryo-Torr 8 (4500 l/s), equipped with vacuum shutter valves HTC GVBB-SS-ISO 200 and 250 and pneumatic valves HTC AVBS-KF50. Forevacuum is provided by pumps (4) Anest Iwata ISP-500B and Edwards XDS35i (total up to $150 \text{ m}^3/\text{h}$).

The residual gas pressure in the expansion chamber P_∞ can vary in the range of $10^{-5} - 10^{-1}$ mbar: measurements in the range of $10^{-4} - 10^{-1}$ mbar are performed by a capacitance vacuum gauge (5) Agilent Technologies CDG-500; measurements in the range of $10^{-5} - 10^{-4}$ mbar are performed by an ionization vacuum gauge (6) Granville-Phillips 356 Micro-Ion Plus. The residual gas temperature in the expansion chamber T_∞ is measured on the chamber wall using a temperature controller (7) Ketotek STC-3008.

The pre-chamber with the installed nozzle block (8) is placed inside

the expansion chamber on a 4-component coordinate mechanism (9) constructed from mechanical positioning components manufactured by LinTech (movement along orthogonal directions – 250-150-150 mm, horizontal rotation $\pm 90^\circ$). The nozzle pre-chamber is connected through an electromagnetic valve Camozzi A321 and gas reducers GCE DIN 200/30 bar, GCE DIN 200/10 bar, PR1-1F11A3G111 250/7 bar to a balloon of pure gas (helium, neon, argon, nitrogen, oxygen, carbon dioxide, methane, sulfur hexafluoride, etc., – impurity content not more than 0.005%) or a gas mixture of known composition. The pressure in the pre-chamber P_0 , in the range 0.03–10 bar, is measured by an absolute pressure transducer (10) Siemens Sitrans P7MF1564; in the range 0.01–1.0 bar – by a vacuum gauge Vacuubrand Vacuu View (installed instead of the absolute pressure transducer). The gas flow is controlled by a regulator (11) Eltochpribor RRG-12 (maximum absolute pressure – 4 bar).

The nozzle pre-chamber, thermally insulated from the coordinate mechanism, can be connected to a heating/cooling module (12) (stagnation temperature T_0 variation – from 260 to 390 K), consisting of a thermoelectric converter TEC1-1206, copper radiator, and cryostatting system LOIP FT211-25. The temperature T_0 is measured on the pre-chamber wall using the second channel of the temperature controller Ketotek STC-3008.

When conducting measurements in pulsed modes, the system is equipped with a pulsed second valve Gems Sensors & Controls A2013 S197 VAC (response time – 9 ms, flow orifice 4.23 mm, operating pressure 0–20.7 bar, operating temperature 270–350 K) or a sub-millisecond electromagnetic valve by Institute of Nuclear Physics SB RAS (response time – 0.15 ms, flow orifices from 0.2 to 1 mm, operating pressure 0–10 bar, operating temperature up to 340 K) [11,41,42].

The high pumping capacities of the LEMPUS-2 vacuum facility allow the use of both single sonic and supersonic (conical or profiled) nozzles, as well as block-nozzle compositions [43]. The parameters of the nozzles used in this work are presented in Table 1.

The electron beam source (13) forming the diagnostic beam is installed in the expansion chamber:

- electron source with hollow cathode manufactured by Elion LLC {now the manufacturer is called TETA LLC} (electron energy up to 10 keV, current up to 100 mA) with a ballast volume evacuated by a turbomolecular pump Shimadzu TMP 303 (320 l/s);
- electron source with heated cathode (lanthanum hexaboride) manufactured by Sumy Electron Optics (electron energy from 30 to 60 keV, current up to 1 A) with a ballast volume evacuated by two turbomolecular pumps Shimadzu TMP 303 (640 l/s).

The output current value of the voltage source (power supply) is set using specialized software on a PC. The multimeter (14) Victor VC97 measures the current of electrons reaching the collector (15) (area $\sim 20 \text{ cm}^2$) in the lower part of the expansion chamber.

A nozzle with an internal ionizer (electron energy in the discharge up

Table 1

The parameters of the nozzles involved in the work: d_s – throat (critical) diameter; d_a – exit diameter; L_n – diverging section length; α_n – half angle of the diverging section.

N ^o	type	d_s , mm	d_a , mm	L_n , mm	α_n , °
1	sonic	0.22	-	-	-
2	sonic	0.24	-	-	-
3	sonic	0.51	-	-	-
4	sonic	1.00	-	-	-
5	sonic	1.12	-	-	-
6	supersonic	0.18	2.55	8.00	8.4
7	supersonic	0.24	1.55	3.00	12.3
8	supersonic	0.26	1.55	3.55	10.3
9	supersonic	0.36	1.30	3.00	8.9
10	supersonic	0.52	3.24	6.00	12.8
11	supersonic	1.00	3.60	6.00	12.2

to 1 keV, current up to 30 mA) can be installed in the prechamber [44].

An optical window (16) with a diameter of 160 mm made of quartz (transmission range – 200–1250 nm, light transmittance – more than 90%) or sapphire monocrystal (transmission range – 0.6–4.0 μm, light transmittance – more than 90%) can be installed in the shell of the expansion chamber perpendicular to its longitudinal axis.

The registration system of the initiated radiation can consist of (see Fig. 1):

- Camera (17) Nikon D7200 with lens Nikon AF-S DX Nikkor 18–300 mm f/3.5–6.3G ED VR or PCO Panda 4.2 bi UV with lens Goyo Optical GMUV57838C-1 (Goyo Optical GMUV42528C) with a selected bandpass filter MidOpt BP Series [45];
- Quartz short-focus lens (18), slit diaphragm (19) and scanner (20) VMK-Optoelectronics MASI-USB with a narrow elongated matrix [46];
- Quartz short-focus lens, waveguide (21) and spectrometer (22) Ocean Optics USB4000-UV-VIS, Ocean Optics HR4000 or DFS-452 with a digital atomic spectrum analyzer module produced by CJSC “Spectral Laboratory” [45];
- Quartz short-focus lens, photomultiplier tube (23) PMT-92 and analog-to-digital converter (24) R-Technology USB-3000 [11].

To exclude interference from external radiation sources, the registration system is protected with light-shielding material. To exclude registration of reflected radiation, the internal walls of the expansion chamber in the observation area are covered with black matte material.

On the end flange of the expansion chamber there is a mount for installing a cone-shaped hollow diaphragm – skimmer (25). Behind the skimmer is located a two-section module of the molecular beam system consisting of post-skimmer (26) and detector (27) sections, separated from each other by a shutter (28) HTC GVB-SS-CF100-P. Between the post-skimmer and detector sections in front of the shutter coaxially with the skimmer, a replaceable collimating diaphragm (29) is installed.

The post-skimmer section is a cylinder with a diameter of 260 mm and length of 325 mm ($\sim 0.02 \text{ m}^3$). The pressure in the post-skimmer section is maintained in the range from 10^{-7} to 10^{-4} mbar by turbomolecular pumps: two Shimadzu TMP 1003LM and one Shimadzu TMP 803LM (total capacity $\sim 3000 \text{ l/s}$).

The detector section is a cylinder with a diameter of 200 mm and length of 600 mm ($\sim 0.02 \text{ m}^3$) equipped with nozzles with CF100 flanges sealed with copper gaskets. The pressure in the detector section is maintained in the range from 10^{-9} to 10^{-6} mbar by a turbomolecular pump Shimadzu TMP-303LM (320 l/s) and a heteroionic pump Thermionics IP-025 (30) ($\sim 25 \text{ l/s}$). The pressure in the post-skimmer P_{ps} and detector P_d sections is measured by ionization vacuum gauges Granville-Phillips 356 Micro-Ion Plus.

In the post-skimmer section, on a single-component coordinate device (movement perpendicular to the molecular beam axis – 150 mm, positioning accuracy – 0.5 mm), a removable intensity sensor (31) with a 2 mm diaphragm is installed, created based on the Granville-Phillips 356 Micro-Ion Plus sensor. Information from the measuring device is transmitted to a PC through an analog-to-digital converter R-Technology USB-3000 (32).

In the detector section, a mass spectrometer (33) suitable for research tasks is installed: Hiden Analytical EPIC1000 (1–1000 amu) or ExTorr RGA300 (1–300 amu).

The molecular beam system features a complex for transporting ions formed in the expansion chamber to the mass spectrometer detector, based on applied potentials to electrically isolated skimmer and collimating diaphragm.

The expansion chamber and post-skimmer section are equipped with atmosphere composition variation modules (34), consisting of a manual regulator (leaker) and an auxiliary gas balloon. The composition of the background environment is determined by the gas analyzer ExTorr RGA-300 (35).

It should also be noted that the LEMPUS-2 vacuum facility can be reconfigured for investigating liquid outflow into a rarefied environment (vacuum). For this purpose, a specialized module is connected to the nozzle block, which includes a liquid filling and degassing chamber, a pressure setting and control chamber, a working liquid temperature control chamber, and liquid supply control equipment. A more detailed description of these capabilities is provided in Refs. [47–50].

3. Rarefied gas flow research

There are many known diagnostic methods and approaches for studying gas flows: schlieren photography [51], Pitot tube [52], PIV [53], electric discharge tracing [54], LDA [55], CARS [56], OES [57], LIF [58], electron beam fluorescence technique (EBFT) [59,60], molecular beam diagnostics (MBD) [61,62], and others. However, the use of many of these methods in rarefied flows is very difficult or inaccessible due to the low particle density and its large gradient. Measurement techniques based on EBFT and MBD have been implemented at the LEMPUS-2 gas-dynamic facility for investigating supersonic rarefied flows. The following subsections present their capabilities, provide validation in non-condensing flows (i.e., at $\Gamma^* \leq 200$), and discuss the specific features of performing measurements with these techniques under conditions of advanced condensation.

3.1. Electron beam fluorescence technique

The electron beam fluorescence technique (EBFT) is based on passing a beam of free electrons through a gaseous object, followed by registration of the initiated radiation. Analysis of the beam-initiated radiation allows determining many properties of particles in the ground state (before interaction with the beam) if the excitation-radiation process path is known. This approach was proposed in the 1930s [60], further developed by various research groups [63–69] and is actively used nowadays [4,70].

3.1.1. Numerical density

According to Refs. [64,71], the radiation intensity I_{ij} of the observed transition from an upper level (i) to a lower (j) initiated by an electron beam is described by the dependence:

$$I_{ij} = k_E \cdot h \cdot c \cdot \nu_{ij} \cdot A_{ij} \cdot \rho_i \quad (3)$$

where c is the speed of light; h is Planck's constant; ν_{ij} is the transition frequency; A_{ij} is the spontaneous transition probability; $\rho_i \sim i_e \cdot \rho_{00} \cdot \sigma_i$ is the number of particles in the upper excited state; i_e is the electron beam current; σ_i is the cross-section for excitation of the upper state; ρ_{00} is the number of particles in the ground state. Considering the specifics of the experiment in a rarefied gas flow at room temperature, the work [72] made the assumption that expression (3) can be represented in form:

$$I_{ij} \sim i_e \cdot \rho \quad (4)$$

where ρ is the numerical density of gas particles. When $\rho > 10^{22} \text{ m}^{-3}$, the particle residence time in the excited state becomes longer than the time between collisions, resulting in the registered intensity being distorted by collisional quenching, described by the Stern-Volmer expression [73]:

$$\frac{I_{ij}}{I_{ij}^{\text{true}}} = \frac{1}{1 + \rho \tau_i K_q} \quad (5)$$

where I_{ij} is the registered radiation intensity corresponding to the $i \rightarrow j$ transition; I_{ij}^{true} is the intensity in the absence of quenching; τ_i is the excited state lifetime; $K_q = \sqrt{\frac{16k_B T}{\pi M_p}} Q_i(T)$ is the quenching constant; k_B is the Boltzmann constant; T is the temperature; M_p is the molar mass of a particle; $Q_i(T)$ is the total collision quenching cross-section for the upper level. For example [74], presents the value of the quenching constant K_q

for the $N_2^+ : B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^+$ transition: $K_q^{(N_2)} = 2.8 \cdot 10^{-10} \text{ cm}^3/\text{s}$. In Ref. [66] a dependence is presented that generalizes (3) and (5) as:

$$I_{ij} = \frac{k_u \rho}{1 + k_q \rho \sigma_q(T) \left(\frac{8}{\pi} \right) R T^{1/2}} \quad (6)$$

where k_u (the ratio of the excitation rate, arising from primary and secondary electron inelastic collisions, to the total spontaneous decay rate for the upper level) and k_q (the ratio of the rate of depopulation, due to atomic or molecular collisions, to the total spontaneous decay rate for the upper level) are constants; R is the molar (universal) gas constant.

To verify the correctness of the adaptation of the traditional method, measurements were carried out under conditions of negligible condensation. Fig. 2 shows an example of using the described method for nitrogen flow from sonic nozzles N^o 4 and 5 (see Table 1) compared with the isentropic distribution (see Appendix A) [75] and numerical simulation (see Appendix B) [18,21] in the modes whose parameters are indicated in the caption.

Similarly [75], the data are presented as a function of the reduced density value ρ/ρ_0 (ρ is the local numerical particle density, ρ_0 is the particle density in the nozzle pre-chamber) versus dimensionless calibers x/d_s (x is the coordinate, d_s is the diameter of the critical/exit section of the sonic nozzle). Radiation intensity registration, according to Ref. [64], recommendations, is performed in the region of “fast” transitions (on the electron beam). For registering I_{ij} intensity, the LEMPUS-2 facility uses an Ocean Optics USB4000-UV-VIS or Ocean Optics HR4000 spectrometer (a). Additionally, it was shown that when conducting similar measurements in pulsed modes, using the integral radiation intensity in a wide spectral range is possible to obtain quality estimates [11]. This allows using PMT-92 data during measurements in pulsed flows (b). For calibrating the radiation intensity values registered in the flow, data obtained in the background environment at a distance from the gas source are used [45].

From the obtained experimental dependencies, it is evident that within the undisturbed jet core, gas expansion occurs according to the isentropic law, which agrees with [76]. The deviation of registered experimental data from the isentropic curve [75] in the far flow field can be explained by the formation of shock-wave structures and the Mach disk, and the density reaching the background environment level due to expansion into a region with finite background pressure. It should be noted that when using PMT, accounting for collisional fluorescence quenching is impossible due to integral intensity registration, which is why experimental data in the near flow field and in the Mach disk region may be distorted. When comparing experimental and calculated data [18], some differences are observed in the internal structure of the Mach disk, however, the position of the Mach disk is predicted quite accurately. Summarizing, the experimental results obtained using the presented technique (taking into account the measurement uncertainty of 10–12%) demonstrate good agreement with available literature data and

numerical models, and therefore can be used for the analysis of various flows.

It was noted in Refs. [2,36] that the development of the condensation process can lead to a distortion of flow isentropicity and, consequently, to a change in the density distribution within the flow. Fig. 3 presents the measured relative density ρ/ρ_0 as a function of the average cluster size $\langle S \rangle$ formed in the flow, determined according to Ref. [33]. All values were recorded at a relative distance of $x/d_{eq} = 30$ downstream of various supersonic nozzles (see Table 1). It is evident that under conditions of low or no condensation, i.e., at $\langle S \rangle < 50$, all relative density values are identical and correspond to the isentropic distribution [77] (see Appendix A). However, as the condensation process develops (with an increase in the condensate fraction and average cluster size), the centerline density decreases. On one hand, this can be explained by the fact that the heat of condensation released into the supersonic flow leads to the expansion of the stream tube, which results in a decrease in relative density and an increase in the characteristic jet expansion angle [2,36]. At the same time, the issue of electronic excitation of clusters remains unresolved: due to intracluster relaxation [78,79], radiation may be observed outside the detected region of the electron beam; furthermore, the excitation/ionization cross-sections of particles within a cluster may differ significantly from those for monomers [80–82]. These aspects may cause EBFT-based measurements to show underestimated relative density values that do not reflect the actual distribution. However, introducing appropriate corrections to account for these factors is only possible by investigating the density distribution using alternative methods (including numerical approaches).

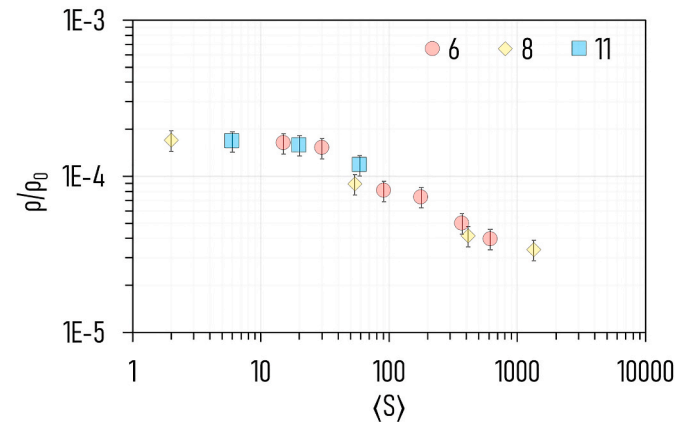


Fig. 3. Dependence of the relative density recorded at $x/d_{eq} = 30$ in argon flows behind supersonic nozzles (Table 1) on the average size of the formed clusters [33].

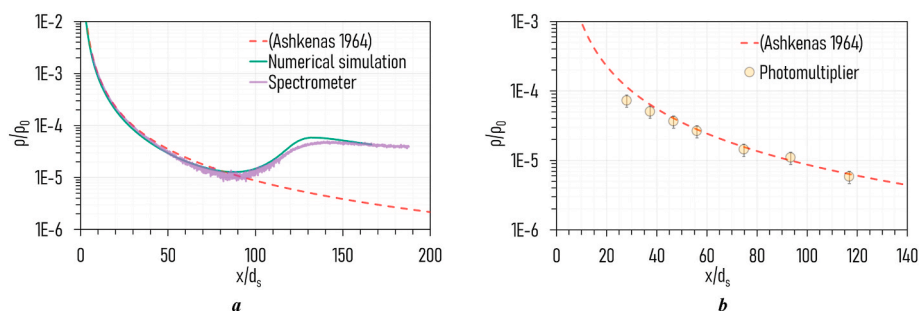
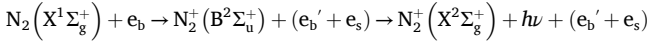


Fig. 2. Results of measuring the number density in a nitrogen flow using a spectrometer (a) [18] and photomultiplier (b) [11] in comparison with the results of numerical calculation [18] and the idealized isentropic curve [75].

a – nozzle N^o5 (Table 1), $P_0 = 0.30 \text{ bar}$, $T_0 = 310 \text{ K}$, $P_\infty = 10.1 \text{ } \mu\text{bar}$, $T_\infty = 297 \text{ K}$;
b – nozzle N^o4 (Table 1), $P_0 = 7.00 \text{ bar}$, $T_0 = 300 \text{ K}$, $P_\infty = < 4.0 \text{ } \mu\text{bar}$, $T_\infty = 298 \text{ K}$.

3.1.2. Rotational temperature

In case of supersonic outflow of gas into a rarefied medium, a sufficient number of paired collisions of molecules (10^1 – 10^2) can be maintained for a long time to establish thermodynamic equilibrium between rotational and translational degrees of freedom, which is why the rotational temperature in these flow conditions fully corresponds to the translational one [83]. Therefore, one of the common methods for determining local temperature in a supersonic flow of rarefied gases is based on EBFT. Let's examine the fundamentals of this method using molecular nitrogen as an example. According to Refs. [65,84], the rotational temperature T_{rot} , included in the Boltzmann distribution function, is determined by the population of rotational levels. In the particular case of the transition used in this work during the interaction of nitrogen molecules with beam electrons, the excitation-emission process shown in Fig. 4 can be described as follows:



where e_b – electron of the diagnostic beam; e_s – secondary electron; $h\nu$ – quantum of light. The excitation-emission process is assumed to be direct (collision processes are not taken into account, $\rho < 10^{22} \text{ m}^{-3}$).

Assuming that the selection rules apply to excitations from $\text{N}_2(\text{X}^1\Sigma_g^+)$ to $\text{N}_2^+(\text{B}^2\Sigma_u^+)$, ionization of the molecule results in a spin change and the formation of P- and R-branches. The intensity distribution in the spectrum of the band (0-0) of the first negative system can be described by the expression [59,65,84,85]:

$$\begin{aligned} -\ln\left(\frac{I_{K'K''}}{I_{\text{ref}}}\right) \frac{1}{(K' + K'' + 1) \left\{ \frac{G_{K'K''} \nu_{K'K''}^4}{[G_{\text{ref}}] \nu_{\text{ref}}^4} \right\}} \right) &= \frac{B_{\nu_1} K'(K' + 1) hc}{k_B T_{\text{rot}}^*} \\ + \frac{\text{const}}{I_{\text{ref}}} G[K', T_{\text{rot}}^*] &= \frac{K' + 1}{2K' + 1} \exp\left\{ \frac{-2B_{\nu_1} (K' + 1) hc}{k_B T_{\text{rot}}^*} \right\} \\ + \frac{K'}{2K' + 1} \exp\left\{ \frac{2B_{\nu_1} K' hc}{k_B T_{\text{rot}}^*} \right\}, \end{aligned} \quad (7)$$

where $I_{K'K''}$ is the intensity of the R-branch lines of the band (0-0) of the first negative system; K' , K'' is the quantum numbers of rotational levels; I_{ref} is the normalization factor (R1 intensity); B_{ν_1} is the rotational constant; ν_1 – transition wave number; T_{rot}^* is the rotational temperature; $G[K', T_{\text{rot}}^*]$ is a function that takes into account the rearrangement of rotational states during the initial excitation of the molecule. Due to the nuclear spin of the molecule, the populations of even and odd rotational levels differ by a factor of two. In Ref. [86] the combination $\theta_{\text{rot}} = B_{\nu_1} hc / k_B$ is called the characteristic rotational temperature of the molecule. For the nitrogen $\theta_{\text{rot}} \approx 2.878 \text{ K}$.

It should be noted that under higher density conditions, a distortion of the Boltzmann plot and an overestimation of the measured temperature are observed due to the appearance of secondary electrons in the measurement region [87,88]. To minimize this effect, it is necessary to use a diaphragm that localizes the measurements in the central part of the electron beam, where the contribution of slow electrons is minimal.

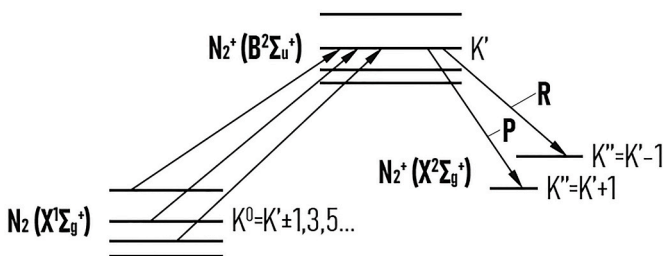


Fig. 4. Schematic representation of the excitation-emission process in a nitrogen molecule.

There are several approaches for obtaining the initial value of T_{rot} for a neutral nitrogen molecule. In Refs. [65,84] an iterative method based on equation (7) was demonstrated: the graph of dependence –

$$\ln\left(\frac{I_{K'K''}}{I_{\text{ref}}}\right) \frac{1}{(K' + K'' + 1) \left\{ \frac{G_{K'K''} \nu_{K'K''}^4}{[G_{\text{ref}}] \nu_{\text{ref}}^4} \right\}} \right) = \xi\{I_{K'K''}\} \quad \text{versus} \quad K'(K' + 1)$$

(Boltzmann plot) represents a straight line, the slope of which corresponds to the rotational temperature. In Refs. [86,89–91], a method based on comparison of calculated and experimental radiation spectra was used. Spectrum calculation and accounting for instrumental function are based on the model presented, for example, in Refs. [92,93].

According to Ref. [94], during inelastic collision, beam electrons transfer additional rotational energy to the molecule ($T_{\text{rot}}^* - T_{\text{rot}}$), equal to:

$$\Delta\varepsilon \sim \sum_K P_{K0} [E_b] K'(K' + 1) \quad (8)$$

where P_{K0} is the vector specifying transition probabilities from the zero rotational level of the ground state to the rotational level K' of the excited state during interaction with beam electrons with energy E_b . In the adiabatic approximation, the value $\Delta\varepsilon$ does not depend on the rotational distribution of molecules in the initial state but has dependence on the value of E_b . Thus, for $E_b = 10$ – 20 keV , the heating $\Delta\varepsilon$ is estimated as $(12.7 \pm 0.3) \text{ K}$, which must be taken into account when calculating the final value of T_{rot} for molecules in the ground state.

In [65] the uncertainty of T_{rot} measurements performed by this method is justified primarily by the accuracy of the assumption about the applicability of selection rules and rotational transition probabilities to the excitation transition. In turn, in Ref. [85], it was shown that the measurement uncertainty has dependence on the value of T_{rot} : at room temperature – less than 10%, at low temperatures – increases up to 30%.

Fig. 5 presents an example of registration of the (0-0) band of the transition $\text{N}_2^+(\text{B}^2\Sigma_u^+) \rightarrow \text{X}^2\Sigma_g^+$ in different modes: 1 – background environment, 2 – rarefied jet, 3 – flooded jet. In mode 1, most of the rotational levels are populated, and the slope of the Boltzmann plot corresponds to $T_{\text{rot}}^{(1)} = 301 \text{ K}$, which agrees well with the results of measurement by the Ketotek STC-3008 sensor on the expansion chamber wall (298 K). In mode 2, only the first rotational levels are populated, and the slope of the Boltzmann plot corresponds to $T_{\text{rot}}^{(2)} = 14 \text{ K}$, which within the specified error can be correlated with the isentropic estimate of translational temperature (10.9 K, see Appendix A) and numerical calculation of rotational temperature (12.8 K, see Appendix B) [18,21]. In turn, mode 3 demonstrates conditions of background gas penetration into the jet, as a result of which, in the first approximation, the radiation spectrum can be considered bimodal: the first mode – cold jet particles ($T_{\text{rot}}^{(3)} = 11 \text{ K}$), the second – hot particles of the background environment that have penetrated inside the jet ($T_{\text{rot}}^{(H)} = 298 \text{ K}$). Separating one mode from another can make it possible not only to determine the value of the rotational temperature, but also to estimate the proportion of background gas penetrating into the flow. For example, in mode 3 shown in Fig. 5, the penetration fraction does not exceed 9%, which is consistent with the results [95]. It should be noted that using this technique, the influence of turbulence on cluster formation in the flow was discovered [96].

Since no argon transition lines are observed in the spectral range of the (0-0) band of the $\text{N}_2^+(\text{B}^2\Sigma_u^+) \rightarrow \text{X}^2\Sigma_g^+$ transition (3890 – 3915 Å), it is possible to measure the temperature in argon by adding molecular nitrogen as a small impurity. Based on the nitrogen emission and assuming thermodynamic equilibrium, the translational temperature of the carrier gas can be determined [97,98]. Of particular interest is the study of non-equilibrium effects in supersonic flows with advanced condensation. Fig. 6 presents the results of temperature measurements in an argon flow downstream of supersonic nozzle N26 (Table 1). The experimental

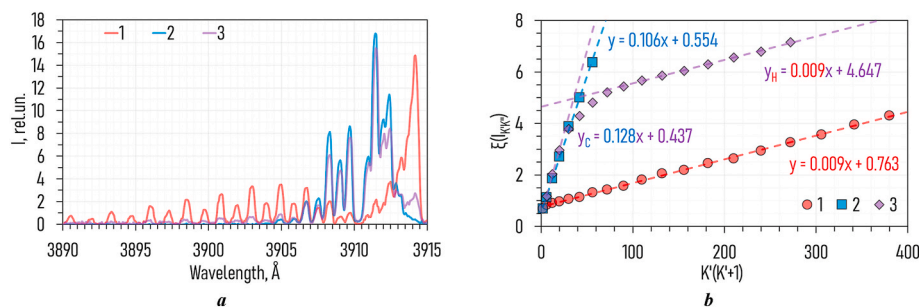


Fig. 5. Results of rotational temperature measurement: *a* – molecular spectrum; *b* – Boltzmann plot. Initial parameters: 1 – background gas, $P_\infty = 64.5 \mu\text{bar}$, $T_\infty = 298 \text{ K}$ (on the wall of the expansion chamber); 2 – rarefied jet, nozzle N^o 3 (Table 1), $P_0 = 0.5 \text{ bar}$, $T_0 = 309 \text{ K}$, $P_\infty = 7.6 \mu\text{bar}$, $T_\infty = 299 \text{ K}$, $x = 10 \text{ mm}$; 3 – flooded jet, nozzle N^o 3 (Table 1), $P_0 = 1.0 \text{ bar}$, $T_0 = 310 \text{ K}$, $P_\infty = 76.0 \mu\text{bar}$, $T_\infty = 297 \text{ K}$, $x = 15 \text{ mm}$.

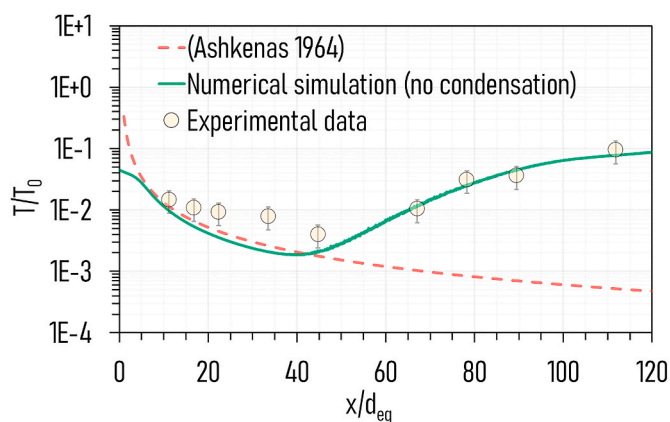


Fig. 6. Results of measuring the temperature in an argon flow with a small admixture of molecular nitrogen in comparison with the results of numerical calculation [18] and the idealized isentropic curve [75]. Initial parameters: nozzle N^o 6, $P_0 = 3.0 \text{ bar}$, $T_0 = 310 \text{ K}$, $P_\infty = 3.1 \mu\text{bar}$, $\Gamma^* = 3167$, $\langle S \rangle = 333$ [33].

data are compared with numerical simulation results neglecting condensation (see Appendix B) [18,21] and the isentropic curve (see Appendix A) [75].

Inside the jet core, numerical calculations correlate well with the isentropic curve, while the experimental data demonstrate overestimated values. It was noted in Refs. [97,98] that in the core of the jet, gas density and collision frequency fall very rapidly and, therefore, a change in internal energy does not manage to follow a reduction in the translational temperature; i.e., there is a delay in the relaxation process until it freezes at some distance from the nozzle edge. It should also be noted that the observed discrepancy may result from the presence of nitrogen in the argon flow (a 5–10% impurity has a slight effect on the effective γ value). At the same time, it was pointed out in Ref. [98] that for flows with stagnation temperatures near room temperature, the formation of van der Waals clusters must be taken into account in the rotational relaxation model. This process leads to the release of latent heat of condensation into the flow. To determine the magnitude of this heat (due to its smallness), a comprehensive study using numerical calculations and alternative measurement techniques is required. Furthermore, beyond the X-configuration, where cluster breakup occurs at the shock wave [99] and the temperature increases due to jet scattering by the background gas, satisfactory agreement between experimental data and numerical calculations is observed.

3.1.3. Visualization and jet dimensions

Registration of the initiated radiation allows not only qualitative analysis of density gradient zones [67], investigation of the flow shape and structure [64,100] (including complex configurations of supersonic

flows from multi-nozzle systems), and observation of kinetic processes of energy exchange [101], but also visualization of flow around various objects [69].

The first visualization method (*electron beam painting*) is based on recording primary emission (transitions from short-lived states with lifetimes of less than $2 \mu\text{s}$) in the region of its initiation (at the electron beam). This approach was previously employed, for example, in Refs. [59,100–103] using photographic film. At the LEMPUS-2 facility, the *electron beam painting* method is implemented using a linear CCD array that records the image of a stationary electron beam, while the nozzle plenum moves translationally orthogonal to the beam axis [46]. Synchronizing the signal integration time with the displacement coordinates allows for the acquisition of a two-dimensional intensity field of the radiation initiated by the electron beam. Replacing the photographic film used in earlier literature as a recording device with a CCD array has significantly expanded the data acquisition capabilities, both in terms of volume and the speed of information processing.

An example of nitrogen jet visualization flowing from sonic nozzle N^o 3 (see Table 1) with the formation of a direct shock wave (Mach disk), obtained using the *electron beam painting* method, is presented in Fig. 7 in comparison with the numerical simulation result [18,21] (see Appendix B). Due to the registration of fast transitions, it is possible to convert the two-dimensional intensity field into a density distribution [46]. This technique, for a reason the registration of integral intensity values, does not allow accounting for collisional fluorescence quenching, however, the comparison of experimental and numerical data demonstrates good agreement in the structure and dimensions of the jet, which may indicate the correctness of the approaches used (on both sides).

The second visualization method (*afterglow registration*), implemented in Refs. [4,59,67,102,104–106], including on the stand LEMPUS-2 in Refs. [45,107], is based on photo-registration of afterglow (radiation observed outside the excitation region due to the drift of excited particles downstream, scattering of the electron beam on background gas, presence of secondary electrons, etc.) outside the static focused electron beam. For the best display of the flow structure, during the post-processing stage, the photo is converted to grayscale, divided into areas with relatively small radiation intensity gradients, each of which is optimized for brightness, contrast, white balance, and depth of dark and light areas. The development of this well-known method at the LEMPUS-2 facility, through the use of windows with a wide transparency range, has enabled the recording of radiation in the visible, as well as in the near-IR and UV ranges. This has made it possible to investigate kinetic processes across a broad spectral range. An example of visualization of the flow regime presented in Fig. 6, obtained using *afterglow registration*, is shown in Fig. 8.

The *afterglow registration* method has some limitations.

- Firstly, the afterglow weakens with increasing stagnation temperature T_0 and, consequently, local temperature of the free flow [59].

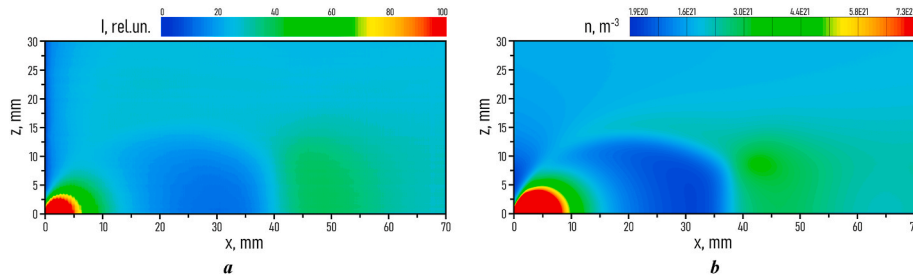


Fig. 7. Visualization of a nitrogen jet by *electron beam painting* (a) in comparison with the result of numerical simulation (b). Initial parameters: nozzle N° 3 (Table 1), $P_0 = 1.0$ bar, $T_0 = 310$ K, $P_\infty = 76.0$ μ bar, $T_\infty = 297$ K.

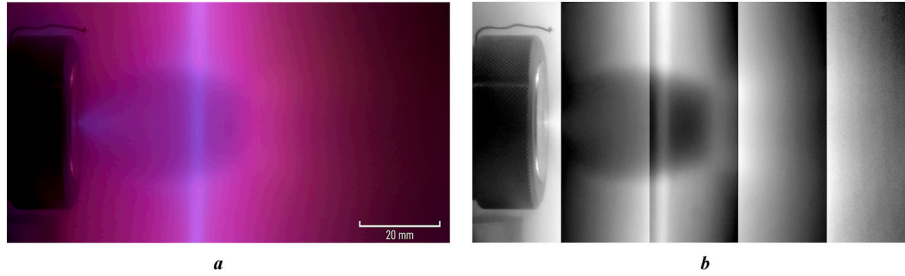


Fig. 8. Visualization of a nitrogen jet by *afterglow registration*. Initial parameters: identical to Fig. 7.

- Secondly, the method is applicable only to those gases in which afterglow is observed outside the diagnostic beam.
- Thirdly, the obtained images cannot be identified as numerical density values, since it is difficult to account for scattering and beam outflow processes, collisional quenching, energy exchange, etc., when registering radiation outside the excitation region.

At the same time, images obtained using *afterglow registration* method allow for rapid study of the shape and structure of the formed flows (the time for registration and processing of an image by *afterglow registration* is significantly less than the corresponding time for obtaining a two-dimensional field by *electron beam painting*), as well as for the study of molecular-kinetic processes of excitation energy exchange in the flow outside the excitation region (including flows with clusters).

Fig. 9 presents visualizations (using the *afterglow registration* method) of the argon jet under conditions of negligible (a) and developed (b) condensation. It can be seen that the traditional spindle-shaped “barrel” is formed in both flows. However, under conditions of developed condensation, a secondary structure of large dimensions is also observed, exhibiting weakly decaying afterglow when the radiation is initiated by a focused electron beam. [108–111]. It should be noted that, despite the differences in the configurations of the nozzles used, due to the used stagnation pressure P_0 values, the gas flow rate (and consequently the numerical density) in flow (a) turns out to be the same as in flow (b).

The nature of the afterglow arising under conditions of advanced condensation is of particular interest. Fig. 10 presents a flow

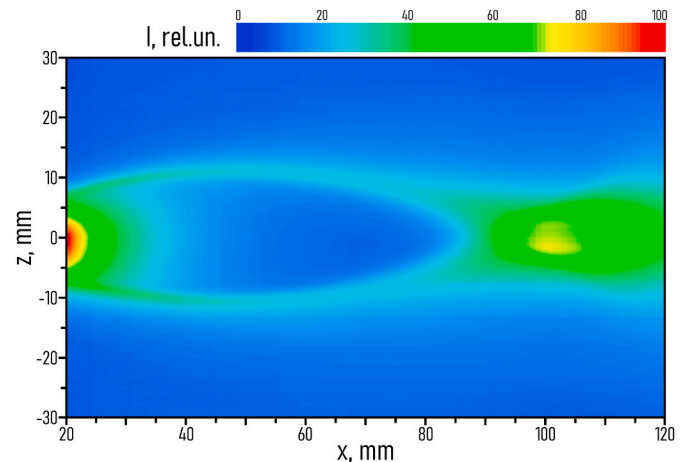


Fig. 10. Visualization of an argon jet by *electron beam painting*. Initial parameters: identical to Fig. 9b.

visualization obtained using the *electron beam painting* technique, with initial parameters identical to those shown in Fig. 9b. It is evident that the jet structures differ significantly: the *electron beam painting* visualization reveals only a spindle-shaped jet, similar to the one shown in Fig. 9a, whereas the visualization obtained via *afterglow registration* also shows a secondary structure with intense luminescence.

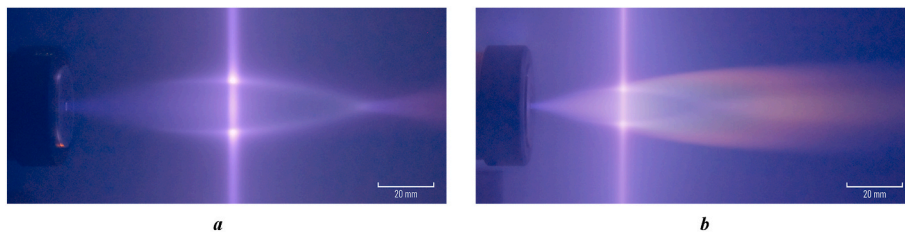


Fig. 9. Visualization by *afterglow registration* of an argon jet with low (a) and developed (b) condensation. Initial parameters: a – nozzle N° 11 (Table 1), $P_0 = 0.2$ bar, $T_0 = 308$ K, $P_\infty = 33.5$ μ bar, $\Gamma^* = 659$, $\langle S \rangle = 19$ [33]; b – nozzle N° 6, $P_0 = 6.0$ bar, $T_0 = 321$ K, $P_\infty = 46.8$ μ bar, $\Gamma^* = 5885$, $\langle S \rangle = 1038$ [33].

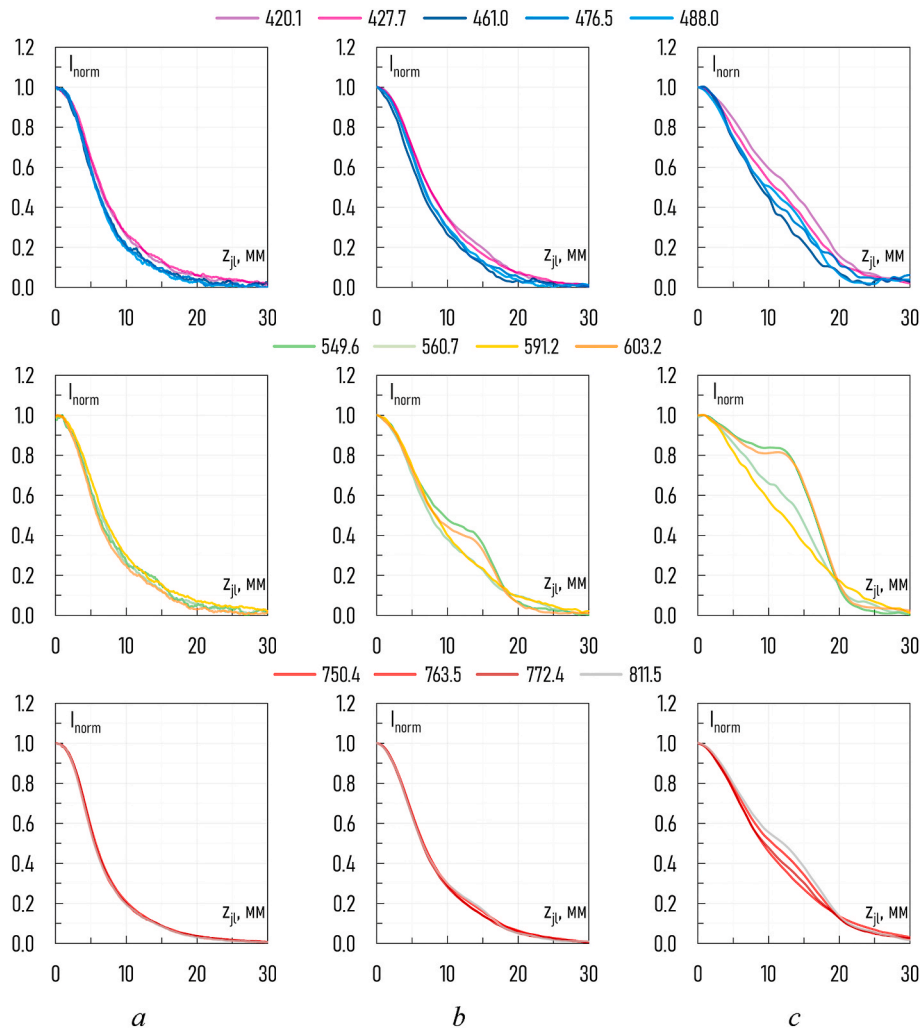


Fig. 11. Comparison of the emission intensity transverse profiles, normalized to the centerline, recorded in the argon flow with the electron beam positioned at a distance of 93.8 mm from the nozzle exit and the measurement location shifted downstream from the electron beam by 0 (a), 5 (b), and 15 (c) mm. Initial parameters: identical to Fig. 9b.

Fig. 11 presents the transverse profiles of the emission intensity, normalized to the centerline value, for various argon transitions (identified according to Ref. [112]). These profiles were recorded using a spectrometer. The electron beam was positioned in the region of secondary structure propagation, and the measurement location was shifted downstream by 0, 5, and 15 mm.

It was found that across the entire available wavelength range (280–900 nm), the intensity profiles of the radiation recorded in the region of direct flow excitation (a), i.e., in the region with short excited-state lifetimes, are identical and demonstrate a distribution similar to that obtained in a non-condensing flow. However, when the measurement region is shifted downstream from the initiating electron beam by 5 mm or more (b–c), the intensity profiles recorded for various argon transitions differ: radiation is detected in the secondary structure region at certain wavelengths, while it is absent at others. At the same time, a primary comparison of the tabulated characteristics of radiative transitions [112] did not reveal any systematic pattern for the observed effect. The results obtained indicate that particle emission outside the spindle-shaped structure is observed only at a distance from the excitation point (based on the electron beam dimensions and an estimate of the flow particle velocity, approximately 10 μ s). It should also be noted that the most intense luminescence in this region was recorded at wavelengths in the yellow-green part of the spectrum, 549.6 nm and 591.2 nm, which correspond to transitions of the $3s^23p^54p - 3s^23p^5nd$

series.

Since this secondary structure is observed only under conditions of developed condensation, we proposed the assumption that clusters formed in the flow are able to overcome rarefied hanging shocks and enter the mixing zone area. Visualization of this accompanying flow, called in Ref. [107] the “cluster wake”, becomes possible due to the peculiarities of energy exchange in clusters capable of retaining excitation due to intracluster relaxation [78,79]. The possibility of cluster concentration in the region of lateral hanging shocks and penetration through these shocks was confirmed in Refs. [99,113,114], and the detected anomaly in the radiation is in Refs. [115–118].

The possibility of installing a nozzle block in the nozzle pre-chamber also allows for the investigation of interaction features between cluster-forming jets under advanced condensation conditions, which are discussed in detail in Ref. [119]. Fig. 12 presents a comparison of visualizations for nitrogen binary jets under conditions of negligible condensation (a) and argon jets under advanced condensation (b) with identical initial parameters. In the far field of both jets, both the repulsion of the primary ‘barrels’ and their merging into a single flow are observed, which is consistent with, for example, [120]. At the same time, in argon (b), the process is complicated by the interaction of not only the primary ‘barrels’ but also the “cluster wake” structures: a low-divergence flow with non-uniform afterglow forms along the geometric axis between the primary jets. The increase in intensity can be

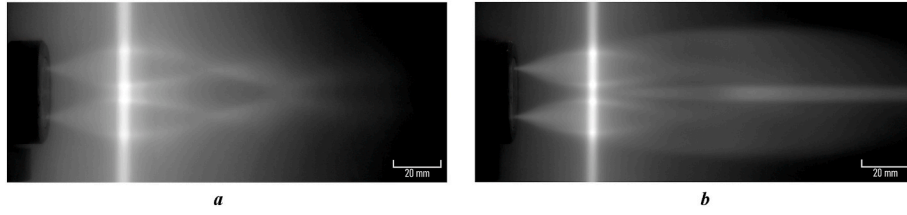


Fig. 12. Visualization by *afterglow registration* of nitrogen (a) and argon (b) binary-jet with low (a) and developed (b) condensation. Initial parameters: nozzle N^o 8 (Table 1), $P_0 = 2.0$ bar, $T_0 = 309$ K, $P_\infty = 26.7$ μ bar, $N^{0.5} = 273.9$, $\langle S \rangle = 5$ (a) and 284 (b) [33].

explained by the overlapping of two “cluster wakes” on the axis of the entire flow. It is also noteworthy that in Fig. 12, the dimensions of the primary jets (spindle-shaped “barrels”) are very similar, despite the difference in the gases used.

Using the *afterglow registration* image processing approach proposed in Refs. [121,122], it is possible to measure the longitudinal and transverse dimensions of the jet. To calibrate these measurements, reference images of objects with known dimensions (taking into account the viewing angle) are required. The longitudinal and transverse dimensions of the nozzle face, as well as the distance from the nozzle exit to the diagnostic electron beam, serve as such objects. Fig. 13 presents the results of measuring the position of the Mach disk or X-wave configuration x_m (a, c) and the barrel shock (including the compressed layer) at the maximum cross-section r_m (b, d) for a nitrogen jet exhausting from sonic and supersonic conical nozzles (Table 1) under conditions of negligible condensation. The results are approximated by

the following dependencies:

$x_m/d_s = k_1 \sqrt{N}$ [75],	a	(9)
$r_m/r_s = k_2 n^{0.5} - k_3 n^{0.25}$ [100],	b	
$x_m/d_a = k_4 \left(N/\bar{F} \right)^{0.5}$ [2],	c	
$r_m/r_a = k_4 \left(N/\bar{F} \right)^{0.5} \text{tg} \theta_m$ [2],	d	

where r_s and d_s are the radius and diameter of the throat (which is also the critical) section of the nozzle; r_a and d_a are the radius and diameter of the outlet (exit) section of the supersonic conical nozzle; $N = P_0/P_\infty$ is the pressure expansion ratio; $n = P_a/P_\infty$ is the degree of mismatch; P_a is the pressure in the nozzle exit section (see Appendix A); $\bar{F} = \frac{F_a}{F^*} = \left(\frac{d_a}{d^*} \right)^2$ is the area expansion ratio; F^* and F_a are the areas of the critical and exit sections of the nozzle; $\theta_m = \arctg \sqrt{\frac{1-J}{J}}$ is the characteristic jet expansion

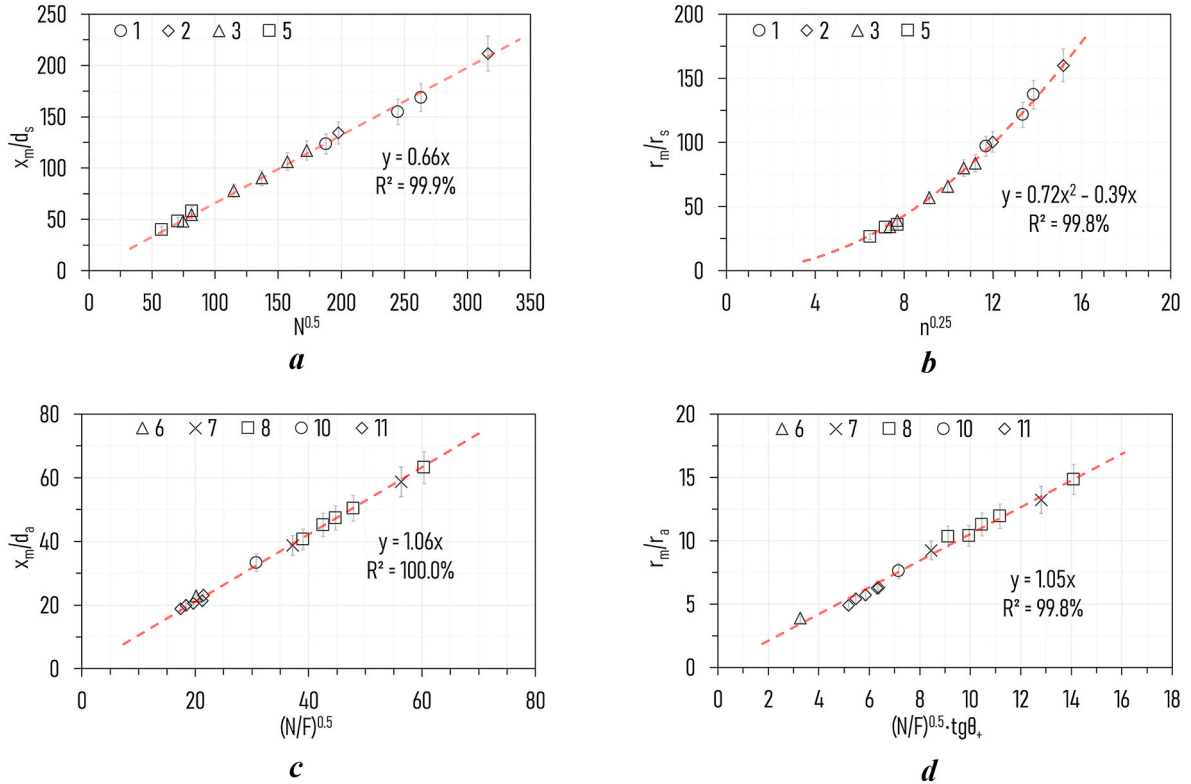


Fig. 13. Measurement results of the position of the direct shock wave (a, c) and the hanging shock in the maximum cross-section (b, d) in a nitrogen jet flowing out of sonic (a, b) and supersonic conical (c, d) nozzles (Table 1).

angle; $\bar{J} = \left(1 + \frac{1}{\gamma M_a^2}\right) \left(1 + \frac{2}{\gamma-1} \frac{1}{M_a^2}\right)^{-0.5}$ is the relative impulse of gas at the nozzle exit; $M_a = u_a/a_a$ is the Mach number at the nozzle exit; u_a and a_a are the directed (hydrodynamic) flow velocity and speed of sound in the nozzle exit; γ is the adiabatic index (ratio of specific heats); $k_1 \approx 0.66$, $k_2 \approx 0.72$, $k_3 \approx 0.39$ and $k_4 \approx 1.05$ are constant numerical coefficients (estimated measurement uncertainty – 8%). According to Refs. [75,100,123–126], coefficient k_1 lies in the range [0.64; 0.69], and in the review [127], data from different research groups are combined into dependence (9a) with coefficient $k_1 = 0.645497$. In Ref. [100], values of coefficients $k_2 = 0.725$ and $k_3 = 0.375$ were obtained. In Ref. [2], when studying carbon dioxide flow from supersonic conical nozzles, dependences (9c) and (9d) were obtained with coefficient $k_4 \approx 1.1$. At the same time, this paper presents the dependence of coefficient k_4 on the adiabatic index γ in the form:

$$k_4 \cong (2.4 \cdot C_1 \cdot C_2)^{0.5}, C_1 = \frac{1}{\sqrt{8}} \left(\frac{2}{\gamma+1} \right)^{\frac{\gamma+1}{2(\gamma-1)}}, C_2 = \frac{\gamma}{(\gamma-1)^{0.5}}, \quad (10)$$

which allows determining the value for nitrogen ($\gamma = 7/5$) $k_4 \approx 1.05$. Thus, it can be concluded that the measurement results obtained using the technique presented in Refs. [121,122] demonstrate good agreement with literature data. This enables its application in studies of complex flow patterns and under non-standard conditions (for example, when investigating the effects of condensation [119,122] or turbulence [96]).

It was noted in Ref. [2] that the formation of the condensed phase, due to the release of latent heat of condensation associated with the formation of van der Waals bonds, leads to an increase in the characteristic jet expansion angle; however, the nature of this broadening was not determined. Fig. 14a presents summarized results of the transverse dimension measurements in an argon flow with varying degrees of condensation. It is evident that the dependence (9d), verified for non-condensing flows using the geometrically calculated Mach number M_a , is unable to describe the data under conditions of advanced condensation, as the relative gas momentum at the nozzle exit $\bar{J}$ changes. The obtained experimental data allow for determining the dependence of the change in relative momentum $\Delta\bar{J} = \bar{J}_{ise} - \bar{J}_{exp}$ (where $\bar{J}_{ise}$ is the theoretical momentum value determined at equilibrium γ and M_a , and $\bar{J}_{exp}$ is the experimental momentum value calculated using the inverse of formula (9d)) on the average cluster size $\langle S \rangle$ determined by (2b) and (2d). The experimental data [45,121,122] obtained for various supersonic nozzles can be generalized as follows (Fig. 14b):

$$\Delta\bar{J} = \Delta\bar{J}_{max} \cdot \left(1 - \exp\left\{-\langle S \rangle / q\right\}\right) \quad (11)$$

where $q = 500$ is a constant coefficient; $\Delta\bar{J}_{max} \approx 0.07$ is the limiting value of $\Delta\bar{J}$. It can be seen that the data form a single curve: when $\langle S \rangle \rightarrow$

1 (in the absence of condensation), the value $\Delta\bar{J}$ is close to zero, and when $\langle S \rangle \rightarrow \infty$, the value $\Delta\bar{J}$ tends to a value $\Delta\bar{J}_{max}$ determined by the limitation of the condensate fraction in the jet. At the same time, it should be noted that cluster formation leads to the appearance of additional (averaged over volume) degrees of freedom of molecules, which causes a decrease in the effective value of the adiabatic index γ determining the value of k_4 in expression (9d). Therefore, in this model correction, it is assumed that the equilibrium value of γ “freezes” in the critical nozzle section, at which $k_4 = 0.987$ remains constant.

Thus, despite the inherent simplicity of the visualization methods, their application provides crucial information regarding flow parameters, including those under condensation conditions, and helps elucidate the observed phenomena.

3.2. Molecular-beam diagnostics

The MBD method is based on the formation of a free-molecular flow from a local region of a supersonic flow using a cone-shaped diaphragm (skimmer). The non-invasive nature of the technique is achieved by selecting an optimal combination of the skimmer diameter and its internal and external opening angles [128–130]. Recording the molecular beam parameters provides information about the state of the particles at the skimmer entrance, i.e., at the specific point of the gas flow under study. This approach was proposed in the middle of the last century [61], subsequently developed by numerous researchers (e.g., Refs. [128, 129,131–136]), and remains in active use today across various scientific fields [44,134,137–140]. Here, we consider only the methods of using molecular beams for the detection of supersonic jets.

3.2.1. Transverse profile of a molecular beam

According to Refs. [141–143], the particle density at an arbitrary point in the molecular beam can be expressed as:

$$\rho_b[\theta_a] = \frac{\rho(x)}{2\sqrt{\pi}} \frac{r_{sk}^2}{L^2} e^{-S_{\perp}^2/t^4} \left\{ 1 + \frac{\sqrt{\pi}}{2} \frac{1}{tS_{\parallel}} e^{\left(S_{\parallel}/t\right)^2} (2S_{\parallel}^2 + t^2) \left[1 + \Phi\left(\frac{S_{\parallel}}{t}\right) \right] \right\},$$

$$t = \sqrt{1 + \text{tg}^2\theta_a S_{\perp}^2/S_{\parallel}^2}, \quad (12)$$

where x is the distance from the point source to the entrance section of a skimmer with radius r_{sk} ; L is the distance from the skimmer to the detection plane; θ_a is the angle between the molecular beam centerline and the line connecting the detection point to the center of the skimmer entrance section; Φ is the Fresnel integral; $S_{\parallel} = W/\sqrt{2k_B T_{\parallel}/m}$ and $S_{\perp} = W/\sqrt{2k_B T_{\perp}/m}$ are the parallel and perpendicular speed ratios, respectively; and W is the average particle velocity along the streamline.

Equation (12) was obtained by integrating over angles and velocities,

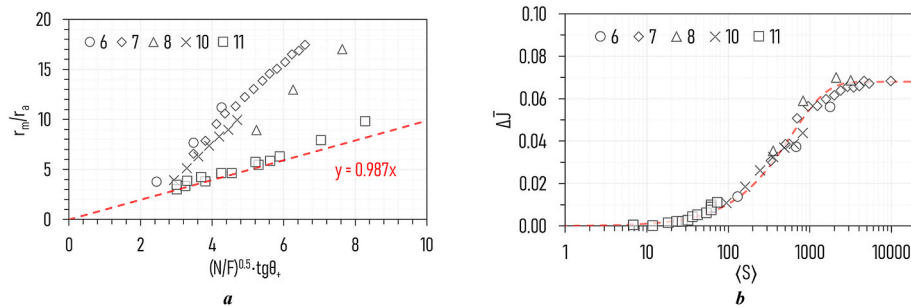


Fig. 14. Measurement results of the position of the hanging shock wave in the maximum cross-section of an argon jet flowing out of supersonic conical nozzles (Table 1) under condensation conditions. The experimental data are presented as a dependence of the reduced radius r_m/r_a on the parameter $\left(N/F\right)^{0.5} \text{tg}\theta_m$ (a) and the change in the relative momentum of the gas at the nozzle exit $\Delta\bar{J}$ on the average cluster size $\langle S \rangle$ (b).

assuming a Maxwellian velocity distribution and under the conditions $(r_{sk}^2/x^2) \ll 4\pi$ and $(r_{sk}^2/L^2) \ll 4\pi$.

In the case where $S_{\perp} = S_{\parallel} = S$ and using the hypersonic approximation ($S > 3$), expression (12) reduces to the form given in Refs. [144, 145]. By normalizing (12) to the corresponding centerline value of the beam, the following can be obtained:

$$S_{\perp}^2 = \left(\frac{1}{\text{tg}^2\theta_a} + \frac{S_{\parallel}^2}{S_{\perp}^2} \right) \left\{ \ln \left[\frac{1}{\tilde{\rho}_b[\theta_a]} \right] + \ln \left[\frac{1}{\left(1 + \text{tg}^2\theta_a S_{\parallel}^2 / S_{\perp}^2 \right)^{5/2}} \right] \right. \\ \left. + \ln \left[1 + \frac{\text{tg}^2\theta_a S_{\parallel}^2 / S_{\perp}^2}{2S_{\parallel}^2 + 1} \right] \right\} \quad (13)$$

where $\tilde{\rho}_b[\theta_a] = \rho_b[\theta_a]/\rho_b[0]$; $\rho_b[0]$ is the particle density on the molecular beam axis. In the case where $S_{\perp}$ and $S_{\parallel}$ are of the same order of magnitude and the angle θ_a is small, the value $\text{tg}^2\theta_a S_{\parallel}^2 / S_{\perp}^2 \ll 1$, and expression (13) reduces to the simplest form:

$$S_{\perp}^2 = \frac{1}{\text{tg}^2\theta_a} \ln \left\{ \frac{1}{\tilde{\rho}_b[\theta_a]} \right\} \quad (14)$$

Given a known W (for instance, the limiting velocity in the flow [146]), formula (14) can be used to determine $S_{\perp}$ through either density or intensity measurements ($I_b = \rho_b \cdot W$). If W depends only weakly on θ_a , its determination is not required. Accordingly, for intensity measurements, formula (14) takes the following form:

$$S_{\perp}^2 = \frac{1}{\text{tg}^2\theta_a} \ln \left\{ \frac{1}{\tilde{I}_b[\theta_a]} \right\} \quad (15)$$

Fig. 15 presents the registered transverse profile of the reduced molecular beam intensity $I_b^{(n)}$ in comparison with the approximation by dependence (13) with the isentropic value $S_{\perp} \approx S = M \cdot (\gamma/2)^{1/2} = 16.4$ (see Appendix A).

Thus, by measuring the transverse profiles of the molecular beam density (or intensity), it is possible to determine the perpendicular speed ratio. Furthermore, in the absence of translational temperature freezing, condensation, skimmer interaction, and background gas penetration, the translational temperature of the flow can also be determined. Fig. 16 compares the measured values of $S_{\perp}$ in a nitrogen flow downstream of various sonic nozzles (Table 1) at different nozzle-skimmer distances with the isentropic values ($S_{\perp} \approx S = M \cdot (\gamma/2)^{1/2}$, Appendix A). Taking

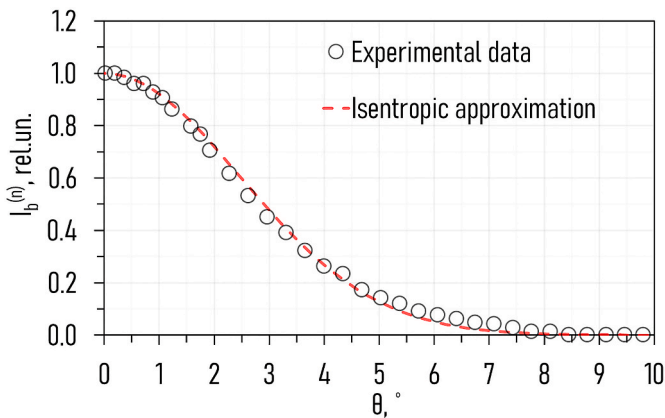


Fig. 15. Transverse profile of the molecular beam vs. dependence (13) with isentropic value $S_{\perp} \approx S = M \cdot (\gamma/2)^{1/2} = 16.4$. Initial parameters: nozzle N^o 3 (Table 1), $P_0 = 0.5$ bar, $T_0 = 299$ K, $P_{\infty} = 26.7$ μ bar, $x/d_s = 49.5$.

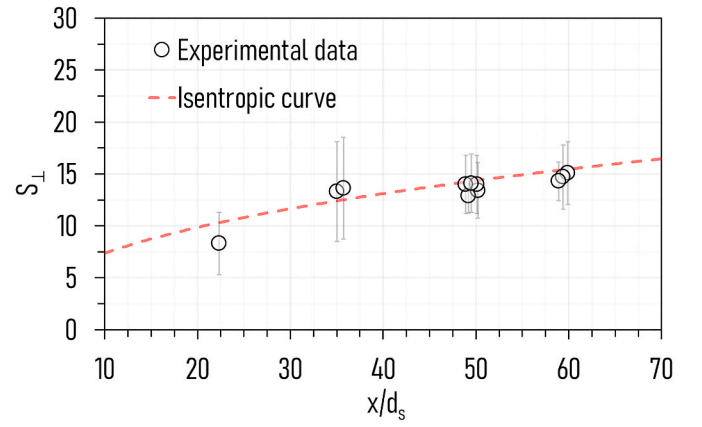


Fig. 16. Experimental data of measurement $S_{\perp}$ in jet behind sonic nozzles N^o 2, 3 and 5 (Table 1) in comparison with the idealized isentropic curve [75] in the sentence $S_{\perp} \approx M \cdot (\gamma/2)^{1/2}$.

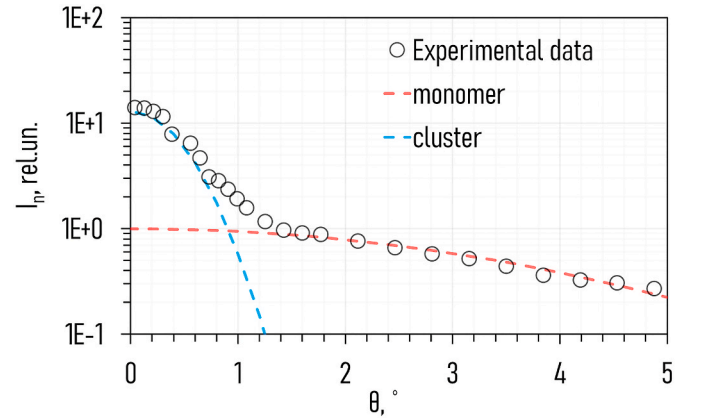


Fig. 17. Transverse intensity profile recorded in a clustered molecular beam with the two-mode approximation. Initial parameters: nozzle N^o 8 (Table 1), $P_0 = 3.0$ bar, $T_0 = 296$ K, $x = 80$ mm, $\Gamma^* = 2723$, $\langle S \rangle = 252$ [33].

into account the experimental error, the results obtained at various initial stagnation parameters are in good agreement with literature data.

The possibility of using this method for measurements in condensing flows is of particular interest. In this case, gas-dynamic particle separation must be taken into account. Based on the results in Ref. [147], it can be assumed that under supersonic rarefied flow conditions, the velocity difference between monomers and clusters (velocity slip) does not exceed 5–7%. Since the speed ratio is proportional to the square root of the particle mass, the $S_{\perp}$ value for clusters is significantly higher than for monomers; as a result, according to (12), the divergence of clusters in the molecular beam is substantially lower. Fig. 17 presents the recorded transverse intensity profile of an argon molecular beam under advanced condensation conditions. It is evident that the resulting distribution is non-uniform: a significant increase in signal intensity (by more than an order of magnitude) is observed near the molecular beam axis. This is explained by the fact that clusters in a stagnation-type (closed-type) detector fragment into monomers due to collisions with the walls, leading to a strong surge in the recorded signal caused by the concentration of large clusters near the axis. Thus, the transverse spatial distribution is a complex function consisting of several components. For instance, it was suggested in Ref. [139] that the function is bimodal (i.e., consists of “cluster” and “cluster” components); from recorded profiles similar to the one shown in Fig. 17, the authors determined the average cluster size in the “cluster” component. However, it should be noted that this approach neglects the contribution of small clusters to the

“monomer” component and the temperature difference between clusters and monomers, which makes it a zero-order approximation model.

3.2.2. Mass spectrometry

Mass spectrometry of physicochemical processes requires low background pressure in the mass spectrometer detector region. This was made possible by using a multi-chamber molecular beam system with differential high-vacuum pumping for each individual section (see Paragraph 2). The pressure in the molecular beam system of LEMPUS-2 is maintained at the level of $10^{-8} - 10^{-4}$ mbar, which allows studying the mass distribution in the free molecular flow. The LEMPUS-2 facility implements mass spectrometric diagnostics with several options for particle ionization:

- particle ionization in the molecular beam using the mass spectrometer ionizer (the EBMS method);
- particle ionization in the jet using a high-voltage electron beam (the HVEB method) [148];
- particle ionization in the diffuser part of the nozzle using an electric discharge (the DIN method) [44].

The HVEB and DIN methods are applied with the mass spectrometer ionizer turned off and using the system for transporting the generated ions to the mass spectrometer detector. Since available programs for optimizing the motion of charged particles in electric or magnetic fields in regimes of flow with particles of different mass are not applicable [149,150], the optimality of ion selection conditions in the skimmer was verified experimentally [148].

It should be noted that when conducting measurements in flows of mixtures (or other systems with a wide mass distribution), according to (12), light particles in the molecular beam diverge at a larger solid angle than heavy ones, as a result of which the mass spectrum at the detection point will differ slightly from the mass spectrum at the location of the skimmer inlet. Also, it should be noted that in the mass spectrum (dependence of signal amplitude A on the ratio of atomic mass to charge M/z), in addition to singly ionized particles ($z = 1$), multiply ionized particles ($z \geq 2$) and dissociation/fragmentation products after interaction with the ionizer electrons are observed, which complicates the original task of studying the mass distribution of particles in the neutral flow.

Fig. 18 presents mass spectra registered in flows of pure gases (nitrogen – *a*, argon – *b*, carbon dioxide – *c*) under conditions of minimizing external factors (skimmer interaction, background gas penetration, condensation, etc.) during gas flow ionization in the mass spectrometer (EBMS method). The measurement results are plotted together with NIST data [151] obtained at similar electron energy of 70 eV. The comparison demonstrates satisfactory agreement with reference data.

In turn, the task of correctly registering the mass composition of a clustered flow is nontrivial, since characteristic binding energies in van

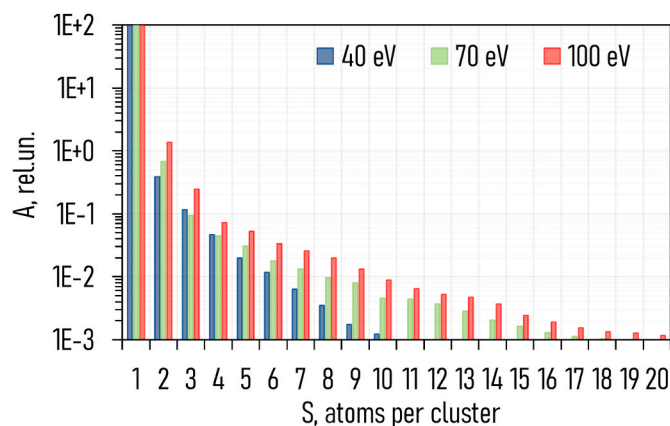


Fig. 19. Distribution of cluster size S recorded in an argon flow with variation of electron energy in ionizer. Initial parameters: nozzle N^o5 (Table 1), $P_0 = 0.45$ bar, $T_0 = 300$ K, $\Gamma^* = 625$, $\langle S \rangle = 18$ [33], $x = 60$ mm, $E_e = 40, 70$ or 100 eV.

der Waals clusters are an order of magnitude lower than in molecules (as noted above, specific binding energy < 80 meV [35]), which leads to strong fragmentation of clusters when using the traditional electron ionization method [152]. As an example, Fig. 19 presents a comparison of mass spectra registered at different electron energies in the mass spectrometer ionizer in an argon flow. It can be seen that as the value of E_e increases, the intensity of high-mass peaks grows, which may be associated with an increase in the probability of interaction between clusters and ionizer electrons [153]. At the same time, according to estimate (2b), for this flow regime $\langle S \rangle = 18$, which allows concluding that fragmentation plays a significant role in the obtained size distributions of clusters. This requires the use of more delicate diagnostic methods (e.g., photoionization [152], Rayleigh scattering [114], diffractive He atom scattering [33], etc.) suitable for working with clusters.

At the same time, molecular beam mass spectrometry allows detecting the formation of various associates in supersonic flow. Fig. 20 presents fragments of a mass spectrum registered in an argon-hydrogen mixture flow (90/10). Under these conditions, not only ions of monomers, dimers, and trimers of argon (Ar_j^+ where $j = 1-3$; $M/z = 40, 80$, and 120) are registered, but also particles that can be identified as the result of protonation of argon (Ar_jH_k^+ where $j, k = 1-3$; $M/z = 41-43, 81-83, 121-123$). Based on the conclusions of the works [82,154,155], it can be assumed that the formation of these ions is the result of electron-stimulated decay of mixed argon-hydrogen clusters formed during supersonic expansion of the gas mixture into rarefied space, since when ionizing by the EBMS method, there are insufficient collisions in the mass spectrometer detector for other variants of ion formation (Ar_nH_m^+). At the same time, it has been shown in Ref. [156] that in the case of ionizing particles with an electron beam in a supersonic flow,

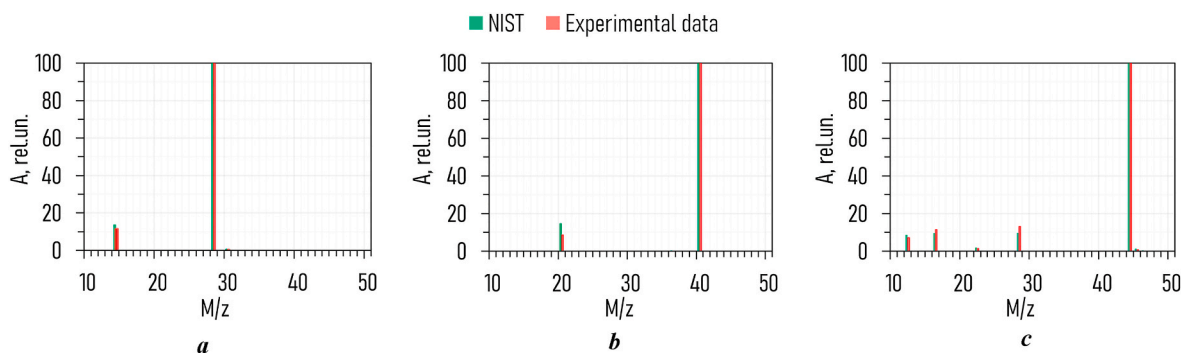


Fig. 18. Mass spectrum of nitrogen (*a*), argon (*b*) and carbon dioxide (*c*) in supersonic molecular beam. Initial parameters: nozzle N^o5 (Table 1), $P_0 = 0.2$ bar, $P_\infty < 5.2$ μ bar, $x = 60$ mm, $E_e = 70$ eV.

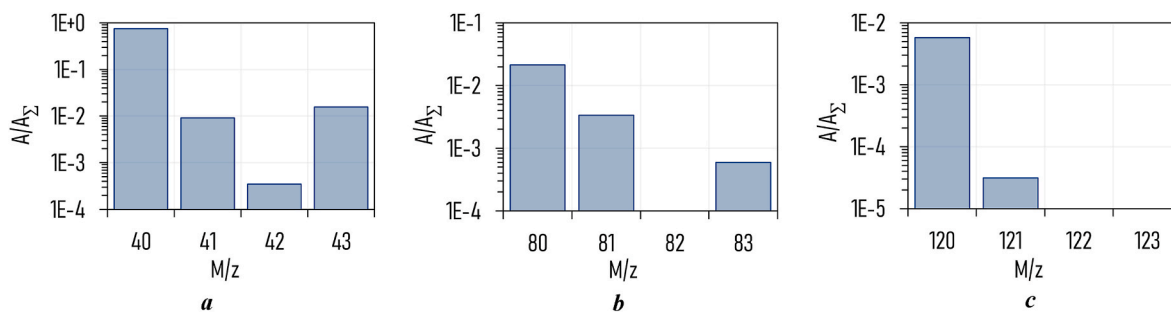


Fig. 20. Mass spectrum fragments recorded in the argon-hydrogen supersonic flow. Initial parameters: nozzle N^o6 (Table 1), $P_0 = 1.0$ bar, $P_\infty < 5.2$ μ bar, $x = 60$ mm, $E_e = 70$ eV.

where collisions are still present, the protonation process proceeds more efficiently.

Mass discrimination in the quadrupole mass spectrometer used in the work when diagnosing a molecular beam under conditions of no clustering and when ionizing neutral particles with 70 eV electrons does not exceed 10%. At the same time, in a clustered molecular beam, even when using the EBMS method, there may be an additional influence of clusters due to differences in particle velocities of different masses, due to their destruction at different times after ionization, as well as non-uniformity of the extraction potential field at the ionizer exit. Nevertheless, based on the mass spectra of argon and nitrogen (see Fig. 18), this influence does not significantly contribute to increasing discrimination. However, when using HVEB and DIN methods, according to Ref. [44], mass discrimination in the range $M/z < 100$ increases significantly. When $M/z > 100$, the contribution of discrimination decreases due to higher initial energies of heavy ions at the mass spectrometer inlet [157,158].

3.2.3. Total scattering cross section

According to Refs. [62,159–161], the process of scattering of molecular beam particles can be described by the expression:

$$\frac{A}{A_{\max}} = \exp\{-\rho_{ps} \cdot L_{ps} \cdot \sigma_b\} \quad (16)$$

where $A_{\max}$ is the amplitude of the registered signal at pressure in the post-skimmer section $P_{ps} \rightarrow 0$; $\rho_{ps} = P_{ps}/k_b T_{ps}$ is the numerical density of particles in the post-skimmer section; L_{ps} is the length of the post-skimmer section; σ_b is the effective cross-section for scattering of molecular beam particles; k_b is the Boltzmann constant. Thus, by measuring the dependence of the signal amplitude on the molecular beam axis on pressure (density) in the post-skimmer section, it is possible to

determine the value of the effective scattering cross-section of flow particles for the used system (for the solid angle within which measurements are taken).

Fig. 21 presents the results of measuring scattering cross-sections of nitrogen molecules (a) and argon atoms (b) using this approach [162]. The obtained results are well described by an exponential dependence (16). The obtained scattering cross-section values for nitrogen and argon are approximately 28.6 and 43.2 $\text{\AA}^2$, respectively. It should be noted that the measured scattering cross-section values σ_b are not differential, which makes them dependent on the solid angle within which unscattered particles reach the detector. In this study, the solid angle φ_{ps} did not exceed $2.7 \cdot 10^{-6}$ sr. If we assume that in the first approximation the relationship between cross-section and solid angle is linear [163], the obtained values can be compared with reference data presented in Refs. [164–166], where the solid angle $\varphi_{ps} \sim 10^{-5}$ sr. For example, in Ref. [166], the obtained value of the effective scattering cross-section for argon is 302 $\text{\AA}^2$ at $\varphi_{ps} \approx 1.1 \cdot 10^{-5}$ sr.

Since clusters move in the flow with a directed velocity close to the monomer one [147], they possess an impulse proportional to their size, as a result of which, during collisions with gas target atoms in the molecular beam, they deviate from the flow axis less than monomers, so their scattering cross-section turns out to be smaller. However, under condensation conditions, it was also found that the registered scattering cross-section of argon atoms ($M/z = 40$), which is a constant value according to Ref. [62], changes under different clusterization conditions. Fig. 22 shows the obtained values of the argon atom scattering cross-section in modes with different degrees of condensation depending on the average cluster size $\langle S \rangle$ [33].

This effect can be explained as follows. It was shown in Ref. [167] that during the development of the condensation process, the number of atoms registered on the molecular beam axis significantly decreases due to gas-dynamic separation of particles by mass in the jet, leading to

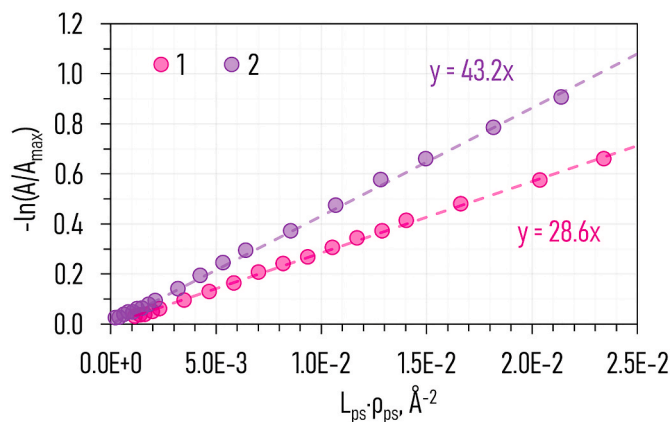


Fig. 21. Measurement results obtained by scattering a molecular beam. Initial parameters: 1 – nitrogen, nozzle N^o 3 (Table 1), $P_0 = 0.5$ bar, $x = 40$ mm; 2 – argon, nozzle N^o 4, $P_0 = 0.25$ bar, $x = 50$ mm.

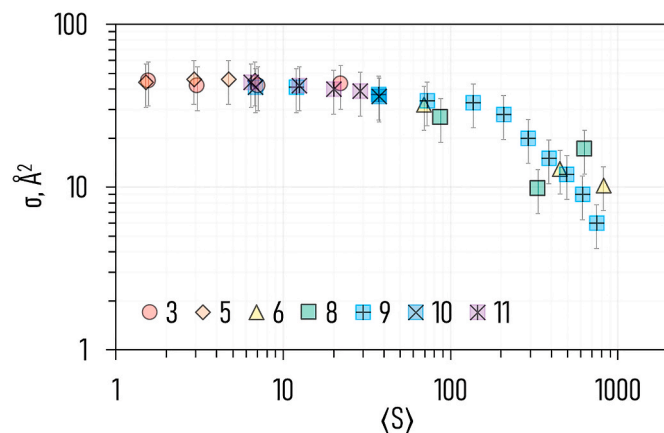


Fig. 22. Measurement results of scattering cross-section of argon atoms in flows with different degrees of condensation behind different nozzles (Table 1).

displacement of light clusters and monomers from the flow axis. In Ref. [168], it was demonstrated by calculation methods that the average condensate fraction in the argon flow does not exceed 30%, however, this value on the molecular beam axis may be higher. The monomers remaining on the molecular beam axis undergo similar collisional scattering described by dependence (16). At the same time, due to collisions with background particles (large clusters, due to their larger surface area, undergo significantly more collisions), clusters can both deviate from the axis and fragment down to monomers, which are subsequently registered by the mass spectrometer and provide an additional increment to the “pure” monomer signal, which, according to (16), will give a lower value of σ . It should also be taken into account that in the mass spectrometer detector, clusters of different sizes can fragment into large fragments or completely disintegrate into atoms.

4. Conclusions

A description of the LEMPUS-2 vacuum facility for studying rarefied gas flows is presented. Compared to the 2017 description [29], the capabilities of the laboratory vacuum setup have been significantly expanded, providing a more detailed study of gas-dynamic and molecular-kinetic processes in supersonic rarefied flows. This enables, among other things, the simulation of processes occurring around spacecraft in the Earth's ionosphere and exosphere, open space, and the rarefied atmospheres of Solar System planets. It is shown that a relatively small laboratory stand, equipped with modern oil-free high-vacuum pumping tools and a universal set of diagnostic equipment, allows for detailed ground-based experimental modeling comparable in resources and functions to large gas-dynamic complexes (e.g., VS-3 and VIKING (Russian Federation) [1], Modular Vacuum Vessel (Italy) [4], PES (China) [5], STG-CT (Germany) [6]), while significantly reducing the costs of full-scale testing for new space technology. A review of diagnostic tools for supersonic rarefied flows used in global practice and available in the laboratory is performed. Methods and approaches based on a comprehensive and multifaceted study of processes have been developed, adapted, and verified, providing information on the shape, structure, and energy parameters of supersonic rarefied flows using EBFT and MBD. The possibility of using existing methodologies for measurements in flows with advanced condensation is also considered.

Based on the obtained data, the following conclusions can be drawn. When modeling discharge processes using similarity parameters [1] at laboratory gas-dynamic facilities, it should be noted that the occurring condensation process can significantly affect the resulting data. At $\Gamma^* < 600$, the influence of condensation on the parameters recorded using the described techniques is negligible; at $\Gamma^* \in [600; 1800]$, the influence of condensation is small. This can be explained by the fact that under these conditions, both the condensate fraction and the average cluster size in the flow are small [168]; therefore, the few small clusters formed are unable to exert a significant influence on the gas dynamics of the entire flow. In turn, at $\Gamma^* > 1800$, the recorded values differ significantly from those obtained in monomeric flows. In this case, the application of these techniques to clustered flows is possible only with the introduction of appropriate corrections and consideration of distorting factors. Advancing diagnostic methods (including the use of delicate

high-sensitivity laser techniques) and developing validated numerical simulation packages for the condensation process in flows will enable a further step in understanding the observed phenomena, which will facilitate the expansion of practical cluster applications in various fields.

Ethical approval

Not applicable.

AI declaration

The authors declare that no AI was involved in the preparation of this manuscript.

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CRediT authorship contribution statement

K.A. Dubrovin: Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Supervision, Validation, Visualization, Writing – original draft. **A.E. Zarvin:** Conceptualization, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Writing – review & editing. **V.V. Kalyada:** Investigation, Methodology, Resources, Validation. **A.S. Yaskin:** Formal analysis, Investigation, Methodology, Visualization, Writing – review & editing. **E.D. Dering:** Data curation, Formal analysis, Investigation, Methodology, Validation, Visualization, Writing – review & editing. **V.E. Khudozhnikov:** Formal analysis, Investigation, Methodology, Validation, Visualization, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Isentropic Gas Expansion: Key Terms

Gas expansion through the nozzle occurs in the isentropic approximation:

$$\frac{P_0}{P} = \left(\frac{\rho_0}{\rho}\right)^\gamma = \left(\frac{T_0}{T}\right)^{\frac{\gamma}{\gamma-1}} \quad (\text{A1})$$

Using the Bernoulli equation for compressible medium without friction consideration:

$$dP + \rho \cdot u \cdot du = 0 \quad (\text{A2})$$

and the differential form of the continuity equation:

$$d(\rho \cdot u \cdot F) = 0 \quad (\text{A3})$$

it is possible to obtain [146]:

$$\frac{P_0}{P} = \left(1 + \frac{\gamma-1}{2} M^2\right)^{\frac{\gamma}{\gamma-1}}, \frac{\rho_0}{\rho} = \left(1 + \frac{\gamma-1}{2} M^2\right)^{\frac{1}{\gamma-1}}, \frac{T_0}{T} = 1 + \frac{\gamma-1}{2} M^2 \quad (\text{A4})$$

In [75], an empirical distribution $M(x)$ for a sonic nozzle was presented in the form:

$$M[x] = A^* \left(\frac{x - x^*}{d_s}\right)^{\gamma-1} - \frac{1}{2} \frac{\gamma+1}{\gamma-1} \left(A^* \left(\frac{x - x^*}{d_s}\right)^{\gamma-1}\right)^{-1} \quad (\text{A5})$$

where d_s is the diameter of the outlet (critical) section of the sonic nozzle; A^* and x^* are constants (see Table A1) depending on γ and determined either by calculation or experimentally.

TABLE A1
Constants for centerline property distribution [75].

γ	1.67	1.4	1.2857
x^*/d_s	0.075	0.4	0.85
A^*	3.26	3.65	3.96

From the consideration that in the critical nozzle section $M = 1$, from equation (A4) we obtain:

$$\frac{P_*}{P_0} = \left(\frac{2}{\gamma+1}\right)^{\frac{\gamma}{\gamma-1}}, \frac{\rho_*}{\rho_0} = \left(\frac{2}{\gamma+1}\right)^{\frac{1}{\gamma-1}}, \frac{T_*}{T_0} = \frac{2}{\gamma+1}. \quad (\text{A6})$$

From (A4) and (A6) we have:

$$\frac{P_0}{P_a} = \left(1 + \frac{\gamma-1}{2} M_a^2\right)^{\frac{\gamma}{\gamma-1}}, \frac{T_*}{T_a} = \frac{1 + \frac{\gamma-1}{2} M_a^2}{\frac{\gamma+1}{2}}, \quad (\text{A7})$$

which allows representing (A3) in the form:

$$\frac{F_a}{F_*} = \frac{\rho_* \cdot u_*}{\rho_a \cdot u_a} = \left(\frac{T_*}{T_a}\right)^{\frac{\gamma+1}{2(\gamma-1)}} \frac{1}{M_a} = \frac{\left(1 + \frac{\gamma-1}{2} M_a^2\right)^{\frac{\gamma+1}{2(\gamma-1)}}}{M_a \left(\frac{\gamma+1}{2}\right)^{\frac{\gamma+1}{2(\gamma-1)}}} \quad (\text{A8})$$

Appendix B. Brief description of numerical modeling approaches

For the numerical simulation of gas expansion from a nozzle into a vacuum, the verified **SMILE++** software package, developed at the Khris-tianovich Institute of Theoretical and Applied Mechanics SB RAS, was used [18,169]. The software implements the majorant frequency scheme for the DSMC method and a two-level Cartesian adaptive grid. The collision cell size is automatically adjusted based on the density gradient and local mean free path. Molecular interactions are modeled using the Variable Hard Sphere (VHS) potential [170].

Since the nitrogen molecule is diatomic, it generally possesses two rotational and one vibrational degree of freedom. As the gas temperature in the flow under consideration is significantly lower than the characteristic vibrational temperature of nitrogen (3374 K), the vibrational degree of freedom is assumed to be frozen.

It is well-established that the relaxation of the molecular distribution function toward the Maxwellian distribution requires a different number of collisions for different degrees of freedom: two or three for translational and approximately ten for rotational. To model the energy exchange between translational and rotational degrees of freedom and to describe the gas distribution function relaxation, a rotational relaxation number — a function of gas temperature — was used to determine the probability of inelastic collision between two molecules [171].

The simulations were performed in an axisymmetric formulation using radial weighting factors.

Nomenclature

A	-	—	amplitude of the registered signal
a_a	m/s	—	speed of sound in the nozzle exit
A_{ij}	s ⁻¹	—	probability of the spontaneous transition (transition $i \rightarrow j$)
A^*	-	—	empirical constant in M
a_1, a_2, a_3, a_4	-	—	empirical constants in Γ^*

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b_1, b_2, b_3, b_4	-	—	empirical constants in $\langle S \rangle$
B_{v_1}	m^{-1}	—	rotational constant
c	m/s	—	speed of light in vacuum
d_a	m	—	exit diameter of the nozzle
d_{eq}	m	—	equivalent diameter of the supersonic conical nozzle
d_s	m	—	throat (critical) diameter of the nozzle
E_e	eV	—	electron energy
F_a	m^2	—	nozzle outlet cross-section area
F_s	m^2	—	nozzle critical cross-section area
$\bar{F}$	-	—	area expansion ratio
$G[K', T_{rot}]$	-	—	function that takes into account the rearrangement of rotational states during the initial excitation of the molecule
h	$\text{J}\cdot\text{s}$	—	Planck's constant
i_e	A	—	electron beam current
I_b	$\text{m}^{-2}\text{s}^{-1}\text{kg}$	—	molecular beam intensity
I_{ij}	-	—	radiation intensity (transition $i \rightarrow j$)
$I_{K'K'}$	-	—	intensity of the R-branch lines of the band (0-0) of the first negative system
I_{ref}	-	—	normalization factor (R1 intensity)
$\bar{J}$	-	—	relative impulse of gas at the nozzle exit
k_1, k_2, k_3, k_4	-	—	constant numerical coefficients
K, K'	-	—	quantum numbers of rotational levels
k_B	$\text{J}\cdot\text{K}^{-1}$	—	Boltzmann constant
K_q	m^3/s	—	quenching constant
k_q	-	—	the ratio of the rate of depopulation, due to atomic or molecular collisions, to the total spontaneous decay rate for the upper level
k_u	-	—	the ratio of the excitation rate, arising from primary and secondary electron inelastic collisions, to the total spontaneous decay rate for the upper level
Kn_{ps}	-	—	Knudsen number for the size of the post-skimmer section
L	m	—	distance between the skimmer inlet and the detection plane
L_n	m	—	diverging section length of the nozzle
L_{ps}	m	—	length of the post-skimmer section
m	kg	—	mass of a particle
M	u	—	relative atomic mass
M_a	-	—	Mach number at the nozzle exit
M_p	kg/mol	—	molar mass
n	-	—	degree of mismatch
N	-	—	pressure expansion ratio
P_a	Pa	—	pressure in the nozzle exit section
P_d	Pa	—	pressure in the detector section
P_{ps}	Pa	—	pressure in the post-skimmer section
P_s	Pa	—	pressure in the critical section of nozzle
P_0	Pa	—	stagnation pressure (in the pre-chamber)
P_∞	Pa	—	background pressure (in the expansion chamber)
q	-	—	empirical factor in $\Delta\bar{J}$
Q_q	m^2	—	total collision quenching cross-section for the upper level
R	$\text{J}\cdot\text{K}^{-1}\cdot\text{mol}^{-1}$	—	molar (universal) gas constant
r_a	m	—	radius of the outlet (exit) section of the nozzle
r_m	m	—	position of the hanging shock (taking into account the compressed layer) in the maximum cross-section
r_{sk}	m	—	radius of the skimmer inlet section
$\langle S \rangle$	-	—	average size of clusters
$S_{ }, S_{\perp}$	-	—	speed ratio values
T_{ps}	K	—	background temperature (in the post-skimmer section)
T_{rot}	K	—	rotational temperature
T_s	K	—	temperature in the critical section of nozzle
T_0	K	—	stagnation temperature (in the pre-chamber)
T_∞	K	—	background temperature (in the expansion chamber)
U_{col}	V	—	potential on the collimator
U_{sk}	V	—	potential on the skimmer
u_a	m/s	—	directed (hydrodynamic) flow velocity
W	m/s	—	average particle velocity along the streamline
x	m	—	longitudinal coordinate (along the jet axis)
x_m	m	—	position of the Mach disk or X-configuration
x^*	m	—	position of the radial expansion point
z	-	—	charge number
α	rad	—	angle between some arbitrarily selected streamline and the jet axis
α_n	rad	—	half angle of the diverging section of the nozzle
β	rad	—	angle between velocity v and the molecular beam axis
γ	-	—	adiabatic index (ratio of specific heats)
Γ^*	-	—	Hagena's correlation complex
θ_a	rad	—	angle between the axis of the molecular beam and the line connecting the center of the skimmer with the detection point
θ_m	rad	—	characteristic jet expansion angle
θ_{rot}	K	—	characteristic rotational temperature of the molecule
ν_1^*	m^{-1}	—	transition wave number
ν_{ij}	Hz	—	frequency (transition $i \rightarrow j$)
ρ	m^{-3}	—	local numerical particle density
ρ_b	kg/m^3	—	particle density at an arbitrary point of the molecular beam
ρ_i	-	—	number of particles in the upper excited state
ρ_{ps}	m^{-3}	—	numerical density of particles in the post-skimmer section

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ρ_s	m^{-3}	—	numerical density of particles in the critical section of nozzle
ρ_0	m^{-3}	—	numerical density in the nozzle pre-chamber
ρ_{00}	-	—	number of particles in the ground state
σ_b	m^2	—	effective cross-section for scattering of molecular beam particles
σ_i	m^2	—	cross-section for excitation of the upper state
τ_i	s	—	excited state lifetime
Ψ_{ps}	rad	—	solid angle

Data availability

Data will be made available on request.

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