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The electron beam diagnostic of the clustered supersonic nitrogen jets

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Abstract. Axial and radial distributions of the rotational temperature and density of $N_2$ molecules in supersonic nitrogen jets formed with conic nozzles (critical diameters $d_{cr}$ of 0.17 and 0.21 mm) were studied using the electron beam fluorescence technique at stagnation pressures $P_0$ of 0.1-0.6 MPa. A rotational temperature $T_r$, equaling a gas temperature $T_g$ owing to fast RT relaxation, was obtained using the rotational line relative intensity distribution in (0-1) vibrational band of the $N_2$ first negative system. Gas density profiles in the jets were obtained using the integral intensity of the band. It is found, $T_r$ at the nozzle outlet is of the order of a few tens of Kelvin and at further expansion $T_r$ drops up to 15-20 K at distance of (100-200) $d_{cr}$. The gas temperature and density distributions in the studied supersonic nitrogen jets are not similar to the isentropic distributions. It is shown, that the lower is the stagnation pressure the faster the gas density and temperature decrease with distance from the nozzle. Increase in $P_0$ leads to elevating $T_g$ in the jets. A reason for this effect may be cluster formation in the jets. Estimations of cluster mean sizes in the jets using Hagen’s parameter show presence of large clusters ($M \geq 200$) at $P_0=0.4-0.6$ MPa.

1. Introduction

For the first time cluster beams have been received in the middle of a past century in Germany at evaporation of a vapor, formed in a source, through a small nozzle into empty space [1]. Now the homogeneous condensation of gas at its expansion at outflow through a nozzle is the most widespread method of cluster production. Cluster beams are usually used for preparation of thin films, fine polish of surfaces and developing new materials [2, 3].

Cluster size depends on stagnation pressure and temperature, nozzle orifice and gas nature [4]. On the other hand, clusters continue to form and grow in expanding free jet until gas density is high enough and gas temperature is low enough. The cluster growth influences on gas density and temperature in the jet, as gas density decreases and some amount of heat is released at gas condensation. Here we study gas density and temperature distributions in supersonic nitrogen free jets using the electron beam fluorescence (EBF) method developed by Muntz [5]. The EBF method was frequently utilized to measure rotational temperature distributions in supersonic nitrogen free jets [6, 7]. However, Marrone pointed that the measurement by the EBF method might be affected by secondary electrons. Some deviations from the Boltzmann distribution (BD) in population of the rotational states were observed in [8, 9]. Coe et al. [7] proposed an analytical model including a quadrupole interaction with an ejected electron as well as a dipole interaction with a primary electron and reduced the spectroscopic data to the BD, concluding that there were no deviations from the BD in the rotational states in a wider range of $P_0 \cdot d$ ($P_0$ is the source pressure and $d$ is the orifice diameter) from 16 to 1016 Torr-mm. Sharafutdinov et al. compared the experimental data obtained by EBF with coherent anti-Stokes Raman spectroscopy (CARS) in the range of $P_0 \cdot d$ from 60 to 1500 Torr mm, the
good agreement was obtained when the density in the jet was high enough to keep the equilibrium conditions [10]. They mentioned that the contribution of secondary electrons is small in low-density conditions. In our experiments $P_0$ of was varied in a range of 128-767 Torr and we suppose the BD in the rotational states. Gas density and temperature distributions at different source pressures were obtained and an attempt to observe some effects of clusters formation in the jets on the distributions was performed.

2. Experimental setup
The jets were obtained in the vacuum chamber of the experimental setup LEMPUS-2 [11]. The expansion chamber is horizontal cylinder with a diameter of 70 cm and a length of 120 cm. The vacuum pumping system provided a total pumping rate of up to 3500 L/s in N$_2$. Stagnation chamber was mounted inside the expansion chamber on the four-directional positioning device possessing three translational and the rotational degrees of freedom. The chamber has laser positioning unit which provide the necessary alignment of the jet axis and the axis of the chamber. Two supersonic nozzles with stationary modes of expansion were used in the experiments: 1) the conic nozzle #1 with inlet diameter $d_{in}$ of 0.17 mm, outlet diameter $d_{out}$ of 2.4 mm and length $L$ of 8.2 mm, and 2) the conic nozzle #2 with $d_{in}$ = 0.21 mm, $d_{out}$ = 3.5 mm and $L$ = 17.5 mm. The stagnation pressure $P_0$ was varied in the range of 0.1-0.6 MPa, gas pressure in the expansion chamber $P_h$ was changed in the range of 0.35-3.6 Pa.

The electron beam (e-beam) was generated by an electron source installed in the upper part of the chamber. The electron source based on a hollow cathode discharge has a ballast volume evacuated by turbomolecular pump and generates a well focused e-beam with electron energies up to 10 keV and the beam current up to 100 mA. The e-beam was crossed the gas jet in perpendicular direction; a position of the jet relatively to the e-beam was assigned by the positioning device. The EBF of the jet was observed through the optical window of 160 mm diameter in the flange on the expansion chamber sidewall. The radiation was focused on one of optical fiber ends while other end of the fiber was installed in front of the spectrometer entrance slit. The spectrometer had a diffraction grating with 1200 grooves/mm and with 30 µm entrance slit it had a resolution of about 0.03 nm. The EBF spectra were recorded by a CCD array connected with a computer via an ADC.

3. The electron beam diagnostics
The electron beam fluorescence (EBF) technique is particularly well suited to low density hypersonic and supersonic flows ($< 10^{16} \text{ cm}^3$) because high fluorescence yields are obtained from excitation of the molecules or atoms of gas flows with high energy electrons. Since its first application in 1953, it has been used in numerous facilities to perform local and non-intrusive measurements of density, vibrational and rotational temperatures and velocity of different species such as N$_2$, NO, CO, CO$_2$ and He [12]. Here we use EBF technique to study rotational temperature and gas density profiles in the supersonic nitrogen jets.

The rotational temperature $T_R$ of N$_2$ molecules was obtained using the relative intensity distribution of rotational lines in (0-1) band of the N$_2$ first negative system (FNS). The theoretical prediction of the relative intensities of the rotational lines in the e-beam excited FNS was developed by Muntz [5]. In case of direct excitation of the $N_2^+ (B^2\Sigma_u^+)$ state by e-beam at electron collisions with nitrogen molecules and the subsequent re-emission unaffected by gas kinetic collisions as well as provided that N$_2$ molecule distribution over rotational levels is a Boltzmann distribution, the $T_R$ is obtained by measuring the relative intensities of the rotational lines in a vibrational band of the FNS and plotting $-\ln[\frac{I_{K^+}K'''}{I_{K''}K''}]/(K''+1)/v''G(K',T_R)$ versus $K''$ [11]). Here $(I_{K^+}K'')$ is intensity of FNS rotational lines in the $(v', v'')$ vibrational band, $K'$ and $K''$ are rotational quantum numbers (Hund’s case (b)), $v'$ and $v''$ are vibrational quantum numbers of $N_2^+ (B^2\Sigma_u^+)$ and $N_2^+ (X^2\Sigma_u^+)$ electronic states, $G(K',T_R)$ is a function of $K'$ and $T_R$, $v$ is the wave number of the emission. Since
$G(K',T_R)$ depends on the rotational temperature $T_R$ and on $K'$, it is necessary to assume a temperature and then obtain the appropriate value $G(K',T_R)$ from a previously calculated look up table. One-two iteration is usually sufficient to obtain a satisfactory temperature measurement.

The total intensity of the vibrational molecular band does not depend on gas or rotational temperature and in case of low gas temperatures (<800K) when practically all molecules are in zero vibrational level ($v''=0$) is proportional to density of the molecules in the ground state $I_n^{R,v''} \propto n_0 (n', \, \text{and} \, n'' \, \text{are electronic quantum numbers})$ [5]. We carried out measurements of the total intensity of (0-1) band of the N₂ FNS along and across the jet to obtain gas density profile in the jet recording the spectrum with 300 µm width of the spectrometer entrance slit. A typical band shapes with resolved and unresolved rotational structure are presented in Fig. 1.

Several experiments were performed to make sure that the effect of secondary electrons and gas heating are small in our conditions. We observe EBF originated from stagnant room temperature nitrogen at the background gas pressure, varying e-beam current in a range of 10-100 mA, and studied dependences of intensity of the (0-1) band of FNS and $T_r$ on e-beam current. The intensity was proportional to e-beam current $i_{eb}$ in a range of 10-50 mA and then declined from the linear dependence showing tendency to saturation. In this range of $i_{eb}$ the $T_r$ was equal to room-temperature within the experimental error (≤5%) and at $i_{eb}$ of 80-100 mA the $T_r$ was higher on 30-50 K. In addition, $T_r$ was measured in X-area of the supersonic jet (nozzle#2, $P_0=0.4 \, \text{MPa}$) at different $i_{eb}$, the measurements also display rise in $T_r$ at $i_{eb}$ of 80-100 mA compared with $T_r$ obtained at $i_{eb}$ of 20-40 mA, in the range of e-beam currents of 20-40 mA $T_r$ was unchanged within the experimental error. Therefore, measurements were carried out at e-beam currents of 20-40 mA.

4. Results and discussion
Photos of the EBF of nitrogen supersonic jets expanding in the vacuum chamber are shown in Fig. 2.

Fig. 1. Typical views of the (0-1) vibrational band of the nitrogen FNS in e-beam fluorescence spectra of the supersonic nitrogen jets with a) resolved and b) unresolved rotational structure; distance from the nozzle is 5 mm; a) nozzle #1, b) nozzle #2.

Fig. 2. Photos of the EBF of the expanding nitrogen supersonic jets (nozzle #1) at stagnation pressure of a) 0.2, b) 0.4 and c) 0.6 MPa.
Fig. 3 show axial profiles of radiation intensity of the (0-1) vibrational band of the FNS and rotational temperature of $N_2$ molecules for the supersonic nitrogen jets. Provided that the intensity is proportional to $N_2$ density $n_0$, the intensity profile displays $n_0$ distribution along the jet centerline. The $T_r$ profile is like the gas temperature distribution along jet centerline because of $T_r=T_g$ due fast rotational-translational relaxation. As it is seen in Fig. 2a at $P_0 \leq 0.2$ MPa there is not formed a typical shape of supersonic jet with one or more barrels [14] so gas density and temperature in the jet decreases at moving away from the nozzle (Fig. 3, curve 1). At $P_0 \geq 0.4$ MPa the EBF photographs clearly show outlines of the first barrel and the shock wave intersection region (X-region). $T_g$ at the nozzle outlet is of the order of a few tens of Kelvin. At moving the jet away from a nozzle the gas density on the jet centerline first decreases at expansion then begins grow due to lateral shock waves influence up to X-region and after then decreases as the jet starts expand again (Fig. 3a, curves 2, 3); at jet expansion the $T_g$ elevates and at compression it falls up to very low values of 15-20 K (Fig. 3b). The $T_g$ and $n_0$ distributions in the nitrogen jets are unlike to the isentropic distributions, for isentropic approximation it is typical decreasing gas density and temperature at jet expansion away from a nozzle.

Radial profiles of the (0-1) band intensity and $T_r$ show that gas density decreases at moving away from jet axis in radial direction and $T_r$ remains almost unchanged in some area around axis and then increases; the last implies on penetration of background gas into the jet.

Fig. 4. Axial profiles of a) radiation intensity of the (0-1) band of the $N_2$ FNS and b) rotational temperature in expanding supersonic nitrogen jets (nozzle #1) as functions of stagnation pressure; $I$ – nozzle #2, $P_0=0.2$ MPa, 2 – nozzle #2, $P_0=0.4$ MPa, 3 – nozzle #1, $P_0=0.6$ MPa.

Fig. 4 shows axial profiles of radiation intensity of the (0-1) band of the $N_2$ FNS and $T_r$ in expanding nitrogen jets (nozzle #1) at different $P_0$. One can see the lower is $P_0$ the faster the gas
density and $T_r$ on the jet centerline decrease with distance from the nozzle. It is worth nothing that the larger is $P_0$, the higher is $T_r$. The reason for this effect may be formation of clusters in the jets.

Cluster mean size in the supersonic nitrogen jets was estimated with Hagena’s parameter $\Gamma^*$ [4]

$$\Gamma^* = K_h P_0 d_{eq}^{0.85} / T_0^{2.29},$$

where $P_0$ is stagnation pressure in mbar, $K_h$ is a coefficient depending on gas nature (528 for nitrogen [8]), $T_0$ is stagnation temperature in K; $d_{eq} = 0.74 d_{cr} / \tan \alpha$ is equivalent nozzle diameter in $\mu$m, where $d_{cr}$ is critical nozzle diameter and $\tan \alpha$ is tangent of a nozzle half-angle. In accordance with [13] the cluster mean size is related to the Hagena’s parameter $\Gamma^*$ for large clusters as $\bar{N} = 2.94 \times 10^{-6} (\Gamma^*)^{2.35}$ and for small clusters as $\bar{N} = 4.62 \times 10^{-3} (\Gamma^*)^{1.64}$. The calculated values of $\Gamma^*$ and $\bar{N}$ for the studied jets are presented in the Table. At cluster formation and growth a heat is released so the larger is the cluster mean size the higher is the gas temperature in supersonic jet. In jet forming with the nozzle #2 at stagnation pressure 0.4 MPa large clusters with mean size of 257 molecules are formed, this is greater than in the jet of nozzle #1 at stagnation pressure of 0.6 MPa. As it is seen in Fig. 3 $T_r$ in jet of nozzle #2 at $P_0 = 0.4$ MPa is higher compared with $T_r$ in jet of nozzle #1 at $P_0 = 0.6$ MPa. This agrees with the hypothesis of the cluster formation effect on gas temperature in the jet.

Table. The supersonic nitrogen jet parameters.

| Nozzle # | $P_0$, MPa | $P_0/P_h$ | $\Gamma^*$ | $\bar{N}$ | Nozzle # | $P_0$, MPa | $P_0/P_h$ | $\Gamma^*$ | $\bar{N}$ |
|----------|------------|-----------|------------|----------|----------|------------|-----------|------------|----------|
| 1        | 0.1        | 285714    | 355        | 7        | 1        | 0.4        | 1421      | 68         |
| 1        | 0.2        | 303030    | 710        | 22       | 2        | 0.4        | 2397      | 257        |
| 2        | 0.2        | 215054    | 1199       | 52       | 1        | 0.6        | 2131      | 195        |

5. Conclusion

Axial and radial distributions of the gas temperature and density in supersonic nitrogen jets were studied using the EBF technique at stagnation pressures of 0.1-0.6 MPa. At the outlet of the nozzle, the gas temperature is of the order of a few tens of Kelvin and at distance of 100-200 $d_{cr}$ the temperature drops up to 15-20K. The gas temperature and density distributions in the studied clustered nitrogen jets are unlike to the isentropic distributions. At pressures $\geq 0.4$ MPa the EBF clearly shows outlines of the first barrel and the shock wave intersection region. It is shown that the lower is $P_0$ the faster decrease the gas density and $T_r$ on the jet centerline with distance from the nozzle and the larger is $P_0$ the higher is the temperature. The reason for this effect may be gas condensation in to clusters in the jets. Mean size of clusters forming in the supersonic nitrogen jets was estimated with Hagena’s parameter. It was found that at $P_0 \geq 0.4$ MPa clusters with mean size of $\geq 200$ M are formed in the jets. It is shown that there can be interdependence between cluster formation and gas temperature in the jets.

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