PHOTOELECTRONIC METHOD FOR MEASURING TIME-OF-FLIGHT SIGNALS

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Molecular beams with a gasdynamic source are now a powerful tool for studying physical, chemical, and kinetic processes, such as condensation, relaxation; chemical reaction, and interactions of molecules and clusters with a surface and with one another [1, 2].

In most works the time-of-flight method is used to measure the velocity distribution function of the molecules (VDF) in the molecular beam. Under certain conditions a relationship can be established between the VDF in the beam and in the jet from which this beam is formed, which enables measuring the VDF in the jet.

The scheme of the time-of-flight molecular-beam method with a gasdynamic source is shown in Fig. 1. A molecular beam, modulated by a chopper 2, is separated from the jet with the help of a special conical diaphragm-skimmer 1. The formed packets of molecules traverse a distance, called the transit baseline 3, and are recorded by the detector 4. The packets of molecules on the baseline spread out in accordance with the VDF, information about which is carried by the detector signal (the time-of-flight signal).

There are two well-known methods for detecting the time-of-flight signals: based on ions and based on metastable particles, forming during the interaction of the electron beam with the molecular beam [2]. In the ion method of detection the chopper consists of a disk with narrow slits, while the detector is an electron beam and an ion collector or a mass-spectrometer. The advantages of this method are the relatively high signal level and insignificant distortions of the VDF accompanying ionization. The disadvantages include the significant and difficult-to-overcome instrumental functions, the complexity of the technical implementation, the indistinguishability of the signal from the molecular beam and the residual gas, and the effect of electromagnetic fields.

When detecting metastable atoms, the chopper is a pulsed electron beam and the detector is an FÉU. The quite simple instrumentation, insignificant instrumental functions, absence of a signal from the background gas, and absence of electric-field effect makes this method competitive, in spite of the low signal level and distortion of the VDF accompanying the formation of metastable particles.

In this work we propose replacing the ion collector with an optical system and a photomultipler, while retaining the chopper and source for exciting the molecules. In this method of recording it is possible to perform a spectral analysis of the time-of-flight signal, which enables the measurement of the VDF in an ensemble of molecules in the same energy state. This method is characterized by weaker signals compared with the ion detector, but the distortion of the VDF by electromagnetic fields and accompanying the excitation of radiation are virtually absent. Another important advantage of this method is the possibility of measuring directly the instrumental functions. Because of the finite time required for opening the chopper and actuating the electronic circuit of the detector as well as because of the finite spatial extent of the region of excitation, significant distortions appear in the time-of-flight signal [3]. They are usually taken into account by introducing the instrumental functions of the detector, its electronic circuit, and the chopper without which it is impossible to perform correct measurements of the VDF and its moments. In the ion method of detection, taking into account the instrumental functions is a quite complicated problem [4], while the photoelectronic method enables their direct measurement. By modeling the molecular beam with a light beam, the overall instrumental function of the chopper and of the electronic circuit of the detector is recorded. The instrumental function of the electron beam, thus far not taken into account in any manner, is determined by measuring the profile of the luminescence of the electron beam in a uniform gas.

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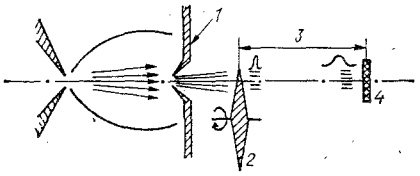

Fig. 1

In deriving the relation between the time-of-flight signal S(t) and the velocity distribution function of the molecules in the flow in front of the skimmer, it is assumed that:

- the change in the velocity distribution function caused by the interaction of the flow with the skimmer is negligibly small;

- the skimmer is very small and it is far enough away from the nozzle that the velocity of the flow can be regarded as directed along the axis of the molecular beam;

- the solid angle of the detector (i.e., the region of intersection of the electron and molecular beams, from which the radiation is collected) relative to the skimmer is small enough that the variation of the velocity in it can be neglected;

- molecular collisions do not occur after passage through the skimmer; and

- the lifetime of the excited states of the molecules is negligibly short.

The coordinate x is measured from the chopper and the time is measured from the moment that the first molecule passes through the slit of the chopper. We denote the instrumental functions of the chopper and electronic circuit of the detector by A(t) and g(t), the length of the detector along the x axis by 2ℓ, and quantities corresponding to the planes of the skimmer, chopper, and center of the detector by the indices s, c, and d.

Under these assumptions the velocity distribution function of the molecules along the x axis at the chopper $f_c(v_x)$ is related with the distribution function in front of the skimmer $f_s(v_x)$ by the relation

$$f_c(v_x) = k_1 v_x^2 f_s(v_x). \tag{1}$$

When the instrumental functions of the chopper and electronic circuit of the detector as well as the size of the region of excitation can be neglected, the time-of-flight signal is given by

$$S^0(t) = k_2 f_c\left(\frac{x_d}{t}\right)\frac{x_d}{t^2}. \tag{2}$$

If the linear density of the electrons along the x axis (integrated in the yz plane over the region of intersection of the electron and molecular beams) B(x) is known, then under the assumption that the density in the molecular beam does not change in the entire recorded region of excitation, the time-of-flight signal (2), taking into account the finite size of the detector, is transformed as follows:

$$M(t) = k_3 \int_{x_d-l}^{x_d+l} f_c\left(\frac{x}{t}\right)\frac{x}{t^2}B(x)\,dx, \tag{3}$$

or

$$M(t) = k_3 \int_{\frac{x_d-l}{t}}^{\frac{x_d+l}{t}} f_c(v)\,vB(vt)\,dv. \tag{4}$$

The distortions introduced into the signal by the instrumental functions of the chopper and electronic circuit are taken into account by the convolution integrals [4]:

$$S(t) = \int_0^t M(t-\lambda)\,\sigma(\lambda)\,d\lambda, \tag{5}$$

where $\sigma(\lambda) = \int_0^\lambda A(\lambda - \tau)\, g(\tau)\, d\tau$ is the total instrumental function. Thus, by determining the functions g(t), A(t), B(x) and solving Eqs. (3) and (5) taking into account (1), it is possible to find from the measured time-of-flight signal S(t) the velocity distribution function of the molecules in the flow in front of the skimmer $f_S(v)$. To reconstruct $f_S(v)$ from (3) and (5), a fast computer with a large memory is required. For a definite form of $f_S(v)$, however, its parameters can be calculated quite simply by the method of moments [5]. The zeroth, first, normalized, and second central moments of the function f(x) are defined as

$$\mu_0\{f(x)\} = \int f(x)\, dx, \quad \eta_1\{f(x)\} = \frac{\int x f(x)\, dx}{\int f(x)\, dx} = \frac{\int x f(x)\, dx}{\mu_0(x)},$$

$$\nu_2\{f(x)\} = \frac{\int (x - \eta_1\{f(x)\})^2 f(x)\, dx}{\mu_0\{f(x)\}} = \eta_2\{f(x)\} - \eta_1^2\{f(x)\}.$$

For the functions appearing in the convolution (5), the following relations exist between the moments:

$$\begin{aligned} \eta_1\{M(t)\} &= \eta_1\{S(t)\} - \eta_1\{\sigma(t)\}, \\ \nu_2\{M(t)\} &= \nu_2\{S(t)\} - \nu_2\{\sigma(t)\}. \end{aligned} \tag{6}$$

From the definition of the moments and Eq. (3) it is easy to obtain

$$\mu_0\{M(t)\} = \mu_0\{f_c(v)\} \int_{x_d - l}^{x_d + l} B(x)\, dx,$$

$$\eta_1\{M(t)\} = \eta_{-1}\{f_c(v)\} \frac{\int_{x_d - l}^{x_d + l} B(x)\, x\, dx}{\int_{x_d - l}^{x_d + l} B(x)\, dx},$$

$$\eta_2\{M(t)\} = \eta_{-2}\{f_c(v)\} \frac{\int_{x_d - l}^{x_d + l} x^2 B(x)\, dx}{\int_{x_d - l}^{x_d + l} B(x)\, dx}.$$

For low gas densities the form of B(x) from [6] is well approximated by a triangular function

$$\begin{aligned} B^-(x) &= 1 - x_d/l + x/l \text{ for } x_d - l < x < x_d, \\ B^-(x) &= 1 + x_d/l - x/l \text{ for } x_d < x < l + x_d. \end{aligned} \tag{7}$$

Then

$$\begin{aligned} \mu_0\{M(t)\} &= \mu_0\{f_c(v)\} l, \quad \eta_1\{M(t)\} = \eta_{-1}\{f_c(v)\} x_d, \\ \eta_2\{M(t)\} &= \eta_{-2}\{f_c(v)\}\left(x_d^2 + l^2/6\right). \end{aligned} \tag{8}$$

Assuming that the velocity distribution function of the molecules in the flow in front of the skimmer is Maxwellian, for $f_c(v)$ we obtain

$$f_c(v) = k_4 v^2 \exp\left[-\frac{m}{2kT}(v - u)^2\right], \tag{9}$$

where u is the flow velocity, T is the temperature, m is the mass of a molecule, k is Boltzmann's constant, k_4 is a constant,

$$\eta_{-1}\{f_c(v)\} = \left(\frac{m}{2kT}\right)^{1/2} \frac{1}{S + r}; \quad \eta_{-2}\{f_c(v)\} = \frac{m}{2kT}\left(\frac{1}{S + r}\right)^2 \left[1 + \frac{1 - 2r(r + S)}{2(S + r)^2}\right],$$

or for S >> r,

$$\eta_{-1}\{f_c(v)\} = \left(\frac{m}{2kT}\right)^{1/2}\frac{1}{S} = \frac{1}{u},\quad \eta_{-2}\{f_c(v)\} = \left(1+\frac{1}{2S^2}\right)\frac{m}{2kTS^2}, \tag{10}$$

where $S = u\left(\frac{m}{2kT}\right)^{1/2}$; $r = \frac{1}{\sqrt{\pi}(1+\operatorname{erf} S)\exp(S^2)}$. Using relations (6), (8), and (10) for a Maxwellian distribution function and B(x) in form (7), we obtain, finally,

$$u = \frac{x_d}{\eta_1\{S(t)\} - \eta_1\{\sigma(t)\}}; \tag{11}$$

$$T = \frac{u^4 m}{4k\left(x_d^2 + \frac{l^2}{6}\right)}\left[\eta_2\{S(t)\} - \eta_2\{\sigma(t)\} - \left(\eta_1^2\{S(t)\} - \eta_1^2\{\sigma(t)\}\right)\left(1+\frac{l^2}{6x_d}\right)\right]. \tag{12}$$

Formulas (11) and (12) enable measuring the flow velocity and the temperature from the known time-of-flight and instrumental functions without solving the inverse problems (3) and (5). The condition S >> r holds well already for S = 2, when the relative error of the approximation is $\Delta u/u = r/S = 2.6\cdot10^{-3}$. Relations (11) and (12) also hold for arbitrary distribution functions of molecules over the velocity components perpendicular to the x axis, and only the function over the x axis remains the same, for example for an ellipsoidal distribution function

$$f(v) = k_5 \exp\left(-\frac{mv_y^2}{2kT_\perp}\right)\exp\left(-\frac{mv_z^2}{2kT_\perp}\right)\exp\left(-\frac{m(v_x-u)^2}{2kT_\parallel}\right).$$

In this case, u and $T_\parallel$ are determined. For an arbitrary function with several unknown parameters the system (8) can be extended by relations between the higher-order moments and the parameters sought can be found from it.

Molecular-beam equipment was developed for studying nonequilibrium processes in low-density jets. The setup for molecular-beam studies (SMBS) is placed inside a gasdynamic tube at the Institute of Thermal Physics of the Siberian Branch of the Academy of Sciences of the USSR (stand VS-4) [7] and supplements previously developed means for diagnostics of rarefied-gas flows [8].

A diagram of the setup and also the equipment used for measurements and primary processing of time-of-flight signals is shown in Fig. 2. The gas expanded into a vacuum chamber through an axisymmetrical acoustic nozzle 1, situated on a three-component coordinate spacer. The axis of the nozzle coincided with the axis of the SMBS.

In order to measure simultaneously the longitudinal and transverse components of the VDF it was necessary to reject the traditional scheme with a collimator and two-chamber pumping. In this case the SMBS consisted of a section with a chopper 2 and a skimmer 3, a section with a detector 4, and an electron gun 5, and the pumping section 6. The chopper was fixed to the coordinate spacer and could be moved away from the axis of the molecular beam for alignment and measurement of the profile of the density across the beam. The electron gun is equipped with systems for focusing and deflecting the electron beam for alignment. The side flange of the detector section had an optical window for extracting radiation onto the FÉU-7 through an optical system consisting of lenses and slits, limiting the region of observation along the axis of the electron beam. The SMBS was evacuated by two N-05 pumps and a panel cooled with liquid nitrogen and serving as an oil trap or, for some gases, a cryogenic pump. A window, behind which an FÉU-8 was located for measuring the instrumental functions, was placed on the back flange. Both FÉU were placed on coordinate spacers with synchrotransmitters and indication blocks for displacement measurements.

The system for recording the FÉU signals was largely the same as that described in [9], where the signal from the ion collector was fed into a synchronous detection block and then into a KSP-4 automatic plotter. The synchropulse was formed from the molecular beam chopper 9. In this work the signal from the synchronous detector 10 was fed through an analog-to-digital converter into the Élektronika 100-I computer 11. The computer accumulated the signals in order to separate them from the background noise and perform the primary processing, including the determination of the hydrodynamic velocity of the flow and the temperature of the gas from formulas (11) and (12) by the method of moments under the assumption of a Max-

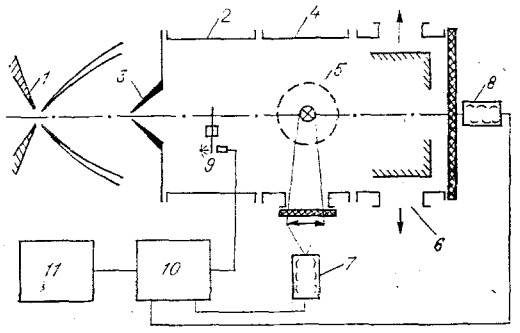

Fig. 2

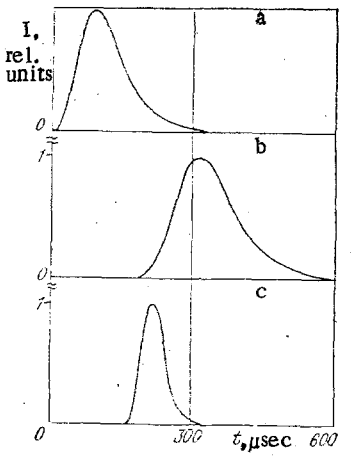

Fig. 3

TABLE 1

Gas	No. of expt.	α	p_0, kPa	p_h 10^{-1} Pa	p_c, MPa	$\bar{u}$, m/sec	σ_u, m/sec	$\tilde{u}$, m/sec	$\bar{T}$, K	σ_T, K	T_*, K	d_*, mm
Ar	1	20,0	1,52	4,79	2,39	545,5	3,4	548,5	3,9	1,2	2,3	5,0
N_2	1	58,5	13,6	5,19	1,73	769,6	1,6	772,2	6,0	0,9	4,2	2,1
	2	24,0	2,53	5,59	2,00	765,9	1,5	768,3	13,1	1,0	8,8	5,0
CO_2	1	20,0	1,46	2,13	0,78	615,2	1,4	611,2	12,4	0,8	10,3	5,0
	2	100	6,65	0,59	0,60	617,1	1,2	619	6.6	0,7	2,8	2,1
	3	100	2,66	0,31	0,29	620,2	2,5	619	8,1	0,8	5,1	2,1

wellian VDF. The finite dimensions of the electron beam were taken into account using formula (7), and the remaining instrumental functions were measured before the time-of-flight signal with the same apparatus and stored in computer memory.

In order to measure the total instrumental function of the chopper and electronic circuit of the detector (their convolutions) the molecular beam was modeled by a light beam recorded by the FÉU-8 on which a diaphragm was placed, corresponding to the region of intersection of the molecular and electron beams. It is more convenient to use two FÉU in this design, but only one FÉU can be used to measure both signals if they are separated enough in time.

A great deal of attention was devoted to carrying out the work to reduce the noise level in order to obtain high-quality time-of-flight signals. It was established that the main sources of luminescence are the heated parts of the electron gun and the collector, as well as excitation of molecules of the background gas, whose density exceeded that of the molecular beam by a factor of 5-20, by the electron beam. In time-of-flight measurements the large constant bias illumination is a source of strong shot noise, which drowns out the weak useful signal. Therefore, the light background must be reduced in every way possible, achieving low pressure in the detector chamber and using effective photon traps and screens. In this sense the proposed scheme without the collimator and differential pumping is much worse than the traditional scheme.

To test the time-of-flight system developed, we measured the time-of-flight signals in N_2, CO_2, and Ar flows behind the sonic nozzles. The temperature in the flow and the hydrodynamic velocity were found, under the assumption that the distribution function over the longitudinal component of the velocity is Maxwellian, by the method of moments. The following quantities remained unchanged in all measurements: the skimmer diameter 1.54 mm; the skimmer-chopper distance 243 mm; the distance between the chopper and the axis of the electron beam 237 mm; the diameter and width of the chopper slits 100 and 4 mm; the half-width of the electron beam [6] 2.5 mm; the energy and current of the electrons 5.5 keV and 17-30 mA; and the size of the region of emission along the electron beam, cut out by the obstacle system,

8 mm. Figure 3 shows the experimentally measured instrumental function (a) and time-of-flight signal (b), measured in a CO_2 jet, flowing from the forechamber with T_0 = 850 K and p_0 = 2394 Pa through the sonic nozzle with a diameter of $d_* = 5$ mm. The pressure in the expansion chamber was $p_h = 0.21$ Pa and the pressure in the SMBS was $p_c = 7.8 \cdot 10^{-4}$ Pa. The time-of-flight signal was measured at a distance of x = 100 mm ($\tilde{x} = x/d_* = 20$) from the cutoff of the nozzle with the chopper rotating with a frequency of f = 188 Hz. To obtain the curves in Fig. 3, 150 accumulations were required. The hydrodynamic velocity and temperature, determined by the method of moments from this signal, are equal to 1050 m/sec and 76 K, respectively. Figure 3c shows the computed time-of-flight signal, corresponding to a zero instrumental function with other conditions remaining the same.

Table 1 shows the hydrodynamic velocity $\bar{u}$ and temperature $\bar{T}$, averaged over several measurements, as well as their variance σ_u and σ_T and the conditions under which the measurements were performed. In all measurements the temperature of the gas in the forechambers was T_0 = 293 K, the rotational frequency of the chopper was f = 187 Hz, and the number of accumulations was equal to 100-200; $\tilde{u}$ is the velocity of the flow calculated with a heat capacity ratio of 1.4 for N_2 and CO_2 and 1.67 for Ar; T_* is the temperature equal to the isentropic temperature for the "nonfrozen" states and the corresponding "frozen" Mach number M_*; if the measurements were performed downstream from the point of freezing, M_* was determined from the model of instantaneous freezing

$$M_* = 1{,}17\,\mathrm{Kn}_0^{-0{,}4},$$

where Kn_0 is Knudsen's number according to the stagnation parameters.

The value of $\tilde{u}$, repeatedly confirmed by experiments [10], can serve as an estimate for the accuracy of the method used to measure u. From the results in Table 1 it follows that the error in the measurement of the hydrodynamic velocity does not exceed 0.4% (the calculation of $\tilde{u}$ is confirmed with approximately the same accuracy). The quantity T_* is much less certain, so that the accuracy of $T_\parallel$ in the flow is evaluated according to σ_T and is equal to 10-20%.

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