

3. G. V. Sklizkov, "Lasers in high-speed photography," in: Laser Handbook, North Holland, Amsterdam (1972).
4. M. Born and E. Wolf, Principles of Optics, Pergamon (1975).
5. V. F. Klimkin and A. G. Ponomarenko, "Study of impulsive electric breakdown of liquids with the help of optical interferometry," Zh. Tekh. Fiz., 49, 9 (1979).
6. V. F. Klimkin and V. V. Pikalov, "Possibilities of microinterferometry in the study of nonstationary processes," Zh. Prikl. Mekh. Tekh. Fiz., No. 3 (1979).

USE OF THERMAL TRANSDUCERS FOR MEASURING THE MOLECULAR  
VELOCITY DISTRIBUTION FUNCTION

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1. In recent years, thermal transducers (bolometers [1-6] or pyroelectrics [7, 8]) have been used as detectors in performing molecular beam measurements. The quantity to be recorded here is the flux of the thermal energy developing as a result of interaction between the beam molecules and the surface of the transducer's sensing element. Modern thermal transducers have a high sensitivity to small molecular fluxes (up to  $10^{12}$  molecules/(m<sup>2</sup>·sec) [1]) along with rather low inertia (the thermal relaxation time amounts to less than  $10^{-4}$  sec [1, 5]). These transducers are especially useful for detecting molecules with excited internal degrees of freedom in the excitation energy range of up to 1 eV, where devices based on the effect of Auger electron emission are inoperative. There is the possibility of investigating the processes of intrinsic molecular energy excitation and relaxation [4-6, 8]. Inclusion of a thermal transducer in the flight-time measuring circuit [9, 10] provides information on the effect of intrinsic energy relaxation on the molecular kinetic energy without adding to the complexity of the experimental equipment. In the case of molecular photoexcitation by laser radiation [4, 8], it is possible to use a generator of short laser pulses instead of the mechanical modulator in the flight-time circuit. The use of a bolometer as the detector in a flight-time measuring circuit was described in [5]. An expression relating the molecular velocity distribution function to the time dependence of the temperature increment of the bolometer's sensing element was also given there. Several simplifying assumptions were used in this case. In the more general formulation given here, we have derived an integral equation relating the molecular velocity distribution to the electric signal received from the output of the thermal transducer (bolometer or pyroelectric). The equation is given in a form suitable for modern methods of experimental data interpretation.

2. As was shown in [3], the energy flux per unit area of the sensing element of the thermal transducer is determined by the expression

$$W = J[\kappa(E_c + E_B - E_S) + (1 - \kappa)\xi(E_c - E_e)], \quad (2.1)$$

where  $E_c = mv^2/2 + E_i$ , where  $J$  is the molecular flux density,  $m$ ,  $v$  and  $E_i$  are the molecular mass, velocity, and intrinsic energy, respectively,  $\kappa$  is the capture coefficient,  $E_B$  is the sublimation energy,  $E_e$  is the energy of molecules in the state of thermal equilibrium with the surface,  $\xi$  is the accommodation factor, and  $E_S$  is the molecular energy in the solid condensate phase at the surface.

For most gases and the deep-frozen, superconducting bolometers used in [1-6], expression (2.1) is considerably simplified in view of the fact that  $\kappa = 1$  and  $E_S \ll E_c$  with sufficiently high accuracy. In this case (2.1) can be approximated by the expression

$$W = J(E_c + E_B). \quad (2.2)$$

The case where  $E_i \geq E_B$  and, as a result of liberation of intrinsic energy,  $\kappa \neq 1$  and  $\xi \neq 1$  is not considered here.

Pyroelectric transducers [7, 8] are presently used to an ever increasing extent in thermal flux measurements. Their main advantage in comparison with bolometers is their operating simplicity, as they don't require equipment for producing low temperatures. In application to pyroelectric transducers (which operate at room temperature), expression (2.1) is simplified and can be represented as

$$W = J\xi(E_c - E_e). \quad (2.3)$$

Expression (2.3) can also be used for approximating the thermal flux to the surface of a deep-frozen bolometer for detecting He molecular beams, which condense poorly at the liquid helium temperature.

3. We shall derive below an equation relating the molecular velocity distribution function to the recorded signal from the flight-time device with a thermal transducer as the basic detector. In deriving the relationship determining the thermal energy flux  $W(t)$  transmitted by the beam molecules to the transducer surface, we shall use the results obtained in [11]. It can be shown that, for the cases characterized by relationships (2.2) and (2.3), the following expression holds:

$$W(t) = \beta S \int_0^\theta \left[ \frac{mL^2}{2(t-\tau)^2} + Q \right] \psi(L, t, \tau) \frac{L}{(t-\tau)} d\tau, \quad (3.1)$$

where  $[0, \theta]$  is the time interval of molecular beam transmission through the chopper ( $t > \theta$ ), while [11],

$$\psi(L, t, \tau) = A(\tau) \frac{L}{(t-\tau)^3} F\left(\frac{L}{t-\tau}\right), \quad (3.2)$$

where  $A(\tau)$  is the instrumental function of the molecular beam chopper,  $L$  is the distance between the chopper and the transducer surface,  $F(v)$  is the molecular velocity distribution function, and  $S$  is the operating surface area of the transducer. For the case (2.2)  $\beta = 1$ , and  $Q = E_i + E_B$ , and for the case (2.3)  $\beta = \xi$ , we have  $Q = E_i - E_e$ . We assume that the accommodation factor  $\xi$  is independent of the velocity of molecules incident to the surface, which in the low energy range ( $10^{-3}$ - $10^{-1}$  eV) has been confirmed experimentally for surfaces coated with an adsorbing layer.

In subsequent presentation, we shall only consider cases where  $Q$  is known. In particular, this occurs 1) for monoatomic gas beams, where  $E_i = 0$ ; 2) for molecular gas beams characterized by rapid processes of V-R-T-relaxation (in beam formation, as a result of gas acceleration by means of the gasdynamic method, the value of  $E_i$  can be neglected, since  $E_i \ll mv^2/2$ ); 3) for molecular beams with slow R-R-T-relaxation processes, where  $E_i$  is determined by the gas parameters in the forechamber; 4) in the case of laser excitation of molecules, where  $E_i$  is determined by the exciting radiation characteristics.

The quantities  $E_B$  and  $E_e$  for the chosen gas are determined by the surface temperature of the thermal transducer. The coefficient  $\beta$  appears in (3.1) in the form of a factor; its value in normalizing the measured signals is negligible.

The relationship between the thermal flux arriving at the transducer's sensing element and the electric signal received from the measuring circuit output depends on the transducer type. We shall first consider the bolometer type of thermal transducer [1-6].

As was indicated in [12], the relationship between  $W(t)$  and the bolometer circuit voltage  $u(t)$  is determined by the equation

$$cdu(t)/dt + G_e u(t) = \mu \epsilon W(t), \quad (3.3)$$

where  $c$ ,  $G_e$ , and  $\epsilon$  are the specific heat, the reduced thermal conductivity, and the absorptive power, respectively, and  $\mu$  is the proportionality factor.

For a pyroelectric transducer, this relationship is determined from the equation [7]

$$Cdu(t)/dt + u(t)/R = I(t), \quad (3.4)$$

while

$$I(t) = \gamma \frac{d}{dt} \left\{ \frac{\varepsilon}{c} \int_0^t \exp \left[ -\frac{G}{c} (t - \tau) \right] W(\tau) d\tau \right\},$$

where  $\gamma$  is the pyroelectric coefficient,  $G$  is the heat transfer coefficient, and  $C$  and  $R$  are the reduced values of the capacitance and the resistance of the circuit.

The integral analog of Eqs. (3.3) and (3.4) is

$$\int_0^t K(t - \tau) W(\tau) d\tau = u(t), \quad (3.5)$$

for a bolometer,

$$K(t) = \frac{\mu\varepsilon}{c} \exp \left( -\frac{G_e t}{c} \right),$$

while, for a pyroelectric transducer,

$$K(t) = K_1(t) - \int_0^t K_1(t - \tau) K_2(\tau) d\tau,$$

where  $K_1(t) = \frac{\varepsilon\gamma}{cC} \exp \left( -\frac{t}{RC} \right)$ ;  $K_2(t) = \frac{G}{c} \exp \left( -\frac{Gt}{c} \right)$ .

As a signal from a bolometric or a pyroelectric element is transmitted through the amplifier unit, it is distorted due to the inertia of the equipment. This effect can be accounted for by introducing the relaxation function of the electronic circuit  $g(t) = \exp(-t/\tau_e)/\tau_e$  [9], where  $\tau_e$  is the time constant.

In order to obtain the final equation relating the distribution function  $F(v)$  to the signal  $r(t)$  from the output of the recording system, it is necessary to substitute (3.1) in (3.5) with an allowance for (3.2) and then produce convolution of the obtained expression with the function  $g(t)$ . As a result, we arrive at the equation

$$\frac{\beta S L^2}{\tau_e} \int_0^t \exp \left( -\frac{t - \theta}{\tau_e} \right) \int_0^\theta K(\theta - \eta) \int_0^\theta \frac{A(\tau)}{(\eta - \tau)^3} \left[ \frac{mL^2}{2(\eta - \tau)^2} + Q \right] F \left( \frac{L}{\eta - \tau} \right) d\theta d\eta d\tau = r(t). \quad (3.6)$$

The problem of determining  $F(v)$  with respect to the experimentally measured signal  $r(t)$  (comprising the experimental noise component) for the known value of  $\tau_e$  and all the parameters characterizing the measuring system consists in solving the integral equation of the first kind (3.6). It is known that, in the classical formulation, this problem is unstable with respect to errors in the signal  $r(t)$ . Regularization methods [13] are now widely used for solving such problems.

We have explored the possibility of solving Eq. (3.6) by means of Tikhonov's method [13]. For this purpose, we used a modification of the regularizing algorithm [14] developed for the processing of flight-time experimental data obtained in measuring the molecular velocity distribution function by means of a density data unit.

Figure 1 shows the results of applying the algorithm given in [14] to test examples. A Maxwellian distribution for argon at the mean velocity of 800 m/sec and an inlet temperature of 170°K (Fig. 1) and a distribution different from the Maxwellian one, which was obtained in [14] (Fig. 2) were used for simulation. These distributions are represented by solid curves in Figs. 1 and 2. The points mark the distributions obtained by solving Eq. (3.6) by means of the regularization method. The initial signal noise level amounted to 2% of the maximum value. The instrumental function of the chopper  $A(t)$  had the shape of an equilateral trapezoid [9, 10], with the center line at 0.1 msec. The values of the parameter  $\tau_e$  and  $L$  were equal to 0.02 msec and 1.06 m, respectively [14], while the bolometer's thermal relaxation time was equal to 0.085 msec in accordance with [5]. We have considered the case of a deep-frozen bolometer, when relationship (2.2) is satisfied. For the zero value of  $E_1$ , the value of  $E_B$  was taken from a handbook of thermophysical properties of materials.

Thus, in using a thermal transducer as the molecular beam detector the flight-time signal carries enough information for measuring the velocity distribution of molecules. By using

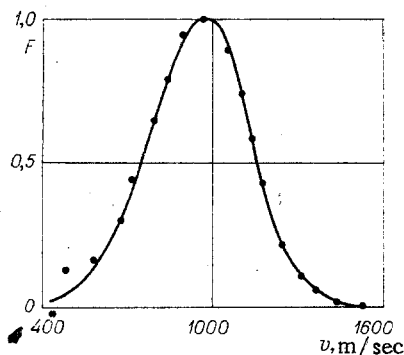


Fig. 1

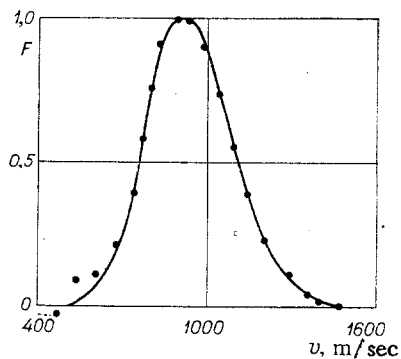


Fig. 2

the regularization method [13] in processing experimental data, we can achieve a fairly high accuracy in calculating this distribution, which is somewhat lower in the case of low-velocity molecules.

#### LITERATURE CITED

1. G. Gallinaro, G. Roba, and R. Tatarek, "Molecular beam detection by fast superconducting bolometer," *J. Phys.*, **E11**, No. 7 (1978).
2. A. A. Vostrikov, S. G. Mironov, et al., "Measurement of the energy of clustered molecular beams by means of cooled bolometers," *Zh. Tekh. Fiz.*, **49**, No. 12 (1979).
3. G. S. Caracciolo, S. Janotta, et al., "Diffractive scattering of H atoms from 001 surface of LiF at 78°K," *J. Chem. Phys.*, **72**, No. 8 (1980).
4. G. R. Coulter, F. R. Grabiner, et al., "Laser pumping of SF<sub>6</sub> in the collisional region of a nozzle beam: bolometric detection of internal excitation," *J. Chem. Phys.*, **73**, No. 1 (1980).
5. M. J. Cardillo, G. E. Becker, and G. D. Kubak, "Granular aluminum superconducting bolometer as a molecular beam detector," *J. Vac. Sci. Technol.*, **17**, No. 1 (1980).
6. A. A. Vostrikov, S. G. Mironov, and B. E. Semyachkin, "Investigation of nonequilibrium processes by means of the molecular beam method," in: *Calculation of Heat and Mass Exchange in Chemical Energy Processes* [in Russian], Institute of Technical Physics, Siberian Branch of the Academy of Sciences of the USSR (ITF SO AN SSSR), Novosibirsk (1982).
7. L. S. Kremenchugskii and O. V. Roitsina, *Pyroelectric Radiation Transducers* [in Russian], Naukova Dumka, Kiev (1979).
8. R. E. Miller, "Infrared laser spectroscopy of molecular beams using a room-temperature beam detector: application to the study of translational freezing in free-jet expansions," *Rev. Sci. Instrum.*, **53**, No. 11 (1982).
9. W. S. Young, "Distortion of time-of-flight signals," *Rev. Sci. Instrum.*, **44**, No. 6 (1973).
10. A. E. Zarvin, "Flight-time method of measuring the molecular velocity distribution function," in: *Nonequilibrium processes in rarefied gas flow* [in Russian], Institute of Technical Physics, Siberian Branch of the Academy of Sciences of the USSR (ITF SO AN SSSR), Novosibirsk (1977).
11. A. I. Sedel'nikov, "Simulation problem of recovering the distribution function with respect to the flight-time signal," in: *Improper Inverse Problems of Atomic Physics* [in Russian], Institute of Theoretical and Applied Mechanics, Siberian Branch of the Academy of Sciences of the USSR (ITPM SO AN SSSR), Novosibirsk (1976).
12. R. Smith, F. Jones, and R. Chesmer, *Detection and Measurement of Infrared Radiation* [Russian translation], IL, Moscow (1959).
13. A. N. Tikhonov and V. Ya. Arsenin, *Methods of Solving Improper Problems* [in Russian], Nauka, Moscow (1979).
14. A. V. Kolosov and A. I. Sedel'nikov, "Effect of the Flight-Time Circuit Parameters on Recovery of the Molecular Velocity Distribution Function," in: *Physical Gas Dynamics* [in Russian], Institute of Theoretical and Applied Mechanics, Siberian Branch of the Academy of Sciences of the USSR (ITPM SO AN SSSR), Novosibirsk (1976).