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TECHNIQUE FOR THE ANALYSIS AND COMPUTERIZED DATA REDUCTION OF TIME-OF-FLIGHT DISTRIBUTIONS OF FREE-JET EXPANSIONS

J. A. Benek, M. R. Busby, and H. M. Powell

ARO, Inc.

October 1970

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FOREWORD

The research reported herein was sponsored by Headquarters, Arnold Engineering Development Center (AEDC), Air Force Systems Command (AFSC), Arnold Air Force Station, Tennessee, under Program Element 64719F.

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This technical report has been reviewed and is approved.

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ABSTRACT

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A velocity distribution function for an aerodynamically accelerated molecular beam is developed from the Maxwell-Boltzmann distribution function for a free-jet expansion. The distribution function is presented in a form compatible with the time-of-flight measurement technique for the experimental determination of such distributions. The expressions for several parameters of general interest, such as static temperature, speed ratio, energy, and velocity, are also presented. A description of a computerized data reduction technique for time-of-flight measurements is given. In addition, the effect of shutter functions and time delays in the accurate measurement of velocity distributions are discussed.

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NOMENCLATURE

| A _c | Area of collimator orifice |
|--------------------------|--|
| AVT | Time of maximum signal |
| С | Total thermal velocity in system at rest |
| C' | Total velocity in moving system |
| Ċ' | Mean velocity |
| C_x, C_y, C_z | Thermal velocity components in system at rest |
| C'_x , C'_y , C'_z | Velocity components in moving system |
| dω | Element of solid angle |
| EV | Beam energy |
| f(v) | Velocity distribution function |
| k | Boltzmann constant |
| ۶ _f | Flight length |
| ℓ _{sc} | Separation distance between collimator orifice and skimmer orifice |
| М | Mach number |
| m | Molecular mass |
| Ń | Defined as dn_t/dt |
| n _t | Number density in "time space" |
| n _v | Number density in "velocity space" |
| R | Gas constant |
| S | Detector signal level |
| s | Nondimensional signal level |
| SR | Speed ratio |
| Т | Temperature |

| To | Source temperature |
|-------|---|
| Ts | Static temperature |
| t | Time of flight |
| tmp | Time of the most probable velocity |
| t_ | Time of flight for a particle having velocity v of flight length ℓ_f |
| V. | Streaming velocity of jet expansion |
| a | $[m/2\pi kT]^{3/2}$ in Maxwellian distribution function |
| β | [m/2kT] ^{1/2} in Maxwellian distribution function |
| γ | Ratio of specific heats |
| θ | Azimuth angle in spherical coordinates |
| τ | Nondimensional time |
| φ | Polar angle in spherical coordinates |
| x[C'] | Speed distribution function for a moving system |

SECTION I

Currently, there is increased interest in the determination of the distribution of velocities in rocket exhaust plumes. Measuring techniques employed in molecular beam devices offer the means to make such measurements in the rarefied portion of the plume expansion. The velocity distribution in any rarefied flow field may be measured by separating (i.e., skimming) a small stream tube from the general flow field (Fig. 1, Appendix). The stream tube is then passed through a collimator to form a molecular beam. The beam is chopped, usually by a rotating disk with radial slots, forming a pulse train of molecules which travel a known flight length before entering a detector. As the molecules traverse the flight length, they separate according to their individual velocities. The detector system then measures the relative number of particles in each of a number of time increments. The output of such a system is shown in Fig. 2.

It is the purpose of this report to illustrate a technique which can be used to interpret these raw data (Fig. 2) in terms of the flow properties of general interest. This will be accomplished by developing an analytical expression for the velocity distribution function of an aerodynamically accelerated molecular beam from the classic Maxwell-Boltzmann distribution function for a beam from an aerodynamic source. The resulting expression will be transformed from "velocity space" to "time space", i.e., to a coordinate system compatible with the physically measurable quantities of the time-of-flight technique. The flow parameters of general interest will then be expressed in terms of the distribution function. Finally, factors affecting the accuracy of the time-of-flight technique will be considered.

SECTION II THEORETICAL DEVELOPMENT OF THE DISTRIBUTION FUNCTION USED FOR DATA REDUCTION

2.1 DISTRIBUTION FUNCTION

2.1.1 Origin

A random distribution of molecular velocities has been found to be accurately described by the classic Maxwell-Boltzmann distribution function obtained from the equilibrium kinetic theory of gases. The function in Cartesian velocity space is of form

$$f[C] = \alpha \exp \left[-\beta^2 (C_x^2 + C_y^2 + C_z^2)\right]$$

where C_x , C_y , and C_z are the components of random velocity, and a and β are functions of temperature and molecular mass. This expression has been shown to give an accurate description of velocity distributions of molecular beams generated from flows from effusive sources (Ref. 1). However, for aerodynamically accelerated beams the above expression must be modified. This can be more easily seen by comparing the distributions from effusive and aerodynamic sources such as is shown in Fig. 3.

2.1.2 Distribution Function for an Aerodynamically Accelerated Beam

The distribution function for the case of an aerodynamically accelerated beam may be obtained from the following assumptions:

- 1. The Maxwell-Boltzmann equilibrium description still remains valid.
- 2. The aerodynamic acceleration superimposes a constant component of velocity, V_{∞} , in the general flow direction. Hence, a Galilean transformation of coordinates is allowed.
- 3. The aerodynamic velocity, V_{∞} , is very much larger than the random components.

It is convenient to choose the coordinate system such that V_{∞} is parallel to one of the axes, for example, the x-axis. The velocity components in the transformed coordinates (denoted by the primes) are given by

$$C_{x}' = C_{x} + V_{\infty}, C_{y}' = C_{y}, C_{z}' = C_{z}$$
 (1)

The velocity in the exponent of the distribution function may be written as

$$C^{2} = C_{x}^{2} + C_{y}^{2} + C_{z}^{2} = (C_{x}' - V_{\infty})^{2} + [C_{y}']^{2} + [C_{z}']^{2}$$

or

$$C^{2} = [C']^{2} - 2C_{x}' V_{\infty} + V_{\infty}^{2}$$

In light of assumption No. 3 above, we may approximate

$$C_x' \simeq C'$$

Then

The resulting expression for the velocity distribution of an aerodynamically accelerated beam is

 $C^2 \simeq [C' - V_{\perp}]^2$

$$f(C) - a \exp \left[-\beta^2 (C' - V_{\infty})^2\right]$$
 (2)

2.2 EXPRESSION FOR PARTICLE DENSITY

2.2.1 In Velocity Space

The output signal from the detection system is proportional to the instantaneous particle density in the ionization region. Hence, it is desirable to obtain an expression for the number density arriving at the detector for a chopped pulse in terms of the distribution function of Eq. (2). The kinetic theory gives the differential number density as

$$dn_y = n_0 f(C) dC_x dCy dC_z$$

where n_0 is the reference number density.

Transforming the coordinates according to Eq. (1), and using the approximation for C^2 , an expression for the number density in the transformed coordinates is of the form

$$dn_{v} = n_{o} \alpha e^{-\beta^{2} [C' - V_{\infty}]^{2}} dC_{x}' dC_{y}' dC_{z}'$$
(3)

It is convenient to define a speed distribution function $\chi[C']$ as

$$X[C'] = \int_{0}^{2\pi} \int_{0}^{\pi} \alpha e^{-\beta^{2} [C'-v_{\infty}]^{2}} [C']^{2} \sin \phi \, d\phi \, d\theta$$

which corresponds to a transformation of the distribution function from Cartesian to spherical coordinates. Performing the integration and substitution of the results into Eq. (3) yields

$$dn_{v} = \frac{4n_{o}}{\pi^{\frac{1}{2}}} \left[\frac{m}{2 k T}\right]^{3/2} [C']^{2} \exp\left\{-\frac{m}{2 k T} [C' - V_{\infty}]^{2}\right\} dC' \qquad (4)$$

where T = temperature and m = molecular mass.

2.2.2 Transformation to Time Space

The form of Eq. (4) is difficult to apply directly since it is not in terms of the readily measurable variables of the system. Therefore, Eq. (3) will be transformed to an equivalent coordinate system with time as the independent variable. The transform is given by

$$C' = \frac{\ell_f}{t}, V_{\infty} = \frac{\ell_f}{t_{\infty}}, \text{ and } dC' = -\frac{\ell_f}{t^2} dt$$
 (5)

where ℓ_f = flight path length, t = time of flight, and t_{∞} = time of flight for a particle having velocity, V_{∞} .

There are two points to note about the transformation. First, it is nonlinear. Second, the appearance of the negative sign which indicates that in the time space the faster molecules arrive first, whereas in the velocity space they arrive last. With this in mind, the negative sign will be dropped in the succeeding development. Thus, at the detector the rate of change of particle density for a pulse is

$${}^{\bullet}_{N} = \frac{dn_{t}}{dt} = \frac{4n_{o}}{\pi^{2}} \beta^{3} \frac{\ell_{f}^{3}}{t^{4}} \exp \left[-\beta^{2} V_{\infty}^{2} \left(\frac{t_{\infty}}{t} - 1\right)^{2}\right]$$
(6)

2.3 EXPRESSION FOR DETECTOR SIGNAL

Data correlation is accomplished by normalization of the intensity (or signal) to maximum signal and time to time of maximum signal. Either Mach number, speed ratio, or temperature may be used as a correlation parameter. In this analysis, speed ratio has been chosen because it is most indicative of the expansion process, i.e., it is a measure of how well the random thermal energy has been converted to the directed kinetic energy of the stream. An analytic expression for the normalized signal can be obtained by recalling that the output signal is proportional to the instantaneous particle density in the ionization region of the detector, or

$$S \sim N = B \frac{1}{t^4} \exp\left\{-(SR)^2 \left(\frac{t_{\infty}}{t} - 1\right)^2\right\}$$
(7)

where B is the constant of proportionality and where SR is the speed ratio and is given by

$$SR = \frac{V_{\infty}}{\sqrt{\frac{2kT}{m}}}$$

The time of maximum signal, t_{max} , can be obtained by differentiating Eq. (7) with respect to time and setting the result equal to zero. Solution of the resulting expression yields

$$\frac{t_{\infty}}{t_{max}} = \frac{1 + \sqrt{1 + 8/(SR)^2}}{2}$$
 (8)

The expression for S_{max} is then

$$S_{max} = B \frac{1}{t_{max}^4} \exp \left\{ -(SR)^2 \left(\frac{t_{\infty}}{t_{max}} - 1 \right)^2 \right\}$$
 (9)

Dividing Eq. (10) by Eq. (12) and introducing the nondimensional quantities,

$$k \equiv \frac{t_{\infty}}{t_{\max}}, r = \frac{t}{t_{\max}}$$

gives the expression for normalized signal

$$\overline{S} = \frac{S}{S_{max}} = \frac{1}{\tau^4} \exp\left\{-(SR^2) \left[\left(\frac{k}{\tau} - 1\right)^2 - (K - 1)^2\right]\right\}$$
(10)

where

$$k = \frac{1 + \sqrt{1 + 8/(SR)^2}}{2}$$

Equation (10) is used to fit the raw data of Fig. 2 as will be explained in Section III.

SECTION III COMPUTERIZED DATA REDUCTION TECHNIQUE

3.1 GENERAL REQUIREMENTS

The basic requirements of the data reduction system are:

- 1. Correction of the data to a reference or zero line. This is to eliminate intrinsic instrumentation error.
- 2. Normalization of data on maximum signal and corresponding time.
- 3. Determination of speed ratio by an iteration procedure which gives best curve fit of data to the distribution function.
- 4. Presentation of data and fitted curve.
- 5. Calculation of additional parameters of interest from the distribution function.

These requirements are most easily satisfied by a computerized data reduction system which gives the additional benefits of speed and accuracy.

3.2 DISTRIBUTION FUNCTION

Correction of the raw data (i.e., the direct output of the detection system (Fig. 2)) for d-c offset is accomplished by comparison of the raw data to a base or reference line (Ref. 2). The base line is obtained, in general, by isolating the detector from the molecular beam and recording the system output. This removes many intrinsic inaccuracies of the system.

An additional correction is made to remove instrument drift during data acquisition. The first 20 points are averaged and fitted with a straight line which will then be used as the zero line for the distribution. Therefore, it should be noted that care must be exercised during data acquisition to assure that these points do not contain part of the distribution.

The corrected data (Fig. 4) are then normalized to the maximum value of signal and the corresponding time. The computer determines these values by searching the corrected data for the maximum value of detector signal. Then, to eliminate the effects of possible scatter in the data, the 11 points preceding and the 11 points following the maximum value are used to determine a fifth order polynomial, least squares curve. This curve is then differentiated and the maximum evaluated. It is this value of signal and corresponding time that is used to normalize the data. The speed ratio represented by the normalized distribution is determined from an iteration process on a least squares fit of the data. The data are fitted to the theoretical expression [Eq. (10)] for a normalized signal, i.e.,

$$\overline{S} = \frac{1}{\tau^4} \exp \left\{ - (SR)^2 \left[\left(\frac{K}{\tau} - 1 \right)^{12} - (K - 1)^2 \right] \right\}$$

where SR is used as a fitting parameter.

The general procedure is to obtain an initial set of residual errors by using a first approximation to the speed ratio as given by Wilmoth and Hagena in Ref. 1. This is

$$\frac{t_{max}}{\Delta t_{Mmax}} = \frac{SR}{1.67} \text{ for } SR > 4 \tag{11}$$

where t_{max} is the time of maximum signal and $\Delta t_{1/2 \ max}$ is the time interval between values of half maximum signal. In normalized form

$$\frac{1}{\Delta r_{\lambda m \, dx}} \simeq \frac{SR}{1.67}$$

A correction of the form $(SR) + \xi$ is then obtained from this set of residuals where ξ is the correction. This correction determines a new value of SR which in turn is used to calculate a set of residuals. The process is repeated until the residuals begin to increase. The minimum residual is then taken as the most probable value of the speed ratio.

3.3 DISCUSSION

At this point it should be noted that data acquired at low values of signal-to-noise ratios can be fitted with an erroneous value of speed ratio. This is due to scatter in the data. The least squares program makes no distinction among the points and finds the best curve through them all. Figure 5 illustrates the difficulty that can result from noisy data. The broken line indicates a better fit.

In addition, it is possible for a "bad" data point to occur such that the maximum value is completely meaningless to the rest of the data. However, since no distinction can be made by the computer, the data will be normalized and speed ratio evaluated according to this value.

It is, therefore, recommended that care be exercised in evaluation of the computer output. The curve fits should be examined visually to assure validity of the data.

3.4 ADDITIONAL PARAMETERS

As noted earlier, several parameters can be evaluated from the distribution function in addition to the speed ratio. An approximation to the value of V_{∞} is obtained

by calculating the most probable velocity of the distribution using a time-of-flight relationship, i.e.,

$$V_{\infty} \simeq \frac{\ell_f}{t_{max}}$$

where t_{max} is determined from the distribution data. The fact that this is a good approximation for large values of the speed ratio may be readily seen from

$$\lim_{\substack{SR \to \infty}} \left(\frac{t_{\infty}}{t_{\max}}\right) = \lim_{\substack{SR \to \infty}} \frac{1 \div \sqrt{1 + B/(SR)^2}}{2} = 1$$

and

$$\lim_{\substack{\text{SR}\to\infty}} \frac{\operatorname{tmp}}{\operatorname{tm}_{4x}} = \lim_{\substack{\text{SR}\to\infty}} \frac{2\beta\ell_{f}}{\operatorname{SR}=\sqrt{SR^{2}-4}} \quad \bullet \frac{1+\sqrt{1+8/SR^{2}}}{2t_{\infty}} = 1$$

where tmp is the time of the most probable velocity obtained by evaluating dx[C']/dC' = 0and transforming into time space according to Eq. (5).

Hence,

$$\lim_{\substack{\text{Im} \\ SR \to \infty}} \frac{\left[\frac{\text{tm}p}{t_{\text{max}}} \right]}{\left[\frac{t_{\infty}}{t_{\text{max}}} \right]} = \frac{\lim_{\substack{\text{Im} \\ SR \to \infty}} \left[\frac{\text{tm}p}{t_{\text{max}}} \right]}{\lim_{\substack{\text{Im} \\ SR \to \infty}} \left[\frac{t_{\infty}}{t_{\text{max}}} \right]} = 1$$

since both limits exist. The static temperature, T_s , of the gas is calculated from the above approximation to V_{∞} and the definition of speed ratio, i.e.,

$$SR = \frac{V_{\infty}}{\sqrt{2 R T_s}}$$
(12)

The one-dimensional form of the energy equation can be used to determine the approximate source stagnation temperature

$$T_o = T_s + \frac{\gamma - 1}{2 \gamma R} V_{\infty}^2$$

Once the speed ratio has been determined, any quantities which depend on the velocity may be calculated from the appropriate moment of the distribution function. Let Q[C'] be a property dependent on the molecular velocities. The mascroscopic or average value, \bar{Q} , is given by

where $\chi[C']$ is the speed distribution. Several of these parameters can be evaluated in closed form by assuming impulsive chopping. Some of these are given on the following page.

The mean speed

$$\overline{C}' = \frac{\int_{0}^{\infty} [C^{2} \times [C^{2}] dC'}{\int_{0}^{\infty} [C^{2}] \times [C^{2}] dC'}$$
(13)

or

$$\overline{C}' = \frac{\ell_{f}}{t_{\infty} SR} \frac{\frac{1}{2} \left[\frac{5}{2} SR + SR^{3} \right] e^{-SR^{2}} + \frac{\sqrt{\pi}}{2} \left[\frac{3}{4} + 3SR^{2} + SR^{4} \right] [1 + erf(SR)]}{\frac{1}{2} [1 + SR^{2}] e^{-SR^{2}} + \frac{\sqrt{\pi}}{2} SR \left[\frac{3}{2} + SR^{2} \right] [1 + erf(SR)]}$$
(14)

The mean square speed

$$(C')^{2} = \frac{\int_{0}^{\infty} [C']^{3} \times [C'] dC'}{\int_{0}^{\infty} [C'] \times [C'] dC'}$$
(15)
$$(C')^{2} = \frac{\ell_{f}^{2}}{\iota_{\infty}^{2} SR^{2}} \frac{\left[1 + \frac{9}{4} SR^{2} + \frac{1}{2} SR^{4}\right] e^{-SR^{2}} + \frac{\sqrt{\pi}}{2} \left[\frac{15}{4} SR + 5SR^{3} + SR^{5}\right] [1 + erf (SR)]}{\frac{1}{2} [1 + SR^{2}] e^{-SR^{2}} + \frac{\sqrt{\pi}}{2} SR \left[\frac{3}{2} + SR^{2}\right] [1 + erf (SR)]}$$
(16)

The average kinetic energy

 $KE = \frac{1}{2} m (C')^2$

The above general expressions contain terms of the form [1 + erf (SR)]. However, since speed ratios encountered in aerodynamically accelerated beams are greater than five, and since

$$[1 + erf(2)]_i = 1.995$$

the approximation [1t + erf (SR)] ≈ 2.0 can be employed. Equations (14) and (16) become

$$\overline{C'} = \frac{\frac{\ell_{f}}{1\infty} \left\{ \frac{1}{2} \left[\frac{5}{2} SR + SR^{3} \right] e^{-SR^{2}} - \sqrt{\pi} \left[\frac{3}{4} + 3SR^{2} + SR^{4} \right] \right.}{\frac{1}{2} \left\{ \left[SR + SR^{3} \right] e^{-SR^{2}} + \sqrt{\pi} SR^{2} \left[\frac{3}{4} + SR^{2} \right] \right\}}$$
$$\overline{(C')^{2}} = \frac{\left(\frac{\ell_{f}}{1\infty} \right)^{2} \left\{ \left[1 + \frac{9}{4} SR^{2} + \frac{1}{2} SR^{4} \right] e^{-SR^{2}} + \sqrt{\pi} SR^{3} \left[\frac{15}{4} SR + 5SR^{3} + SR^{5} \right] \right\}}{\frac{1}{2} \left\{ \left[SR^{2} + SR^{4} \right] e^{-SR^{2}} + \sqrt{\pi} SR^{3} \left[\frac{3}{2} + SR^{2} \right] \right\}}$$

Figure 6 shows the reduced input data of Fig. 2 and the corresponding values of the above parameters. Note once again that the above applies to an impulsive shutter function. This will be discussed further in Section 4.1.

SECTION IV CONSIDERATIONS ARISING FROM THE APPARATUS

4.1 SHUTTER FUNCTIONS

The preceding analysis has assumed that the molecular beam was chopped instantaneously by the chopper. However, when there is a finite time required for the chopping process, the detector system will not record a Maxwellian distribution. The signal will be distorted in some manner peculiar to the type of chopper and chopping frequency employed (Ref. 3).

In practice, the chopper will approximate an impulsive shutter function if

$$\frac{\Delta t}{t_{\max}} \leq 0.05$$

is satisfied (Ref. 4). Here, Δt is the time required for the chopper wheel slot to cross the beam (i.e., time interval between leading and trailing edges of slot crossing the beam) and t_{max} is the detector system time of maximum signal. Most systems in operation maintain $\Delta t/t_{max} \leq 0.05$ by proper choice of shutter speed and width and flight time. The case of $\Delta t/t_{max} > 0.05$ is discussed in Ref. 4 for various types of shutter time dependence.

4.2 TIME DELAYS

4.2.1 Amplifier Freqency Response

Because the amplifier gain depends upon the bandwidth and frequency response of the amplifier circuits, care must be exercised in their application (Ref. 5). A phase shift can result and can become significant at higher chopping frequencies. This shift does not change the distribution shape, but it does make the velocities appear to be less than they actually are. A 150-kHz bandwidth has been shown to be adequate for most applications (Ref. 2).

4.2.2 Quadrupole Flight Times

Significant time delays occur in the quadrupole ion source and mass filtering section. These delays are dependent upon the ion mass and quadrupole ionization energy; however, these do not vary inversely as the mass number of the ion (Ref. 2). Figure 7 shows how these delays vary with mass number and ion energy. It is important to determine these delays accurately and to correct the distribution appropriately. This is accomplished in general from empirical curves such as shown in Fig. 7.

SECTION V TYPICAL DATA

Figures 8 and 9 show typical time-of-flight distributions obtained from various source conditions. These data were obtained from a molecular beam apparatus and illustrate the distributions resulting from exceptionally high signal-to-noise ratio.

Figure 10 shows a time-of-flight distribution obtained by a plume sampling probe. This distribution illustrates the effect of poor signal-to-noise ratio.

SECTION VI SUMMARY

The theoretical time-of-flight distribution has been determined for an aerodynamically accelerated molecular beam. This distribution has been properly transformed to be applicable to the time-of-flight measurement technique. A technique for the computerized reduction of data has been outlined. In addition, problem areas which may influence the accuracy of the data are noted and references cited.

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APPENDIX ILLUSTRATIONS

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Fig. 1 Typical Time-of-Flight System Configuration



Waveform Eductor Base Line

Fig. 2 Typical Output of Time-of-Flight System



Fig. 3 Comparison of Effusive and Aerodynamic Sources



Fig. 4 Corrected Raw Data of Figure 2



Fig. 5 Time-of-Flight Distribution Determined from Excessively Noisy Data

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Fig. 6 Final Reduction of Data of Figure 1



Fig. 7 Ion Flight Times in a Quadrupole Mass Spectrometer



Fig. 8 Typical Time-of-Flight Distribution Obtained from a 1400° K Source



Fig. 9 Typical Time-of-Flight Distribution Obtained from a 2400°K Source



Fig. 10 Typical Time-of-Flight Distribution Obtained from a Mass Spectrometer Sampling Probe

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| A velocity distribution function for a molecular beam formed from an aerodynamically accelerated source is developed from the Maxwell- Boltzmann distribution function for an effusive source. The distribu- tion function is presented in a form compatible with the time-of-flight measurement technique for the experimental determination of such distributions. The expressions for several parameters of general interest, such as static temperature, speed ratio, energy, and velocity, are also presented. A description of a computerized data reduction technique for time-of-flight measurements is given. In addition, the effect of shutter functions and time delays in the accurate measurement of velocity distributions are discussed. | | | | | | | |

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