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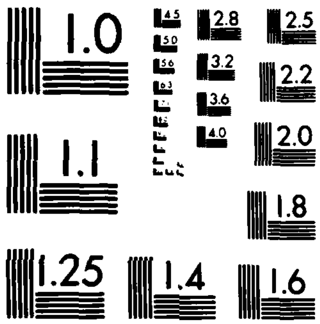

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STUDY OF SUBMICRON PARTICLE SIZE DISTRIBUTIONS BY LASER DOPPLER MEASUREMENT OF BROWNIAN MOTION

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STUDY OF SUBMICRON PARTICLE SIZE DISTRIBUTIONS BY LASER DOPPLER MEASUREMENT OF BROWNIAN MOTION

Annual Technical Report

Prepared by

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SUMMARY

Few nonintrusive techniques are available for particle measurements in the submicron size range (< 0.1 μm diameter), yet measurement of these particles is basic to an understanding of important processes in combustion, such as soot formation and oxidation. The objective of the present research is the development and application of a technique for measurement of individual submicron particles in a gas stream. The approach is to measure the inertial relaxation time of individual particles in Brownian motion, by statistical analysis of the time-resolved (100 MHz) heterodyne signal obtained in an interferometer system resembling a more conventional laser Doppler velocimeter. Progress to date has included the development of an optical system and fast data acquisition system for conducting the experimental studies. A detailed Monte Carlo numerical simulation of the Brownian motion of isolated particles has been developed and applied to the calculation of the time dependent signals —>

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19. Abstract (Continued)

expected in the experiment. These simulated signals have been analyzed for correlations between the particle size and statistical properties of the signal. The mean time between signal extrema has been found to be proportional to the square root of the product of the particle relaxation time, a quantity which is proportional to the particle diameter, and the data sampling interval.

Continuing research on this program will be initially focused on validation of the experimental approach, by statistical analysis of signals obtained with the interferometer for particles of known size. Following the demonstration of this approach, the technique will be applied to the study of the size evolution of combustion particulates.

TABLE OF CONTENTS

LIST OF ILLUSTRATIONS

1. RESEARCH OBJECTIVES (STATEMENT OF WORK)

The research objectives for this program are outlined in the Statement of Work, which includes the following tasks:

Task I - The contractor shall construct a laser diagnostic apparatus to be used for submicron particle size analysis by measurement of Brownian motion. The development of this apparatus shall include:

a) Construction and assembly of relevant optical components;

b) Adaptation of suitable data acquisition and recording apparatus; and

c) Development of appropriate software for analysis of signals obtained from the Brownian motion instrument.

Task II - Using the optical apparatus and data recording and analysis tools developed in Task I, the contractor shall investigate the development of optical heterodyne techniques to be used in measurement of the Brownian motion of individual particles. Specifically, techniques are required which permit measurements of motion where the particle velocity changes in distances shorter than one light wavelength.

Task III - Using the instrumentation and techniques developed in Tasks I and II, the contractor shall analyze the statistics of particle motion in the gas to investigate the particle mass, friction coefficient, and equilibrium temperature. These studies shall be conducted under carefully controlled flow conditions, initially using standard size particles. Subsequent development of the technique may utilize a well-characterized flat-flame burner.

Task IV - The contractor shall investigate the measurement of the refractive index of particles in the Rayleigh scattering regime by analysis of the scattered light intensity and particle mass (size) obtained using the Brownian motion sensor.

Task V - By application of the Brownian motion sensor in a flat-flame burner, the contractor shall assess the application of this technique for in-situ sizing of submicron particles in combustion streams.

2. RESEARCH STATUS

The research effort during the first year of the program has focused on development of the theory of single particle sizing by measurement of Brownian motion. The approach has been to use a numerical simulation of the Brownian motion of individual particles to predict the behavior (temporal characteristics) of the signals which would be measured by an experimental device. This approach has proven to be extremely useful, both in devising methods of analysis of the signal statistics for determination of particle size and in guiding the design of an experimental apparatus (including data acquisition system) for testing the measurement concept. This model of the experimental technique is described in detail in the Appendix to this report, and the important results from the simulation as they bear on the experiments are summarized below.

An experimental optical system and a data acquisition system for the Brownian motion measurements were assembled during the first year of the program. These systems are described below. Finally, the research plan for the second year of the program is outlined in this section.

2.1 Theory of Measurement Technique

Much of the first-year effort was concentrated on development of the theory of the Brownian motion measurement technique. This effort included the development of a detailed Monte Carlo simulation of the experimental system. The important feature of this model is that it simulates time dependent signals arising from particles of different size and facilitates the development of approaches to signal analysis in order to discover correlations between the particle size and statistical properties of the signals. The details of the simulation approach are discussed in the Appendix, but highlights of the approach and important simulation results are summarized in this section.

The measurement approach utilizes a laser interferometric system for measurement of an interference signal which is sensitive to particle displacement. Such a system is shown schematically in Figure 1. Although the system shown is physically similar to the familiar Laser Doppler Velocimeter, important differences in its implementation form the basis of the present approach. This approach, in simple terms, is to measure the time-dependent interference signal arising from a single submicron particle (≤ 0.1 μm diameter) passing through the measurement volume formed by the intersection of the laser beams in Figure 1 and to determine the particle size by statistical analysis of the signal. To understand this approach, it is important to appreciate some of the characteristic aspects of the Brownian motion of individual particles.

A schematic representation of the time dependent motion of a 0.1 μm particle is shown in Figure 2 in order to indicate the time scales characteristic of the motion. In this figure, random changes in particle velocity, of order 10^{-5} m·s^{-1}, occur in characteristic times of 10^{-14} seconds due to collisions with gas molecules. Given sufficient time resolution to measure instantaneous particle velocities, one would observe a Maxwellian velocity distribution if a sufficiently large sample is measured. The characteristic time for the Maxwellian distribution to evolve is the "relaxation time" t_{relax} ($\approx 10^{-7}$ s in Figure 2), which is a function of particle size and fluid properties. Clearly if one wants to obtain any information on the Brownian motion of an individual particle, he must sample the time history of particle motion in an interval short compared to the relaxation time, or the effects will be averaged out. Conversely, one cannot monitor the instantaneous Brownian velocity of the particle, because electronics and signal-to-noise limitations constrain realizable sampling intervals to be much longer than the 10^{-14} s characteristic collision time. The measurement strategy, then, is to sample with an interval on the order of 10^{-8} s, which is fast compared to the relaxation time. The type of signal

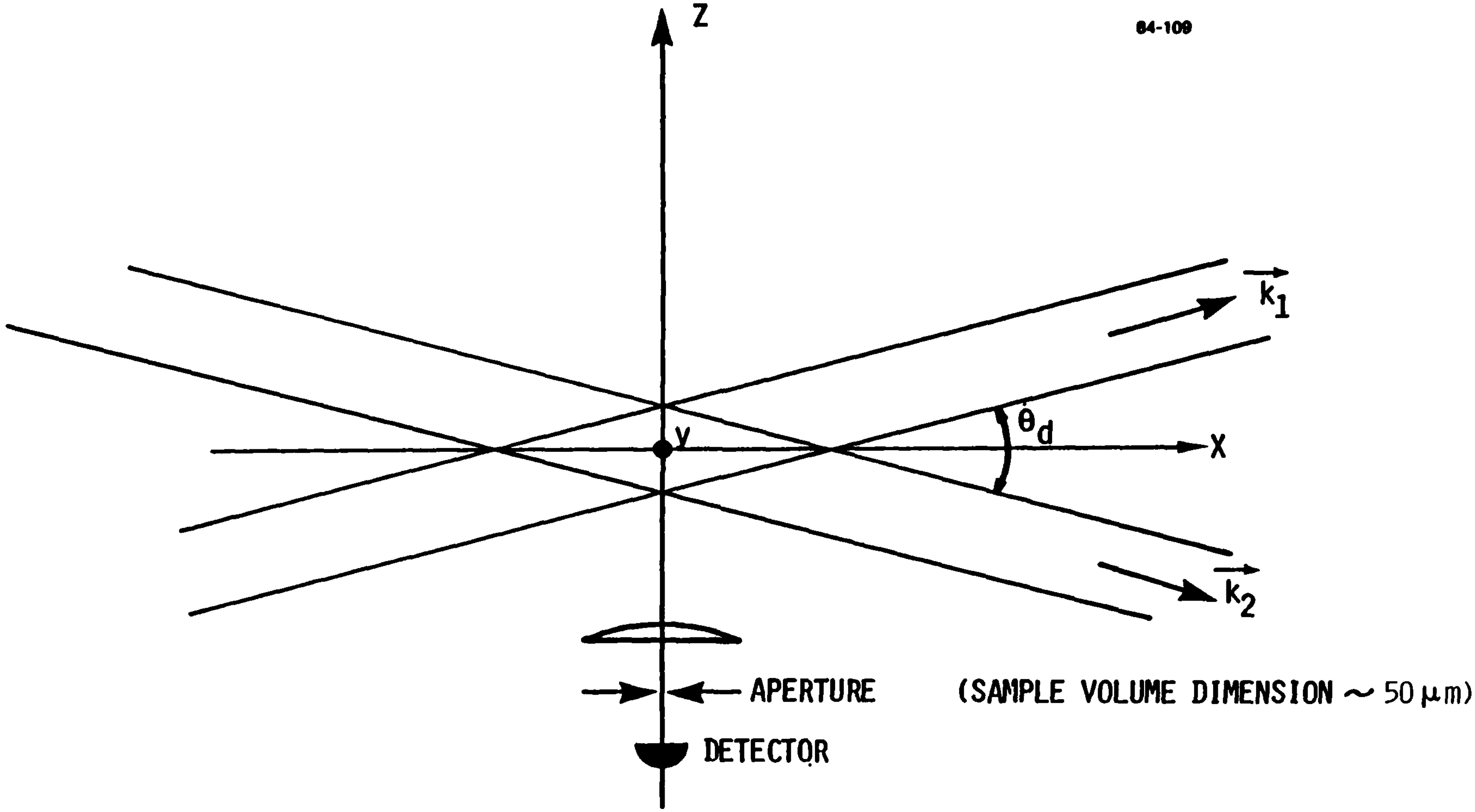


Figure 1. Laser Interferometric System for Measurement of Brownian Motion.

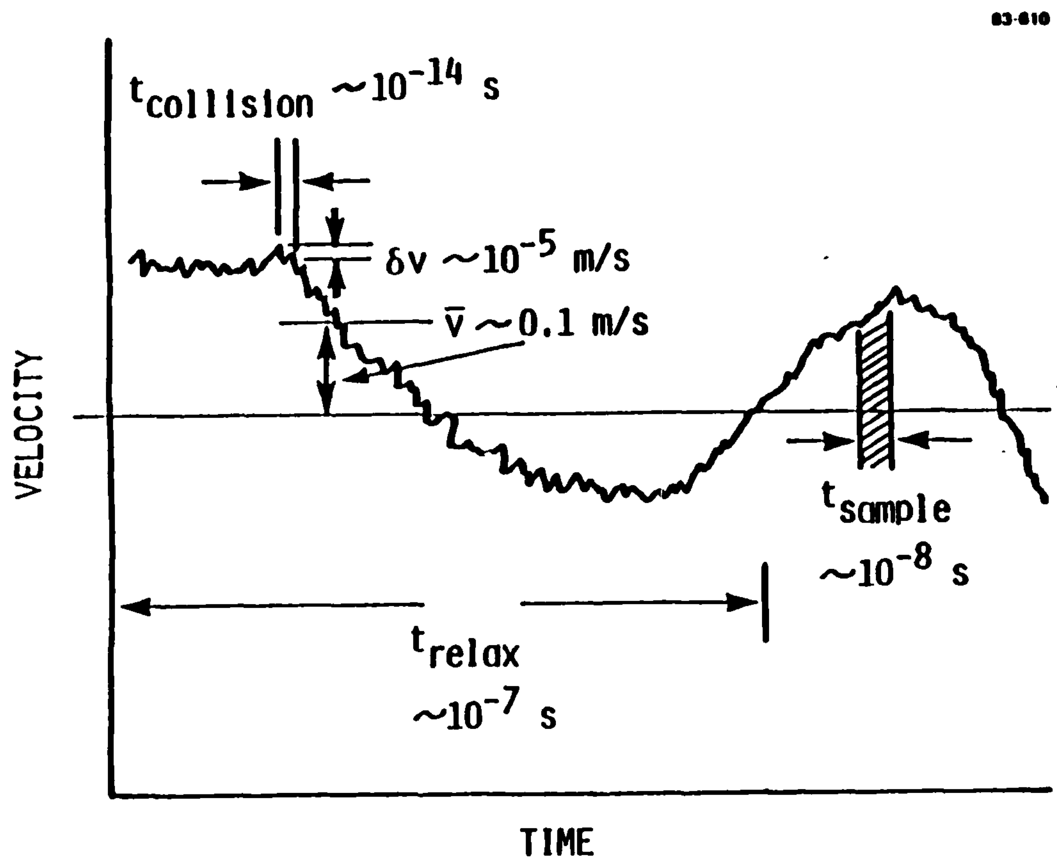


Figure 2. Time Dependent Motion of a 0.1 μm Diameter Particle in Air at 300 K and Atmospheric Pressure.

measured with this kind of time resolution will be a superposition of many random events, but the time dependence of this signal should exhibit statistical properties representative of the Brownian motion of a particle of a particular size.

A second important concept is mentioned here both to avert possible misconceptions about the "LDV" nature of the measurement and to underscore the idea that the signal contains time (but not velocity) information which must be analyzed in a statistical sense. This concept is that the mean excursion distance for the particles in Brownian motion is much smaller than the wavelength of visible light, as shown in Figure 3. As discussed in greater detail in the Appendix, the consequence of this characteristic of the measurement is that the magnitude of signal variations is not meaningful (because the motion may be occurring in different regions of the LDV fringe pattern). The frequency characteristics of the signal are significant, however, in that they can be related to the statistics of particle motion. The identification of such relationships is the motivation for the computer simulation of the signal.

The computer simulation is a solution to the equation of motion for a particle, with a random excitation term representing random collisions with fluid molecules. A Monte Carlo technique is used to simulate the velocity histories of such particles at regular sampling intervals Δt. Particle velocities are chosen from a distribution with the constraint that the distribution is Maxwellian in the long time limit. With the velocity history simulated in this fashion, the particle trajectory through the crossed beam measurement volume of the Brownian motion sensor can be determined by integration over time (a mean fluid velocity may also be superimposed). Finally, the light intensity scattered by the particle from each of the crossed laser beams is readily calculated, and the interference signal at the detector may be obtained as a function of time. This calculated interference signal is the basic result of the simulation. The next step is to consider

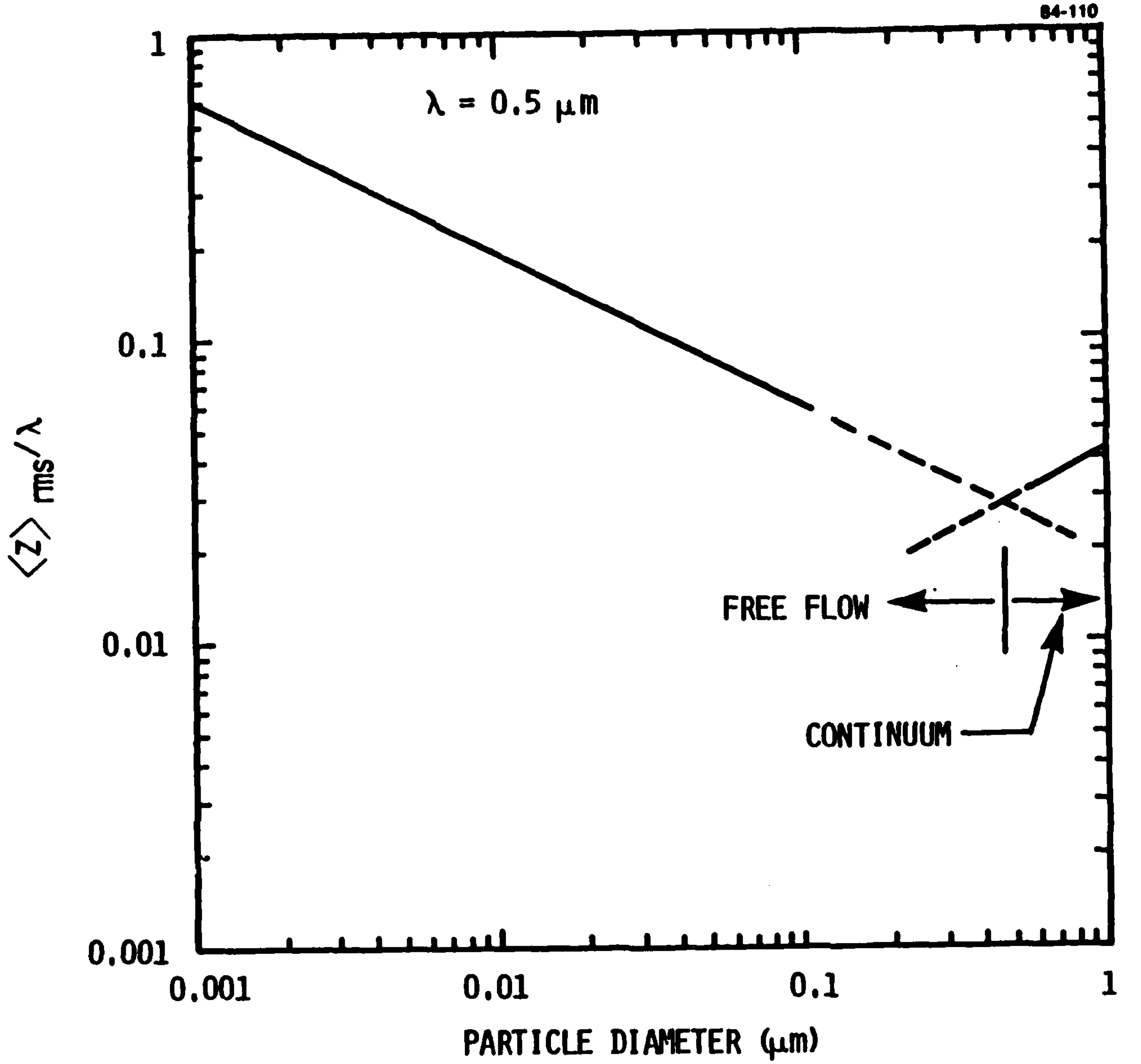


Figure 3. Mean Excursion Distance, Compared with the Wavelength of Visible Light, for Particles (Specific Gravity = 2) in Brownian Motion in a Gas at T = 300 K, p = 1 Atmosphere.

methods for analysis of the signal in order to relate the particle size to characteristics of the signal.

As discussed in the Appendix, a statistical analysis which appears to provide a suitable correlation is an analysis of the mean time between local extrema (first derivative zero crossings) in the signal. As shown in Figure 4, when this parameter is plotted against a parameter depending on the sampling interval, Δt, and the particle relaxation time, $1/\beta$, a straight line correlation is obtained. (Curvature away from the linear relationship occurs when the sampling interval becomes comparable to the relaxation time). Thus, from a measurement of a statistical parameter (the mean time between extrema), the particle relaxation time and, hence, the particle mass or size may be determined. This approach to data analysis is the approach which we expect to pursue in our analyses of experimental data.

2.2 Experimental Development

The experimental development during the first year of the program has concentrated on the design and assembly of a laser optical system suitable for interferometric measurements of the motion of small particles and on the design of a data acquisition system with the very fast data rate capabilities (~100 MHz) required by the measurement technique.

The optical system which has been assembled for this study is shown schematically in Figure 5. A Spectra-Physics Model 165 argon ion laser with a single-line power of approximately 2 watts at 514.5 nm is used. The output of this laser is expanded and collimated to a beam size of approximately 3 cm diameter and is then split with a 50/50 cube beamsplitter. The two parallel beams are brought to a common focus using a multi-element lens system. The focused laser spot size (diagnostic measurement volume) is approximately 50 μm. The measurement volume is imaged through an aperture onto a fast photomultiplier which views the light scattered at 90°. An interference filter is used to reject light outside a 1 nm band centered at 514.5 nm.

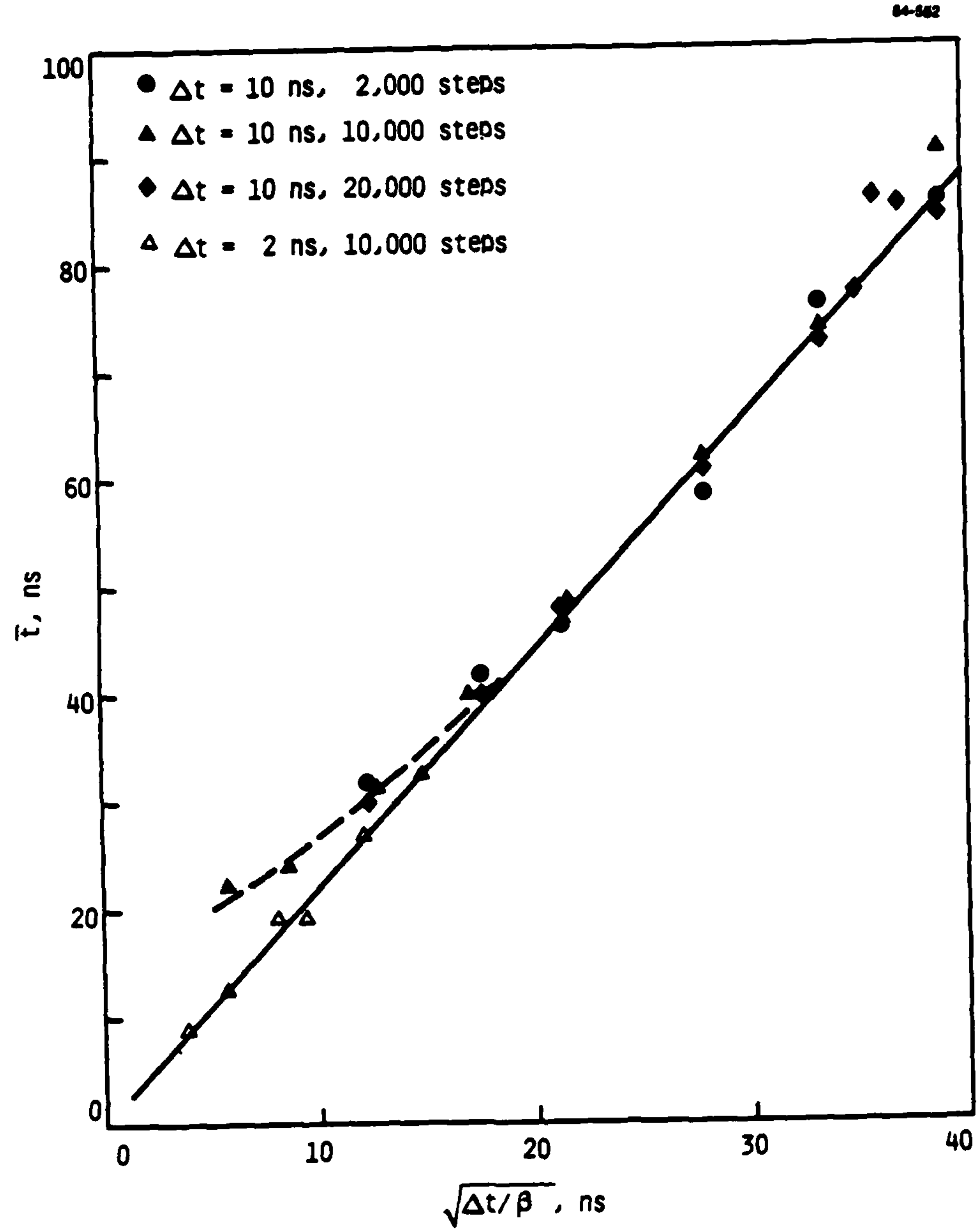


Figure 4. Correlation Between the Mean Time Between Signal Extrema and the Particle Relaxation Time, β^{-1}.

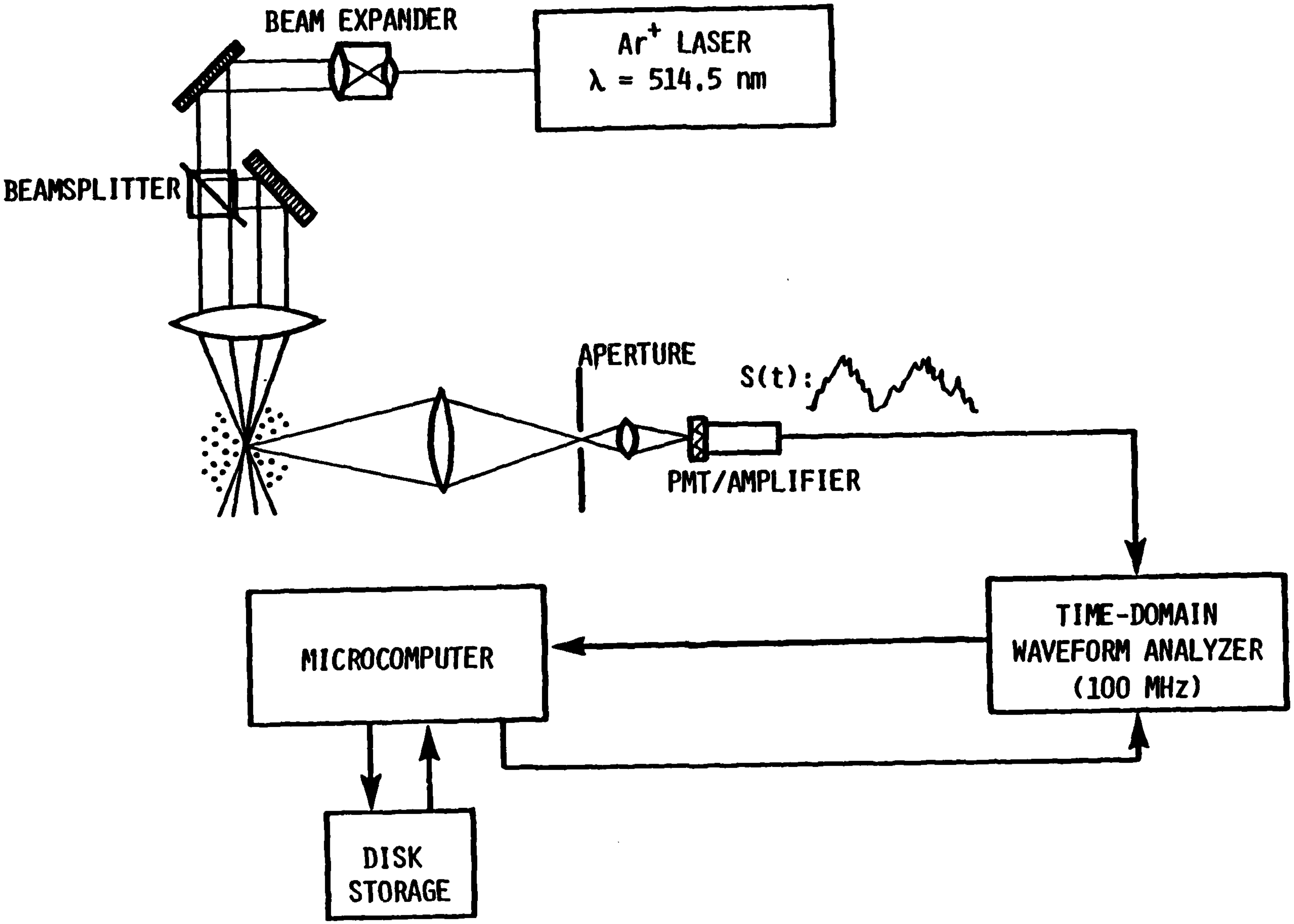


Figure 5. Schematic of Experimental Optical System for the Study of Brownian Motion Measurement Techniques.

A microcomputer-based data acquisition system with very fast time resolution capability has been assembled for this project. This system is an IBM Personal Computer XT with a CAMAC interface (Transiac Corp., Mountain View, Calif.). This system permits considerable flexibility in that any CAMAC-compatible module may be used. Fast time resolution in data acquisition is achieved with the appropriate choice of CAMAC hardware in combination with fast memories for temporary data storage. The contents of the data memories are transferred into the computer memory via a DMA interface at data rates of approximately 400 kHz. Large volumes of data may then be stored on the system's fixed disk.

Presently we have set up and tested this system with a fast amplifier and transient recorder, enabling acquisition of up to 32,000 (8-bit) data points at a maximum rate of 100 MHz (10 ns sampling interval). We anticipate that this recorder will be used in our initial experiments for testing the Brownian motion measurement concept. Because the data analysis requires statistical analysis of the time characteristics of the signal, we may ultimately find that a time-domain hardware configuration (e.g., for time distribution measurements of the signal) is more suitable. The CAMAC system provides the flexibility for using this type of data acquisition hardware without redesign of the entire data acquisition system.

2.3 Research Plan

The research plan for the second year of the program emphasizes experimental validation of the measurement concept. Specifically, we anticipate performing the following tasks:

1. We will initially test the measurement approach by measurement of the Brownian motion of individual submicron particles suspended in liquids. Commercially available particles of known size are used in these measurements, which also avoid the added complications of dispersing the particles in a gas stream. These tests, in combination with the Monte Carlo simulations, should establish

whether the signal fluctuations exhibit the expected behavior for particles of different size and should provide a crucial test for the measurement theory.

2. If the tests with liquid suspensions conclude with favorable results, we will proceed with measurements of submicron particles suspended in gas flows. These measurements will require either dispersal of the liquid-suspended particles into a gas stream or perhaps chemical generation of small particulates from the gas phase. Measurements of submicron particles suspended in well-controlled gas flows should be critical in establishing the capabilities and limitations of the technique as a particle diagnostic and should provide the foundation required for investigations of diagnostic applications in the third year of the program.

3. PUBLICATIONS

No results to date from this program have been published in technical journals. We anticipate that the work in the coming year on experimental validation of the Brownian motion measurement approach will be submitted to a journal, e.g., Journal of Applied Physics, Applied Optics, Physics of Fluids, etc., for publication.

4. PERSONNEL

The Principal Investigators for this work are Dr. Alan Stanton, who is a Principal Research Scientist at Aerodyne Research, Inc., and Dr. Wai Cheng, Senior Research Scientist (part-time) at Aerodyne and Assistant Professor of Mechanical Engineering, MIT. Dr. Stanton has concentrated his research efforts to date for this program on the development of optical and data acquisition systems for the experimental program, while Dr. Cheng has concentrated on the measurement theory, through development of the Monte Carlo Brownian motion simulation model. In addition to his work at Aerodyne on this project, Dr. Cheng has supervised the work of undergraduate and graduate students in the Department of Mechanical Engineering, MIT. This work has been in support of the Brownian motion simulation model, funded by a subcontract to MIT.

5. INTERACTIONS

Three presentations were made at AFOSR Contractors' Meetings on this work during the first year of the contract. These presentations were:

A.C. Stanton and W.K. Cheng, "Techniques for Submicron Particle Sizing", 1983 AFOSR Contractors' Meeting on Air Breathing Combustion Dynamics Research, Scottsdale, AZ, September 19-22, 1983.

W.K. Cheng and A.C. Stanton, "Single Particle Sizing by Measurement of Brownian Motion", 1984 AFOSR Research Meeting on Diagnostics of Reacting Flows, Yale University, March 21-22, 1984.

A.C. Stanton and W.K. Cheng, "Single Particle Sizing by Measurement of Brownian Motion", 1984 AFOSR/ONR Contractors' Meeting on Airbreathing Combustion Research, Pittsburgh, PA, June 20-21, 1984.

6. NEW DISCOVERIES OR INVENTIONS

The experimental approach under study in this program, i.e., determination of particle size by measurement of the Brownian motion of individual particles, is a new approach which has not previously been demonstrated. We anticipate that this approach, if successful, will permit the development of research instrumentation for the study of the growth of submicron particles, based on new concepts in measurement and data analysis.

APPENDIX

COMPUTER SIMULATION OF THE BROWNIAN MOTION SENSOR SIGNALS

Prepared by

Wai K. Cheng

APPENDIX

1. Summary

Approaches to signal processing for the Brownian motion sensor are examined by simulating the signal from such a system on the computer using a Monte Carlo method. In Section 2 the nature of the Brownian motion is discussed, and the necessary statistical tools to generate a Monte Carlo simulation of the process are introduced. Then the results of the simulation and the qualitative behavior of the signal from a laser Doppler system observing the particle are analyzed. Because the amplitude of the Brownian motion is smaller than the fringe spacing of the probing laser beam, the wavelength of the laser is not a relevant parameter, and only temporal information (and hence, not velocity) can be extracted. In Section 3, the statistics of the temporal content of the signal are examined. We find that the mean period of the signal may be related to the relaxation time, from which the mass, or size, of the particle may be calculated.

2. Simulation of the Brownian Motion Sensor Signal

A numerical simulation of the signal obtained from the Brownian motion sensor is used to study the nature of the signal, and to devise an algorithm for extracting the information pertinent to the particle size. In what follows, the method of simulating the signal is described and the qualitative nature of the signal is discussed. Then a method is described to analyze the signal.

2.1 Nature of the Brownian Motion

The motion of a particle in thermal equilibrium with the surrounding fluid is described by the Langevin equation,[1]

$$\frac{d\underline{u}}{dt} = \beta \underline{u} + \underline{A}(t) \tag{1}$$

where $\underline{u}$ is the velocity and β is the damping coefficient. The random excitation $\underline{A}(t)$ has the property that the solution of Eq. (1), which is

$$\underline{u} - \underline{u}_o e^{-\beta t} = e^{-\beta t} \int_o^t e^{\beta \xi} \underline{A}(\xi)\, d\xi \tag{2}$$

has the Maxwellian velocity distribution in the limit of large t. The time scale for approaching this limit is

$$\tau_{relax} = 1/\beta \quad . \tag{3}$$

In continuum flow ($\lambda_{mfp} << d$), for a spherical particle of diameter d and mass m, β is given by

$$\beta = 3\pi\mu\, d/m \quad , \tag{4}$$

where μ is the viscosity of the fluid medium. For the free flow limit ($\lambda_{mfp} >> d$), the value of β is given by the Epstein formula,

$$\beta = \frac{\pi d^2}{m} \rho_{air} \frac{\bar{c}}{3} (1 + \frac{\pi\alpha}{8}) \tag{5}$$

where $\bar{c}$ is the mean thermal speed of the fluid molecules and α is the accommodation coefficient. A value of $\alpha = 0.5$ is used in the simulation. Values for the relaxation time are shown in Figure A-1.

As an example, we consider a particle of 0.2 μm diameter with a specific gravity of 2. For air at 300 K and at 1 atmosphere pressure, the relaxation time is $\tau_{relax} = 150$ ns. To obtain information about the velocity

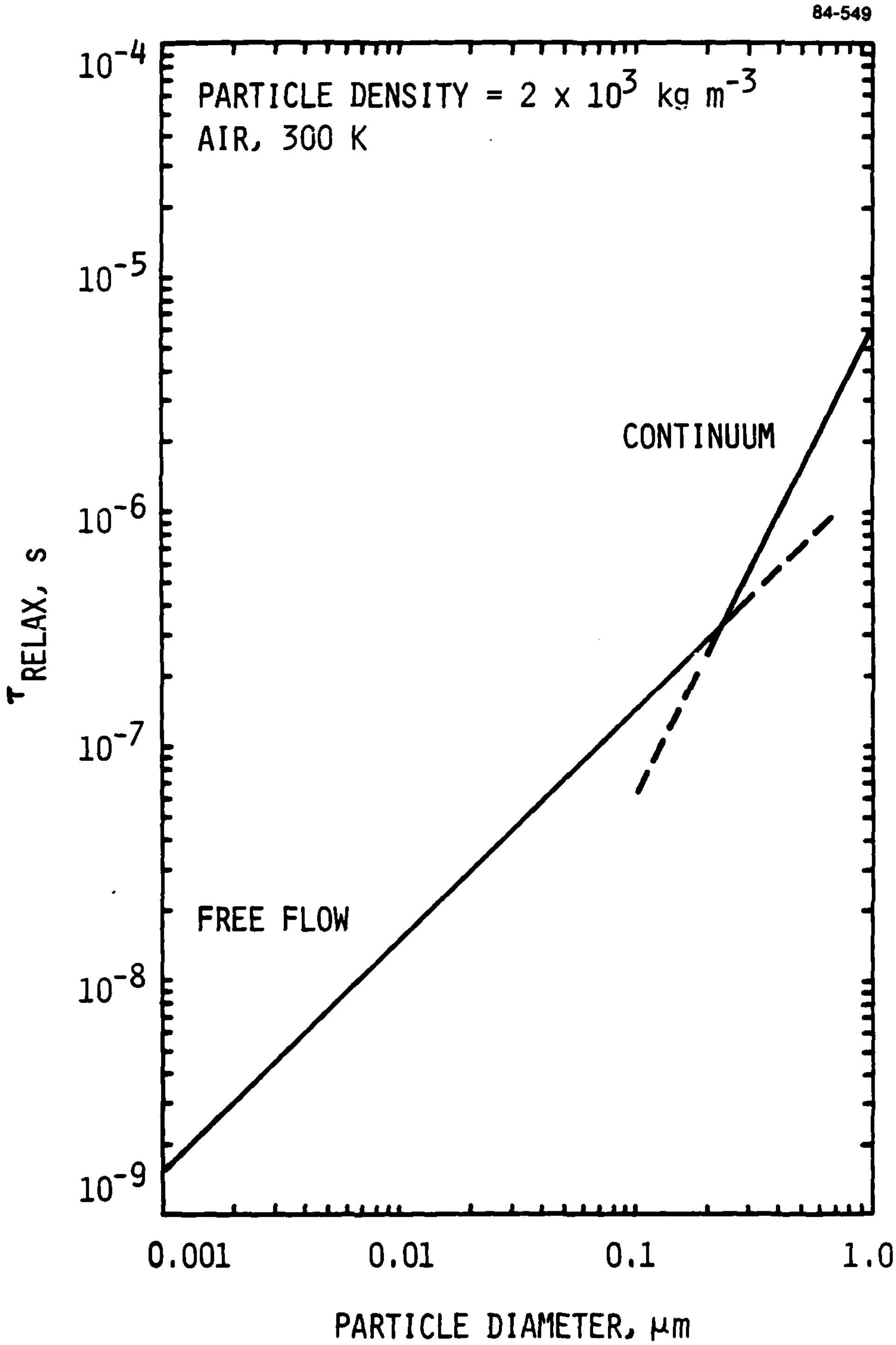


Figure A-1. Characteristic Relaxation Time for Particles in Brownian Motion.

distribution, the sampling time should be less than τ_{relax}. A sampling time Δt of 10 ns is used in the following analysis. Furthermore, a time between samples of 10 ns is assumed.

The time scale for the fluctuating excitation A(t) is of the order of the collision time τ_c between the fluid molecules and the particle, which is ~10^{-14} s. During each collision, the particle changes its velocity by

$$\frac{\sqrt{8kT\tilde{m}}}{m} \tag{6}$$

where $\tilde{m}$ is the mass of a fluid molecule. The value for δV is ~10^{-5} $m \cdot s^{-1}$.

Since the sampling time Δt is large compared to the collision time τ_c, the velocity sampled is the average over Δt of the time dependent velocity given by Eq. (2). The requirement that the velocity distribution has the asymptotic limit of a Maxwellian distribution requires the sampled velocity $\underline{w}_n$ at $n\Delta t$ to have a probability distribution of,[1]

$$p(\underline{w}_n, \Delta t \; ; \; \underline{w}_{n-1}) = \left[\frac{m}{2\pi kT \; (1-e^{-2\beta\Delta t})} \right]^{3/2} \exp \left[\frac{-m \left| \underline{w}_n - \underline{w}_{n-1} e^{-\beta\Delta t} \right|^2}{2kT \; (1-e^{-2\beta\Delta t})} \right] . \tag{7}$$

Therefore, if $\underline{w}_n = (u_n, v_n, w_n)$, and if the Doppler system is sensitive to the velocity component w, the probability distribution function for w_n is

$$p(w_n, \Delta t \; ; \; w_{n-1}) = \left[\frac{m}{2\pi kT \; (1-e^{-2\beta\Delta t})} \right]^{1/2} \exp \left[\frac{-m \; (w_n - w_{n-1} e^{-\beta\Delta t})^2}{2kT \; (1-e^{-2\beta\Delta t})} \right] . \tag{8}$$

2.2 Simulation of the Brownian Velocity

Using the probability distribution of Eq. (8), we may generate, using a pseudo-random number generator, the velocity history of w with the correct statistical properties. In general, if R is a random number with uniform distribution in the interval $[0,1]$, and $p(x)$ is the probability distribution of a variable x, R may be mapped into x by equating the probability of finding x in $[x_1, x_2]$,

$$\int_{x_1}^{x_2} p(x)\ dx = \int_{R_1}^{R_2} 1\ dR = R_2 - R_1 \quad . \tag{9}$$

In particular let $x_1 = -\infty$ and $R_1 = 0$; then

$$\int_{-\infty}^{x} p(x)\ dx = \int_{0}^{R} 1\ dR = R \quad . \tag{10}$$

For the distribution of Eq. (8), it is convenient to rewrite the expression as

$$p\ (w_n, \Delta t;\ w_{n-1}) = \frac{A}{\sqrt{\pi}} \exp \left[-\ A^2 (w_n - w_{n-1} h)^2 \right] \tag{11}$$

where

$$A^2 = \frac{m}{2kt(1-h^2)} \tag{12}$$

$$h = e^{-\beta \Delta t} \quad . \tag{13}$$

Then Eq. (10) becomes

$$\frac{A}{\sqrt{\pi}} \int_{-\infty}^{w_n} \exp\left[-A^2(w_n-w_{n-1}h)^2\right] dw = R \quad , \tag{14}$$

from which w_n may be solved from the random number R. The solution of Eq. (14) is

$$w_n = \frac{\xi_n}{A} + w_{n-1}h \quad , \tag{15}$$

where

$$\xi_n = \text{erf}^{-1}\,(2R_n-1) \quad . \tag{16}$$

To summarize, the algorithm for generating the Brownian velocity history works as follows. The initial velocity w_1 is obtained randomly from the Maxwellian distribution

$$p(w_1) = \left(\frac{m}{2\pi kt}\right)^{1/2} \exp\left(\frac{-mw^2}{2kt}\right) \quad , \tag{17}$$

or

$$w_1 = \left(\frac{2\pi kt}{m}\right)^{1/2} \text{erf}^{-1}(2R_1-1) \tag{18}$$

The subsequent velocities w_n, with sampling time Δt, are then given by Eqs. (15) and (16).

2.3 The Doppler Signal

Consider a crossed laser beam system with beam waist r_o as shown in Figure 1. The intensities of the two beams scattered by the particle at $\underline{x}$ = (x,y,z) (in a coordinate system centered on the probe volume) are

$$I_1 \sim \frac{\exp\left[- \left|\underline{x} - (\underline{x} \cdot \hat{\underline{k}}_1)\hat{\underline{k}}_1\right|^2 \right]}{r_o^2} \tag{19}$$

$$I_2 \sim \frac{\exp\left[- \left|\underline{x} - (\underline{x} \cdot \hat{\underline{k}}_2)\hat{\underline{k}}_2\right|^2 \right]}{r_o^2} \tag{20}$$

Here the unit propagation vectors are

$$\begin{aligned} \hat{\underline{k}}_1 &= (x \cos \frac{\theta_d}{2}, 0, -z \sin \frac{\theta_d}{2}) \\ \hat{\underline{k}}_2 &= (x \cos \frac{\theta_d}{2}, 0, z \sin \frac{\theta_d}{2}) \quad . \end{aligned} \tag{21}$$

Therefore the expressions for I_1 and I_2 become

$$I_{\substack{1\\2}} = \frac{\exp\left[-(x \sin^2 \frac{\theta_d}{2} \pm z \sin \frac{\theta_d}{2} \cos \frac{\theta_d}{2})^2 + y^2 + (z \cos^2 \frac{\theta_d}{2} \pm x \cos \frac{\theta_d}{2} \sin \frac{\theta_d}{2})^2\right]}{r_o^2} \tag{22, 23}$$

The mean particle trajectory is

$$\underline{x}(t) = \underline{x}_o + \underline{\bar{v}}\, t \quad , \tag{24}$$

where $\underline{x}_o$ is the reference point and $\underline{\bar{v}}$ is the mean velocity. Substitution of $\underline{x}(t) = (x(t), y(t), z(t))$ from Eq. (24) into Eqs. (19) and (20) would give the scattered intensity profile as the particle traverses the beam volume.

At the detector, because of the interference between the two beams, the light collected is a modulated Doppler signal. The signal may be written as

$$S \sim I_1 + I_2 + 2\sqrt{I_1 I_2} \cos \left(\Delta k_z \cdot \int^t v_z dt + \phi\right) \quad , \tag{25}$$

where

$$\Delta k_z = \frac{2\pi}{\lambda} \left(2 \sin \frac{\theta_d}{2}\right) \quad , \tag{26}$$

and the z velocity component contains both a mean and random velocity,

$$v_z = \bar{v}_z + w(t) \quad . \tag{27}$$

The phase shift ϕ is a constant depending on the reference point definition.

The power received at the detector may therefore be written as

$$P = \left(\frac{I_o}{2\lambda} H \int \frac{d\sigma}{dr} dr\right)$$

$$\cdot \left\{f_1 + f_2 + 2\sqrt{f_1 f_2} \cos \left(\frac{2\pi}{\lambda} \cdot 2 \sin \frac{\theta_d}{2} \cdot \left[\bar{v}_z\, t + \int_{-\infty}^{t} w(t) dt\right]\right)\right\} \quad . \tag{28}$$

The first term represents the absolute intensity function which is proportional to the beam intensity I_o, the aperture and transmissivity function H of the receiving optics, and the scattering cross-section integrated over the appropriate field of view. The functions f_1 and f_2 are defined by the right hand sides of Eqs. (19) and (20). The cosine term represents the heterodyned Doppler signal.

2.4 Simulation Results and Discussion

The Brownian velocity of a 0.1 μm diameter spherical particle with a specific gravity of 2 in 300 K air at atmospheric pressure has been simulated using the Monte-Carlo calculation described above. The velocity history is shown in Figure A-2 for 500 velocity samples with a sampling time of 10 ns each. The velocity statistics are shown in the histogram in Figure A-3. The result agrees with the Maxwellian distribution which is also plotted in the figure.

Calculations were then carried out to examine the behavior of the Doppler signal, which is described by Eq. (28). The term of interest is

$$S = \frac{1}{2}\{f_1 + f_2 + 2\sqrt{f_1 f_2} \cos (\frac{2\pi}{\lambda} \cdot 2 \sin \frac{\theta_d}{2} [v_z t + \int_{-\infty}^{t} w(t)dt] + \phi)\} \quad , \qquad (29)$$

with f_1 and f_2 defined by the right hand sides of Eqs. (19) and (20). To obtain the most favorable signal, the particle is assumed to have mean velocity in the y direction only, and is traversing the center of the sample volume along the y axis. The reference phase value is set arbitrarily to (-0.5π) which corresponds to aligning the y axis along the "boundary" of the

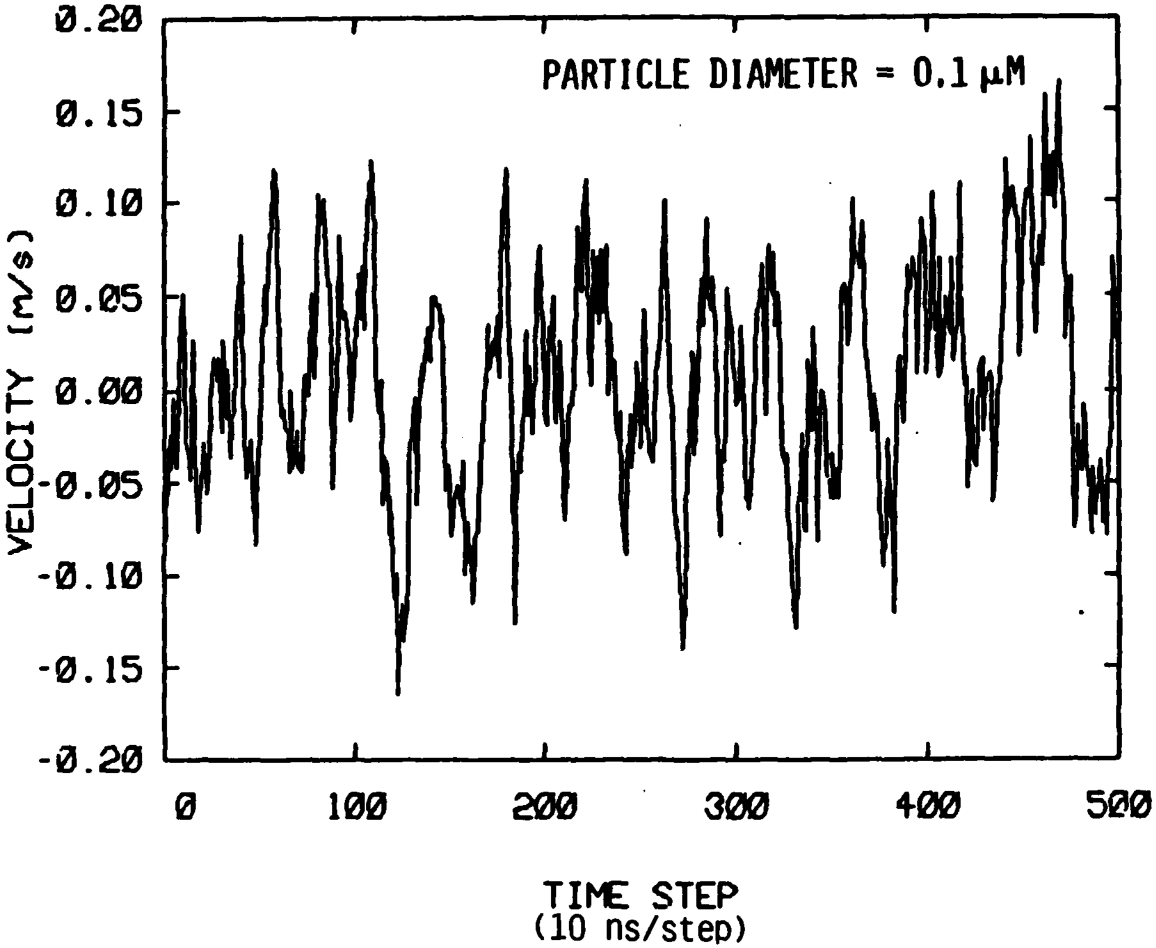


Figure A-2. Simulated Brownian Velocity History of a 0.1 μm Diameter Particle (Specific Gravity = 2) in Air at T = 300 K, p = 1 atm.

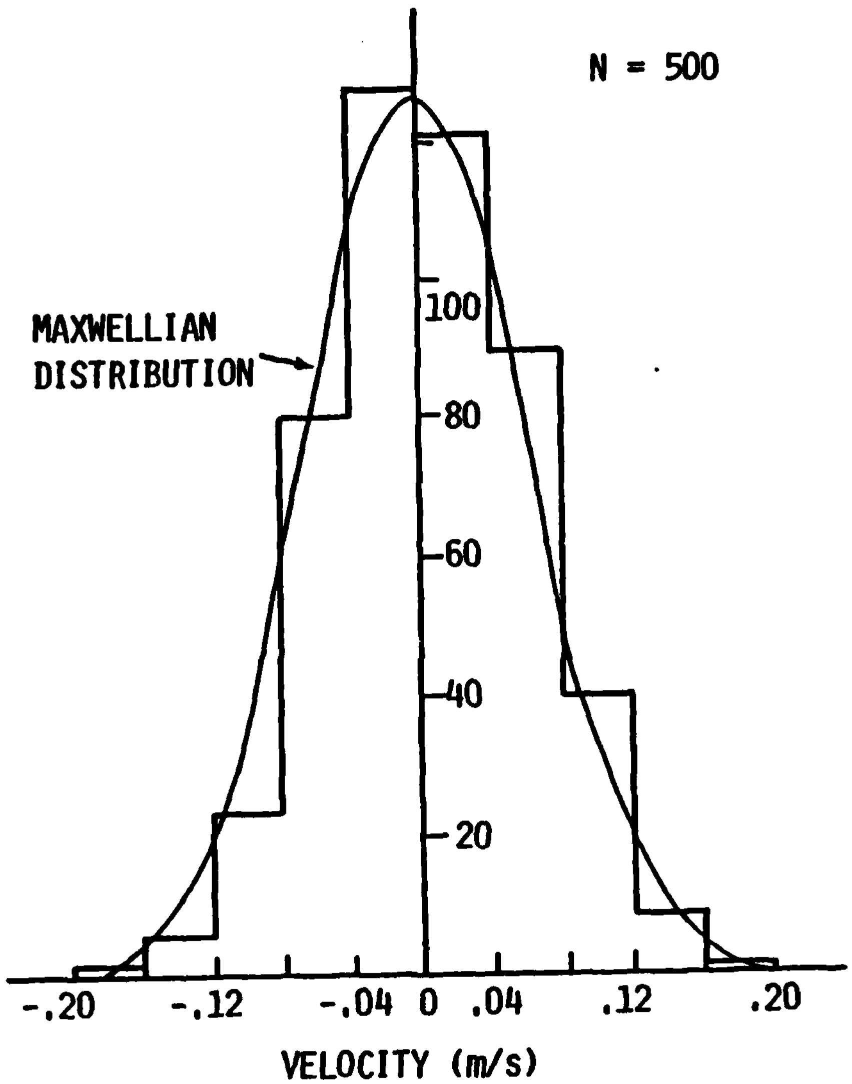


Figure A-3. Comparison of the Velocity Distribution Obtained from the Monte Carlo Simulation with a Maxwellian Distribution.

bright and dark fringes in the beam sample volume. The laser beam waist r_o is 50 μm and the beam crossing angle is set at $\theta_d = 10°$. These values are typical of a practical system.

Under the above conditions, the simulated Doppler signal for a 0.1 μm particle with $v_y = 1\ m \cdot s^{-1}$ traversing the center part of the beam volume, starting from $\underline{x}$ = (0., −10 μm, 0.), is shown in Figure A-4(a). The horizontal axis is the number of time steps, with each time step equal to 10 ns. The signal fluctuation due to the Brownian motion is clearly detectable.

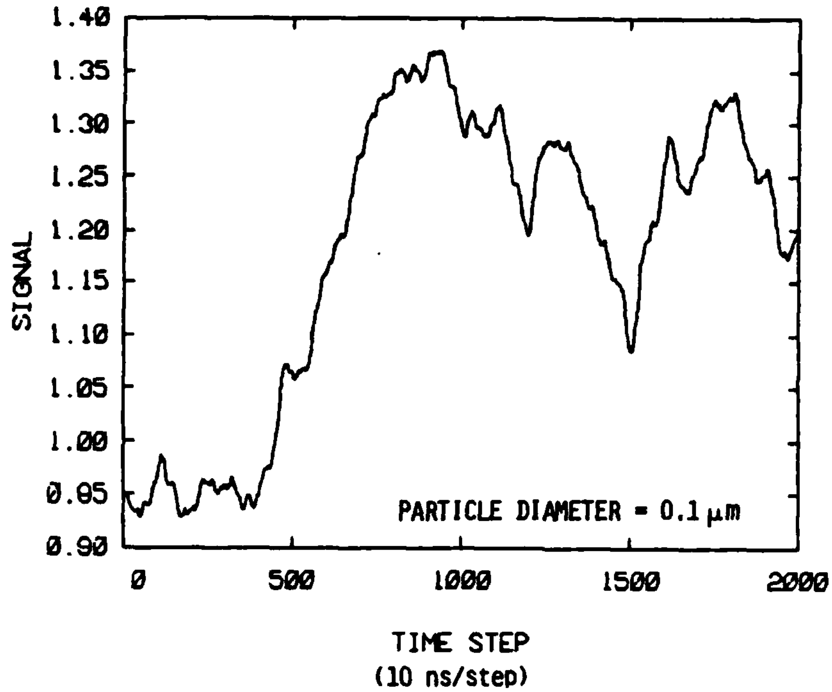
The simulation is repeated, under the same conditions, with a 0.01 μm diameter particle. The simulated signal for the smaller particle is shown in Figure A-4(b). The fluctuation is of a much higher level than those exhibited by the larger diameter particle. Furthermore, because of the behavior of the cosine function, the modulation is reduced when the argument of the cosine is close to $\pm n\pi$, n = 0,1,2,.... For the signal in Figure A-4(b), the particle is very close to the center of the beam volume as the absolute intensity is approximately constant. Therefore, the signal has a magnitude of

$$S \sim 1 + \cos \left\{\frac{2\pi}{\lambda} \cdot 2 \sin \frac{\theta_d}{2} \left[\bar{v}_z t + \int w(t)dt\right] + \phi\right\} \quad . \tag{30}$$

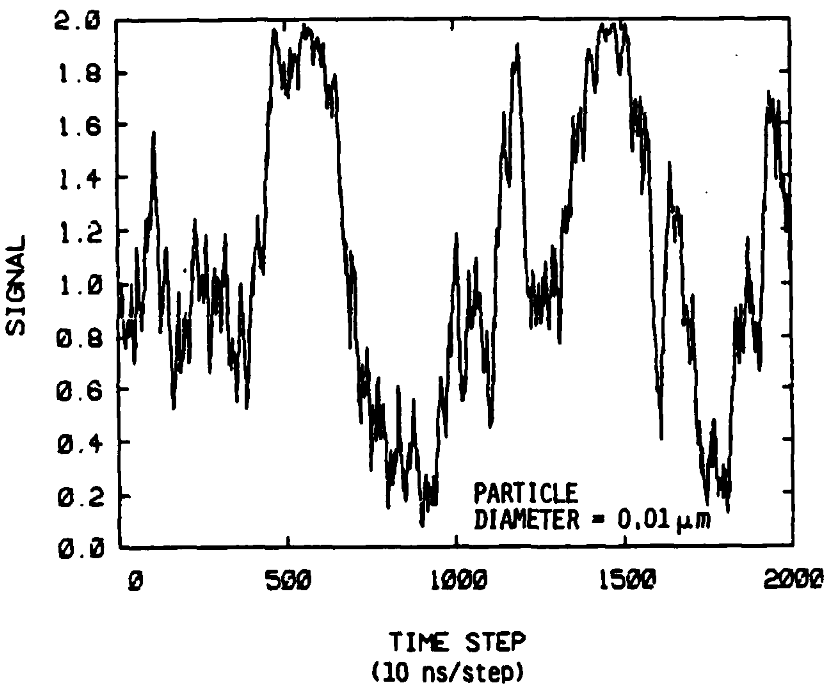
Thus, at regions where S ~1 or 2, the magnitude of the modulation is reduced. This effect is evident in Figure A-4(b).

The signal modulation in Eq. (29) is a result of the cosine term, which is rewritten here as

$$\text{modulation} \sim \cos(k'z + \phi) \tag{31}$$



(a)



(b)

Figure A-4. Simulated Time Dependent Interference Signals:
(a) 0.1 μm Diameter Particle
(b) 0.01 μm Diameter Particle

where

$$k' = \frac{2\pi}{\lambda} \cdot 2 \sin \frac{\theta_d}{z} \tag{32}$$

and

$$z = \bar{v}_z t + \tilde{z} \tag{33}$$

$$\tilde{z} = \int^t w(t)dt \quad . \tag{34}$$

The root-mean-square value $\langle\tilde{z}\rangle_{rms}$ of the Brownian displacement is shown in Figure A-5. For particles of 100 to 1000 Å diameter, $\langle\tilde{z}_m\rangle/\lambda \sim 0.1$. Therefore the rms displacement is much less than the fringe spacing k'^{-1}. As a result, the length scale λ does not play any role in the determination of the Brownian velocity w. The signal, therefore, only contains information about the time scale and not any length scale, and the determination of a meaningful velocity is, in principle, not possible.

The essence of the above discussion is illustrated in Figure A-6, which exhibits the transfer function of the detection system [the right hand size of Eq. (29)]. For typical Brownian motion modulation (AB), the signal modulation is (A'B'). The signal modulation is drastically reduced where the slope of the transfer function vanishes (CD). The amplitude of the modulation is a function of the position of the particle relative to the fringe pattern. Since this is not known a priori, the amplitude modulation, therefore, does not provide useful information on the nature of the Brownian motion.

When there is a fluid velocity component $\bar{v}_z$ perpendicular to the fringe pattern, the particle position will be represented by the trajectory (EF),

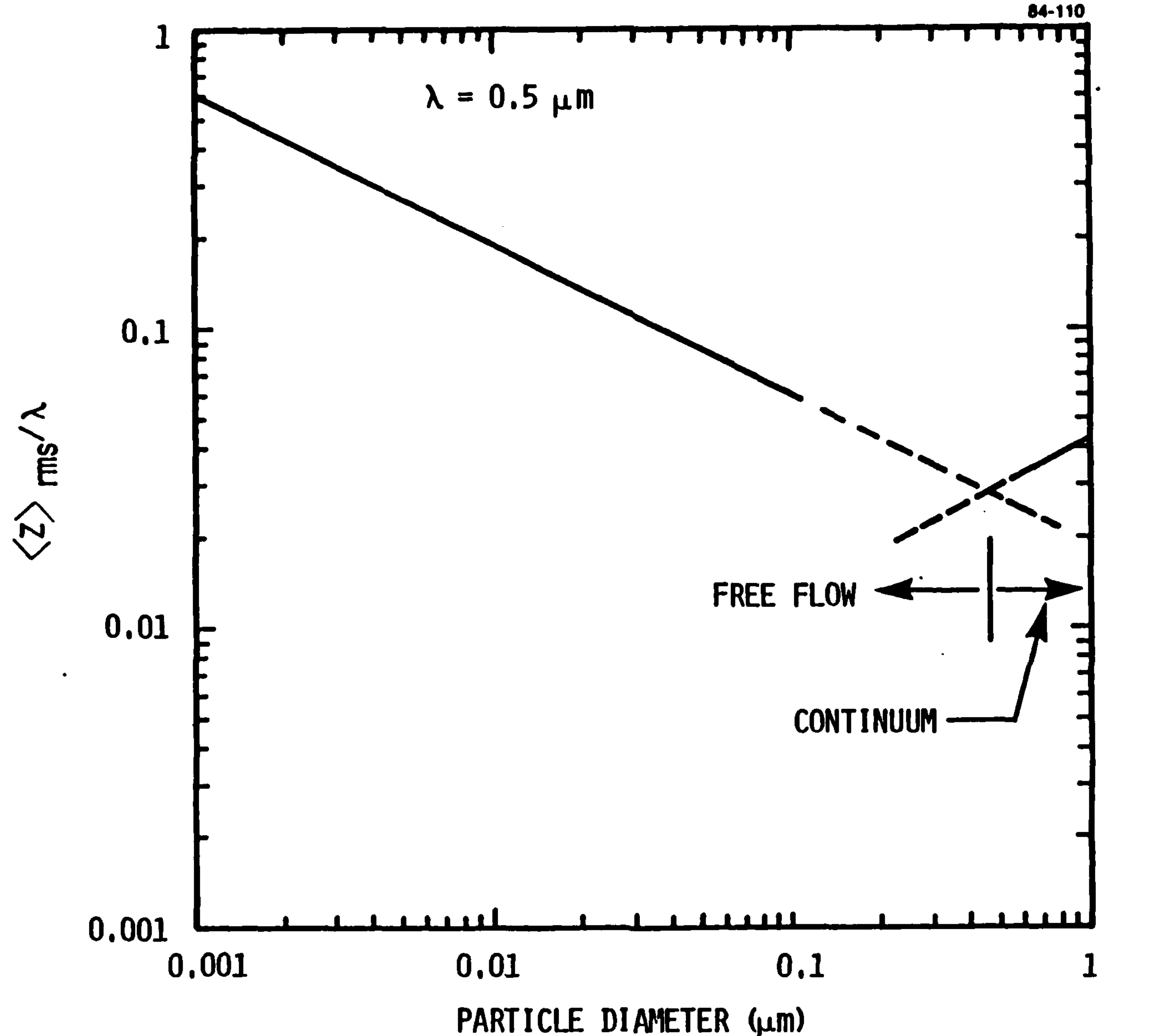


Figure A-5. Mean Excursion Distance, Compared with the Wavelength of Visible Light, for Particles (Specific Gravity = 2) in Brownian Motion in a Gas at T = 300 K, p = 1 Atmosphere.

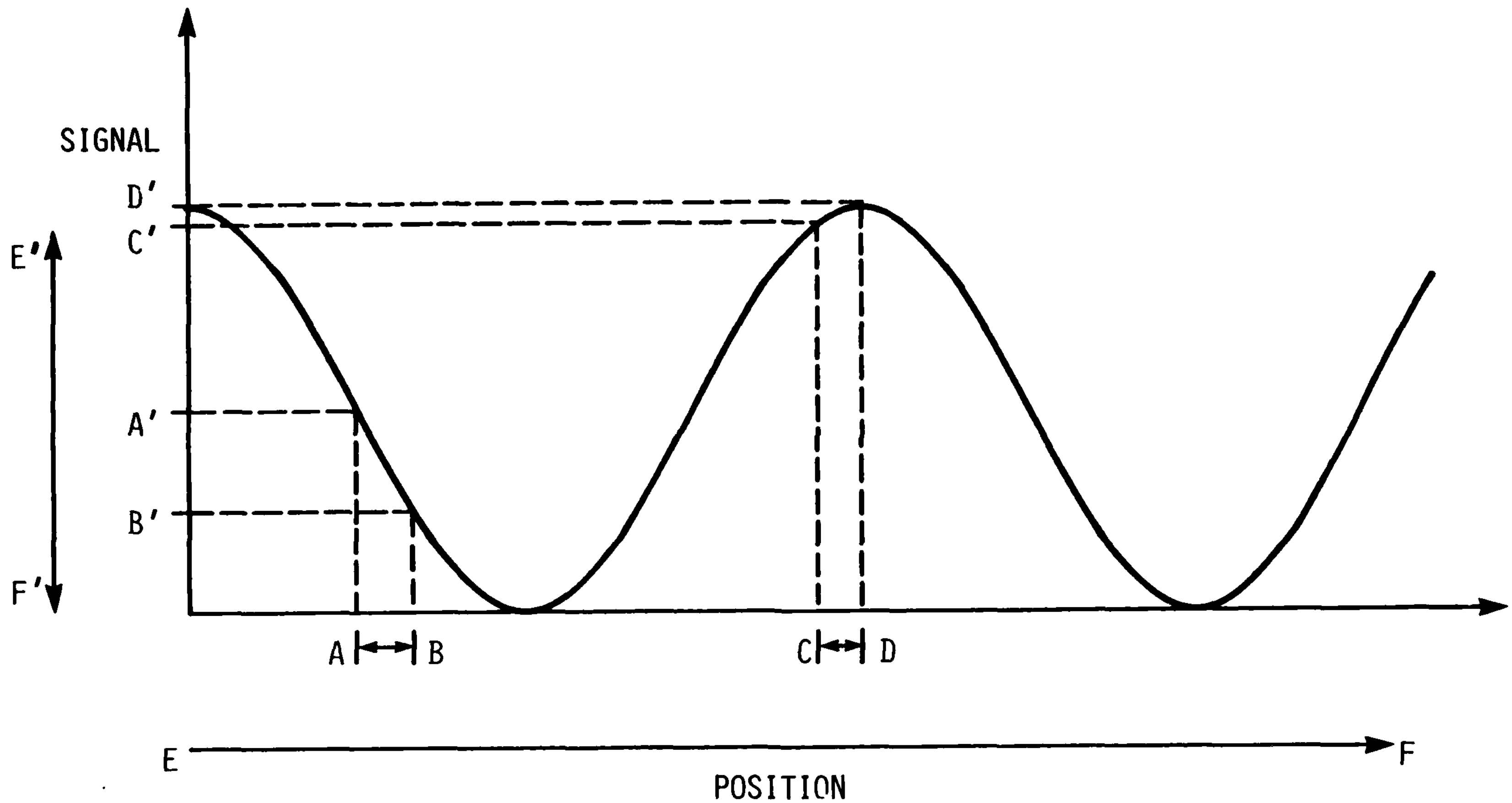


Figure A-6. Modulation of the Doppler Signal as a Function of Particle Position.

with the Brownian modulation such as (AB) superimposed on top. The signal will exhibit full modulation (E'F') (regular Doppler signal) with the Brownian motion modulation (A'B') superimposed on top. When $\overline{v}_z$ is large compared to the Brownian velocity w, it may be difficult to recover the Brownian motion modulation from the regular Doppler signal. This effect is illustrated in Figures A-7(a) and A-7(b) in which velocity $\overline{v}_z$ of 0.1 and 0.3 $m \cdot s^{-1}$ respectively are superimposed onto the Brownian velocity. In the latter case, the Brownian motion signal is almost completely masked and the signal resembles the usual Doppler velocimeter signal. For small particles with a higher Brownian velocity, good quality signal may still be obtained with a significant $\overline{v}_z$. This is illustrated in Figure A-8 by the simulation of the signal from a 0.01 μm particle with $\overline{v}_z = 0.3\ m \cdot s^{-1}$. The mean Brownian velocity $\overline{w}$, where

$$w = \sqrt{\frac{8KT}{\pi m}} \tag{35}$$

is approximately 0.1 $m \cdot s^{-1}$ for a 0.1 μm diameter particle at a gas temperature of 300 K. Thus one expects that for particles in the 0.01 to 0.1 μm diameter range, reasonable signal quality may be obtained if $\overline{v}_z$ is less than 0.1 $m \cdot s^{-1}$.

The above discussion indicates that the strategy for analyzing characteristics of the Brownian motion lies in analysis of the time information. The fundamental time scale of the fluctuation is the relaxation time $1/\beta$. The information on the particle size may be calculated from β using Eqs. (4) and (5). Furthermore, the signal must be filtered to remove the regular Doppler component ($\overline{v}_z$) for proper interpretation.

3. Processing of the Brownian Motion Sensor Signal

As discussed above, only time information may be obtained from the Brownian motion sensor. Since the time scale relevant to the Brownian motion is much faster than any other time scales (e.g., the transit time of the

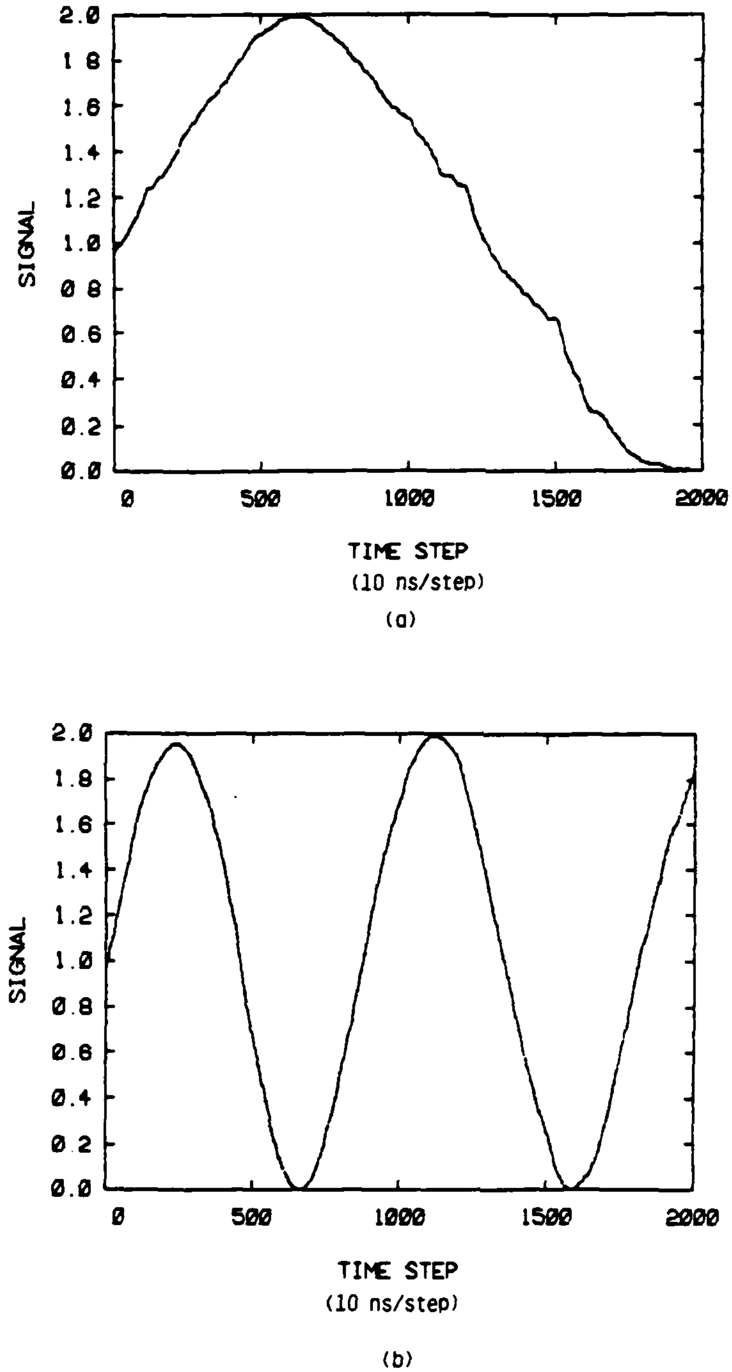


Figure A-7. Simulated Brownian Motion Signals for a 0.1 μm Diameter Particle with Fluid Velocity Component $\bar{v}_z$ Superimposed

(a) $\bar{v}_z = 0.1\ m.s^{-1}$
(b) $\bar{v}_z = 0.3\ m.s^{-1}$

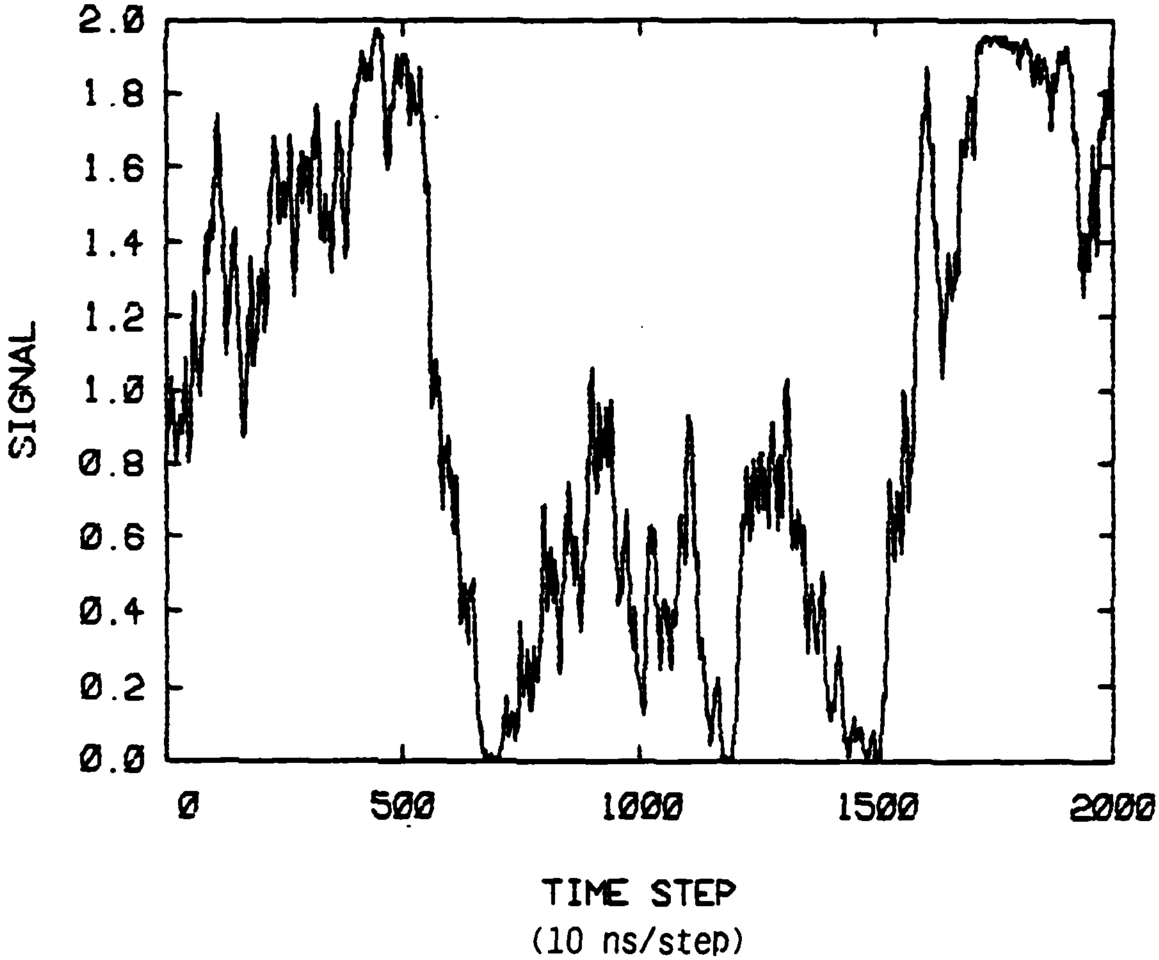


Figure A-8. Simulated Brownian Motion Signal for a 0.01 μm Diameter Particle with $\overline{v}_z = 0.3\ m.s^{-1}$ Superimposed.

particle due to its mean velocity), the latter may be filtered out easily. In the following discussion we shall assume that the signal has already been filtered to contain the Brownian information only. The appropriate statistics of the temporal information signal are discussed.

The time scale of the fluctuating signal may be analyzed in terms of the time between zero crossings (assuming that the mean has already been removed by subtraction of a moving average), or in terms of the time between extrema of the signal (zero crossing of the first derivative). The particular implementation depends on the hardware configuration. Nevertheless the principle remains the same. We shall use the time between extrema method in the following analysis. Also the analysis considers only the mean time between extrema, and does not attempt analyze the full spectral content of the signal, so that possible aliasing is not an issue. (The method amounts to doing statistical sampling rather than fixed interval sampling, so on the average, the higher frequency content is "represented.")

The signal processing is simulated on the computer using the Monte Carlo technique described previously. The Brownian motion of a particle with specific gravity of two, embedded in room temperature air, is analyzed. The mean time between extrema $\bar{t}$ as a function of the particle diameter d is shown in Figure A-9. The values of $\bar{t}$ do not seem to follow a simple linear relationship with d, notwithstanding the fact that the only time scale involved is the relaxation time $\tau_{relax} = 1/\beta$. With β proportional to 1/d in the free flow regime, the relaxation time should be proportional to d. Furthermore, depending on the sampling interval Δt, the mean time $\bar{t}$ is different. The relevant parameter should therefore clearly involve Δt. This issue is resolved when the data are replotted against $\sqrt{\Delta t/\beta}$ (with the dimension of time) (Figure A-10). The data points collapse on to a single straight line passing through the origin. The relationship between sampling and adequate statistics is also illustrated, as at large values of $\sqrt{\Delta t/\beta}$, a

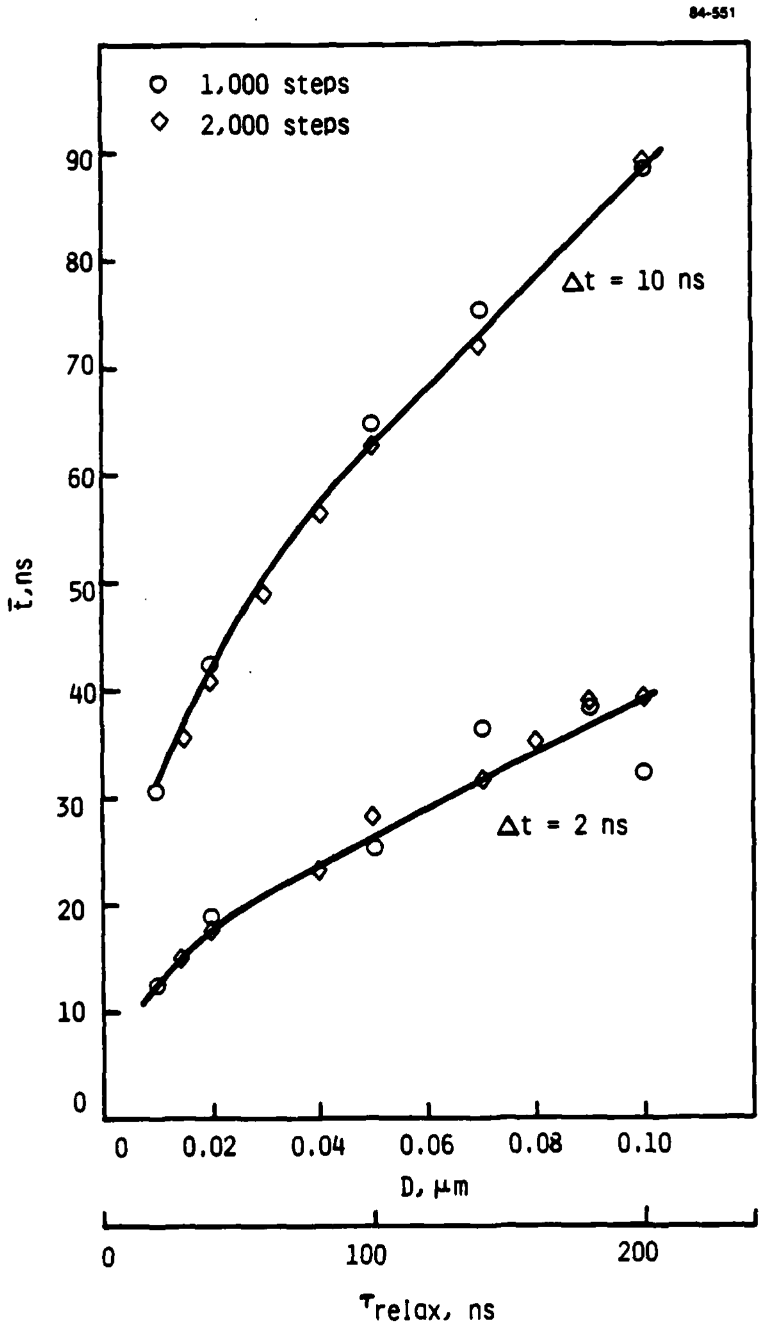


Figure A-9. Mean Time Between Extrema in Brownian Motion Interference Signal, for Two Sampling Intervals.

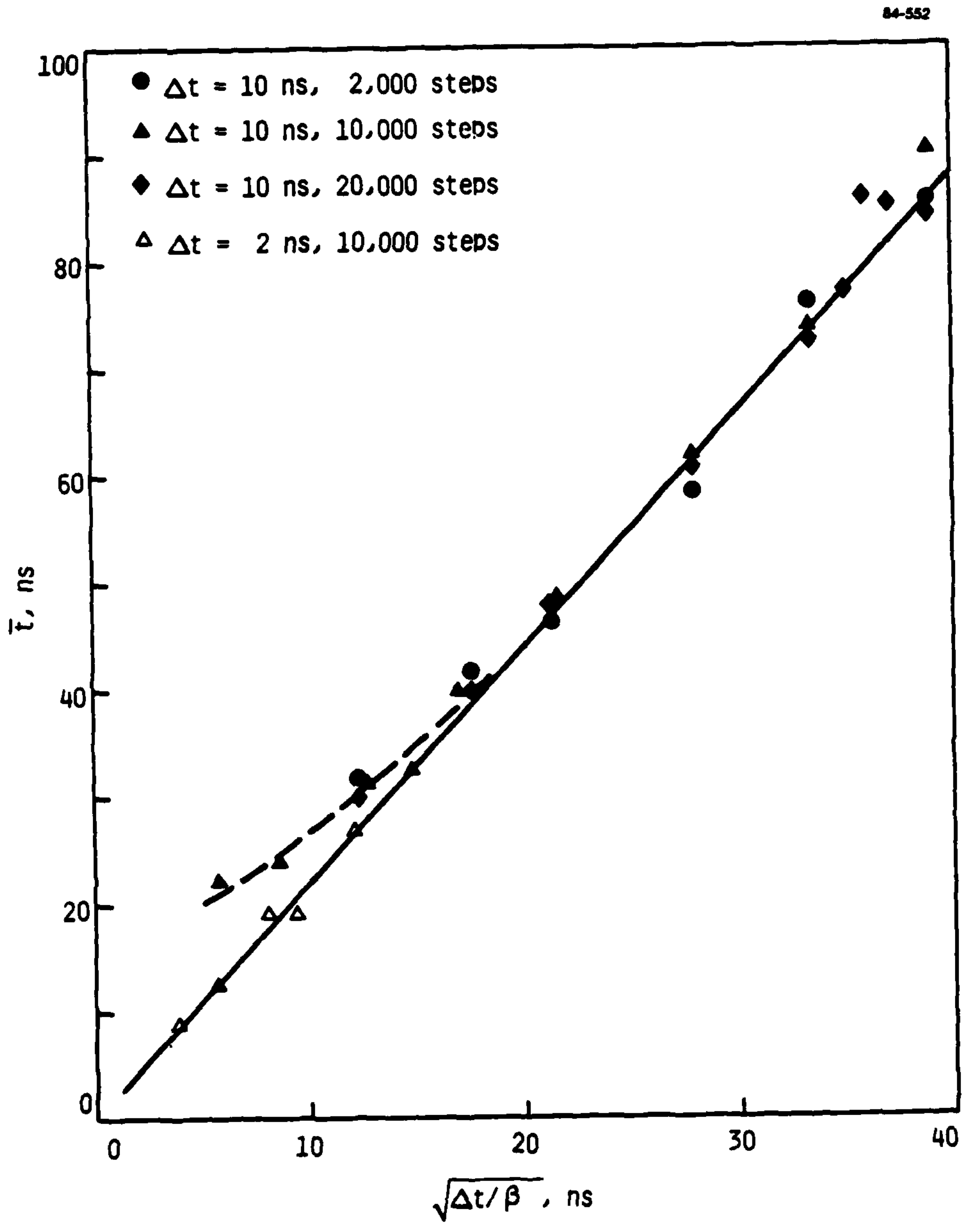


Figure A-10. Correlation Between the Mean Time Between Signal Extrema and the Particle Relaxation Time, β^{-1}.

long string of data is required to produce a good estimate of t. For small values of $\sqrt{\Delta t/\beta}$, the sampling interval of $\Delta t = 10$ ns is not adequate to resolve the Brownian motion time scale, and as a result, $\bar{t}$ is overestimated.

4. Reference

1. S. Chadrasekhar, "Stochastic Problems in Physics and Astronomy," in Review of Modern Physics, Vol. 15, 1943, pp. 1-89.

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