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FEASIBILITY STUDIES FOR LIGHT SCATTERING EXPERIMENTS TO DETERMINE
THE VELOCITY RELAXATION OF SMALL PARTICLES IN A FLUID

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The extent by which the motion of a small particle in a fluid deviates from ideal Brownian motion has been the center of considerable interest in the past few years. According to the theory of Brownian motion in its simplest form[1], the velocity autocorrelation function of a small object moving under the action of molecular bombardment in a fluid at rest decays exponentially with time. The decay constant is a function of the particle's mass and radius, and of fluid viscosity. This description, which is typical of a Markoffian system with short memory, was believed to be appropriate for Brownian motion even for times longer than the velocity relaxation time[2]. Thus it was both surprising and exciting when Alder and Wainwright[3] reported a much longer-lived velocity correlation function from molecular dynamics computer calculations. The long-time persistence of the velocity autocorrelation function, approximately a $t^{-3/2}$ decay law, is indicative of previously unsuspected memory effects at the molecular level. Subsequent theoretical studies revealed that this long-time behavior was consistent with the hydrodynamic description of the particle motion. In the hydrodynamic regime, the presistent memory of the velocity autocorrelation function has been traced to the existence of local fluid vortices surrounding the moving object. In fact, even at the molecular level, vortex-like patterns of neighboring molecules were observed around test particles[5] in computer simulations. This behavior appears to be unrelated to the detailed form of the intermolecular forces, as more recent computer calculations by Levesque and Ashurst demonstrated with a Lennard-Jones interaction potential[6].

The first clear experimental verification of the long-time tail of the velocity autocorrelation function was given by Kim and Matta[7]. These investigators used a shock wave apparatus to accelerate small latex spheres

in an atmosphere of argon or air and observed the decaying velocity in a region of the shock tube where the superposition of the forward and reflected shock waves caused the gas to come to rest locally. The experiment provided conclusive evidence that the velocity autocorrelation function did not decay exponentially for all time. The details of the long time decay, however, were shown to be rather sensitive to the particle's history prior to the beginning of the observation. Thus, the data indicated a long time tail with a decay law which was somewhat intermediate between the $t^{-1/2}$ and $t^{-3/2}$ power laws.

Attempts to exhibit the $t^{-3/2}$ power law decay by light scattering techniques have yielded less convincing evidence[8]. In these experiments, accuracy is limited by the dual requirements that many particles be illuminated and that mutual interactions between scatterers be minimized. By varying the concentration of scatterers, it is possible to satisfy the first requirement. However, it is very difficult to obtain a reliable estimate of the effect of particle interactions[9].

Over the past year, we have investigated a third approach for measuring the non-Markoffian component in the relaxation mechanism of a Brownian particle which combines desirable features of both the shock wave experiment and conventional light scattering experiments. We suggest using the radiation pressure generated by a C.W. laser to guide an individual spherical particle to terminal velocity; at an appropriate time, the beam intensity is suddenly lowered to a value at which the radiation pressure is negligible, and the ensuing velocity relaxation is measured directly. Recent experiments in optical levitation[10] have shown that spherical particles can easily be trapped and held stably in a C.W. laser beam operating in the TEM_{00} mode having a Gaussian intensity profile.

In this report we first review the theoretical background and then describe the main features of the proposed experiments which are designed to provide information on the velocity relaxation of a small particle.

Brownian motion is the apparently random motion of a small particle immersed in a fluid. The original investigations of this phenomenon were made on various objects of colloidal size. The theory, however, has been successfully applied to many other situations, from the description of the thermal noise of galvanometers, to the motion of ions and polymer molecules in solution, even to the influence of rotational diffusion on NMR line widths.

In its general form, the theory of Brownian motion need not refer to the description of a real particle at all, but instead, to the behavior of some collective property of a macroscopic system, (e.g., the instantaneous total electric dipole moment of a macroscopic sample of fluid. If $\vec{X}$ represents a typical macroscopic coordinate, its random motion can be described by the Langevin equation

$$\frac{d^2\vec{X}}{dt^2} = -\beta \frac{d\vec{X}}{dt} + \vec{F}(t) , \qquad (1)$$

where $-\beta d\vec{X}/dt$ represents the macroscopic damping force and $\vec{F}(t)$ is the total external force acting on the appropriate degree of freedom. Usually $\vec{F}(t)$ is composed of various deterministic contributions and of a random part which simulates the uncontrollable collision effects. If the random component of the force $\vec{F}(t)$ has a very short memory (or if it is δ- correlated), the velocity correlation function will decay exponentially to zero with a rate constant which is given by the damping rate β:

$$\langle v(t) \ v(0) \rangle \approx e^{-\beta t} . \qquad (2)$$

For a spherical particle in a homogeneous fluid, for example, one finds

$$\beta = \frac{6\pi\eta R}{m} , \qquad (3)$$

where R is the radius of the particle, m is its mass, and η is the fluid shear viscosity.

A more realistic description of the random motion of a particle in a fluid is obtained by the addition of the so-called Basset-Boussinesq non-local correction which takes into account drag memory effects. The modified Langevin equation takes the form

$$m \frac{d\vec{v}}{dt} = -6\pi\rho R\vec{v} - \frac{2}{3}\pi\rho R^3 \frac{d\vec{v}}{dt} - 6R^2\sqrt{\pi\eta\rho} \int_{-\infty}^{t} \left[\frac{d\vec{v}}{ds} \frac{ds}{\sqrt{t-s}} \right] + \vec{F}(t) \qquad (4)$$

where ρ is the fluid density. The first term on the right hand side is the steady state Stokes drag. The second term is the inertial correction associated with the fact that the sphere must displace a volume of fluid equal to its own volume before it can proceed forward. The third term is the Basset-Boussinesq term which arises from the analysis of the linearized Navier-Stokes equation; it introduces memory effects as a result of streamlines induced in the fluid by the particle motion. The last term contains possible additional deterministic forces and the usual random force describing the collisions of the Brownian particle with the molecules of the fluid.

If a particle is injected in the fluid with a non-zero initial velocity, the velocity relaxation to equilibrium is described on the average by an equation with the same formal structure as the modified Langevin equation (4) without the random force (since the average random force is zero). The linearity of this equation also insures that the velocity autocorrelation function will relax to its equilibrium value according to the same equation as that satisfied by the average velocity. The key point in this discussion is that the presence of the non-local term causes a significant departure for long times from the exponential decay which is expected for both the average velocity autocorrelation function according to the simple Langevin description. Thus, we can obtain the long time behavior of the autocorrelation function by measuring the long time behavior of the average velocity. From Eq. (4) we see that if $\frac{dv}{ds} < 0$ for all $s < t$, the Basset term causes a delay in the damping

process so that the particle drift velocity approaches zero over a longer time scale than the usual characteristic damping time m/(6πηR) of ordinary Brownian motion.

This is not very surprising from a hydrodynamic point of view (the streamlines of the perturbed fluid curl around the moving sphere and "push" it from behind in the direction of the motion). What is surprising is that Eq. (4), in spite of its complicated structure, can be solved exactly if the sphere's initial velocity is constant, $v(t < 0) = v_0$, and if the mean value of the external force is also constant, $\langle F(t) \rangle = F$. First we consider the case $F = 0$. Because Eq. (4) is linear in the velocity v, the average drift velocity of the Brownian particle satisfies the same equation as (4) without the random driving force. In the following development we will have no further need to consider the random velocity v so that without danger of confusion, the symbol $v(t)$ from now on will denote the average drift velocity. Its equation of motion is obtained by taking the average value of each term in Eq. (4):

$$\frac{dv}{dt} + \beta v + \gamma \int_0^t ds \; \frac{1}{\sqrt{t-s}} \; \frac{dv}{ds} = 0 \qquad (5)$$

where

$$\beta = \frac{+6\pi\eta R}{m+m_f} \qquad (6)$$

$$\gamma = \frac{6R^2(\pi\eta\rho)^{1/2}}{m+m_f} \qquad (7)$$

and

$$m_f = \frac{2\pi}{3} R^3 \rho \qquad (8)$$

On the surface it might appear that the drift motion of the Brownian particle is controlled independently by its Stokes damping rate β and by the strength γ of the memory term. In reality, the memory effect in the context of this model imposes a kind of "universal" deviation from the usual Brownian motion

if the time scale of the evolution is measured in units of β^{-1}. We can see this clearly if we introduce the dimensionless time $\xi = \beta t$ in Eq. (5). The evolution equation becomes

$$\frac{dv}{d\xi} + v + \frac{\gamma}{\sqrt{\beta}} \int_0^\xi d\xi' \; \frac{dv}{d\xi'} \; \frac{1}{\sqrt{\xi - \xi'}} = 0 \qquad (9)$$

Hence the extent to which the Basset term affects the particle velocity is controlled uniquely by the parameter

$$\frac{\gamma}{\sqrt{\beta}} = \frac{3}{\sqrt{\pi}} \sqrt{\frac{m_f}{m + m_f}} = \frac{3}{\sqrt{\pi}} \sqrt{\frac{\rho_f}{2\rho_s + \rho_f}} \qquad (10)$$

where ρ_s is the mass density of the sphere. Surprisingly enough, the scaled equation is no longer governed directly by the fluid shear viscosity η so that different fluid media with the same mass density ρ_f will have the same effect on the particle velocity. This is, of course, true only if the observed time dependence of the drift velocity is analyzed on a time scale such that the unit of time corresponds to one Stokes decay time.

Equations (9) and (10) also indicate that in order to enhance the deviation from ordinary Brownian motion, one should use a fluid medium with a relatively high mass density. This, in fact, is not entirely feasible because high fluid density also implies large shear viscosity and short Stokes relaxation times. Thus, time resolution problems may become serious if one should consider a fluid-like water where the Stokes relaxation time for 10 μm radius particle is only of the order of 20 μsec.

By contrast, a 10 μm radius particle in air at a pressure of 1 atmosphere and a temperature of 20°C relaxes to equilibrium in a time of the order of 1 msec., which is easily measured, e.g., by laser Doppler spectroscopy techniques. However, as noted above, the relative importance of memory effects

decreases when the fluid density is small. Thus, in the case of 10 μm glass beads in air at standard conditions, one finds $\frac{\gamma}{\sqrt{\beta}} \approx 4 \times 10^{-2}$.

We now consider the solution of Eq. (9) in order to put our comments on a more quantitative basis. The exact solution of Eq. (9) can be derived by a rather straightforward application of Laplace transform techniques. The velocity decay is rather complicated:

$$\frac{v(\xi)}{v_0} = \frac{\mu+\sqrt{\mu^2-1}}{2\sqrt{\mu^2-1}} \, \exp\left[(\mu-\sqrt{\mu^2-1})^2\,\xi\right] \, \mathrm{erfc}\left[(\mu-\sqrt{\mu^2-1})\sqrt{\xi}\,\right]$$

$$- \frac{\mu-\sqrt{\mu^2-1}}{2\sqrt{\mu^2-1}} \, \exp\left[(\mu+\sqrt{\mu^2-1})^2\,\xi\right] \, \mathrm{erfc}\left[(\mu+\sqrt{\mu^2-1})\sqrt{\xi}\,\right] \tag{11}$$

where
$$\mu = \frac{\gamma}{2}\sqrt{\frac{\pi}{\beta}} \tag{12}$$

We can obtain the long-time behavior of the velocity by performing an asymptotic analysis of Eq. (11) in the limit $\xi \to \infty$ (since $\xi = \beta t$) with the result

$$v(\xi) \sim \xi^{-1/2} \,. \tag{13}$$

We are thus able to demonstrate mathematically how collision memory effects lead to an inverse power law decay in the velocity relaxation over long times, which should be compared to the exponential decay of simple Brownian motion.

Up to this point, we have considered decay in the absence of external forces. However, if the external forces F in Eq. (4) are constant, we can easily obtain the velocity decay from Eq. (11) by rescaling the velocity. In fact, we simply subtract the quantity $\widetilde{F} = F/[(m + m_f)\beta^2]$ from $v(\xi)$ and v_0.

In view of the size of the memory term parameter $\gamma/\sqrt{\beta}$, we have also developed an accurate perturbative solution to first order in $\gamma/\sqrt{\beta}$, which although not limited to constant F, can be applied to the case F = 0. After taking a Laplace transform of Eq. (5) we find

$$\frac{\widetilde{v}(s)}{v_0} = \frac{1 + \gamma'/\sqrt{s}}{s+1+\gamma'/\sqrt{s}} \tag{14}$$

where

$$\gamma' = \frac{\gamma}{\sqrt{\beta}} \sqrt{\pi} \tag{15}$$

$$\widetilde{v}(s) = L(v(\xi)) \tag{16}$$

To first order in γ', Eq. (14) takes the form

$$\frac{\widetilde{v}(s)}{v_0} = \frac{1}{s+1} + \frac{\gamma'}{\sqrt{s} \, (s+1)^2} + O(\gamma'^2) \tag{17}$$

Eq. (17) can be easily inverted with the result

$$\frac{v(\xi)}{v_0} = e^{-\xi} + \frac{2\gamma}{\sqrt{\beta}} e^{-\xi} \int_0^{\sqrt{\xi}} e^{\lambda^2} \, d\lambda - \frac{2\gamma}{\sqrt{\beta}} \xi^{1/2} \, {}_1F_1(2,3/2;-\xi) \tag{18}$$

The second term on the right hand side of Eq. (18) is the so-called Dawson function[11]; the third is the better known confluent hypergeometric function. It is clear that for short times ($\xi \leq 1$) the dominant contribution is the exponential term that describes the usual relaxation of a Brownian particle (this is especially true because of the smallness of the term $\gamma/\sqrt{\beta}$). For long times instead, a careful asymptotic expansion of the Dawson function and of the confluent hypergeometric function leads to the following limiting behavior of $v(\xi)$

$$\frac{v(\xi)}{v(0)} \to \frac{1}{2} \frac{\gamma}{\sqrt{\beta}} \xi^{-1/2} (1+\xi^{-1}+ \cdots) \tag{19}$$

Clearly, the asymptotic behavior of the drift velocity for a particle subject to a zero external force (for example, in a gravity-free environment) is drastically different from an exponential. There remains to see at what stage of the evolution a clear depature from the memory-free behavior becomes observable. This is shown in Fig. 1 for several values of the ratio $\gamma/\sqrt{\beta}$.

The experimental tests which we have proposed to study memory effects in an earthbound laboratory, envision the suspension of a small glass bead in a

powerful (a few watts) focused C.W. laser beam. The particle is initially at rest in air at various pressures and can be trapped stably in the restoring radiation pressure field provided by the Gaussian laser beam. If the beam intensity is suddenly changed by a small amount, the particle will seek a new equilibrium position undergoing a modification of the usual damped harmonic motion. The modification will come about as a result of the memory term in the equation of motion.

The Langevin equation for the drift velocity of a particle subject to viscous damping and a harmonic restoring force has the form

$$\frac{dv}{d\xi} + v + \frac{\gamma}{\sqrt{\beta}} \int_0^t d\xi' \; \frac{dv}{d\xi'} \; \frac{1}{\sqrt{\xi - \xi'}} = -kz \qquad (20)$$

where ξ is the usual dimensionless time, k is the scaled restoring force constant and z is the instantaneous distance of the particle from equilibrium. Unlike the case of a free particle where an exact solution of the Langevin equation can be obtained without much effort in this case, the Laplace inversion requires the solution of a cubic equation. It is clearly preferable to forgo the search for an analytic solution, and to take advantage instead of the smallness of $\gamma/\sqrt{\beta}$ to obtain a perturbative solution which is correct up to terms of order $(\gamma/\sqrt{\beta})^2$. We consider again the Laplace transform of Eq. (20) and eliminate the variable $z(\xi)$ with the help of the relations

$$\frac{dz}{d\xi} = v$$

$$s\,\tilde{z}(s) - z_0 = \tilde{v}(s) \qquad (21)$$

where $\tilde{z}(s) \equiv L\,(z(\xi))$ and $z_0 = z(\xi = 0)$. After some minor algebraic steps we find

$$\bar{v}(s) = \frac{(1 + \gamma'/\sqrt{s}) \, v_0 - k \, z_0/s}{s + 1 + \gamma'\sqrt{s} + k/s}$$

$$= \frac{s \, v_0 - k \, z_0}{s^2 + s + k} + \gamma'\sqrt{s} \; \frac{[k \, z_0 + v_0] \, s + k \, v_0}{(s^2 + s + k)^2} \tag{22}$$

where $\gamma' \equiv \frac{\gamma\sqrt{\pi}}{\sqrt{\beta}}$. The inversion of Eq. (22) is rather laborious, but straightforward. The result is

$$v(\xi) = A \, e^{-P\xi} + B \, e^{-Q\xi}$$

$$- \frac{2\gamma \, C}{\sqrt{\beta}} \left\{ \sqrt{P} \; e^{-P\xi} \int_0^{\sqrt{P\xi}} e^{\lambda^2} \, d\lambda - \sqrt{Q} \; e^{-Q} \int_0^{\sqrt{Q\xi}} e^{\lambda^2} \, d\lambda \right\}$$

$$+ \frac{2\gamma}{\sqrt{\beta}} \, \xi^{1/2} \, [D \, {}_1F_1 \, (2, 2/2; \, -P\xi) + E \, {}_1F_1 \, (2, 3/2; \, -Q\xi)] \tag{23}$$

Before we proceed to identify the numerous parameters of this solution, we must point out at once that the last two terms on the right hand side of Eq. (23) represent the memory correction. Furthermore, the evolution of $v(\xi)$ proceeds according to two distinct time scales set by the rates P and Q. We now discuss the various symbols appearing in Eq. (23). The parameters P and Q are solutions of the quadratic equation $s^2 + s + k = 0$ (see Eq. (22)), i.e.,

$$P = \frac{1}{2} (1 + \sqrt{1 - 4k}) \tag{24}$$

$$Q = \frac{1}{2} (1 - \sqrt{1 - 4k}) \tag{25}$$

They can be real or complex as with the ordinary problem of a damped harmonic oscillator. The constants A and B which govern the ordinary (non-memory related) part of the solution are given by

$$A = \frac{k \, z_0 + P \, v_0}{P-Q} \tag{26}$$

$$B = \frac{k \, z_0 + Q \, v_0}{Q-P} \tag{27}$$

Note that in the typical experimental situation which is being considered we expect to have $v_o = 0$

$$C = - \frac{k z_o + v_o - 2k v_o}{(P - Q)^3} \qquad (28)$$

$$D = \frac{k v_o - P(k z_o + v_o)}{(P - Q)^3} \qquad (29)$$

$$E = \frac{k v_o - Q(k z_o + v_o)}{(P - Q)^2} \qquad (30)$$

We can easily verify that if $k \to 0$, Eq. (23) properly reduces to Eq. (18) (force free case). The long time limit of Eq. (23) however provides us with a surprising result which helps in dispelling some of the early attempts in the literature to associate excessive fundamental significance to the $\xi^{-1/2}$ behavior of the long time force free case. In fact, the long time limit of Eq. (16) is seen to be of the form

$$v(\xi) \sim \frac{\gamma}{2\sqrt{\beta}} \left[C \left(\frac{1}{Q} - \frac{1}{P} \right) - \left(\frac{E}{Q^2} + \frac{D}{P^2} \right) \right] \xi^{-3/2} \qquad (31)$$

The power law is now of the type $\xi^{-3/2}$ indicating that the asymptotic behavior is apparently a sensitive function of the external field of force.

We should note here that while the solution of Eq. (23) converges to the one obtained in the force free case for $k \to 0$, it is no longer possible to try to compare Eq. (31) with its long time counterpart in the $k \to 0$ limit. This is because as $k \to 0$, the parameter Q approaches zero thus violating the asymptotic conditions for $P\xi$ and $Q\xi$ which are necessary in order to derive Eq. (31).

It is interesting to observe that in the overdamped case ($k < 1/4$), three phases of the relaxation should be investigated carefully:

a) $P\xi$ and $Q\xi < 1$

   In this case, the motion is essentially indistinguishable
   from that of an overdamped oscillator

b) $P\xi > 1$ but $Q\xi < 1$

   Here, one should be able to detect evidence of memory
   effects

c) $P\xi \gg 1$, $Q\xi \gg 1$

   This corresponds to the asymptotic decay region which is
   governed by the power law (Eq. (31)). This situation appears
   difficult to study experimentally because of the long time
   scale involved.

In addition to the exact and perturbative solutions to special cases of

the modified Langevin equation, we also constructed a numerical solution which,

is applicable to arbitrary forces F(t). This solution was based on the Adams-

Bashforth-Moulton predictor/corrector technique for solving differential equa-

tions. In extensive tests between the numerical and analytical solutions,

agreement to four significant digits was demonstrated throughout the region

of interest. The numerical solution can be used to predict the motion of a

levitated glass bead when the laser undergoes small sinusoidal fluctuations

in power. By varying the frequency of these fluctuations, the resonance of

the system can be probed to yield critical information on the size of the

memory term. This point, however, needs additional study, and will be

analyzed in subsequent progress reports.

We discuss now a few possible ways to verify the predictions detailed

above. We are especially interested in measuring the velocity relaxation

of a small glass sphere (diameter of the order of 10-40 μm) in a gas. In

an earthbound laboratory, a possible way to accomplish this goal is to

levitate the particle using the radiation pressure produced by a focused

Argon-Ion laser having a Gaussian intensity profile. As shown schematically

in Fig. 2, the laser beam is deflected upward and focused on a collection of particles resting on a microscope slide. A cylindrical piezoelectric crystal is used to break the Van der Waals attraction between the glass beads and the slide. With some careful handling, a sphere can be trapped in the laser beam and propelled upward above the focal plane of the lens where the effects of gravity, radiation pressure and radiometric forces (if present) balance each other out. The beam profile is such that the suspended spheres are trapped stably as a result of radial restoring forces if sufficient precaution is exercised in shielding the system from external disturbances (air drafts, mainly). Once the particle is levitated, the laser power is to be suddenly increased or decreased to a new level by means of an electro-optic device and the ensuing motion will be measured.

A direct recording of the motion can be done with a high speed streak camera of the rotating drum type. The required resolution of 0.1 μsec appears to be available commercially. At a subsequent time, we will develop an optical heterodyne detection scheme for measuring the particle velocity directly through the Doppler shift of the backscattered light. Real-time frequency measurements of the beat signal between a reference beam and the Doppler shifted one will give direct information on the relaxation process of interest. These measurements appear quite feasible because of the high intensity of the backscattered radiation, but it is difficult to predict how accurate the readings can be for the required long times. Of special interest is the analysis of the scaling relation developed in the theoretical section. For this purpose, we will modify the cell design (Item 6, Fig. 2) so that the atmospheric pressure can be varied in a controlled way. This is, of course, quite easy to do because the required operating pressures will be well within the reach of a small roughing pump (a few torr to a few hundreds

of torr). Under low pressure conditions, the particle motion toward the perturbed equilibrium configuration should be underdamped and easy to follow with moderate time resolution.

It appears interesting to force a sinusoidal modulation of the laser intensity at various frequencies and to observe the particle response. As the modulation frequency is varied across the particle resonance, additional information on the non-Markoffian components of the velocity will be sought.

Certain aspects of the velocity relaxation cannot be measured in an earthbound laboratory owing to the presence of gravity. In particular, the $t^{-1/2}$ power law derived previously in this paper applies only to velocity relaxation in the absence of forces. It is this relaxation which we will measure in a gravity-free enivronment. Now, the laser can be used to propel a sphere to terminal velocity in the scattering cell. After the laser is reduced to the level of a probe signal, the velocity relaxation will be measured using the same heterodyne detection system. Other systems for zero-gravity experiments may also be developed. Since there is no gravitational force to overcome, low power lasers may be used to propel the spheres. In addition, the spheres may be trapped by configuring two laser beams of equal intensity antiparallel to each other.
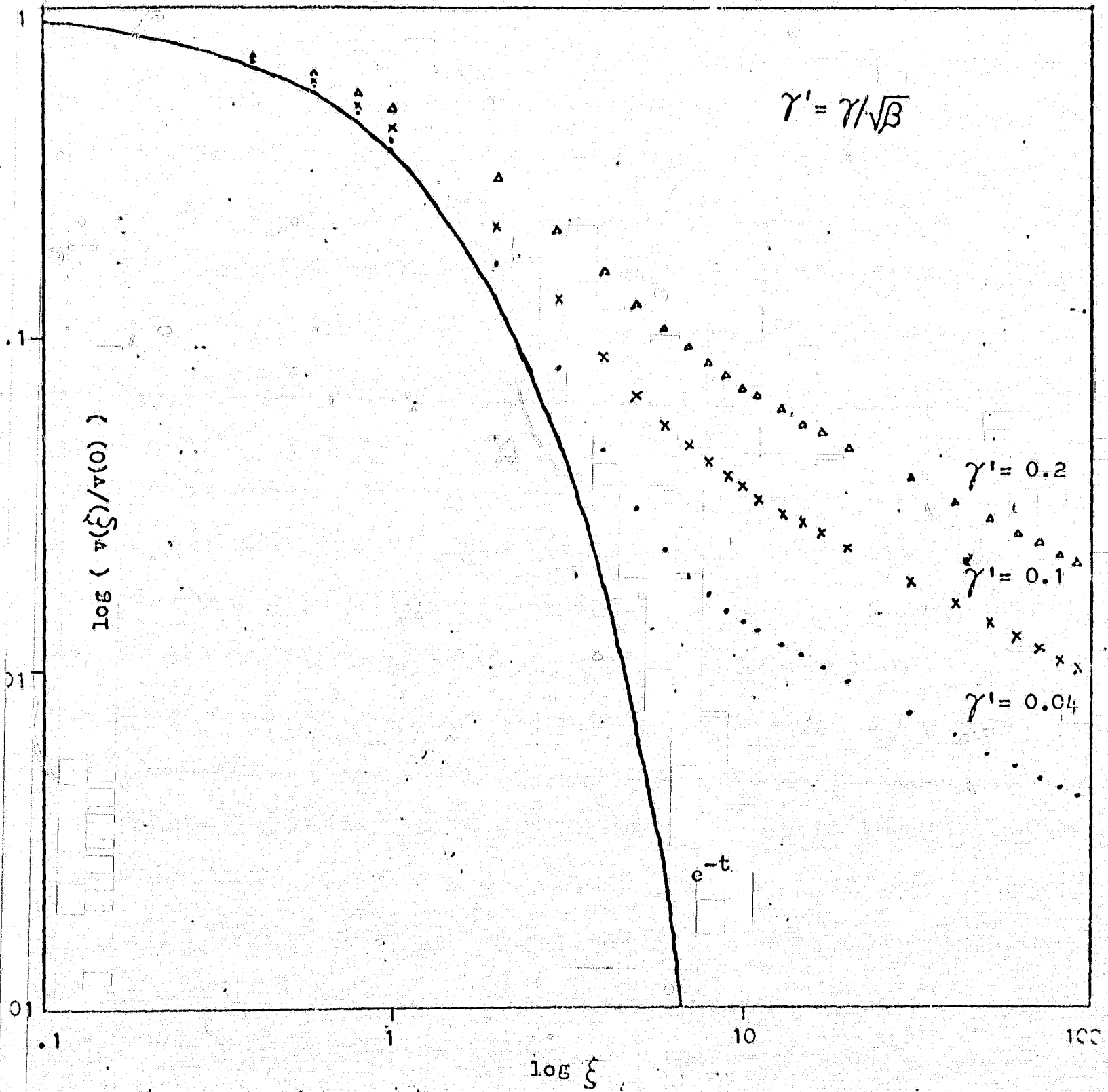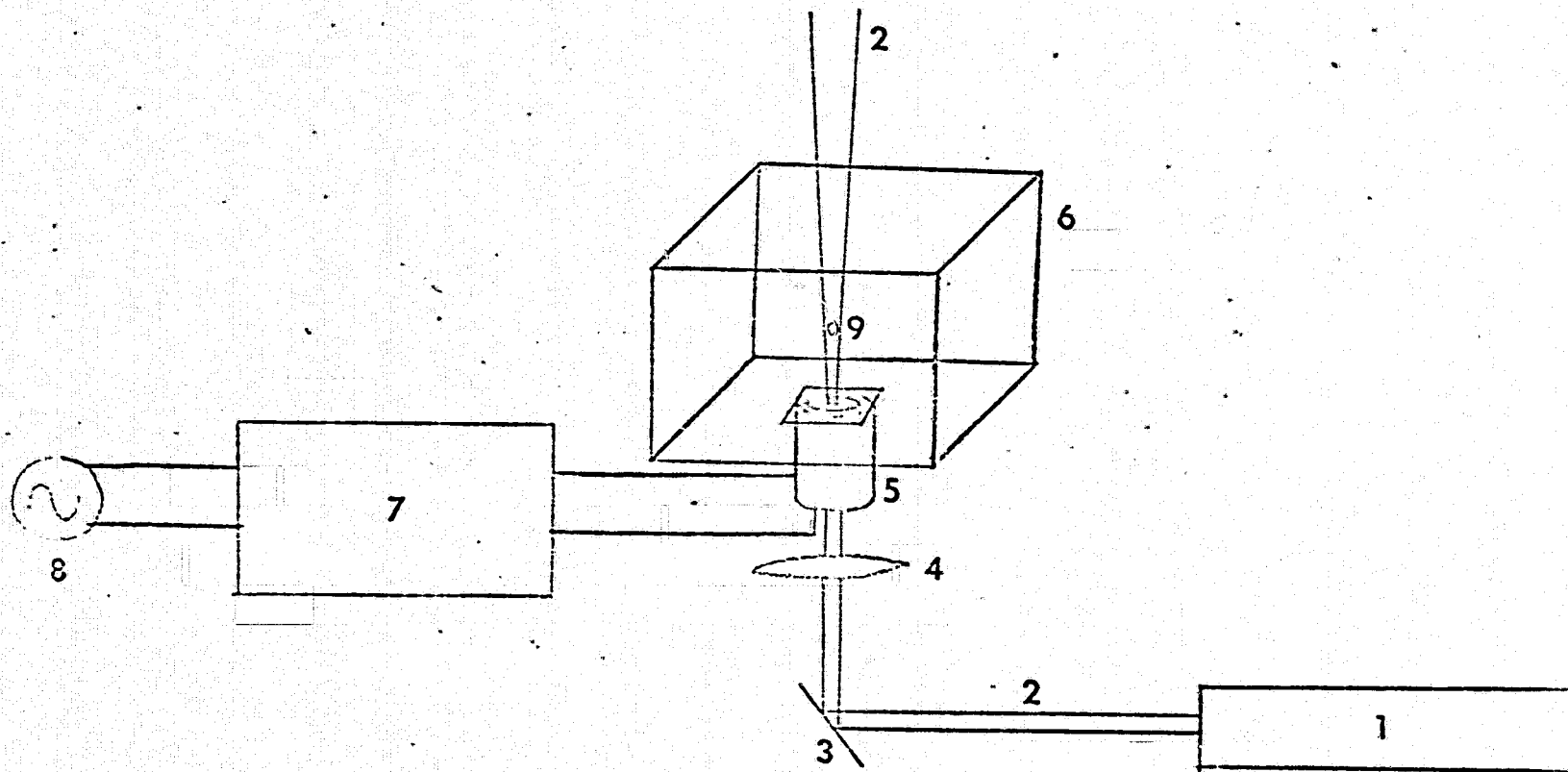
FIG. 1

FIGURE 2

1. Argon ion laser
2. Laser beam
3. Mirror
4. Focusing lens
5. Piezoelectric crystal with
   glass cover slip
6. Plexiglas shield
7. Wide band amplifier
8. Function generator
9. Suspended particle

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