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Long-Time Behavior of the Velocity Autocorrelation: A Measurement*

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The persistent velocity autocorrelation, first discovered by Alder and Wainwright through molecular dynamic computations, is confirmed experimentally in gaseous air and argon for the first time. Analysis is based on hydrodynamic considerations.

The long-time behavior of the velocity autocorrelation function has in the past few years been a center of considerable interest. A slowly decaying tail with a $t^{-3/2}$ dependence (where t denotes time) was initially discovered by Alder and Wainwright[1] in the course of molecular dynamic computations. Many investigators[2-9] have subsequently shown by a number of methods that such a behavior of the velocity autocorrelation for large times is consistent with both kinetic and hydrodynamic theory. The persistent autocorrelation is thought to be related ultimately to the collective effects of the surrounding fluid, specifically, in the form of vorticity.[10,11] Such a long-time behavior has been deduced for the Brownian as well as molecular motion in fluids. We wish to report in this Letter on the first laboratory confirmation of the Alder-Wainwright effect.

The design of the experiment is based on the following analysis: Along the line of reasoning first suggested by Alder and Wainwright,[10,11] Zwanzig and Bixon,[3] and Widom[7] (see also Ref. 8 for a general treatment), we have obtained the $t^{-3/2}$ dependence by making a provision for the time-dependent nature of the fluctuations. The motion of a small sphere of mass m and radius R moving with a small velocity in a fluid at rest in the absence of external force satisfies the modified Langevin equation[12,13]

$$m\,d\vec{V}/dt=-6\pi\eta R\vec{V}(t)-\tfrac{2}{3}\pi R^3\rho\,d\vec{V}/dt-6R^2(\pi\eta\rho)^{1/2}\int_{-\infty}^{t}(t-s)^{-1/2}[d\vec{V}(s)/ds]\,ds+\vec{f}(t), \tag{1}$$

where η and ρ are the shear viscosity and density of the fluid, respectively. The third term on the right-hand side of Eq. (1) is called the Basset term. The last is the fluctuating force in the fluid.

For a particle of 2.02 μm diam moving in gaseous air or argon with an initial velocity of the order of 10^3 cm/sec, it can be shown that $\vec{f}(t)$ is negligible. Along the direction of the particle motion, Eq. (1) can then be written as

$$dV/dt=-\alpha V(t)/\tau-\beta(\pi\tau)^{-1/2}\int_{-\infty}^{t}(t-s)^{-1/2}[dV(s)/ds]\,ds, \tag{2}$$

where $\alpha=(1+m_f/2m)^{-1}$, $\tau=m/6\pi\eta R$, $m_f=\frac{4}{3}\pi\rho R^3$, and $\beta=3[\alpha(1-\alpha)]^{1/2}$. It can also be shown that the velocity autocorrelation function satisfies a differential equation which is exactly identical in its physical origin and structure to Eq. (2). The only difference between the two is that the fluctuating force in Eq. (1) becomes exactly zero upon taking an ensemble average in constructing the velocity autocorrelation function, whereas in obtaining Eq. (2) $\vec{f}(t)$ has been neglected.

The initial condition for $V(t)$ is $V(t=0)=V_0$, but Eq. (2) must be supplemented by a boundary condition determining $V(t)$ for $t<0$. The two extreme cases are (i) $V(t)=0$ for $t<0$, (ii) $V(t)=V_0$ for $t<0$. Case (i) corresponds to the situation in which the sphere gains a finite velocity instantaneously at $t=0$ through a spontaneous fluctuation. The computation by Alder and Wainwright satisfies this boundary condition. The solution of Eq. (2) is given, when the boundary condition (i) holds, by

$$V(t)/V_0 = e^{at}[\cos bt + \beta(4\alpha-\beta^2)^{-1/2}\sin bt] + (\pi\tau)^{-1/2}\int_0^t (t-s)^{-1/2}e^{as}[-\beta\cos bs + (2\alpha-\beta^2)(4\alpha-\beta^2)^{-1/2}\sin bs]\,ds, \tag{3i}$$

where $a = -(2\alpha-\beta^2)/2\tau$ and $b = \beta(4\alpha-\beta^2)^{1/2}/2\tau$.

A comparison of the solution (3i) with the Alder-Wainwright data[11] has been made at the fluid density which is $\frac{1}{3}$ of the close-packing density. $V(t)/V_0$ is evaluated for various t/τ with $m_f/m = \pi/9\sqrt{2}$. The initial portion of the $V(t)/V_0$-versus-t/τ plot is then fitted with an exponentially decaying function to determine a new decay constant τ_p, which in general is different from τ because of the Basset term. $V(t)/V_0$ is replotted as a function of t/τ_p. Also, the Alder-Wainwright data are similarly displayed by finding their own τ_p in units of the Alder-Wainwright collision time. Agreement between the two is good for $t \geq 3.5\tau_p$. Such agreement is significant in view of the fact that in the asymptotic limit[14] $V(t)/V_0$ of Eq. (3i) becomes $\alpha_D t^{-3/2}$, with $\alpha_L = 2(4\pi\nu)^{-3/2}/3\alpha n$ as compared to $\alpha_D = (8\pi\nu)^{-3/2}/n$ of Kawasaki[2] and $2[4\pi(\nu+D)]^{-3/2}/3n$ of Ernst, Hauge, and Van Leeuwen[4] and Dorfman and Cohen.[6] ν and D are the kinematic viscosity and self-diffusion coefficient of the fluid, respectively.

Case (ii) accounts for the situation in which the particle maintains a steady motion with velocity V_0 for all times before $t=0$. The right-hand side of Eq. (2) now remains finite as $t\to 0$, and its solution exhibits an exponential decay for small times. When the boundary condition (ii) is applied, Eq. (2) is satisfied by

$$V(t)/V_0 = e^{at}[\cos bt - \beta(4\alpha-\beta^2)^{-1/2}\sin bt] + 2\alpha[(4\alpha-\beta^2)\pi\tau]^{-1/2}\int_0^t (t-s)^{-1/2}e^{as}\sin bs\,ds. \tag{3ii}$$

Equation (3i) shows the $t^{-3/2}$ dependence of the particle velocity for large times, but Eq. (3ii) has no such dependence and shows, in fact, a $t^{-1/2}$ dependence.

The preceding discussion demonstrates that the hydrodynamic description of the persistent velocity autocorrelation is reasonable and secondly that a laboratory test of the Alder-Wainwright effect can be made by determining the decay of particle velocity in a fluid at rest.

The experiment[15] is carried out in a shock tube because of the advantage that a uniform flow can be switched on with a rise time of the order of a collision time. An aerosol of latex spheres (1.060 ± 0.010 g/cm^3 in density) is formed in gaseous air or argon near the end of the 1.5-in.-diam shock tube. The particle diameter is varied from 2.02 to 3.21 μm. Typically, a shock of low shock Mach number ($1.05 < M_s \leq 1.12$) is generated initially, which triggers the particles into motion. By the time the reflected shock returns from the shock-tube end wall, the particle is observed to be maintaining a uniform motion with the flow behind the primary shock. In other words, the vorticity around the particle has died off completely when the particle plunges into the reflected shock flow which is at rest in the lab frame. The particle trajectory is recorded directly on film in a rotating drum camera with time resolution of 0.1 μsec., from which the particle velocity is obtained as a function of time. Experimental details will be reported elsewhere.

Figure (1) shows the results from three different runs, all pertaining to the decay of particle velocity starting from the instant of the reflected-shock arrival. One measurement was made in air and two in argon, each with different-size

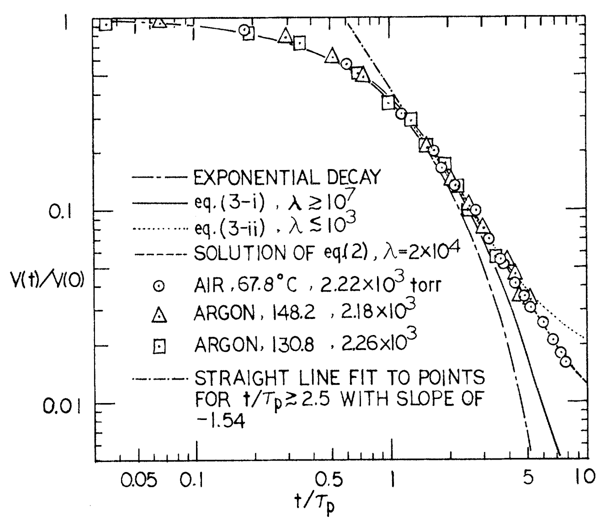

FIG. 1. Dimensionless velocity $V(t)/V_0$ (or velocity autocorrelation function) versus dimensionless time t/τ_p. Measured velocities of latex particles in air (circles, $\tau_p = 11.1 \pm 0.4$ μsec) and in argon (triangles, $\tau_p = 12.8 \pm 0.4$; squares, $\tau_p = 18.4 \pm 0.6$ μsec) are compared with the predictions of Eqs. (3i) and (3ii), which give the upper and lower bounds to the data. The measuring error in $V(t)/V_0$ is smaller than the size of the circles in the figure.

latex particles. All three runs have, however, been made in a narrow pressure range of $(2.18-2.26)\times10^3$ Torr. The particle relaxation time τ_p has been determined from the initial portion of the decay for each particle. For the run in air the measured τ_p, τ_p of Eq. (3i), and τ_p of Eq. (3ii) are 0.93τ, 0.92τ, and 1.11τ, respectively. As can be predicted from Eqs. (3i) and (3ii), the three measured curves fall right on top of each other when $V(t)/V_0$ is plotted as a function of dimensionless time, t/τ_p.[16] Equations (3i) and (3ii) are also shown, both evaluated for the average pressure of 2.20×10^3 Torr at 67.8°C.

Deviation from the exponential decay starts at about $1.5\tau_p$, and the $t^{-3/2}$ dependence is seen for times greater than about $2.5\tau_p$. This $t^{-3/2}$ dependence appears to be a coincidence because the measured points do not quite fall in the asymptotic region.[14] The experimental results differ considerably from the prediction of Eq. (3i), while a better agreement is indicated with that of Eq. (3ii). This observation can be understood in the following physical picture: In view of the fact that the reflected-shock thickness is of the order of $\frac{1}{10}$ of the particle diameter, it takes a small but finite length of time for the shock to run over the particle. During this period, the deceleration does not quite begin, and consequently some vorticity, similar to the case of steady motion, can be generated in the surrounding fluid. This, together with the vorticity associated with deceleration, preserves the memory of the initial velocity longer than Eq. (3i) predicts.

In any event Eqs. (3i) and (3ii) definitely give rise to the lower and upper bounds to the shock-tube data, as shown in Fig. 1, and this is borne out in a numerical simulation. If, for $t<0$, one chooses to express $V(t)$ as

$$V(t)=V_0e^{\lambda t},$$

the boundary conditions (i) and (ii) are satisfied in the limits $\lambda\to\infty$ and $\lambda\to0$, respectively. For the shock-tube experiment, λ should be of nonzero, finite value. At $\lambda=2\times10^4\ \mathrm{sec}^{-1}$ the solution of Eq. (2) agrees with the measurement, as also shown in Fig. 1.

This particular value of λ has, however, no connection to the shock structure and therefore should not be taken too seriously. In this case, it appears to be model dependent and stems from the particular choice of $V(t)=V_0e^{\lambda t}$ for $t<0$. dV/dt thus remains very large at $t=0$ for a wide range of λ, and, consequently, the term,

$$\beta(\pi\tau)^{-1/2}\int_{-\infty}^{0}(t-s)^{-1/2}[dV/ds]\,ds$$

in Eq. (2) deviates from $\beta V_0(\pi\tau t)^{-1/2}$ toward weaker time dependence only as a slow function of decreasing λ. This is physically unrealistic. If $V(t)$ for $t<0$ is, however, modeled after the density rise across the shock front, one might find the critical value of λ which is related to such parameters as the collision frequency and τ.

Aside from the slight ambiguity in the boundary condition, the experiment is consistent with the Alder-Wainwright effect. In the light of the property of Eqs. (3i) and (3ii), the Alder-Wainwright effect, on the other hand, may have to be viewed in a less stringent form than that of the $t^{-3/2}$ dependence. Namely, the long-time behavior of the velocity autocorrelation function depends strongly on the boundary condition, although in any case it persists longer than the exponential decay prescribes.

Remember that this conclusion is based only on the hydrodynamics. The kinetic calculation of Dorfman and Cohen[6] should provide a more general conclusion on this point, but it is not clear at this moment. It should also be noted that the hydrodynamic calculation based on the Navier-Stokes equation fails to describe at moderate densities the Alder-Wainwright data for short times. Furthermore, the hydrodynamic calculation of Eq. (2) cannot produce the negative autocorrelation at very high densities. These add to the shortcomings of the hydrodynamic formulation.

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Parametrically Driven Ion Cyclotron Waves and Intense Ion Heating*

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Finite-amplitude plasma waves excited by plates outside a plasma column are shown to decay into other plasma waves and electrostatic ion cyclotron waves, accompanied by ion heating.

A number of experiments[1] on parametric instabilities have been reported recently, including measurements of energy deposition into the tails of the particle velocity distributions[2,3] and electron heating.[4] In this Letter we report measurements of strong heating of the bulk of the ions. The increase in ion temperature coincides with the onset of ion cyclotron waves driven parametrically unstable by nearly perpendicularly propagating electron plasma waves resulting from rf electric fields applied to a plasma column. The experimental results include an identification of the pump wave as a Trivelpiece-Gould mode, and measurements of the dispersion relation of the parametric ion cyclotron wave, the threshold conditions, pump-field depletion accompanying instability onset, and ion temperatures up to a factor of 100 higher than the initial temperature.

The importance of this experiment rests on the following points. First, the pump-wave frequency ω_0 is close to the lower-hybrid frequency ω_{LH} which in fusion-reactor plasmas ($\omega_{LH}/2\pi \sim 2$ GHz $\sim \omega_{pi}/2\pi$, where ω_{pi} is the ion plasma frequency) represents the practical upper limit of available high-power sources.[5] Second, in accordance with the Manley-Rowe relation, the present parametric process, when compared with others has a high frequency ratio ($\sim \Omega_i/\omega_{pi}$) of the low frequency component of the decay wave to the pump, allowing a relatively high level of power to be stored in the ion wave. Third, the frequency of the ion cyclotron wave can be very close to the ion cyclotron frequency Ω_i; thus heating of the bulk of the ions should be expected.

Linear calculations of the relevant parametric processes,[6,7] using the dipole approximation, show that two branches ($k_\parallel \ll k_\perp$ and $k_\parallel \sim k_\perp$) of ion cyclotron and ion sound waves may be parametrically destabilized. Their frequencies for the lowest thresholds (in the usual notation) are, for $k^2 c_s^2/\Omega_i^2 \ll 1$,

$$\omega^2/\Omega_i^2 = 1 + k^2 c_s^2/\Omega_i^2, \quad k_\parallel \ll k_\perp, \tag{1a}$$

$$\omega^2 = k_\parallel^2 c_s^2, \quad k_\parallel \sim k_\perp; \tag{1b}$$

and for $k^2 c_s^2/\Omega_i^2 \gg 1$,

$$\omega^2 = k^2 c_s^2, \quad k_\parallel \ll k_\perp, \tag{2a}$$

$$\omega^2 = \Omega_i^2 (k_\parallel^2/k_\perp^2), \quad k_\parallel \sim k_\perp. \tag{2b}$$

The parametric process described by Eq. (2a) has been previously reported.[8] The corresponding threshold condition[7] for these instabilities is

$$(\vec{k}\cdot\vec{\epsilon})^2 = 2\left(\frac{kv_e}{\omega_{pe}}\right)^2 \frac{\nu_e}{\omega_0}\frac{\nu_i}{\Omega_i}\frac{T_e}{T_i},$$

where $|\vec{k}|$ is the instability wave number, $\vec{\epsilon}$ is the particle displacement due to the rf field $\vec{E}$ at frequency ω_0, v_e is the electron thermal velocity, ν_e is the collisional damping of the high-frequency component of the parametric instability, and

$$\frac{\nu_i}{\Omega_i} = 2\sqrt{\pi}\,\frac{k^2 v_i}{\Omega_i^2}\frac{\omega}{k_\parallel v_i}\exp\left[-\left(\frac{\omega-\Omega_i}{k_\parallel v_i}\right)^2\right]$$

denotes ion cyclotron damping. The dominant