Brownian dynamics in a thin sheet with momentum decay

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The velocity autocorrelation function (VACF) of a disk moving in a two-dimensional viscous fluid is calculated under the condition that the momentum leaks out of the fluid with a relaxation time τ. In the absence of any memory effects, VACF decays exponentially. Even in the presence of memory effects, VACF essentially decays exponentially being accompanied by the correction of algebraic decay. This correction depends on the coupling strength between the sheet and the outer fluid, i.e., $\sim e^{-t/\tau}t^{-3/2}$ and $\sim e^{-t/\tau}t^{-1}$ for strong- and weak-coupling limits, respectively. The correction of t^{-1} reflects the two-dimensional character of the fluid sheet.

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I. INTRODUCTION

The hydrodynamical problem of a diffusing particle in a thin plane sheet of viscous fluid embedded in another viscous fluid has received great attention in connection with biophysics, for example, diffusion properties of proteins or lipid molecules within biological membranes. From the theoretical point of view [1–7], this problem has been treated under the assumption that the velocity field is constant over any cross section of the fluid sheet. It then turns out that the anisotropic nature (or two dimensionality) of the fluid sheet plays an essential role in such a system. The diffusing particle lying in the sheet has been represented by a cylindrical disk whose axis is perpendicular to the plane of the sheet.

In a two-dimensional hydrodynamic model, however, the Stokes approximation (inertialess limit) cannot provide any solution of the velocity field such that the boundary conditions are satisfied at the surface of the cylinder and simultaneously vanish at infinity. In other words, a steady force per unit length applied to the cylinder induces an infinite velocity causing infinite mobility [8]. This contradiction is called the "Stokes paradox." One way to recover a finite mobility is to partially take into account the inertial term, for instance, by the Oseen approximation. Unfortunately, even this approximation is not sufficient to provide the diffusion coefficient in the sense that the mobility does not take a constant value. In fact, it depends on the velocity of the disk since there is no linear relation between the velocity and the drag it experiences. Hence the argument used in deriving the Einstein relation fails.

These difficulties are intrinsic to a two-dimensional hydrodynamical model with momentum conservation. In the biomembrane problem, however, the fluid membrane is not an isolated system. Typical biomembranes which consist of freely moving lipid molecules exhibit certain fluidity and are sandwiched by the surrounding water. The momentum contained in the fluid sheet can be transferred to the adjacent bulk (three-dimensional) fluid due to the coupling between the water molecules and the polar heads of the lipid molecules. When the momentum leaks into the surrounding outer fluid, the Stokes paradox can be eliminated, and one can obtain a finite, constant mobility of the disk.

Saffman and Delbrück were the first to come to this point [1,2]. In their theory, Brownian motion of a cylinder was investigated by a purely hydrodynamical model in which both the thickness of the fluid sheet and the viscosity of the outer fluid were taken into account. The transfer of momentum to the adjacent fluid was incorporated through boundary conditions at the membrane surfaces. The resulting translational diffusion coefficient exhibits only a weak (logarithmic) dependence on the particle size. Despite this success, it was still not easy to obtain the general solution which satisfies the dual boundary conditions. Their calculation was limited to the case where the viscosity of the membrane is sufficiently large compared to the viscosity of the adjacent fluid.

In order to grasp a more intuitive understanding of the problem, several authors have proposed two-dimensional hydrodynamic equations in which the total momentum is nonconserved [4–7]. Particularly, Izuyama introduced the idea of a phenomenological decay time characterizing the leak process of the total momentum in the fluid sheet [5]. This decay time is inversely proportional to the coupling strength between the fluid sheet and the outer fluid. Corresponding to the result by Saffman and Delbrück, the diffusion coefficient shows logarithmic size dependence in the weak-coupling limit (slow momentum-relaxation limit) [6,7] (see also Sec. VI). The same hydrodynamical model as that appearing in Ref. [7] was independently proposed by Evans and Sackmann in a somewhat different context where they considered a fluid membrane associated with a rigid substrate such as the Langmuir-Blodgett film [4]. The consequences of this model are briefly summarized in Sec. III.

The main purpose of this paper is to investigate the velocity autocorrelation function (VACF) of the Brownian disk moving in a two-dimensional fluid with momentum decay. In order to describe the motion of the diffusing particle, we used the generalized Langevin equation which takes memory effects into account. From the point of view of statistical mechanics, one of the advantageous outcomes of the present hydrodynamical model with momentum decay is that one can rely on the fluctuation-dissipation theorem (FDT) as our basis. As described above, the mobility becomes infinite when the momentum should be conserved, which in turn causes the breakdown of FDT in the long-time limit. In fact, Saffman was obliged to rely on more a obscure argument when he calculated the mean-square displacement of the disk diffusing in the isolated two-dimensional fluid [2].

Meanwhile our interests in the dynamical properties of a Brownian particle naturally originate from the predicted hydrodynamic long-time tails over twenty years. The pioneering molecular-dynamics simulation by Alder and Wainwright suggested that the VACF of a tagged particle decays algebraically rather than exponentially at long times [9,10]. In a d-dimensional system, the simulation data were consistent with an asymptotic decay as $\sim t^{-d/2}$. The appearance of the long-time tails was attributed to the coupling between particle diffusion and shear modes in the fluid. Several authors have theoretically rederived this behavior by using, for instance, the lowest-order mode-coupling theory [11] or kinetic theory [12].

Nevertheless, the above explanations for long-time tails give rise to a serious breakdown of two-dimensional hydrodynamics. According to the Green-Kubo relation [see (4.15) later], the diffusion coefficient diverges as $\sim \ln t$ when the VACF simply decays as $\sim t^{-1}$. In order to overcome this difficulty, several theoretical arguments have been proposed insisting on a faster-than-t^{-1} decay, such as $(t\sqrt{\ln t})^{-1}$ [13,14]. Since this time, many attempts have been made to compare these predictions directly with computer simulations. However, the expected correction to the t^{-1} decay was too small to be observed, and most of the simulations could confirm only the existence of the t^{-1} tails, contrary to their aims [13,15,16]. Quite recently, possible evidence of faster-than-t^{-1} decay has been reported [17,18] by using a lattice-gas cellular automaton [19]. What has been observed in these studies is, however, only the onset of the crossover to the $(t\sqrt{\ln t})^{-1}$ behavior.

We will show that within the present two-dimensional fluid model with momentum decay, the VACF essentially decays exponentially even in the presence of memory effects. Moreover, we find algebraically decaying corrections to the exponential decay depending on the coupling strength between the adjacent fluid.

The outline of this article is as follows. In the next section, the hydrodynamical model with momentum decay is explained in detail. Following the calculation by Evans and Sackmann [4], the drag on the disk is obtained both for stationary and time-dependent cases in Sec. III. In Sec. IV, a generalized Langevin equation is introduced to describe the particle motion on the basis of FDT. Our main results concerning the VACF are given in Sec. V for both the cases without and with memory effects.

II. HYDRODYNAMICAL MODEL WITH MOMENTUM DECAY

In order to provide a two-dimensional Newtonian fluid theory appropriate for the diffusion process of a Brownian particle within the fluid sheet, a simple phenomenological hydrodynamical model was independently proposed by two different groups though in the fairly different contexts [4–7]. In this section, we explain the basic idea of this model in detail following Ref. [6], but with some new aspects added. We consider an infinite plane sheet of viscous fluid with a dynamic viscosity η. This fluid sheet is assumed to be incompressible, being characterized by a constant density ρ.

Let us consider a disk with radius a representing a diffusing particle. When the disk moves with velocity $-\mathbf{U}(t)$, a velocity field $\mathbf{u}$ is induced around the disk. This velocity field should vanish at infinity. In ordinary hydrodynamics, the equation of motion of the fluid is written using a tensor notation as [20]

$$\frac{\partial}{\partial t}(\rho u_i) = -\frac{\partial \pi_{ik}}{\partial x_k} , \tag{2.1}$$

where π_{ik} is the momentum-flux-density tensor

$$\pi_{ik} = -\sigma_{ik} + \rho u_i u_k , \tag{2.2}$$

and σ_{ik} is the stress tensor

$$\sigma_{ik} = -p\delta_{ik} + \eta \left[\frac{\partial u_i}{\partial x_k} + \frac{\partial u_k}{\partial x_i} \right] . \tag{2.3}$$

In (2.3), p is the pressure, and the incompressible condition has been imposed. Since the velocity $\mathbf{u}$ vanishes at infinity, (2.2) is consistent with the conservation rule of the total momentum of the fluid $\mathbf{\Pi}$, i.e.,

$$\frac{\partial \Pi_i}{\partial t} = \frac{\partial}{\partial t} \int d\mathbf{r}(\rho u_i) = 0 , \tag{2.4}$$

where the integration is extended to the whole area of the fluid.

In the present problem, the fluid sheet is not an isolated system but coupled to the adjacent fluid. The momentum within the fluid sheet thereby may leak to the outer fluid. Consistent with such considerations, Suzuki and Izuyama have proposed a hydrodynamic equation that does not conserve the total momentum [5–7]. They introduced a phenomenological momentum relaxation time τ which should be inversely proportional to the coupling strength between the sheet and the outer fluid. The dissipation of the momentum should be given by

$$\frac{\partial \Pi_i}{\partial t} = -\frac{1}{\tau}\Pi_i . \tag{2.5}$$

A hydrodynamic equation which is consistent with this total-momentum decay is expressed as

$$\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u}\cdot\mathrm{grad})\mathbf{u} = -\frac{1}{\rho}\mathrm{grad}\, p + \nu\nabla^2\mathbf{u} - \frac{1}{\tau}\mathbf{u} , \tag{2.6}$$

where $\nu=\eta/\rho$ is a kinematic viscosity. Here both grad and ∇^2 represent two-dimensional operators. The last term on the right-hand side indicates the transfer of momentum due to the interaction at the interface.

It is worthwhile to point out that the momentum-decay mechanism should, in principle, operate on the difference between the velocity at a given point and that at infinity. So far we have used a coordinate system such that the two-dimensional fluid is at rest at infinity. Hence the momentum-relaxation effect is incorporated through the expression $-\mathbf{u}/\tau$ in (2.6).

As a linearization of (2.6), we adopt the Stokes approximation, neglecting the convective acceleration term $(\mathbf{u}\cdot\text{grad})\mathbf{u}$. Consequently, our model is reduced to

$$\frac{\partial \mathbf{u}}{\partial t}=-\frac{1}{\rho}\text{grad}\,p+\nu\nabla^2\mathbf{u}-\frac{1}{\tau}\mathbf{u}\ , \tag{2.7}$$

together with the incompressible condition

$$\text{div}\,\mathbf{u}=0\ . \tag{2.8}$$

Notice that (2.7) and (2.8) are equivalent to equations proposed by Evans and Sackmann who considered a fluid membrane associated with a rigid substrate such as the Langmuir-Blodgett film [4].

It is important to notice that the present two-dimensional fluid is only a "fluid" on time scales short compared to the momentum decay time τ. Therefore the analysis given in this paper concerns the decay in two-dimensional hydrodynamics on some intermediate-time scale, but does not capture true asymptotics. The expected behavior of the two-dimensional fluid in the superlong-time regime $(t\gg\tau)$ will be separately discussed in Sec. VI.

III. DRAG ON THE DISK MOVING IN THE FLUID SHEET

Several people have recently calculated the translational drag which is felt by the disk moving with a time-independent constant velocity $-U$ along the x axis [4–7]. For the purpose of extending the model to the nonstationary case, the outline of previous arguments is briefly summarized here.

Starting from the linearized steady-flow equation

$$-\frac{1}{\rho}\text{grad}\,p+\nu\nabla^2\mathbf{u}-\frac{1}{\tau}\mathbf{u}=\mathbf{0},\quad \text{div}\,\mathbf{u}=0 \tag{3.1}$$

and the nonslip boundary condition at the surface of the disk

$$(u_x,u_y)=(-U,0)\quad \text{at } r=a\ , \tag{3.2}$$

we obtain the following drag force exerted by the surrounding fluid along the x axis [4]:

$$F=\left[\pi\eta(\kappa a)^2+4\pi\eta\frac{(\kappa a)K_1(\kappa a)}{K_0(\kappa a)}\right]U\ . \tag{3.3}$$

In the above, κ is the important parameter defined by [5–7]

$$\kappa^2=\frac{1}{\nu\tau}\ , \tag{3.4}$$

and K_0 and K_1 are modified Bessel functions of the second kind, order zero and one, respectively. The linear dependence of F on U provides the well-defined drag coefficient Λ such that

$$\Lambda=\pi\eta(\kappa a)^2+4\pi\eta\frac{(\kappa a)K_1(\kappa a)}{K_0(\kappa a)}\ . \tag{3.5}$$

The diffusion coefficient is simply related to the mobility b (defined as the inverse of the drag coefficient) by the Einstein relation

$$D=k_BTb=\frac{k_BT}{\Lambda}\ . \tag{3.6}$$

The time-dependent drag coefficient $\Lambda(t)$ exerted by the surrounding fluid on the disk, moving now with the time-dependent velocity $-U(t)$ along the x axis, can be easily obtained by extending the previous results. Let us introduce the Fourier-Laplace (or one-sided Fourier) transform defined for a function $f(t)$ by

$$f[\omega]=\int_0^\infty dt\, e^{-i\omega t}f(t)\ . \tag{3.7}$$

By taking the Fourier-Laplace transform of (3.1), we obtain

$$-\frac{1}{\rho}\text{grad}\,p[\omega]+\nu\nabla^2\mathbf{u}[\omega]-\frac{1}{\tau'}\mathbf{u}[\omega]=0\ , \tag{3.8}$$

where

$$\frac{1}{\tau'}=\frac{1}{\tau}+i\omega\ . \tag{3.9}$$

In (3.8), we have used the fact that the surrounding fluid is at rest at $t=0$. We require that the nonslip boundary condition (3.2) holds at any time:

$$(u_x(t),u_y(t))=(-U(t),0)\quad \text{at } r=a\ , \tag{3.10}$$

or, equivalently, in ω space

$$(u_x[\omega],u_y[\omega])=(-U[\omega],0)\quad \text{at } r=a\ . \tag{3.11}$$

In view of (3.8) and (3.11), a frequency-dependent drag coefficient can be simply deduced by replacing the constant decay time τ in the stationary case with the frequency-dependent decay time τ' defined as in (3.9). By using the new abbreviation

$$\kappa'^2=\frac{1}{\nu\tau'}\ , \tag{3.12}$$

the drag coefficient is then given by

$$\begin{aligned}\Lambda[\omega]&=\pi\eta(\kappa' a)^2+4\pi\eta\frac{(\kappa' a)K_1(\kappa' a)}{K_0(\kappa' a)}\\ &=\pi\rho a^2 i\omega+\pi\eta(\kappa a)^2+4\pi\eta\frac{(\kappa' a)K_1(\kappa' a)}{K_0(\kappa' a)}\ .\end{aligned} \tag{3.13}$$

Note that in the $\omega\to 0$ limit, this expression reduces to (3.5). In the next section, we use (3.13) in order to construct the generalized Langevin equation that describes the Brownian motion of the disk.

IV. GENERALIZED LANGEVIN EQUATION

For the description of the Brownian motion of the particle, we employ the Langevin equation of the disk moving with the time-dependent velocity $U(t)$ (it is sufficient to study only one-dimensional motion, say, along the x axis). We also consider the case where the drag force can be retarded. Then the Langevin equation should be generalized to

$$m\frac{d}{dt}U(t)=-\int_{-\infty}^{t}dt'\Lambda(t-t')U(t')+R(t)\ . \quad (4.1)$$

In the present problem, m is the mass of the disk, $\Lambda(t)$ is the inverse Fourier-Laplace transform of $\Lambda[\omega]$ in (3.13), and $R(t)$ is a random force. Since the first term on the right-hand side of (3.13) is proportional to $i\omega$, one can consider a renormalized mass m^* which takes into account the additional inertia due to the dragging motion of the fluid, i.e.,

$$m^*=m+\pi\rho a^2\ . \quad (4.2)$$

Consequently, we are led to an effective generalized Langevin equation instead of (4.1) given by

$$m^*\frac{d}{dt}U(t)=-\int_{-\infty}^{t}dt'\lambda(t-t')U(t')+R(t)\ , \quad (4.3)$$

with

$$\lambda[\omega]=\pi\eta(\kappa a)^2+4\pi\eta\frac{(\kappa' a)K_1(\kappa' a)}{K_0(\kappa' a)}\ . \quad (4.4)$$

The random force $R(t)$ in (4.3) is assumed to vanish when averaged over the ensemble of molecular motions, i.e.,

$$\langle R(t)\rangle=0\ . \quad (4.5)$$

In addition, the random force should have properties consistent with the equipartition of energy. This condition is given through the correlation function of $R(t)$, such as by [21]

$$\langle R(t_0)R(t_0+t)\rangle=k_BT\lambda(t)\ . \quad (4.6)$$

As will be described below, (4.6) is equivalent to the knowledge of the power spectrum of the random force. The power spectrum of a real random variable $f(t)$ is generally defined as

$$I_f=\lim_{\Delta t\to\infty}\langle|f(\omega)|^2\rangle\frac{\Delta t}{2\pi}\ , \quad (4.7)$$

where Δt is the time interval of the observation, and $f(\omega)$ is the Fourier transform of $f(t)$

$$f(\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty}dt\, e^{-i\omega t}f(t)\ . \quad (4.8)$$

[Here we have distinguished between the Fourier components and the previous Fourier-Laplace components according to the shape of the bracketing; see (3.7).] By using these notations, (4.6) can be rewritten as

$$I_R(\omega)=\frac{m^*k_BT}{\pi}\mathrm{Re}\{\tilde{\lambda}[\omega]\}\ , \quad (4.9)$$

with

$$\tilde{\lambda}[\omega]=\frac{\lambda[\omega]}{m^*}\ . \quad (4.10)$$

This is what is called the fluctuation-dissipation theorem.

After taking the Fourier transform of (4.3), one finds that the power spectrum of $U(t)$ and $R(t)$ are combined by

$$I_U(\omega)=\frac{1}{m^{*2}}\frac{I_R(\omega)}{|i\omega+\tilde{\lambda}[\omega]|^2}\ . \quad (4.11)$$

Since $I_R(\omega)$ is given by (4.9), (4.11) yields $I_U(\omega)$, which in turn gives the velocity autocorrelation function $\langle U(t_0)U(t_0+t)\rangle$ according to the Wiener-Khintchine theorem;

$$\begin{aligned}\phi(t)&\equiv\langle U(t_0)U(t_0+t)\rangle\\&=\int_{-\infty}^{\infty}d\omega\, e^{i\omega t}I_U(\omega)\\&=\frac{k_BT}{m^*}\frac{1}{2\pi}\int_C d\omega\frac{e^{i\omega t}}{i\omega+\tilde{\lambda}[\omega]}\ . \end{aligned}\quad (4.12)$$

In deriving (4.12), we have used the fact that $\tilde{\lambda}[\omega]$ is analytic in the lower half-region of the complex ω plane and the integration path C is that depicted in Fig. 1. In the limit of $t\to 0+$, the sum of the residues of $(i\omega+\tilde{\lambda}[\omega])^{-1}$ is equal to the residue around the infinity, i.e., $\omega=\infty$. If $\tilde{\lambda}[\omega]$ remains finite in this limit, the residue is simply equal to unity. Therefore (4.9) [or (4.6)] ensures the equipartition law

$$\lim_{t\to 0+}\phi(t)\equiv\langle U^2\rangle=\frac{k_BT}{m^*}\ . \quad (4.13)$$

The mean-square average displacement of the disk within the time interval $[0,t]$ is given by

$$\begin{aligned}\langle x^2(t)\rangle&=\int_0^t dt'\int_0^t dt''\langle U(t')U(t'')\rangle\\&=2\int_0^t dt'(t-t')\phi(t')\ . \end{aligned}\quad (4.14)$$

Equation (4.14) can be transformed into an expression for the diffusion coefficient called the Green-Kubo relation

$$D=\lim_{t\to\infty}\frac{\langle x^2(t)\rangle}{2t}=\int_0^{\infty}dt'\phi(t')\ . \quad (4.15)$$

When the last integral converges, we obtain a finite diffusion coefficient.

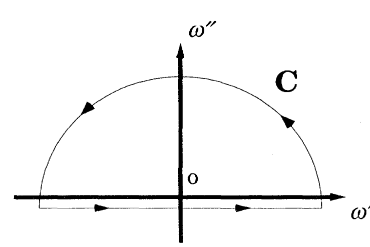

FIG. 1. Integration path C in the complex ω plane. ω' and ω'' represent the real and imaginary parts of ω, respectively.

V. VELOCITY AUTOCORRELATION FUNCTION

In this section, we calculate the velocity autocorrelation function of the disk for the cases without and with memory effects in accordance with the Wierner-Khintchine theorem [see (4.12)].

As discussed in Sec. III, when we consider the long-time limit, i.e., $\omega \to 0$, the drag coefficient (4.4) reduces to a finite constant value which no longer depends on the frequency,

$$\tilde{\lambda} = \frac{1}{m^*}\left[\pi\eta(\kappa a)^2 + 4\pi\eta\frac{(\kappa a)K_1(\kappa a)}{K_0(\kappa a)}\right] . \tag{5.1}$$

In such a case, Brownian motion simply turns out to exhibit the Markovian nature in which the retardation effects disappear. Substituting (5.1) into (4.12) and performing the contour integral along the path C, we obtain the expected exponentially decaying VACF as

$$\phi(t) = \frac{k_B T}{m^*}\exp\left[-\frac{t}{m^*}\left[\pi\eta(\kappa a)^2 + 4\pi\eta\frac{(\kappa a)K_1(\kappa a)}{K_0(\kappa a)}\right]\right] . \tag{5.2}$$

We find that this fast decay of $\phi(t)$ is consistent with the finite diffusion coefficient even, in the two-dimensional fluid.

In the problem of a hard sphere moving in a three-dimensional fluid, Landau and Lifshitz calculated the frequency-dependent correction to the Stokes formula $F = 6\pi\eta a U$, where a is now the radius of the sphere [20]. The physics behind the correction term is the coupling to the shear modes governed by the diffusion equation. Zwanzig and Bixon calculated the frictional force on a moving sphere for arbitrary frequency, compressibility, and viscoelasticity, with arbitrary slip of the fluid on the surface of the sphere [22]. By using the frequency-dependent drag force, we can take the memory effects into account. The observed hydrodynamical long-time tail $\phi(t) \sim t^{-3/2}$ in a three-dimensional fluid can be essentially explained by this correction according to the hydrodynamical description [22]. Here we follow the same arguments as that used in the case of the two-dimensional fluid model with momentum decay. Since it is impossible to handle (4.4) in general, we consider here two limiting cases, namely, the strong- and weak-coupling limits. The relevant asymptotic forms of the modified Bessel functions are provided in the Appendix.

In the strong-coupling limit ($\kappa' a \gg 1$), we use (A1) to obtain the asymptotic drag coefficient as

$$\begin{aligned}\tilde{\lambda}[\omega] &\approx \frac{1}{m^*}[\pi\eta(\kappa a)^2 + 4\pi\eta\kappa' a] \\ &= \frac{1}{m^*}[\pi\eta(\kappa a)^2 + 4\pi\eta\kappa a\sqrt{1+i\omega\tau}] ,\end{aligned} \tag{5.3}$$

which is then substituted into (4.12). Changing the variable to $s = i\omega + (1/\tau)$, we can express the VACF as

$$\phi(t) = \frac{k_B T}{m^*}\frac{1}{2\pi i}e^{-t/\tau}\int_{C'} ds \frac{e^{st}}{s + \alpha\sqrt{s} + \beta} , \tag{5.4}$$

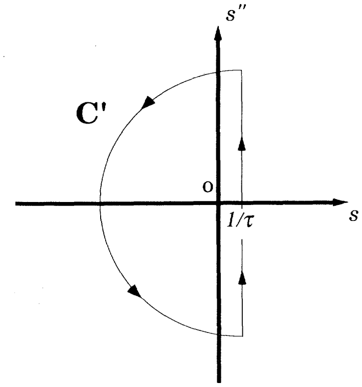

FIG. 2. Integration path C' in the complex s plane. s' and s'' represent the real and imaginary parts of s, respectively.

where

$$\alpha = \frac{1}{m^*}4\pi\eta\kappa a\sqrt{\tau} , \tag{5.5}$$

$$\beta = \frac{1}{m^*}\pi\eta(\kappa a)^2 - \frac{1}{\tau} = -\frac{m}{\tau m^*} . \tag{5.6}$$

The integration path C' is a straight line parallel to the imaginary axis of the complex s plane (see Fig. 2). Since singular points lie only on the negative real axis, the path C' can be deformed to the real negative axis, as shown by path Γ in Fig. 3 [21]. Hence the VACF can be further calculated as

$$\begin{aligned}\phi(t) &= \frac{k_B T}{m^*}\frac{1}{2\pi i}e^{-t/\tau}\left[\int_{-\infty}^{0} ds \frac{e^{st}}{s+\beta-\alpha\sqrt{|s|}i} + \int_{0}^{-\infty} ds \frac{e^{st}}{s+\beta+\alpha\sqrt{|s|}i}\right] \\ &= \frac{k_B T}{m^*}\frac{\alpha}{\pi}e^{-t/\tau}\int_0^\infty ds \frac{\sqrt{s}e^{-st}}{(s-\beta)^2+\alpha^2 s} \\ &= \frac{k_B T}{m^*}\frac{\alpha}{\pi}e^{-t/\tau}t^{-3/2}\int_0^\infty d\zeta \frac{\sqrt{\zeta}e^{-\zeta}}{(\beta-\zeta/t)^2+\alpha^2\zeta/t} .\end{aligned} \tag{5.7}$$

For large t, we have

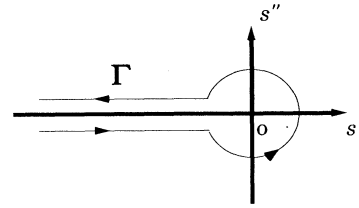

FIG. 3. Integration path Γ in the complex s plane. s' and s'' represent the real and imaginary parts of s, respectively.

$$\phi(t) \approx \frac{k_B T}{m^*} \frac{\alpha}{\pi} e^{-t/\tau} t^{-3/2} \int_0^\infty d\zeta \frac{\sqrt{\zeta} e^{-\zeta}}{\beta^2}$$
$$= \frac{k_B T}{2m^*} \frac{\alpha}{\sqrt{\pi}\beta^2} e^{-t/\tau} t^{-3/2} . \quad (5.8)$$

Since $1/\tau$ is large in this limit, the VACF essentially decays exponentially. It is interesting to note that the exponential decay is corrected by the algebraically decaying term $t^{-3/2}$.

Finally, we consider the weak-coupling limit ($\kappa' a \ll 1$). Upon using (A2) and (A3), the drag coefficient takes the following asymptotic form:

$$\tilde{\lambda}[\omega] \approx \frac{4\pi\eta}{m^*[\ln(2/\kappa' a) - \gamma]}$$
$$= \frac{4\pi\eta}{m^*}[-\ln(\tfrac{1}{2}\kappa a\sqrt{1+i\omega\tau}) - \gamma]^{-1} . \quad (5.9)$$

We substitute (5.9) into (4.12) and change the variable to $s = i\omega + (1/\tau)$, as above. It then follows that

$$\phi(t) = \frac{k_B T}{m^*} \frac{1}{2\pi i} e^{-t/\tau} \int_{C'} ds \frac{e^{st}}{s + (4\pi\eta/m^*)[-\ln\sqrt{a^2 s/4\nu} - \gamma]^{-1} - 1/\tau}$$
$$= \frac{k_B T}{m^*} e^{-t/\tau} \frac{1}{2\pi i} \int_{C'} ds \frac{[-\ln\sqrt{a^2 s/4\nu} - \gamma] e^{st}}{(4\pi\eta/m^*) + [s - (1/\tau)][-\ln\sqrt{a^2 s/4\nu} - \gamma]} . \quad (5.10)$$

Since our concern is the large t behavior of $\phi(t)$, we take the limit of $\omega \to 0$, i.e., $s - (1/\tau) \to 0$. By performing the inverse Laplace transform of the remaining function, the asymptotic form of VACF turns out to be

$$\phi(t) \approx \frac{k_B T}{m^*} e^{-t/\tau} \frac{1}{2\pi i} \int_{C'} ds \frac{m^*}{4\pi\eta} [-\ln\sqrt{a^2 s/4\nu} - \gamma] e^{st}$$
$$= \frac{k_B T}{8\pi\eta} e^{-t/\tau} t^{-1} . \quad (5.11)$$

The algebraic correction to the exponential decay is now t^{-1}, reflecting the two-dimensional character of the fluid sheet.

When $1/\tau = 0$, our model reduces to the usual two-dimensional fluid in which the total momentum is conserved. In this case, one can easily find from (5.11) that the VACF decays only algebraically,

$$\phi(t) \approx \frac{k_B T}{8\pi\eta} t^{-1} , \quad (5.12)$$

corresponding to the observed long-time tails in two-dimensional fluids. [Notice that we cannot put $1/\tau = 0$ in (5.8) for the strong-coupling limit.] The retardation effect shows up as a very slow decay of the VACF. According to the Green-Kubo relation, (4.15), the diffusion coefficient diverges as $\sim \ln t$, exhibiting the annoying Stokes paradox. Equation (5.11) implies that the presence of a nonzero $1/\tau$ ensures the exponential decay of the VACF, which then eliminates the Stokes paradox. This is the essential reason why we could obtain the finite diffusion coefficient as in (3.6).

VI. SUMMARY AND DISCUSSIONS

We have calculated the velocity autocorrelation function of a disk moving in a two-dimensional viscous fluid under the condition that the momentum leaks out of the fluid sheet with a characteristic relaxation time τ. In the long-time limit, the VACF decays exponentially, supporting the fact that the memory effects disappear. The obtained results are consistent with the previous calculations of diffusion coefficients for stationary flow [4].

The introduction of the momentum decay mechanism leads to the recovery of the fluctuation-dissipation theorem with which we can calculate the VACF under the influence of retardation effects. It has been shown that the VACF essentially decays exponentially even in the presence of memory effects. This exponential decay eliminates the Stokes paradox and ensures the finite mobility of the disk. We have also obtained algebraically decaying correction in the VACF depending on the coupling strength between the fluid sheet and the outer fluid. The results are

$$\phi(t) \sim e^{-t/\tau} t^{-3/2} \quad (6.1)$$

and

$$\phi(t) \sim e^{-t/\tau} t^{-1} \quad (6.2)$$

in the strong- ($\kappa' a \gg 1$) and the weak- ($\kappa' a \ll 1$) coupling limits, respectively. The correction in the weak-coupling limit t^{-1} reflects the two-dimensional character of the fluid sheet.

It is instructive to argue here the physical interpretation of the phenomenological relaxation time τ. According to (3.5) and (3.6), the translational diffusion coefficient in the weak-coupling limit ($\kappa a \ll 1$) is given by [7]

$$D = \frac{k_B T}{4\pi\eta} \left[\ln \frac{2}{\kappa a} - \gamma \right] . \quad (6.3)$$

This logarithmically weak size dependence corresponds to the result by Saffman and Delbrück [1,2]. They considered a system such that a membrane of dynamic viscosity η and thickness h is surrounded by a three-dimensional fluid which has smaller dynamic viscosity η' satisfying $\eta' \ll \eta$. It can be shown that (6.3) coincides with the result of Saffman-Delbrück theory

$$D = \frac{k_B T}{4\pi\eta h} \left[\ln \frac{h\eta}{a\eta'} - \gamma \right] , \quad (6.4)$$

by putting

$$\tau=\frac{1}{\nu}\left[\frac{h\eta}{2\eta'}\right]^2 , \tag{6.5}$$

except for the presence of h in the denominator of (6.4). This difference simply comes from the definition of the diffusion coefficient; we considered the force per unit thickness, while they treated the force itself.

As mentioned in Sec. II, the two-dimensional fluid model discussed so far is only a fluid on time scales shorter than τ. In the superlong-time regime ($t \gg \tau$), the flow velocity is no longer a slow mode, and the system has only two slow (diffusive) scalar modes, collective diffusion, and heat diffusion. According to the mode-coupling theory, the long-time tail is given by $\sim t^{-2}$, as in Lorentz gases. This argument has been supported by the recent quantitative analysis and corresponding simulation of the VACF for a two-dimensional lattice gas model of interacting particles [23,24].

It is worth pointing out that in the calculation of the VACF, one can also consider the long-time decay for the general case with τ being fixed. Upon expanding $\tilde{\lambda}[\omega]$ in (4.10) [see also (4.4)] as

$$\tilde{\lambda}[\omega]\approx\tilde{\lambda}+i\omega\tilde{\lambda}_1+O(\omega^2) , \tag{6.6}$$

we obtain the exponentially decaying VACF with a modified relaxation constant such that

$$\phi(t)=\frac{k_B T}{m^*}\exp\left[-\frac{\tilde{\lambda}t}{1+\tilde{\lambda}_1}\right] . \tag{6.7}$$

Finally, we remark that our result is in contrast to that obtained by Serra and Rubí [25]. They found that the VACF decays as $\sim t^{-1}$ and therefore exhibits a long-time tail. We found that the VACF always decays exponentially.

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APPENDIX

Asymptotic forms of modified Bessel functions are summarized in this appendix. As $z \to \infty$

$$K_n(z)\sim\sqrt{\pi/2z}\,e^{-z} . \tag{A1}$$

As $z \to 0$

$$K_0(z)\sim-(\ln\tfrac{1}{2}z+\gamma)+\tfrac{1}{4}z^2(-\ln\tfrac{1}{2}z-\gamma+1)+\cdots , \tag{A2}$$

$$K_1(z)\sim\frac{1}{z}+\tfrac{1}{2}z(\ln z-\tfrac{1}{4}+\tfrac{1}{2}\gamma)+\cdots , \tag{A3}$$

where γ is Euler's constant $\gamma=0.5772\ldots$.

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