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Quasi-two-dimensional diffusion of single ellipsoids: Aspect ratio and confinement effects

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We report on video-microscopy measurements of the translational and rotational Brownian motions of isolated ellipsoidal particles in quasi-two-dimensional sample cells of increasing thickness. The long-time diffusion coefficients were measured along the long ($D_a$) and short ($D_b$) ellipsoid axes, respectively, and the ratio, $D_a/D_b$, was determined as a function of wall confinement and particle aspect ratio. In three dimensions (3D), this ratio ($D_a/D_b$) cannot be larger than 2, but in quasi-two dimensions, wall confinement was found to substantially alter diffusion anisotropy and substantially slow particle diffusion along the short axis compared to 3D.

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I. INTRODUCTION

In many biological and industrial processes, diffusing particles are often nonspherical and often move in confined geometries. Examples of such particles include proteins diffusing in membranes [1] and fine grains migrating through the pores of micro- and nanoporous media. To date, few quantitative measurements of anisotropic particle diffusion in confined geometries have been reported. However, new particle fabrication schemes and imaging technologies with novel image analysis tools now make direct measurement of the diffusion of anisotropic particles readily possible. In this contribution, we investigate anisotropic diffusion and hydrodynamic confinement effects associated with the motions of isolated ellipsoidal particles between two parallel plates. The paper expands upon related results presented cursorily in the supplementary online materials of a short recent report [2].

In particular, the problem is given historical and theoretical context, the experimental methods for measurement of particle confinement and tracking errors are described at length, the systematic effects of counter-ion screening and vertical displacements are assessed, and the anisotropy results are discussed at length and in the context of a simple qualitative picture.

The Brownian diffusion coefficient $D$ of an isolated spherical particle is well understood. It is inversely proportional to the drag (or friction) coefficient $\gamma$ via the Einstein relation

$$D = k_B T / \gamma,$$

where $k_B$ is the Boltzmann constant and $T$ is the temperature. For a prolate spheroid with long axis of length $2a$ and two short axes of length $2b$, translational diffusion is anisotropic and described by diffusion coefficients $D_a = k_B T / \gamma_a$ along the long axis and $D_b = k_B T / \gamma_b$ along the short axes. The rotational diffusion coefficient of the prolate spheroid about its short axes is $D_\phi = k_B T / \gamma_\phi$. Generally, the drag coefficients $\gamma_a$, $\gamma_b$, and $\gamma_\phi$ depend on the shape and size of the ellipsoid. Brownian motion of anisotropic particles was first seriously considered by F. Perrin [3,4], who computed these drag coefficients analytically for a sphere diffusing in three dimensions (3D). Interestingly, the ratio $D_a/D_b$ varies from 1 to 2 in 3D, as the sphereoid aspect ratio, $\phi = a/b$, varies from 1 to infinity.

The problem of diffusion in confined geometries, such as quasi-two-dimensional (2D) media, is different from the 3D case as a result of a complex interplay between hydrodynamic drag, the boundaries of the medium, and the particle geometry. Surfaces near a moving particle modify fluid flow fields, often increasing particle hydrodynamic drag. A full theoretical formulation of wall hydrodynamic effects has been developed for one sphere (or ellipsoid) confined to one wall [5]. However, for more complicated situations such as a sphere or an ellipsoid confined by two parallel walls, the only available analytical solutions are for weak confinement in a few special symmetric configurations [5]. Recent numerical calculations [6], on the other hand, have been developed to derive the hydrodynamic drag of a single sphere and a linear chain of spheres confined more strongly in quasi-2D.

On the experimental side, the hydrodynamic drag of single spheres in weak confinement has been measured [7,8] and video microscopy has been applied recently to measure anisotropic particle diffusion, including ellipsoids in quasi-2D [2] and 3D [9], colloidal clusters near one wall [10], and carbon nanotubes in weak confinement [11]. In the present contribution, we report measurements of hydrodynamic drag on ellipsoids in quasi-2D, confined between two parallel walls. We explore the strong confinement regime where drag coefficients are not readily available from theory and simulation, and we report on a light interference method to accurately measure the confinement. We find that the diffusion anisotropy is made stronger and the diffusion along ellipsoid short axes is dramatically slowed by wall confinement. Note that the present work examines how spatial confinement affects long-time scale traditional diffusion coefficients: this aim contrasts with the investigation of short-time scale time-dependent translation-rotation coupling and non-Gaussian Brownian dynamics of a single ellipsoid, which was the focus of Ref. [2].
II. THEORY BACKGROUND

When a spheroid with semiaxes \((a, b, c)\) moves along one of its principle axes with velocity \(v\), through an unbounded quiescent fluid with viscosity \(\eta\) at low Reynolds number, then the translational and rotational (about short axis) drag coefficients affecting the spheroid are

\[ \gamma = 6\pi \eta G, \]

\[ \gamma_\theta = 6\eta VG_\theta, \]

where \(V\) is the volume of the spheroid and \(G\) is the geometric factor that renders the ellipsoid different relative to the case of a sphere. The geometric factors for prolate spheroids diffusing in 3D are analytically derived from Perrin’s equations [5]

\[ G_a = \frac{8}{3} \frac{1}{1 - \frac{dB}{2a}} + \frac{2}{(\phi - 1)^2} \ln\left(\frac{\phi + \sqrt{\phi^2 - 1}}{\phi - 1}\right), \]

\[ G_b = \frac{8}{3} \frac{1}{\phi^2 - 1 + \frac{2}{(\phi - 1)^2} \ln\left(\phi + \sqrt{\phi^2 - 1}\right)}, \]

and [3,12]

\[ G_\theta = \frac{2}{3} \phi^3 - 1 \frac{1}{\phi_{-1}} \ln\left(\phi + \sqrt{\phi^2 - 1} - 1\right). \]

Here, \(\phi = a/b\) is the ellipsoid aspect ratio. When \(\phi = 1\), then \(G = G_a = 1\) and Eq. (2) reduces to the translational and rotational Stokes laws for a sphere. Note also that Eqs. (2a) and (3) are obtained using stick boundary conditions, valid when the particle is much larger than the fluid molecules [13,14]. In Fig. 1, Eq. (3) is plotted as a function of \(\phi\) for \(\phi < 10\). When the aspect ratio \(\phi \gg 1\), Eqs. (1), (2a), (2b), and (3a)–(3c) yield

\[ D_a = \frac{k_BT \ln \phi}{2\pi \eta a}, \]

\[ D_b = \frac{k_BT \ln \phi}{4\pi \eta b}. \]

The ratio between these diffusion coefficients along long and short axes, i.e., \(D_a/D_b = G_b/G_a\), increases monotonically from 1 to 2 as \(\phi\) increases from 1 to infinity (in 3D). In quasi-2D, however, \(D_a/D_b\) can be larger than 2.

III. EXPERIMENT

The diffusion of micrometer size polymethyl methacrylate (PMMA) and polystyrene (PS) ellipsoids was measured in water confined between two glass walls. Both PS and PMMA ellipsoids are synthesized by the method described in Ref. [15]. Briefly, we placed 0.5% (by weight) PS spheres into a 12% (by weight) aqueous polyvinyl alcohol (PVA) solution residing in a Petri dish. After water evaporation, the PVA film was stretched at 130 °C. The PS (or PMMA) spheres embedded in the film are readily stretched under these circumstances because their glass transition temperatures are below 130 °C. After cooling to room temperature, the PVA was dissolved and ellipsoids obtained. Note that the initial PMMA or PS spheres must not be cross-linked, otherwise they cannot be stretched. We measured the size of ellipsoids by scanning electron microscopy (SEM) and by optical microscopy.

The ellipsoid solutions were cleaned and stabilized with 7 mM sodium dodecyl sulfate (SDS). The ellipsoids were not expected to have strong interactions with the glass surfaces because the solution ionic strength was more than 0.1 mM and the Debye screening length for the particles was correspondingly less than 30 nm. However, it is difficult to estimate the ionic strength accurately in a thin cell because the glass surfaces can release Na\(^+\) ions [16]. Nevertheless, we found that the addition of 2 mM salt to the solution did not induce a detectable change in particle diffusion coefficients. This observation suggests the double layers are not significantly affecting particle diffusion.

Glass surfaces of the sample cell were rigorously cleaned in a 1:4 mixture of hydrogen peroxide and sulfuric acid by sonication. Then the glass was thoroughly rinsed in deionized water and quickly dried with an air blow gun. Typically 0.3 μL solution spread over the entire 1.8 × 1.8 mm\(^2\) coverslip area and ellipsoids did not stick to the surfaces. Because the ellipsoid’s gravitational height, \(k_BT/mg\), is much larger than the cell thickness, \(H\), the ellipsoids were readily suspended around midplane between the two walls. Finally, the cell was sealed with UV-cured adhesive (Norland 63).

We measure the wall separation by light interference. When the cell thickness is below a few micrometers, then the interference colors produced by reflections from the two inner surfaces of the sample walls in white light illumination can be observed by eye or in the reflection mode of the microscope [see Figs. 2(b) and 2(c)]. When the wall separation \(H = 0\), the effective light path difference is \(\Delta l = \lambda/2\) due to the \(\pi\) phase shift of reflection at the lower surface. Thus all wavelength components of the white light yield a dark black color in interference at \(H = 0\). When \(H > 0\), the reflected light in the normal direction is a mixture of light with various wavelengths, and different wavelengths contribute with different weights to the observed color. White light interference from a wedge, for example, will yield bands of colors as in the Michel-Levy chart [17]. By comparing the experimentally observed color to the Michel-Levy chart, one can effec-
ent uncertainty as described above, the relative values of different colors generated by interference of the top/bottom sample cell walls. Color changes due to variation in the separation between the top sample wall and the ellipsoid top surface are diffusive with period $D_{bt}$, where $D_{bt}$ may be subject to misreading the color by one or more periods, we established a reference of their corresponding MSDs [18]. Using these procedures, we assign an orientation resolution of $1^\circ$ and spatial resolutions of 0.5 pixel=40 nm along the particle’s short axis and 0.8 pixel=64 nm along its long axis because of the superimposed small tumbling motion [2].

From the image analysis, we obtained the trajectory of a particle’s center-of-mass positions $x(t_n)=[x(t_n),y(t_n)]$ in the laboratory frame and its orientation angle $\theta(t_n)$ relative to the $x$ axis at times $t_n=n(1/30)$ s [see Fig. 1c of Ref. [2]]. We define each 1/30-s time interval as a step. During the $n$th step, the particle’s position changes by $\delta x(t_n)=x(t_n)-x(t_{n-1})$ and its angle by $\delta \theta(t_n)=\theta(t_n)-\theta(t_{n-1})$. To obtain the drag coefficients along long and short axes, we need to convert the measured displacements from the fixed laboratory frame to the local body frame. Step displacements $\delta \mathbf{x}_{n}$ relative to the local body frame and step displacements $\delta \mathbf{u}_{n}$ relative to the fixed laboratory frame are related via

$$
\begin{pmatrix}
\delta x_n \\
\delta y_n
\end{pmatrix} =
\begin{pmatrix}
\cos \theta_n & \sin \theta_n \\
-\sin \theta_n & \cos \theta_n
\end{pmatrix}
\begin{pmatrix}
\delta u_n \\
\delta v_n
\end{pmatrix},
$$

where $\theta_n=[\theta(t_{n-1})+\theta(t_n)]/2$ [see Fig. 1c of Ref. [2]]. In practice, choosing $\theta_n=\theta(t_{n-1})$ or $\theta_n=\theta(t_n)$ has little effect on our results because $\theta$ barely changes during 1/30 s.

IV. RESULTS AND DISCUSSION

In both the laboratory and the body frame, MSDs are diffusive with $\langle (\Delta x(t) )^2 \rangle = 2 D_x$, $\langle (\Delta y(t) )^2 \rangle = 2 D_y$, $\langle (\Delta x(t) )^2 \rangle = (D_x+D_b) t = 2 D_x$, and $\langle (\Delta \theta(t) )^2 \rangle = 2 D_{bt}$ [2]. Here the brackets, $\langle \rangle$, denote data averaged over all possible time intervals in the whole trajectory of an ellipsoid. For example, an N-step total time trajectory (with 30 steps per second due to our frame rate of 30 Hz) contains $N \sim 30 n$ mean-square displacement data points for a correlation time interval of $t$ seconds. Figure 3 shows MSDs of a 2.4 $\times$ 0.3 $\times$ 0.3 $\mu$m$^3$ ellipsoid confined in an 846-nm-thick cell. All curves exhibit diffusive behavior and the diffusion coefficients, $D=\text{MSD}/(2t)$, shown in the figure are derived from the best fit lines. The slopes show that $D_x=0.161$ $\mu$m$^2$/s, $D_y=0.179$ $\mu$m$^2$/s, $D_a=0.044$ $\mu$m$^2$/s, and $D_x=D_y=(D_a+D_b)/2$. 
The error bars associated with each point in the figure, i.e., the standard deviations of the MSDs, were measured to be proportional to $t^{1/2}$. This can be understood from the probability theory. As an example, we derive the error of the total MSD, $(\Delta r)^2=\langle \Delta x^2 \rangle + \langle \Delta y^2 \rangle + \langle \Delta z^2 \rangle$, $\Delta a$ and $\Delta b$ are statistically independent since $\gamma_a$ and $\gamma_b$ are constant independent of angle. Consequently, the variance $\sigma^2(\Delta r)^2=\sigma^2(\Delta a)^2+\sigma^2(\Delta b)^2$. In probability theory [19], given two probability distribution functions (PDFs), $f_\alpha(x)$ and $f_\beta(x)$ for $\alpha$ and $\beta$ statistically independent, the joint PDF of their product is

$$f_{\alpha\beta}(x) = \int_{-\infty}^{\infty} f_\alpha(x_1)f_\beta(x-x_1)dx_1.$$  

(6)

Since $\Delta a$ follows a Gaussian distribution [2] $f_{\Delta a}(x)=\frac{1}{\sqrt{2\pi\sigma_a}}e^{-x^2/2\sigma_a^2}$, the probability density function of $(\Delta a)^2$ is

$$f_{(\Delta a)^2}(x) = \begin{cases} \frac{1}{\sqrt{2\pi\sigma_a}}e^{-x^2/2\sigma_a^2}, & x \geq 0 \\ 0, & x < 0 \end{cases}.$$  

(7)

This distribution of $(\Delta a)^2$ was confirmed in our data. Hence the variance of $(\Delta a)^2$ is

$$\sigma^2(\Delta a)^2 = \int_{-\infty}^{\infty} f_{(\Delta a)^2}(x)x^2dx$$

$$= \int_{0}^{\infty} \frac{1}{\sqrt{2\pi\sigma_a}}e^{-x^2/2\sigma_a^2}x^2dx = 3\sigma_a^4 = 12\sigma_a^2.$$  

(8)

Here, $t$ is the step time. Thus the standard deviation of $(\Delta r)^2$ is

$$\sigma = \sqrt{12\sigma_a^2t + 12\sigma_b^2t} / \sqrt{t_{end}} = t^{1/2},$$  

(9)

where $M$ is the number of independent steps during the whole experimental time $t_{end}$. (Note that the error bars of the figures in Ref. [2] were similarly calculated, albeit no theoretical explanation was given.)

![FIG. 3. (Color online) MSDs of a 2.4×0.3×0.3 μm³ ellipsoid confined in an 846-nm-thick cell. The four lines arranged from top to bottom, respectively, are MSDs along $a$, $x$, $y$, and $b$ axes. Symbols represent experimental data and lines represent linear fits to these data. (Insets) Angular MSD (circles) and the best fit (line).](image)

![FIG. 4. (Color online) Ratio of theoretical 3D diffusion coefficient [5] along the ellipsoid long axis, $D_a^{3D}$, to the measured diffusion coefficient, $D_a$, for ellipsoids confined at $2b/H$. Diamonds: 2.4×0.3×0.3 μm³ ($\phi=8$) ellipsoids; circles: 3.3×0.635×0.635 μm³ ($\phi=5.2$) ellipsoids; solid circles: from samples with no added salt; open circles: from samples with 2 mM added salt; green solid circle: from sample with bovine serum albumin (BSA) covered glass surfaces; squares: all other samples—lower aspect ratio spheroids with particle aspect ratios labeled below each data point. The accuracy of $2b/H$ for these measurements is similar to other samples. Dashed curves: guides for the eyes. Solid curve ($\phi=1$): replot of the numerical prediction in Fig. 1 of Ref. [6] for a sphere strictly in the $H/2$ midplane.](image)

![FIG. 5. (Color online) Ratio of theoretical 3D diffusion coefficient [5] along the ellipsoid short axis, $D_b^{3D}$, to the measured diffusion coefficient, $D_b$, for ellipsoids confined at $2b/H$. Symbols are the same as those in Fig. 4. Solid curves from left to right: theoretical weak confinement predictions [5] for aspect ratios $a/b=3,2$ and numerical result [6] for aspect ratio $a/b=1$ ranging over both weak and strong confinement regimes.](image)

We repeated the experiments described above for different ellipsoids under different confinement conditions. From the slopes of their MSDs, we obtain $D_{\alpha}, D_{\beta}$, and $D_\gamma$ of different particles as a function of confinement condition as shown in Figs. 4–6, respectively. Specifically, the normalized quantities, $D_i^{3D}/D_i = \gamma_i/\gamma_i^{3D}$, for $i=\alpha, \beta, \gamma$, are plotted as a function of increasing confinement, $2b/H$. Here the 3D normalization constants, $D_i^{3D}$ (alternatively, $\gamma_i^{3D}$), are calculated from Eqs. (2) and (3). (Note that these figures contain the same infor-
be small due to the high ionic strength of the suspension. As the regime where ionic concentration was low and spheroids been observed with depolarized dynamic light scattering in length of the ellipsoid. This effect in rotational diffusion has tional drag is proportional to the volume rather than the down diffusion, especially rotational diffusion because rota-
suspension increase their hydrodynamic diameter and slow

distinguishable. The analytical Eq. (3) shows that the 6.9 nm screening layer of 2 mM solution will lower the 3D diffusion coefficients by less than 2%.

Notice that 2b/H=0 corresponds to the 3D limit wherein $D^{3D}/D=1$. As expected, hydrodynamic drag increased and the diffusion coefficients correspondingly decreased as the confinement became stronger. The larger positive slopes exhibited by the more needlelike spheroids are indicative of motions more sensitive to confinement. Furthermore, the slopes of the same ellipsoids in Fig. 5 are larger than those in Fig. 4, indicating that diffusion along the ellipsoid short axis is more strongly affected by the confinement than diffusion along the long axis. Limited comparisons to all available analytical and numerical predictions (i.e., the solid curves in Figs. 4 and 5) suggest that our data exhibit the generally expected trends with increasing confinement. Note that the solid curves of analytical and numerical predictions in Figs. 4 and 5 are for particles forced to sit in the midplane. In real experiments, the measured drag is an average over different $z$. In our experiments, there are no detectable interfer-
color changes at the centers of ellipsoids. Consequently $z$ fluctuations are less than $-50$ nm, i.e., $\sim H/20$. Numerical results in Ref. [6] indicate that the drag on a sphere at $H/3$ is very close ($<10\%$) to the drag at $H/2$. Thus the $z$ fluctuations of our ellipsoids should have only a very small effect on the reported particle drag.

Another question that our data hold potential to explore concerns the effect of electric double layers on ellipsoid diffu-
sion. The electric double layers around charged particles in suspension increase their hydrodynamic diameter and slow down diffusion, especially rotational diffusion because rota-
tional drag is proportional to the volume rather than the length of the ellipsoid. This effect in rotational diffusion has been observed with depolarized dynamic light scattering in the regime where ionic concentration was low and spheroids small [20]. In our systems, such effects might be expected to be small due to the high ionic strength of the suspension. As can be seen in Figs. 4–6, diffusion coefficients for the samples with 2 mM added salt and no added salt are indistin-
guishable. The analytical Eq. (3) shows that the 6.9 nm screening layer of 2 mM solution will lower the 3D diffusion coefficients by less than 2%.

Finally, Fig. 7 shows the impact of aspect ratio on the translational diffusion anisotropy ratio $D_a/D_b$. Here, it is evi-
dent that diffusion in quasi-2D is quite different from diffu-
sion in 3D. For 3D, $D_a/D_b$ asymptotes to 2 at large aspect ratio, as shown by the solid theoretical curve. For quasi-2D, on the other hand, $D_a/D_b$ grows very rapidly with increasing aspect ratio. Since we expect the stick boundary condition to hold in this system, the observation that $D_a/D_b < 2$ is purely due to confinement.

A schematic to qualitatively capture this basic effect is given in Fig. 8. Imagine fluid flowing past the ellipsoid. In 3D, the fluid flow pathways will be displaced by distances of order $2b$ in order to “go around” the ellipsoid. This fluid flow displacement is the approximately the same, whether the spheroid is orientated either parallel or perpendicular to the flow, and therefore $\gamma_0$ and $\gamma_z$ are thus comparable. In 2D, however, the fluid flow displacement pathway is approxi-
mately $2b$ (or $2a$) when the spheroid is oriented parallel (per-
collinear) to the flow, so that $\gamma_0/\gamma_z$ diverges with increasing $a/b$. This very simple qualitative picture also explains our observation that diffusion along the ellipsoid short axes is more strongly affected by the confinement than diffusion.
along the long axis. The rotational diffusion is also not so strongly affected by the confinement (e.g., similar to $D_{z}$) because “easy” flow pathways exist to accommodate this ellipsoid motions. Finally, we note that in quasi-2D confinement, $D_{z}/D_{y}$ increases with increasing aspect ratio and should eventually saturate according to numerical calculations [6] at a value much larger than 2 because some fluid will always “leak” between the particle and walls.

In summary, we have found that the anisotropic drag coefficients for ellipsoid diffusion substantially increase when the ellipsoids are strongly confined between parallel plates, especially diffusion along their short axes. In the future, many questions about these systems will be exciting to explore, including the effects of neighboring ellipsoids and the effects of other confinement geometries. For example, quasi-one-dimensional (1D) confinement of an ellipsoid will align the ellipsoid along the diffusion direction. This effect may compensate drag from the boundaries and lead to an optimal diameter for ellipsoid diffusion speed in a quasi-1D cylinder.

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