Diffusion of a Deformable Body in a Random Flow

Gady Frenkel, Moshe Schwartz

Raymond and Beverly Sackler Faculty of Exact Sciences
School of Physics and Astronomy
Tel Aviv University, Ramat Aviv, 69978, Israel

We consider a deformable body immersed in an incompressible liquid that is randomly stirred. Sticking to physical situations in which the body departs only slightly from its spherical shape, we calculate the diffusion constant of the body. We give explicitly the dependence of the diffusion constant on the velocity correlations in the liquid and on the size of the body. We emphasize the particular case in which the random velocity field follows from thermal agitation.

PACS: 05.40.-a, 82.70.-y, 83.50.-v

Keywords: drop diffusion, noise, deformable body, size effect, deformation

Deformable bodies that are nearly spherical can be found in various soft matter systems such as emulsions and complex fluids, where deformable bodies are immersed in a host fluid. It is easy to envisage a situations in which the host fluid is stirred randomly. For example, mixing of bodies in an emulsion by external mechanical vibrations (e.g., ultrasound). Another example is thermal agitation of a complex fluid. Hence, it is interesting to investigate the dynamics of deformable bodies in random flow. In this work we study the movement of a single deformable body in a random velocity field, which is uncorrelated in time and correlated in space in a general way. We focus on three aspects of this subject: derivation of the explicit equation of motion for the body’s center, the Mean Squared Displacement (MSD) of the center and the effect of the body’s size on the diffusion constant. Consider a single deformable body immersed in a host fluid. The system is chosen to have
the following characteristics:

1. The host fluid is incompressible, \( \nabla \cdot \vec{v} = 0 \). Moreover, we assume that the Reynolds number is small so that the Stokes approximation is applicable.

2. The body is characterized by an energy that depends on its shape. The shape of minimum energy is a sphere. The energy may be surface tension \( \mathbb{8} \), Helfrich bending energy \( \mathbb{1,5} \), etc. The body’s surface shape is described by the equation: \( \psi(\vec{r}) = 0 \) where \( \psi(\vec{r}) \) is a scalar three dimensional field.

3. A deformation of the body induces a force density on the host fluid. As a result, the fluid’s velocity is given by the sum of \( \vec{v}_{ext} \), which is caused by external sources, and \( \vec{v}_\psi \), which is induced by the body.

4. The external velocity, \( \vec{v}_{ext} \), is random and is chosen to have zero average and known correlations. It is convenient to define the external velocity in terms of its spatial Fourier transform. Since the fluid is incompressible, we will express the velocity in a general form that has no longitudinal part:

\[
v_{ext}(\vec{q}) \equiv \sum_j \left( \delta_{ij} - \frac{q_i q_j}{q^2} \right) u_j(\vec{q}),
\]

where \( \vec{u} \) is some general vector field. Next, we define the correlations of the velocity by the correlations of \( \vec{u} \),

\[
\langle u_i(\vec{q}, t) \rangle = 0 \quad \text{and}
\]

\[
\langle u_i(\vec{q}, t_1) u_m(\vec{p}, t_2) \rangle = \delta_{im} \delta(\vec{q} + \vec{p}) \phi(\xi q) \delta(t_2 - t_1),
\]

where \( \delta_{im} \) is the Kronecker delta, \( \delta() \) is the Dirac delta function and \( \xi \) is the velocity correlation length.
5. The surface elements of the body are carried by the host fluid, i.e. each surface point moves according to

\[ \dot{\vec{r}} = \vec{v}_{\text{ext}}(\vec{r}) + \vec{v}_\psi(\vec{r}). \]  

(3)

6. We assume that the external velocity is weak enough to cause only minor shape fluctuations of the body.

We will be interested in the following in the Mean Squared Displacement (MSD) of the center. Since the body is deformable the definition of its center is not unique. We will choose a specific definition later. It turns out, however, that the value of the MSD at long times does not depend on the specific choice, because for long times the MSD (according to any definition) is much larger than the size of the body.

Following the line of derivation of Schwartz and Edwards, equation (3) may be turned into a continuity equation for \( \psi \),

\[ \frac{\partial \psi}{\partial t} + (\vec{v}_{\text{ext}} + \vec{v}_\psi) \cdot \nabla \psi = 0. \]  

(4)

Consider a deformable body, carried by the host fluid in such a way that at any instant it is nearly spherical. Its state can thus be characterized by the position of its center, \( \vec{r}_0(t) \), and a deformation function \( f(\Omega, t) \) that describes the shape by the equation

\[ \frac{\rho}{R} + f(\Omega, t) - 1 = 0, \]  

(5)

where \( \rho \) is the distance of the surface from the center in the direction of the solid angle \( \Omega \) and \( R \) is the radius of the body when not deformed. The deformation function \( f \) can be expanded in spherical harmonics, \( f(\Omega, t) = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} f_{l,m}(t) Y_{l,m}(\Omega) \). The center of the shape, \( \vec{r}_0(t) \), is defined as that point around which \( f_{1m}(t) = 0 \) for \( m = -1, 0, 1 \). Parameterizing now the gauge field \( \psi \) as \( \psi(\vec{r}, t) = \frac{|\vec{r} - \vec{r}_0|}{R} + f(\Omega, t) - 1 \), equation (3) leads to a linear equation for each
\[ f_{l,m}, \]
\[ \frac{\partial f_{l,m}}{\partial t} + \lambda_l f_{l,m} + \frac{1}{R} [\hat{\rho} \cdot (\vec{v}_{\text{ext}} - \vec{r}_0)]_{lm} = 0, \tag{6} \]

where \( \hat{\rho} \) is a unit vector directed outwards from the center in the direction of \( \Omega \), and
\[ [\hat{\rho} \cdot (\vec{v}_{\text{ext}} - \vec{r}_0)]_{lm} = \int d\Omega \left\{ \hat{\rho} \cdot \left[ \vec{v}_{\text{ext}}(\vec{r}_0 + R(1 - f)\hat{\rho}) - \vec{r}_0 \right] Y_{l,m}^* (\Omega) \right\} \tag{7} \]

The eigenvalues \( \lambda_l \)'s characterize the decay rate of a slightly deformed sphere into a sphere in the absence of the external velocity. Note that the term \( \lambda_l f_{lm} \) in Eq. (6) results from the shape induced velocity, \( \vec{v}_\psi \), in Eq. (4) \[7\]. Different physical systems are characterized by different sets of \( \lambda_l \)'s. For example, Schwartz and Edwards \[7\] calculate \( \lambda_l \) for a droplet with a constant surface tension and equal viscosities inside and outside the droplet, Gang at al. \[2\] give \( \lambda_l \) for a droplet with a surface tension under the assumption that the viscosity of its interior is much greater than that of the surrounding fluid, Milner and Safran \[8\] do the same for a surface controlled by bending energy and Dörries and Foltin add to it in-plane dissipation \[9\]. In all of these cases, it is obvious that the decay must depend only on \( l \) because of the spherical symmetry. Therefore, our treatment is general and applicable to a large class of physical systems in which the decay rate depends only on \( l \).

Equation (6) implies that in order that \( f_{1,m} \) stays zero for all times we must have as an equation determining the location of the center
\[ [\hat{\rho} \cdot (\vec{v}_{\text{ext}} - \vec{r}_0)]_{1m} = 0, \quad m = -1, 0, 1. \tag{8} \]

For \( l \neq 1 \) it is clear that \( \vec{r}_0 \) can be dropped from the last term on the left hand side of Eq. (6). Therefore \( f(\Omega, t) \) is linear in \( \vec{v}_{\text{ext}} \) (for long enough times the initial deformations have already decayed). Consequently we can always drop, for small enough \( \vec{v}_{\text{ext}} \), \( f \) in the argument of \( \vec{v}_{\text{ext}} \) on the right hand side of Eq. (7). The result is decoupling of the deformation degrees of freedom from that of the center of the sphere. The equation for the motion of the center can thus be given, using linear combinations of \( Y_{1,m} \), in vector form as
\[ \int d\Omega \hat{\rho} \left( \hat{\rho} \cdot \vec{r}_0 \right) = \int d\Omega \hat{\rho} \left( \hat{\rho} \cdot \vec{v}_{\text{ext}}(\vec{r}_0 + R\hat{\rho}) \right), \tag{9} \]
Direct integration of the left hand side, Fourier decomposition of \( \vec{v}_{\text{ext}}(\vec{r}_0 + R\dot{\rho}) \) on the right hand side and the use of the partial waves decomposition \([10,11]\):

\[
e^{-i\vec{q} \cdot (\vec{r}_0 + R\dot{\rho})} = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} (-i)^l 4\pi j_l(qR) Y^*_l m(\Omega_q) Y_{lm}(\Omega),
\]

where \( \Omega_q \) is the solid angle in the \( \vec{q} \) direction, yields

\[
\dot{\vec{r}}_0 = 3 \int d\vec{q} e^{-i\vec{q} \cdot \vec{r}_0} \left( \frac{1}{3} j_0(qR) + j_2(qR) A \right) \vec{v}_{\text{ext}}(\vec{q},t),
\]

where \( j_0 \) and \( j_2 \) are spherical Bessel functions and the matrix \( A(\vec{q}) \) is given by

\[
A_{ij} = -\frac{2}{3} \delta_{ij} + \left( \delta_{ij} - \frac{q_i q_j}{q^2} \right).
\]

It may seem that \( A \) on the right hand side of Eq. \([11]\) mixes directions. However, the bracketed term in Eq. \([12]\) is just a projection operator on the transverse direction. The external velocity is incompressible and hence already transverse. Consequently, this term acts as a unity operator, \( \delta_{ij} \) and Eq. \([11]\) leads to

\[
\dot{\vec{r}}_0 = \int d\vec{q} e^{-i\vec{q} \cdot \vec{r}_0} (j_0(qR) + j_2(qR)) \vec{v}_{\text{ext}}(\vec{q},t).
\]

Equation \([13]\) is the explicit equation of motion for the center of the body. In the limit \( R \to 0 \) the approximation, \( \vec{r}_0 = \vec{v}_{\text{ext}}(\vec{r}_0, t) \) is obtained. Note that this equation is general and describes the motion of the center for any given (small enough) external velocity field.

Next, we calculate the MSD, \( \langle (\Delta \vec{r}_0)^2 \rangle \), as a function of the elapsed time, \( t \). The starting point is the trivial relation

\[
\Delta \vec{r}_0(t) = \int_0^t \dot{\vec{r}}_0(t')dt'.
\]

Assuming the decomposition \([12,13]\)

\[
\left\langle v_{\text{ext}i}(\vec{q}_1, t_1) v_{\text{ext}j}(\vec{q}_2, t_2) e^{-i\vec{q}_1 \cdot \vec{r}_0(t_1)} e^{-i\vec{q}_2 \cdot \vec{r}_0(t_2)} \right\rangle =
\left\langle v_{\text{ext}i}(\vec{q}_1, t_1) \right\rangle \left\langle v_{\text{ext}j}(\vec{q}_2, t_2) \right\rangle \left\langle e^{-i\vec{q}_1 \cdot \vec{r}_0(t_1)} e^{-i\vec{q}_2 \cdot \vec{r}_0(t_2)} \right\rangle
\]

(15)
and using equations (1), (2) and (13) the MSD is

$$\langle (\Delta \vec{r}_0(t))^2 \rangle = 8\pi \int_0^\infty q^2 dq \phi(q) (j_0(qR) + j_2(qR))^2 t \equiv 3Dt. \quad (16)$$

We expect the diffusion coefficient to depend on the ratio $\gamma = \frac{R}{\xi}$ in the following way. As $\gamma$ increases the diffusion coefficient decreases. This is due to the fact that as $\gamma$ increases, different regions of the surface become less correlated and move in different directions. In the limit $\gamma \rightarrow \infty$, the movement of the center ceases and the MSD is always zero. In the limit $\gamma \rightarrow 0$, the bracketed Bessel term in Eq. (16) can be replaced by unity. A close inspection of the derivation reveals that this limit produces the same MSD equation as the approximated equation $\dot{\vec{r}}_0 = \vec{v}_{ext}(\vec{r}_0)$ that is accurate in the limit of infinite correlation length, or point particles.

We turn to evaluate the dependence of the diffusion coefficient on $R$ and $\xi$ in the above limiting cases. Consider the correlation function $\phi(q) = C \ (q\xi)^\alpha g(q\xi)$, where $g$ is a cutoff function and $g(0) = C_0 > 0$. The diffusion constant is given by

$$D = \frac{8\pi C\xi^3}{3R^3+\alpha} \int_0^\infty du \ u^{2+\alpha} g(\frac{\xi}{R}u)[j_0(u) + j_2(u)]^2. \quad (17)$$

In the limit $R/\xi \rightarrow \infty$ we distinguish between two cases: $\alpha < 1$ and $\alpha > 1$. Since the large $u$ dependence of $j_0(u) + j_2(u)$ is proportional to $\cos(u)/u^2$ we find that

$$D \propto \begin{cases} \frac{C\xi^\alpha}{R^{3+\alpha}} & \text{for } \alpha < 1 \\ \frac{C\xi^2}{R^3} & \text{for } \alpha > 1. \end{cases} \quad (18)$$

In the opposite limit $R/\xi \rightarrow 0$ we find that regardless of $\alpha$

$$D \propto \frac{C}{\xi^3}. \quad (19)$$

(Note here that we have written the power law dependence of $\phi(q)$ as $q^\alpha \xi^\alpha$ but having other dimensional constants in the model may make $C$ depend on $\xi$, so that eqs. (18) and (19) may be considered only as equations that yield the dependence of $D$ on the radius $R$).

It is interesting to consider the case where the fluctuations in the velocity field are due to
thermal agitation. General considerations show that the correlations of the external velocity must be, in this case, of the form

\[
\langle v_{ext,i}(\vec{q},t_1)v_{ext,j}(\vec{p},t_2)\rangle = \left(\delta_{ij} - \frac{q_i q_j}{q^2}\right) \delta(\vec{q} + \vec{p})\delta(t_2 - t_1)\frac{c}{q^2},
\]

where \(c\) is a dimensional constant (Strictly speaking the above is valid for \(qa > 1\), where \(a\) is the inter-particle distance). We use these correlations with Eq. (16) and obtain

\[
D = \frac{8\pi^2 c}{5R}.
\]

Dimensional analysis reveals that \(c\) must be proportional to \(K_B T/\eta\) (with a dimensionless proportionality constant). A detailed calculation yields a proportionality constant equal to \((2\pi)^{-3}\). We do not represent here the detailed calculation, because it yields a result identical to that obtained in the past [14,15] using two totally different approaches (that we wanted to check by a more direct calculation. Actually, Hadamard calculated the mobility that can be related to the diffusion constant by the Einstein relation). Therefore, for \(R\) larger than the inter-particle distance in the liquid,

\[
D = \frac{K_B T}{5\pi \eta R}.
\]

Note that this result, for a liquid membrane that has liquid inside as well as outside, is different from the well known Stokes result for a hard sphere and from the result for a polymer subjected to thermal fluctuations [16] (The difference is in the prefactors).

Fig. 1 depicts \(D\) for typical correlation functions while Fig. 2 depicts the dependence of the diffusion coefficient on \(R, \xi\), for two noise realizations. The first is just a cutoff function, \(\phi(q) = C \exp(-q^2\xi^2)\). The second corresponds to the temperature driven randomness, \(\phi(q) = C' (q\xi)^{-2}\exp(-q^2\xi^2)\). We keep \(\xi\) constant and vary \(R\). There are two distinct regimes: For \(R/\xi < 1\) the diffusion coefficient is not sensitive to \(R\), while for \(R/\xi > 1\) the slope of the graph turns towards -3 for the cutoff function and towards -1 for the temperature driven randomness.
[1] X. L. Wu and A. Libchaber, Phys. Rev. Lett. 84, 3017 (2000).

[2] Hu Gang, A. H. Krall, and D. A. Weitz, Phys. Rev. E 52, 6289 (1995).

[3] Y. Navot, Phys. of fluids 11, 990 (1999).

[4] H. J. Deuling and W. Helfrich, J. Physique 37, 1335 (1976).

[5] V. Lisy, B. Brutovsky, and A. V. Zatovsky, Phys. Rev. E 58, 7598 (1998).

[6] S. F. Edwards and M. Schwartz, Physica A 167, 595 (1990).

[7] M. Schwartz and S. F. Edwards, Physica A 153, 355 (1988).

[8] S. T. Milner and S. A. Safran, Phys. Rev. A 36, 4371 (1987).

[9] G. Dörries and G. Foltin, Phys. Rev. E 53, 2547 (1996).

[10] D. Bohm, Quantum Theory, Prentice-Hall (1951).

[11] R. H. Landau, Quantum Mechanics II, Wiley-Interscience Publication, 2nd Ed. (1996).

[12] G. Frenkel and M. Schwartz, Europhys. lett. 50, 628 (2000).

[13] M. Schwartz and R. Brustein, J. Stat. Phys. 51, 585 (1988).

[14] S. F. Edwards and M. Schwartz, Physica A 178, 236 (1991).

[15] J. S. Hadamard, Comp. Rend. Acad. Sci. 152, 1735 (1911).

[16] M. Doi and S. F. Edwards, The Theory of Polymer Dynamics, Oxford Science Publications (1986).
Figure captions

Fig. 1
The dependence of the diffusion coefficient $D$ on the ratio of the radius $R$ of the sphere to the correlation length $\xi$, for typical correlation functions. The axis are presented in non-dimensional units, where $D_0 = C/\xi^3$.

Fig. 2
The dependence of the diffusion coefficient on $R$. We keep $\xi$ constant and vary $R$. For negative $\ln(R/\xi)$ there is no $R$ dependence, while for positive values $D$ is proportional to $R^\mu$ where $\mu$ tends to (a) -1 for thermal agitation and (b) -3 for a simple cutoff function.
\[ \frac{D}{D_0} \]

\[ \phi(q) = C \, \delta(q\xi - 1) \]

\[ \phi(q) = C \, e^{-q^2\xi^2} \]

\[ \phi(q) = C \, \Theta(1 - q\xi) \]

**FIG. 1.**
(a) $\phi(q) = C \left( q\xi \right)^{-2} e^{-q^2\xi^2}$
(b) $\phi(q) = C e^{-q^2\xi^2}$