Stokes Drag on a Sphere in a Nematic Liquid Crystal

J. C. Loudet,* P. Hanusse, P. Poulin

A sphere of radius R moving in a fluid of viscosity η experiences a Stokes drag force $F_S = 6\pi\eta R v$, where v is the velocity of the sphere (*1*). For anisotropic liquids, an anisotropic drag force is predicted (*2*, *3*). We experimentally determined the Stokes drag coefficients of a Brownian sphere immersed in a nematic liquid crystal phase. Unlike previous falling ball experiments (*3*), which can only yield an average viscous drag, we used video microscopy coupled with particle tracking routines (*4*) to quantitatively measure the anisotropic diffusion coefficients.

A nematic liquid crystal possesses an orientational order specified by a unit vector, **n**, called the director (*5*). In the absence of any perturbation, the director field is uniformly aligned. The presence of a particle alters the orientational order of the nematic, which yields to elastic distortions and topological defects (*6*, *7*).

We studied a binary mixture composed of a thermotropic nematic phase (E7, Merck) and an isotropic dispersed phase (silicone oil, Aldrich, a copolymer of dimethylsiloxane and methylphenylsiloxane). At high temperature, the mixture ($\phi_{E7} > 95$ weight percent, where ϕ is the weight fraction) forms an isotropic phase. When a sample is thermally quenched, the subsequent phase separation leads to the formation of isotropic droplets embedded in the nematic solvent (*8*). We focused on the early stages of the phase separation, when the small coarsening oil droplets exhibit the Saturn or surface ring configuration, in which a disclination line surrounds the sphere at its equator (Fig. 1A, inset) (*8*, *9*). This configuration is stable against thermal fluctuations and has quadrupolar symmetry (*7*).

Working with very dilute suspensions ($\phi_{oil} \leq 0.6$ weight percent), these noninteracting quadrupoles, the diameters of which range from 1 to 2 μm, perform Brownian motion. Using video microscopy and particle tracking routines (*4*), we analyzed the Brownian fluctuations and determined the particle positions at regular time steps. A typical trajectory derived from 1000 snapshots is shown in Fig. 1A. The self-diffusion coefficient of a random walker is given by the Stokes-Einstein relation $D = k_B T/6\pi\eta R$, where k_B is the Boltzmann constant and T the temperature (*5*). Stokes drag forces can therefore be computed, provided that D (and hence η) can be experimentally determined. Because the nematic phase has a rotational symmetry axis, the Brownian motion is governed by two independent diffusion coefficients, namely $D_\parallel$ and $D_\perp$, which characterize the diffusion along and perpendicular to the director, respectively. They are given by $D_{\parallel/\perp} = k_B T/6\pi\eta_{\parallel/\perp} R$, where $\eta_\parallel$ and $\eta_\perp$ are related to the intrinsic viscosity coefficients of the liquid crystal material (*2*, *3*).

From the analysis of the Brownian fluctuations, it is possible to derive $D_\parallel$ and $D_\perp$. The probability that a particle will diffuse a distance δ in the plane in time τ obeys the Gaussian distribution (*5*): $P(\delta|\tau) = P_0(\tau)\exp\left(\frac{-\delta^2}{\Delta^2(\tau)}\right)$, where $P_0(\tau)$ is a normalization constant and $\Delta(\tau)$ is the width of the distribution. Averaging over 18,000 trajectory steps, Fig. 1B shows the histograms of the particle displacements $\delta = |\mathbf{r}(t+\tau) - \mathbf{r}(t)|$, where $\mathbf{r}(t)$ refers to the distance of the particle's center of mass to the origin of time t, in both the x ($\parallel$ **n**) and y ($\perp$ **n**) directions. The histograms are well fitted by a Gaussian distribution the width of which, Δ, is directly related to D through $\Delta^2_{\parallel/\perp} = 4D_{\parallel/\perp}\tau$. We see that $\Delta_\parallel > \Delta_\perp$, which implies that $D_\parallel > D_\perp$. It is therefore easier for the quadrupoles to diffuse along the director than perpendicular to it. From the values of $\Delta_{\parallel/\perp}$, we compute $D_\parallel$, $D_\perp$, and the anisotropy ratio $D_\parallel/D_\perp$ (Table 1). These results are in fair agreement with recent Stokes drag calculations that take into account the distortions of the director field (*2*, *3*).

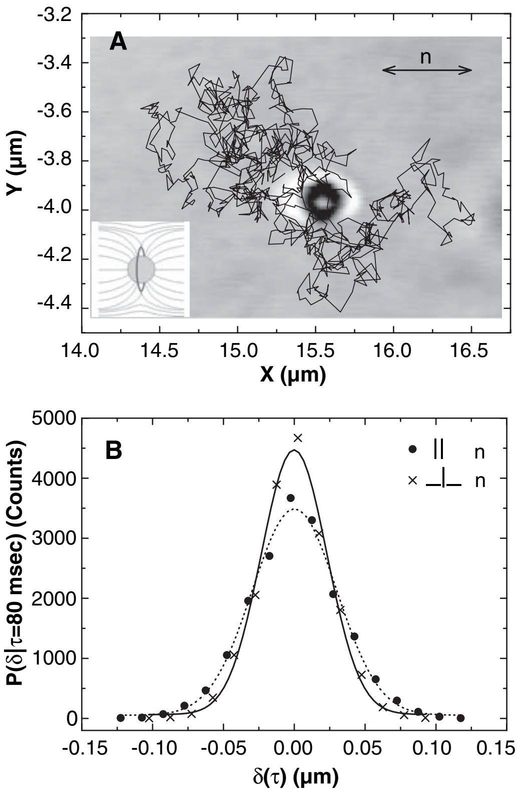

Fig. 1. (**A**) Brownian trajectory of a 1-μm-diameter droplet embedded in a nematic continuous phase. The time τ between two steps was 0.125 s. Background picture: The director around the drop exhibits the Saturn ring configuration (inset), and the black double arrow indicates the director alignment far from the particle. The particle size and the graph axes do not have the same scale. (**B**) Histograms of particle displacements along ($\parallel$) and perpendicular ($\perp$) to the director for τ = 0.08 s from a sample of 18,000 trajectory steps. The solid and dotted lines are Gaussian fits.

Table 1. Diffusion coefficients and anisotropic ratios derived from experiments and simulations [see (*3*), values of liquid crystal 5CB] for a Saturn ring droplet of radius R = 0.55 μm.

Diffusion coefficients	Experiments	Simulations
$D_\parallel$ (10^{-3} μm²/s)	7.7	7.9
$D_\perp$ (10^{-3} μm²/s)	4.8	4.6
$D_\parallel/D_\perp$	1.6	1.72

References and Notes

1. J. Happel, H. Brenner, *Low Reynolds Number Hydrodynamics* (Klüwer, Dordrecht, Netherlands, 1991).
2. R. W. Ruhwandl, E. M. Terentjev, *Phys. Rev. E* **54**, 5204 (1996).
3. H. Stark, D. Ventzki, *Phys. Rev. E* **64**, 031711 (2001).
4. J. C. Crocker, D. G. Grier, *J. Coll. Interf. Sci.* **179**, 298 (1996).
5. P. M. Chaikin, T. C. Lubensky, *Principles of Condensed Matter Physics* (Cambridge Univ. Press, Cambridge, 1995).
6. P. Poulin, H. Stark, T. C. Lubensky, D. A. Weitz, *Science* **275**, 1770 (1997).
7. H. Stark, *Phys. Rep.* **351**, 387 (2001).
8. J. C. Loudet, P. Barois, P. Poulin, *Nature* **407**, 611 (2000).
9. J. C. Loudet, P. Poulin, *Colloid Chemistry I* (Topics in Current Chemistry 226, Springer, Berlin, 2003).
10. Supported by the CNRS, Aquitaine Region, and Bordeaux University.

16 July 2004; accepted 24 September 2004

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Science, **306** (5701), .
DOI: 10.1126/science.1102864

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