Supporting Online Material for

**Brownian Motion of Stiff Filaments in a Crowded Environment**

Nikta Fakhri, Frederick C. MacKintosh, Brahim Lounis, Laurent Cognet, Matteo Pasquali*

*To whom correspondence should be addressed. E-mail: mp@rice.edu

DOI: 10.1126/science.1197321

**This PDF file includes:**

- Materials and Methods
- Fig. S1
- Table S1
- References

**Other Supporting Online Material for this manuscript includes the following:**
(available at www.sciencemag.org/cgi/content/full/330/6012/1804/DC1)

- Movie S1
Brownian motion of stiff filaments in a crowded environment

Nikta Fakhri¹, Frederick C. MacKintosh², Brahim Lounis³, Laurent Cognet³, Matteo Pasquali¹*  
¹Department of Chemical and Biomolecular Engineering, Department of Chemistry, The Smalley Institute for Nanoscale Science & Technology, Rice University, Houston, Texas 77005, USA  
²Department of Physics and Astronomy, Vrije Universiteit, 1081 HV Amsterdam, The Netherlands  
³Centre de Physique Moléculaire Optique et Hertzienne, Université de Bordeaux and CNRS, Talence F-33405, France  
*E-mail: mp@rice.edu

Materials and methods:  

Sample preparation:  
1. SWNT dispersion: SWNTs with diameter between 0.7 and 1.2 nm were obtained from Rice University HiPco reactor (batches 162.8 and 189.2 without purification). Dilute (~1ppm) aqueous suspensions of SWNTs in 1 wt% sodium deoxycholate surfactant (NaDOC, Sigma) were prepared by brief tip ultrasonication (7 W, 10 s, Microson, Misonix) to minimize SWNTs breaking (S1, S2).  
2. Agarose gel: Purified agarose (Multi Purpose, Roche) was used without further purification. Agarose gels were prepared by adding 1 mL of NaDOC surfactant solution (1 wt %, pH = 7.0) to 0.05 g agarose powder (5 wt % agarose gel concentration) in a sealed glass vial; the vial was then heated to about 90°C in a water bath until complete dissolution of the polymer. Solutions were kept sealed at 80°C at above gelation point for further steps. If necessary, drops of hot water were added to keep the polymer fraction constant.  
3. We determined the scaling of agarose pore size (ξ) with agarose concentration (c) through bulk rheological measurements. The elastic (G') and viscous (G'') moduli of the agarose gels were measured by a strain-controlled shear rheometer in parallel plates
configuration (ARES, Rheometrics Scientific, now TA Instruments). The plateau elastic modulus $G_e$ was obtained from dynamic data ($G'$ vs. frequency). For physical gels, rubber elasticity theory (S3) predicts that $G_e$ is proportional to $c^2$. De Gennes (S4, S5) has predicted a limiting behavior $G_e \propto c^{2.25}$ by noting the parallel between the osmotic pressure of semi-dilute polymer solutions and the modulus of gels. Figure S1 shows the data points from bulk measurements and the line is de Gennes’ predicted scaling for $G_e$ and concentration. From the relationship between pore size and elastic modulus given by rubber elasticity theory (S3, S6), $\xi \propto (G_e)^{1/3}$, we determined the pore size dependence for different concentration of the gels as $\xi \propto c^{-0.74}$, in agreement with the value predicted by de Gennes (S7) for a network of flexible chains from scaling arguments.

4. SWNTs in agarose gel: In a typical experiment (S8), a few μL of SWNT/NaDOC dispersion were added to few 100 μL hot clear solution of agarose and were mixed together using vortex mixer to get the final agarose concentration of 0.5-2.5 wt%. It is important to note that SWNTs stabilized by NaDOC do not interact with agarose gel (S9).

5. Slide preparation: 5-10 μL of solution was sandwiched between a heated #1.5 coverslip and microscope slide. The cell was sealed using vacuum grease and cooled to room temperature (which is below the gelling temperature of 37°C) and thus triggering the gelation of the agarose.

**Agarose pore size distribution:**

In agarose, large fibrous polymers form heterogeneous elastic structures that support macroscopic stress but allow the motion of particles in confined pores. The polydispersity in pore size has been estimated to be of order 20% (S10). The theories of Odijk and Doi assume a distribution of pore sizes with a characteristic pore size, $\xi$. A SWNT explores different static microenvironments over the course of the measurement, resulting in a time-averaged characteristic pore size that is not a representation of any instantaneous microenvironments. Because we follow each SWNT for $\tau_c \gg \tau_d$, each SWNT explores many pores (of the order of $\frac{\tau_c}{\tau_d} \propto \left( \frac{L}{\lambda} \right)^2 \frac{L_p}{L} \geq 25$) and each SWNT samples a reasonable pore distribution.

**Experimental setup:**
Nanotubes were excited by either a frequency doubled YAG laser, a tunable dye laser (rhodamine) or a tunable Ti:Sa laser depending on the chirality of the SWNTs to be resonantly excited at their second order transition \((S11, S12)\). The beams were focused into the back aperture of a high NA objective (100x or 60x, Numerical Aperture 1.4), with excitation intensities between 0.1-10 kW/cm\(^2\) of circularly polarized light. The fluorescence was collected with the same objective and imaged on a low noise Si-CCD camera (Micromax, Roper Scientific) or NIR InGaAs camera (Xenics) depending on the chirality of the nanotubes under study, to produce wide-field images of individual nanotubes. The emission spectrum of SWNTs was collected by a cryogenically cooled 1D InGaAs detector (OMA V, Roper Scientific) placed at the output of a spectrometer; At these irradiation levels, there were no noticeable effects on the SWNT dynamics \((S13)\). Images of SWNT dynamics were recorded at 30 frames per second.

**Image analysis:**

From the image analysis we obtained data sets consisting of SWNTs center of mass position in the lab frame and its orientation angle relative to the x-axis.

The overall shape of the SWNT was analyzed by enveloping its trace by a best-fit ellipse that encompassed the shape of the SWNT and computed the lengths of the major and minor axes as well as the orientation of the resulting ellipse, \(\theta_i\), where \(i\) represents frame number spaced by 30 ms acquisition time.

The center of mass position of SWNT in each frame is determined by weighting each pixel by its absolute gray level intensity, \(r_i = [x_i, y_i]\). In this method the brighter pixels indicate a greater amount of SWNTs and give weight to where SWNT mass exist. This method could introduce error for the case of highly curved backbones because the centroid can be located off the filament and any off-filament centroid would introduce error in mean square displacement data analysis in which the assumption is that the centroid is a point on the SWNT. However, because nanotubes are stiff and \(L < L_p\) and relatively short, measuring the centroid through this method is sufficient and we verified the accuracy of the method for our images \((S14)\). For experimental conditions where filaments are long, an alternate method should be employed where the filament center rather than its centroid is chosen.

The backbone of the SWNTs and their contour length were extracted using previously described method \((S1)\).
The displacements of the center of mass (MSD) or angle (MSAD) can be specified by a time averaged autocorrelation function given by ($X$ is either angle (MSAD) or Cartesian coordinates (MSD)):

$$MSD(MSAD) = \langle \Delta X^2(\tau) \rangle = \left\langle (X(t + \tau) - X(t))^2 \right\rangle$$

where $\tau$ corresponds to various lag times.

For the case of translational motion, the center of mass displacements were decomposed into its components parallel ($\Delta n^2$) and perpendicular ($\Delta s^2$) to the running time-averaged reptation tube,

$$\Theta_\tau = \left\langle \theta(t) \right\rangle_\tau = \frac{1}{\tau} \int_0^\tau \theta(t') dt'$$

and applying the rotation matrix $R_{\Theta_\tau} = \begin{pmatrix} \cos \Theta_\tau & \sin \Theta_\tau \\ -\sin \Theta_\tau & \cos \Theta_\tau \end{pmatrix}$

to the Cartesian displacements,

$$\begin{pmatrix} \Delta s(t, \tau) \\ \Delta n(t, \tau) \end{pmatrix} = R_{\Theta_\tau} \begin{pmatrix} \Delta x(t, \tau) \\ \Delta y(t, \tau) \end{pmatrix}$$

and computing the time-average,

$$\begin{pmatrix} \Delta s^2(\tau) \\ \Delta n^2(\tau) \end{pmatrix} = \frac{1}{\tau} \int_0^\tau dt \begin{pmatrix} \Delta s^2(t, \tau) \\ \Delta n^2(t, \tau) \end{pmatrix}.$$
Figures:

Figure S1: Dependence of plateau elastic modulus on agarose concentration. The solid line denotes de Gennes’ predicted scaling behavior.
### Table S1: Chirality of the SWNTs imaged in this study and used in Figure 2.

<table>
<thead>
<tr>
<th>Agarose concentration (w/w)</th>
<th>0.5%</th>
<th>1.0%</th>
<th>1.5%</th>
<th>2.0%</th>
<th>2.5%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chirality</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(6, 5)</td>
<td>[ ]</td>
<td>[ ]</td>
<td>[ ]</td>
<td>[ ]</td>
<td>[ ]</td>
</tr>
<tr>
<td>(9, 4)</td>
<td>[ ]</td>
<td>[ ]</td>
<td>[ ]</td>
<td>[ ]</td>
<td>[ ]</td>
</tr>
<tr>
<td>(9, 7)</td>
<td>[ ]</td>
<td>[ ]</td>
<td>[ ]</td>
<td>[ ]</td>
<td>[ ]</td>
</tr>
<tr>
<td>(8, 6)</td>
<td>[ ]</td>
<td>[ ]</td>
<td>[ ]</td>
<td>[ ]</td>
<td>[ ]</td>
</tr>
<tr>
<td>(7, 6)</td>
<td>[ ]</td>
<td>[ ]</td>
<td>[ ]</td>
<td>[ ]</td>
<td>[ ]</td>
</tr>
<tr>
<td>(10, 5)</td>
<td>[ ]</td>
<td>[ ]</td>
<td>[ ]</td>
<td>[ ]</td>
<td>[ ]</td>
</tr>
</tbody>
</table>
References:


