Supplemental Material for "Accounting for inertia effects to access the high-frequency microrheology of viscoelastic fluids."

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Microrheology with $a = 1.47\,\mu$m beads.

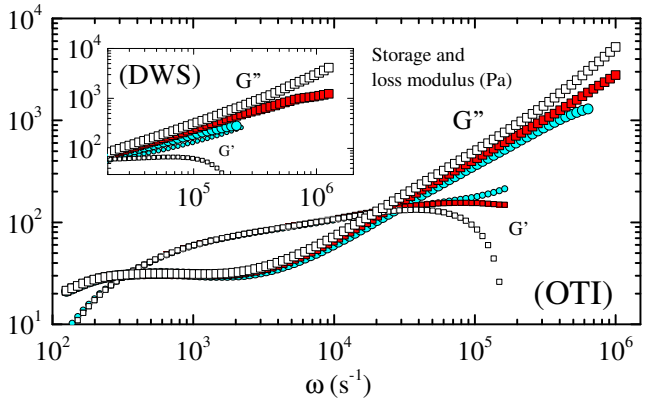

FIG. 1. Comparison of the microrheology results using OTI and DWS (Inset) for one bead size $a = 1.47\,\mu$m in a viscoelastic micelle solution with and without taking into account the effects of inertia. Data without inertia correction ( ), corrected using Indei-Schieber methodology ( ) and using the corrected-MSDs shown in Fig. 1 of the main text ( ).

Fig. 1 shows the results of $G^*(\omega)$ for the micellar solution when using beads of size $a = 1.47\ \mu$m for DWS and OTI. The results are very similar to Fig. 2 of the main text for $a = 0.94\ \mu$m. The discrepancies between DWS and OTI arise because of the differences in the MSDs, which are related with the interaction of the bigger beads with the optical trap in the polymer-based solution.

Comparison between Evans' and Mason-Weitz's methodologies.

An alternative methodology to connect the MSDs with the rheological properties of the fluid was reported by Evans, Tassieri *et al* [1], which provides a complex modulus which we call $G^*_{\rm E}$. Using Evans' methodology:

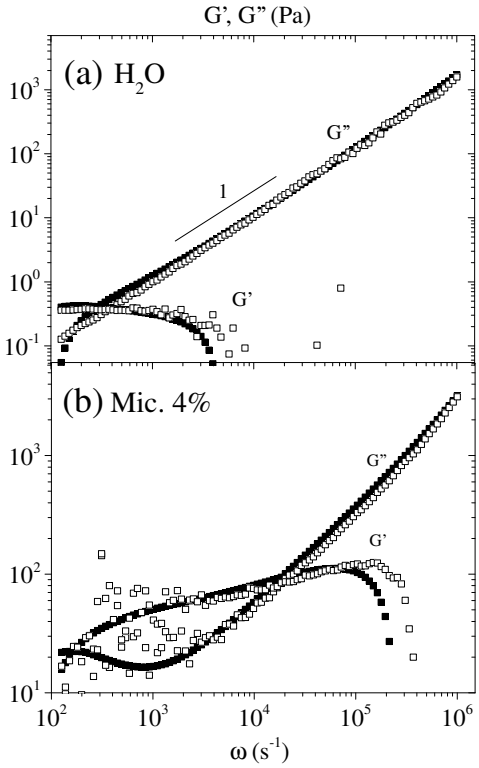

FIG. 2. Mason-Weitz ( ) and Evans' methodologies ( ) for OTI data when using $a = 0.94\ \mu$m beads. (a) Water, (b) Micelles solution at 4%

$$\frac{i\omega}{G^*_{\rm E}(\omega)} = \frac{e^{-i\omega t_N}}{\eta_0} + iC\omega\left\langle\Delta r^2(0)\right\rangle + \\ + C(1-e^{-i\omega t_1})\frac{\left\langle\Delta r^2(1)\right\rangle - \left\langle\Delta r^2(0)\right\rangle}{t_1} + \\ + \sum_{k-2}^{N} C(e^{-i\omega t_{k-1}} - e^{-i\omega t_k}) \times \\ \times \left(\frac{\left\langle\Delta r^2(t_k)\right\rangle - \left\langle\Delta r^2(t_{k-1})\right\rangle}{t_k - t_{k_1}}\right)$$

where $C \equiv \pi a/k_B T$, and η_0 and $\langle \Delta r^2(0) \rangle$ have to be known. Figure 2 shows the results when using Mason-Weitz approach (MW) and Evans' methodology applied to the MSDs obtained through OTI for $a = 0.94$ μm beads. The complex moduli are very similar, with the exception of the viscoelastic fluid at low frequencies, where $G^*_{\rm E}$ does not provide a good curve in comparison with MW. We have only used the MW approach for the results summarized in the main text because both methods are theoretically equivalent and because MW, which is standard in the field of microrheology, provides better results with our experimental data.

$G'(\omega)$ break down for the micelles solution.

The Mason's approximation uses $\alpha(s) \equiv (d \ln \mathrm{MSD}(t)/\ln t)|_{t=1/s}$, to expand the MSD locally around the frequency of interest s. As Mason commented in his work, if $\alpha \sim 1$ over a large temporal range, the estimate for the dominant $G''(\omega)$ will be excellent, but $G'(\omega)$ will degrade. This is the observed behavior in the micelles solution microrheology, where the elastic component shows a bad behavior at high frequencies, specially when it needs to change its curvature quickly. Besides, it is known that the elastic component is very sensitive to artifacts [2, 3], when it is calculated by simply using the Mason-Weitz methodology. However, $G'(\omega)$ works very well for low frequency values: for water experiments the spring constant k value used in the optical trap of OTI experiments can be recovered using the calculated low-frequency value of $G'(\omega)$, which is $G' \sim 0.4$ Pa, as it can be seen in Fig. 2 (a). The spring constant is then $k = 6\pi a G' \cong 7$ μm/N in good agreement with the value obtained from calibration.

The $G' \sim \omega^{3/4}$ behavior at high frequencies is not recovered in our experiments, but theoretically speaking, G' and G'' are not independent quantities through they are related by Kramers-Kronig relationships [4, 5]. To exemplify this point, we can use the approximation made by Booij and Thoone to the generalization of Kramers-Kronig transforms for viscoelastic liquids [6]:

$$G'(\omega) - G'(0) \cong -\frac{\omega\pi}{2}\left(\frac{d[G''(u)/u]}{d\ln u}\right)_{u=w}$$

Using that expression with $G''(u) = k_0 \sin(3\pi/8) u^{3/4} + \eta_s u$, where $k_0 \equiv \frac{1}{15}\rho_m \kappa l_p (2\xi/\kappa)^{3/4}$, and deriving with $x \equiv \ln u$, it is easy to obtain that $(G'(\omega) \gg G'(0))$:

$$G'(\omega) \cong \frac{\pi}{8} k_0 \sin\left(\frac{3\pi}{8}\right) \omega^{3/4}$$

which is almost identical to $G'_{\rm GMK} = k_0 \cos(3\pi/8)\, \omega^{3/4}$ because $\frac{\pi}{8}\sin(3\pi/8) = 0.36$ and $\cos(3\pi/8) = 0.38$.

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