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Supporting Material

Diffusion of the Reaction Boundary of Rapidly Interacting Macromolecules in Sedimentation Velocity

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Supporting Material

The Supporting Material contains a more detailed derivation of Eqs 2, 3 and 4, as well as Figures S1 – S3.

Derivation of Eqs 2, 3, and 4

For the rapidly interacting A + B ←→ AB system considered here, the Lamm equations Eq. 1 are:

$$\frac{\partial c_A}{\partial t} + \frac{1}{r}\frac{\partial}{\partial r}\left[c_A s_A \omega^2 r^2 - D_A \frac{\partial c_A}{\partial r} r\right] = q_A$$

$$\frac{\partial c_B}{\partial t} + \frac{1}{r}\frac{\partial}{\partial r}\left[c_B s_B \omega^2 r^2 - D_B \frac{\partial c_B}{\partial r} r\right] = q_B \qquad \text{(Eq. S1)}$$

$$\frac{\partial c_{AB}}{\partial t} + \frac{1}{r}\frac{\partial}{\partial r}\left[c_{AB} s_{AB} \omega^2 r^2 - D_{AB} \frac{\partial c_{AB}}{\partial r} r\right] = q_{AB}$$

From mass conservation, it follows that $q_A = -q_{AB}$, and adding the first and third equation gives, and the second and third equation, respectively, gives

$$\begin{aligned}\frac{\partial (c_A + c_{AB})}{\partial t} + \frac{1}{r}\frac{\partial}{\partial r}\left[(c_A s_A + c_{AB} s_{AB})\omega^2 r^2 - (D_A \frac{\partial c_A}{\partial r} + D_{AB} \frac{\partial c_{AB}}{\partial r})r\right] = 0 \\ \frac{\partial (c_B + c_{AB})}{\partial t} + \frac{1}{r}\frac{\partial}{\partial r}\left[(c_B s_B + c_{AB} s_{AB})\omega^2 r^2 - (D_B \frac{\partial c_B}{\partial r} + D_{AB} \frac{\partial c_{AB}}{\partial r})r\right] = 0\end{aligned} \qquad \text{(Eq. S2)}$$

Now using the definition of the constituent concentrations $c_{Atot} = c_A + c_{AB}$ we can write for A

$$\frac{\partial c_{Atot}}{\partial t} + \frac{1}{r}\frac{\partial}{\partial r}\left[\frac{(c_A s_A + c_{AB} s_{AB})}{c_{Atot} s_{Atot}} c_{Atot} s_{Atot} \omega^2 r^2 - \frac{(D_A \frac{\partial c_A}{\partial r} + D_{AB} \frac{\partial c_{AB}}{\partial r})}{D_{Atot} \frac{\partial c_{Atot}}{\partial r}} D_{Atot} \frac{\partial c_{Atot}}{\partial r} r\right] = 0 \quad \text{(Eq. S3)}$$

and define the new quantities s_{Atot} and D_{Atot} such that

$$\frac{(c_A s_A + c_{AB} s_{AB})}{c_{Atot} s_{Atot}} = 1 \qquad \text{(Eq. S4)}$$

, i.e.

$$s_{Atot} = \frac{c_A s_A + c_{AB} s_{AB}}{c_{Atot}} = \frac{c_A s_A + K c_A c_B s_{AB}}{c_A + K c_A c_B}, \qquad \text{(Eq. S5)}$$

and

$$\frac{(D_A \frac{\partial c_A}{\partial r} + D_{AB} \frac{\partial c_{AB}}{\partial r})}{D_{Atot} \frac{\partial c_{Atot}}{\partial r}} = 1 \qquad \text{(Eq. S6)}$$

, or

$$D_{Atot} = \frac{D_A \frac{\partial c_A}{\partial r} + D_{AB} \frac{\partial c_{AB}}{\partial r}}{\frac{\partial c_{Atot}}{\partial r}} = \frac{D_A \frac{\partial c_A}{\partial r} + D_{AB} K \left[c_A \frac{\partial c_B}{\partial r} + c_B \frac{\partial c_A}{\partial r} \right]}{\frac{\partial c_A}{\partial r} + K \left[c_A \frac{\partial c_B}{\partial r} + c_B \frac{\partial c_A}{\partial r} \right]} \quad . \qquad \text{(Eq. S7)}$$

With symmetric operations for B follow trivially from here Eq. 1, 2 and 3 in the paper.

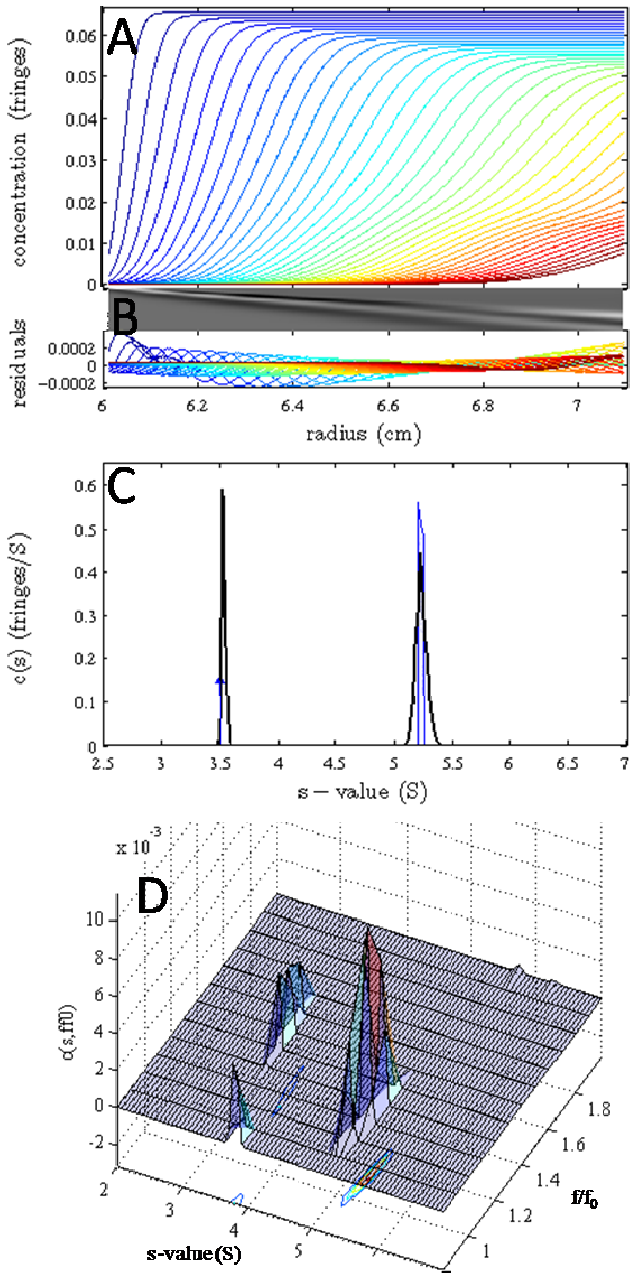

Figure S1

Fitting concentration profiles of interacting systems with distributions of non-interacting species in the case of low polydispersity. Panel A: Lamm PDE solutions for the interacting systems of Figure 2 were calculated at equimolar loading concentrations $c_{Btot} = c_{Atot} = 0.2\ K_D$ (solid lines). A fit of the concentration profiles with *c(s)* (dashed lines) gives an rmsd of 0.0096-fold the loading signal. Panel B: Residuals in overlay and bitmap format (28). Panel C: The resulting sedimentation coefficient distribution *c(s)* (black line), and the asymptotic boundary $d\hat{c}/dv$ (blue patch, scaled) and the undisturbed boundary (blue stem, in units of fringes) as predicted from Gilbert-Jenkins theory. *c(s)* was calculated with maximum entropy regularization on a level of P = 0.95. A two discrete species model assigning a single *s*-value and *D*-value to the reaction boundary leads to an rmsd 0.0104-fold the loading signal (fit not shown). Panel D: Size-and-shape distribution $c(s,f/f_0)$ fit to the same data , producing an rmsd of 0.0006-fold the loading signal. $c(s,f/f_0)$ was calculated with Tikhonov regularization at a level of P = 0.95. The gridlines indicate the discretization of *s*-dimension and the f/f_0-dimension.

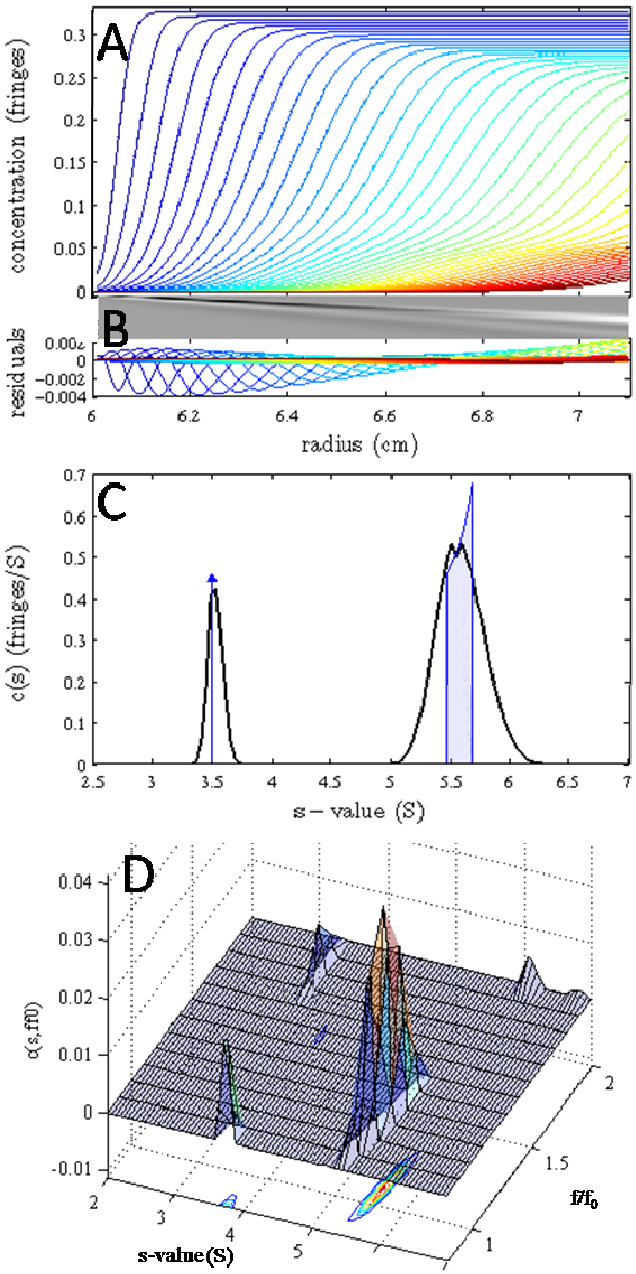

Figure S2

Fitting concentration profiles of interacting systems with distributions of non-interacting species in the case of medium polydispersity. Panel A: Lamm PDE solutions for the interacting systems of Figure 2 were calculated at equimolar loading concentrations $c_{Btot} = c_{Atot} = K_D$ (solid lines), and a fit of the concentration profiles with *c(s)* (dashed lines). Panel B: Residuals, with an rmsd of 0.0017-fold the loading signal. For comparison, a two discrete species model assigning a single *s*-value and *D*-value to the reaction boundary leads to an rmsd 0.0022-fold the loading signal (fit not shown). Panel C: The resulting sedimentation coefficient distribution *c(s)* (black line), $d\hat{c}/dv$ (blue patch, scaled) and the amplitude of the undisturbed boundary (blue stem, in units of fringes). Panel D: Size-and-shape distribution $c(s,f/f_0)$ fit to the same data, leading to an rmsd of 0.0011-fold the loading signal.

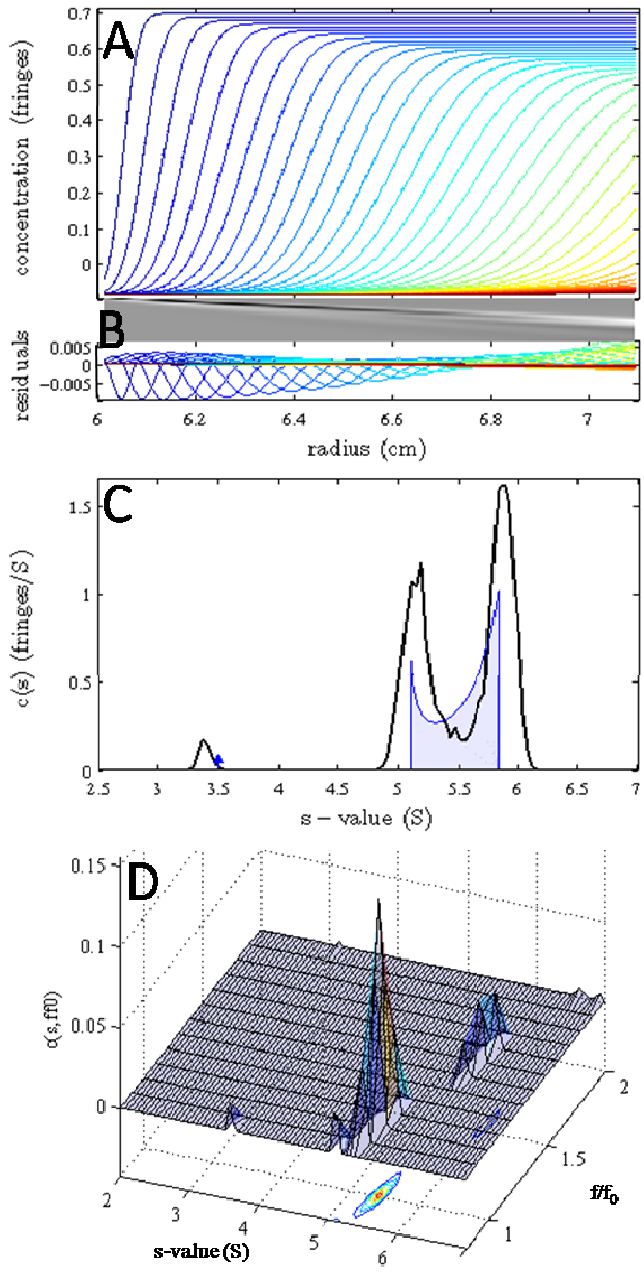

Figure S3

Fitting concentration profiles of interacting systems with distributions of non-interacting species for the case of high polydispersity. Panel A: Lamm PDE simulations as in Figure 6, but for loading concentrations c_{Btot} = 3 K_D and c_{Atot} = 1.5 K_D, which is at the phase transition line by EPT in the region of stronger polydispersity of the reaction boundary (Figure 5). A fit of the concentration profiles with *c(s)* (dashed lines) gives an rmsd of 0.0019-fold the loading signal, with residuals as presented in Panel B. For comparison, a two discrete species model assigning a single *s*-value and *D*-value to the reaction boundary leads to an rmsd 0.0043-fold the loading signal (fit not shown). Panel C: The resulting sedimentation coefficient distribution *c(s)* (black line), $d\hat{c}/dv$ (blue patch, scaled) and the amplitude of the undisturbed boundary (blue stem, in units of fringes). Panel D: The size-and-shape distribution $c(s,f/f_0)$ fit to the same data produces an rmsd of 0.001-fold the loading signal. In *c(s)*, the second peak within the reaction boundary implies apparent *M*-values of 72 kDa, whereas in $c(s,f/f_0)$ the secondary peak at ~ 6 S and f/f_0 implies apparent *M*-values 1.5-fold the complex molar mass.