Comment on “Nanoconfinement-Enhanced Conformational Response of Single DNA Molecules to Changes in Ionic Environment”

In a recent Letter [1], Reisner et al. study the ionic strength dependence of DNA extension in nanochannels. One of the central claims of the work is that the variation of DNA persistence length with ionic strength is alone insufficient to explain the observed variation of DNA extension. To this end, the authors argue that it is necessary to introduce an ionic-strength dependent effective polymer width to account for the effects of increased self-avoidance. This is plausible at high ionic strengths and large channel cross-sections, i.e., when the DNA polymer is expected to exhibit blob-like behavior [2]. However, in the regime of low ionic strength, where the interaction of the DNA backbone with the confining channel walls precludes intersegmental repulsion, the relevance of an effective polymer width is debatable and warrants some scrutiny. In the present Comment, we argue that the data of Reisner et al. at low ionic strengths $I \leq 30$ mM are in fact described very well without the notion of an effective polymer width using the entropic depletion theory due to Odijk [3] with an ionic-strength dependent DNA persistence length.

The conclusion of Reisner et al. that the model based solely on persistence length fails to explain the experimental observations stems from a comparison of the DNA extension data with their Eq. (4) derived in [4]. However, this expression is expected to hold only when the extended length $r$ of DNA confined in a nanochannel approaches its contour length $L$, i.e., $(L-r)/L \ll 1$. It is apparent from Fig. 1 of [1] that this condition is not satisfied for a majority of the data points. Therefore, we compare these data with the expression derived in [3] (incidentally, also cited in [1]), which is free from the above approximation. In this case, the extended length of DNA in a square nanochannel with the cross-section of side $A$ is $r = R_e (\cos \eta) = R_e \left(1 - \langle \eta^2 \rangle / 2\right)$, where $\langle \eta^2 \rangle = 0.340 (A/P)^{2/3}$, $R_e^2 = 2 L g - 2 g^3 (1 - \exp (-L/g))$, $L$ is the polymer contour length, and $g$ is the global persistence length (for details, see [3]). The experimental data on the ionic strength dependence of the DNA persistence length relevant to single-molecule DNA experiments obtained in [7] are described well by an Odijk–Skolnick–Fixman (OSF) type formula [3] [6] (see Fig. 1b). Using this dependence, we compare the theory [3] with the data of [1] and find very good agreement at low ionic strengths (Fig. 1b).

Furthermore, in order to describe their data, the authors of [1] invoke the scaling-based de Gennes theory [2] which, as it turns out, is quite insensitive to particular functional dependences of both the DNA persistence length and width on the ionic strength. In fact, agreement with data for $I > 10^{-2}$ M can be achieved by selecting an appropriate numerical prefactor [1], although it still fails for lower $I$. By contrast, Odijk’s theory [3] is extremely sensitive to the particular functional dependence $P(I)$ and has no tunable parameters. It not only predicts the correct magnitude of the effect, but also describes experimental data well for $I \leq 30$ mM (Fig. 1b).

Note that this theory is not expected to describe the data at higher ionic strengths. Thus, we demonstrate that contrary to the conclusions made by Reisner et al. [1], the ionic strength dependence of the DNA persistence length alone is indeed sufficient to explain the experimental data on DNA extension in nanochannels at low ionic strengths.

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