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Simulation of longitudinal string waves through a single polymer molecule

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HIGHLIGHTS

- Feasibility of single-molecule acoustics was investigated by simulation.
- DNA strand was subjected to longitudinal oscillation at one end.
- Resulting propagating oscillation was shown to be wave-like.
- Proof of concept experiment using optical tweezers setup proposed and simulated.

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ABSTRACT

The capability of an individual polymer molecule to carry an acoustic wave along its length was investigated by a Brownian dynamics simulation using the algorithm of Ermak and McCammon and the discrete worm-like chain model. A 3 μm long DNA strand featuring 50 nm persistence length was subjected to longitudinal oscillations (2 kHz to 25 kHz) at one end, and the properties of the resulting propagating interaction were studied and shown to be wave-like. A proof of concept experiment is proposed, where an optical tweezers dumbbell experiment is conducted to induce and receive the wave, and the result is compared to that of a control experiment with the DNA strand absent. Simulations were conducted to show what one might expect to see in such an experiment.

1. Introduction

To our knowledge, acoustics has never been applied on the scale of a single macromolecule acting as a wave-transmitting medium. Studies have, however, been made of related phenomena, such as diffusive propagation of tension within polymer strands [1] and the appearance of localized disturbances in polymer strands when subjected to rapid external perturbations [2].

Acoustics has a large number of applications, such as structural property characterization [3], structural mapping [4], defect detection [5] and micromanipulation [6]. Possible applications of single-molecule acoustics could be envisioned as nano-scale analogies to macroscopic applications, such as the characterization of quantities like persistence length, detection of irregularities such as breakages and DNA knots, or even the study of nanostructures built by DNA self-assembly (so-called "DNA origami" [7]).

In this work, the feasibility of doing single-molecule acoustics was investigated by simulation. A DNA molecule in water with optically trapped micron-sized beads attached to each end (a dumbbell assay) was simulated. DNA was chosen because of its availability and because its mechanical properties are known [8,9]. The discrete worm-like chain (DWLC) polymer

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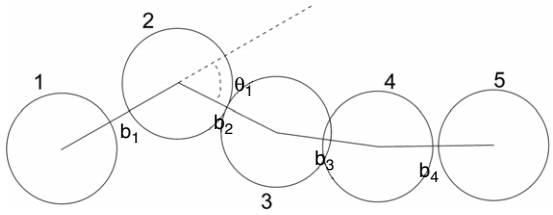

Fig. 1. The discrete worm-like chain model, first five subunits, and the bond vectors b and angles θ. Here b_2 is at equilibrium length, b_3 compressed, and b_1 and b_4 extended.

model [10] was used together with the algorithm of Ermak and McCammon [11], developed to simulate Brownian dynamics of assemblies of identical spherical particles immersed in fluid. This method was applied to polymers by Allison [12] and has since been used in many different studies [13–16]. The model and its implementation is described in Section 2.

An "ideal" simulation unconstrained by the experimental limitations of the setup was conducted to show that the structure of DNA can transmit a propagating wave-like interaction, and to determine the properties of the interaction. In this simulation, thermal motion of the end beads and hydrodynamic coupling between them was ignored. An oscillation was induced into one of the end-beads, inducing a wave that propagated along the DNA strand, which then was picked up by the other bead. The time-series of the positions of both beads, as well as every polymer subunit, were recorded. The obtained results are discussed in Section 3.1.

A DNA strand is too narrow to be observed by optical microscopy. Fluorescence can be used to infer the approximate position of the strand, but it is not feasible to expect it to be able to reveal details such as the displacement of an individual, oscillating polymer subunit. Hence, in a validating experiment, the presence of a wave must be deduced from the time-series of the positions of the end-beads. This raises an issue when interpreting experimental data: how does one know that the oscillation picked up by the receiving bead is due to a mechanical wave transmitted by the strand, rather than due to some other interaction? Two alternative hypotheses should be examined: I. The received oscillation is due to the hydrodynamic coupling between the sending and the receiving bead, with the strand having no effect. II. The strand acts as an elastic rod with no internal parts and effectively instantaneous relaxation, a spring that applies forces of equal magnitude and opposite direction to both beads. I. can directly be tested by making two experiments where, in one experiment, the entire setup with beads and a DNA strand in between is used whereas in the other one, the strand is removed while the beads are retained, and everything else is kept equal. Then, any observed difference can be attributed to the presence of the DNA strand. II. is trickier since no corresponding paired control experiment can be conducted. Based on the above arguments, a simulation is valuable for suggesting validation experiments and for interpreting data collected during such experiments.

Simulations were carried out, including the aforementioned experimental limitations of Brownian motion and hydrodynamic interaction, to show what one might expect to see in such an experiment. The simulation using the *DWLC* model was compared to models corresponding to hypotheses I and II: The *beads-only* model without DNA where hydrodynamic coupling was the only mechanism driving the receiving bead, and the *elastic rod* model where the DNA was modelled as a spring following the equilibrium force–extension relationship of a worm-like chain. The effects of excitation frequency and inter-bead distance on the amplitude ratio and phase difference between the sending and receiving beads were mapped. The results are presented and discussed in Section 3.2.

2. Theory and methodology

2.1. Theory

The DWLC model, Fig. 1, models a polymer as a chain of N spherical subunits linked by virtual bonds. These bonds can either be inflexible or flexible. Inflexible bonds correspond to the standard WLC model, for which the contour length of the chain is constant and stretching the chain merely straightens bends in the chain. Collisions with the molecules of the ambient fluid cause the subunits to undergo thermal motion and the chain to bend, counteracting the straightening effect of a stretching force. The relationship between the force with which a chain is stretched and the distance between its endpoints is referred to as its force–extension relationship. For the standard WLC model, the following approximate force–extension relationship, derived by Marko and Siggia, applies [17]

$$F = \frac{k_B T}{P}\left(\frac{1}{4\left(1-\frac{x}{L_0}\right)^2} - \frac{1}{4} + \frac{x}{L_0}\right) \tag{1}$$

where F is the magnitude of the extending force, x is the end-to-end distance of the chain, L_0 is the contour length of the chain, P is the persistence length of the chain, k_B is Boltzmann's constant, and T is the absolute temperature. We note that x/L_0 asymptotically approaches unity when F goes to infinity. Stretching experiments such as those by Wang et al. [18] have shown DNA to be slightly elastically extensible and to account for this, they introduced the modified Marko–Siggia