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Single-Molecule Studies of DNA Dynamics and Intermolecular Forces

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Typically, polymeric fluids are experimentally investigated by examining the bulk properties of the fluid, so the individual molecular dynamics can only be inferred from the fluid properties by using theoretical predictions that relate the two. However, DNA has been shown to be a model system for probing fundamental questions in polymer science, and the recent development of the single-molecule approach using DNA has allowed for direct probing of the molecular conformations and dynamics of polymers. Here, DNA molecules were used to investigate how molecular length, topology and concentration influence the dynamical properties of polymers. Fluorescence microscopy and single-molecule tracking were used to determine self-diffusion coefficients of DNA molecules, and a new experimental approach was developed, using optical tweezers, to measure the intermolecular forces confining entangled DNA molecules. Scaling of diffusion with concentration was determined for the four possible topological combinations of linear and circular molecules. At higher concentrations topology had a dramatic effect on the diffusion, and scaling was in agreement with the reptation model, predicted to describe the dynamics of entangled polymers. The notable exception was the strongly hindered diffusion of a circular molecule in an entangled solution of linear DNA. Using the new optical tweezers method, a tube-like field confining a single entangled molecule was measured, in accord with the key assumption of the reptation model. A time-dependent harmonic potential opposed displacement transverse to the molecular contour, and the force relaxations following displacement were composed of three distinct modes. A characteristic tube radius for the entangled solution was also determined, close to the classically predicted value. The dependence of the above findings on molecular topology and concentration was also investigated.