Entangled polymer dynamics beyond reptation

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Many theoretical and experimental studies have aimed at understanding polymer dynamics at the molecular level that give rise to its bulk phase properties. While much progress has been made in the field over the past ~60 years, many aspects of polymers are still not well understood, especially in complicated systems such as entangled fluids and polymers. Despite the importance of molecular level polymer dynamics in both materials and life science researches, a highly complex nature of the crowded polymers makes it particularly difficult to understand their dynamics at the molecular level using ensemble-averaged experimental methods. While there are many experimental studies aimed to explain the conformational dynamics of a single DNA molecule in crowded environment, the characterization of its motion and dynamics in an equilibrium condition has been challenging. We have been developing a new single-molecule tracking techniques to address these issues [1-3].

Here, we report 3D super-resolution fluorescence microscopy of linear (λDNA, 48.5 kbp) and cyclic (Charomid DNA, 42 kbp) that directly visualizes nanoscale conformational structure of entangled DNA chain in equilibrium state [4]. Using the astigmatism-based 3D localization microscopy, we measured the super-resolution images of Cy5 fluorescently labeled DNA. The spatial resolution of the experiment on a surface-deposited molecule and a molecule in the solution was 14 nm and 30 nm, respectively. Since the persistence length of DNA is approximately 50 nm (i.e., the molecule can be treated as a rigid rod in the length scale of the persistence length), we were able to capture the actual conformational state of the chains and their dynamics in our experiment. The motion and conformational state of the fluorescently labeled DNA under entangled conditions were captured by recording time-lapse super-resolution images in a concentrated solution of unlabeled DNAs. The results showed that our approach enabled to directly visualize both overall motion of the molecule and local motion along the chain at nanoscale under equilibrium conditions. The results also demonstrated that the motion of the linear λDNA, a model system for semi-flexible polymers, is described by the standard reptation theory, whereas the motion of the cyclic Charomid DNA is clearly distinct from that of the linear DNA [4].

References:

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