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Entropy and barrier-controlled fluctuations determine conformational viscoelasticity of single biomolecules

Bhavin Khatri, Masaru Kawakami, Katherine Byrne, D. Alastair Smith, and Tom C..B.McLeish

Polymer and Complex Fluids Group, School of Physics and Astronomy, University of Leeds, United Kingdom *Present Address: Soft Condensed Matter Group, Department of Physics, University of Surrey, Guildford, United Kingdom

Abstract: Biological macromolecules have complex and nontrivial energy landscapes, endowing them with a unique conformational adaptability and diversity in function. Hence, understanding the processes of elasticity and dissipation at the nanoscale is important to molecular biology and emerging fields such as nanotechnology. Here we analyze single molecule fluctuations in an atomic force microscope, using a generic model of biopolymer viscoelasticity that includes local “internal” conformational dissipation. Comparing two bipoymers, dextran and cellulose (polysaccharides with and without local bistable transitions), demonstrates that signatures of simple conformational change are minima in both the elastic and internal friction constants around a characteristic force. A novel analysis of dynamics on a bistable energy landscape provides a simple explanation: an elasticity driven by the entropy, and friction by a barrier-controlled hopping time of populations between states, which is surprisingly distinct to the well-known relaxation time. This nonequilibrium microscopic analysis thus provides a means of quantifying new dynamical features of the energy landscape of the glucopyranose ring, revealing an unexpected underlying roughness and information on the shape of the barrier of the chair-boat transition in dextran. The results presented herein provide a basis toward probing the viscoelasticity of macromolecular conformational transitions on more complex energy.



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