

AFM-based single molecule force spectroscopy of complementary quadruple hydrogen-bonded systems

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Self-assembly is one of the most intriguing phenomena in the world of chemistry, physics and especially biology. The 3D conformation of DNA and a wide variety of proteins are only a few of the fascinating examples of nature's ability for self-assembly, which provides the human body for instance with a swift response to small environmental changes and fast enzymatic activity at body temperature.

Self-assembly is controlled by intermolecular interactions^[1] which determine the behavior and properties of bulk materials. Although the last few decades have provided significant insights into the understanding of these bulk properties, knowledge on a single molecule level is often lacking. Current developments in nanotechnology and biology require fundamental understanding of single molecule behavior. This becomes crucial in cases where bottom-up approaches are necessary for the development of, for instance, nano-scale devices or single molecule motors.

To understand the fundamentals of intermolecular forces of hydrogen-bonded systems, in-situ Atomic Force Microscopy-based single molecule force spectroscopy was used^[2] in this work to investigate the formation and rupture of quadruple hydrogen-bonds. A comparative study between different quadruple hydrogen-bond moieties in solution was performed to provide a better understanding of structure-property relationships like the influence of configuration and conformation on polymer (bond) strength^[3]. This approach not only provides statistic data on a single molecule level, but can also be used to determine, understand and predict resulting bulk properties. The research performed here clearly supports the notion that theoretical calculations based solely on hydrogen-bond interactions itself can give a substantial underestimate of the actual bond strength due to additional effects like for instance intermolecular stabilization of the hydrogen bond moiety.

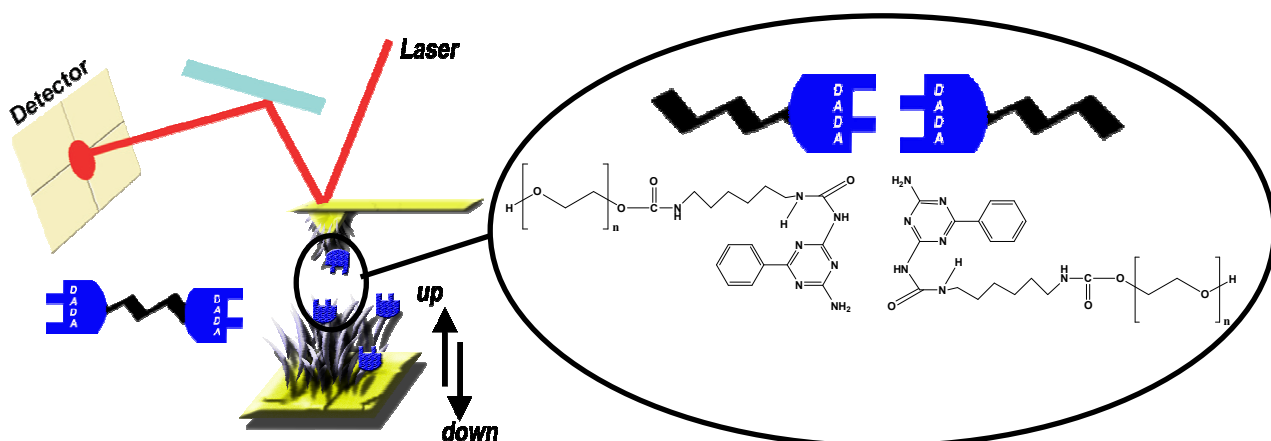


Figure 1. Schematic representation of Atomic Force Microscopy-based Single Molecule Force Spectroscopy of a supramolecular DADA hydrogen-bond array. Either supramolecular dimers or supramolecular polymers can be probed in situ by adding a bifunctional PEG-linker. Bond strength of these hydrogen bonds can be determined by varying the loading rate during the experiment.

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