Abstract—The power spectrum of the hydrogen-bond energy fluctuation of DNA is investigated by molecular dynamics simulation [1,2]. A $1/f$ frequency response is found, even for simple configurations of DNA composed only with thymine bases and with the equal presence of complementary strands. This suggests that various relaxation processes are involved as a general trend. The noise amplitude strongly depends on the temperature. These results suggest that experimental measurements of the hydrogen-bond energy noise may provide additional degrees of freedom for a wide range of studies from DNA nanotechnology to biosensors.

Keywords—DNA, hydrogen-bond energy, $1/f$ noise, biosensors, DNA nanotechnology

Hydrogen-bond (hb) interactions are ubiquitous in nature, and their dynamics is expected to play a key role in chemical reactions and biomolecular recognition [3,4]. Recent experimental setups allowing the direct observation of single-molecule hydrogen-bond dynamics with single-molecule resolution for solvent/surface interaction open new opportunities for the study of hb dynamics [4]. It was previously shown that liquid water yields $1/f$ noise in hb energy, whereas simple liquids such as liquid argon exhibit a near white spectrum [5]. This observation was related to network rearrangement dynamics. In DNA systems, hb interactions are typically studied from temperature ramps to assess a melting temperature $T_m$ and measured by fluorescence. The noise in these experimental systems is typically too high to directly assess hb energy fluctuations; however, the utilization of fluorescence correlation microscopy at single-molecule level is very promising [6] for such studies. With the recent progress in molecular dynamics (MD) simulations that nicely reproduce experimental results, in particular for DNA [2], we investigate by MD (Fig.1a) the dynamics of hb energy, and the potential interest of using DNA hb fluctuations as an additional signal for studies in DNA nanotechnology and biosensors.

We first confirm that hb energy follows a sigmoid dependence ($T_w$ = 55°C; Fig.1b). Interestingly, the power spectrum of the hb energy fluctuation $S_{hb}$ exhibits a $1/f$ frequency response (Fig.1c), even for the single-molecule configuration. We attribute these results to multiple relaxation processes induced by the fluctuation of π-π molecular interactions [7] as well as possible partial hybridization and a statistical distribution of these relaxation processes in the case of multiple molecules. The temperature dependence of the noise (Fig.1d) allows obtaining additional information to the sigmoid response (Fig.1b). We suggest that the investigation of DNA sequence, length, concentration and ionic strength may provide useful data for future experiments. An analytical model for hb noise is under development.

References