

## Conformational Memory of Single Photosynthetic Pigment-Protein Complexes.

### A Precursor of Non-Photochemical Quenching?

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Proteins are supramolecular machines that carry out a wide range of different functions many of which require flexibility. The current picture is that proteins can assume many different conformations or conformational substates in order to fulfill their tasks. Generally these structural fluctuations can be made visible by optical spectroscopy of chromophores that are embedded in the protein matrix. Since the energies of the electronic energy levels of a chromophore are very sensitive to its interactions with the local surrounding, conformational fluctuations of the protein lead to changes in the chromophore-protein interactions that show up as spectral fluctuations (spectral diffusion) of the probe molecule. However, as a consequence of the conformational heterogeneity, protein ensembles exist in a broad variety of structures, which manifests itself as a dramatic increase in dynamic heterogeneity reflecting the distribution of the associated barriers that separate the various structures. In order to elucidate information that is commonly washed out by ensemble averaging single-molecule techniques have been exploited. This allows these dynamic processes to be observed that are usually obscured by the lack of synchronization within an ensemble, because a single protein that undergoes conformational fluctuations is at any time in a distinct, well-defined substate.

In order to learn more about the conformational fluctuations of a protein we exploit the phenomenon of fluorescence intermittency, also termed blinking. Up until now spontaneous conformational fluctuations of proteins have always been assumed to reflect a stochastic random process. The present single-molecule study shows a system where a protein, the LH2 complex from a purple photosynthetic bacterium, displays clear conformational memory. We argue that such a behaviour is exactly the process that can facilitate the evolution of control of switching between two conformational states that can then be used to regulate protein function. If changing between different conformational states was random than it would be much more difficult to understand how conformational control of protein function could have evolved. For the LH2 complexes the conformational memory behaviour seen could provide a pathway by which non-photochemical quenching evolved.

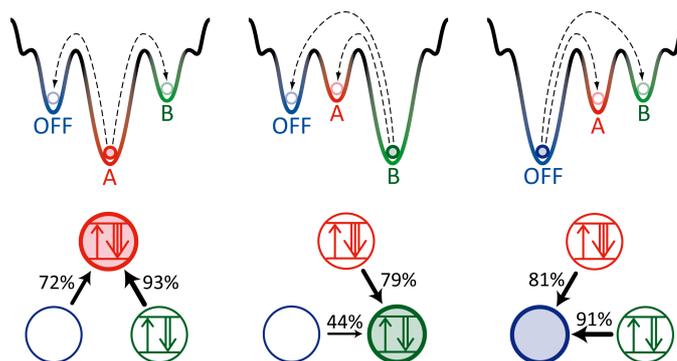


Fig1. Schematic sketch of the observed memory in the intensity fluctuations of LH2. At the top a schematic energy landscape with minima at A, B, and OFF, respectively, is displayed. The minima are associated with the initial intensity levels of the emitted fluorescence of the protein. The numbers next to the arrows at the bottom correspond to the probability to return within two consecutive intensity jumps back to the initial state. The probability for a stochastically independent random event amounts to 50% and is observed only for the transition from OFF  $\rightarrow$  B if B is the initial state (bottom centre), whereas all other probabilities exceed this value clearly.