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Turn over in enzyme catalysis using large deviation theory

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Enzymatic processes[1] act like machines which catalyse biochemical reactions with high dynamic variability often passing through large conformational fluctuations, which decides the catalytic turnover. In various single molecule events, techniques are developed to understand the fluctuating trajectories to obtain their kinetic pathways[2]. Recently introduced nonequilibrium thermodynamics is extensively studied to learn how the chemical force in enzymatic processes can be utilized for mechanical work under the heading of molecular motor, for example, many theoretical efforts have been made on single molecule pulling experiments in this light[3]. However, from the viewpoint of nonequilibrium thermodynamics, the effect of external force on the chemical reaction, specially on the kinetic efficiency[4] and time dependent energetics[5], are not well understood and that too for large deviation[6,7,8] in the chemical process is not available. Here we have considered nonequilibrium turn over statistics of enzyme catalysis in a chemiostatic flow with externally controllable parameters, like substrate injection rate and mechanical force. Large deviation theory gives the thermodynamic force of nonequilibrium steady state, as is usually recorded experimentally by single molecule technique, which plays a key role responsible for the dynamical symmetry of the scaled cumulant generating function and large deviation rate function. Using some special properties of Legendre transformation here we have provided the relation involving the deviations of fluxes and dissipation rates which can be utilized to understand the role of chemical potential at nonequilibrium steady state to control the Fano factor.

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Page 50