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Inhomogeneous Single-Molecule Interfacial Electron Transfer Dynamics: A Single-Molecule Approach[1] V. GOVIND RAO, B. DHITAL, H. P. LU, Bowling Green State University — Interfacial electron transfer (ET) plays a critical role in energy science, photocatalysis, and surface chemistry. The inherent complexity of the interfacial ET dynamics is often difficult to resolve by conventional ensemble-averaged spectroscopic or electrochemical measurements alone. Single-molecule spectroscopy provides insightful details about interfacial ET dynamics which are beyond the realms of conventional ensemble-averaged analyses; particularly with regard to complex mechanism and spatiotemporal heterogeneity. Combination of single-molecule fluorescence spectroscopy approach with various other techniques such as atomic force microscopy, electrochemistry, and Raman spectroscopy can further facilitate inspection of multiple-parameters with high chemical selectivity and wide temporal and spatial resolutions. We have used photonstamping spectroscopy correlated with electrochemical techniques to dissect complex interfacial ET dynamics by probing individual Zn-porphyrin molecule anchored to TiO_2 NP surface in aqueous electrolyte solution while electrochemically controlling the energy states of TiO_2 NPs. Our work advanced a critical understanding of the interfacial ET dynamics by revealing the intermittency of ET dynamics.

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