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Single-Molecule Interfacial Electron Transfer Dynamics at Dye-Sensitized TiO₂ Nanoparticles H. PETER LU, YUANMIN WANG, YUFAN HE, Bowling Green State University, Department of Chemistry, Center for Photochemical Sciences, Bowling Green, OH 43403 — Interfacial electron transfer dynamics is important for environmental and catalytic reactions. Extensive ensemble-averaged studies have indicated inhomogeneous and complex dynamics of interfacial ET reaction. To characterize the inhomogeneity and the complex mechanism, we have applied single-molecule spectroscopy and correlated AFM/STM imaging to study the Interfacial ET dynamics of dye molecules adsorbed at the surface of TiO₂ nanoparticles. The interfacial ET activity of individual dye molecules showed fluctuations and intermittency at time scale of milliseconds to seconds. The fluctuation dynamics were found to be inhomogeneous from molecule to molecule and from time to time, showing significant static and dynamic disorders in the dynamics. Furthermore, we have applied site-specific AFM-Raman spectroscopy on analyzing ET associated mode-specific vibrational reorganization energy barriers. Our experiments revealed site-to-site variations in the vibrational reorganization energy barriers in the interfacial ET systems. Our recent experiments on single-molecule metal-to-ligand electron transfer (3) and single-molecule STM manipulation will also be discussed.

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