

Intermittent Single-Molecule Interfacial Electron Transfer Dynamics

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We have applied single-molecule spectroscopy to study photosensitized interfacial electron transfer (ET) processes in Coumarin 343 (C343)-TiO₂ nanoparticle (NP) and Cresyl Violet (CV⁺)-TiO₂ NP systems. By probing single-molecule fluorescence fluctuations and systematic control experiments, we observed interfacial ET reaction rate fluctuations, associating redox reactivity intermittency with the fluctuations of molecule-TiO₂ electronic and Franck-Condon coupling. The intermittent interfacial reaction dynamics that likely occur among single molecules in other interfacial and surface chemical processes can typically be observed by single-molecule studies, but not by conventional ensemble-averaged experiments.