Many photochemical transformations involve coupling between nuclear and electronic motions that prompt structural changes in concert with relaxation of excited molecular electronic states. There is continuing excitement for harnessing this nonadiabaticity for light-driven applications, yet efficient control of these photoresponses requires understanding of how the details of molecular structure facilitate desired pathways between multiple potential energy surfaces. This talk will present our experimental efforts to illuminate relative efficiencies of competing deactivation channels of reversible photoresponsive systems and how they depend on details of molecular/material structure and photonic inputs.