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Light, Molecules, Action: Broadband UV-visible transient absorption studies of excited state dynamics in photoactive molecules

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Broadband UV-visible transient absorption spectroscopy provides a powerful tool for the investigation of the dynamics of electronically excited molecules in the condensed phase. It is now possible to obtain transient spectra on a routine basis spanning the range from <300 nm to >800 nm with femtosecond time resolution. We have used this method to study the excited state dynamics and internal conversion of a range of molecular systems with potential application as optically powered molecular devices. The cyclohexadiene ring-opening reaction is the basis of a class of important optical switches and of the biological synthesis of previtamin D3. The ring-opening reaction is ultrafast, occurring on a picosecond to subpicosecond times scale depending on the substituents around the ring. These have a significant influence on the dynamics and electronic structure of the electronically excited molecule. The results of a series of transient absorption studies as a function of chromophore substitution and environment will be presented. The cis-trans isomerization of polyene molecules, especially substituted stilbenes, provides another important class of functional molecular transformations. Again the excited state dynamics can be ultrafast with photochemistry controlled by details of the curve crossings and conical intersections. Finally the photochemistry of the even more complex set of cobalamin chromophores with a photoalabile C-Co bond has been proposed as a tool for spatio-temporal control of molecule delivery including drug delivery. Broadband transient absorption spectroscopy has been used to investigate the ultrafast electronic dynamics of a range of cobalamin compounds with comparison to detailed theoretical calculations. The results of these studies will be presented.