

Evolution of the Triplet Excited State in PtII Perylenediimides

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R sum  en anglais

Here, we present the ultrafast dynamics of a series of metal complexes developed to permit access to the perylenediimide (PDI) triplet manifold that preserves the desirable colorfastness and visible light-absorption properties associated with these dyes. To this end, three PtII complexes each bearing two PDI moieties tethered to the metal center through acetylide linkages emanating from one of the PDI bay positions have been thoroughly examined by static spectroscopic methods, electrochemistry, laser flash photolysis, and ultrafast transient absorption spectrometry. Upon ligation to the PtII center, the bright singlet-state fluorescence ($\Phi = 0.91$, $\tau = 4.53$ ns) of the free PDI-CCH chromophore is quantitatively quenched, and no long wavelength photoluminescence is observed from any of the PtII-PDI complexes in deaerated solutions. Ultrafast transient measurements reveal that upon ligation of PDI-CCH to the PtII center, picosecond intersystem crossing ($\tau = 224$ ps) from the 1PDI excited state is followed by vibrational cooling ($\tau = 1219$ ps) of the hot 3PDI excited state, whereas only singlet-state dynamics, including stimulated emission, were observed in the "free" PDI-CCH moiety. In each of the Pt-PDI chromophores, quantitatively similar transient absorption difference spectra were obtained; the only distinguishing characteristic is in their single-exponential lifetimes ($\tau = 246$ ns, 1.0 μ s, and 710 ns). These long-lived 3PDI excited states are clearly poised for bimolecular electron and energy transfer schemes. In the present case, the latter is demonstrated through bimolecular sensitization of singlet oxygen phosphorescence at ~ 1270 nm in aerated dichloromethane solutions, producing reasonable 1O_2 quantum yields ($\Phi = 0.40-0.55$) across this series of molecules.

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