S1. Picosecond pump-probe transient absorption decay trace obtained using 355-nm excitation of the DUR/TCNB CT complex and observed at 468 nm polarized at the magic angle (open circles). The line indicates the best fit of the observed trace using a single exponential decay.
S2. Top panel: Picosecond pump-probe transient absorption decay traces obtained using 355-nm excitation of the DUR/TCNB CT complex and observed with 468-nm light polarized parallel (open boxes) and perpendicular (filled boxes) to the excitation. The lines show the best fit of the individual dichroic traces assuming a single exponential decay component to the dichroism decay in addition to the parameters obtained by fitting the magic angle trace (S1). Bottom panel: A plot of the orientation factor as a function of time, calculated using the above dichroic traces (filled circles). The solid line indicates the fit of orientation factor based on the parameters obtained from the fit of the individual traces above. The dashed line indicates a K value of 0.333.
S3. Picosecond pump-probe transient absorption decay trace obtained using 355-nm excitation of the MES/TCNB CT complex and observed at 468 nm polarized at the magic angle (open circles). The line indicates the best fit of the observed trace using a single exponential decay.
S4. Top panel: Picosecond pump-probe transient absorption decay traces obtained using 355-nm excitation of the MES/TCNB CT complex and observed with 468-nm light polarized parallel (open boxes) and perpendicular (filled boxes) to the excitation. The lines show the best fit of the individual dichroic traces assuming a single exponential decay component to the dichroism decay in addition to the parameters obtained by fitting the magic angle trace (S3). Bottom panel: A plot of the orientation factor as a function of time, calculated using the above dichroic traces (filled circles). The solid line indicates the fit of orientation factor based on the parameters obtained from the fit of the individual traces above. The dashed line indicates a K value of 0.333.