TR-2PPE Studies of Ultrafast Charge Separation at Organic Photovoltaic Interfaces S.W. ROBEY, G.J. DUTTON, NIST, W. JIN, J.E. REUTTER-ROBEY, University of MD — Dissociation of excitons in organic photovoltaic (OPV) devices occurs exclusively at interfaces between donor and acceptor molecular materials. To help understand critical charge separation processes, we have performed time-resolved two-photon photoemission (TR-2PPE) studies of sub-picosecond exciton dynamics at well-characterized organic donor-acceptor interfaces. Interfaces between phthalocyanines and C₆₀ were engineered using organic MBE and characterized using STM, STS, and UPS. Ultrafast TR-2PPE measurements were performed on CuPc\C₆₀ structures by pumping the lowest optical $\pi \rightarrow \pi^*$ transitions (Q-band) to generate CuPc singlet (S₁) excitons and probing this population with a time-delayed UV pulse. For thick films, CuPc S₁ decay is dominated by vibrational relaxation (several 100’s femtoseconds) and singlet-to-triplet conversion ($\sim$ 1 picosecond). Directly at the interface, however, charge transfer to C₆₀ dominates decay of S₁ exciton, ($\sim$ 100 femtoseconds). We also find evidence for important recombination routes from the charge separated state back to lower-lying CuPc T₁ triplet excitons. To test the impact of intersystem crossing to triplet levels, we have performed analogous investigations for H₂Pc\C₆₀ interfaces. Results for this interface will be compared and contrasted with the CuPc\C₆₀ case.