Abstract: Rapid changes in membrane potential facilitate the unique physiology of electrically-excitable cells like neurons and cardiomyocytes. Despite the central importance of membrane potential dynamics, methods to accurately monitor voltage rely on highly invasive electrodes or indirect optical measurements, such as calcium imaging with fluorescent sensors. Direct visualization of voltage changes has the potential couple the speed and sensitivity of electrode-based methods with the spatial resolution of imaging approaches. However, the development of fast, sensitive, and bright voltage-sensitive fluorescent indicators remains an outstanding challenge. Here, I present our efforts to design, synthesize, and apply fluorescent indicators that use photoinduced electron transfer (PeT) as a voltage-sensing trigger to achieve fast and sensitive voltage imaging in a variety of biological contexts.