Abstract
Photoluminescence (PL) blinking, or random switching between states of high- (ON) and low (OFF) emissivities, is a universal property of molecular emitters exhibited by organic molecules and artificial nanostructures. For the past fifteen years, colloidal nanocrystals have been used as a model system for studies of this phenomenon. The occurrence of OFF periods in nanocrystal emission has been commonly attributed to the presence of an additional charge, which leads to PL quenching by nonradiative Auger recombination. However, the “charging” model was recently challenged in several reports. Here, to clarify the role of charging in PL intermittency, we perform time-resolved PL studies of individual nanocrystals while controlling electrochemically the degree of their charging. We find that two distinct mechanisms can lead to PL intermittency. We identify conventional blinking (A-type) due to charging/discharging of the nanocrystal core when lower PL intensities correlate with shorter PL lifetimes. Importantly, we observe a different blinking (B-type), when large changes in the PL intensity are not accompanied by significant changes in PL dynamics. We attribute this blinking behavior to charge fluctuations in the electron-accepting surface sites. When unoccupied, these sites intercept hot electrons before they relax into emitting core states. Both blinking mechanisms can be controlled electrochemically and under appropriate potential blinking can be suppressed. These results offer an explanation to recent controversial observations on PL intermittency and should assist in the development of nanocrystals with nonblinking emission for applications from quantum information to high-sensitivity detection of chemicals and tracking of biological molecules. C.G and V.I.K acknowledge support of the Center for Advanced Solar Photophysics, an Energy Frontier Research Center funded by the U.S. Department of Energy (DOE), Office of Science, Office of Basic Energy Sciences (BES). Y.G. and A.S. are supported by Los Alamos National Laboratory Directed Research and Development Fund. M.S., J.A.H, and H.H. are supported by NIH-NIGMS Grant 1R01GM084702–01. This work was conducted, in part, at the Center for Integrated Nanotechnologies, a DOE/BES user facility.

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