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Fluorescence Intermittency and Nanodot Evolution in Graphene Oxide¹ ANTHONY RUTH, Univ of Notre Dame, HAYASHI MICHITOSHI, National Taiwan University, MATTHEW MCDONALD, JIXIN SI, YURI MOROZOV, Univ of Notre Dame, PETER ZAPOL, Argonne National Laboratory, MASARU KUNO, BOLDIZSAR JANKO, Univ of Notre Dame — In recent experiments, micron-sized reduced graphene oxide (rGO) flakes were observed to exhibit strong photoluminescence intensity fluctuations, or blinking. Although blinking has been observed in a wide variety of nanoscale emitters, and striking universalities exist across these very different systems, rGO is the first quasi-two dimensional emitter that shows blinking. Despite the widespread presence of blinking at nanoscale, a microscopic mechanism behind this phenomenon remains elusive. Here we provide density functional theory results, analytical calculations, and Monte Carlo simulations to connect the fluorescence trajectories observed in the experiment to microscopic processes. Through Monte Carlo simulations of chemical processes occurring on the graphene oxide surface, we observe the formation and destruction of carbon nanodots. Finally, we use emission characteristics of carbon nanodots from Ab Initio methods to reconstruct the photoluminescence of the macroscopic flake. In particular, we are investigating whether fluorescence intermittency in reduced graphene oxide is an intrinsic optoelectronic property of the nanodot constituents or the result of reversible chemical processes capable of changing the size and number of graphene nanodots.

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