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Effect of Air Exposure on Carrier Relaxation Dynamics in Colloidal Quantum Dots MILAN SYKORA, ALEXEY KOPOSOV, JOHN MCGUIRE, ROLAND SCHULZE, JEFF PIETRYGA, VICTOR KLIMOV, Los Alamos National Laboratory — The development of wet-chemistry synthetic routes to high-quality lead chalcogenide (PbS, PbSe, and PbTe) nanocrystals (NCs) created an opportunity for exploitation of these materials in practical applications where the tunability of electronic properties in the near-infrared region of the optical spectrum is of particular importance. One potential obstacle to broad application of lead chalcogenide NCs is their limited chemical and photochemical stability. Several recent studies of air exposed PbS and PbSe NC films provided evidence that under aerobic conditions the dominant degradation mechanism is surface oxidation of the NCs by atmospheric O_2. In the present work, we show that exposure of solutions of QDs to air leads to rapid oxidation of QDs that has pronounced effect on their chemical composition, electronic structure and carrier relaxation dynamics. Dramatic variations in PL quantum yield, observed following air exposure, are explained in terms of changes in the efficiencies of surface carrier trapping and nonradiative interband relaxation. After accounting for enhanced carrier trapping and oxidation-induced reduction in NC core size, we demonstrate that the dramatic changes in the surface properties of oxidized NCs do not significantly affect the dynamics of the Auger relaxation or the efficiency of Carrier Multiplication process.

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