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Ultrafast X-ray and optical time-resolved investigation of CuInS2 quantum dots

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The interest in copper-indium-sulfide (CIS) quantum dots (QDs), as heavy metal-free nanomaterials, has increased significantly in the past few years. CIS quantum dots (QDs) have been studied for many applications like photodynamic therapy, solar cells, LEDs, and bioimaging [1]. They show exciting optoelectronic properties, such as broad photoluminescence (PL) with a large Stokes shift and long charge carrier lifetimes. Several mechanisms for the radiative recombination in CIS QDs have been proposed over past years. In this work we aim to understand and confirm the possibility of radiative recombination resulting from an electron in the conduction band and a hole in a so-called confined hole state (CHS) or localized hole [2-3], which is a mid-gap state arising from a defect of Cu+ that can get oxidized to Cu2+ by the hole. The range of possible defects would explain the broad PL and large Stokes shift. In addition, we aim to understand the effects of stoichiometry and Zn doping on the formation process of the CHS.[4] To these ends, the element and oxidation state specificity of X-ray techniques can be a great tool to carry out a more direct observation of such processes, allowing us to confirm the proposed mechanisms. We approached these questions through a combination of laser, synchrotron and XFEL techniques, including our recent results from SACLA XFEL and ALBA. We focused on tracking in real-time the oxidation state changes of Cu via femtosecond-resolved Cu K-edge XANES and comparing the structure of the different samples through steady-state EXAFS. We have complemented our investigation in the X-ray range with time-resolved optical studies monitoring ultrafast transient absorption and photoluminescence (with fluorescence up-conversion spectroscopy).

References:
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