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# Assessing feasibility of detecting photogenerated charge carriers in photocatalysts via transmission electron microscopy: simulation study

Monia Runge Nielsen[1,2], Shima Kadkhodazadeh[1,2], Michael S. Seifner[1,2]

[1]Nanolab - National Centre for Nano Fabrication and Characterization, Technical University of Denmark, Kongens Lyngby, Denmark, [2]NanoPhoton - Center for Nanophotonics, Technical University of Denmark, Kongens Lyngby, Denmark

## Background

In the face of global energy and climate crises, the necessity for sustainable solutions has never been more pressing. Photocatalysts, which use sunlight to drive chemical transformations, offer hope in this quest. Among their many applications, one of the most promising lies in the conversion of carbon dioxide (CO2) into fuels, presenting a dual opportunity for reducing atmospheric CO2 levels and generating sustainable energy.1 However, the effectiveness of photocatalysts depends on the complex interaction of light absorption, charge carrier dynamics, and catalytic activity, particularly in the context of poorly understood steps such as charge carrier accumulation.1-3

## Methods

Taking advantage of the fact that the accumulation of charge carriers in photocatalyst will induce local alterations in the atomic structure affecting parameters such as nuclear charge, atomic/ionic radius, and chemical bonding.3 These alterations, potentially originating from selective reduction of cations at a specific nanocrystal (NC) active surface facet,4 can manifest as local phase shift variations in transmission electron microscope (TEM) images. The goal of this study is to identify visible light-induced phase shift variations by extracting phase images from reconstructed exit wave functions corresponding to specific NC surface facets under various conditions. Advanced image simulations will provide valuable insights into the detectability of photogenerated charge carriers in TEM experiments.

## Results

In this study, ab initio Transmission Electron Microscopy (abTEM) simulations5 was employed to assess the feasibility of detecting photogenerated charge carriers at the interface of photocatalysts under light irradiation, in the TEM. By creating a model photocatalytic system comprising facet engineered NCs, where photons are absorbed on the active facet during reaction, leading to the accumulation of photogenerated charge carriers. Simulating the structural and electronic properties of these photocatalysts under illumination, we aim to elucidate whether charge carrier accumulation at the interface is detectable in the TEM. I will present both high resolution TEM (HRTEM) and integrated differential phase contrast STEM (iDPC-STEM) imaging techniques and assess whether they are sufficiently sensitive to detect the potential difference arising from the slight change in nuclear charge.

## Conclusion

This simulation study provides a crucial insight on the sensitivity of TEM techniques to subtle changes in potential and chemical reactivity induced by photogenerated charge carriers, providing insights into the feasibility of using TEM for monitoring photocatalytic reactions. This serves as a crucial step towards using advanced microscopy techniques for probing the dynamics of photocatalysis at the nanoscale, ultimately facilitating the design and optimization of efficient photocatalytic systems for renewable energy generation and addressing environmental issues.

Keywords:

Photocatalyst, charge carriers, abTEM simulations

**Reference:**

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