Metal-oxide based systems for efficient visible light conversion

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ON-SITE - S3 Seminar Room, Third Floor, Physics Building
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Speaker

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Abstract

Research in renewable energy, particularly solar-to-chemical conversion via photocatalysis, is vital for mitigating climate change and fossil fuel scarcity. Cerium oxide, combined with plasmonic nanoparticles (NP), hold promise as a material meeting critical efficiency and stability criteria.

In order to completely understand and then to optimize a photocatalytic material it is crucial to characterize it in terms of optical, morphological and electronic properties, and to understand how these properties can influence each other. Moreover, to obtain photocatalysts with increased visible light harvesting efficiency and with an optimized density of long-living excited states, it is fundamental to characterize not only their static properties, but also their dynamics, to properly describe the light-induced excitations. We analyzed CeO$_2$ thin films, either alone or combined with Au NPs by using advanced time-resolved spectroscopy techniques. First, we studied the electron injection efficiency and the steady state absorbance at different photon energies for a composite system made of Au NPs embedded in a cerium oxide matrix. We characterized the ultrafast dynamics of excited states induced by UV and visible-light excitation in the system, aimed at understanding the excitation pathways, by means of femtosecond transient absorbance spectroscopy. Secondly, we investigated the ultrafast dynamics of photoexcited states in cerium oxide at the European X-Ray free electron laser (FEL). Ultra-short and ultra-intense FEL pulses, tunable in energy in the x-ray energy range, can provide information on the dynamics of photoinduced modifications of the local electronic and atomic structure around the individual atomic species via pump-probe X-ray absorption spectroscopy (XAS). We successfully measured pump-probe Ce L$_3$-edge XAS spectra in the total fluorescence yield acquisition mode in the near and extended energy range at different delay times and we measured the dynamics of the observed variations at different photon energies within a delay time range extending up to 250 ps, with a resolution of 100 fs. We could also acquire pump-probe Ce L$_a$ X-ray emission spectroscopy (XES) spectra at different photon energies across the Ce L$_3$ edge, exciting the Ce L$_2$ edge and detecting the Ce L$_{a1}$ XES intensity – at different delay times. The data show clear dynamic changes in the electronic properties and in the local atomic structure as a function of the delay time between the pump and the probe.

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