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Real-Space Mapping of Surface Trap States in Cigse Nanocrystals Using 4D Electron Microscopy

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Abstract : This work reports visualization of charge carrier dynamics on the surface of copper indium gallium selenide (CIGSe) nanocrystals in real space and time using four-dimensional scanning ultrafast electron microscopy (4D S-UEM) and correlates it with the optoelectronic properties of the nanocrystals. The surface of the nanocrystals plays a key role in controlling their applicability for light emitting and light harvesting purposes. Typically for quaternary systems like CIGSe, which have many desirable attributes to be used for optoelectronic applications, relative abundance of surface trap states acting as non-radiative recombination centre for charge carriers remains as a major bottleneck preventing further advancements and commercial exploitation of these nanocrystals devices. Though ultrafast spectroscopic techniques allow determining the presence of picosecond carrier trapping channels, because of relative larger penetration depth of the laser beam, only information mainly from the bulk of the nanocrystals is obtained. Selective mapping of such ultrafast dynamical processes on the surfaces of nanocrystals remains as a key challenge, so far out of reach of purely optical probing time-resolved laser techniques. In S-UEM, the optical pulse generated from a femtosecond (fs) laser system is used to generate electron packets from the tip of the scanning electron microscope, instead of the continuous electron beam used in the conventional setup. This pulse is synchronized with another optical excitation pulse that initiates carrier dynamics in the sample. The principle of S-UEM is to detect the secondary electrons (SEs) generated in the sample, which is emitted from the first few nanometers of the top surface. Constructed at different time delays between the optical and electron pulses, these SE images give direct and precise information about the carrier dynamics on the surface of the material of interest. In this work, we report selective mapping of surface dynamics in real space and time of CIGSe nanocrystals applying 4D S-UEM. We show that the trap states can be considerably passivated by ZnS shelling of the nanocrystals, and the carrier dynamics can be significantly slowed down. We also compared and discussed the S-UEM kinetics with the carrier dynamics obtained from conventional ultrafast time-resolved techniques. Additionally, a direct effect of the state trap removal can be observed in the enhanced photoresponse of the nanocrystals after shelling. Direct observation of surface dynamics will not only provide a profound understanding of the photo-physical mechanisms on nanocrystals' surfaces but also enable to unlock their full potential for light emitting and harvesting applications.

Keywords : 4D scanning ultrafast microscopy, charge carrier dynamics, nanocrystals, optoelectronics, surface passivation, trap states
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