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Probing the Surface Photovoltage Effect by Imaging Photo-assisted Secondary Electron Emission

Bolin Liao, Yu Li and Usama Choudhry

University of California, Santa Barbara, Santa Barbara, California, United States

Scanning ultrafast electron microscope (SUEM) has emerged as a photon-pump-electron-probe imaging tool with combined high spatial and temporal resolutions, which is particularly suitable to visualize photocarrier dynamics on surfaces or at interfaces of photonic materials[1,2]. Originally developed in Zewail group at Caltech, SUEM has been utilized to image the photocarrier separation at a silicon p-n junction[3], the spontaneous spatial separation of hot electrons and holes in hydrogenated amorphous silicon[4], and the anisotropic photocarrier transport in black phosphorous[5], among other examples. In this talk, I will first introduce our development of an SUEM setup at the University of California, Santa Barbara, where we have implemented a few technical improvements, including a mechanical coupling between the SEM suspension system and the optical system, and an in-chamber lens holder to push for smaller optical pump beam size. Then I will discuss our recent effort to systematically understand the contrast mechanisms of SUEM, which has only been qualitatively discussed before. In particular, I will describe our observation of the surface photovoltage effect (SPV) on the SUEM contrast of doped semiconductors. SPV originates from the surface band bending of doped semiconductors due to the Fermi-level pinning by surface in-gap defect states. For example, in heavily doped p-type (n-type) silicon, the surface band bends downward (upward) and thus helps (suppresses) secondary electron emission from the surface. Photoexcited electrons and holes migrate under the surface built-in potential and will compensate for the surface band bending effect. Therefore, p-type (n-type) silicon is expected to show dark (bright) contrast under photo-illumination compared to the case without the optical pumping. We observed that (1) the SPV effect only affects the SUEM contrast for primary electrons with lower energy (< 5 keV), mainly because electrons with higher energy penetrate much deeper than the surface band bending area; (2) under sufficiently strong photoillumination, the surface band bending can be fully compensated, indicated by a contrast reversal (from dark to bright) in p-type silicon; (3) The dependence of the contrast intensity on the optical pump modulation frequency reveals the timescale of the surface charge transport driven by the SPV effect. This study paves the way for a quantitative understanding of SUEM contrasts in future experiments. This work is partially supported by DOE (DE-SC0019244), ARO (W911NF-19-1-0060) and NSF (DMR-1905389).

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