doi:10.1017/S143192762200349X
Microsc. Microanal. 28 (Suppl 1), 2022

Imaging Atomically Thin Transition Metal Dichalcogenides Using Deep Ultraviolet Photoelectron Emission Microscopy

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Photoelectron emission microscopy enables us to probe the materials properties by harnessing light-matter interactions. [1]. Because the photoelectron yield is a convolution of the electric field strength and the occupied electron density of states, photoelectron emission microscopy (PEEM) has the potential to explore the electronic structure and the optical response at the nano-meter length scale [2]. In this talk we will highlight two exemplars focusing on the nanoscale heterogeneity at transition metal dichalcogenide (TMD)-gold interfaces [3] and the optical response of TMD-embedded dielectrics cavities [4].

There is an intensive effort to control the interactions between ultrathin semiconductors and metals and to understand their impact on the electronic properties at the junction. We employ a tailored sample structure, in which the junction between WS_2 and Au crystallite forms a semi-epitaxial relationship to directly compare how the electronic structure of WS_2 changes when the intimate interface is established with Au. By conducting photoelectron spectroscopy using PEEM, we directly probed the highest occupied states at the Brillouin zone center as a function of WS_2 thickness with sub-micron spatial resolution. In multilayer WS_2, we discovered variations in the electronic states that spatially align with the crystalline grains of underlying Au. Corroborated by density functional theory calculations, we attribute the electronic structure variations to stacking variations within the WS_2 films. We propose that strong interactions exerted by Au grains cause slippage of the interfacing WS_2 layer with respect to the rest of the WS_2 film. Our findings illustrate that the electronic properties of transition metal dichalcogenides, and more generally 2D layered materials, are physically altered by the interactions with the interfacing materials, in addition to the electron screening and defects that have been widely considered.

Imaging of fabricated nanostructures or nanomaterials covered by dielectrics is highly sought after for diagnostics of optoelectronics components. We show imaging of atomically thin MoS_2 flakes grown on SiO_2-covered Si substrates and buried beneath HfO_2 overlayers up to 120 nm in thickness using PEEM with deep-UV photoexcitation. Comparison of photoelectron yield to modeled optical absorption evinced the formation of optical standing waves in the dielectric stacks (i.e., cavity resonances of HfO_2 and SiO_2 layers on Si). The presence of atomically thin MoS_2 flakes modifies locally the optical properties of the dielectric stack and thus the cavity resonance condition, resulting in image contrast. Finally, we show the cavity resonances are imaged in other dielectric stacks (Al_2O_3 and SiO_2 layers on Si substrate, and SiO_2 layers on Si substrate), suggesting the broad applicability of this imaging concept. This subsurface sensitivity underscores the role of optical effects in photoelectron imaging with low-energy photons. This approach can be extended to nondestructive imaging of buried interfaces and subsurface features needed for analysis of microelectronic circuits and nanomaterial integration into optoelectronic devices.

Overall, the work presented here underscores the opportunities to elucidate the materials' electronic properties, optical responses, and their combined phenomena at the nano-meter length scale using PEEM [5].

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[5] We acknowledge insightful discussions with N. Bartelt, M. B. Sinclair, and W. T. S. Luk. We thank G. Copeland for his assistance in the PEEM measurement. The work at Sandia National Laboratories was supported by Sandia's LDRD program and the Center for Integrated Nanotechnologies user program, an Office of Science User Facility operated for the U.S. Department of Energy (DOE) Office of Science (DEAC04-94AL85000). The work at the U.S. Naval Research Laboratory (NRL) was funded by the Office of Naval Research. Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525. The views expressed in the article do not necessarily represent the views of the U.S. Department of Energy or the United States Government.