Ultrafast Dynamics of Carriers at Hybrid Organic / Inorganic Semiconductor Interfaces

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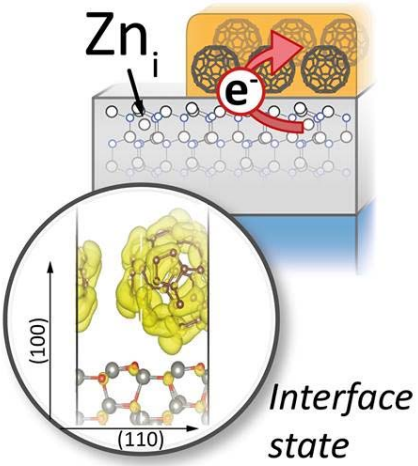
Interfaces between organic and inorganic semiconductors have an essential function in many next-generation optoelectronic technologies. Hybrid organic electronic devices combine the ease of tailoring organic semiconductors with the superior electronic properties of inorganic semiconductors to achieve high-efficiency in photovoltaic devices, light-emitting diodes, micro-scale lasers, spin-valves and transistors. Moreover, inorganic semiconductors are used as functional interlayers of complex device architectures in fully organic devices, where they may prevent unwanted carrier recombination or act as charge-selection layers. Yet, little is known about the principles that determine the emerging electronic structure at such hybrid organic / inorganic semiconductor interfaces, and the microscopic mechanism and carrier dynamics are unclear.



In this talk, I will discuss the challenge of understanding such interfaces, focusing on the factors that determine the rich physics taking place in the presence of poor screening, bandgaps and gap states. Using the prototypical wide-bandgap semiconductor ZnO, I will show how the naturally ultrafast dynamics of carriers in the conduction band are disrupted upon interface formation [1,2], and offer a detailed atomistic understanding of the observed localization dynamics [3-6]. Indeed, I will demonstrate how hybridization between localized states in the inorganic semiconductor and electronic levels of the π-conjugated organic molecule, ranging from C_{60} to various perylene derivatives, fundamentally determines the interfacial electronic structure and how it is responsible for the observed ultrafast dynamics. Our work has also explored the range of conditions under which the proposed model is valid, tying high-sensitivity photoelectron spectroscopies, optical spectroscopy and time-resolved photoemission experiments together and indicating the important role of screening and the nature of the

participating wavefunctions.

Finally, I will show results from initial studies of electronic structure and ultrafast dynamics in van der Waals layered (quasi-)2D materials, ranging from topological insulators [7] to semiconductors [8-10]. These results demonstrate how interfacial hybridization can be chemically controlled, opening new avenues for rational tailoring of electronically active low-dimensional materials.

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