Coupling between 0D and 3D: hot electrons, coherent phonons, and solar energy conversion

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Semiconductor nanocrystals (NCs) are called artificial atoms because of their atom-like discrete electronic structure due to quantum confinement. They can also be assembled into artificial molecules or solids, thus, extending the toolbox for material design. We address the interaction of artificial atoms or molecules with bulk semiconductor surfaces. These interfaces are model systems for understanding the coupling between localized and delocalized electronic structures. In many perceived applications, such as nanoelectronics and solar energy conversion, interfacing semiconductor NCs to bulk materials is a key ingredient. Here, we apply the well-established theories of chemisorption and interfacial electron transfer as conceptual frameworks for understanding the coupling between NCs and bulk semiconductor surfaces. In the model system of PbSe NCs adsorbed on $TiO_2(110)$, we show that strong interfacial electronic coupling allows hot electron transfer from photo-excited NCs. Moreover, we discovered that the transient electric field resulting from ultrafast charge separation across the PbSe-TiO_2 interface excites coherent vibrations of the TiO_2 surface atoms, whose motions can be followed in real time. These results indicate hot electron transfer from semiconductor NCs to a technologically relevant electron acceptor is possible. This effect is expected to be of general significance to other 0D/3D interfaces and may be utilized in the design of the ultimate solar cell.