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Ultrafast Excited State Dynamics and Nonlinear Optical Spectroscopy at Interfaces

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We employ time- and angle-resolved photoemission spectroscopy to obtain a direct momentum resolved view of the ultrafast excited state dynamics and interlayer charge transfer in semiconducting transition metal dichalcogenides (TMDCs). While in bulk TMDCs the overall crystal structure is inversion symmetric, individual layers lack this property and in combination with strong spin-orbit coupling the energy degeneracy of electronic bands of opposite spin polarizations is lifted. Applying circularly polarized light leads to momentum- and spin-selective selective excitation of spin-polarized electrons in the K-valleys of the TMDC band structure. We show that, even in centrosymmetric samples of 2H-WSe_2 we can generate spin-, valley- and layer-polarized excited states in the conduction band which are localized within individual layers at the K points. Subsequent ultrafast scattering populates states at the Σ-valley with a three-dimensional character facilitating optical control of interlayer charge transfer. Another important type of interfacial charge transfer processes occurs in electrochemical environment at solid-liquid interfaces. These are difficult to probe by interface specific techniques. Here I will report on our recent progress using nonlinear optical spectroscopy to study the structure of interfacial water and oxide formation on gold electrodes under potential control.