CHYN MEETS HARBOR

Droplet etching for quantum structures with tunable wavefunction

Leonardo Ranasinghe

Probing the UV-induced photochemistry of the L-Cysteine disulfide in aqueous solution via femtosecond X-ray absorption spectroscopy

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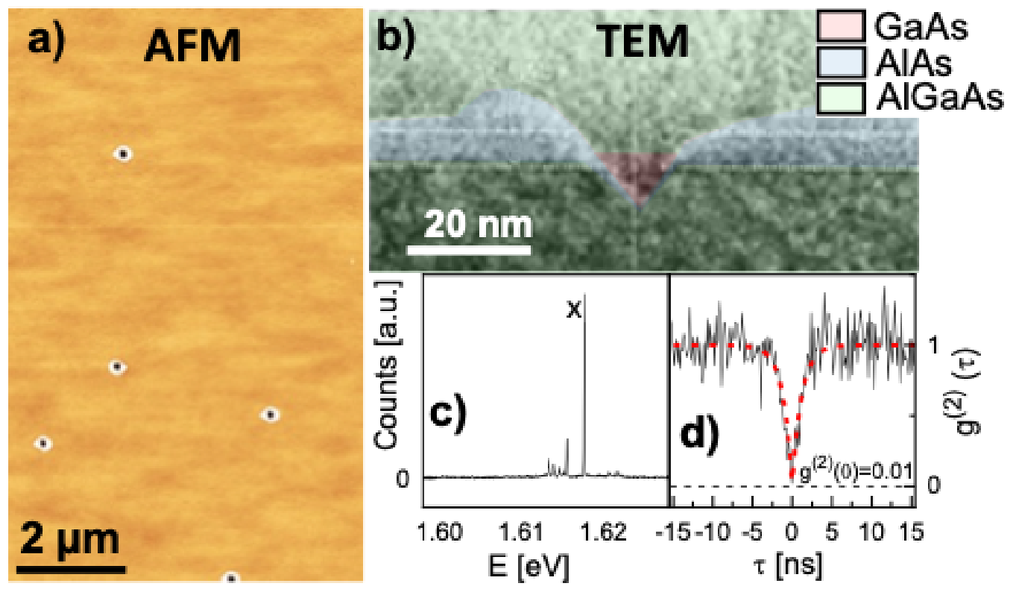
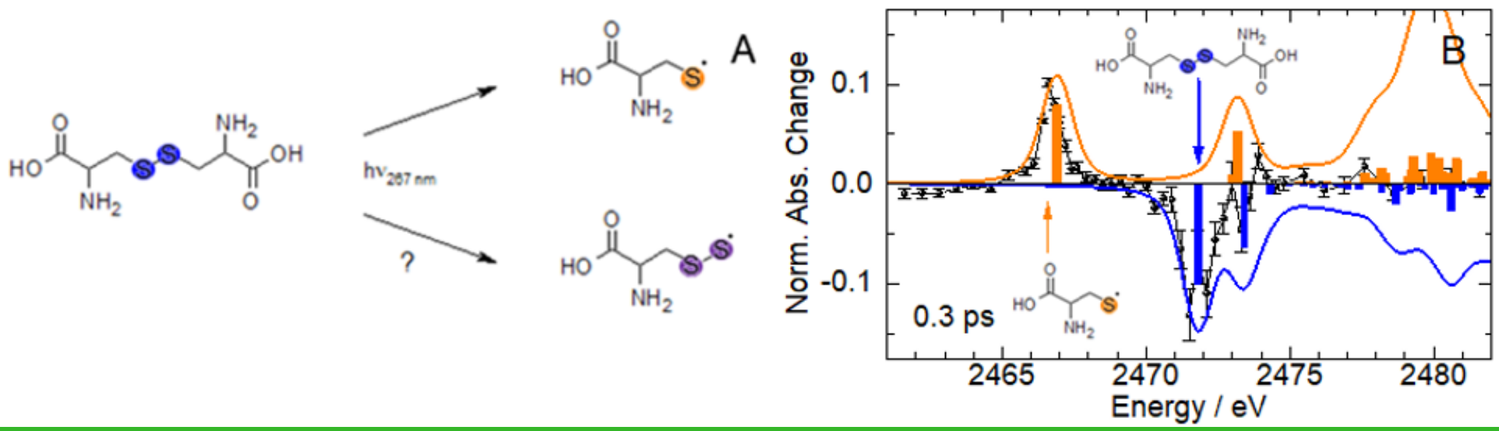
Abstract Talk 1: Semiconductor quantum dots (QDs) are important building blocks for quantum information processing and cryptography. We fabricate GaAs QDs by molecular beam epitaxy (MBE) and study their structural and optical properties. The fabrication process starts with the self-assembled etching of low-density nanoholes in AlAs or AlGaAs surfaces using hot Al droplets as etchant (Fig. 1a). Subsequently, the nanoholes are filled with GaAs for the generation of QDs with a precisely controlled density, size, and shape (Fig 1b).

Single-dot photoluminescence (PL) spectroscopy demonstrates sharp excitonic lines with linewidth down to 25 µeV (Fig. 1c) and autocorrelation measurements establish perfect single-photon emission (Fig. 1d). Simulations predict that a vertical electric field induces a controlled dot to ring transformation and a tunable elongation of the QD radiative lifetime up to milliseconds, which suggests these dots as a quantum memory for light. Furthermore, the creation of quantum dot molecules with strong coupling is demonstrated by filling of a nanohole with two QDs.



Abstract Talk 2: The disulfide bond motif is an important structure-making moiety in proteins, whereby two spatially adjacent L-cysteinyl residues in an amino acid chain can react to form a covalent disulfide bond. These disulfide bonds stabilize the proteins tertiary structure and can act as a UV radiation shield and as radical scavengers.However, many of the underlying processes are still unknown. This is especially true for the earliest timescales where the initial disulfide bond breakage occur.Therefore, the UV photochemistry of sulfur-containing amino acids and in particular of the disulfide bond are of interest in the field of ultraviolet photochemistry in proteins. Such reactions have therefore been studied extensively with both conventional and time-resolved methods. We demonstrated that time-resolved X-ray absorption spectroscopy (TRXAS) at the sulfur K-edge is a chemically sensitive tool to observe the UV photochemistry of simple organosulfur compounds in nonpolar solvent environments [1,2]. However, for a better understanding of the photochemistry under physiological conditions, these model systems need to be extended to the natural amino acid L-Cysteine in aqueous solution. Figure 1: Panel A shows possible reaction pathways of L-Cystine after UV-excitation. In panel B is the differential X-ray absorption spectrum of L-Cystine at 0.3 ps after 267 nm excitation is displayed. The negative absorbance change at 2471.8 eV signals the emergence of either excited state parent molecules or new sulfur-containing species. The characteristic absorption lineshape of the sulfur radical at 2466.5 eV suggests ultrafast generation of thiyl radicals.

Herein, we report the first results of the photodissociation dynamics of L-Cystine, the disulfide dimer of L-Cysteine, in aqueous solution upon UV irradiation with 267 nm light using TRXAS at the sulfur K-edge with femtosecond time resolution. We observe the emergence and subsequent decay of the photoproduct on timescales of hundreds of femtoseconds up to 800 ps.



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