



Seminar announcement

Monday, June 2, 2025

10:00 am

WSI, Seminar room S 101

Exclusively in person

“Dynamic landscape engineering in semiconducting photoelectrodes for light-driven organic transformations”

Photoelectrocatalysis presents a sustainable strategy for solar-driven chemical transformation by coupling light absorption with interfacial redox reactions. Transition metal oxides (TMOs) such as BiVO₄ and CuFeO₂ offer operational stability in aqueous media and favorable band alignment, yet their performance is hampered by polaron-induced charge transport losses. To elucidate carrier relaxation pathways and energy-loss mechanisms, we developed a chemical-solution deposition method combined with rapid thermal annealing to synthesize highly epitaxial BiVO₄ and CuFeO₂ thin films. Resonant inelastic X-ray scattering (RIXS) reveals anisotropic, orbital-symmetry-dependent electron-phonon coupling, providing insights into energy dissipation pathways in these materials.

This talk will also highlight two photoelectrocatalytic organic transformations. First, selective oxidation of benzyl alcohol to benzaldehyde is achieved using a BiVO₄ photoanode in acetonitrile. Interfacial states formed with the redox mediator N-hydroxy-succinimide (NHS) facilitate carrier recombination and reduce photocurrent. Optimized film deposition and dopant incorporation suppress these detrimental states, enabling a benzaldehyde production rate of 20.6 $\mu\text{mol cm}^{-2} \text{ h}^{-1}$ with nearly-unity Faradaic efficiency. Second, we tune the conductivity type of inverse spinel CuFe₂O₄ from n-type to p-type by controlling annealing conditions, passivating oxygen vacancy donors and introducing copper vacancy acceptors. The resulting p-type CuFe₂O₄ photocathode achieves stable photoelectrocatalytic hydrogenation of benzaldehyde to benzyl alcohol over 16 hours.

Finally, we report the fabrication of ultrathin Ta₃N₅ photoanodes via the disproportionation of metastable Ta₂N₃. These 100 nm Ta₃N₅ films on silicon substrates eliminate the need for Ta foils and outperform thicker films derived from conventional TaOx precursor, reducing tantalum usage while delivering a photocurrent density of 3.86 mA cm^{-2} . Together, these studies underscore the importance of engineering carrier dynamics and interfacial energetics at both semiconductor-electrolyte and semiconductor-substrate interfaces to advance selective and efficient photoelectrocatalytic transformations.

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