Controlling Hot Electron Photoemission Dynamics in Plasmonic Nanostructures

David J. Nesbitt

JILA, University of Colorado and National Institute of Standards and Technology, and Department of Chemistry and Biochemistry, University of Colorado, Boulder, Colorado 80309-0440

Abstract

A variety of advanced photonic applications rely on efficient generation of hot carriers within metal nanoparticles and charge transfer to surrounding molecules or materials. The optimization of such processes requires a detailed understanding of the excited carrier spatial, temporal, and momentum distributions, which also leads to opportunities for active optical control over hot carrier dynamics at the space-time limit of nanometer and femtosecond scales. This talk will summarize recent progress on controlling/measuring ultrafast electron photoemission dynamics in plasmonic nanostructures, with topics (time permitting) in the following complementary areas. 1) There is a growing appreciation in the literature for clarifying mechanistic differences between volume- vs. surface-mediated electron photoemission processes. With our SPIM apparatus, we exploit angle-resolved velocity map imaging (VMI) to distinguish volume and surface contributions to nanoplasmonic hot electron emission from gold nanorods as a systematic function of aspect ratio (from highly prolate to spherical). 2) We have explored purely isotropic plasmonic nanostructures, specifically gold nanospheres and nanoshells, which result in surprisingly anistropic hot electron emission distributions that can be continuously controlled via laser polarization. 3) We have developed theoretical models based on plasmonic electric field distributions within the nanoparticle volume and ballistic Monte Carlo modeling of the hot electron dynamics, that recapitulate the SPIM/VMI experimental results in near quantitative detail. The results demonstrate a remarkably predictive understanding of the underlying physics and possibilities for ultrafast spatiotemporal control over hot carrier dynamics.