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Optically Controlling Femtosecond Hot Electron Currents in Nanoplasmonic Systems[1]

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Plasmonic metal nanoparticles concentrate optical field energy into deeply sub-wavelength volumes, producing high densities of excited (hot) electrons and holes. While predicting and controlling the spatial and momentum distributions of these hot carriers remain significant challenges, such capabilities introduce exciting opportunities for actively controlling ultrafast currents in a variety of photocatalytic, photovoltaic, and nanoelectronic applications. Toward these ends, some recent insights from single-nanoparticle angle-resolved photoemission spectroscopy studies will be presented. Gold nanostars, for instance, behave as multi-tip photocathodes with simultaneous frequency- and polarization-selective tip hot spots for photocurrent directionality control. Gold nanorods, on the other hand, provide a unique testbed for distinguishing fundamental surface- vs. volume-mediated photoemission pathways and their corresponding hot electron spatial/momentum distributions. These investigations are complemented by a combination of classical finite element electrodynamics, semi-classical Monte Carlo, and fully quantum modeling for predictively understanding hot electron dynamics in arbitrary nanoplasmonic geometries.

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