Noble metal nanoparticles have been of tremendous interest because of their intriguing size- and shape-dependent plasmonic and catalytic properties. The combination of tunable plasmon resonances with superior catalytic activities on the same nanoparticle, however, has long been challenging because plasmonics and catalysis require nanoparticles in two drastically different size regimes. Tunable plasmon resonances are a unique feature of sub-wavelength metallic nanoparticles, whereas heterogeneous catalysis requires the use of sub-5 nm nanoparticles as the catalysts. My group has recently demonstrated that desired plasmonic and catalytic properties can be integrated on the same particle by controllably creating high-index facets on individual sub-wavelength metallic nanoparticles. The capabilities to both nanoengineer high-index facets and fine-tune the plasmon resonances through deliberate particle geometry control allow us to use these nanoparticles for a dual purpose: as substrates for plasmon-enhanced spectroscopies and efficient surface catalysts. Such dual functionality enables us to gain quantitative insights into the facet-dependent molecular transformations on Au nanocatalysts using surface-enhanced Raman spectroscopy as an ultrasensitive spectroscopic tool with unique time-resolving and molecular fingerprinting capabilities.

I will also talk about our latest progress on plasmonic hot electron-driven photoreactions. It has been recently observed that the energetic hot electrons generated during plasmon decay can be harnessed to drive or enhance a series of interesting chemical or photochemical reactions on metallic nanoparticle surfaces. However, the detailed mechanisms of these plasmon-mediated reactions are still poorly understood and under intense debate. We use surface-enhanced Raman spectroscopy to precisely monitor, in real time, the plasmon-driven photocatalytic reactions at the molecule-nanoparticle interfaces, which allows us to pinpoint the effects of plasmon excitations, molecular adsorption states, local field enhancements, and photothermal processes, on the plasmon-driven photoreactions.

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