Optics at the Nanoscale: Light Emission in Plasmonic Nanocavities

Thesis by
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For my parents, David and Judy Ross,
who taught me to embrace the pursuit of knowledge,
in memory of my grandmother, Patricia Ross,
whose laughter and smile have always been an inspiration,
and with love and gratitude to Doug
for his encouragement along the way.

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Abstract

Nanophotonics has greatly benefited from the unique ability of surface plasmons to confine optical modes to volumes well below the diffraction limit of light. Plasmonics is an emerging area of research that opens the path for controlling light–matter interactions on the subwavelength scale, enabling truly nanophotonic technologies that are unattainable with conventional diffraction-limited optical components. Novel surface plasmon devices exploit electromagnetic waves confined to the interface between a metal and a dielectric, and permit the researcher to shrink light to dimensions previously inaccessible with optics. The extremely high and localized fields in plasmonic nanocavities are finding applications in research areas such as single-molecule sensing, nano-lasers, and photothermal tumor ablation, among others.

This thesis explores, both experimentally and theoretically, light emission in a number of plasmonic nanostructures. We present cathodoluminescence imaging spectroscopy as a new method of characterizing surface plasmons on metal films and localized in nanocavity resonators, with experimental observations supported by analytical calculations and electromagnetic simulation. This technique enables extremely localized surface plasmon excitation, a feature we exploit in both planar metal geometries and plasmonic nanocavities. We also study a specific nanocavity geometry, the plasmonic core-shell nanowire resonator, investigating both passive and active semiconductor core materials. This geometry allows precise control of the local density of optical states (LDOS), exhibiting the highest LDOS and smallest mode volumes in structures with dimensions as small as $\lambda/50$. Moreover, we discuss the Purcell effect as it applies to plasmonic nanocavities, and calculate enhancements in the radiative decay rate of more than 3000× in the smallest structures. These results demonstrate the promise of plasmonics to enable truly nanophotonic technologies and to manipulate light at the nanoscale.

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Portions of this thesis have been drawn from the following publications:

“Enhancing the rate of spontaneous emission in III-V semiconductor plasmonic core-shell nanowire resonators.” C. E. Hofmann, F. J. García de Abajo, and H. A. Atwater, submitted.

“Conjugated polymer/metal nanowire heterostructure plasmonic antennas.” D. M. O’Carroll, C. E. Hofmann, and H. A. Atwater, *Advanced Materials* **22**, 1223-1227 (2010).

“A plasmonic ‘bull’s-eye’ nanoresonator.” C. E. Hofmann and H. A. Atwater, *SPIE Newsroom* 1088-2008-03-04 (2008).

“Plasmonic modes of annular nanoresonators imaged by spectrally resolved cathodoluminescence.” C. E. Hofmann, E. J. R. Vesseur, L. A. Sweatlock, H. J. Lezec, F. J. García de Abajo, A. Polman, and H. A. Atwater, *Nano Letters* **7**, 3612-3617 (2007).

“Direct imaging of propagation and damping of near-resonance surface plasmon polaritons using cathodoluminescence spectroscopy.” J. T. van Wijngaarden, E. Verhagen, A. Polman, C. E. Ross, H. J. Lezec, and H. A. Atwater, *Applied Physics Letters* **88**, 221111 (2006).