Chapter 7

Summary and Outlook

The field of plasmonics has undergone significant growth in recent years, primarily due to advances in nanofabrication techniques and full-field electromagnetic simulation. We are now able to not only investigate optics at the nanoscale, but to precisely control and predict these phenomena in a wide array of architectures. This thesis has been devoted to the study of light emission in a variety of plasmonic geometries, illustrating new methods of exciting and manipulating light in complex nanocavities.

In the early chapters, we demonstrated that cathodoluminescence imaging spectroscopy is a powerful tool for investigating metallic films and nanostructures. The light emission resulting from electron beam excitation was used to determine the propagation distance of near-resonance surface plasmons, and distances as short as 400 nm were measured on Ag films. We also examined a metal-coated semiconductor nanowire with this technique, and were able to excite localized resonances in the metal coating, as well as bright emission from the semiconductor nanowire. Modes of a more complex geometry, the annular nanoresonator, were also investigated with spectrally resolved cathodoluminescence. We mapped out the spatial profiles of annular nanoresonator modes in both Ag and Au structures with extremely high spatial resolution. Supported by full-field simulation, our observations led to the conclusion that CL emission in a particular resonator mode is brightest for electron beam excitation in a position of high field intensity.

We then chose to study a specific plasmonic nanocavity, the core-shell nanowire resonator, with a number of different semiconductor core materials and an optically thick Ag coating. Theoretical calculations of the Si-Ag geometry proved that the LDOS could be carefully tuned by adjusting the dimensions of the resonator, and that

the highest LDOS is achieved in the smallest structures. Remarkably, a Si-Ag core-shell nanowire resonator with dimensions on the order of $\lambda/50$ still sustained modes with $Q > 25$, permitting dramatic enhancements in the total and radiative decay rates. Further, we presented two routes of fabricating the Si-Ag core-shell nanowire resonators, one relying on top-down lithography and RIE etching techniques, and the other based on CVD growth, oxidation, and wet chemical etching. In both cases, Si cores with diameters of <50 nm were achieved, and fully fabricated resonators were characterized with dark-field optical spectroscopy. Switching to III-V semiconductor core materials, we illustrated that active plasmonic core-shell nanowire resonators are a promising design for fast, bright, and directional on-chip light sources. Structures with GaAs, AlGaAs, and InGaN active emitters were chosen, and all exhibited extremely high field confinement for deeply subwavelength mode volumes. Again here, the smallest structure (InGaN) provided the most dramatic decay rate enhancements of more than 8000 times with quantum efficiency of >40%.

One area of ongoing work involves designing metal nanostructures to enhance the rate of spontaneous emission from GaAs nanowires, inspired by the theoretical results of Chapter 6. Our preliminary experiments include dropcasting high quantum efficiency GaAs NWs grown by molecular beam epitaxy [92] on a variety of substrates (Si, Ag, and Ag coated with a thin Cr layer) and comparing the GaAs PL intensity and lifetime decay traces. Single nanowire spectroscopy is performed using the setup shown in Figure 7.1, with a picosecond supercontinuum laser source operating at 40 MHz passed through filters for excitation between 550 and 750 nm. The laser is sent through an inverted optical microscope with a 100× objective (0.9 NA) and focused to a 15 μm spot on the nanowire. In the detection path, a CCD and a single photon avalanche diode (SPAD) permit collection of single nanowire PL spectra and PL lifetime decay traces, respectively. Figure 7.2a shows a TEM image of an individual GaAs NW approximately 15 μm long, and with a diameter that tapers from 150 to 80 nm. The NW is composed of a GaAs p-i-n radial junction and thin $Al_{0.4}Ga_{0.6}As$/GaAs cap 10 nm thick. Once dropcast onto the various substrates, PL spectra (Figure 7.2c) and PL lifetime decays (Figure 7.2d) are measured for a

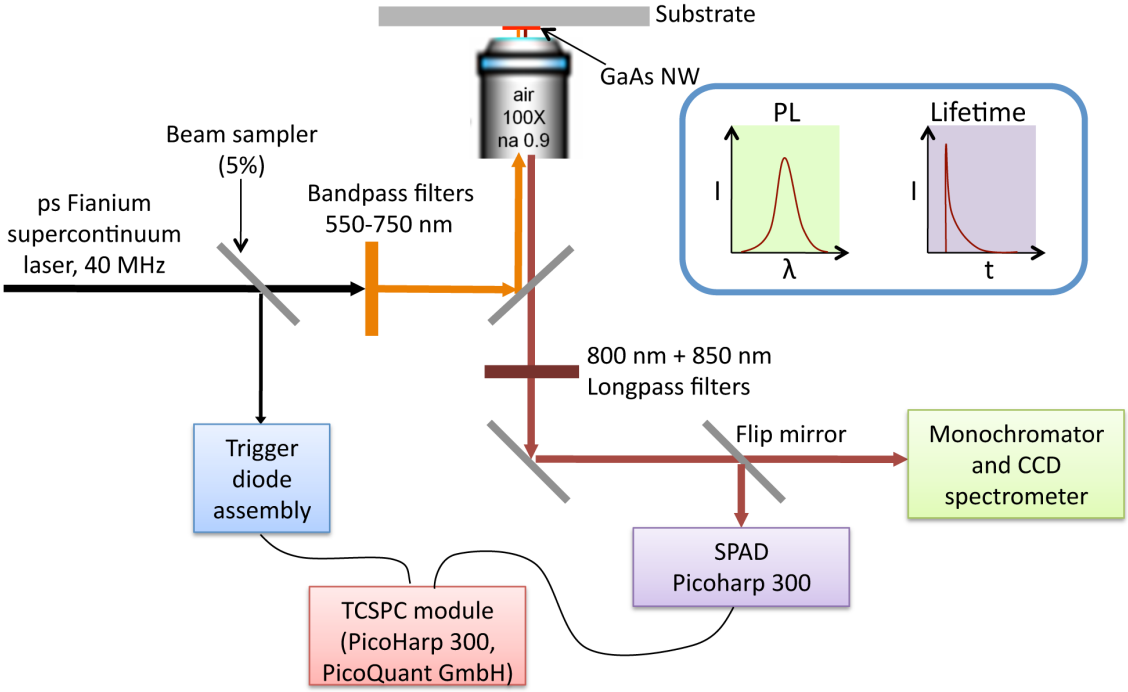

Figure 7.1. Schematic of single nanostructure PL and lifetime decay experimental setup. A picosecond supercontinuum laser source operating at a fixed repetition rate of 40 MHz is sent through bandpass filters at wavelengths of 550–750 nm, into an inverted optical microscope operating in reflection equipped with a 100× objective (NA 0.9), and focused onto a single nanostructure. The emitted PL is sent through a pair of longpass filters (800 and 850 nm) and then, via a flip mirror assembly, into either a monochromator and CCD spectrometer for collection of PL spectra, or into a single photon avalanche diode (SPAD) detector and time-correlated single photon counting (TCSPC) module for lifetime measurements. A beam sampler (5%) sends a portion of the incident laser light into a trigger diode to sync the source and detector pulses.

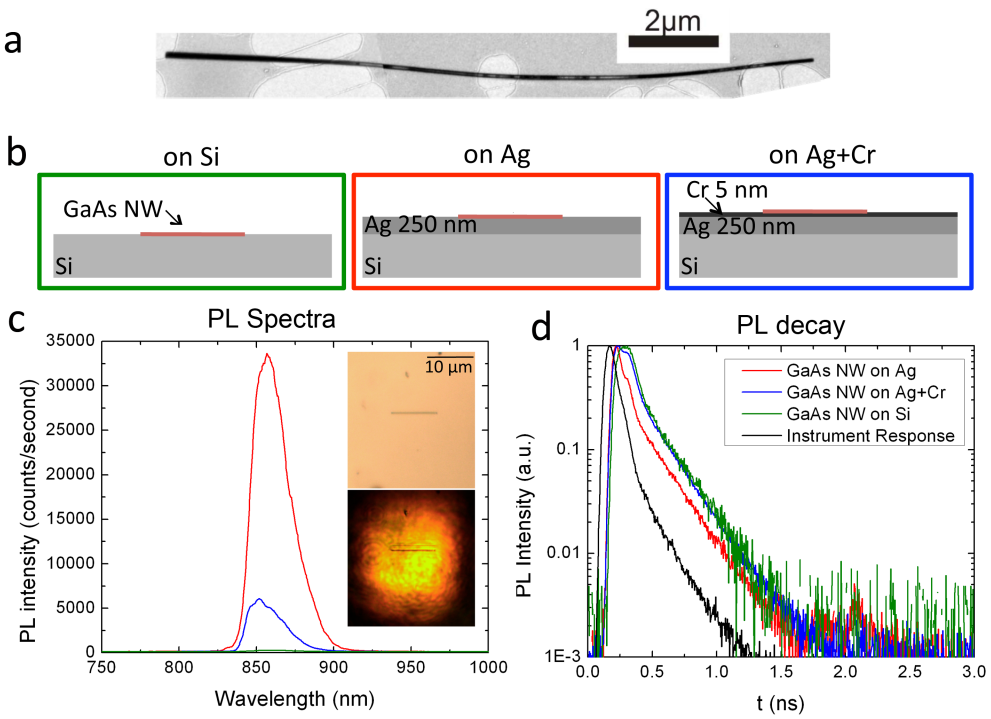

Figure 7.2. (a) TEM image of a single GaAs NW 15 μm long and tapered from 150 to 80 nm diameter. The nanowire core consists of a GaAs p-i-n radial junction clad with 8 nm $Al_{0.4}Ga_{0.6}As$ and a 2 nm GaAs cap. (b) Schematics of GaAs NWs dropcast on three different substrates: Si, Ag (250 nm) on Si, and Ag (250 nm) + Cr (5 nm) on Si. (c) PL intensity and (d) PL lifetime decay for single GaAs NWs on each substrate: Si (green), Ag (red), and Ag+Cr (blue), with the instrument response for decay measurements plotted in black. The PL spectra show a 5-fold enhancement in PL intensity for a NW on Ag when compared to a NW on Ag+Cr, and the PL decay shows a 3× decay rate enhancement for a NW on Ag compared to NWs on bare Si and Ag+Cr films. Insets in (c): (top) Bright-field reflected light image, and (bottom) laser spot focused on a single nanowire.

number of NWs. Because of absorption in the Si at both the pump wavelengths and at the PL emission wavelength, the emission intensity for the GaAs NW on Si is too low to plot alongside the others in Figure 7.2c. The PL spectra for GaAs NWs on the other two substrates, however, show a 5× enhancement in the emission intensity for a NW on Ag when compared to a NW on Ag+Cr. We expect the thin Cr layer to damp SPs while not significantly diminishing the reflectivity of the Ag, and therefore attribute the PL intensity enhancement to coupling into SPs. Furthermore, the PL decay traces in Figure 7.2d illustrate shorter PL lifetimes τ for a GaAs NW on Ag ($\tau = 43$ ps) compared to NWs on Si and Ag+Cr ($\tau = 125$ ps). These results encourage further experimental investigation of additional plasmonic nanostructures for enhancing spontaneous emission, a variety of which can be easily fabricated using FIB milling of metal films. We are currently pursuing this work with a goal of observing >100× enhancement in the emission rate of GaAs NWs.

Overall, this thesis has demonstrated that plasmonics enables truly nanophotonic technologies and provides a means of manipulating light at the nanoscale. The relatively young field of plasmonics has already seen rapid progression in the miniaturization of traditional optical components such as waveguides and resonators, achieved by judiciously designing metallodielectric structures that exhibit extremely high and localized field intensities and extremely short wavelengths. Though these early prototype devices are impressive in their own right, the true potential of plasmonics will be realized by exploiting the hybrid electronic/photonic characteristics of surface plasmons to create a new class of applications. From investigations of cathodoluminescence emission in plasmonic nanostructures, one can envision the development of an electrical surface plasmon source that could be integrated for chip-based nanophotonic networks or sensors. New plasmonic nanocavity designs that support dramatic spontaneous emission enhancements may enable nanoscale LEDs that are brighter and faster than chip-based laser sources. There are, of course, myriad opportunities, with the only limits being our imagination and determination to realize the promise of plasmonics.