Silicon nanoparticles are promising new materials for photovoltaic applications that combine materials property tunability on the nanoscale with silicon’s established performance in photovoltaics. We have succeeded in synthesizing crystalline silicon nanoparticles in a continuous flow plasma reactor and established control over particle size. By carefully tuning the plasma parameters, we have synthesized particles with radial precision of a couple atomic layers and characterized them structurally and optically. Such tight size control is the first step toward assembly of nanoparticles into a device layer to produce silicon-based PV devices with tunable bandgaps.

Si nanocrystals (NCs) less than 5 nm in diameter exhibit a size-dependent tunable band gap, visible photoluminescence (PL), and multiple exciton generation (MEG). These properties of Si NCs have led to an increased interest in their utilization in third-generation photovoltaic devices. In this project, we have synthesized Si NCs in the 3-7 nm range via nucleation and growth in a low-temperature SiH$_4$/Ar plasma. In these discharges, while the gas temperature is close to room temperature, the particles crystallize due to highly exothermic ion-electron and radical-radical recombination reactions on the surface: these reactions, in the absence of collisional cooling, lead to particle heating by several hundred degrees above the gas temperature. The NCs are transported by gas flow, and collected downstream onto a grid for transmission electron microscopy (TEM) and advanced optical characterization. TEM images (top left and middle figures) and the corresponding electron diffraction data indeed confirm that the particles are crystalline. The NC size can be varied by controlling the gas residence time in the plasma volume, power input to the plasma source, and power modulation of the plasma on the time scale of NC generation, which is a few milliseconds. Room temperature PL (top right panel) from ~7 nm NCs has an emission peak centered at 850 nm, which blue shifts to 650 nm as the crystal size decreases to ~3 nm (bottom left graph). Transmission infrared (IR) measurements show that the surface Si atoms of the as-synthesized NCs are H-terminated with monohydride, dihydride, and trihydride species; however the NCs rapidly oxidize over just a few minutes, which is also apparent from the IR data. The PL intensity from the oxidized NC is enhanced due to surface defect passivation with a corresponding blue shift as a result of a smaller Si core.

Our overall goal is to assemble nanoparticles into electrically well-coupled, efficient, thin film dot arrays. By controlling nanoparticle size, we will effectively produce a silicon based thin film with a bandgap that can be tuned to the solar spectrum. Nanoparticles within these arrays must be monodispersed in size for efficient carrier transport. Hence, the ability to control nanoparticle size, and, in particular, to obtain a tight size distribution (bottom right graph) is a critical first step to building up nanoparticle thin films.