NONTHERMAL PLASMA SYNTHESIS OF PHOTONIC SILICON NANOCRYSTALS

U. Kortshagen
University of Minnesota, Mechanical Engineering, 111 Church Street SE, Minneapolis, MN 55455, USA

Chemically reactive nonthermal plasmas at low pressure are an interesting medium for the growth of nanocrystals. Molecular precursors are dissociated by electron impact reactions and the resulting molecular fragments and radicals, many of them charged, nucleate to form clusters and nanocrystals. Energetic surface reactions heat these initial clusters to temperatures that exceed the gas temperature by hundreds of Kelvin.[1] This enables plasmas to form crystalline nanoparticles even of materials with very high melting points. In this presentation, we will discuss the plasma synthesis of silicon quantum dots with diameters on the order of ~3 nm for luminescent applications as well as the synthesis of larger silicon nanocrystals, 60-200 nm in diameter, that show interesting scattering behaviors. We also discuss how the temporary trapping of nanoparticles in the plasma contributes to the very narrow size distributions observed in both cases.[2]

Plasma-synthesized silicon quantum dots have shown good luminescence properties with photoluminescence quantum yields of up to 60%. One application of luminescent silicon quantum dots is in luminescent solar concentrators (LSCs), semitransparent waveguides that are doped with silicon quantum dots.[3] The silicon dots absorb solar radiation primarily in the blue range of the spectrum and reemit it in the near-infrared. Waveguiding concentrates this radiation on small solar cells that can be edge or surface mounted. We discuss results of a numerical model that investigates the application of silicon LSCs to greenhouses and demonstrate that LSC roof panels have the ability to enable net-zero-energy greenhouses in certain climates.[4]

We also discuss recent progress in the plasma synthesis of larger, highly monodisperse silicon nanocrystals with diameters of 60-200 nm.[5] These nanocrystals exhibit intriguing optical scattering through overlapping electric and magnetic dipole modes. We show that ensembles of silicon nanocrystals produced by plasmas show scattering behavior that is essentially consistent with single particle scattering models due to their very narrow size distribution.

This work was supported by the Army Research Office MURI grant W911NF-18-1-0240.

(1) Mangolini, L.; Thimsen, E.; Kortshagen, U. High-Yield Plasma Synthesis of Luminescent Silicon Nanocrystals. *Nano Letters* **2005**, *5* (4), 655–659. https://doi.org/10.1021/nl050066y.

(2) Xiong, Z.; Lanham, S.; Husmann, E.; Nelson, G.; Eslamisaray, M. A.; Polito, J.; Liu, Y.; Goree, J.; Thimsen, E.; Kushner, M. J.; Kortshagen, U. R. Particle Trapping, Size-Filtering, and Focusing in the Nonthermal Plasma Synthesis of Sub-10 Nanometer Particles. *Journal of Physics D: Applied Physics* **2022**, *55* (23), 235202. https://doi.org/10.1088/1361-6463/ac57de.

(3) Meinardi, F.; Ehrenberg, S.; Dhamo, L.; Carulli, F.; Mauri, M.; Bruni, F.; Simonutti, R.; Kortshagen, U.; Brovelli, S. Highly Efficient Luminescent Solar Concentrators Based on Earth-Abundant Indirect-Bandgap Silicon Quantum Dots. *Nature Photonics* **2017**, *11* (3), 177–185. https://doi.org/10.1038/nphoton.2017.5.

(4) Liu, Y.; Keil, J.; Ferry, V. E.; Kortshagen, U. R. Energy and Thermal Performance Analysis of Quantum Dot Luminescent Solar Concentrators in Greenhouses. *Advanced Sustainable Systems* **2023**, 2300107. https://doi.org/10.1002/adsu.202300107.

(5) Eslamisaray, M. A.; Wray, P. R.; Lee, Y.; Nelson, G. M.; Ilic, O.; Atwater, H. A.; Kortshagen, U. R. A Single-Step Bottom-up Approach for Synthesis of Highly Uniform Mie-Resonant Crystalline Semiconductor Particles at Visible Wavelengths. *Nano Lett.* **2023**, *23* (5), 1930–1937. https://doi.org/10.1021/acs.nanolett.2c05084.