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## Design of Si-based particles for infrared-active metamaterials

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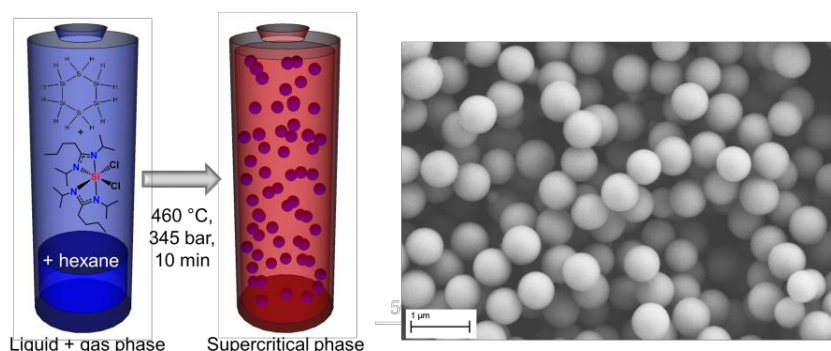
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**Abstract:** We report the synthesis and self-assembly of Si@SiO<sub>x</sub>N<sub>y</sub> core-shell particles, scattering infrared light. They were produced by decomposing a Si coordination complex alongside cyclohexasilane, under supercritical conditions. Core and shell dimensions were controlled through precursor stoichiometry and relative concentration. The electric and magnetic multipoles were characterized using a custom-built static light scattering spectrometer. Simulations show that the magnetic and electric responses are respectively located in the core and shell. The particles were self-assembled into metasurfaces and optically characterized using ellipsometry.

Core-shell particles were designed to achieve optical metamaterials with forward light scattering properties in infrared frequencies. Forward light scattering, referring to the light transmitted forward with respect to the incidence angle, with no reflection in the backward direction, is a property useful in light harvesting and anti-reflective screens. In a previous work we demonstrated that similar particles with reduced size (~350 nm) more efficiently supported forward light scattering, and over a broader region of the visible spectrum, than pure silicon particles.<sup>1</sup> Here, the scattering frequencies are shifted to the infrared, with the synthesis of larger particles (~550 nm) obtained from the use of a different silane precursor. The dimensions of the particles were tuned by varying the concentration of precursor during the synthesis. The mechanism involved in the particles' formation in supercritical n-hexane from silicon bisamidinate and cyclohexasilane are currently under investigation. The bottom-up approach (Figure 1) employed here led to particles fulfilling the requirements for efficient light scattering: being smaller than the wavelength of incident light and large enough to support Mie resonances at optical frequencies and being monodisperse in size.



**Figure 1.** Synthesis of core-shell particles supporting magnetic and electric dipole resonances at optical frequencies.

The optical response obtained from polarized resolved static light scattering<sup>2</sup> confirmed the presence of the electric and magnetic dipole resonances in the infrared light spectrum. Their overlap is a mandatory condition for broadband forward light scattering (first Kerker condition).<sup>3</sup> Previous theoretical simulations demonstrated that this condition is achieved in such particle structures with the magnetic dipole resonance located in the core, and the electric dipole resonance predominately localized in the shell in the forward direction.<sup>1,4</sup> The optical electric and magnetic dipolar resonances depend upon both the dimensions and the refractive index of the core and the shell.<sup>4-6</sup> Consequently, tailoring these parameters through the bottom-up preparation of Si@SiO<sub>x</sub>N<sub>y</sub> core-shell particles enabled tuning their optical response toward the superimposition of electric and magnetic dipole resonances with comparable intensities.

Furthermore, we pursued the study with the self-assembly of the particles onto 2D substrates resulting in metasurfaces. Dip-coating the particles from a suspension in high viscosity solvents (chloroform and isopropanol) allowed us to obtain monolayers over large surfaces. The optical properties of the collection of single scatterers were experimentally studied using ellipsometry, and the results were compared to simulations.

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