Enhanced LSPR absorption from dielectric anisotropy in Si nanowires

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We recently demonstrated that phosphorus-doped segments embedded along the length of Si nanowires support mid-infrared localized surface plasmon resonances (LSPRs) [1,2]. Control of doped segment aspect ratio and carrier density (> 10^{20} cm^{-3}) permits tuning of resonant frequency (700 – 2200 cm^{-1}). Here, we show that the 1-D geometry of nanowires, particularly the anisotropic dielectric environment that surrounds each embedded resonator, can substantially increase LSPR absorption cross-section and local field intensity. Si nanowires with a range of doped segment aspect ratios and phosphorus densities are synthesized via the vapor-liquid-solid (VLS) technique in a custom-built ultrahigh vacuum system. The spectral response of as-grown nanowire arrays is determined with *in-situ* transmission infrared spectroscopy and simulations within the discrete dipole approximation (DDA) provide a quantitative description of the underlying physics.

Figure 1 (left, middle) compares the experimental and simulated spectral response of Si nanowires containing phosphorus-doped segments with a range of aspect ratios. Whereas the transverse LSPR is always weak, the absorption cross-section for the longitudinal LSPR is strong and increases as a function of aspect ratio. A comparison with the simulations for an isotropic medium in Figure 1 (right) reveals that the anisotropic arrangement of the undoped regions in nanowires significantly enhances the polarization. As displayed in Figure 2, an investigation of alternative dielectric materials (e.g. ZnO, GaP, Ge, etc.) reveals that this effect scales with refractive index and that absorption cross-section enhancements of up to 35 times are possible relative to vacuum. Our findings show that an anisotropic dielectric medium offers a new route to engineer localized surface plasmon resonances and underscore the usefulness of the VLS technique for this purpose. We expect that this approach will be applicable to traditional and non-traditional plasmonic materials alike, but is predicated on the availability of syntheses that can yield the requisite structural anisotropy.

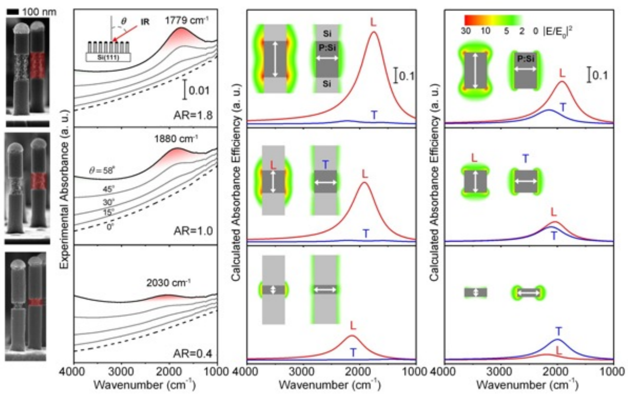

Figure 1: Experimental and simulated spectral responses of embedded (i.e. in nanowires) or isolated (i.e. in vacuum) Si resonators as a function of aspect ratio.

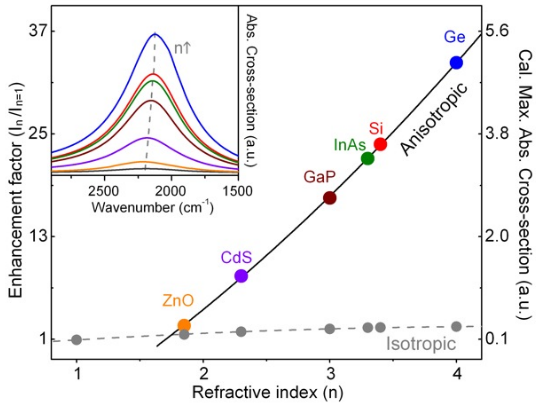

Figure 2: DDA simulations of absorption cross-section enhancement as a function of refractive index for resonators with an aspect ratio of 0.4.

[1] L.-W. Chou, N. Shin, S. Sivaram, M. Filler, *J. Am. Chem. Soc.,* **134**, 16155, (2012).

[2] L.-W. Chou and M. Filler, *Angew. Chem. Int. Ed.,* **52**, 8079, (2013)