Optical Response of High Refractive Index Nanoparticles with Metallic Impurities

J. M. Sanz, R. Alcaraz de la Osa, A. I. Barreda, J. M. Saiz, F. González, F. Moreno

Grupo de Óptica, Dpto. Física Aplicada, Universidad de Cantabria,
Avda. Los Castros s/n, 39005 Santander, Spain
sanzjm@unican.es

Abstract
The influence of the degree of purity of a high refractive index nanoparticle (silicon or other semiconductor material in the VIS-NIR) on its resonances, either electric or magnetic, is assessed by using Mie theory as well as finite-element simulations. In particular, it is shown that the main effect of the increase of absorption due to metallic pollutants is observed in the magnetic resonances. Concerning directionality conditions (zero backward/forward conditions) of electromagnetic scattering, it is found that they are spectrally shifted when the material's purity is varied. Resistive losses confirm the quenching of magnetic resonances, showing that the region of influence in the magnetic dipole resonance is much larger than in the electric one, although, for instance, for silicon, it has been found that losses are not critical when its purity is over 99.50%.

Introduction and Methods
The resonant response of nanoparticles as a reaction to an external electromagnetic field, makes them attractive for a wide range of applications, such as real-time sensors, surface enhanced Raman spectroscopy (SERS), light energy guiding, nanolensing or electromagnetically induced transparency, among others [1]. The theoretical behavior of small particles exhibiting both electric and magnetic dipolar resonances was predicted several decades ago [2]. These particles scatter with coherent effects between both dipoles, thus enabling control of the scattered radiation [3].

Currently, ohmic losses in metallic nanoparticles have turned attention to dielectric materials and, in particular, to high-permittivity low-loss nanoparticles and nanoparticle structures, for instance, made of silicon (Si) [4, 5, 6]. These researches have been done under the assumption that those high refractive index materials, apart from not having ohmic losses at all, they do not contain any sort of contamination, i.e. they have a purity of 100 %. However, materials with 100% of purity are non-realistic cases since industry can provide pure materials under certain conditions only and massively increasing manufacturing costs. From an industrial point of view, pure Si can be obtained with metallurgical-grade (MG-Si, < 98%) and upgraded up to solar-grade (SG-Si, > 99.999%) [7] with the corresponding increased price.

In this research, Si particles of size smaller than the incident wavelength are studied. Specifically, the influence of the material's degree of purity on both electric and magnetic resonances is analyzed. The main metallic contaminants in Si powder are usually metals, like Fe, Al and Ti [7, 8]. In order to evaluate their influence, the composition of an industrial sample of Si powder (provided by Elkem [8]) was taken as a starting point, varying the Si content of the particle from 99.0% to 100.0%. The resulting dielectric function was obtained by performing a weighted average, by simply taking into account the relative concentration of each pollutant and their optical constants, which, due to their metallic nature, show important absorption losses. Near- and far-field computations were done by using Mie theory [9] as well as finite-element simulations [10].

Results and Discussion
Fig. 1 shows the spectral extinction (top) and absorption (bottom) efficiencies (λ in the interval [900; 2100] nm) for an isolated spherical particle of Si (radius a=230 nm) as the purity grade is reduced from 100.0% to 98.98%. Obviously, the Si fingerprint can be observed as an absorption peak for multipolar resonances in the visible region. The well-known spectral pattern of a pure Si sphere is distorted when the purity grade is reduced. Initially (for high purity grades), this effect only slightly affects magnetic resonances (b_1 and b_2 peaks) but it is spread through the entire spectrum when the purity grade is further reduced. Accordingly, absorption efficiency peaks (especially the magnetic ones) grow and broaden when purity decreases, with the overall absorption efficiency background rising as well.

Fig. 2 shows maps of the norm of the electric near-field and resistive losses of the magnetic (top) and electric (bottom) dipole resonances at the wavelength they occur for several purity grades. As can be seen, the electric dipole resonance only shows slight changes, even for low purity grades. On the other hand, the magnetic dipole resonance dramatically changes for Si purity grades < 99.50%. Resistive losses summarize these two situations: while the maximum value of the losses is similar in both dipolar resonances, the area of influence in the magnetic dipole resonance is much larger than in the electric one.

Acknowledgements

The authors thank the United States Army International Technology Center Atlantic (USAITC-A) for financial support through grant W911NF-13-1-0245. We also thank Elkem [8] for providing Si data and samples.

References

[1] P. N. Prasad, Nanophotonics. John Wiley & Sons, Inc., 2004.

[2] M. Kerker, D.-S. Wang, and C. L. Giles, "Electromagnetic scattering by magnetic spheres," J. Opt. Soc. Am., **73**-6 (1983) 765.

[3] J. M. Geffrin, B. García-Cámara, R. Gómez-Medina, et al., "Magnetic and electric coherence in forward-and back-scattered electromagnetic waves by a single dielectric subwavelength sphere," Nat. Commun., **3** (2012) 1171.

[4] L. Shi, J. T. Harris, R. Fenollosa, et al., "Monodisperse silicon nanocavities and photonic crystals with magnetic response in the optical region," Nat. Commun., **4** (2013) 1904.

[5] A. Garcia-Etxarri, R. Gómez-Medina, L. S. Froufe-Perez, et al., "Strong magnetic response of submicron silicon particles in the infrared," Opt. Express, **19**- 6 (2011) 4815.

[6] Y. H. Fu, A. I. Kuznetsov, A. E. Miroshnichenko, et al., "Directional visible light scattering by silicon nanoparticles," Nat. Commun., **4** (2013) 1527.

[7] N. Yuge, M. Abe, K. Hanazawa, et al., "Purification of metallurgical-grade silicon up to solar grade," Progress in Photovoltaics: Research and Applications, **9**-3 (2001) 203.

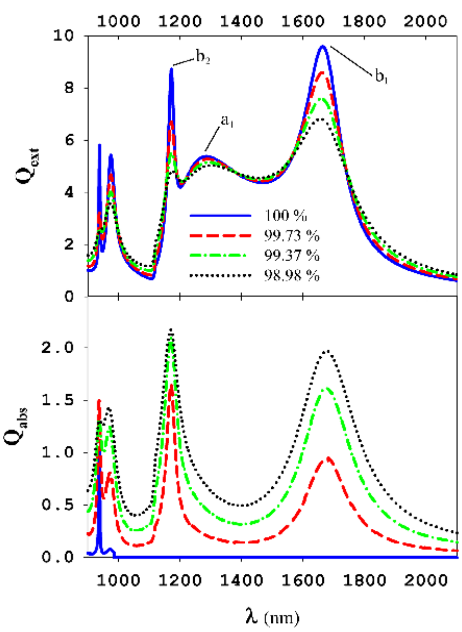
[8] Elkem AS Silicon Materials: http://www.elkem.com/Global/ESM/qualitysafety/product-data-sheets/siliconproducts/silgrain/Silgrain-pds.pdf.

[9] C. Bohren and D. Huffman, Absorption and Scattering of Light by Small Particles, 2nd ed. Wiley-VCH, 2010.

[10] Comsol Multiphysics 4.3a, Comsol Inc., Burlington, MA.

Figures

1-



2-

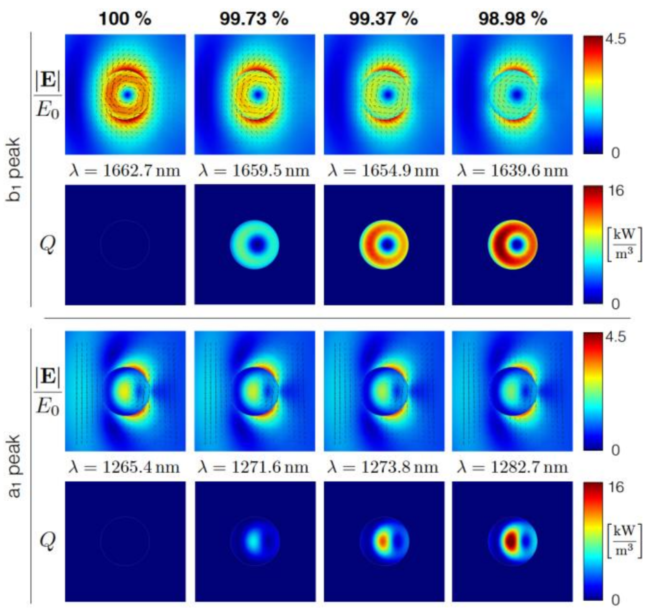


Fig. 1: Spectral extinction (top) and absorption (bottom) efficiencies (λ in [900; 2100] nm) for an isolated spherical particle of Si (radius *a* = 230 nm) as the purity grade is reduced from 100.0% to 98.98%.

Fig. 2: Maps of the near-field norm and resistive losses of the magnetic (top) and electric (bottom) dipole resonances at the wavelength they occur for several purity grades.