Nanoplasmonics from large-scale *ab initio* calculations: opposite trends in Ag and Na clusters

Marc Barbry[1], Peter Koval[1,2], Natalia E. Koval[1], Javier Aizpurua[1], Daniel Sánchez-Portal[1,2]

[1]Material Physics Center, San-Sebastian, Spain
[2]Donostia International Physics Center, San-Sebastian, Spain
marc.barbry@mailoo.org

An accurate description of electronic excitations is indispensable for understanding material properties and designing nanoscale devices. For instance, using large-scale TDDFT calculations, we have recently demonstrated the importance of taking into account the details of the atomic-scale structure [1] and the quantization of electron transport [2] in metal nanostructures in order to accurately describe their plasmonic properties. In this contribution we will compare the surface plasmon resonance of sodium and silver clusters within the same framework of iterative TDDFT [3]. Recent progress in our implementation made it possible to perform calculations of large clusters of diameters ranging from a few Å to 4–5 nm (Figure 1), counting up to 5000 silver atoms and using only modest computational resources (a 32-core node with 500GB RAM). With these new capabilities, we have characterized the size-scaling of the SPR frequency for both sodium and silver clusters. As expected these two materials show opposite behaviors that can be related to the different spill out of charge at the surface and to the additional screening created by the 4d electrons in silver.

References

[1] M. Barbry et al. Nano Letters, 15 (2015) 3410.
[2] F. Marchesin et al, ACS Photonics, 3 (2016) 269.
[3] P. Koval et al. J. Phys.: Condens. Matter, 28 (2016) 214001.

Figures

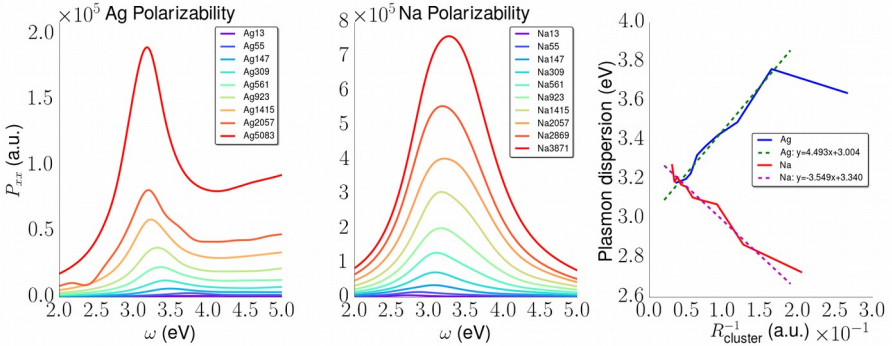

Figure 1: First and second column: optical polarizability for icosahedral silver and sodium clusters of different sizes. Third column: plasmon dispersion for silver and sodium cluster from ab-initio calculations, the dashed lines show the linear interpolations.