

Spatial Nonlocality in the Optical Response of Metal Nanoparticles

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Experimental access to particle sizes and interparticle spacings of below 10 nm is now available for metallic dimers [1], tips [2], and shell structures [3], in which spatial dispersion (nonlocality) in the materials response is known to play an important role. However, most electromagnetic calculations of nanostructures rely on local, frequency-dependent dielectric functions [4], sometimes incorporating a phenomenological damping to account for finite-size effects [5].

Spatial nonlocality is known to play an important role at distances of a few nanometers [6] leading to significant plasmon broadening and blue shift [7], but few efforts have been made to theoretically investigate nonlocal effects in a rigorous way. The optical response of nanosized metallic structures is greatly influenced by quantum confinement of their conduction electrons, which adds up to the intrinsic nonlocality in the response of homogeneous bulk media.

In this talk, we present two different approaches to account for nonlocality in metal nanoparticles: (a) the non-retarded specular reflection model (SRM) [8-9] and (b) the retarded hydrodynamical model [10-11]. Comparison with available experiments results in excellent agreement with our parameter-free modeling of nonlocal effects, which produce dramatic changes with respect to the customary local theory. We show that nonlocal effects in both models produce sizable plasmon blue shift and broadening in single metal nanoparticles as well as in dimers (see Fig. 1).

Analysis of the plasmon resonance dependence on the interparticle spacing and nanoparticle size allows us to separate nonlocal and retardation effects, but there are common conditions for which both of them coexist, giving rise to competing mechanisms of field enhancement and mode displacement. This study is particularly relevant for broad, active areas involving applications of local field enhancement to biosensing and nonlinear optics.

References

- [1] M. Danckwerts, L. Novotny *Phys. Rev. Lett.* **98** (2007), 026104.
- [2] P. S. Kumar, I. Pastoriza-Santos, B. Rodríguez-González, F. J. García de Abajo, L. M. Liz-Marzán, *Nanotechnology* **19** (2008), 015606.
- [3] E. Prodan, C. Radloff, N. J. Halas, P. Nordlander, *Science* **302** (2003), 419–422.
- [4] V. Myroshnychenko, J. Rodríguez-Fernández, I. Pastoriza-Santos, A. M. Funston, C. Novo, P. Mulvaney, L. M. Liz-Marzán, F. J. García de Abajo, *Chem. Soc. Rev.* **37** (2008), 1792–1805.
- [5] U. Kreibig, C. v. Fragstein, *Z. Physik* **224**, (1969) 307–323.
- [6] U. Kreibig, M. Vollmer, *Optical Properties of Metal Clusters*, Springer-Verlag, Berlin, 1995.
- [7] J. Renger, R. Quidant, N. van Hulst, L. Novotny, *Phys. Rev. Lett.* **104** (2010), 046803.
- [8] R. H. Ritchie, A. L. Marusak, *Surf. Sci.* **4** (1966), 234–240.
- [9] F. J. García de Abajo, *J. Phys. Chem. C.* **112** (2008), 17983–17987.
- [10] F. Bloch, *Z. Phys.* **81** (1933), 363–376.
- [11] F. J. García de Abajo, *Rev. Mod. Phys.* **82** (2010) 209–275.

Figures

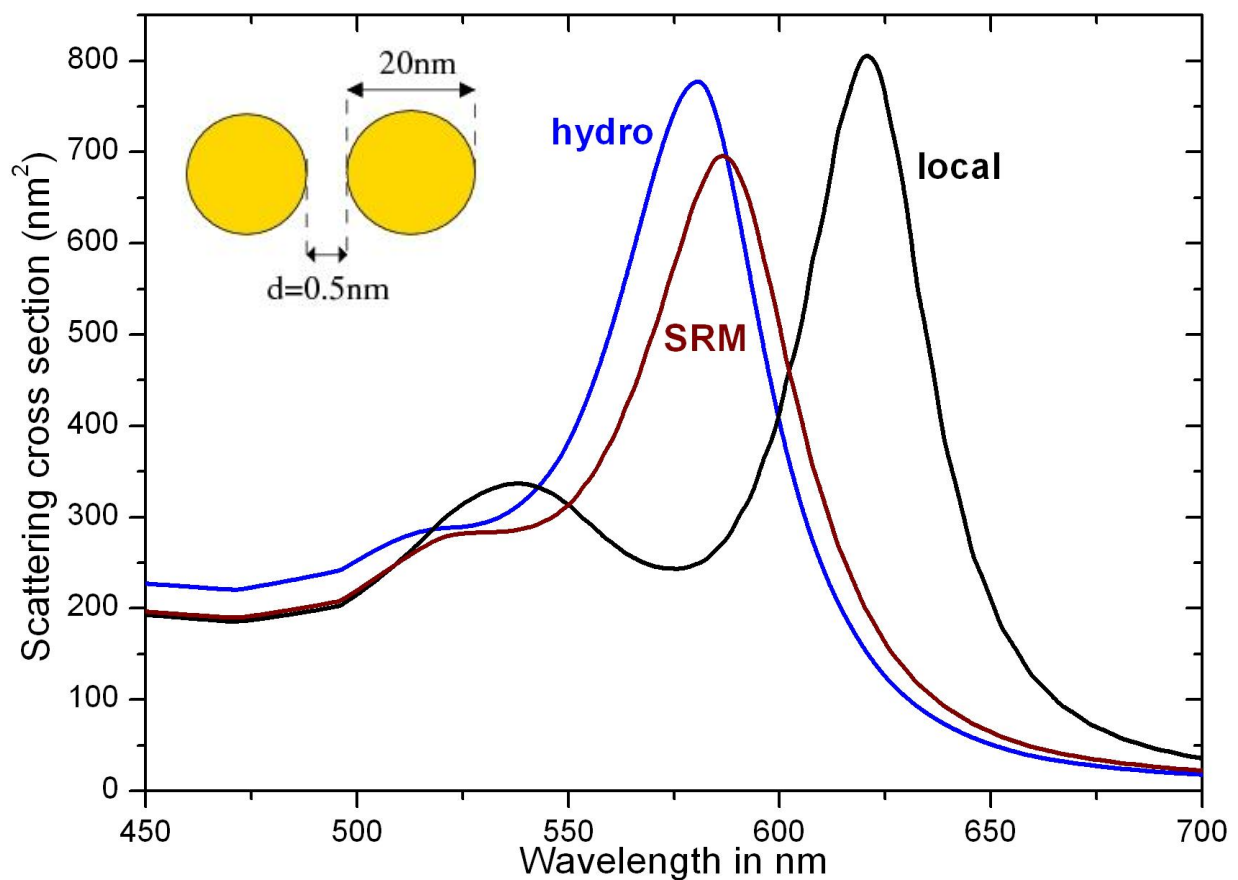


Figure 1. Extinction spectra for a gold dimer with 0.5 nm spacing between Au spherical particles of 20 nm in diameter. The external field is parallel to the interparticle axis. Local and nonlocal calculations are compared. Nonlocal calculations are presented both using the specular reflection model (SRM) and the hydrodynamic model (hydro).