Electronic Supplementary Material (ESI) for Nanoscale.

Boosting Fano Resonances in Single Layer Concentric Core-Shell Particles: Supplementary Information

Jordi Sancho-Parramon* and Denis Jelovina**

*Rudjer Boskovic Institute, Bijenicka cesta 54, Zagreb 10000, Croatia
**Chemistry Department, Faculty of Sciences, Universidad Autónoma de Madrid, Ciudad Universitaria de Cantoblanco, 28049 Madrid, Spain

August 15, 2014

1 Electromagnetic model for Fano resonances applied to the scattering spectra of core-shell particles

The asymmetric line-shape that was deduced by Ugo Fano in his original study of atomic systems assumed the existence of a continuum and a discrete state [1, 2]. This model can qualitatively describe the Fano resonances in plasmonic systems but may be inaccurate for a quantitative description. Gallinet and Martin proposed an ab-initio model for electromagnetic Fano resonances that considers the interference of a continuum of radiative waves and a non-radiative mode that spectrally overlap [3]. The interference between light radiated by direct excitation of the continuum and by the indirect excitation of the dark state through the excitation of the continuum results in an asymmetric line-shape (σ_A). The continuum of radiative waves (σ_S) is assumed to have a symmetric Lorentzian line-shape. The response of the system is then given by product of both asymmetric and symmetric resonances:

$$\sigma = \sigma_S\sigma_A = \frac{a^2}{\left(\frac{\omega^2-\omega_S^2}{2W_S\omega_S}\right)^2 - 1} \frac{\left(\frac{\omega^2-\omega_A^2}{2W_A\omega_A} + q\right)^2 + b}{\left(\frac{\omega^2-\omega_A^2}{2W_A\omega_A}\right)^2 + 1} \quad (1)$$

where ω is the light frequency, ω_S and ω_A the central frequency of the symmetric and asymmetric resonances and W_S and W_A their respective widths. The parameter a is the amplitude of the symmetric resonance. The line-shape of σ_A is characterized by two parameters: the asymmetry parameter q, closely connected to the original Fano parameter and the modulation damping parameter b, connected to the absortion losses of the system b. This model has been succesfully applied to a wide range of plasmonic structures showing Fano resonances [4] including dolmen-like structures, oligomers, grating and metallic photonic crystals. It has been also applied to describe the scattering properties of multilayer core-shell systems [5]. Figure 1 shows the fits of the scattering spectra of the dielectric-core metal-shell particles described in Fig.1 of the manuscript for different values of the core radius r_c. A good fitting of the spectra is achieved in all the cases, although the fitting quality decreases for large values of r_c, i.e., when hybridization becomes strong and the spectral overlapping between the bonding and anti-bonding modes decreases. Table 1 presents the model parameter values resulting from the data fit. The absolute value of q increases with r_c while the value of b is minimum for a small value of r_c. Hence, the the characteristic Fano line-shape is best observed for moderate values of r_c.

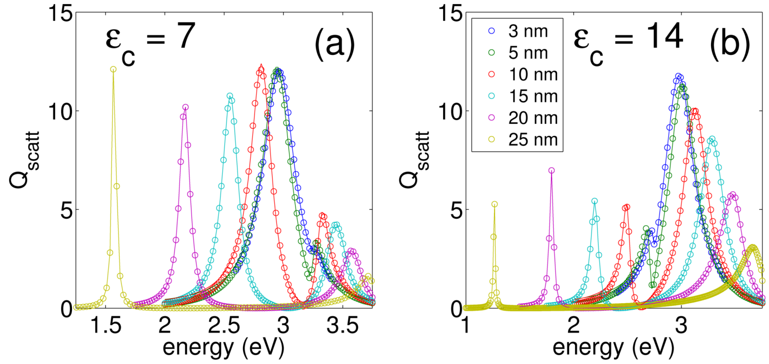

Figure 1: Scattering efficiencies (symbol) and fit to the electromagnetic model for Fano resonances described by Equation 1 (solid lines) for silver-shell dielectric-core particles embedded in a water-like environment ($\epsilon_m = 1.77$) with $r_s = 30$ nm for $\epsilon_c = 7$ (a) and $\epsilon_c = 14$ and different values of r_c

Table 1: Parameter optimal values of the model described by Equation 1 resulting from the fitting of the scattering spectra shown in Fig.1

ϵ_c	r_c (nm)	ω_A (eV)	W_A (eV)	ω_S (eV)	W_S (eV)	a	b	q
7	3	3.281	0.04196	2.965	0.1611	3.537	1.198	0.1167
	5	3.259	0.03447	2.950	0.1475	3.730	0.9498	0.5331
	10	3.315	0.0650	2.822	0.1087	4.872	0.8829	1.996
	15	3.434	0.0887	2.558	0.0797	5.918	3.541	4.157
	20	3.570	0.1015	2.169	0.0512	7.283	12.95	7.701
	25	3.685	0.1055	1.573	0.017	16.88	10.48	14.14
14	3	2.753	0.0195	2.981	0.1655	3.472	0.7137	-0.09781
	5	2.706	0.0263	3.005	0.1556	3.560	0.4561	-0.6224
	10	2.503	0.04099	3.122	0.1374	3.679	1.048	-2.288
	15	2.203	0.03811	3.280	0.1299	3.295	10.38	-3.787
	20	1.796	0.02099	3.457	0.1348	2.49	81.69	-4.282
	25	1.264	0.008461	3.650	0.1848	1.689	149.4	-2.223

2 Scattering of core-shell particles with gold-shell

In the core-shell systems studied in the manuscript, Ag has been used as the metal of choice. The reason is that Ag presents the smallest losses and interband transitions among noble metals and thus more clearly show the appearance of FR. However, gold is the primary choice for many metallo-dielectric core-shell systems [6, 7, 8, 9]. Figure 2 shows the extinction, scattering and absorption efficiencies for a particle similar to the one studied in Fig. 1 of the manuscript but Au is taken as shell material instead of Ag. The FR are significantly weaker than in the case of Ag. Figure 3 shows how increasing of the particle size improves the excitation efficiency of the FR and leads to excitation of higher order FR for large enough particles. In these simulations, the dielectric function of the core has been set to $\epsilon_c = 9$, that correspons to a value comparable to core-shell particles that have been already fabricated [8, 9].

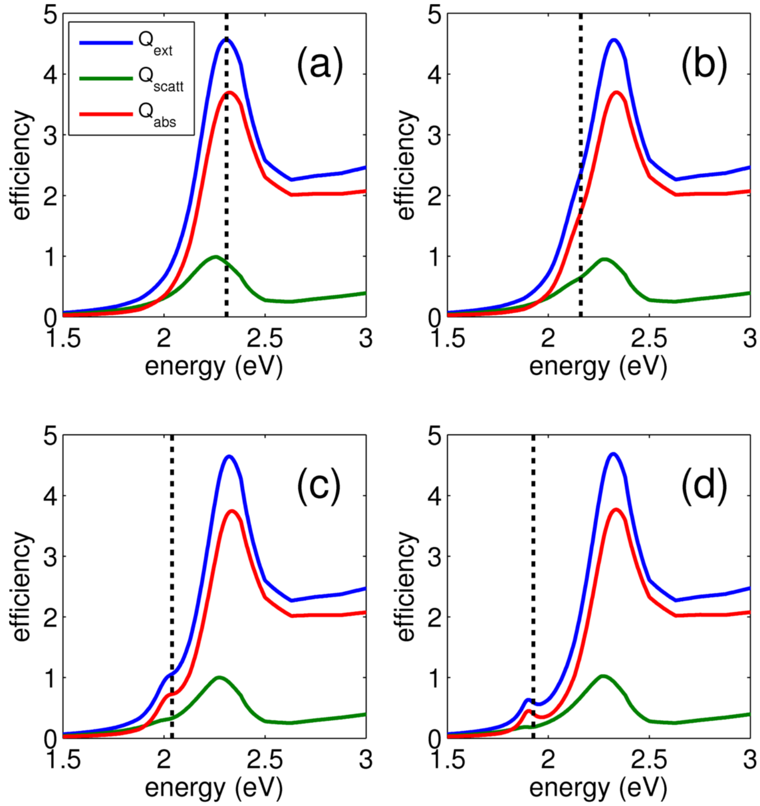

Figure 2: Extinction, scattering and absorption efficiency of a gold-shell dielectric-core particle embedded in a water-like environment with $r_s = 30$ nm for $\epsilon_c = 10$ (a), $\epsilon_c = 15$ (b), $\epsilon_c = 20$ (c) and $\epsilon_c = 25$ (d). Optical constants of gold are taken from Johnson and Christy data. The position of the anti-bonding cavity-like mode is given by the Frölich condition $\epsilon_c = -2\epsilon_s$ and is indicated by vertical dashed lines. Due to the large absorption losses in gold, the excitation of FR can be only observed for large values of ϵ_c. In this case, the resonance take place at lower photon energies where Au absoption is lower.

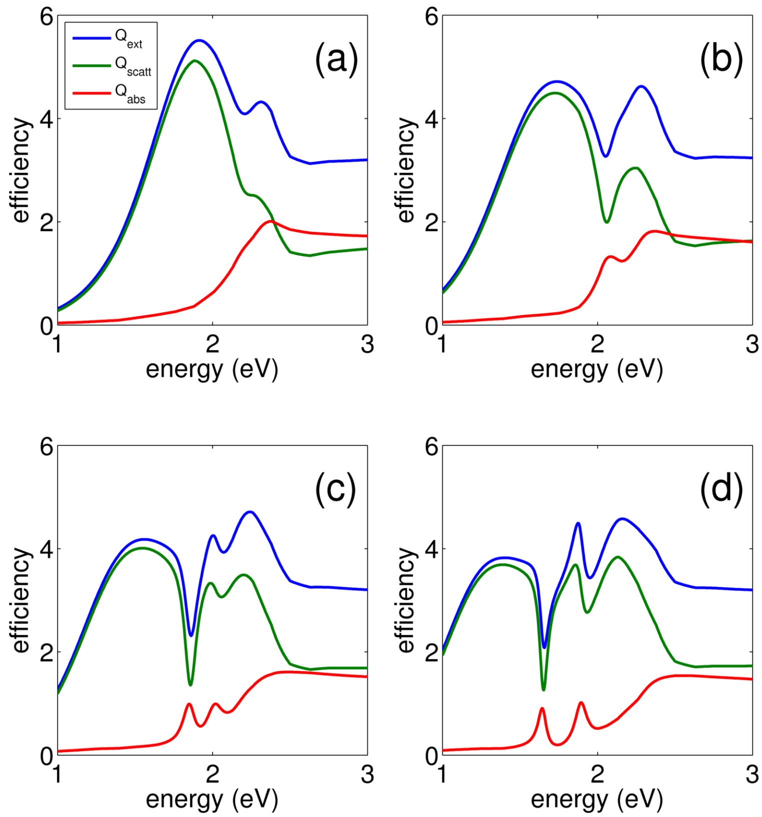

Figure 3: Extinction, scattering and absorption efficiency of a gold-shell dielectric-core particle embedded in a water-like environment with $\epsilon_c = 9$ for $r_c = 25$ nm, $r_s = 75$ nm (a), $r_c = 40$ nm, $r_s = 90$ nm (b), $r_c = 55$ nm, $r_s = 105$ nm (c) and $r_c = 70$ nm, $r_s = 120$ nm (d). As particle size increases, the cavity mode shifts to lower energies where absorption is lower and the FR becomes more pronounced. Quadrupolar FR can be observed for the largest particles.

References

[1] Boris Luk'yanchuk, Nikolay I Zheludev, Stefan A Maier, Naomi J Halas, Peter Nordlander, Harald Giessen, and Chong Tow Chong. The fano resonance in plasmonic nanostructures and metamaterials. *Nature Materials*, 9(9):707–715, 2010.

[2] Andrey E Miroshnichenko, Sergej Flach, and Yuri S Kivshar. Fano resonances in nanoscale structures. *Reviews of Modern Physics*, 82(3):2257, 2010.

[3] Benjamin Gallinet and Olivier JF Martin. Ab initio theory of fano resonances in plasmonic nanostructures and metamaterials. *Physical Review B*, 83(23):235427, 2011.

[4] Benjamin Gallinet and Olivier JF Martin. Influence of electromagnetic interactions on the line shape of plasmonic fano resonances. *ACS Nano*, 5(11):8999–9008, 2011.

[5] Ovidio Peña-Rodríguez, Antonio Rivera, Mariano Campoy-Quiles, and Umapada Pal. Tunable fano resonance in symmetric multilayered gold nanoshells. *Nanoscale*, 5(1):209–216, 2013.

[6] Carly S Levin, Cristina Hofmann, Tamer A Ali, Anna T Kelly, Emilia Morosan, Peter Nordlander, Kenton H Whitmire, and Naomi J Halas. Magnetic-plasmonic core-shell nanoparticles. *ACS Nano*, 3(6):1379–1388, 2009.

[7] Jiunn-Woei Liaw, Huang-Chih Chen, and Mao-Kuen Kuo. Plasmonic fano resonance and dip of au-sio2-au nanomatryoshka. *Nanoscale Research Letters*, 8(1):1–8, 2013.

[8] Rizia Bardhan, Nathaniel K Grady, Tamer Ali, and Naomi J Halas. Metallic nanoshells with semiconductor cores: Optical characteristics modified by core medium properties. *ACS Nano*, 4(10):6169–6179, 2010.

[9] Shaunak Mukherjee, Heidar Sobhani, J. Britt Lassiter, Rizia Bardhan, Peter Nordlander, and Naomi J. Halas. Fanoshells: Nanoparticles with built-in fano resonances. *Nano Letters*, 10(7):2694–2701, 2010.