Contribution ID: **99** Type: **Invited Talk**

Nonlinear plasmonic nanostructures for quadratic nonlinear optics

Thursday, November 7, 2024 1:00 PM (30 minutes)

We report the quadratic nonlinear optical responses of noble metal nanoparticles with various size and shapes, and their corresponding hyperpolarizability β values as measured via the Harmonic Light Scattering technique at 1.064 μm.
First we investigate the influence of surface area of gold and silver nanospheres and nanorods on their β values, and explore the validity limit of their purely dipolar origin. By studying gold and silver nanorods with different surface areas, we evidence the predominance of these surface effects over shape factors.
Second, we report the synthesis and characterization of platinum "nanoflowers"(PtNFs)with different sizes. These PtNFs display exceptionally strong first hyperpolarisabilities, but particle surface corrugation is shown to govern these huge β values, with a very limited contribution from plasmonic effects.
In a third part, we synthesize and investigate the β values of non-centrosymmetric gold nanoprisms (NPrs). Their β's not only display a linear dependence with surface area but also strongly depend on the sharpness of NPr corners. Their very high values are assigned mainly to the enhancement of electromagnetic fields due to geometrical effects (sharp extremities). This phenomenon dominates over centrosymmetry breaking. These results open the way to the investigation of various nonlinear noble metal nanopolyhedra families.

Type of presence

Presence online

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Session Classification: Advances in Nonlinear Optics and Laser-Matter Interactions