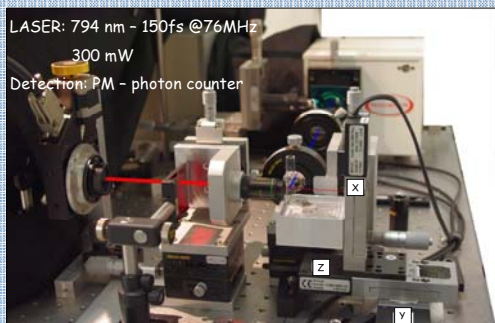


SECOND HARMONIC GENERATION FROM A SINGLE METALLIC NANOPARTICLE IN A HOMOGENEOUS MEDIA

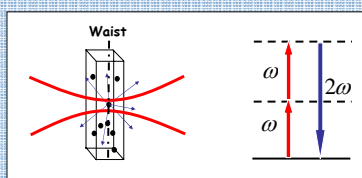


J. Butet, J. Duboisset, G. Bachelier, I. Russier-Antoine, E. Benichou, Ch. Jonin, P. F. Brevet
 Laboratoire de Spectrométrie Ionique et Moléculaire, UMR CNRS 5579, Université Claude Bernard Lyon 1

Experimental setup

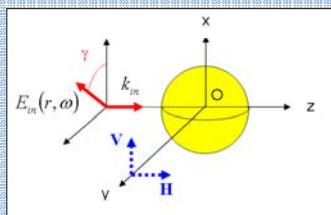


What is SHG?



Two photons at the fundamental frequency are converted in one photon at the harmonic frequency.

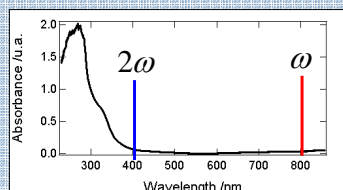
Scattering geometry



Harmonic frequency collected perpendicular to the incident beam

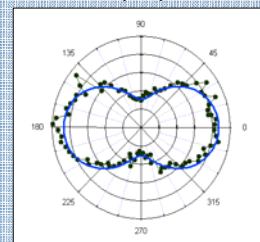
Optical properties of the gelatin matrix

Absorbance spectrum



Transparent at both fundamental and second harmonic wavelengths

Vertically polarized SHG intensity as a function of γ the input polarization angle



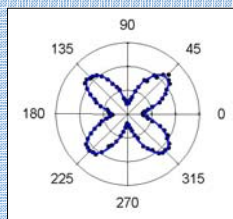
Dipolar term

Hyper Rayleigh Scattering from gold nanoparticles 150 nm

Ensemble measurement in solution

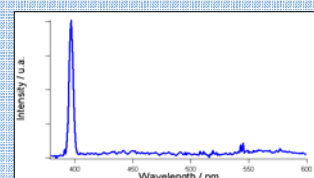
Average on size and shape distribution

Vertically polarized HRS intensity as a function of γ



Quadrupolar source

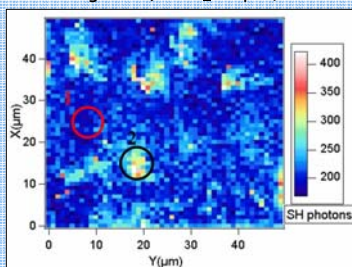
SH emission spectrum



SH signal, not two-photon fluorescence

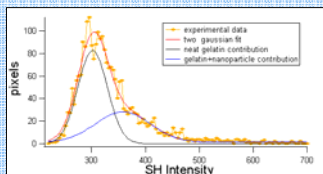
3D imaging of 150 nm gold nanoparticles dispersed in 3D homogeneous gelatin matrix

The sample scan is carried out perpendicularly to the incident beam direction collecting the SHG signal at $\gamma=45^\circ$. Step: 1 μ m and Time: 8s/pixel



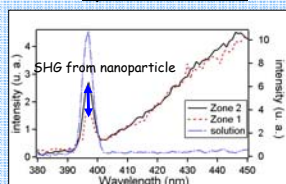
1. No nanoparticle 2. a single gold nanoparticle

Counts distribution of the first map



The SH signal intensity is a function of the distance of the nanoparticle from the beam waist along the beam propagating direction.

Spectrum emission

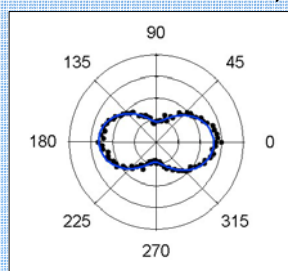


The SH signal from zone 1 is more intense than the one from zone 3. The increase in SHG intensity corresponds to a single nanoparticle with $\beta_{SHG} = 1.0 \pm 0.15 \times 10^{-23}$ as expected (see J. Duboisset and *al.*)

The multi-excited luminescence comes from the gelatin matrix

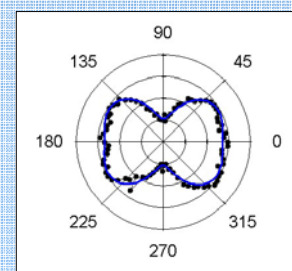
SHG from a single gold nanoparticle

Vertically polarized SHG intensity from zone 3 as a function of γ



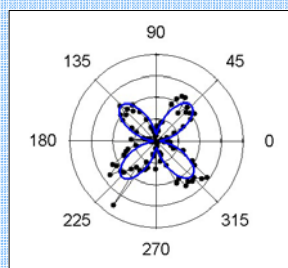
Similar to the graph obtained for neat gelatin

Vertically polarized SHG intensity from zone 1 as a function of γ

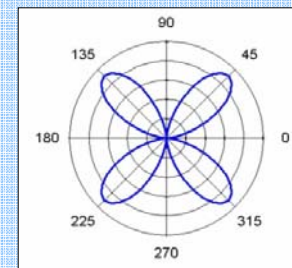


Vertically polarized SHG intensity from a single nanoparticle as a function of γ

Gelatin signal is rejected by subtraction:



Signal zone 1 - Signal zone 2



FEM calculation

Quadrupolar source as expected

Ref. :- R. Jin, J. E. Jureller, H. Y. Kim, and N. F. Scherer, "Correlating second harmonic optical responses of single Ag nanoparticles with morphology", *J. Am Chem Soc.*, **127**, 12482-12483 (2005)

- I. Russier-Antoine, E. Benichou, G. Bachelier, C. Jonin and P.F. Brevet, "Multipolar contributions of second harmonic generation from silver and gold nanoparticles", *J. Phys. Chem. C*, **111**, 9044-9048 (2007)
- J. Duboisset, I. Russier-Antoine, G. Bachelier, E. Benichou, C. Jonin and P.F. Brevet, "Single metallic nanoparticle sensitivity with Hyper Rayleigh Scattering", *J. Phys. Chem. C*, **113**, 13477-13481 (2009)