Nonlinear Optical Response of Nanoparticle Single Layers: Polarization and Structural Effects

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Metallic nanoparticles embedded in a dielectric matrix are simple structures which combine the ease of preparation with interesting nonlinear response, waveguiding effects and optical switching applications. In this work the linear and nonlinear optical response of Gallium-doped glasses are investigated. Gallium is adopted as the metallic nanoparticle constituent due to its high second-order nonlinear response [1] and to the possibility of driving a phase transition between the solid and the liquid state in the range from liquid nitrogen to room temperature. This possibility has been in effect exploited in macroscopic Ga aggregates proposed as optical switching elements [2]. Here we focus our attention on the nonlinear response at the second harmonic generated (SHG) frequency as obtained by impinging with femtosecond high repetition-rate laser pulses on mono- and multi-layer structures.

The composite structures are prepared by evaporation-condensation (Vollmer-Weber mode) of high purity Gallium on a dielectric transparent matrix film (SiO_x, TiO_2, MgF_2). The nanoparticles are solidified at low temperatures and then covered with a protective additional matrix layer. The metal doped film is supported on an optically transparent substrate (SiO_2 or sapphire). Nanoparticle average radii are ranging between 6 and 80 nm. The samples are carefully characterized by linear reflectance and transmittance measurements that allow to control the surface plasma resonance response in the visible spectral region and by atomic force microscopy as a feedback for size and shape effects on the nonlinear response.

The nonlinear response has been excited by femtosecond laser pulses in the 780 – 820 nm range so that SHG frequency is resonantly coupled to the surface plasma linear absorption. This causes the SHG to be drastically enhanced as already evidenced in the solid and liquid states and as predicted in a nonlinear Mie theory [3].

When these favorable conditions for resonance are met we measure the SHG response of Ga monolayers by varying sample orientation around the surface normal (azimuth) at different angles of incidence. A strong and regular anisotropy results from these scans, apparently determined by the nanoparticle shape and orientation. Of relevant interest is the polarization dependence of the SHG, when input and output polarizations are properly selected and are measured as a function of the angle of incidence. *p* polarization results to be favoured over the *s* one, and both are dominant over the substrate and pure matrix material response. This fact points to a polarization activity induced by the metallic nanostructures which may turn out of practical use.

By properly combining the two effects of nonlinear nanoparticle activity and of effective-medium dielectric properties one can tune the Ga/matrix layer thicknesses in a multilayer structure in a search for phase matching conditions in SHG. To this aim Ga nanoparticles have been grown in different matrix materials, whose dielectric function has been checked by spectroscopic ellipsometry. The size dependence of SHG has been measured and different resonant behaviors have been found to depend on the host matrix material. The multilayer structures have been designed and realized in Ga/SiO_2 and amplification of the nonlinear signals has been measured versus number of periods.

[1] G.T. Boyd *et al.,* Phys. Rev. B **30**, 519 (1984).
[2] K.F. MacDonald *et al.*, Appl. Phys. Lett. **82**, 1087 (2003).
[3] A.M. Malvezzi *et al.*, Phys. Rev. Lett. **89**, 087401 (2002).

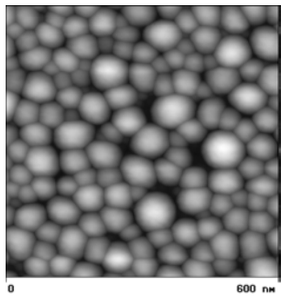

Figure 1. AFM image of a monolayer of Ga nanoparticles of average radius on 20 nm embedded in a silica matrix.

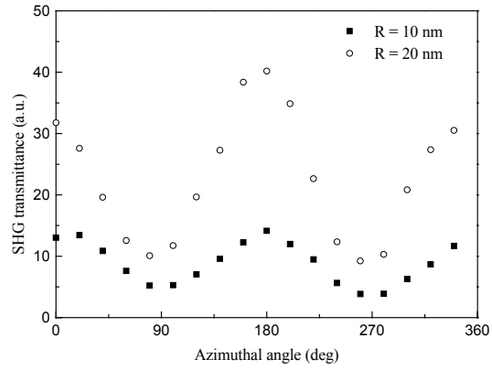

Figure 2. Azimuthal dependence of SHG transmitted signal as detected in two different samples of Ga monolayers embedded in TiO_2 matrix.

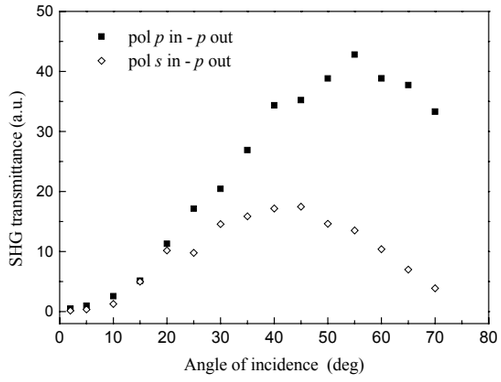

Figure 3. *p* polarized component of transmitted SHG as a function of the angle of incidence for both *s* and *p* polarized pump beam as measured on Ga nanoparticle monolayers (R = 30 nm) in TiO_2 matrix.