Temporal and Spectral Eependence of the Nonlinear Optical Properties of $Au:Al_2O_3$ and $Cu:Al_2O_3$ Composite Films

H. Fernández, R. Del Coso, J. Solis, J. Gonzalo, and C. N. Afonso
Instituto de Óptica, CSIC, Spain

Third order optical non-linearities in metal-dielectric nanocomposites have received considerable attention over the last years. These nanocomposites show large third order susceptibilities in the vicinity of the surface plasmon resonance with ultrafast build-up times. These characteristics make them promising for several applications in the field of information technologies like all-optical switching, signal regeneration or high speed demultiplexing [1]. Particularly, a very large third order nonlinear optical susceptibility has been recently reported for Cu nanocomposites near the percolation threshold that has been related to the appearance multiple particle interactions and giant local field enhancement effects [2].

The aim of this work is to investigate the spectral and temporal dependence of the third order non-linear response of Cu and Au nanocomposites embedded in Al_2O_3 with large metal volume fractions. Alternate pulsed laser deposition (PLD) is used to produce the samples. The nanocrystals are organized in layers that are separated by Al_2O_3. The total number of nanocrystal layers are of 10 and 5 respectively for the case of the $Cu:Al_2O_3$ and $Au:Al_2O_3$ samples while the spacing layers are 6 nm - thick and in both cases. In order to analyse the effect of the metal content and morphology in the non-linear optical properties of the nanocomposites, the dimensions and shape of the nanocrystals in each layer have been varied in different samples by increasing the metal content up to a limit close to the percolation threshold. The nonlinear optical properties of the films have been analyzed by degenerate four wave mixing and z-scan in the wavelength interval from 500 to 620 nm using laser pulses durations in the 100 fs - 10 ps interval.

For all the nanocomposites analysed (Cu and Au, and no matter the metal volume fraction), at wavelengths close to that of the surface plasmon resonance, the third order susceptibility values determined with laser pulses in the ps range are considerably higher (five to twenty times) than the ones obtained under fs laser pulses. The observed increase of the third order susceptibility for increasing pulse durations is bigger in the Cu than in Au nanocomposites. In both cases the third order susceptibility value observed saturates for pulse durations around 5 ps. These results are discussed in terms of the different physical mechanisms contributing to the effective third order susceptibility of the nanocomposites, including the possible contribution associated to the ultrafast heating of the nanoparticles related to the temperature dependence of their dielectric function.

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