# Enhancement of the nonlinear optical response of Cu:$Al_2O_3$ nanocomposite films by multiple particle interaction effects

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Metal-dielectric nanocomposites have received considerable attention over the last years as they exhibit prominent non-linear optical properties. The latter have been pointed out as promising for several applications in the field of information technologies like all-optical switching, signal regeneration or high speed demultiplexing [1]. Particularly, very large third order nonlinear optical susceptibility values along with an ultrafast response time have been reported for Cu nanocomposites [2]. Recent theoretical studies also point out to the feasibility of achieving a large improvement of the nonlinear response of metal-dielectric nanocomposites via multiple particle interactions and giant local field enhancement effects. In spite of this, there are no previous studies reporting the non-linear response of Cu nanocomposites in a wide range of metal concentrations or nanocrystals dimensions.

The aim of this work is to investigate the role of multiple scattering interactions in the third order non-linear response of Cu nanocomposites through the dependence of their response as a function of the volume fraction of nanocrystals up to the percolation threshold. Alternate pulsed laser deposition (PLD) is used to produce Cu nanocrystals embedded in an amorphous $Al_2O_3$ matrix. The nanocrystals are organised in layers that are separated by 6 nm of $Al_2O_3$ having 10 nanocrystal layers, the total film thickness being approximately 110 nm The nanocrystal shape and dimensions are varied from spherical particles with a 2 nm average diameter to a quasi-continuous network of metal in the vicinity of the percolation limit threshold. These limits correspond to volume fractions of Cu nanocrystals ranging from approximately 10% to 50%. The non-linear optical properties of the films have been analyzed by degenerate four wave mixing in the wavelength interval from 580 up to 620 nm with 12 picosecond laser pulses and also at 580 nm with 450 femtosecond laser pulses.

In the case of quasi-spherical crystals with average sizes below 4 nm and volume fractions smaller than 20%, the nonlinear response of the composites can be well described in the frame of the Maxwell-Garnett effective medium approximation. For higher volume fractions, the effective nonlinearity of the composites increases sharply with the volume fraction and the nonlinear third-order susceptibility values increase up to $2x10^{-7}$ esu in the vicinity of the surface plasmon resonance of the composite. This value is about one order of magnitude higher than that predicted by the Maxwell-Garnett effective medium theory indicating that the electromagnetic interaction between particles plays a dominant role in the material response. The importance of multiple scattering interactions and giant local field enhancement effects gets higher as the nanocrystals become bigger and closer to each other. The build-up time of the non-linearity in samples with nanocrystal volume fractions around 30% has been found to be shorter than 450 femtoseconds

References

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