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Directional Selective Nonlinear Transmission of Femtosecond Pulses in Glass-Metal Nanocomposites

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Third-order nonlinear optical materials have gained special attention due to their unique role in all optical switching, optical limiting and optical communication applications. Composite materials based on nanoparticles (NP) in transparent matrix proved to be promising candidates whose $\chi^{(3)}$ can be adequately tailored by tuning the material parameters such as size, shape, and filling factor of nanoparticles [1].

The samples studied are metallic nanocomposites consisting of glass containing spherical silver nanoparticles (Ag NP) arranged in a two-layer geometrical structure. The nanoparticles are distributed within a thin surface layer of glass of thickness few microns, while the remainder of the sample thickness (1mm), consists of pure soda-lime silicate glass (SLSG). A femtosecond Z-scan technique has been used for nonlinear characterization. In general one measures the transmission change as a function of intensity [2]. A Ti:Sa laser (wavelength $\lambda=800\text{nm}$, pulse duration $\tau=100\text{fs}$, repetition rate 1kHz) is used as excitation source. The excitation at 800nm pump wavelength results in considerable two-photon absorption (2PA) for Ag NPs due to their surface plasmon resonance (at 410nm), whereas glass UV absorption requires at least three photons absorbed simultaneously. To investigate the nonlinear properties of these two-layer systems the Z-Scan measurements are performed by irradiating from particle as well as substrate side of the sample [3].

Fig.1. (a): OA Z-Scan curves measured in forward and backward directions; (b): Measured optical limiting behaviour of NP containing sample in forward and reverse direction of laser beam; (c): Results of numerical calculation representing the variation of anisotropy transmission ΔT with relative particle thickness $L_{\text{Ag}}/L_{\text{sam}}$, calculated for different Rayleigh lengths.

Characteristic examples for results on Ag NP glasses are presented in Fig. 1a & b. Experimental measurements showed an anisotropic transmission (optical diode like) behaviour when irradiating the sample in forward and backward direction, respectively. The observed anisotropy consists of an enhancement in nonlinear absorption upon irradiating from glass side of the sample (Fig. 1a: red open triangle) in comparison to the NP layer (open blue triangle). On the basis of a numerical model, this observation is explained by interplay between the self-focusing from glass substrate and 2PA at the Ag NP layer. The intensity dependent contraction of the beam radius, due to self-focusing upon irradiation from the glass side of the sample leads to higher peak intensity – and thus to higher 2PA – in the particle layer than the direct entrance into the layer. The experimental and numerical studies (Fig. 1b & c) showed an increase in anisotropy, when the sample thickness (L_{sam}) approaches the Rayleigh length (Z_0) of the laser beam. A similar directional selectivity in nonlinear absorption is also observed in other two-layer systems comprising of titanium oxide (TiO_2) thin films coated on SLSG substrates (Fig. 1b (ii)). In this context the developed numerical model could disentangle the contribution from individual material layers which can be implemented for the Z-scan analysis of multi-layer based material structures.

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