Leaky-modes excitation in thermally poled nanocomposite glass and their exploitation for saturable absorption

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Abstract: Thermal poling is used to create a reduced index layer in a soda-lime/nanocomposite film. Leaky-modes have been exploited to enhance interaction of light with Au-nanoparticles and demonstrate saturable absorption characteristics in line with state-of-the-art technology.

OCIS codes: (190.4400) Non-linear materials; (160.4236) Nano-materials; (160.1245) Artificially engineered materials

1. Introduction

Thanks to their distinct linear and nonlinear optical properties, noble metal nanoparticles are rapidly finding applications in a variety of fields ranging from bio-chemical sensing to spectroscopy, from chemistry to photonics. Among other applications they are now also emerging as a saturable absorber material for ultra-short pulse generation. Semiconductor saturable absorber mirrors (SESAMS) are the benchmark material for the latter application. Despite the complex and costly clean-room fabrication systems SESAM has the important advantage that the key parameters can be engineered to match the specific application. Recently, techniques that allow a unique flexibility in tailoring the optical properties of noble metal nanoparticles have been demonstrated [1]. In addition, the ultrafast response time makes metal nanoparticles, which are relatively inexpensive to manufacture, attractive as saturable absorber media. The engineering of the modulation depth, which is the maximum change of absorbance, proves more difficult and the highest light induced transmittance change $\Delta T/T$ of about 22% was reported in copper nanoparticles [2]. In this work we present a technique to increase and control the interaction length of the light with 15 nm Au nanoparticles dispersed in a sol-gel silica film deposited on a soda-lime substrate. Our approach exploits a thermal poling to create a layer of reduced refractive index below the film. The layer act as a leaky-waveguide and a 1 mm long interaction length is demonstrated resulting in a massive light-induced reflectance change of 370%. Achieved modulation depth of 48.8%, saturation fluence of 278 $\mu J/cm^2$ and non-saturable losses of 26.9% are comparable to SESAMs and other established saturable absorber materials.

2. Sample preparation

For experiments a soda-lime glass plate (22 x 22 x 3 mm^3) with a nano-composite 130 nm-thick film on top was employed. The film was constituted by ~ 15 nm gold nanoparticles dispersed in a SiO_2 matrix with a volume fraction estimated in 2.3% (Fig. 1a). The sample was heat-treated at 280°C while simultaneously applying voltage from 0 to 1 kV in steps of 200 V every 10 min. The voltage was applied through a 9 x 7 mm^2 stainless steel anode. A thin gold metal wire (25 μm diameter) was inserted between the anode and the sol-gel film near one of the edges, to prevent complete bleaching of the nanoparticles [3, 4]. The thermal/electric treatment induced the drift of Na^+ in highly ionic conducting glasses such as soda-lime causing a reduction of the refractive index of ~1% in a micron-thin layer under the nano-composite film [5]. A scanning tunnelling electron microscope image of the sample cross section (Fig. 1b) confirmed that a modified layer, by ~ 1 μm thickness, had been created. The excitation of the leaky-modes by launching light through the substrate is a technique that has been employed in the past for thin-film characterisation. It is analogue to the two-layer Kretschmann geometry used to excite surface plasmons in a metallic film at both the air/metal and glass/metal interface. This is however the first time that such geometry is realized monolithically in the same piece of glass and used to enhance surface plasmons excitation.

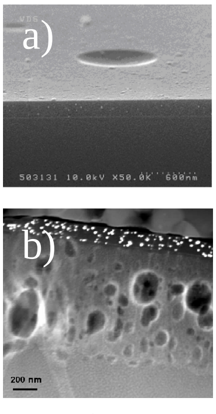

Figure 1 a) SEM image of the pristine sample and b) STEM image taken after thermal- electric treatment (courtesy of Prof. Zayats)

3. Results and discussion

Successful coupling of light into the first leaky-mode in the TE-polarisation was obtained by launching 10mW CW laser light at 532 nm from the side of the sample at grazing angle (Fig. 2). Guided light was observed for a length of ~ 1 mm corresponding to the size of the anode electrode (Fig 2). The corresponding far-field distribution of the reflected beam presented a dip in the middle which indicated the coupling of the incident light to the leaky waveguide. The coupling of light into the leaky mode was also associated to the onset of strong light scattering indicating interaction of the light with the gold nanoparticles. In this low-power regime 88.2% of the laser light power coupled to the leaky waveguide was absorbed. The scattered power was estimated to be not more than 1% of the total incident power. The nonlinear absorption was tested by focusing the light from a frequency doubled Nd:YVO_4 laser (λ =532 nm, τ_{pulse}= 8 ps, w_0 = 205 μm^2) onto the sample oriented at the angle for maximum coupling to the leaky waveguide. The pulse fluence was increased up to 480 $\mu J/cm^2$ by varying the laser repetition rate while maintaining the average power constant at 10 mW to mitigate thermal effects. As a result of the coupling to the leaky modes and the consequent mm-long interaction length of the light beam with the gold nanoparticles, the reflectance was enhanced from the 11.8% in the small signal regime to a 55.6% at the highest pulse fluence (Fig. 3). It corresponded to a $\Delta R/R$ = 371% increase which is by far larger than what is typically obtained with nanoparticles dispersed in solutions or in a glass matrix [2].

A simple two-level saturable absorber model was employed to characterise the sample (Fig.3-inset). Fit to the experimental data in Fig. 3 resulted in saturation fluence, F_{sat} = 278.4 ± 51.6 mJ/cm^2 , non-saturable losses A_{ns} = 26.9 ± 3.9% and small-signal-absorption A_0 equal to 75.6 ± 7.5%. The modulation depth, defined as A_0 - A_{ns} resulted as high as 48.8% ± 3.6%. Most importantly we have here shown a technique that promise to allow engineering of the desired modulation depth by appropriately choosing the parameters of the leaky-waveguide by controlling the depletion layer thickness during poling. The recovery time in gold nanoparticles is typically ~2.5 ps. Despite further improvements are certainly required the characteristics of the saturable absorber shown in this work make gold nanoparticles embedded in a silica matrix already comparable to other established saturable absorber materials like SESAM or semiconductor quantum dots. The typical value for the saturation fluence in SESAMs is ~50 $\mu J/cm^2$ and recovery times can be adjusted from ~100 fs to tens of ps.

4. Conclusions

In conclusion we have shown that with a simple and inexpensive thermal-electric treatment a leaky-waveguide can be produced in a soda-lime glass with a nanocomposite film deposited on top and exploited to enhance the interaction with gold nanoparticles. Our approach was used to demonstrate a saturable absorber based on noble metal nanoparticles having key properties of the same order as the other state-of-the-art technologies. The proposed technique allows the demonstration of modulation depths as high as 49%. We envisage applications of the monolithic glass/leaky-waveguide geometry for laser mode-locking and Q-switching.

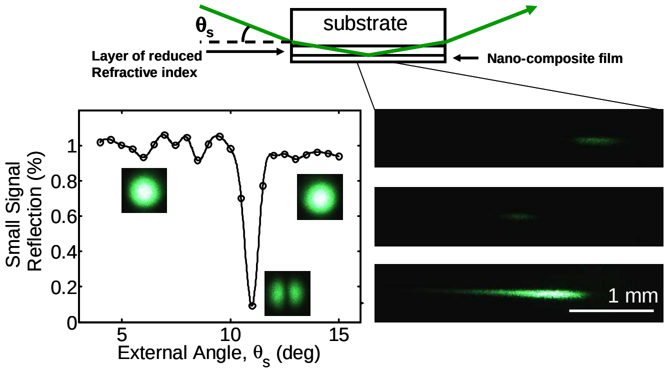

Figure 2 (top) Schematic illustrating the geometry of the coupling of light to the leaky modes and (left) the measured reflectance with far-field images of the beam reflected through the sample. (Right) image of the guided mode.

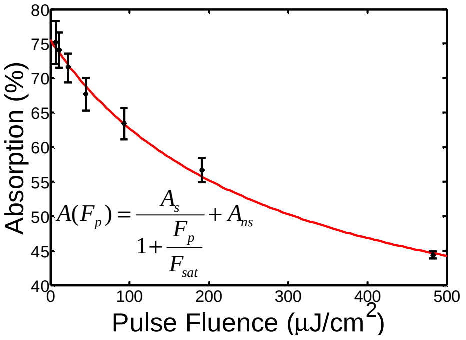

Figure 3 Light induced change of absorption

5. References

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