Femtosecond laser assisted rewriting 3D data storage in composite glass with Ag nanoparticles

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Abstract: 3D anisotropic modifications induced in glass with spherical Ag nanoparticles by multicolor fs laser irradiation was demonstrated as promising technique for 3D storage by spectral data coding. Reversible by heating at 600 ^{o}C or irradiation by 200 ns laser pulses at 527 nmmodifications allowproducing of rewriting data storage devices.

1. Introduction

In the last two decades composite materials containing metal nanoparticles found various applications in different fields of science and technology [1-3]. Linear and nonlinear optical properties of metal clusters are determined by surface plasma oscillations and the fact that the surface plasmon (SP) strongly depends on shape, distribution and concentration of metal nanoparticles as well as on surrounding dielectric matrix. This offers the opportunity for manufacturing of new promising nonlinear materials, nanodevises and optical elements. For instance, the dependence of surface plasmon resonance from the shape of Ag nanoparticles was used for optical data storage by spectral coding [4-5]. In combination with the fs laser assisted shape modification of spherical Ag nanoparticles embedded in glass [6] this method opens up a route for using composite material containing Ag nanoparticles as medium for optical data storage.

As it has earlier been shown, the capacity of optical data storage devices can be extremely enhanced using multiple wavelengths and multiple coding layers [7-8]. In this paper, we present a technique for spectral coding using multicolor fs laser pulse irradiation of spherical Ag nanoparticles distributed with depth gradient of fill factor in glass which in turn could be used in rewriting high density 3D data storage devices.

2. Experimental

The samples containing Ag nanoparticles used in our experiments were provided by CODIXX AG (prepared as intermediate product for manufacturing of broadband polarizers) and prepared using the well-known Ag-Na ion exchange method of float soda-lime glass with following annealing in H_2 reduction atmosphere [9]. This technique results in formation of spherical Ag nanoparticles of 30-40 nm mean diameter in thin surface layers of approximately 6 µm thickness. Size and distribution of Ag nanoparticles in the depth of glass sample strongly depend on temperature and time of Na-Ag ion exchange as well as annealing in reduction atmosphere. In

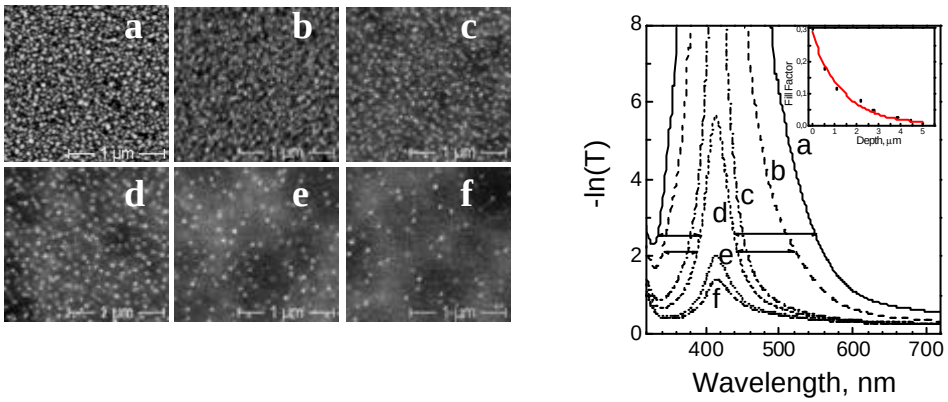

Fig.1. SEM pictures of etched in 12% HF samples with Ag nanoparticles (a – the fill factor 0.29; b – 0.25; c – 0.18; d – 0.08; e – 0.02; f –0.006) and corresponding extinction spectra; inset: fill factor gradient of Ag inclusions in the depth.

particular, the fill factor of Ag nanoparticles near to the surface of the samples used in our experiment was as high as 0.29 and strongly decreased in the depth, as shown in Fig.1(inset). In turn the fill factor of Ag nanoparticles was defined as volume of the inclusions in unit volume of the composite material and was estimated from Scanning Electron Microscopy (SEM). The fill factor gradient of Ag nanoparticles in the depth and the thickness of the layer with metal clusters were measured using samples etched in 12% HF acid with

different detention time (Fig.1). Moreover, the etching technique also enables us to prepare samples with low fill factor of silver nanoparticles (about of 0.006), which in turn were also used in photomodification experiments.

As it can be seen on the fig.1, original samples in extinction spectra demonstrate a strong and broad SP band corresponding to the spherical Ag nanoparticles incorporated in the glass matrix with high fill factor up to 0.29. Etching of the sample in the HF acid results in fading of absorption band caused by decrease of the thickness of the silver-containing layer as well as by sinking of fill factor nanoparticles in the sample. Moreover, from extinction spectra it can be seen, that upper metal-rich layers (f>0.18) are responsible for shift of red shoulders of SP band towards longer wavelengths (see arrows), which obviously results from rise of collective dipolar interactions between aggregated Ag nanoparticles.

Laser induced modifications of Ag nanoparticles in glass were studied using a Ti:sapphire mode locked laser with regenerative amplification. Irradiation was performed at three different wavelengths; by linearly polarized pulses at 400 nm derived from second harmonic generator (SHG), also 500 nm and 550 nm generated by optical parametric amplifier (OPA) with following sum frequency of signal and fundamental wavelength at 800nm. In each case, pulses with energy of 20 μJ and temporal width of 150 fs were focused on the sample using a lens with focal length of 300mm yielding to a spot diameter of approximately 150μm. The sample was mounted on a motorized X-Y translation stage with its glass surface towards the incident beam. In each case square areas of approximately 3x3 mm^2 was written on the sample using multi shot regime of the laser with a pulse density of approximately $2x10^4$ shots/mm^2.

3. Results and discussion

First we studied the effect of fs laser irradiation at 400 nm on the spherical Ag nanoparticles embedded in glass with low fill factor (0.006). As it can be seen from Fig.2 the original plasmon band peaked at 413 nm in extinction spectra is split into two polarization dependent bands centered at 390 nm and 470 nm for s- (perpendicular to the laser polarization) and p-(parallel to the laser polarization), respectively. These two bands

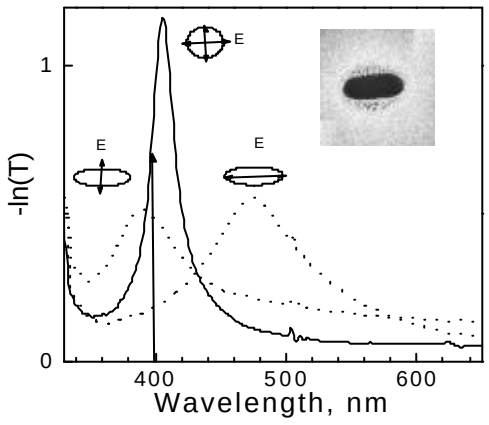

Fig.2. Polarised extinction spectra of glass sample containing single spherical Ag nanoparticles (f=0.006), sample subjected to the irradiation by intense fs pulses at 400 nm; inset: TEM picture of irradiated particle.

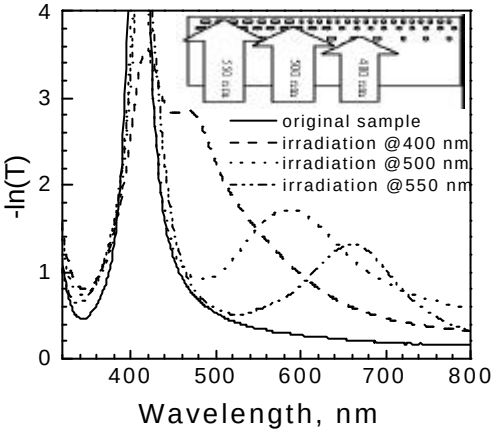

Fig.3. Polarised extinction spectra of the sample with fill factor gradient of silver clustersand irradiated fs laser pulses at 400 nm, 500 nm and 550 nm. The spectra measured in polarization parallel to the laser polarization

can be attributed to SP resonances along short and long axis of the modified (oblong) silver clusters. These shape modifications are also confirmed by transmission electron microscopy (inset Fig.2). Here we have to point out that irradiation of spherical Ag nanoparticles at wavelengths beyond the SP resonance didn't evoke any changes in the extinction spectra.

Similar behavior in extinction spectra was observed in the samples with high fill factor of Ag nanoparticles in glass irradiated by fs laser pulses. However, due to inhomogeneous broadening of the SP band caused by fill factor gradient the spectral hole burning in SP band is associated with modification of Ag nanoparticles distributed in appropriated depth, which in turn could also be defined by wavelength of the incident radiation.

In fact irradiation from the glass substrate side of the glass layers containing Ag nanoparticles resulted in wavelength dependent dichroism, where maximum of SP band in extinction spectra for p-polarisation shifted towards longer wavelengths. From fig. 3 it can be seen that irradiation at 400 nm leads to the appearance of SP band at 470 nm, while irradiation at 500 nm results in SP band peaked at 570 nm, and finally irradiation at 550 nm shifts SP band to 660 nm. Etching of the sample in 12% HF acid testified the fact that the depth of fs laser induced modifications depends on the excitation wavelength. In other words, irradiation at 550 nm affects mostly Ag nanoparticles with highest fill factor and placed near to the surface of the glass layer. Shift of the irradiation

wavelength to 500 nm results in modifications in deeper intermediate region. The changes in extinction spectra induced by excitation at 400 nm are caused by modifications in the deepest layer where collective interactions between Ag nanoparticles are negligible.

Also the obtained results lead to the fact that glass samples with fill factor gradient of the spherical Ag nanoparticles can be used as a medium for high-density 3D optical data storage. Moreover, the capacity of the written information can be increased by simultaneously using both, multicolor irradiation and coding in single layer by the rotation of laser polarization

Indeed by irradiation of a single layer using three different orientations of the laser polarization (angle between the laser polarization and polarization of analyzer is 0^o, 45^o and 90^o), it is possible to achieve up to four independent logic states of the system defined by set of four extinction spectra of modified Ag clusters (see fig. 4) measured by constant polarization direction of the analyzer. It corresponds to the writing of two bits of information. Using three-wavelength irradiation of the glass sample with fill factor gradient of Ag nanoparticles

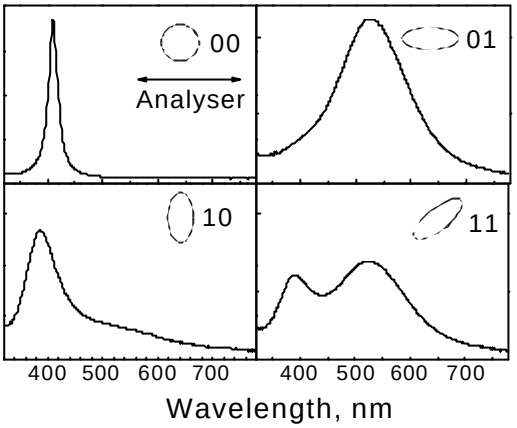

Fig.4 Spectral coding of two bits of information by variation of writing laser polarization and shape modification of spherical Ag nanoparticles in glass

the capacity of a logic unit can be increased up to 6 bits by the coding of information in discrete anisotropic layers in the depth. Hence this technique, the data volume of optical discs can be increased up to 28-30 Gb, which is higher as modern HD-DVD and Blue-Ray Discs. Additional increase of wavelengths used for multicolor irradiation, optimization of fill factor gradient and usage of multilayer discs could significantly enhance the capacity of the data storage.

In addition, it has to be pointed out that observed fs laser induced modifications in composite glass with Ag nanoparticles are reversible by heating at approximately 600 ^{o}C or re-radiation of the modified region by a cw or Q-switched laser at 532 nm near to the SP band. This allows producing of rewriting sources for optical data storage.

In conclusion, we have demonstrated the formation of discrete anisotropic dichroic layer in the glass containing spherical Ag nanoparticles by multicolor fs laser irradiation. The depth and position of absorption band maximum of modified layers is strongly dependent on excitation wavelength resulted from fill factor gradient of Ag nanoparticles in the sample. Restoration of the spherical shape of Ag nanoparticles by heating or irradiation by 200 ns laser pulses at 532 nm, near to SP band, has also been observed, which in turn allows to use this technique in rewriting sources for high density optical data storage.

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