Influence of 532 and 355 nm Nanosecond Laser Pulses on Photodestruction of Silver Nanoparticles in Photo-thermo-refractive Glasses

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Abstract: In this research we investigate an influence of wavelength of nanosecond laser radiation on the process of silver nanoparticles photodestruction in photo-thermo-refractive glass. Second and third harmonic of nanosecond YAG:Nd laser have been applied to irradiate photo-thermo-refractive silicate glasses with silver nanoparticles and different halogenides (F, Cl, Br) as dopants. Optical spectroscopy and X-ray diffraction analysis have revealed a presence of core/shell nanoparticles Ag/AgBr and Ag/$Na_{0.8}Ag_{0.2}Cl$. Irradiation of samples by third harmonic at 355 nm wavelength causes a red shift of surface plasmon resonance band (35 nm) whereas nanosecond laser radiation at 532 nm does not cause significant shift of the surface plasmon resonance band. Such a difference is caused by mechanisms involved in the photodestruction process.

1 INTRODUCTION

Phototermorefractive (PTR) glasses are perspective material for recording of highly efficient volume phase holograms operating in red visible and near IR spectral range (700-3000 nm) (Dubrovin et al., 2014; Ignatiev et al., 2015). The PTR-glasses allow to precipitate silver nanoparticles (SNPs) possessing high absorption coefficient in visible spectral range (λ_{max}=414-490 nm) and nanosize crystalline phase of NaF in local area of the glass host by photo-thermo-induced crystallization (Nikonorov et al., 2010). Unfortunately, this absorption band of SNPs restrict to the using the holograms in short visible range. There are several investigation devoted to the photodestruction of silver nanoparticles in glass matrix. An influence of femtosecond laser radiation on SNPs in glass matrix after ion-exchange was widely investigated (Podlipensky et al., 2004; Stalmashonak et al., 2007). However, the mechanisms of photodestruction of relatively big nanoparticles larger than 20 nm differ from the small ones and require particular attention. The influence of laser irradiation with wavelength 532 nm was well described for silver-containing silicate glass. Nevertheless, it is still very important to use a laser radiation with an appropriate wavelength to reduce the possible damage to the hologram efficiency in such glasses. In the present work we demonstrated a possibility of reduction of the absorption band by bleaching technology with the use of pulse (9 ns) laser radiation with two wavelengths at 355 and 532 nm. The features of SNPs with different shells and surrounding in glass matrix are considered using X-ray diffraction analysis and optical spectroscopy.

2 EXPERIMENTAL

In our studies we have used PTR glass of sodium-alumina-silicate system, $Na_2O – Al_2O_3 – ZnO – SiO_2 – NaF –NaCl(Br)$, activated by CeO_2(0.007 mol%), Sb_2O_3(0.02 mol%), and Ag_2O(0.007 mol%).

The glasses were synthesized in fused silica crucibles at 1500 °C in the environment air. Stirring with a Pt thimble was used to homogenize the liquid. After melting, the glasses were cooled down to 500 °C , then annealed at glass transition temperature (T_g = 494 °C for Ag-Br and Ag-Cl samples, 473 °C for Ag-Br-F sample) for 1h, and cooled down to room temperature with a rate of 0.15K/min. The samples were prepared as the polished plates with the thickness 1 mm.

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Table 1: Glass compositions.

Components	Ag-Br, (mol.%)	Ag-Cl, (mol.%)	Ag-Br-F, (mol.%)
Ag_2O	0.006	0.006	0.006
Br	1.5	0	1.5
Cl	0	1.5	0
F	2	2	6

Samples were irradiated by high pressure mercury lamp. Then a thermal treatment of samples was carried out in programmable muffle furnaces (Neibotherm) at 520 °C at 90 min. The irradiation of the samples were carried out by pulsed (9 ns) YAG:Nd^{3+} laser TII LS-2131M (Lotis) with second harmonic at 532 nm, pulse energy 70 mJ and third harmonic at 355 nm, pulse energy 22 mJ at frequency 10 Hz. The spectral measurements were carried out by means of spectrophotometer Lambda 650 (Perkin-Elmer) in the range 200-800 nm with the step of 1 nm. The X-ray diffraction spectra were obtained on an Ultima IV diffractometer (Rigaku).

3 RESULTS AND DISCUSSIONS

After the mercury lamp irradiation and thermal treatment, x-ray diffraction analysis of the samples has been conducted. Fig.1a,b shows that Ag-Cl and Ag-Br samples contain $Na_{0.8}Ag_{0.2}Cl$ and AgBr crystalline phase. It's been suggested (Nikonorov et al., 2010) that such a crystal can be formed at SNPs as shell. The formation of NaF crystalline phase occurs at relatively high concentration of F^- ions (6 mol.%) and it happens only when glass composition involves Cl or Br ions (Fig.1c). Therefore, it is reasonable to suggest that depending on glass composition SNPs have $Na_{0.8}Ag_{0.2}Cl$ shell in Ag-Cl sample, AgBr shell in Ag-Br sample and AgBr and NaF shells in Ag-Br-F sample (inset on Fig.1a-c). A significant decrease of glass refractive index after UV irradiation and thermal treatment also proves the formation of NaF surroundings (Ivanov et al., 2015).

Curves 1 and 2 at Fig.2 present optical density spectra of investigated glasses before and after mercury lamp irradiation and followed thermal treatment. One can see that initially glasses were colorless down to 350 nm. After the treatment an absorption peak at 453 nm has appeared (Curve 1 at Fig.1). This absorption is connected with surface plasmon resonance (SPR) of SNPs in dielectric matrix. Generally, SNPs are characterized by SPR peak at 408-411 nm (Garcia, 2011), but in case of Ag-Br and Ag-Br-F samples it is shifted to 453 nm due to AgBr shell, which has larger value of refractive index (Nikonorov et al., 2009). Maximum of SPR peak for Ag-Cl samples is located at 419 nm. Such a difference in SPR peak wavelength can be

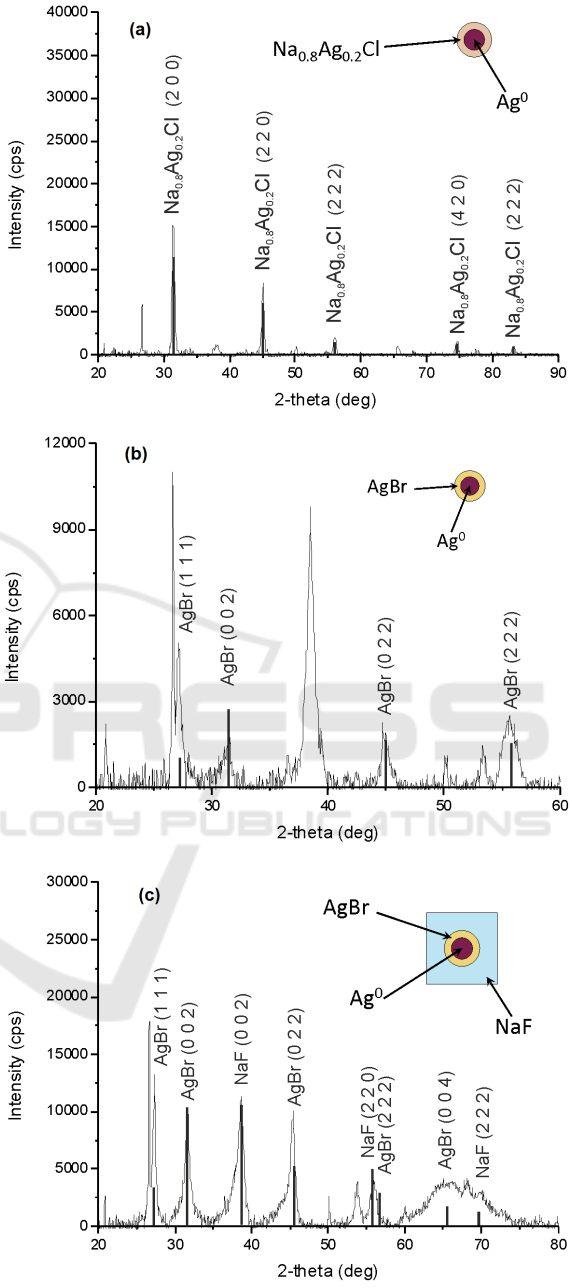

Figure 1: (a) Ag-Cl sample X-ray diffraction data PTR glasses containing 0.13 mol% Ag_2O and 2.2 mol% NaCl (b) Ag-Br sample X-ray diffraction data for PTR glasses containing 0.06 mol% Ag2O and 1.5 mol% NaBr (c) Ag-Br-F sample X-ray diffraction data for PTR glasses containing 0.06 mol% Ag2O, 1.5 mol% NaBr and 6.0 mol% NaF.

related with the width of halide shell or refractive index of the shell (Nikonorov et al., 2009), but this question won't be considered in this research.

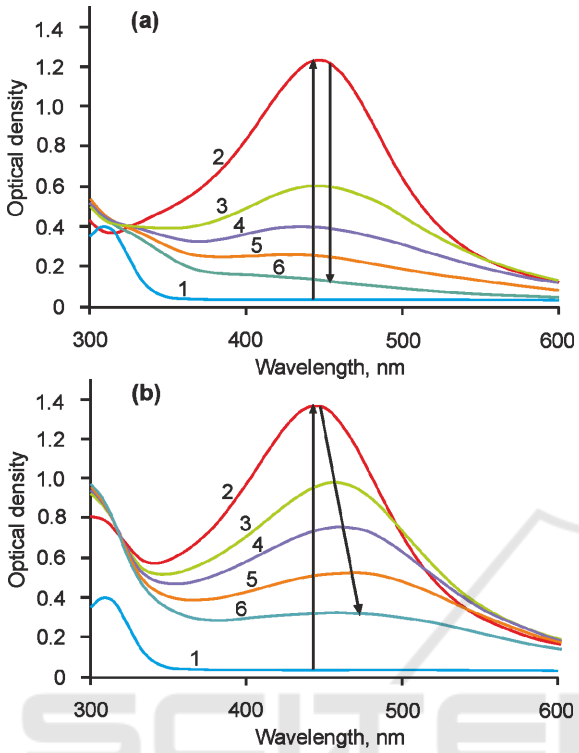

Figure 2: (a) Ag-Br-F after 532. 1- as-prepared glass, 2 – before laser irradiation, 3 – 150 pulses, 4 – 300 pulses, 5 – 450 pulses, 6 – 1800 pulses. (b) Ag-Br-F after 355. 1 - as-prepared glass, 2 – before laser irradiation, 3 – 600 pulses, 4 - 1800 pulses, 5 – 3000 pulses, 6 – 6000 pulses.

Following laser irradiation was conducted using two harmonics of nanosecond laser at 532 and 355 nm with energy density 0.5 and 0.2 J/cm^2 respectively. Fig.2 shows the result of the action of nanosecond laser pulses at 532 nm on Ag-Br-F sample. One can see that the intensity of SPR peak decreases with the increase of laser radiation dose, whereas the SPR band position does not change. An influence of nanosecond laser pulses at 355 nm wavelength is differs significantly. Curves 3-8 also shows the decrease of SNP band intensity accompanied with its red shift on 30-35 nm. Also there is a slight increase in absorption at 270-325 nm spectral range, which can be associated with the formation of glass network defects and a formation of silver molecular clusters Ag_m^{n+}(Klyukin et al., 2014).

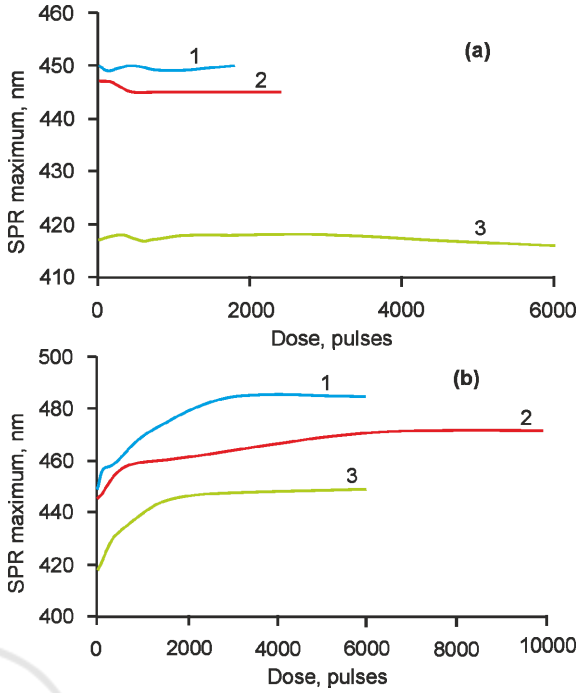

Figure 3: Influence of laser pulses with 532 nm (a) and 355 nm (b) wavelength on Ag-Br-F (1), Ag-Br (2) and Ag-Cl (3) samples.

Fig.3 shows the influence of laser radiation dose on the shift of SPR peak of each sample. One can see that the similar behavior is observed for all the glasses. Namely, the position of SPR peak doesn't alter significantly after laser pulses at 532 nm (Fig.3a). Only Ag-Cl sample demonstrates slight blue shift after 2000 pulses, which can be associated with the decrease of refractive index around nanoparticles or size of SNPs. However, the position of SPR peak of SNPs alters dramatically during the laser irradiation by pulses at 355 nm wavelength (Fig.3b). Each sample demonstrates 10-15 nm red shift after the first 500 pulses and it continues with smaller rate until the glass is fully transparent. Table 2 contains the maximum SPR peak shift for each sample. Clearly that only after the laser irradiation at 355 nm significant SPR peak shift occurs. It should be noted that laser irradiation of the samples was conducting until the full bleaching. Therefore, it is possible to compare the rate of photobleaching of the samples for particular pulse energy. For Ag-Cl sample it took approximately 6000 pulses to bleach it fully by laser radiation at 532 nm, whereas for Ag-Br and Ag-Br-F the number of pulses doesn't exceed 2500 pulses. So Ag-Br and Ag-Br-F glass samples have an advantage to compare with Ag-Cl glass as long as they can be photobleached by the nanosecond laser at 532 nm almost 2 times faster.

This result can be explained by closer position of SPR peak to the laser wavelength at 532 nm, so the probability of laser absorption much higher in case of Br-containing samples with SPR peak near 450 nm. Nevertheless, in case of laser irradiation at 355 nm there is no similar dependence, where the photobleaching process of Ag-Br sample takes 10000 pulses, whereas Ag-Br-F and Ag-Cl samples become colorless even after 6000 pulses. It is also reasonable to suggest that the photobleaching by laser pulses at 532 nm wavelength took less pulses because of higher energy density (0.5 J/cm^2) to compare with laser pulses at 355 nm (0.2 J/cm^2).

Table 2: SPR band shift.

Laser wavelength, nm	Ag-Br, nm	Ag-Cl, nm	Ag-Br-F, nm
532	2	1	1
355	27	32	35

According to the theoretical research (Nikonorov et al., 2009) the position of SPR peak depends on the permittivity of the nanoparticle itself, its shell and surrounding dielectric. It is assumed that SNP in the investigated glasses are surrounded by high refractive index shell (AgBr, $Na_{0.8}Ag_{0.2}Cl$), which causes the initial SPR peak red-shifted position. The following laser irradiation results in the photodestruction of SNP, which can occur in different ways (Hashimoto et al., 2012). It seems that two main mechanisms of the photodestruction take place: photothermal evaporation and Coulomb explosion. Near-field ablation hardly involved in the photodestruction of considered glasses as long as it requires high intensity density, which can be achieved rather by femtosecond laser pulses. Therefore, it reasonable to explain the red shift of SPR peak after laser irradiation at 355 nm by photodestruction of SNP through the photothermal evaporation of silver ions that come from the nanoparticles core to the surroundings. That process causes a local increase of refractive index of SNP shell and following red-shift of SPR peak. This process occurs gradually because the laser pulse wavelength slightly overlap with SPR band. Coulomb explosion mechanism is likely involved in the photodestruction of SNPs by laser radiation at 532 nm. In this case SNPs are breaking at smaller ones and as long as it occurs in condensed material like glass with high value of viscosity the smaller parts of SNPs can not overcome the glass matrix and they stay near each other surrounded by shell with the same refractive index. The reason of involving of such mechanisms of photodestruction for particular wavelengths is not quite understood yet.

4 CONCLUSIONS

In conclusion, we have demonstrated a possibility of reduction of the absorption band by bleaching technology with the use of pulse (9 ns) laser radiation with two wavelengths at 355 and 532 nm. X-ray diffraction analysis have revealed the existence of AgBr, $Na_{0.8}Ag_{0.2}Cl$ and NaF crystalline phase in the investigated glasses after the mercury lamp irradiation and following thermal treatment. Such a crystalline phase is located around SNPs and affects on the SPR band position. During the irradiation the SNPs absorption band decreases depending on the exposure dose. This process accompanies with a red shift of SPR band (35 nm) after the laser pulses at 355 nm, whereas there is no significant shift of the absorption band after the laser pulses at 532 nm. The photothermal evaporation is responsible for the photodestruction of SNPs in case of laser pulses at 355 nm, whereas the Coulomb explosion can explain the results of the action by the nanosecond laser pulses at 532 nm. The technology allowed us to control the size of the silver nanoparticeles in PTR glasses and record the phase holograms in visible range.

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