Polarization selective metasurface formed by interference laser writing

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Abstract. Polarization dependent optical response of the periodically nanostructured SiO₂-Ag nanocomposite material is investigated. As a result of the laser-based surface structuring, the reflection is found to be particularly enhanced and blueshifted for parallel polarization. This result is attributed to the interaction between photonic and plasmonic resonances, namely to the coupling of the grating caused Rayleigh-Wood anomaly mode (described by using grating theory) with localised surface plasmon modes of the embedded Ag nanoparticles (described by using effective medium approach). The laser-induced formation of such metasurface allows precise control of the effect that is essential for practical application in photonics devices and optical filters.

1. Introduction

Structures with a combination of plasmonic properties if NPs and photonic properties of the matrix nanorelief are of great practical interest in designing metasurfaces for photonics, for various sensors, for optics and for security [1]. Recently, prospects were shown for using 2D periodic nanostructures obtained by femtosecond recording in transparent films as high-performance diffractive optical elements [2]. Such meta-surfaces were used to geometrically manipulate the phase of laser beams, obtain an optical vortex, encode and decode optical information [3].

The control over the properties of functional surfaces in their design and fabrication can be achieved by using various methods of colloidal and cluster chemistry often involving self-assembly and organization of nanoparticles on a large scale [4]. On the other hand, methods of laser recording of structures [5], growing nanoobjects of complex shape [6] and synthesis of nanoparticles [7] are effectively used to control the properties of functional elements.

Metasurfaces composed of periodic structures or particles arrays have many attractive applications in optics and photonics. Fabrication of such materials is possible by different methods, among which electron beam lithography and laser formation are recommended to be the most effective. Recently, a number of papers appeared related to the photo-controlled spontaneous organization of metallic NPs in porous films, as well as in glasses, where nanostructures were formed under the influence of CW laser radiation and femtosecond laser pulses [8, 9]. Nevertheless, the controlled synthesis of metallic NPs in nanostructures and deep modification of the structure of the SiO₂ matrix itself remains a complex gear. To create functional surface with new optical properties, it is promising to use laser for writing a periodic nanorelief on a mesoporous film along with the synthesis of NPs and their spatial self-organization.
In this work, we demonstrate the effect of polarization selection with a nanocomposite metasurface obtained by laser treatment. The synthesis of periodic structures in a sol-gel film impregnated with silver nitrate is possible in a two-beam interference scheme with picosecond laser pulses. The paper considers the possibility of simultaneous synthesis of silver nanoparticles (Ag-NPs) and periodic modification of the structure of mesoporous thin films. The obtained combined nanostructures exhibit resonant scattering due to excitation of localized surface plasmon resonance and Rayleigh anomaly in the nanograting. This effect can be utilized for different functional elements in photonics and advanced optics.

2. Results and discussion

2.1. Experimental details and periodic nanostructures formation

Thin mesoporous SiO$_2$ films with a thickness of 170±10 nm elaborated by sol-gel technique on a glass substrate were used as initial samples. The nanocomposite material containing silver NPs with a diameter less than 5 nm as well as silver atoms and ions is obtained. Third harmonic of solid-state Nd:YAG picosecond laser was used as a laser source (pulse duration is 30 ps at 10Hz repetition rate). The scanning speed was set to be up to 5000 μm/s. The direction of the interference pattern was along the scanning direction. The laser beam was split by the phase diffraction grating and then two beams of the 1$^{st}$ diffraction order were subtracted by the spatial filter. The obtained two beam interference pattern was used for a controlled formation of a grating-like structure on the surface, as shown in figure 1(a). The period of the interference pattern was controlled by varying the angle between two interfering beams. Rather large exposure time (equal to 1000 pulses) and laser energy $E_{av}$=33 μJ were found to result in the best uniformity.

![Figure 1](image)

**Figure 1.** The principal of interference laser writing method: the two beam interference pattern initiates periodically distributed field in the thin film. The resonant absorption in the small NPs leads to temperature increase and NPs growth (a); SEM microimage of the periodical nanostructure obtained after laser treatment $\Lambda = 540$ nm (b); SEM microimage of NPs embedded in the film after laser treatment (c).

Under such irradiation conditions, two different effects were mentioned. The first one is the modification of thin film geometry i.e. the formation of the periodic structures with a period of $\Lambda = 540$ nm that is clearly observed under scanning electron microscopy (SEM) (figure 1 b). The modification of the film is possible due to the second effect which is the resonant absorption in small Ag nanoparticles leads to sample heating and consequent NPs growth up to 30±10nm diameter (figure 1 c). Thus, the sample after laser treatment represents a periodic grating-like metasurface containing the embedded chaotically located NPs.
2.1.1. Optical properties of the obtained structures
The obtained nanostructure possesses interesting optical properties, particularly when it is illuminated by a polarized light. Figure 2 (a, b) demonstrates the transmission microimages of the area obtained by laser scanning with the overlapping along one axis N_\text{x} = 1000 and E_{\text{av}} = 33 μJ. The higher contrast of the structure is attained when the polarization vector is along the direction of periodic nanostructures (figure 2 a). On the other hand, the area is hardly visible for the orthogonal direction of polarization and periodicity (figure 2 b). The indicated effect reveals the anisotropy of the obtained nanostructures, which is characteristic for the nanocomposite materials containing metallic NPs [10].

![Figure 2](image)

**Figure 2.** The transmission microimage of the irradiated area in the crossed polarizers: polarization vector is parallel to the periodic nanostructures (a); polarization vector is orthogonal to the periodic nanostructures (b). Diffuse scattering of the sample before and after laser treatment (c). White double arrow represents the polarization direction of the illumination light.

Diffuse scattering of the sample is also found to depend on the polarization of the illumination that is demonstrated in Figure 2 c. The scattering spectra exhibit a noticeable peak at about 500 nm when the sample is illuminated by perpendicularly polarized light. However, the scattering peak in a perpendicular direction (when the polarization vector is aligned perpendicularly to the grating-like structure) redshifts to 588 nm and its amplitude is much lower.

Optical characteristics of such structures are influenced by both the periodic nanostructures and the presence of resonant metallic nanoparticles. For the parallel orientation of polarization vector to the nanostructures the LSPR modes on the Ag nanoparticles enhance a Rayleigh anomaly mode on the periodical lattice leading to the appearance of the well-pronounced peak. For the orthogonal orientation, a detuning between arising on the periodic nanostructures LSPR mode and Rayleigh anomaly mode occurs. The resonant peak in this case is weaker and the redshift corresponds to the decrease of LSPR contribution from the metallic NPs.

3. Conclusions
In this work we have fabricated polarisation-selective nanocomposite structures via laser irradiation by two-beam interference field. The presence of small initial nanoparticles and their growth during the laser writing process make laser light absorption possible. The resulting composite can be described by using an effective medium approximation. Due to the periodic surface relief, the obtained metasurface and possesses unique optical properties namely polarisation selection typical for diffuse surface gratings. The shown dependence of the scattering spectra for different polarization is attributed to a coupling between Rayleigh-Wood anomaly typical for gratings with plasmonic behavior typical for nanoparticle containing effective medium. The observed effect is promising for many applications, such as nonlinear optical filtration, polarisation splitting and switching.

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