Complex holographic elements in photo-thermo-refractive glass for the visible spectral region

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ABSTRACT

Planar holographic optical elements (volume Bragg gratings, VBGs) recorded in photo-thermo-refractive (PTR) glass are widely used for fine spectral filtering and laser beam control. PTR glass provides photosensitivity in near UV region. Therefore, while planar holographic elements operate in the whole window of transparency - near UV, visible and near IR spectral regions, application of complex (nonplanar) elements is restricted to near UV. A method has been proposed to create high-efficiency diffractive optical elements in PTR glass using visible light. The method employs excited state absorption in PTR glass doped with Tb^{3+}. UV radiation was used for excitation to a metastable level of Tb^{3+} and pulsed radiation at 532 nm was used for hologram recording. Both planar VBGs and holographic lenses operating at 532 nm were demonstrated. Complex holographic optical elements in PTR glass can provide attractive solutions for lasers and spectroscopy replacing conventional optical components.

Keywords: Volume Bragg gratings, photo-thermo-refractive glass, complex holograms, holographic optical elements, laser beam control

1. INTRODUCTION

Holographic optical elements (HOE) are the optical elements such as lenses, mirrors, optical filters, diffraction gratings, beam splitters, etc. produced by holographic recording. Diffractive properties are determined by spatially varying refractive index change inside a holographic material, produced by an interference pattern formed between two incident laser beams of actinic optical radiation. Applications of HOEs include beam scanners, laser beam control systems, beam combiners, wavelength multiplexers / demultiplexers, fine filters etc. [1, 2, 3]. The elements can be used to control parameters of a laser beam such as direction, power, polarization, and wavefront shape. There are several advantages of employing HOEs rather than traditional optical elements in an optical system. First, HOEs are generally more compact compared to their mechanical counterparts, besides, some holographic materials allow for formation of several HOEs in the same area. All of above leads to reduction in optical system dimensions when HOEs are used. Furthermore, diffractive holographic elements are often also less expensive compared to traditional optical elements, and are easily reproducible. Finally HOEs are wavelength selective which provides filtering capabilities. The elements with thickness larger than the wavelength are called volume HOEs, and those exhibit the highest diffraction efficiencies, which theoretically can be as high as 100%.

1.1 LIDAR application of holographic optical element

Visible light applications of HOE such as visible laser beam control are among the highly demanded, and frequently encountered. A LIDAR application for atmosphere sensing and meteorology can be an example of advantageous implication of a holographic element into an optical systems [4]. As LIDAR probe beam, normally at a wavelength of 532 nm, is reflected from different areas of the atmosphere, the reflected signal is collected, and then analyzed by the device. The signal light is first incident on a mirror, operating at a signal wavelength, whereupon the light falling into the appropriate bandwidth is reflected into the detector. However the system with traditional mechanical mirrors exhibit substantial noise contribution due to limited reflection bandwidth of a mirror. The problem can be solved by using a diffraction element, where fine filtering is readily available due to wavelength selectivity of a HOE. Theoretically a very high signal to noise ratio can be expected in a system, where a HOE is employed, yet real world systems demonstrate results that are well below the expectations. The disagreement can be accounted for relatively poor holographic capabilities of real world systems that are quite often limited by the properties of a holographic material.

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1.2 Dichromated gelatin for fabrication of holographic optical elements

For a long period of time dichromated gelatin (DCG) has been the most common material used for production of HOEs [5]. Optical element formation begins by coating a glass plate with a solution of gelatin and dichromate in water. The coated plate then undergoes a long-term exposure due to low sensitivity of DCG as compared to silver halide materials. Successive treatment, which includes chemical fixing, washing, and drying, finalizes the hologram formation. Sensitivity response of original DCG is limited to wavelengths below 532 nm, however the range can be extended into the red region by adding methylene dye sensitizer to the material [6]. Dichromated gelatin was found to possess a number of properties giving it an advantage over other holographic materials such as photoemulsions. High diffraction efficiency resulting from large refractive index modulation (RIM) values along with low scattering noise can be sited as an example of such properties. In reality, properties of the resulting hologram are strongly dependent on thermal treatment, bias hardening, and dehydration rate, which often leads to poor performance of a HOE. Besides, the holographic capabilities of DCG are limited due to several other drawbacks such as hygroscopicity and low resolution. In addition, a HOE for high power applications is not possible to fabricate in DCG, since the material with notable absorption would fail when exposed to high power beam. Thus, creating a holographic material for fabrication of efficient HOEs for visible light applications including high power applications remains relevant.

1.3 Photo-thermo-refractive glass

Shortcomings of gelatin described above can be overcome by using a photosensitive glass as a holographic material. Glasses, generally provide better resolution than other holographic media, besides, a holographic structure fabricated in glasses is not affected by humidity. In fact, a photosensitive glass with advanced properties, such as photo-thermo-refractive (PTR) glass allows for fabrication of a HOE that can withstand high power incident radiation. PTR glass is a sodium-zinc-aluminum-silicate glass doped with cerium, silver, tin and antimony [7]. The glass is widely used for fabrication of holographic structures such as volume Bragg gratings (VBGs) [8-12] and phase masks [13] that, in turn, find their application in lasers and imaging systems. The process of hologram fabrication in PTR glass comprises two main stages, namely, exposure to UV radiation and thermal development. Photosensitivity of PTR glass is determined by presence of the following dopants: silver ion acting as a photosensitive agent and trivalent cerium ion as a sensitizer. First UV exposure stage, when glass is illuminated by UV radiation in the range between 280 nm and 350 nm, results in excitation of trivalent cerium ion (Ce^{3+}) from the ground state to the $5d^1$ band, which is located above the electron mobility threshold in PTR glass. As a consequence Ce^{3+} ion is converted to a Ce^{4+} ion, and the released electron is captured by a silver ion Ag^+. As a result a neutral Ag^0 atom is created. It should be noted that no refractive index change is observed as a result of the first stage, rather a latent image of the interference pattern is formed. UV exposure is followed by a thermal development procedure, where exposed glass is subjected to heat treatment. Thermal development also consist of two stages with two different development temperatures. At the first stage of development glass is heated to temperatures above T_g leading to formation of silver containing clusters. After that glass is cooled, and then developed again, this time at higher temperature. Here silver containing clusters serve as nucleation centers for growth of sodium fluoride nanocrystals. Once crystals are grown, a negative refractive index change takes place in the exposed areas of the glass sample. Values of RIM as large as 10^{-3} (1000 ppm) can be obtained in PTR glass. Such high magnitude of induced refractive index and high spatial frequencies of 9000/mm or higher allow for recording of high efficiency VBGs with ultimate spatial frequencies.

1.4 Advantages of holographic optical structures recorded in photo-thermo-refractive glass

Induced refractive index obtained in PTR glass using the procedure described above is permanent owing to the thermal development [14]. As a consequence, a hologram cannot be destroyed by incident radiation of any wavelength in such manner as it happens to materials similar to Fe-doped $LiNBO_3$. In fact an optical structure recorded in PTR glass is not affected by exposure to high power radiation given that the beam intensity does not exceed the damage threshold of the glass. This property is accounted for exceptionally low absorption (below 10^{-4} cm^{-1}) in the visible and near IR spectral regions. Apart from that, VBGs recorded in PTR glass are tolerant to heating at temperatures of up to 350°C. Thermal and irradiation stability gives an important advantage to VBGs fabricated in PTR glass over the other holographic materials. The advantage renders them possible to use for high power laser applications including mode selection, spectral beam combining, spectral stabilization of high-power laser diode arrays, stretching/compressing of ultrashort pulses, etc. [12].

1.5 Limitations of original photo-thermo-refractive glass

Owing to exceptional optical properties of PTR glass, VBG recorded in PTR glass exhibit considerably better performance than their counterparts created in materials such as DCG. However, application of the original PTR glass for visible HOE fabrication is strongly encumbered. In fact, only trivial planar HOEs, such as volume Bragg gratings can be recorded in PTR glass for visible applications. This is accounted for the photosensitivity of the original PTR glass which results from its sensitizer – Ce^{3+}. Since Ce^{3+} has its absorption band located in the UV region between 280 nm and 340 nm, any holographic recording can be only carried out using radiation at wavelength within the indicated range. Meanwhile a complex HOE can be only fully reconstructed using radiation at the same wavelength as used for the recording [15]. Hence complex HOEs in the original PTR glass can be fabricated for only UV applications, while planar gratings can be used in the visible and the IR regions as well. Yet, a possibility to record HOEs in PTR glass using visible radiation would be of a great benefit, as the holographic structure would offer performance unachievable by the other holographic materials.

2. HOLOGRAPHIC OPTICAL ELEMENTS FOR VISIBLE APPLICATIONS IN PHOTO-THERMO-REFRACTIVE GLASS

A new approach for fabrication of a HOE in PTR glass for visible application using upconversion excitation of Tb^{3+} doped PTR glass (Tb:PTRG) was described in a recent publication [16]. Negative RIM of several hundred ppm resulting from concurrent green and UV exposure was obtained in the Tb:PTRG. Holographic capabilities of the material are discussed in this paper, and the efficiency of the recording technique is analyzed.

2.1 Tb^{3+} doped photo-thermo-refractive glass

Tb:PTRG is a type of PTR glass, where Ce^{3+} dopant is replaced with another rare earth element – Tb^{3+}. Photoionization here is performed in a similar fashion as in regular cerium doped PTR glass where excitation to a 5d band, placed above an electron mobility threshold. Trivalent terbium has a 5d band with energy higher than that of cerium, consequently direct excitation of Tb:PTRG requires beams with very high photon energies corresponding to the far UV region [17, 18]. Yet Tb^{3+} allows for excitation of the 5d band using two photon excited state absorption (ESA) process [19]. ESA is an upconversion process, where an ion is excited to a high energy state via an intermediate level by undergoing two successive upward transitions. Each of the transitions is attended by absorption of a photon. Therefore two photons instead of one are required for such excitation. On the other hand two photons might have lower energies which makes it possible for photoionization of the glass by visible light. An important requirement for ESA is for an intermediate level to be metastable. Tb^{3+} has two energy levels that can have sufficiently long lifetime so they might be considered metastable (Fig. 1): upper $^5D_3$ level, which is placed 3.3 eV above the ground state, and the lower $^5D_4$ level, situated 2.6 eV above the ground state. The lifetime of the upper $^5D_3$ level is strongly affected by concentration of Tb^{3+} in glass decreasing as Tb^{3+} concentration increases [20]. As a result, a glass with terbium concentration exceeding 0.1 at. % has only one metastable level – $^5D_4$, which can be used for ESA. Thus, while increased dopant concentration however leads to larger number of ions undergoing ESA as ground state absorption increases, the only $^5D_4$ level could be used for excitation.

2.2 Excited state absorption in Tb^{3+} doped photo-thermo-refractive glass

PTR glass doped with 0.7 at% terbium was chosen for the purpose of recording of a HOE with visible light using ESA upconversion excitation process. The type of ESA implemented in the experiments, employs two photons with different energies for ground state absorption and ESA transitions. This type of ESA is known as double-wavelength ESA (Fig. 1, left). Here the photon providing excitation from a ground state 7F6 is called a pump photon, whereas the photon responsible for the excitation from the metastable state 5D4 is known as a signal photon. In the experimental environment double-wavelength ESA was performed by concurrently exposing a glass sample to two different sources emitting at two different wavelengths.

2.3 Implementation of excited state absorption in Tb^{3+} doped photo-thermo-refractive glass

A UV LED emitting at 375 nm and delivering 450 mW of power was employed as a pump source. The LED provided excitation of Tb^{3+} ion from a ground state to the $^5D_3$ level. However, $^5D_3$ level is no longer metastable in the glass due to high concentration of Tb^{3+}. For that reason the upward transition was followed by nonradiative relaxation to the lower

$^5D_4$ level, which is still metastable. Consequent ESA was carried out using a high power nanosecond laser with high coherence length and pulse energy of up to 200 mJ emitting at 532 nm as a signal source. ESA took place from the lower $^5D_4$ metastable level to the 5d band of terbium, and resulted in photoionization followed by refractive index change after thermal development. The signal source is hence responsible for any holographic recording performed by the means of the ESA upconversion process.

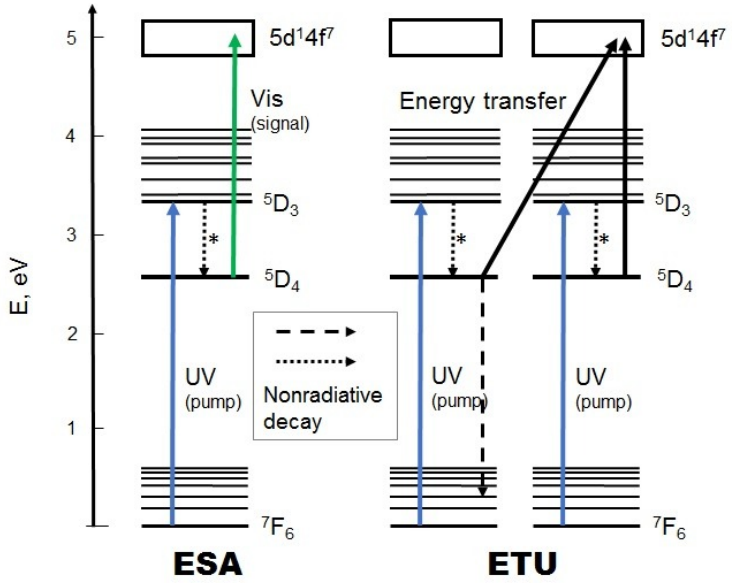

Figure 1. Energy levels of Tb^{3+}. Excited state absorption and energy transfer upconversion in Tb:PTRG are shown with arrows.

2.4 Requirements for efficient excited state absorption

Efficient ESA in Tb:PTRG takes place if the requirements, imposed onto intensities and dosages of radiation from pump and signal sources, are fulfilled [21]. Requirements for signal source are quite straightforward, and claim that signal beam intensity should be as high as possible for efficient ESA. The refractive index change in that case will be proportional to the dosage of visible (signal) light. Constraints imposed on the pump source require that a certain ratio of UV and visible beam intensities is maintained. Intensity of the UV source should be high enough, and its dosage should be large enough to provide efficient pumping to the metastable level. It was determined in the course of the experiments that ESA requires the minimum UV dosage of 150 J/cm^2 when pumping is carried out by the 375 nm UV LED at full power. At the same time, excessively large dosages of pump radiation are unwanted, as they would result in refractive index change due to UV radiation. RIM due to UV radiation results from another upconversion process known as energy transfer upconversion (ETU) (Fig. 1, right). ETU involves an energy transfer between two Tb^{3+} ions both in the intermediate level, and comes into effect at high pump intensities. ESA and ETU are competing processes, in fact the latter leads to excitation of the 5d band as well. Thus, UV light intensity should be kept significantly lower than that of the visible light, and the UV dosage has to be maintained below a certain value in order to keep the UV induced RIM reasonably low. The UV dosage of 2 kJ/cm^2 was found to result in RIM of 50 ppm due to UV radiation after 120 min of thermal development. Here, any refractive index change caused by UV radiation will decrease the dynamic range of RIM due to visible light, consequently the performance of resulting holographic elements is expected to deteriorate. For the reasons described above, the parameters of UV and visible sources were chosen so that to provide the most efficient ESA upconversion possible.

2.5 Excited state absorption efficiency evaluation based on refractive index modulation

The efficiency of ESA in Tb:PTRG can be determined from the photosensitivity of the glass to visible light, which in turn can be assessed by the amount of refractive index change caused by exposure to visible light. Photosensitivity of Tb:PTRG to visible radiation at 532 nm from the high power laser mentioned above was characterized by the induced

refractive index. Samples of polished Tb:PTRG were irradiated with the stripe having a Gaussian lateral intensity profile, which allows for precise measurement of RIM. Exposure geometry is presented in Fig. 2a. A collimated beam from the green laser was focused onto the sample surface using a cylindrical lens, so that the illuminated spot was a stripe with a close to Gaussian lateral profile in vertical direction. However, the longitudinal beam profile obtained in such geometry is inhomogeneous due to nonuniform intensity profile of the collimated beam in horizontal direction. The longitudinal inhomogeneity problem was fixed by sliding the sample back and forth along the stripe axis (Fig. 2b). This pattern at 532 nm was produced by the signal beam, whereas the 375 nm UV LED, mentioned above, provided the pumping. UV beam was focused into a square with a side of 7.5 mm with uniform intensity across the beam area. Beam alignment was performed so that a stripe-shaped signal beam was imposed upon the center of a square-shaped LED beam (Fig. 2b). A set of Tb:PTRG samples was irradiated with different dosages of visible radiation, followed by thermal development. RIM in stripes was measured using a technique based on the shearing interferometer, described in details in [22].

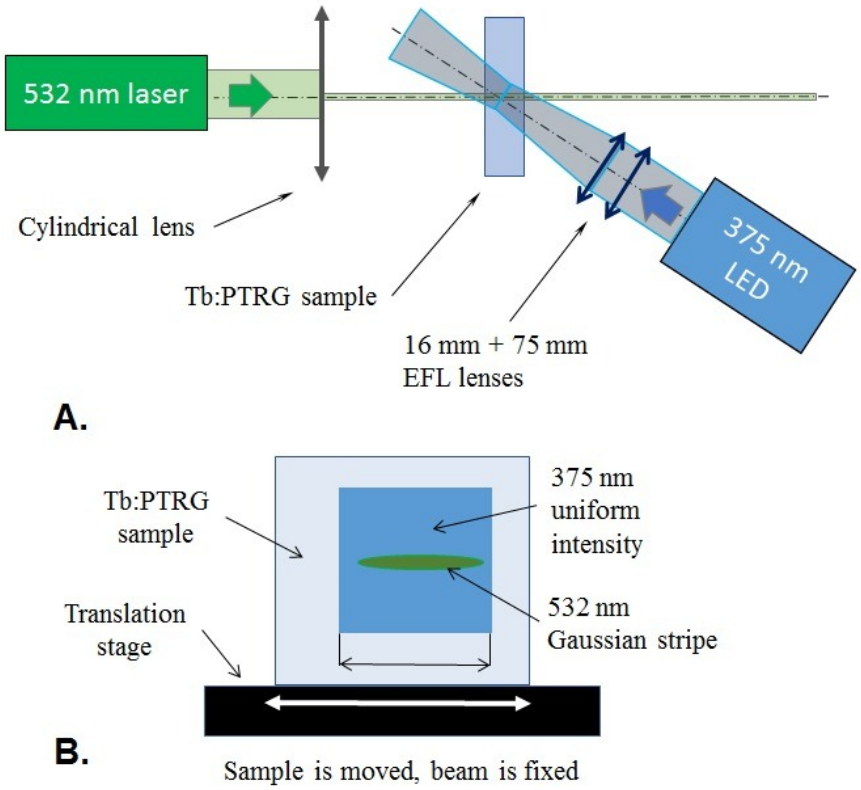

Figure 2. Experimental setup for concurrent exposure of Tb:PTRG sample to a Gaussian stripe at 532 nm and square patter at 375 nm. A – geometry of beam combining, B. geometry of exposure patterns.

2.6 Photosensitivity of Tb^{3+} doped photo-thermo-refractive glass

RIM dependence on dosage of visible radiation for Tb:PTRG is presented in the Fig. 3. The glass underwent two series of exposures with beams of 0.5 and 1 J/cm^2 pulse energy densities. The magnitude of refractive index change depends not only on the parameters of the visible beam, but on the additional conditions as well, including UV light dosage, development time and temperature. The results presented in the figure are the highest values of RIM that were possible to obtain throughout the experiments. It can be seen from the graph that the maximum achievable refractive index change approaches 400 ppm in Tb:PTRG. This value allows for recording of VBGs with diffraction efficiency of 99% and better. It can be then inferred from the obtained results that the Tb:PTRG demonstrates photosensitivity sufficient for holographic recording in the glass.

3. HOLOGRAPHIC CAPABILITIES OF TB^{3+} DOPED PHOTO-THERMO-REFRACTIVE GLASS

HOEs were actually recorded in Tb:PTRG at 532 nm using concurrent exposure to UV and visible beams based on the ESA upconversion. Transmitting (TBG) VBGs was recorded in the glass, since the efficiency of such an optical structure can be easily determined [23]. An optical setup shown in Fig. 4 was devised for recording of a TBG. A collimated beam at 532 nm was split by means of a beam splitter, and then the two beams were combined at the full incidence angle of around 6°. A sample of Tb:PTRG was placed at the intersection of the beams, so that the interference pattern can be recorded in the glass plate. Pumping was carried out by the 375 nm LED beam focused from the backside onto the sample in a 7.5x7.5 mm2 square. The sample was exposed to visible radiation with the total dosage of 32 kJ/cm2, while the UV dosage was found to be 1 kJ/cm2. The TBG was formed in the glass sample after the exposure and subsequent thermal development. The period of the grating was measured to be approximately 5 μm, and its diffraction efficiency was 50%. Thus, characteristics of a HOE recorded with visible radiation in Tb:PTRG are still inferior to those fabricated with the standard UV exposure in regular PTR glass, however the efficiency is expected to increase once the recording process is optimized.

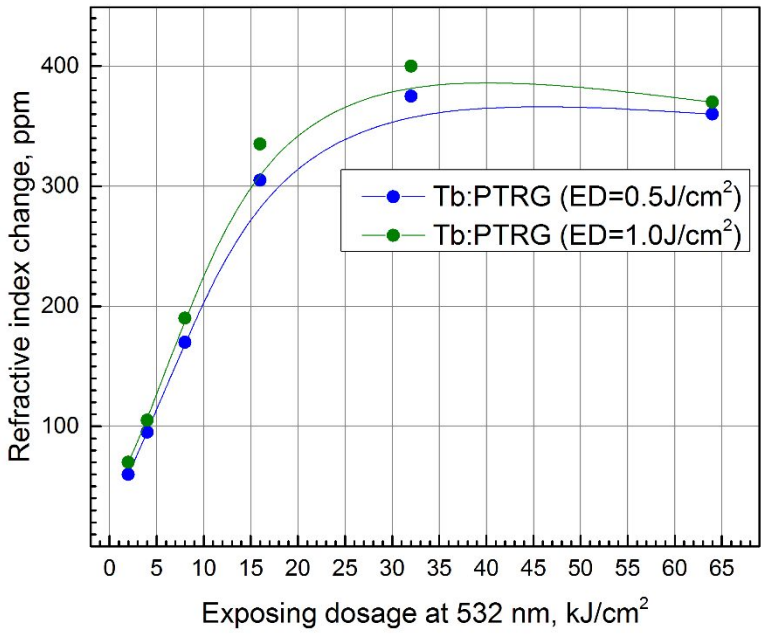

Figure 3. Photosensitivity to visible radiation after exposure to 532 nm beam with different pulse energy densities (ED) for Tb:PTRG

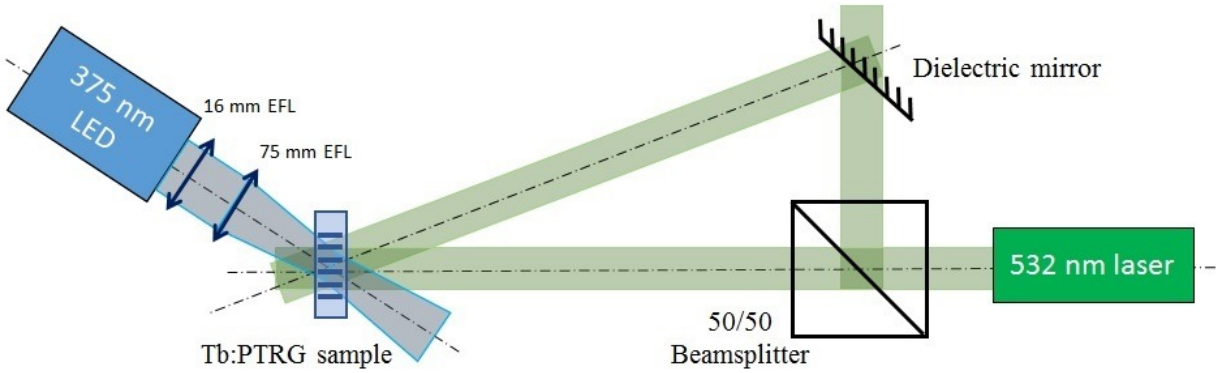

Figure 4. Setup for TBG recording in Tb:PTRG

3.1 Planar holographic optical elements in Tb^{3+} doped photo-thermo-refractive glass

A reflective VBG (RBG) was recorded in Tb:PTRG as well. In that case the beams were combined in a way as it is shown in Fig. 5, so that the converging angle could be easily adjusted. RBGs recording, however, require much higher stability of the recording stage as their period is usually much smaller compared to that of TBGs. Phase stabilization systems hence are required for efficient recording of reflective gratings, and those systems are currently being developed [24].

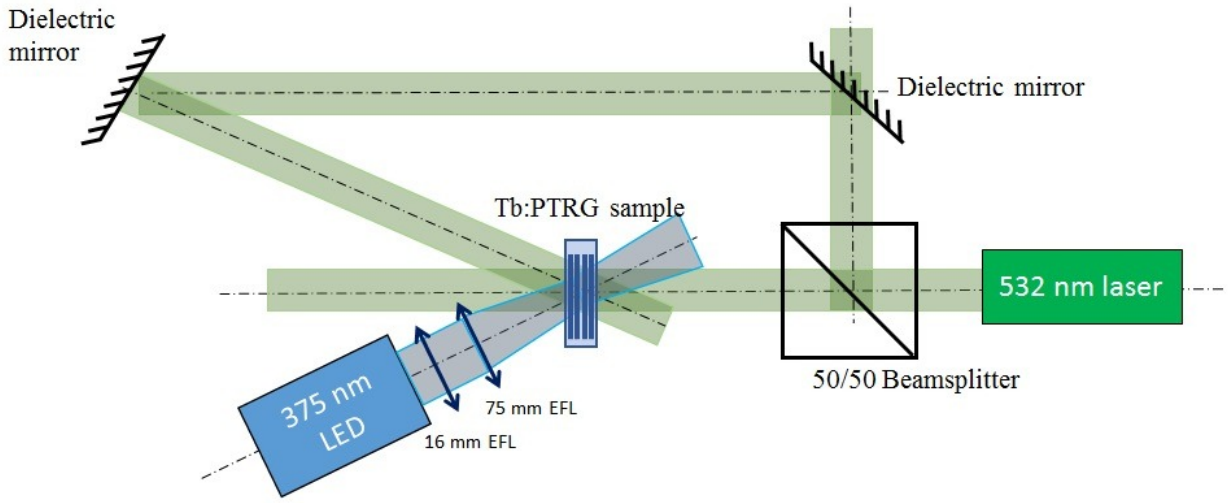

Figure 5. Setup for RBG recording in Tb:PTRG

3.2 Planar holographic optical elements in Tb^{3+} doped photo-thermo-refractive glass

Although planar Bragg gratings allow for fair estimation of the holographic capabilities of a material, they were not of most concern for the present paper, since those structures can be recorded in regular PTR glass with UV radiation. A hologram of a positive convex lens with a focal length of 200 mm was recorded in a sample of Tb:PTRG as an example of a complex HOE utilizing the same laser beam at 532 nm for both recording and reconstruction. The recording setup is shown in Fig. 6 and resembles that used for a regular TBG recording except for one modification. The lens was placed in one of the arms of the setup so that the appropriate beam was focused, and then defocused after reaching its minimum size. The distance between the lens and the sample was chosen in such manner that the focused beam returned back to its initial size as it reached the recording beams intersection point. Therefore the two beams had the same size at the intersection point, which enabled recording a hologram with the largest possible diameter. Such a hologram is basically a two-dimensionally chirped TBG where surfaces of constant refractive index are not planar and grating period is varied along the side of the hologram. As the beam is diffracted by such a hologram, it experiences convergence in a similar way as it would after passing through a lens, or divergence depending on the direction of the incident beam. Operation of the holographic lens in the focusing mode is demonstrated in Fig. 7. The diffracted beam starts to converge upon emerging from the grating (Fig. 7a), and reaches its minimum size as it goes through the focal point. After that, the beam begins to diverge, so its size eventually larger than that of the transmitted beam (Fig. 7b). Efficiency of such HOE however still remains fairly low, as, similarly to RBGs, those structures are highly sensitive to instabilities in the recording stage, and the air movement in particular. Nevertheless the optical elements with higher efficiency are possible to achieve by incorporating a phase stabilization system. A hologram of a curved mirror for instance can be fabricated in Tb:PTRG by modifying the RBG recording setup in a similar way to that described above – placing a lens in one of the arms.

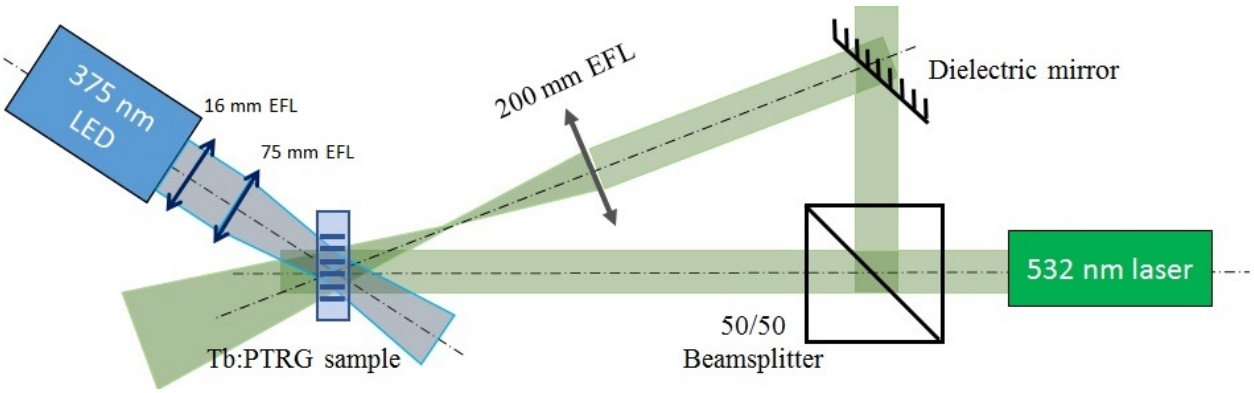

Figure 6. Setup for recording of chirped TBG in Tb:PTRG

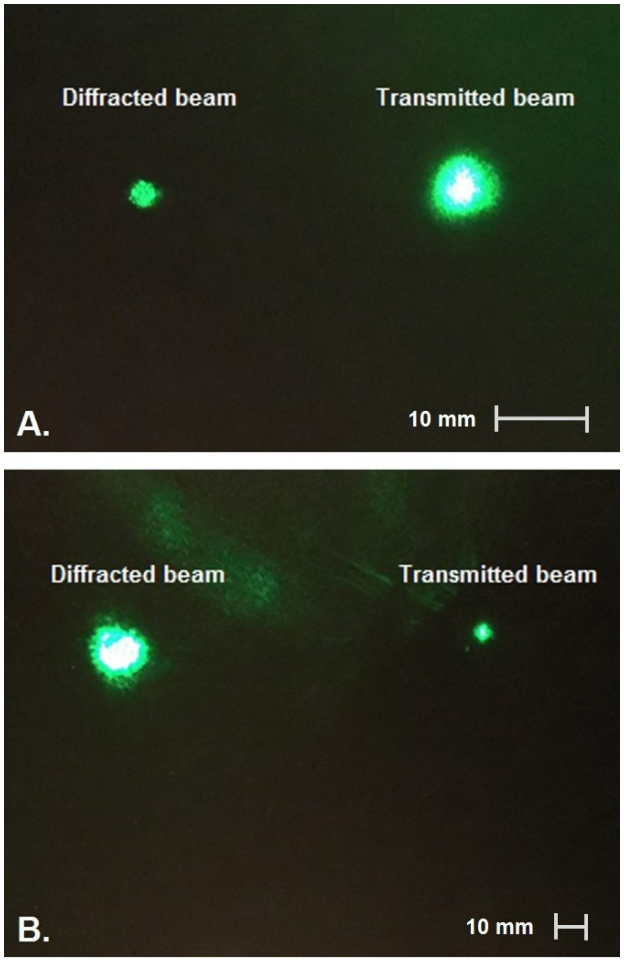

Figure 7. Holographic lens operation, focusing of diffracted beam a) transmitted and diffracted beams 30 mm from the lens; b) transmitted and diffracted beams 80 mm from the lens. Photos are with different magnification – diameter of the transmitted beam is the same at both photos.

4. CONCLUSION

In conclusion, a technology for fabrication of complex holographic optical elements in an inorganic photosensitive glass for visible applications has been developed. The technology involves concurrent exposure of Tb^{3+} doped PTR glass to UV and visible radiation. UV radiation provides excitation to a metastable level of Tb^{3+} ion while subsequent excitation by laser radiation at 532 nm results in photoionization and, after proper thermal development, refractive index change in PTR glass. Thus holographic optical structures could be recorded with visible radiation. Both planar volume Bragg gratings and nonplanar HOEs (holographic lenses) operating in visible spectral range were recorded with this technology. This result paves a way for creation of compact monolithic holographic elements for applications in visible spectral range.

5. ACKNOWLEDGMENTS

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