Holographic recording in bismuth tellurite using nanosecond laser pulses

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Abstract: Bismuth tellurite, Bi$_2$TeO$_5$, is a promising photorefractive material for holographic data storage. Due to its self fixing capabilities, recorded volume holograms do not require any further fixing. Thereby bismuth tellurite accomplishes non-volatile readout of holograms without additional system technology and overcomes an crucial drawback of other inorganic storage materials. Another typical shortcoming of holographic data storage in inorganic crystals using cw laser beams are the long medial writing times. Consequently, in this article the characteristics of recording two dimensional data pages in undoped bismuth tellurite crystals are studied when using a pulsed laser at 532 nm with pulse durations in the nanosecond time scale. Build-up, decay and diffraction efficiency of the photorefractive gratings are examined under pulse excitation. Additionally, the quality and durability of recorded digital data pages are investigated and compared to those recorded into iron doped lithium niobate.

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Introduction

Digital volume holography is one of the most promising next generation optical storage technologies. Advantages are huge storage capacities, fast access times as well as unmatched search capabilities in order to find unindexed information in large data bases [1,2,6]. One of the most crucial components in holographic storage systems is the storage material. Appropriate materials must exhibit high diffraction efficiency, high recording sensitivity, low scattering and long term stability. Photorefractive crystals like lithium niobate have been used for volume holographic recording since many years. Among the different appropriate holographic materials inorganic crystals still provide the highest optical qualities and possess acceptable recording sensitivities. Their major disadvantage is the poor durability of holograms during permanent readout. Especially when multiplexing several holograms in one location of the storage material, the volatility problem necessitates special recording schemes, in order to guarantee equal diffraction efficiencies of recorded holograms [13].
By employing bismuth tellurite crystals, Bi$_2$TeO$_5$, this problem can be overcome. This material enables self-fixing of holograms during the recording process rendering any further fixing technique unnecessary. In 1990 first samples with acceptable optical quality for basic holographic storage investigations were grown [5]. The photorefractive properties of Bi$_2$TeO$_5$ in FWM experiments with cw lasers revealed the existence of a decay component that not only lasts many years in the dark, but also persists under permanent illumination [3, 8]. Basically the effect is accomplished by converting the electro-optically induced refractive index modulation into an electrically neutral modulation of the spatial oxygen distribution. The performance of the Bi$_2$TeO$_5$ crystals has already been studied under CW illumination [7, 8]. However, another critical shortcoming of volume holographic storage in inorganic crystals are the relatively long writing times. These could be reduced by the use of pulsed writing beams [14].

Therefore, our main objective in this article is to study the characteristics of pulsed (3.5 ns, 532 nm) holographic data storage in undoped BiTeO$_5$, while maintaining high diffraction efficiencies and suitable image qualities required for storage applications. The deterioration of reconstructed analog and digital data pages under permanent and pulsed readout is examined and in order to enable qualitative classification of the results, all experiments are also performed in iron doped LiNbO$_3$.

**Experiment**

In order to investigate the recording characteristics, at first simple diffraction gratings are stored. This allows simple comparison and direct measurement and calculations of several important characteristics, such as refractive index modulation, saturation diffraction efficiency and holographic sensitivity. Subsequently the quality of reconstructed data pages is investigated when storing analog and digital data pages.

The employed undoped Bi$_2$TeO$_5$ crystals and reference iron doped LiNbO$_3$ crystals are grown by the diameter controlled Czochalski technique. The samples are x-ray oriented, (45$^\circ$-)cut and polished to optical quality. The impurity concentrations of the Bi$_2$TeO$_5$ samples are $1.0 \times 10^{-6}$ for Cr and $1.2 \times 10^{-5}$ for Fe. Further technical details of the material preparation and the crystal growth process can be found in [4].

The storage experiments are performed in a setup in the 90$^\circ$ configuration (G||45$^\circ$ ab, P||[001]) as depicted in Figure 1. Main advantages of the 90$^\circ$ geometry are insensitivity to holographic scattering and fanning as well as higher angular selectivity, which is of major importance when aiming the superimposition of many holograms in one storage location [11]. The recording beam is split into a signal and a reference beam of equal intensities. In order to record digital data, either a mask or a spatial light modulator is used to imprint the two dimensional bit information onto the expanded collimated laser beam. The signal beam is then focused into the storage material where it interferes with the reference wave. The crystals are mounted inside a vacuum chamber to prevent the breakdown of air molecules in the focal points when recording with high light intensities. All beams are ordinary polarized and the obtained spatial frequency is about 1100 lines/mm. As recording laser a frequency doubled (532 nm), q-switched Nd:YAG pulse laser is used with a pulse duration of 3 ns at a maximum repetition rate of 100 Hz. In order to characterize grating build-up and decay in Bi$_2$TeO$_5$ under laser pulse illumination, the diffraction efficiency was monitored in realtime using a cw He-Ne laser, which was carefully adjusted to fit the appropriate Bragg angle of the grating for the longer wavelength. This condition is vital, since in the setup the angular half-
widths are typically much less than 0.1° and a small deviation from the Bragg angle would cause a large drop in diffraction efficiency. The diffracted red readout beam is separated from the reflected and scattered green light by the use of a cut-off filter in front of the detector. All measurements were carried out at room temperature. The setup is adjusted to obtain an optimal temporal and spatial overlap of the two beams. A typical writing sequence consists of 1000 pulses with a maximum of 15 mJ/cm² at 10 Hz. Signal waves are reconstructed either by one pulse of the original reference beam or by use of a cw beam of the same wavelength. The overlap of the recording beams is about $3 \times 3 \times 3 \text{ mm}^3$. A high resolution CCD camera is used to detect the reconstruction of stored data pages. For comparison reasons all measurements are also carried out in LiNbO₃ in the same setup using cw illumination (532 nm).

**Results and Discussion**

**Photorefractive performance**

In order to analyze the performance of bismuth tellurite as holographic storage material in pulsed systems, the temporal evolution of the diffraction efficiency is measured after recording single holograms. Bi₂TeO₅ crystals show a multicomponent decay of the photorefractive signal. The decay is made up of three major components consisting of an excited state grating, a trapped charge related grating and an ion grating which is responsible for the self fixing process [9]. In this article we only study the slowly decaying signal components, which are mainly important for storage applications. The slowest decay component originates from a grating induced by oxygen ion displacements, where $\text{O}^{2-}$-ions move to open neighboring positions in the oxygen sublattice. This ionic displacement alters the local density and thus produces a refractive index grating, which persists after the decay of the space-charge field.
In order to verify the non-volatility, the gratings recorded with laser pulses are continuously read out over several hours using a low power cw laser, figure 2 a. Additionally the grating development is analyzed under sequential read out over nearly one week, figure 2 b. In both cases the holograms have been recorded with a pulse fluence of approx. 10 mJ/cm² and a fixed pulse repetition rate of 10 Hz. In this experiment the grating period of the holograms was 1.3 µm. Both experimentally obtained curves in figure 3 show the expected theoretical behavior. In the beginning of readout the short time constants of the excited state grating and the trap charge relocation lead to a fast drop of the diffraction efficiency, until the remaining fraction is reached, which is based on the oxygen ion displacement. The continuous readout revealed a remaining diffraction efficiency of more than 10% of its original value. Subsequent illumination with cw light did not result in any further measurable grating degradation. Prior FWM experiments with high intensity cw lasers already suggested that, even after hours of permanent illumination, about 10% of the original saturation diffraction efficiency remains in undoped Bi₂TeO₅ [3]. However, instead of the obtained, almost ideal curve as shown in figure 2 a, some measurements also showed oscillatory effects during the writing and reading cycles. These are very similar to those known from lithium niobate and originate most likely from large drift components [15]. It turns out that only a few pulses are necessary in order to establish holographic gratings that exhibit clearly discernible diffraction efficiencies. The actual achieved saturation diffraction efficiency and grating build-up time depend on the incident intensity. The corresponding saturation diffraction efficiency is direct proportional to the total recording energy. However, changes of the pulse repetition rate did not affect the material response.

In figure 2 b the dark decay of hologram gratings is studied during sequential readout. In this case the remaining diffraction efficiency is approx. 40%. After the first measurement point, all fast signal components have already decayed except for the one of the oxygen ionic origin. The gratings have been probed once per hour for one second. Fluctuations in an hourly timescale may be due to power fluctuations in the incident reading beam or thermal gradients as the crystal was
not temperature stabilized.

The diffraction efficiencies achieved in the different pulse experiments in this article is found to be around 5%. With the model of a sinusoidal phase gratings, the maximum refractive index change can be calculated to be around $1 \times 10^{-5}$, using Kogelnik’s coupled wave theory. The saturation diffraction efficiency obtained in undoped Bi$_2$TeO$_5$ samples is limited by the charge carrier and charge trap concentrations. Performance enhancing dopants investigated for Bi$_2$TeO$_5$ are Fe and Cr. Cr dopants enhance holographic sensitivity and initial diffraction efficiency but weakens long time stability of the grating. Fe dopants increase the long living decay component but also promote light scattering [3].

*Image recording*

In order to investigate the image quality of pulsed volume holographic storage in Bi$_2$TeO$_5$ several analog and digital test pattern are recorded. Figure 3 shows an analog and a digital data page reconstructed by cw light, pointing out well suited storage capabilities of Bi$_2$TeO$_5$. However, they also indicate that light scattering is not negligible. Illumination with a reference beam, without any previously recorded hologram, Bi$_2$TeO$_5$ and LiNbO$_3$ reveal already different optical qualities. In this case illumination with the reference beam indicates strong light scattering in Bi$_2$TeO$_5$, while there is virtually no background noise in LiNbO$_3$. Scattering noise is critical in holographic storage systems, since it significantly affects the systems performance during digital data storage. In a noise limited system, lowering the noise level is equivalent to increasing the capacity or obtaining a better signal-to-noise ratio. Although light scattering can occur due to surface inhomogeneities and nonlinear recording of the signal wave, crystal defects are suggested to be the main noise source in Bi$_2$TeO$_5$ crystals. Apart from scattering, data pages recorded in Bi$_2$TeO$_5$ show sharp contours and high contrast values but are still not equivalent good to the image quality achieved in LiNbO$_3$.

![Fig. 3. Cut-out of a reconstructed digital (left) and analog (middle) data page that have been recorded in Bi$_2$TeO$_5$ using laser pulses and for comparison a analog hologram recorded in LiNbO$_3$ (right). The reconstruction of the data pages is performed by cw illumination of the same wavelength.](image)

Important goals of the present investigation are the reduction of data transfer rates when recording in or reading from the employed inorganic crystals. Therefore, at first an analog image, a USAF 1951 test pattern, was recorded by use of several laser pulses and reconstructed just by a
single pulse. The result is shown in figure 4 a. Figure 4 b shows the reconstruction of the same analog page under cw illumination. In figure 4 a shadow images are observable, which may be evoked by internal reflections inside the not anti-reflection coated crystals. However, both images propose a similar fidelity down to number 4 in group 3 of the test charts in the specific optical setup. One factor with a strong influence on image quality is the inhomogeneity of the beam profile compared to the cw laser beam. Additionally, at pulse energies > 5 mJ/cm² the gratings are accompanied by an observable change in the absorption spectrum producing a pale spot in the illuminated area, which leads to an explicit deterioration of the image quality.

Fig. 4. The hologram of an USAF 1951 test chart has been recorded by pulsed writing beams. The analog image is subsequently reconstructed either by a single laser pulse (left) or by illumination with cw light (right).

Conclusions

The photorefractive performance of bismuth tellurite crystals has been investigated under pulsed excitation. The main advantage of Bi₂TeO₅ compared to other photorefractive crystals is that non-volatile storage is obtained without the need for e.g. an external electric field or thermal heating. The durability of holograms without a separate fixing process is a unique property of Bi₂TeO₅ crystals. Although the attained diffraction efficiencies are slightly below those observed in previous investigations using cw light [10], the presented experiments when employing pulsed laser light again point out these beneficial capabilities of Bi₂TeO₅ for holographic storage applications. It turned out that the photorefractive build-up time of the gratings is much shorter when using pulsed writing beams compared to the recording process under cw illumination. Digital and analog data pages have been recorded in Bi₂TeO₅ using pulsed signal and reference laser beams. Although scattering is still a crucial problem, the reconstructed data pages suggested an expedient quality of the material for digital volume holographic data storage in systems employing pulsed lasers beams.

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References and Links