

Three-Dimensional Holographic Recording by Femtosecond Pulses

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ABSTRACT

We demonstrate fabrication of two-dimensional (2D) photonic crystal (PhC) structure inside the volume of SU-8 resist film by four beams interference. This structure, actually, is a 3D structure fabricated inside the film without sample scanning or laser beam steering. The mechanism of recording in transparent dielectrics is discussed in terms of third harmonic generation, white light continuum, and thermal emission by “hot” electrons.

Keywords: Photonic crystals, holographic recording, third harmonic generation, thermal emission

1. INTRODUCTION

The field of photonics has been of ever-growing importance in recent decades, matching even the progress in microelectronics and approaching its impact.^{1,2} One of the main issues in photonic applications concerns the spectral and spatial manipulation of light. Both tasks are usually accomplished by a grating, which is recorded holographically in an appropriate photosensitive medium, such as crystalline material,^{3,4} liquid crystals,^{5,6} or photo-thermo-refractive (PTR) glass.⁷

The aim of present work was to record holographically a three-dimensional (3D) structure inside a resist film using multi-beam interference pattern. We, first, describe our experimental setup and samples (Sec. 2), then numerical simulation of 5-beams interference patterns will be given in Sec. 3.1. Then, the main experimental results on 4-beams holographic recording in resist film (Sec. 3.2.1) are presented. The possible mechanisms of non-linear absorption of 800 nm wavelength irradiation in transparent dielectrics (glasses, crystals, and polymers) are discussed: a third harmonic generation (Sec. 3.4) and thermal emission of hot electrons (Sec. 3.5).

2. EXPERIMENTAL

2.1. Holographic recording by fs-pulses

An optical setup used for recording of the multi-beams interference patterns is described in details elsewhere.⁸ Briefly, it is based on a diffractive beam splitter (DBS), which divide input laser beam into several, four of them were used for recording. The beams are collected to the sample by two lenses. The beams necessary for the designed interference pattern are selected by an aperture, which is placed between the two lenses. The lens, which was used for focusing into a sample was dry objective lens *Olympus Upo 20×* magnification with numerical aperture $NA = 0.75$. Since the optical distances of the divided beams are the same within the pulse length, temporal overlap of divided pulses is achieved without adjustment. As for a spatial overlap, there is an additional advantage to use the diffracted pulses instead of those obtained by a beam-splitter division, especially, in the case of ultra-short pulses.⁹ An overlap of two split pulses is determined by their longitudinal dimension,

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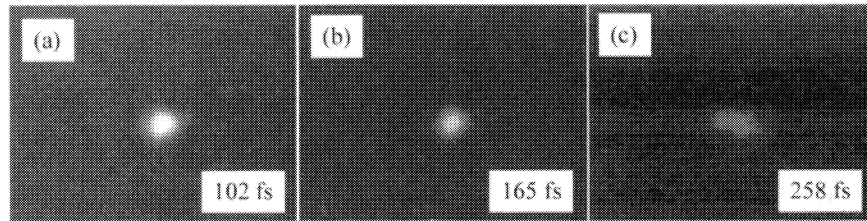


Figure 1. GRENUILLE *time × spectrum* traces of fs-laser (*Tsunami* oscillator) pulses: at the output of oscillator (a), just before oil-immersion objective lens (b), and after the focus, which was placed inside cover glass (c). Pulse durations at FWHM retrieved by FROG algorithm are shown on each image. Horizontal size of the images corresponds to 1779 fs; the vertical - 33.7 nm.

$l_p = c\tau_p$, which is about $30\ \mu\text{m}$ for a 100 fs pulse. Hence, the width of the spatial overlap is $w = c\tau_p / \sin(\Theta_{air})$ for 2-beam interference, where Θ_{air} is the angle between the central axis and the beams in air. In contrary, for diffracted pulses the wave front is not perpendicular to the direction of propagation and $w = d / \cos(\Theta_{air})$, where d is the cross-section diameter of the beam. Thus, sub-mm-scale patterns can be routinely produced using even a tight focusing optics (with numerical aperture, $NA > 0.5$). Four beams create a 4-fold symmetric (tetragonal) interference pattern. The results given here were obtained using $\Theta_{air} = 33.6^\circ$, if otherwise is not stated.

2.2. Laser setup

Femtosecond (fs) pulses were obtained from a setup of Ti:Sapphire oscillator (*Tsunami*) with regenerative amplifier (*Spitfire*, both from *Spectra Physics*) operating at the 800 nm wavelength with pulse duration of $t_p = 130 \pm 10$ fs at FWHM (100 ± 10 fs from oscillator) at the repetition rate of 1 kHz.

To evaluate a temporal fs-pulse spread out after passing a microscope objective lens we have carried out a separate experiment. Pulse duration at different locations along the optical path and at the focus was measured by GRENUILLE technique¹⁰ (*Swamp Optics*). This technique allows to record *time × spectrum* image of the pulse, and then, pulse duration (FWHM) can be retrieved by the frequency-resolved optical gating (FROG) algorithm (*Femtosecond technologies*). To measure pulse duration at the focus, we used an additional solid immersion lens, which collects strongly divergent light before introducing it into the GRENUILLE setup. Figure 1 shows *time × spectrum* image of the pulse and the retrieved pulse durations using FROG algorithm at: the output of fs-oscillator (a), after passing optical alignment bench and part of a microscope till the entrance of objective lens (b), and after the passing of the focus of an oil-immersion $NA = 1.3$ objective lens. For the all retrieved pulse durations a FROG error was lower than 2%. Pulse duration in the case of holographic recording setup, which had smaller amount of optical elements and a $NA = 0.75$ objective lens was evaluated to be about 160 ± 15 fs (the pulse duration at the output of regenerative amplifier was measured by GRENUILLE and found to be 130 fs).

2.3. Media for holographic recording

In our experiments we used a film of negative photoresist SU-8 (Microlithography Chemical Corp.). Samples of SU-8, which absorbs at $\lambda < 400$ nm, were spin-coated on a cover-glass plate with a thickness about of $5 - 6\ \mu\text{m}$, and prebaked before exposure. After irradiation, the samples were postbaked to enhance a photo-initiated crosslinking reaction. By the successive development, the non-crosslinked regions were washed out and 3D structures were obtained. Typical laser power before DBS was 0.21 W, and 1.67 mW at the sample place for 4-beams hologram, and the exposure time was 5 – 90 s in the case of SU-8 film. In terms of energy per pulse it corresponds to $0.42\ \mu\text{J}$ (fluence - $2.4\ \text{mJ}/\text{cm}^2$ with the irradiance of $1.6 \times 10^{10}\ \text{W}/\text{cm}^2$ for a $150\ \mu\text{m}$ diameter spot and $t_p = 150$ fs pulses). Maximum intensity at the maximum of 4-beams interference amounts to 16 times of that in a single pulse. For observation in scanning electron microscope (SEM) a thin layer of Au (< 20 nm) was coated by sputtering.

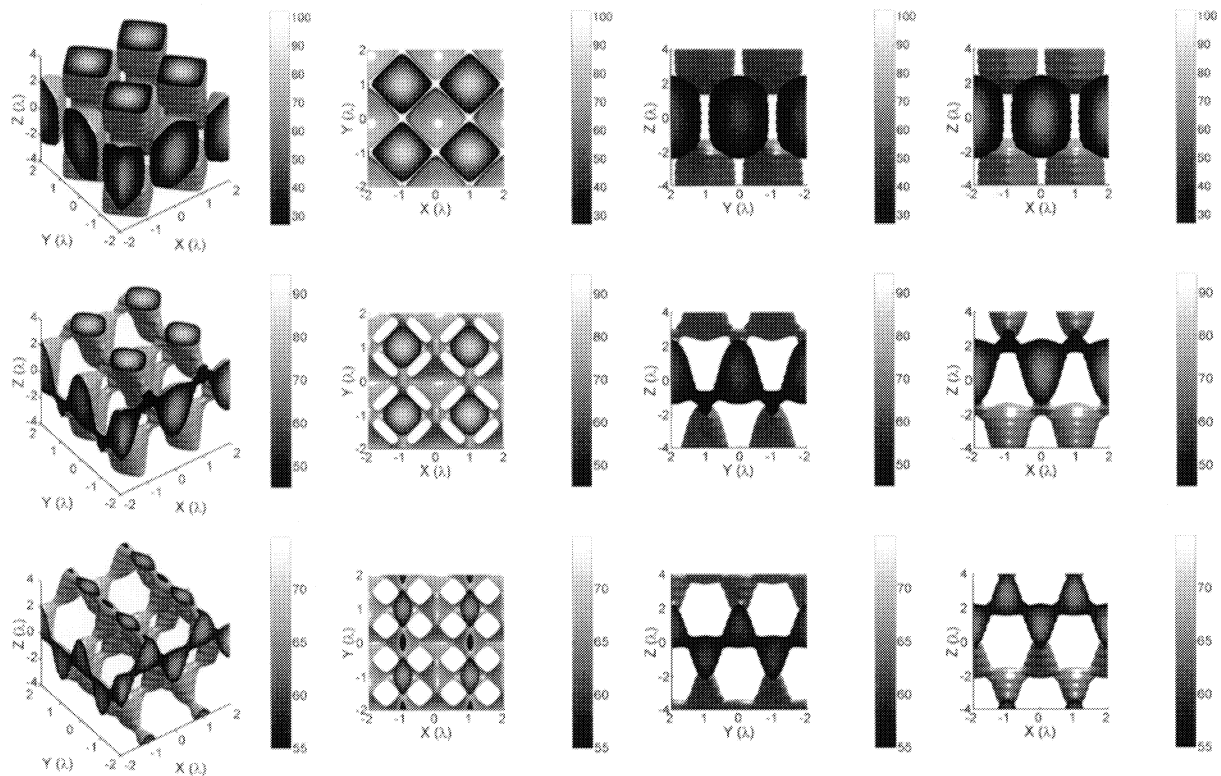


Figure 2. Spatial light intensity distributions generated by 5-beam (plane waves) interference. E-fields of all beams were: $(4e^{-i0}(\text{central}); 1e^{-i0}; 1e^{-i0}; 1e^{-i0}; 1e^{-i0})$ for the top row, $(4e^{-i0}(\text{central}); 1e^{-i\pi/4}; 1e^{-i\pi/4}; 1e^{-i0}; 1e^{-i0})$ for the middle row, and $(4e^{-i0}(\text{central}); 1e^{-i\pi/2}; 1e^{-i\pi/2}; 1e^{-i0}; 1e^{-i0})$ for the bottom row, respectively. Threshold intensity at which iso-surfaces are drawn are different for each row: 27, 45, 52. Spatial dimensions are normalized to the wavelength of the recording light, λ . $\Theta_{air} = 30^\circ$ was taken for calculations. Note the scale of z-axis differs from the others.

3. RESULTS AND DISCUSSION

3.1. Numerical Simulations of Multi-Beam Interference

The expected structures were calculated from the light intensity pattern, a hologram, created by the corresponding number of beams as given by:

$$I(\vec{r}) = \left\langle \left| \sum_{i=1}^4 E_i \cos(\vec{k}_i \cdot \vec{r} - \omega t + \varphi_i) \right|^2 \right\rangle, \quad (1)$$

where E_i is the light field amplitude, k_i , φ_i are the wavevector and phase of the corresponding beam; while ω is the cyclic frequency, r – the coordinate vector and t is for time.

Figure 2 shows 3D light intensity distribution created by the colliding 5-plane waves according to eqn. 1. To obtain the highest contrast in hologram the E-field amplitudes should obey the 4(central beam): 1 : 1 : 1 : 1 ratio for a 5-beam interference. The effect of phase control on the morphology of a hologram was also simulated. In order to avoid a spatial translation of an entire structure, the phases of a pair of beams (counter-propagating in z-projection (Fig. 2)) was set equal. In such a way, a smooth transformation of a body-centered cubic (bcc) structure (upper row in Fig. 2) into a diamond structure (bottom row) can be induced by a change of a phase from 0 to $\pi/2$. For the development procedure of an exposed resist film, it is very important to have access to

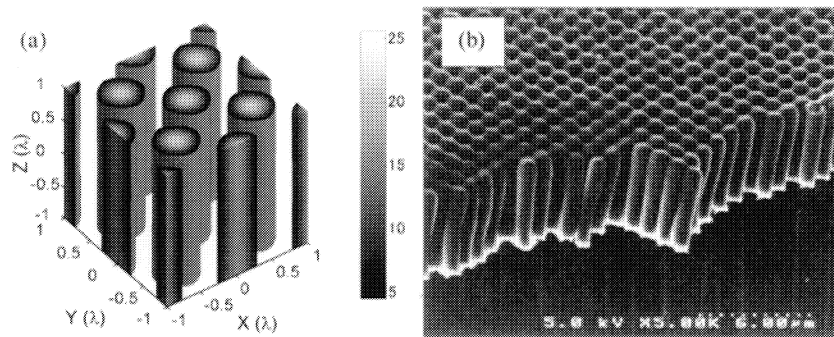


Figure 3. (a) Calculated 3D light intensity patterns (2D PhC structure) of 4-beams interference. The electric field amplitude of each beam was 1, the phases were 0, and the dimensions of pattern are expressed in the wavelength units, λ . (b) SEM images of structures recorded by 4-beams hologram in SU-8. Period in the lateral plane (XY) was $\simeq 1 \mu\text{m}$ (refractive index of SU-8 is 1.67). The height of the developed hologram corresponds to the $5 \mu\text{m}$, which is a thickness of spin-coated SU-8 resist.

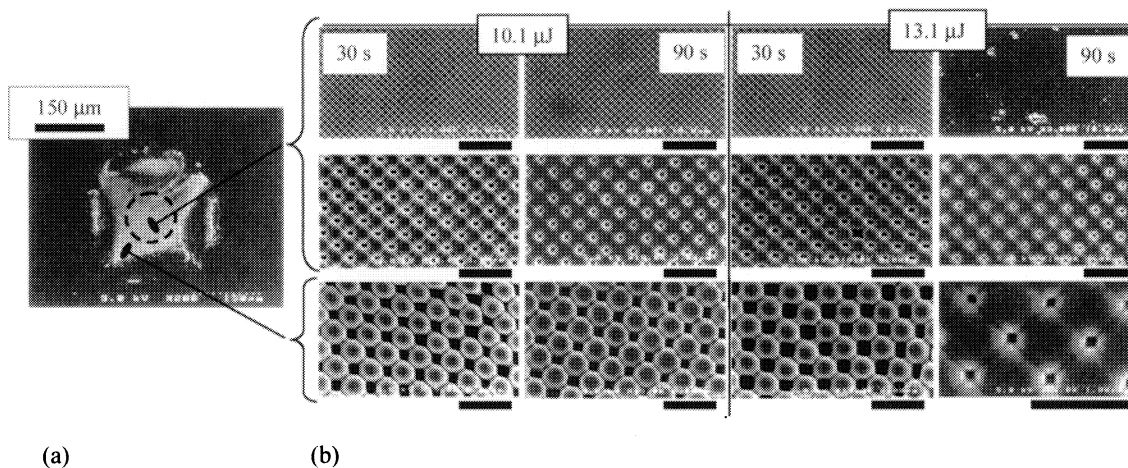


Figure 4. (a) SEM image of 4-beams fabricated structures in SU-8 film. Exposure wavelength was 800 nm. (b) Examples of patterns fabricated at different exposure length and pulse energies. Scale bars are: 10, 3, and $2 \mu\text{m}$ for the top, middle, and bottom rows, respectively.

the interior of a structure. This is, actually, impossible for a fully crosslinked bcc structure. Also, in the case of diamond structure, the high intensity regions are more spatially localized, what makes it more favorable for 3D development in solution after exposure.

3.2. Holographic recording of 3D structures

3.2.1. Recording in resist SU-8

First, we recorded a 3D rod structure (a 2D PhC structure) by 4-beams interference in a film of resist. The pattern recorded into a resist was expected to be similar to that shown in Fig. 3(b), where $\varphi_I = 0$ was taken for calculations. Judgment for this was based on our previous experiments, since the difference in phases of the beams should bring about the corresponding changes in structure as it is predicted by numerical simulation (Fig. 2) for 3D photonic structures, however different morphologies of fabricated structures were never observed experimentally (a detailed consideration of the influence of the phase on interference pattern in the case of 2D

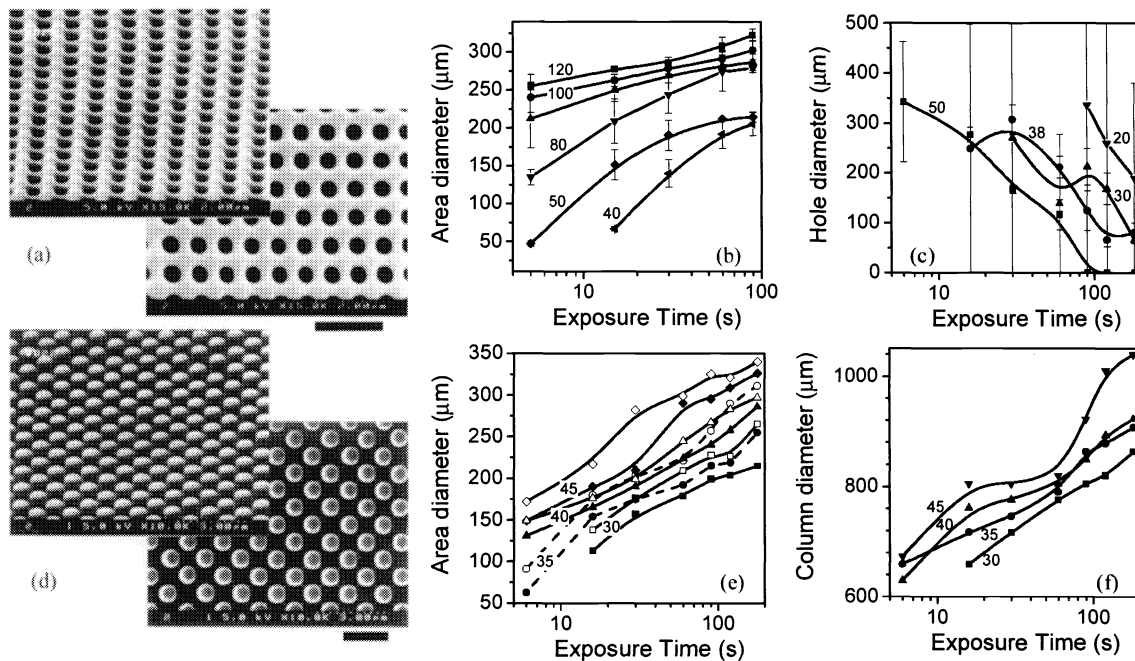


Figure 5. (a) Slanted and top-view SEM images of structures recorded in SU-8 resist by (2 + 2)-exposure (after the exposure by two beams, the sample was additionally exposed by two beams, which were in a plane oriented perpendicularly to the first beam pair) at 800 nm wavelength. Area (b) and hole (c) diameters of the recorded structures vs. the exposure time. Total energy of the combined beams is given by the corresponding curve. (d-f) Same as (a-c) but for a four-beam exposure. Filled and hollow markers in (e) denote the crosssections of the recorded area in two perpendicular dimensions. Scale bars (a,d), 2 μm .

photonic structures is given in ref.¹¹). Recorded structures were closely following the expected ones as it is shown by SEM images in Fig. 3(b). Translation period was found 1 μm exactly equal to the expected value of $\lambda/(\sqrt{2} \sin \theta_{\text{air}}) = 1.02 \mu\text{m}$.

Figure 4 shows SEM images of the fs-pulses exposed and developed SU-8 film. We observed a pattern, which corresponds to a $\varphi_I = 0$ condition. The only difference can be observed was a degree of exposure at different locations of the fabricated spot. Exact conditions when the largest volume of unexposed resist is removed must be found experimentally Fig. 4(b). Films shown in Fig. 4 were prepared by slightly different procedure. First, the film of SU-8 was spincoated and fully exposed. Then, the layer for pattern fabrication was coated over in a standard way. This yielded in a film, which was better withstanding a development and a structures of higher aspect ratio were obtained.

By using 4-beams exposure two different patterns can be recorded into the resist film. Figure 5 shows patterns of holes (a-c) and columns (d-f) recorded in SU-8 film. The hole-pattern was created by combining two consecutive exposures by 2 beams. The column-pattern was recorded in a single 4-beam exposure. Typical area and hole/column diameters are given as a function of pulse energy and exposure time in Fig. 5. Large uncertainties depict a scattering between the largest and the smallest diameters observed inside a fabricated region.

Recently, it was found that a thick phase grating can be recorded in photo-thermo-refractive (PTR) glass¹² by similar holographic exposure to 4-beams generated pattern.¹³ The mechanism of PTR glass exposure (which needs a 308 nm wavelength light) is still unclear in the case of hologram recording at 800 nm by fs-pulses.¹³ Let us discuss below several possibilities relevant to photo-modifications in photo-sensitive glasses and resists

in general: three photon absorption, absorption of white light continuum, third harmonic generation (THG), and absorption of a thermal emission of hot electrons (thermally not equilibrated with the ions). The first two mechanisms usually proves themselves ineffective. For example, a three-photon absorption was absent at 700-800 nm spectral range in PTR glass as it was found by direct measurement using a multi-photon absorption spectroscopy. White-light continuum (WLC) generation as a probable source of photo-sensitive glass and resist exposure is discussed in the next Section 3.3.

3.3. White light continuum generation

White-light continuum (WLC) is typically observed in transparent dielectrics (crystals and glasses) at about 10^{11} W/cm² irradiance. For example, in PTR glass WLC was observed at $(6 \sim 7) \times 10^{11}$ W/cm² irradiance at 775 nm wavelength. If WLC would be generated at a local maximum of the interference field inside PTR glass, it could produce a periodical phase modulation. However, the interference of WLC should be disregarded as a mechanism of periodical structure formation because WLC propagates collinearly with first harmonics. This means that it will interfere with unequal periods and destroy a regular 3D pattern.

Also, it is noteworthy, that a kind of WLC observed during optical damaging of glasses by tightly focused (numerical aperture $NA \geq 1$) ultra-short pulses can not be explained by the standard models of non-linear optics due to a short length of propagation inside a region of high light intensity, which length can be estimated as a Rayleigh length, $z_R = \pi w_0^2 / \lambda$, here $n = 1.473$ is the refractive index, and the beam waist of a Gaussian beam is $w_0 = 0.61 NA / \lambda$ with $NA = 1.35$ being the numerical aperture of the objective lens. For the wavelength $\lambda = 800$ nm one finds $z_R = 1.5$ μ m.

Let us evaluate spectral broadening of a Gaussian beam via self-phase modulation (SPM) after passing a high intensity focal region at the irradiance close to dielectric breakdown. Light induced damage threshold (LIDT) of a BK7 (*Schott*) glass was found at irradiance $I_{LIDT} = 6.6 \times 10^{12}$ W/cm² for $\tau_p^{FWHM} = 200$ fs pulses (pulse duration was measured after focus by GRENOUILLE technique (see, Sec. 2)). Nonlinear refractive index of silica is $n_2 = 2.5 \times 10^{-16}$ cm²/W,¹⁴ where the intensity dependent refractive index is given by $n(I) = n_0 + n_2 I$. The cause of spectral broadening is a change of phase of the passing pulse.¹⁵ According to the classical theory,¹⁵ the distance over which the phase is changed by 1 rad is called a nonlinear length and is given by $L_{nonl} = ((2\pi/\lambda)n_2 I)^{-1} = 77$ μ m for $I = I_{LIDT}$. Then, the maximum shift of the frequency, which is the same for the Stokes and anti-Stokes modes is $\Delta\omega = \sqrt{2/e^1} \varphi_{max} \Delta\omega_0 = 4.4$ cm⁻¹, where the maximum phase shift is $\varphi_{max} = (2\pi/\lambda)n_2 I \cdot 2z_R$ and the spectral width at (e^{-2})-level is $\Delta\omega_0 = 260$ cm⁻¹ (10 nm FWHM spectral bandwidth at $\lambda = 800$ nm). Spectral shift of $\Delta\omega = 4.4$ cm⁻¹ is small, indeed, for our pulses of $\omega_0 = 12500$ cm⁻¹ ($\lambda = 800$ nm).

Theory of SPM,¹⁵ which accounts for self-steepening of the pulses predicts even smaller spectral broadening after passing a focal region due to small parameter $Q = 2z_R n_2 I / (c(2\sqrt{\ln 2})\tau_p^{FWHM}) \ll 1$ for the conditions considered above. Indeed, Stokes and anti-Stokes spectral shifts are given by $\Delta\omega_{s,a} = \omega_0(0.25(\sqrt{Q^2 + 4} \mp Q) - 0.5) < 1$ cm⁻¹ at the dielectric breakdown irradiance. This suggests that according to the classical WLC generation models there is no spectral broadening, which could produce the light at the wavelength suitable for PTR glass or SU-8 exposure by one-photon absorption.

3.4. Mechanisms of Third Harmonic Generation in Glasses

THG is a mechanism, which may produce the light strongly absorbed in isotropic transparent materials, such as photo-sensitive glasses or resists. During exposure of glass by fs-pulses, which overlap in space and time, strong transient effects of pulse energy exchange are expected via the transient grating induced by the interference of two beams. This two-wave mixing geometry is schematically depicted in a wave-vector presentation in Fig. 6(a). For the every pair of pulses used in hologram recording such an effect of self-diffraction is expected. This is analogous to Bragg diffraction. Recently, we have reported on an efficient THG in a Bragg grating pre-recorded in a PTR glass, when the readout was made by 130 fs pulses at 775 nm wavelength.¹⁶ The diffraction efficiency $\eta = I_1/I_0 \simeq 0.05\%$ was observed, here I_1 and I_0 are the first order diffracted and incident intensities, respectively.

Third harmonic generation (THG) is most effectively produced at the interfaces, e.g. the air/glass boundary has been well studied.¹⁷⁻¹⁹ However in our experiments, we had not detected any THG at the interface

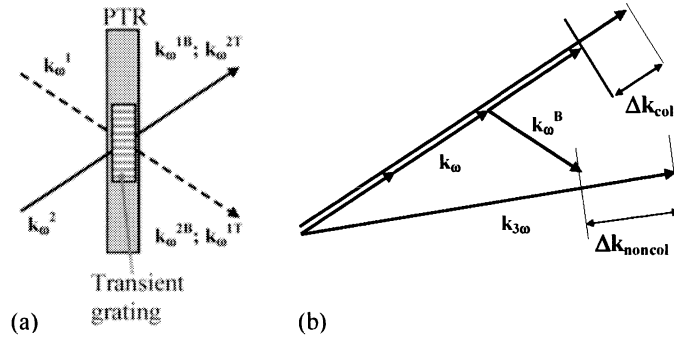


Figure 6. (a) Schematic presentation of two-wave mixing (two beams interference or Bragg grating readout by one beam). (b) Wave vector diagram for collinear and noncollinear THG for geometry shown in (a).

during hologram recording (Pt-coated grating was used to avoid absorption of TH). Moreover, the direction of propagation of high order harmonics (such as TH) generated at interface is collinear with the first harmonics, which was absent in our experiment.

Let us consider here PTR glass as a case material to discuss the mechanism of THG. PTR glass exhibits normal dispersion, therefore the least mismatch is observed in the collinear geometry of THG:

$$\Delta k_{col} = 3k_{\omega} - k_{3\omega} = \frac{3\omega}{c} (n(\omega) - n(3\omega)) < 0. \quad (2)$$

According to the dispersion curve fitting equation for PTR glass, $n(\omega) = 1.49$ at 775 nm and $n(3\omega) = 1.54$ at 258 nm,²⁰ which results in $\Delta k_{col} = -12160 \text{ cm}^{-1}$ for the collinear case.

Fig. 6(b) clearly shows that larger mismatch is obtained in any noncollinear four-wave-mixing geometry. When the angle between the two fundamental waves inside the medium is 5° , the resulting mismatch is $\Delta k_{noncol} = -12470 \text{ cm}^{-1}$. However, one would still expect noncollinear THG process to dominate under certain conditions. When two fundamental waves E_1 and E_2 are present in the medium, the electric field of the third harmonic is given by:

$$E_3 \sim \chi_{eff}(E_1 + E_2)^3 = \chi_{eff}E_1^3 + \chi_{eff}E_2^3 + 3\chi_{eff}E_1^2E_2 + 3\chi_{eff}E_1E_2^2, \quad (3)$$

where the first and second terms describe third harmonic waves generated collinearly to the input and diffracted fundamental waves respectively, while the third and fourth terms represent noncollinear THG process. When $E_1 \sim E_2$ (e.g., high diffraction efficiency or mixing of two waves of equal intensities), noncollinear terms will dominate due to the different degeneracy factors appearing in the process of wave mixing. This degeneracy will result in a 9-fold decrease of collinear third harmonic irradiance as compared to the noncollinear process.

3.4.1. Efficiency of THG

Let us evaluate the efficiency of THG. Electric field of third harmonic could be expressed as follows:

$$E_3 = \frac{i3\omega}{n_{3\omega}c} \chi_{1111}^{(3)}(-3\omega, \omega, \omega, \omega) 3E_1^2E_2J_3(\Delta k, z_0, z), \quad (4)$$

where SI units and Maker-Terhune convention²¹ for the third order nonlinear susceptibility $\chi^{(3)}$ are used. In this convention the polarization responsible for THG is $\mathbf{P}_{NL} = 4\epsilon\chi^{(3)}\mathbf{EEE}$. E_1 – incident fundamental wave, E_2 – Bragg diffracted fundamental wave, E_3 – noncollinear third harmonic (factor of 3 in front of $E_1^2E_2$ product in eqn. 4 is a degeneracy factor due to wave mixing. Also,

$$J_3(\Delta k, z_0, z) = \int_{z_0}^z \frac{e^{i\Delta kz'}}{(1 + iz'/b)^2} dz', \quad (5)$$

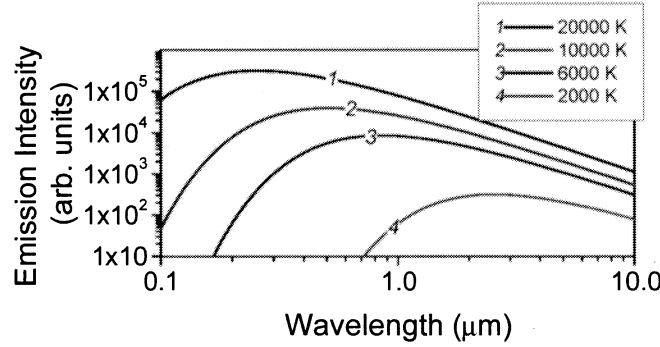


Figure 7: Spectral distribution of blackbody radiation (eqn. 9) at several temperatures.

where z_0 is the value of z at the entrance of the medium, $b = \pi w_0^2/\lambda$ (w_0 is the waist of the beam at focus). In our case b is large compared to the values of z in the integral ($b = 10$ mm for f-number $f\# = 100$ at 755 nm), so that the plane wave approximation can be used:

$$J_3(\Delta k, z_0, z) = \int_{z_0}^z e^{i\Delta k z'} dz' = \frac{2}{\Delta k} e^{i\Delta k L/2} \sin(\Delta k L/2) \quad (6)$$

where $L = (z - z_0) = 1$ mm is the thickness of the sample.

Value of $\chi_{1111}^{(3)}$ for PTR glass is close to $\chi_{1111}^{(3)}$ of BK7 (4.7×10^{-23} m²/V²) and fused silica (3.6×10^{-23} m²/V²) and is assumed to be 4.0×10^{-23} m²/V² in our calculations. $\Delta k_{\text{noncol}} = -12470$ cm⁻¹ and Bragg diffraction efficiency is 30%. Using $I = (nc\varepsilon_0/2)|E|^2$ for irradiance we get:

$$I_3 = \frac{(3\omega)^2}{n_{3\omega} n_{\omega}^3 c^4 \varepsilon_0^2} (\chi_{1111}^{(3)})^2 9 I_1^2 I_2 \frac{\sin^2(\Delta k L/2)}{(\Delta k/2)^2}, \quad (7)$$

after averaging for temporal and spatial Gaussian pulse shape and taking $I_1 = 0.7I_0$, $I_2 = 0.3I_0$ ($I_0 = 5 \times 10^{11}$ W/cm² is incident irradiance) we obtain for the THG efficiency:

$$\eta = \frac{1.323}{3^{3/2}} \frac{(3\omega)^2}{n_{3\omega} n_{\omega}^3 c^4 \varepsilon_0^2} (\chi_{1111}^{(3)})^2 I_0^2 \frac{\sin^2(\Delta k L/2)}{(\Delta k/2)^2} = 4.3 \times 10^{-7}, \quad (8)$$

which is about 3 orders of magnitude lower than what we measured experimentally.¹⁶ This implies that some kind of a new phase matching process is relevant for THG in a Bragg grating illuminated by fs-pulses of high intensity at conditions of Bragg incidence. We conclude that no conventional quasi phase matching mechanism could be established in this case. The presence of the grating is essential for this phase matching to occur. So, that the observed experimentally THG should be a phase-matched process¹⁶ similar to that reported in fs-two wave mixing, where, so called, self-phase matching was observed.²²

Once present, third harmonics can effectively expose photosensitive glass or resist and record a grating. Indeed, the period in the lateral direction for a small intersecting beam angle, 2θ , at the fundamental wavelength, λ , is given by $p_\lambda = \lambda/(\sqrt{2}\sin\theta) \cong \lambda/(\sqrt{2}\theta)$, while for the third harmonic it is, also, the same: $p_{\lambda/3} = (\lambda/3)/(\sqrt{2}\sin(\theta/3)) \cong \lambda/(\sqrt{2}\theta)$. This implies that a three photon processes may be responsible for the formation of volume phase grating with the same lateral period as that produced at first harmonic. Further investigations are planned in order to elucidate the influence of THG by using a 5-beam recording. In that case, the difference in the axial periodicity of the recorded structure should be large for the fundamental and third harmonic wavelengths. This conjecture is currently under investigation.

3.5. Thermal emission of hot electrons

The other mechanism relevant to photo-sensitive glass (or resist) exposure at 800 nm wavelength is a thermal emission of “hot” electrons in a plasma locally created at the high light intensity regions. Thermal emission of a black body at frequency, ν , is given by Plank’s formula²³:

$$I_\nu = \frac{2\pi k_B^3 T^3}{c^2 h^2} \frac{x^3}{e^3 - 1}, \text{ where } x = \frac{h\nu}{k_B T}. \quad (9)$$

Here, h and k_B are Plank and Boltzmann constants, respectively; T is the absolute temperature. It is well established those electrons can possess a non-equilibrium energy of a few electronvolts (1 eV $\rightleftharpoons$ 11605 K) in the case excitation by ultrashort pulses (< 1 ps) of high intensity.²⁴ Energy of these hot electrons can be described in terms of non-equilibrium temperature T_e , which is much higher than that of ions, T_i . Condition $T_e > T_i$ is fulfilled for a time period comparable with or up to several times longer than the pulse duration and it is dependent on pulse irradiance. Frequency of electromagnetic emission, at which the intensity has its maximum for a fixed temperature follows from eqn. 9 and is known as Wien’s law: $\nu_{max} = 2.82k_B T/h$. Hence, the temperature of hot electrons, at which thermal emission has maximum at the 308 nm Ce^{3+} absorption band in PTR glass is 16500 K or just 1.42 eV. Such an thermal emission of electrons should take place at the high intensity regions inside PTR glass. Highly spatially localized plasma state matter created inside the bulk of solid state material by ultra-short pulses should yield in a short (1-100 ps) thermal emission by “hot” electrons. Such an emission can be a probable mechanism of a grating recording in glass/resist at 800 nm wavelength, for which those materials are otherwise transparent. White light emission by non-equilibrium electrons is an attractive light source, which can be “lightened” inside a solid state material (as well as in a liquid or gaseous media for this matter). This can be used, for example, for a 3D optical memory readout. Condition for the creation and dynamics of an ionized plasma inside a solid state media need further investigation.

4. CONCLUSIONS

In conclusion, we have demonstrated a 3D holographic recording of a multi-beam interference pattern inside the resist film using exposure of femtosecond pulses. 3D structures were recorded in a single step, without sample (or laser beam) translation. The mechanism of this 3D recording was a nonlinear process at a 800 nm wavelength of fs-laser emission, most probably a linear absorption of a third harmonic or thermal emission of hot electrons. Identical structures were recorded in PTR glass¹³ and SU-8 resist film. The readout of the recorded structure in glass¹³ indicated that the structure is a phase grating, whose parameters (refractive index modulation, period, and thickness) were consistent with the values calculated from the geometry of the experiment and from Kogelnik’s coupled wave theory.²⁵ This was the first report¹³ to describe a phase hologram recorded in glass using interference of IR irradiation.

4.1. Acknowledgments

We are grateful to Dr. A. Rode for fruitful discussions.

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