Vilnius University
Faculty of Physics
Laser Research Center

Laboratory assignment No. MNFT-4

MODIFICATION OF REFRACTIVE INDEX OF GLASS EMPLOYING ULTRASHORT PULSED IRRADIATION

For master students of Laser Physics and Optical Technologies and Laser Technology programmes

Vilnius 2026

1 Objectives

1. Become familiar with the modification of the refractive index in the volume of glass by femtosecond laser pulses, with the types of the induced modifications (type I and type II modifications, damage) and with the direct laser writing system used to produce them.

2. Understand how the writing parameters – the average laser power, the writing velocity and the polarization – influence the modifications, and why soda-lime glass and fused silica respond to them differently.

3. Gain practical experience in creating modifications in the volume of glass (below its surface) and in characterizing them by optical microscopy (including observation between crossed polarizers) and by diffraction measurements.

2 Tasks

1. Using the direct laser writing system, fabricate line gratings in the volume of fused silica and soda-lime glass with varied average laser power and, in fused silica, with two different writing velocities and two orientations of the laser beam polarization.

2. Explore the effect of the laser beam polarization: in both materials, fabricate squares of closely overlapping lines with different orientations of the laser beam polarization and different average laser powers.

3. In soda-lime glass, fabricate multilayer gratings with different periods and average laser powers for the diffraction measurements.

4. Using an optical microscope (bright field and crossed polarizers), evaluate the modifications produced with different exposure parameters: identify the types of the induced modifications (type I, type II, damage), measure the width of the modification as a function of the average laser power and examine the squares written with different polarization.

5. Using the optical characterization setup (HeNe laser and screen), measure the diffraction efficiency of the multilayer gratings, calculate their periods from the diffraction pattern and compare them with the values set in the fabrication program.

6. Compare the results obtained in fused silica and soda-lime glass.

3 Questions for the defence

1. A typical optical setup for laser modification of glass: its main components and their purpose.

2. Modifications in transparent materials and their types.

3. How can type I and type II modifications be distinguished with an optical microscope?

4. Parameters of the laser irradiation required to induce modifications.

5. Influence of the laser beam polarization on the modification of glass with ultrashort laser pulses.

6. Phase gratings written in glass.

7. Applications of the modifications in transparent materials.

4 Equipment and materials

During the laboratory assignment a direct laser writing (DLW) system with the femtosecond laser "Pharos" will be used (Fig. 1). The polarization of the laser beam is rotated with a half-wave ($\lambda/2$) plate in a rotation mount. A power meter is used to measure the average laser power. The modifications are formed in soda-lime glass (microscope slides, 1 mm thick) and in fused silica samples (the thickness may vary from sample to sample). The fabricated structures are characterized with an optical microscope equipped with crossed polarizers and with an optical characterization setup consisting of a HeNe laser ($\lambda = 632.8$ nm), a screen and a ruler.

Other laboratory supplies: microscope slides and adhesive tape (for fixing small samples), tweezers, isopropanol and cleaning tissues (for cleaning the samples), disposable gloves and laser safety goggles.

The generalized scheme of the DLW system is depicted in Fig. 1. The main component of the system is a femtosecond solid-state laser "Pharos" (*Light Conversion, Lithuania*). The active medium is an Yb:KGW crystal, and the central wavelength of the fundamental radiation is $\lambda_f = 1030$ nm. The laser generates pulses of $\tau_{imp} < 300$ fs duration. The repetition rate can be adjusted in the range of 1–200 kHz. The experiments are performed using the fundamental radiation, which is directed straight to the focusing objective.

The average power of the laser is set by a two-stage attenuator. The first stage, consisting of a manually rotated $\lambda/2$ phase plate and a Brewster angle polarizer, is used to set the approximate average power of the beam entering the optical system. The second stage, consisting of a $\lambda/2$ phase plate housed in a computer-controlled rotator and a Brewster angle polarizer, is for precise average power adjustment during laser writing. Automatic power calibration is performed by placing the power meter sensor in the path of the laser beam. A $2\times$ beam expander *BE* is installed in the beam path to ensure that the beam fills the aperture of the focusing objective. The beam is focused into the volume of the glass with a dry (non-immersion) objective, which will be specified by the instructor. The fabrication process is imaged coaxially through a dichroic mirror with the *CMOS* camera.

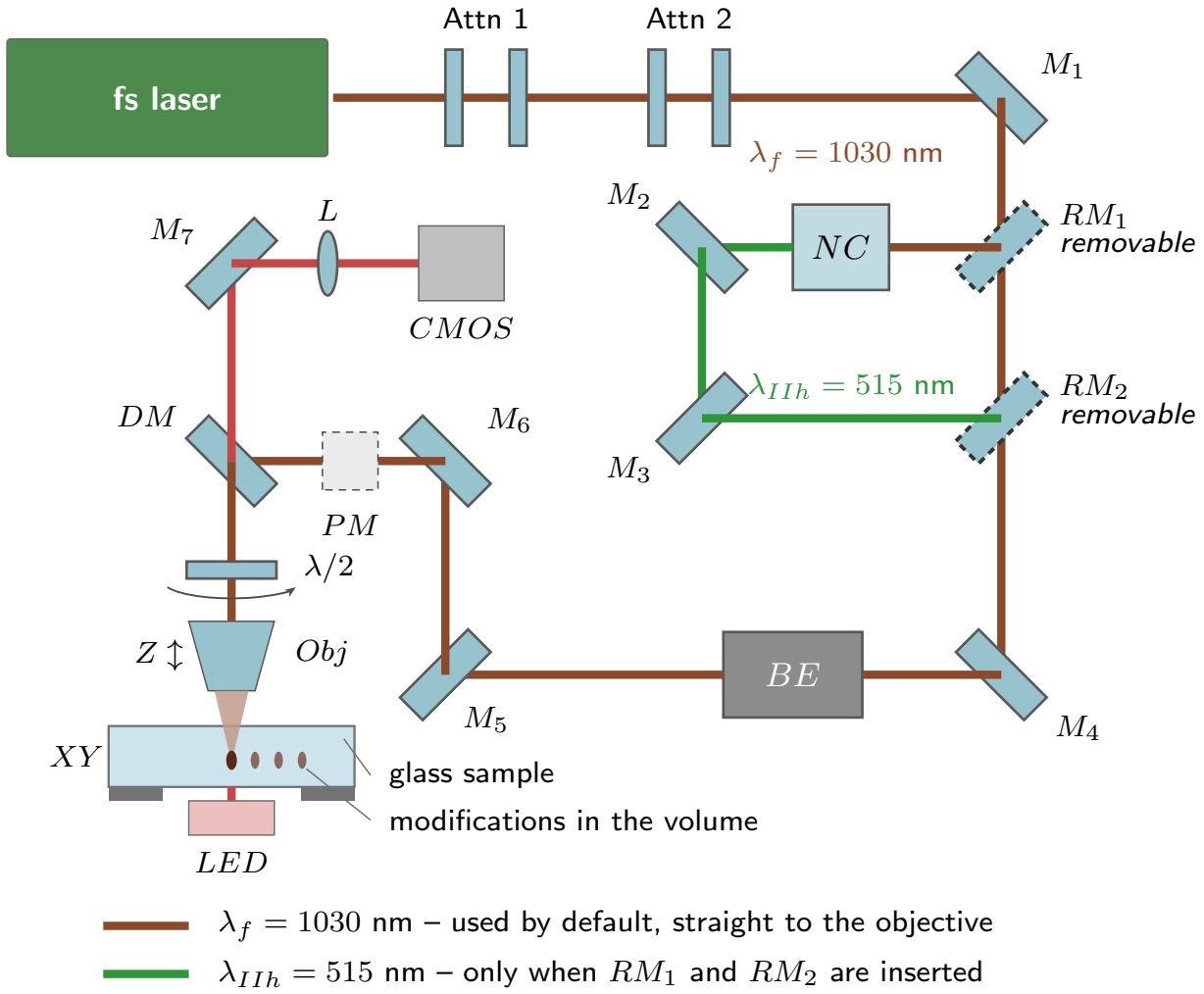


Fig. 1 Generalized scheme of the DLW system; only the main components are shown, and the actual layout may differ. The beam reaches the objective at the fundamental wavelength $\lambda_f = 1030$ nm; the second harmonic $\lambda_{IIh} = 515$ nm is obtained only when the removable mirrors RM_1 and RM_2 are inserted, routing the beam through the nonlinear crystal NC. Meanings of the abbreviations: fs laser – femtosecond laser source, Attn 1, Attn 2 – power attenuator stages, M_1–M_7 – mirrors, RM_1, RM_2 – removable mirrors, NC – nonlinear crystal, BE – beam expander, PM – power meter, DM – dichroic mirror, $\lambda/2$ – rotatable half-wave plate, Obj – objective, L – lens, $CMOS$ – camera, LED – LED illumination source, XY, Z – sample translation coordinates.

Because of the Brewster angle polarizers of the attenuator, the beam reaching the objective is linearly polarized. The polarization is rotated using the half-wave plate mounted in a rotation mount directly above the objective. A half-wave plate rotates the direction of linear polarization by twice the angle between the incident polarization direction and the plate's fast axis. Consequently, rotating the plate by an angle Δ rotates the polarization of the transmitted beam by 2Δ, regardless of the initial polarization direction. Thus, rotating the plate by 15°, 30° and 45° rotates the polarization direction

by 30°, 60° and 90°, respectively.

Submicrometric precision positioning stages ANT130-110 (*Aerotech, USA*) with linear motor drives are used for the translation of the sample in the XY plane of the laser beam waist. Translation in the Z axis is performed by positioning the focusing objective with an ANT130-060 linear stage. Such a positioning system ensures ± 75 nm position repeatability in each axis and allows the samples to be translated at a maximum velocity of 300 mm/s. The sample is held on the stages by a vacuum suction holder. The "3DPoli" (*Femtika, Lithuania*) software package is used to automate the system.

5 Workflow

Before the laboratory assignment, remember laser safety requirements. In this work you will use a class 4 laser, which means that even scattered radiation can cause eye or skin injuries if the safety requirements are not fulfilled.

All the structures in this assignment are written in the volume of the samples, below the surface. The work is done in the following order: first all the structures are written in fused silica (line gratings and squares), then in soda-lime glass (line gratings, squares and multilayer gratings for the diffraction measurements). Both samples are then examined with an optical microscope, and finally the diffraction of the multilayer gratings is measured with a HeNe laser.

The workflow below gives the order of the work and the approximate sizes of the structures. It may change depending on the situation in the laboratory – always follow the assignment sheet given to you by the instructor at the beginning of the work: it contains all the detailed instructions and the fabrication parameters (average powers, periods, number of layers, $\lambda/2$ plate positions). The fabrication programs are prepared in advance and will be provided to you – you do not need to write them yourself; only the writing parameters at the end of each program file are changed, as given in the assignment sheet.

For every written structure record the average power, the writing velocity, the period (or line spacing dx), the number of layers and the distance between them dz, and the $\lambda/2$ plate position – you will need them for the report.

1. **Checking the laser system.** Together with the instructor, verify that the system is ready to use: the wavelength (1030 nm), the pulse duration (300 fs), the pulse repetition rate (100 kHz) and the beam alignment.

2. **Computer preparation.**

   (a) Start the *3DPoli* software and the „wxPropView“ camera program used to observe the fabrication process live. The instructor will help with the first launch. For convenience, the camera window is displayed on the auxiliary system monitor.

(b) In the *3DPoli Fabrication* program, check the fabrication settings: home all the axes and calibrate the laser power. Check the parameter file for the objective in use if needed.

3. **Placing the fused silica sample.** Clean the fused silica sample with isopropanol and cleaning tissue, so that no dust or fingerprints remain on its surface; depending on the size of the sample, it may need to be taped to a microscope slide first. Put the sample into the holder of the translation stages and turn on the vacuum suction, which keeps the sample in place during the fabrication. Put the $\lambda/2$ plate into the rotation mount directly above the objective and set it to its reference (0°) position given by the instructor.

4. **Finding the surface of the sample.** Lower the objective towards the sample with the *3DPoli Fabrication* program, first in larger and then in smaller steps, while watching the camera image, until the upper surface of the glass (dust particles, scratches) is seen – this is the approximate position of the surface. NOTE: to get a clear image of the surface, it may be necessary to move the lens in front of the camera. Then set the average power given in the assignment sheet, open the laser shutter and move the objective up and down until a dark dot appears in the camera image: the sample has been damaged, so the focal spot is at or below the surface. Now find the surface more precisely: move the sample to the side in 5 μm increments and, at every new position, lift the objective by a 5 μm step in Z. While the focal spot is still in the glass, a dark dot appears at each new position; once the objective has been lifted too far from the glass, the dot no longer appears, because the focal spot is above the sample. The Z position at which the dots stop appearing is approximately the surface of the sample. Close the laser shutter and move the objective towards the glass by the distance given in the assignment sheet; the structures are written at this depth.

5. **Line gratings in fused silica.** Keep the $\lambda/2$ plate in the reference position.

   (a) Write small (approximately 100×50 μm) single-layer line gratings at a writing velocity of 100 μm/s, one grating per average power, varying the average power.

   (b) Write a set of larger (approximately 300×50 μm) single-layer line gratings with the same average powers at a writing velocity of 1000 μm/s.

   (c) Turn the $\lambda/2$ plate by 45° (the polarization rotates by 90°) and write a copy of the gratings of step 5b. Then return the plate to the reference position.

6. **Squares with different polarization in fused silica.** Write an array of squares (approximately 50×50 μm) filled with closely overlapping parallel lines, at a writing velocity of 100 μm/s. The columns of the array are written with different average powers, the rows with different positions of the $\lambda/2$ plate: 0°, 15°, 30° and 45°, i.e. with the polarization rotated by 0°, 30°, 60° and 90°. Start with the plate in the reference position; after each row the program pauses –

during the pause turn the plate by 15°. When the fabrication is finished, lift the objective away from the sample, turn off the vacuum suction and only then remove the sample from the holder.

7. **Placing the soda-lime glass sample.** Clean the soda-lime glass sample (a 1 mm thick microscope slide) with isopropanol, put it into the holder, turn on the vacuum suction, set the $\lambda/2$ plate back to the reference position and find the surface of the sample in the same way as for fused silica (step 4).

8. **Line gratings in soda-lime glass.** Write single-layer line gratings (approximately 300×50 μm) at a writing velocity of 1000 μm/s, one grating per average power, varying the average power.

9. **Squares with different polarization in soda-lime glass.** Write an array of squares in the same way as in fused silica (step 6). Then return the $\lambda/2$ plate to the reference position.

10. **Multilayer gratings for the diffraction measurements in soda-lime glass.** Return to the surface of the sample and move the objective deeper into the glass, as given in the assignment sheet. Fabricate several large (approximately 1×1 mm) gratings consisting of several layers, varying the grating period and the average power, at a writing velocity of 2000 μm/s. When the fabrication is finished, lift the objective away from the sample, turn off the vacuum suction and remove the sample.

11. **Characterization of the samples with an optical microscope.** Examine both samples in bright field (BF) and between crossed polarizers (CP).

    (a) *Line gratings.* In BF, take overview images of each set of gratings and images of the grating written with each power; measure the width of the modification and note at which powers damage (cracks, irregular, light-scattering lines) appears. In CP, examine the gratings written with different powers and determine which type of modification (type I, type II or damage) was induced: an isotropic (type I) modification remains dark, except for possible bright halos at the edges caused by stress-induced birefringence, whereas a birefringent (type II) modification appears bright. Compare the gratings written with different writing velocities (steps 5a and 5b) and with the polarization rotated by 90° (steps 5b and 5c) – did anything change?

    (b) *Squares.* In BF, take an image of the whole array of squares. In CP, observe the array while turning the analyzer and the polarizer (keeping them closed all the time), and note which squares go dark and how their brightness changes. Compare the effect of the polarization in fused silica and soda-lime glass.

12. **Diffraction measurements with a HeNe laser.** For each multilayer grating in soda-lime glass:

    (a) measure the power of the incident HeNe beam and of the beam transmitted through the grating;

(b) with the beam at normal incidence to the grating, measure the distance x_1 between the zero- and first-order diffraction maxima on the screen and the distance L from the sample to the screen, calculate the diffraction angle $\theta_1 = \arctan(x_1/L)$ and the grating period from the diffraction equation

$$m\lambda = \Lambda \sin\theta_m, \tag{1}$$

where m – order of the diffraction maximum ($m = 1$), $\lambda = 632.8$ nm – wavelength of the HeNe laser, Λ – period of the grating, θ_m – angle between the normal to the grating and the direction to the m-th diffraction maximum. Compare the calculated period with the one set in the fabrication program;

(c) tilt the grating to the angle at which the diffraction is strongest and measure the power of the stronger first-order diffraction maximum; calculate the diffraction efficiency as the ratio of this power to the incident (or transmitted) beam power.

13. **Report and conclusions.** In the report, give the fabrication parameters of every written structure (power, writing velocity, period or dx, number of layers and dz, $\lambda/2$ position), the microscope images (BF and CP) and the graphs of the modification width as a function of the average power. Compare your observations in fused silica and soda-lime glass, discuss your results and write the conclusions.

6 Theory

6.1 Modification of transparent materials

The existence of a threshold intensity at which laser damage occurs opens up unique possibilities for processing transparent materials. By controlling the laser pulse intensity, conditions may be selected so that the threshold intensity is exceeded only in the focus of the focusing lens. In this way, the area of laser-induced damage will be strictly confined to a defined region in the volume of the transparent material. By gradually changing the position of the focus in a transparent medium it is possible to form various three-dimensional objects, photonic elements or integrated optical systems with micrometric or submicrometric resolution.

In the literature, the term laser-induced material damage commonly refers to an irreversible change in the properties of a medium caused by laser radiation, normally detected by optical methods. However, under certain conditions, structures form in the material that stand out by their extremely high regularity, are homogeneous and can be clearly classified. Such structures are formed when a laser pulse whose intensity is close to the damage threshold changes the structure of the material in a regular manner. To single out these structures from the others, the term laser-induced material modification will be used in this description. The concept of material damage will be used only for irregular formations, visually detectable cracks in the material and so on.

6.2 Volumetric material modification

To achieve the laser pulse intensities greater than TW/cm^2 required to modify the material, the laser beam inevitably has to be focused. Focusing, on the other hand, makes it possible to selectively choose the regions where the radiation intensity is sufficient for nonlinear absorption.

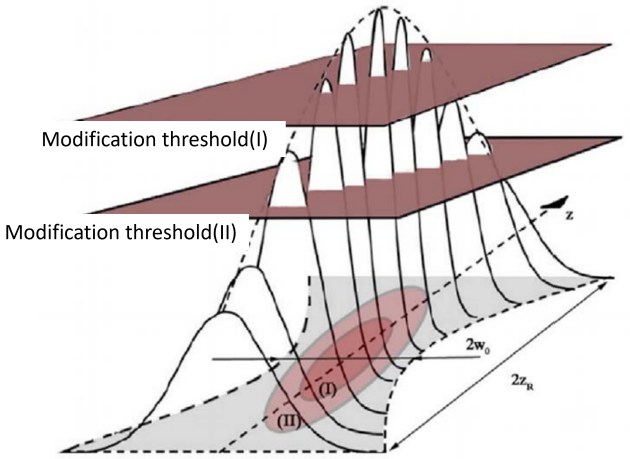


Fig. 2 Gaussian beam intensity distribution in the focus (z – beam propagation direction). The material can be modified only in the limited region where the threshold intensity is exceeded. Depending on the threshold value (or on the radiation intensity), the size of the modified region may be smaller than the diffraction-limited diameter ($2w$) or the waist length ($2z_r$).

Diffraction limits the minimum region of space into which the laser beam can be focused. If a plane wave is focused with a lens of numerical aperture NA, the minimum possible beam radius in the beam waist is approximately $w = 0.61M^2\lambda/NA$, where λ – wavelength, M^2 – beam quality parameter. This size in fact determines the radius of the formed Airy disk. The profile of the laser beam is usually closer to a Gaussian distribution, but if the spatial energy distributions of such beams were compared, the difference would be very small: in the case of the plane wave, 83 % of the energy is concentrated in the Airy disk, and in the case of the Gaussian beam, 86 % of the energy falls within the $1/e^2$ level of the Gaussian envelope. Therefore, the formula above describes the size of the focal spot quite accurately, although it does not take the aberrations of the focusing optics into account. The waist length of the Gaussian beam, the so-called confocal parameter, corresponds to twice the Rayleigh length (the distance at which the on-axis radiation intensity decreases by half): $2z_r = 2\pi nw^2/\lambda$, where n – refractive index of the medium. As already mentioned, in order to modify a material, it is necessary to reach a radiation intensity above a certain threshold. As can be seen from Fig. 2, it is easy to realize a situation where the threshold intensity is exceeded only in the focus. Thus, it is possible to create modified structures in the volume without damaging the surface of the material. When the intensity is chosen close to the damage threshold, it is possible to create structures that are smaller than the diffraction-limited beam diameter. Such a technology is particularly successful in

the creation of three-dimensional structures with submicrometric resolution in polymers. A similar resolution can also be achieved in glasses, but due to nonlinear processes such as light filament formation the modified region of the material increases. Nevertheless, the formation of three-dimensional microstructures is successfully implemented.

6.3 Types of modifications

By selecting different parameters of the laser radiation, various types of modifications can be created in a transparent material. Although the nature of the modified region often depends on the physical properties of the substance, certain types of modifications can be distinguished (Fig. 3). When a substance is exposed to a pulse whose intensity is lower than the modification threshold, short-lived unstable changes, manifested by altered absorption or refractive index, may already occur in the medium. Such modifications are called unstable. When the radiation intensity only just exceeds the threshold for the formation of a permanent modification, a homogeneous region with an altered refractive index is produced, also known as a type I modification. With increasing intensity, the homogeneity of the modified region begins to decrease, and birefringence of light appears (type II modification). By increasing the intensity even more and selecting appropriate focusing conditions, microcavities can be formed in the material. Illustrations of the modifications are provided in Fig. 4. In some materials one type of modification may predominate and another may not occur at all. In other materials, such as fused silica, all types of modifications can be realized.

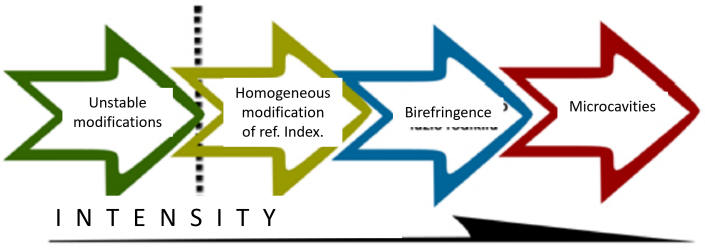


Fig. 3 Types of modifications induced by ultrashort laser pulses.

Modification of the refractive index

When the intensity exceeds a certain critical value, the modification of the substance becomes permanent. Regularly formed transparent modifications in various types of glass were demonstrated for the first time by Davis et al. in 1996. The induced modifications were found to have an isotropic change of the refractive index that can reach up to 10^{-2}. Such a change is sufficient to form waveguides in the medium, which were demonstrated by the same group of scientists in later works. These works greatly pushed the development of integrated optical technology. Despite abundant research,

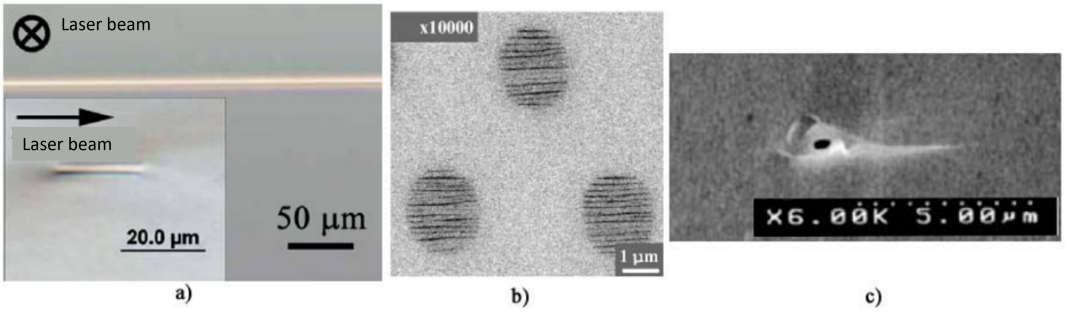


Fig. 4 Materials modified by ultrashort laser pulses: (a) optical waveguide written in phosphate glass (longitudinal and transverse view, type I modification); (b) nanograting in fused silica (type II modification); (c) microcavities in fused silica.

the physical mechanisms that lead to changes in the refractive index are still not fully understood today. Many different processes that can change the refractive index in one way or another can take place in a solid medium exposed to an intense laser pulse. There are three possible models describing changes in the refractive index: the thermal model, the colour centre (defect) model and the lattice deformation model (Fig. 5).

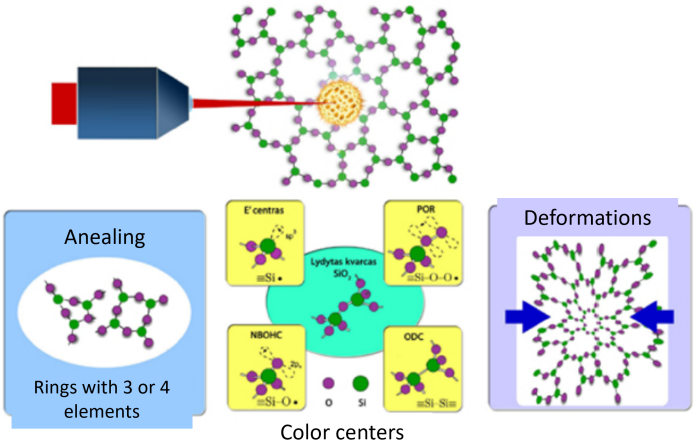


Fig. 5 Mechanisms of the refractive index change in fused silica. Material affected by intense radiation may become denser after remelting: due to a change in the chemical structure, 3- and 4-membered rings are formed instead of the dominant 5- and 6-membered rings. Point defects – colour centres – may also be formed. On rapid heating, internal pressure builds up in the substance, resulting in deformation of the lattice. All of these phenomena affect the change of the refractive index of the material.

Thermal model

This model is based on the fact that the temperature of the material in the focus can become high enough for a phase transformation of the material to take place. The thermal conductivity of a dielectric medium is low and the hot zone is concentrated in a small region of space; therefore, even at low pulse energies (500 nJ), the material can be heated up to 2000 K and more. After heating, the region cools rapidly due to the high temperature gradient (typical relaxation time > 1 μs). In this way, the medium is remelted and acquires the properties of a supercooled liquid, which differ from the properties of the unaffected material. The refractive index of fused silica is known to increase (by $\sim 10^{-3}$) if the silica is rapidly cooled from a high-temperature state. This change is due to an anomaly of this material – compaction: the fused silica network consists of rings formed from –O–Si–O– units. Ordinary fused silica is dominated by 5- and 6-membered rings, but in silica rapidly cooled from a hot state the number of 3- and 4-membered rings increases significantly, making the material denser. Indeed, Raman spectroscopic studies of the laser-modified zone confirm this ring formation, and experimental work demonstrates an increase of the refractive index in fused silica. In most other glasses, by contrast, the supercooled melt phase has the usual lower density, so a decrease of the refractive index is expected after exposure to a laser pulse; experiments in borosilicate, phosphate and other glasses indeed confirm such a decrease. Laser heating also causes a redistribution of ions in the material, which affects the change of the refractive index as well.

Such an interpretation of the refractive index modification, based on the thermal behaviour of the material, allows the possible changes of the refractive index to be predicted qualitatively, but it is far from sufficient to explain all the features of the modified region. Microscopic analysis of the modified regions shows that the distribution of the refractive index in the region affected by the laser pulse is quite complex: various variations of the refractive index are observed, which, moreover, depend strongly on the irradiation parameters. Typically, if there is an increased refractive index at the centre of the modification, regions of reduced refractive index are observed at the periphery, and vice versa. By selecting appropriate pulse parameters, zones with opposite signs of the refractive index change can be induced in the same material. This suggests that laser pulse absorption induces rapid dynamic processes that cause additional lattice deformations, which also influence the change of the refractive index.

Deformation model

Relaxation of the free electrons generated by a femtosecond pulse proceeds very rapidly (< 1 ps). Immediately after relaxation, a large amount of energy is quickly transferred to the lattice; therefore its temperature rises and a high thermoelastic pressure is created in a confined space. The relaxation of the stress in the material can launch strong acoustic waves that can induce plastic deformations of the lattice. The existence of such waves has been confirmed experimentally. The propagating wave interacts with the heated lattice and creates inhomogeneous density variations that affect the change

of the refractive index. Such compression is also confirmed by the change of the molecular bond angle in SiO_2 recorded in Raman spectra. Local changes in density inevitably cause a stress field in the surrounding region not affected by the laser, which is visible under a polarizing microscope. However, simulations of the refractive index change showed that lattice deformations account for only about 10 % of the total observed refractive index change; hence this model alone cannot explain in detail the origin of the refractive index change.

Colour centres

The development of electron paramagnetic resonance (EPR), luminescence and light absorption measurement methods in the middle of the last century allowed the identification of various glass defects and the observation of their dynamics under exposure to various radiation, bombardment with high-energy particles, etc. Glasses have an amorphous structure, which makes them rich in various lattice defects. Even glasses of the same composition can have a different thermal history, so the nature and amount of defects also differ. Therefore, the study of defects is quite complicated.

In the case of pure fused silica, the four most common groups of defects can be distinguished: E′ centres (≡Si•, where • stands for an unpaired electron), non-bridging oxygen hole centres – NBOHC (≡Si–O•), peroxy radicals – POR (≡Si–O–O•) and oxygen deficiency centres – ODC (≡Si–Si≡).

All of the listed defects can be induced both by ultraviolet laser radiation and by femtosecond infrared radiation. Defects of a similar type are found in other glasses: germanosilicate glass is dominated by germanium–oxygen hole centres, borosilicate glass by boron–oxygen hole centres and so on.

The influence of the colour centres on the change of the refractive index is debatable. EPR measurements show that up to 10^{19} cm^{-3} defects may be formed in the exposed material, which would correspond to a refractive index change of up to 10^{-3}. On the other hand, direct absorption measurements show that the defects cause a much smaller change of the refractive index (10^{-8}). These differences are most likely caused by different laser pulse and focusing parameters; however, it is generally agreed that the colour centres are not stable in the long term and can be destroyed by heat treatment (annealing at 200 °C), whereas the refractive index modifications themselves are relatively stable up to 700 °C.

The actual change of the refractive index can probably be explained only by all three models together. The change of the refractive index is usually thought to be due to the thermal action on the material, which in turn causes lattice deformations. However, the thermal model cannot fully explain the occurrence of a refractive index change under laser radiation of low intensity (insufficient to cause a phase transition) but high pulse repetition rate. On the other hand, the deformation and colour centre models predict a much smaller change than is observed.

Birefringence. Nanogratings

At higher energies, the homogeneity of the modified zone decreases; moreover, these modifications are characterized by birefringence of light. To distinguish them from the others, these modifications were named type II modifications. Only relatively recently has the cause of the birefringence been found: it results from the nanostructuring that takes place in the region affected by the laser.

Self-organized nanostructures formed on the surface of materials under laser irradiation were noticed already at the dawn of laser research. Surface nanogratings (the so-called “ripples”) that appear during ablation have a period close to the laser wavelength and an orientation that depends on the polarization of the radiation; they are observed in practically all materials – metals, semiconductors and dielectrics. Analysing type II modifications in fused silica, Shimotsuma et al. found that nanogratings can also form in the bulk of the material. The lines of such a nanograting are < 10 nm thick, its period is several times smaller than the wavelength, and its orientation is perpendicular to the polarization of the incident radiation. The density of the material varies periodically in the nanograting, and the lines of the grating are etched in hydrofluoric (HF) acid up to 300 times faster than the unaffected material between them. Similar gratings were also observed in tellurium dioxide (TeO_2); in other materials such nanogratings had not been registered at the time [1].

The physics of the formation of such nanostructures is not yet understood, as there is still a lack of experimental data. One interpretation is based on the fact that the interference of the intense electromagnetic wave with the electric field generated by the electron plasma can form such structures. Because of this interference, the laser light modulates the plasma density and essentially divides the plasma into separate nanoplanes, which are separated from each other by a period equal to half the light wavelength in the medium. In other works, however, the grating period is much smaller than half a wavelength. In addition, it has been observed that the grating period is affected by the pulse repetition rate. These results call into question the interference theory of grating formation, and the formation of the plasma nanoplanes is instead explained by plasmonic effects. The conditions required for such a plasma density distribution are unclear. It should be noted that the periodicity and regularity of the gratings are maintained over macroscopic distances when the sample is slowly translated during writing with respect to a fixed focal position. Therefore, it can be said that the redistribution of the plasma density can also be guided by material defects: the defects created by one laser pulse "remember" the primary plasma distribution and pass this information on to the following pulses arriving at the same place or during slow translation of the sample.

The presence of nanogratings oriented in one direction explains the emergence of birefringence in type II modifications. By changing the radiation polarization, the intensity and the modified area, this birefringence can be controlled. It has already been shown that phase plates can be formed on this principle.

The influence of the polarization was studied in fused silica with the same type of laser system as the one used in this assignment (Yb:KGW laser "Pharos", 1030 nm, 300 fs, 100 kHz, 0.42 NA

lens) [1]. Lines written there were observed with an optical microscope in the phase contrast and crossed-polarizer regimes. Under crossed polarizers, the lines rotate the polarization of the transmitted light, and this effect grows with the pulse energy; it is caused by birefringence and is one of the indicators of the type of modification. In type I modifications birefringence may appear only at the periphery of the modified zone, where it is caused by stress, whereas in type II modifications it is caused by nanogratings. Even at the lowest pulse energies above the modification threshold, the lines written in fused silica were type II modifications.

The nanogratings are always oriented perpendicular to the polarization of the writing beam, and their periodicity is maintained over distances much larger than the modified zone. When the polarization was parallel to the sample translation (writing) direction, the period of the nanograting was 260 nm; when it was perpendicular, the period was somewhat larger, 340 nm. Thus, when a line is written with the polarization parallel to the writing direction, the nanoplanes are perpendicular to the line, and when the polarization is perpendicular to the writing direction, the nanoplanes lie along the line – the optical axis of the induced birefringence follows the polarization, not the writing direction. Between crossed polarizers the brightness of a birefringent region is proportional to $\sin^2 2\varphi$, where φ is the angle between its optical axis and the polarizer; therefore, rotating the writing polarization shifts the angle of the crossed polarizers (or of the sample) at which the structure goes dark, but a rotation by exactly 90° cannot be distinguished with crossed polarizers. Stress birefringence, in contrast, follows the geometry of the structure. Besides the orientation, the polarization relative to the writing direction also changes the nanograting itself (its period, regularity and the resulting retardance), which shows up as differences in brightness and uniformity. A regular nanograting forms only when each point of the material is exposed to a large number of pulses: homogeneous lines were obtained when at least 100 pulses hit the same spot (translation velocity < 1 mm/s at 100 kHz). With fewer pulses the nanogratings form irregularly or not at all, the birefringence under crossed polarizers weakens and becomes non-uniform, and random light-scattering centres appear along the line. Because nanogratings had been observed only in fused silica and TeO_2, the effect of the polarization in soda-lime glass may be weak or absent – comparing the two materials is one of the tasks of this assignment.

6.4 Influence of radiation parameters on modifications

Modifications induced by ultrashort laser pulses were discussed in the previous section. However, we did not analyze in detail the laser radiation parameters required to create such modifications. Radiation intensity, wavelength, pulse duration, pulse repetition rate, focusing sharpness, optical and physical properties of the material and other parameters may affect the type or quality of the induced modifications. Moreover, the physical mechanisms that determine the occurrence of modifications are not yet fully understood, which complicates the analysis of such light–matter interaction. Therefore, to induce the desired type of modification with a selected laser system, it is still necessary to select the required radiation parameters experimentally. Nevertheless, the relatively wide range of research

on the modification of transparent media allows the influence of the essential writing parameters on the formation of modifications to be understood.

Most of the studies were conducted using the most popular Ti:sapphire-based laser systems. They generate pulses at 800 nm wavelength, the duration of which is less than 100 fs. In such systems the pulse repetition rate is low, typically 1 kHz. With this type of system, homogeneous refractive index modifications in fused silica are observed at pulse intensities of about 40 to 200 TW/cm^2 (fluence 4 to 20 J/cm^2). Type II modifications start to occur at intensities between 200 and 600 TW/cm^2 (20-60 J/cm^2), and microdamage occurs at intensities greater than 600 TW/cm^2 ($>$ 60 J/cm^2). These values are obtained by focusing the radiation with a lens with a numerical aperture of 0.65.

The intensity can be varied by using focusing optics of different sharpness, but the focal length of the lens itself can greatly change the nature of the created modification. When lenses of small numerical aperture ($NA < 0.2$) are used, the modification threshold is reached only when the radiation power already exceeds the self-focusing threshold of the material. The resulting modifications are then heavily affected by the phenomenon of filamentation. During filament formation the energy is redistributed over a large region, so the longitudinal dimension of the modification changes significantly and can even reach a few millimetres. The increased concentration of free electrons in the focus reduces the dielectric constant of the material, so the radiation is always partially defocused, and filamentation begins even when lenses of medium focusing sharpness ($NA \approx 0.4 - 1$) are used. For this reason the morphology of the modification becomes inhomogeneous. In the region close to the focus the change of the refractive index is usually larger, and its dimensions are close to, or even smaller than, the waist width of the focal spot or the depth of focus. Beyond this zone, a weakly modified region forms along the direction of pulse propagation; its length depends on the parameters of the focusing lens. Thus, the formation of regular microcavities is possible only with very sharply focusing lenses ($NA > 1$), because, in order to initiate microexplosions, all the radiation energy must be concentrated in the smallest possible volume, and therefore filamentation must be kept to a minimum.

As already mentioned, the relaxation times of a substance exposed to ultrashort laser pulses reach microsecond scales. Therefore, when laser systems with a high pulse repetition rate are used, thermal accumulation is possible, because the material does not have time to cool down before the next pulse arrives. This accumulation changes the nature of the modification: under such conditions the modified region becomes larger and its dimensions start to depend on the duration of the laser exposure. It is important to note that this effect is not observed in all materials: it is particularly pronounced in borosilicate glasses but very weak in fused silica. This material selectivity of the effect is not fully understood, but it is believed that the reason could be the larger band gap of fused silica and its higher phase transition temperature (1800 °C). The pulse repetition rate at which the thermal accumulation effect becomes noticeable is 0.5 MHz (pulses arrive in the material every 2 μs). At lower repetition rates (< 0.2 MHz) this effect is observed only at intensities significantly exceeding the damage threshold.

Another important parameter that determines the type of modification is the pulse duration. Here

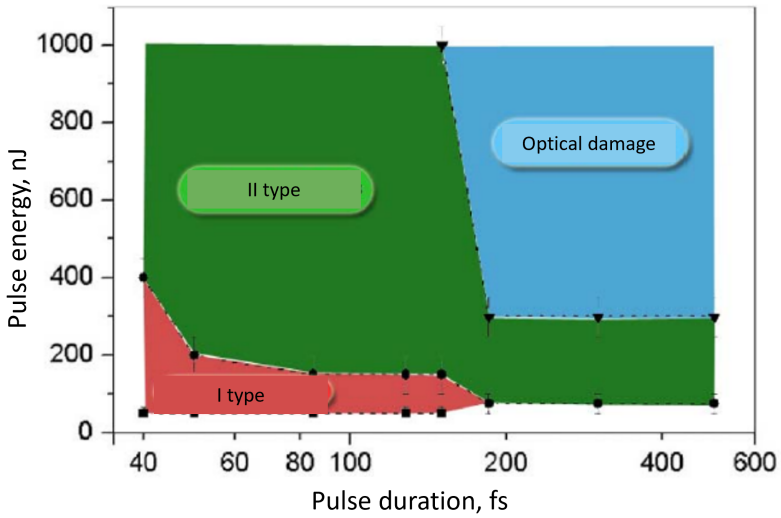


Fig. 6 Influence of the laser pulse duration on the type of modification in fused silica (radiation wavelength 800 nm, focused with a 0.65 NA lens).

we will discuss only the experimental results related to the induction of refractive index modifications. The change of the modification morphology with the pulse duration has not been studied consistently. Only a few works have addressed this problem, and the writing regimes of the modifications in them differ considerably. In a study of modifications induced in fused silica by a light filament, using weakly focusing optics ($NA < 0.1$), it was found that the refractive index modification (the type of this modification was not investigated in that work) can be induced only when the pulse duration is shorter than 250 fs. At longer pulse durations, strongly light-scattering optical damage is formed. The influence of the pulse duration on the morphology of modifications made with a sharper focusing objective was investigated by Hnatovsky et al. It can be seen in Fig. 6 that type I modifications are formed only at durations shorter than 200 fs; at longer pulse durations the modifications already exhibit birefringence. In borosilicate glass, in which no type II modifications are observed, homogeneous changes of the refractive index are also obtained with pulses lasting a few picoseconds.

The effect of the radiation wavelength on the induction of modifications remains highly debatable. When the thresholds of modifications induced with different wavelengths are measured in different transparent materials, no significant difference is observed: the difference between the thresholds at 800 nm and 400 nm was only 10 %. According to Keldysh theory, shorter wavelengths increase the probability of multiphoton ionization, whereas at longer wavelengths the tunnelling ionization rate increases accordingly. Based on these considerations, it can be said that the overall nonlinear absorption coefficient varies little with the wavelength. However, there is still a lack of research that could provide a stronger basis for this claim.

6.5 Photonic elements in transparent materials

It has been observed that ultrashort laser pulses can be used to modify the optical properties of transparent media. Even without a definitive understanding of the nature of this phenomenon, the potential applications of this technology have been demonstrated by a great variety of photonic elements written in various types of glass. A detailed overview of the elements created by this method can be found in the review by Della Valle et al. and the references therein. Therefore, in this section we will review only the essential applications demonstrating the use of various types of modifications for practical purposes.

Already the very first works on the modification of transparent media, carried out by the group of Hirao, demonstrated the waveguiding effect due to the locally altered refractive index of the material. By slowly translating the sample relative to the focal position, a waveguide is formed directly; its shape and structure depend on the translation geometry of the sample and on the parameters of the writing beam. Although different materials respond differently to femtosecond pulses, various technological solutions allow waveguides to be written in various types of materials: fused silica, borosilicate glass, multicomponent glasses, nonlinear crystals, polymers, etc. The flexibility of this technology is demonstrated by more complex photonic elements such as integrated multichannel waveguide splitters and combiners, interferometers and other microdevices. In terms of efficiency, such elements still lag behind waveguides integrated on silicon substrates, but that technology is not as universal and its application possibilities are rather limited. Direct writing technology is promising for the production of unique elements. As an example, a waveguide written in fused silica in the same processing step together with a Bragg grating may be given.

Another wide field of application is the production of diffractive elements. Thin one-dimensional gratings, volume Bragg gratings, Dammann gratings, Fresnel lenses and holograms written with femtosecond pulses have been demonstrated in various types of glass. Usually such photonic elements are not very efficient, as there is currently no method that can accurately control the required change of the refractive index and the homogeneity of the modification itself. Other types of modifications are also used in various fields. For example, microcavities can be used as bits of information that could compactly fill the whole volume of a transparent material. Type II modifications make it possible to create polarization-sensitive waveguides or gratings and other elements; moreover, their sensitivity to etching in HF acid enables the formation of microchannels for microfluidic applications. Despite this, applied works based on the modification of transparent materials by ultrashort laser pulses show the great potential of this technology.

The theoretical part of this description is adapted from the PhD dissertation of Dr. Domas Paipulas [1], where the original references can be found.

References

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