VILNIUS UNIVERSITY
INSTITUTE OF PHYSICS

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THE LIGHT SCATTERING IN OPTICAL COATINGS AND LASER COMPONENTS IN A WIDE SPECTRAL RANGE

Summary of doctoral dissertation

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Introduction

Performance of laser systems directly depends on the quality of optical components. Therefore, the components for laser systems should fulfill a number of strict requirements, which can be achieved only by using the latest manufacturing technology combined with modern equipment for parameter characterization. The progress in laser technologies is tightly related to the quality in manufacturing of the optical components. The fabrication process mainly defines fundamental optical properties of the components. Reflection, transmission, absorption and scattering of light should be measured to characterize propagation of laser radiation through an optical element completely. Procedures of measurement of all listed parameters are described by international standards. The international ISO-13696 standard defines the method for the standardized measurement of the total light scattering in visual and near infrared spectral range [1-5]. The total scattering measurements provide a lot of information about the production process or properties of the already produced optical element and are a method for relatively rapid control of surface roughness of the optical element [3-8]. They rather quickly provide information about the entire surface of the element, because the laser spot diameter generally can be changed from microns to a few millimeters. Atomic force or electron microscopes can directly determine the surface quality, but with these methods takes some time to make measurement and they provide information only about the very small (a few tens of microns) surface area [2-5]. Fast and effective characterization of the optical quality can be achieved by combining the light scattering and microscopy techniques [3, 4]. The light scattering is one of the mechanisms which limits the maximum reflectance of optical mirrors, and decreases the surface damage threshold of the optical elements [3]. In the case of optical coatings, the scattering should be measured at the wavelength of the optical coating design [4]. In order to obtain the detailed information about the surface quality of optical elements, it is desirable that the tests can be carried out using different wavelength radiation. The light scattering measuring system with a wide spectrum is needed to make such measurements. However, to date, the most of scattering measurements were performed using a single wavelength. As the scattering intensity of optical coating correlates with the reflection of this coating, the single-wavelength approach does not allow investigate samples coated with optical coatings. Therefore, the aim of this work was to create the light scattering measurements system suitable for measurement of such optical coatings. Optical parametric oscillators (OPO) are currently the best laser sources in such system for characterization of optical components and optical coatings, because they have the widest wavelength tuning range of the existing sources of coherent radiation [3, 4]. However, to obtain a wide spectral tuning range and high conversion efficiency, short laser pulses are needed to pump OPO. The high pulse repetition rate Q-switched nanosecond laser sources are an optimal cost-effective solution for such applications.

This work describes the system for the light scattering measurements in a wide spectral range, which uses the light parametric oscillators and harmonic generators pumped with a nanosecond pulses and summarizes the research in various types of coatings on optical components and inside nonlinear optical crystals.

The scientific tasks in this work were:

1. Using OPO's and the harmonic generators pumped by the high repetition rate nanosecond pulses, to create a system for the proper measurement of total light scattering in the wide spectral range;
2. To investigate the scattering of the high reflectivity optical coatings made by the various methods, and to find the relation between the value of the scattering losses and the technology of the optical coating deposition;
3. To investigate the total scattering losses of the antireflective sol-gel coatings;
4. To investigate the bulk scattering losses of the non-linear $LiInS_2$ and $LiInSe_2$ crystals in infrared region;
5. To investigate defects in a fast-rapid grow KDP crystals, using the laser scattering tomography method.

The innovations presented in the thesis are:

1. The light scattering measurements of optical coatings for the first time were performed in a wide spectral range using pulses of tunable radiation of OPO's.

2. The total scattering losses were characterized in the infrared region for a new promising in nonlinear optics $LiInS_2$ and $LiInSe_2$ crystals.
3. It was shown that the laser scattering tomography, previously used for investigation of volume defects in semiconductor crystals, can be applied in the nonlinear optical quality control.
4. Scattering losses of high reflection and antireflection coatings deposited by e-beam, e-beam with ion assistance and sol-gel methods were evaluated at specific wavelengths or wavelength regions.

Propositions to be defended

1. Parametric light oscillators, pumped with a nanosecond laser pulses, are suitable light sources for the total light scattering measurements and in addition provide capability to tune the wavelength, which was not possible with the cw laser sources.
2. The scattering losses for the high reflective optical coatings increased with the increase in a number of optical films of the coating, and they correlated with the increase in surface roughness of the coating. The scattering losses as well as surface roughness were higher in the coatings deposited without ion-assistance.
3. The sol-gel antireflection coatings and the optical coatings deposited in vacuum with ion assistant have the higher scattering losses compared to losses of the bare substrate. It is shown that the coatings increased the surface roughness of substrate.
4. The light scattering losses in volume of the $LiInS_2$ and $LiInSe_2$ single crystals in the 1–2 μm spectral region are large (5–10% over the 1 cm length) and can not be neglected.
5. The laser scattering tomography method is suitable for the identification of scattering centers in such nonlinear crystals as KDP. The centres usually are the impurity absorption centers, and normally initiate the laser induced damage.

Summary of doctoral dissertation

"Introduction"

In this part of the dissertation, the relevance of the investigation is motivated and the main task and propositions to defend are formulated.

Chapter I. "Literature review"

In this chapter the scattering theory in optical components is discussed. Vector scattering ([9, 10]) and scalar scattering ([5, 7]) models of the rough surface, volume scattering in optical layers, defects in real optical components (mirrors, crystals), methods of scattering measurements (total light scattering, angle resolved scattering, light scattering tomography) and previously performed experiments is described. It was shown the relation between amount of uncoated component surface scattering and it surface roughness.

Chapter II. "Experimental technique"

In this chapter, the systems for total light scattering and laser scattering tomography are described. The multiple-stage laser system, which serves as the light source for this systems presented in Fig. 1. A Lambda Physik StarLine model 2030 laser operating at a repetition rate of 1 kHz is used as the master laser oscillator. The 8 mJ, nearly diffraction limited, 1064 nm output of this diode-pumped, Q-switched Nd:YAG laser allows efficient nonlinear optical frequency conversion.

The two lower-peak-power OPO's are operated at up to 1 kHz repetition rate. Infrared radiation tuneable in the range 1.40–4.5 μm is generated by an OPO based on PPLN and pumped directed by the output of this Q-switched laser oscillator. A portion of the laser oscillator output can be converted to 532 nm radiation by second-harmonic generation, and the second-harmonic radiation can be combined with the remaining 1064 nm fundamental laser output to produce third-harmonic pulses at 355 nm by the nonlinear process of optical sum-frequency generation. An OPO based on BBO pumped by the

355 nm pulses can be used to generate 420 to 2200 nm tuneable pulsed coherent radiation. These two OPO's have relatively low peak power.

For the measurements of sol-gel and PMMA coatings scattering, a Standa model STA-01 Nd:LSB microlaser operating at a repetition rate of 1 kHz with a pulse energy of 10 μJ was used.

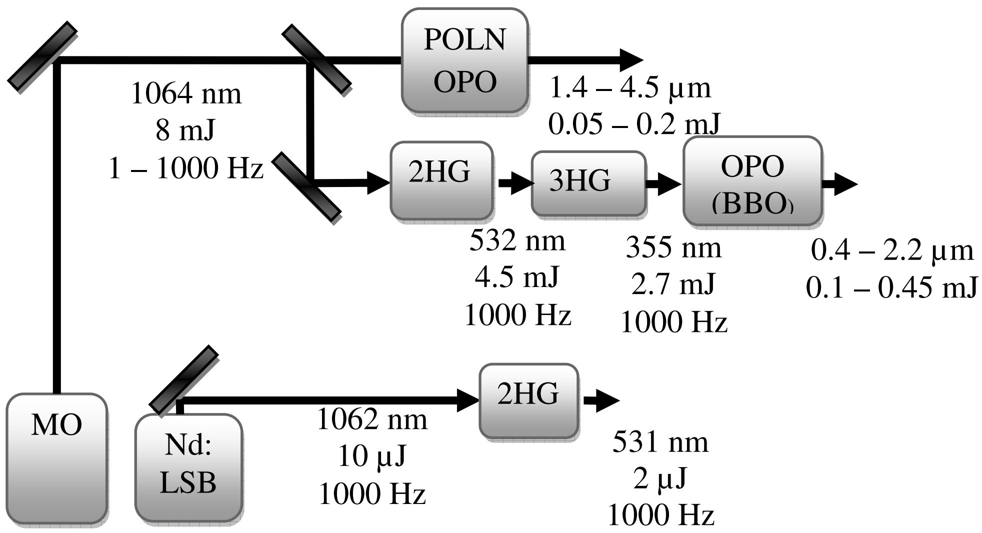


Fig. 1. Laser system: MO – diode-pumped, Q-switched master laser oscillator StarLine 2030; 2HG – second-harmonic generator; 3HG – third-harmonic generator; OPO (PPLN) – OPO based on periodically poled lithium niobate; OPO (BBO) – OPO based on BBO crystal; Nd:LSB – STA-01 Nd:LSB microlaser.

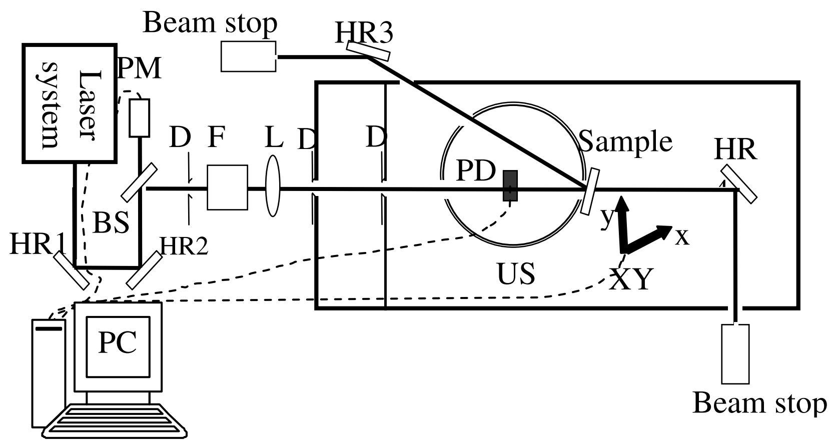


Fig. 2. Experimental set-up based on the Ulbricht sphere (US) for the total light scatter measurements in a wide spectral range: D – aperture; F – variable attenuator; FD – photodiode; L – lens (focal length 1 m); PM – power meter.

The total light scattering measurements system (Fig. 2) is equipped with the Ulbricht sphere. Sensitivity of the system was determined by measurements with the unloaded sphere. The background level due to the Rayleigh scattering in the air and diffraction by the elements used in the system, was as low as 10^{-5}. For the determination of the total backward scatter values, the specimen was attached to the exit port of the sphere. For the measurement of forward scatter signal, the procedure was repeated with the specimen located at the entrance port. A 16-bit-resolution analogue-digital converter (ADC) was used in the experiment. The experiment was controlled by the computer. In order to control the total scattering measurements, a procedure based on the programming package LabView was created. The input of the technical parameters of the sample (the material of the sample, type of coating, deposition technology, laser power and etc.) was needed to start the measurements. The sample was mounted on the xy translation stage, and the surface area was divided in to hexagon spot matrix. The program controlled the positioning of the sample to the first point of the matrix, saved the measurement data (scattering losses and the coordinates of the point position) and translates the sample to another point. In this way, all surface of the sample was scanned with the laser beam.

Laser Scattering Tomography (LST) system was used to investigate submicroscopic precipitates and the patterns of decorated defects in crystals. The properties of LST correspond mainly to that of dark-field optical microscopy. Fig. 3 shows our experimental arrangement. The second harmonic of a nanosecond Nd:YVO_4 laser (532 nm, 1 W, 10 kHz repetition rate) was extended by a telescope (*T*). The subsequent cylindrical lens (*C*) formed a strip of light. A strip of light irradiated the scatter centers inside the sample, and it was possible to observe 2D LST images in live modus. The height of the light strip in the y direction was up to a few mm. The width of the light strip (FWHM) was about 30 μm.

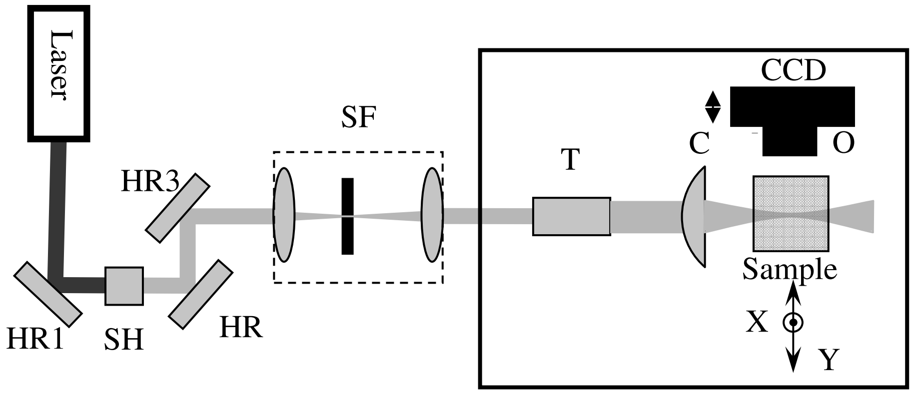


Fig. 3. Experimental set-up for the Laser scattering tomography measurements. HR1-HR3 – mirrors; SH – second harmonic crystal; SF – spatial filter; T – telescope; C – cylindrical lens; CCD – CCD camera with objective.

An objective (*O*) imaged the irradiated scatter centers onto the target of a CCD matrix camera (8 bit, 373 x 490 pixels, exposure times up to 30 sec) selected for a sufficient sensitivity at the laser wavelength.

The sample could be moved by the translation stages in x-, y- and z-direction with the fixed laser beam and the camera in y-direction. During y-movements of the sample, the camera was translated to conserve the optical conjugation between irradiated plane and CCD. The shift of the camera was (n-1)/n of the shift of the sample with refractive index *n*. Such corrected translations were essential for the layer-by-layer imaging and the 3D computer reconstruction.

Chapter III. "Measurements of the optical scattering losses of various optical coatings in a wide spectral range"

In this chapter the results on the measurements of the scattering losses of optical components, made using various materials and by different manufacturing technologies, are discussed. In the first section, the scattering losses of the high reflection (at 1064 nm) SiO_2/TiO_2 coatings deposited using Ion-assisted vapor deposition are described.

The scattering of high reflection ion-assisted SiO_2/TiO_2 coatings at 1064 nm spectral range

Ion-assisted bombardment is a coating deposition technique that can offer unique benefits under certain circumstances. The ion assistance during the deposition process leads to a higher atomic or molecular packing density in the thin-film layers. This results in a higher refractive index and, most important, better stability of the spectral parameters.

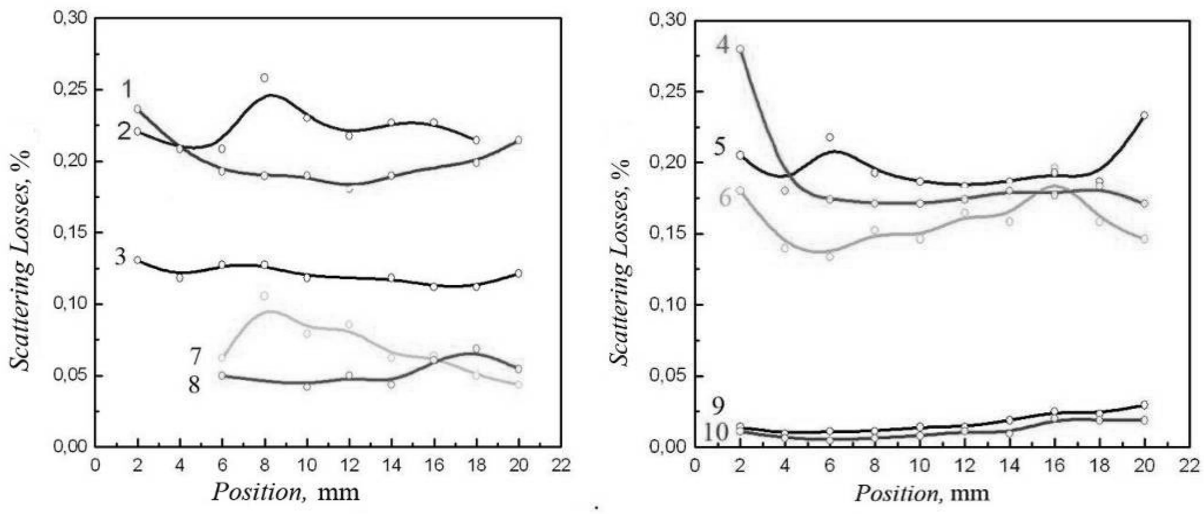


Fig. 4. Scattering losses of investigated samples scanned in one dimension at 1064 nm: 1–6 samples with SiO_2/TiO_2 coatings; 7, 8 samples with SiO_2/Ag coating; and 9, 10 substrates without coating (bare).

The light scattering measurements were performed using 1064 nm laser radiation with the total scattering measurement system described in Fig. 2. The laser beam energy was 340 μJ per pulse at a beam diameter of 1 mm. The results are presented in Fig. 4. The scattering losses for uncoated substrates depended on the surface roughness σ, and the scattering measurements provided an assessment of the roughness. The scattering losses of the bare optical substrates used for production of laser mirrors was less than 0.02 % and corresponded to the surface roughness of 0.9 nm. The scattering losses of the metallic mirrors additionally coated with one dielectric layer were in the range of 0.05-0.07 %. The high reflection mirrors coated with multilayer stacks of 21 thin films had the scattering losses in range from 0.12 % to 0.25 %. It can be shown that in one optical layer deposited by e-beam with ion assistant is scattered 0.01 % of radiation power. The scattering losses of the coating deposited without the ion-assistance described in the next section.

Scattering losses in optical coatings on the ZnSe substrate in the NIR spectral range

Zinc selenide is the most popular material for infrared applications. Due to a very wide transmission range covering from 0.6 to 22 μm, the chemical-vapor-deposition-grown ZnSe as a high optical quality material is used to manufacture optical components for high power IR lasers. It is the high-index component (*n* ~ 2.4) used in combination with low-index fluoride compounds to construct the coatings in the IR region at wavelengths longer than 2 μm. Low-index companion layers materials include YF_3, YbF_3, IRX and ThF_4. Zinc selenide layers are relatively soft and insoluble in water. The scattering losses of some layers deposited on the ZnSe substrate without the ion assistance are presented in Fig. 5.

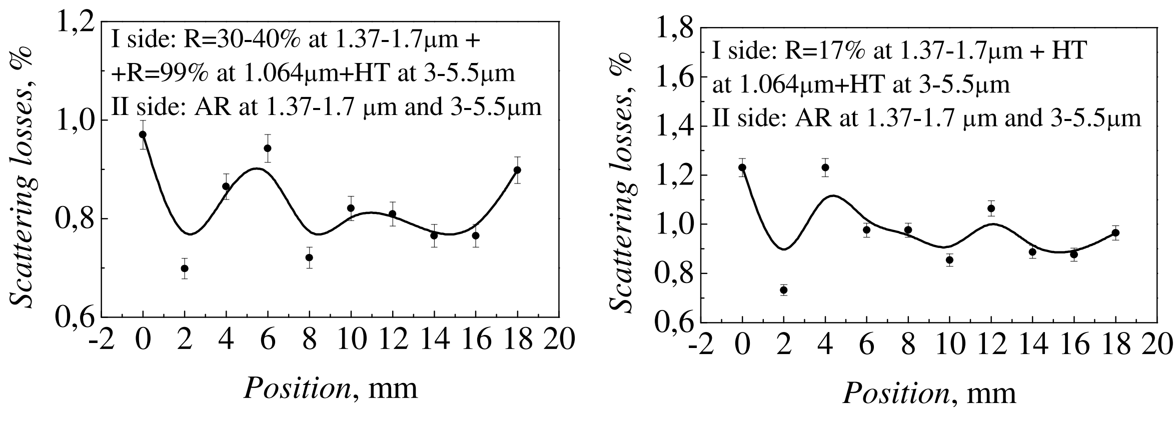


Fig. 5. Scattering losses as a function of position of the partially reflecting coatings on the ZnSe substrate. The solid line is a polynomial fit to the data shown as dots.

The scattering measurements were performed using the 1.5 μm PPLN OPO radiation. It can be see than scattering losses of these coatings are much larger that scattering losses of the SiO_2/TiO_2 coatings deposited with the ion assistance.

Scattering losses of optical coatings on the CaF_2 substrate

We have performed the total scattering measurements of the dielectric coatings on two different CaF_2 substrates by using the tunable PPLN-OPO radiation. For the scattering measurements two different samples were chosen.

The mirror with the high reflectivity in the range of 700–870 nm and with the high transmission in the range of 1.4–4.5 µm on CaF_2 was measured by using 1.65, 1.84 and 1.92 µm wavelengths. The scattering map of the surface of this mirror is presented in the Fig. 6. Scattering losses as a function of position of the coatings on the same CaF_2 mirror are demonstrated in the Fig. 7.

Another sample for the scattering measurements was the CaF_2 beamsplitter. The scattering map of the surface of this mirror at different OPO wavelength is presented in the Fig. 8. From the measured scattering results, it can be seen that there are some scattering centers on these samples. The later was proven by

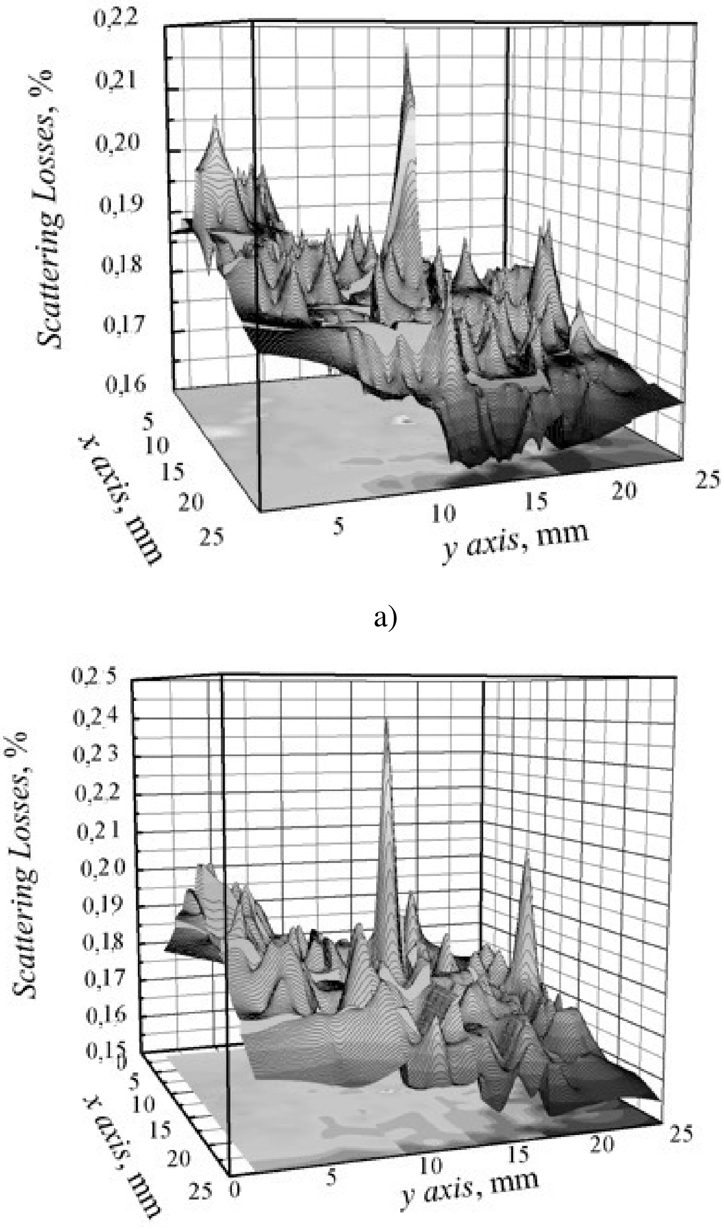


Fig. 6. Scattering losses of the CaF_2 mirror (HR at 700–870 nm, $R > 95$ %, HT at 1.4–4.5 µm) at various surface positions using 1.84 µm (a) and 1.65 µm (b) radiation.

investigation of these samples by microscope. The influence of the wavelength of the radiation on the scattering correlated with the transmission spectra of these samples measured by the spectrometer.

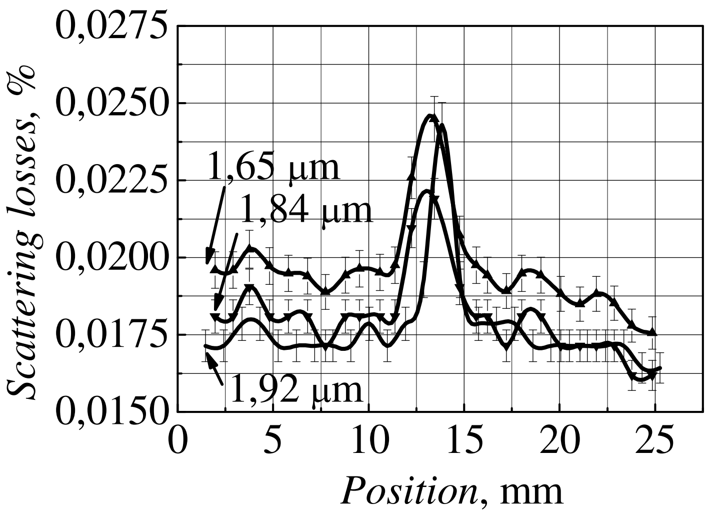
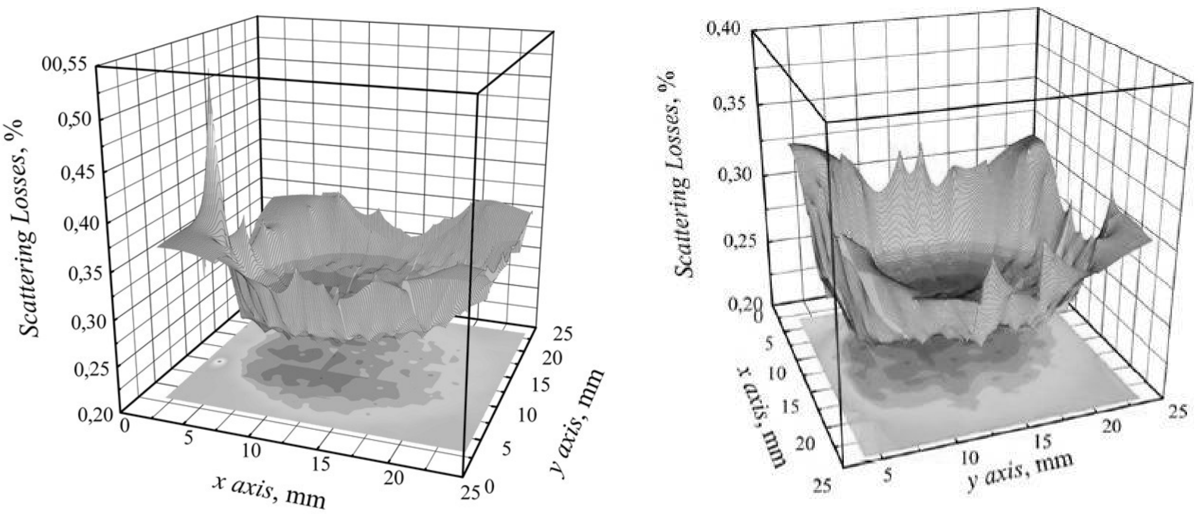


Fig. 7. Scattering losses as a function of position on the CaF_2 substrate. The solid lines are polynomial fits to the data shown as dots.



a) b)

Fig. 8. Scattering losses of the CaF_2 beamsplitter (HR at 800 nm, HT at 2.0–3.0 μm) at various surface positions using 1.84 μm (a) and 1.65 μm (b) radiation.

Scattering of samples deposited by sol-gel technique

In this section the scattering losses of antireflective coatings deposited by sol-gel methods are described. Sol-gel process is an alternative, inexpensive technique that does not require vacuum or high temperature as previously discussed deposition methods. The samples were produced by spin-coating and dip-coating.

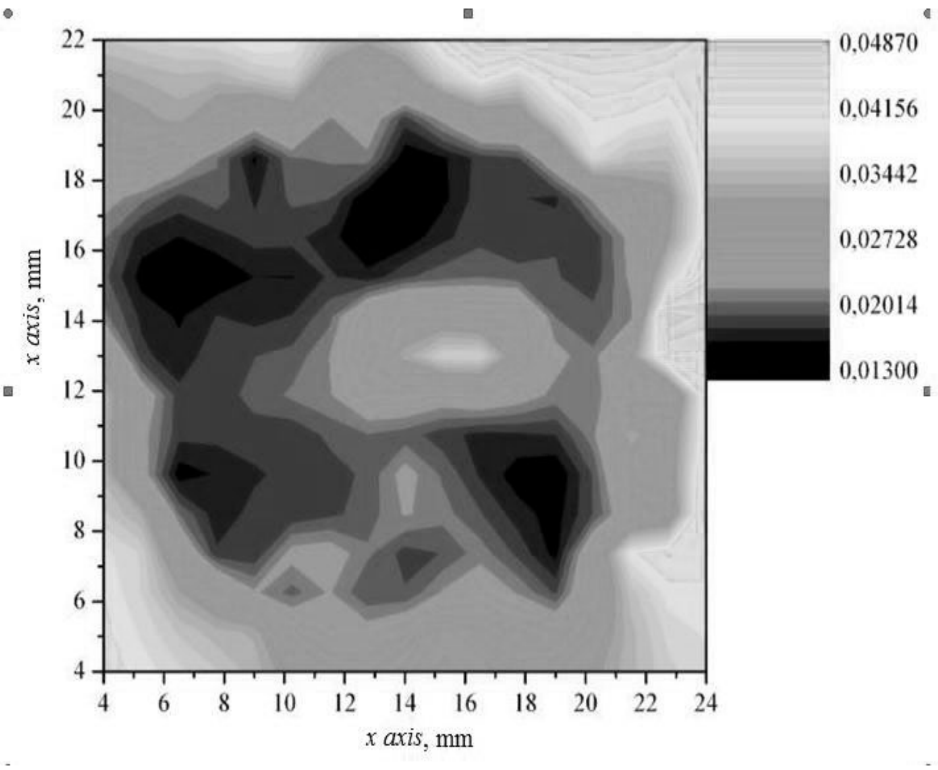


Fig. 9. The scattering losses of the sol-gel coating deposited by spin-coating.

Spin coating has been used for several decades for the application of thin films. In this process, a drop of a sol solvent is placed on the substrate, which is then rotated at high speed. Rotation is continued until the fluid spins off the edges of the substrate, and the desired thickness of the film is achieved. The applied solvent is usually volatile, and simultaneously evaporates. So, the higher the angular speed of spinning, the thinner the film. The thickness of the film also depends on the concentration of the solution and the solvent.

For the total scatttering measurements, a system showed in Fig. 2 was used. Radiation of the Nd:LSB microlaser was focused into 1 mm beam diameter with a 10 μJ pulse energy. In this case, the system dynamic range was as high as 10^6. As shown in Fig. 9, using this deposition method, the scattering losses are significantly higher near edges of the sample.

Dip coating refers to the immersing of a substrate into a tank containing coating material, removing the piece from the tank, and allowing it to drain. The coated piece can then be dried by force-drying or baking. The dip-coating technique commonly used for liquid-deposition, implies a safety hazard due to coating solution handling and storage in the case of large amounts of highly flammable solvent use [11]. Fig. 10 (a) shows the scattering losses map of the fused sillica substrate. The scattering-loss value of this sample was 0.06 %. In Fig. 10 (b) the scattering losses of this substrate with one the LD TM coating deposited by the dip coating sol-gel method are shown. The calculated value of scattering losses was 0.07 %. Thus, it can be concluded that a single

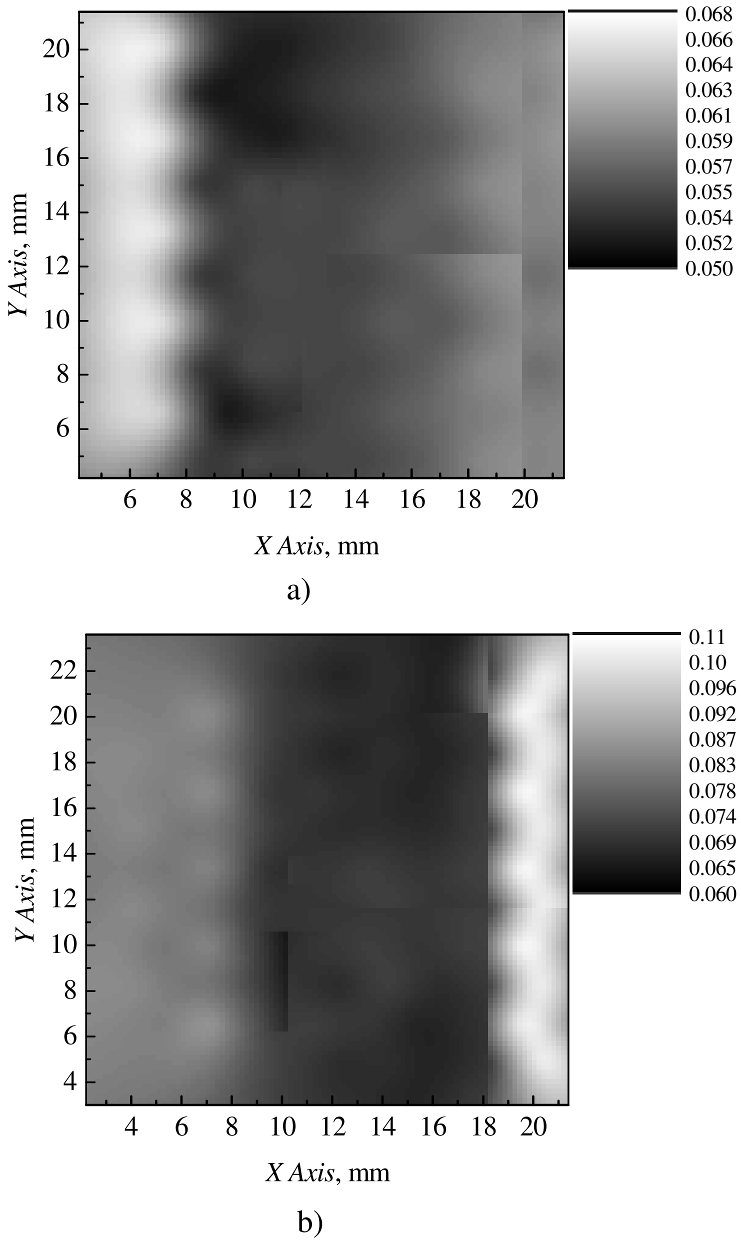


Fig. 10. Scattering losses of fused silica (a) and fused silica with the one LD-TM coating (b).

LD TM coating increases by 0.01 % the scattering losses. The value is sufficiently small and is of the same magnitude as for the ion-assisted coating scattering losses.

Scattering losses of PMMA waveplates

Polymethyl methacrylate (PMMA) is a versatile polymer that is a proper material for fabrication of many commercial optical components: lenses, fibers, windows, phase waveplates and others. Our focus was to characterize the total scattering value of achromatic zero-order waveplates made of anisotropic PMMA. The most successful combination of those plates made of the same material was proposed by Pancharatnam (Fig. 11a) [12].The end components have coincident optical axes and equal retardation. The middle component has a retardation $\tau_2 = 180^o$ and its optical axis is rotated by an angle τ_2 relative to the axes of the end components. Further improvements of this design were made by Kucherov et al. for an arbitrary number of components. It was shown ([13]) that additional pairs of components (every component of a pair has a symmetrical position relative to the central one) improve considerably the waveplate characteristics. The optical axes in every additional pair of waveplates must be parallel and their retardations must be equal to 180^o at the central wavelength λ_o. The five-component design is shown schematically in Fig. 11, b.

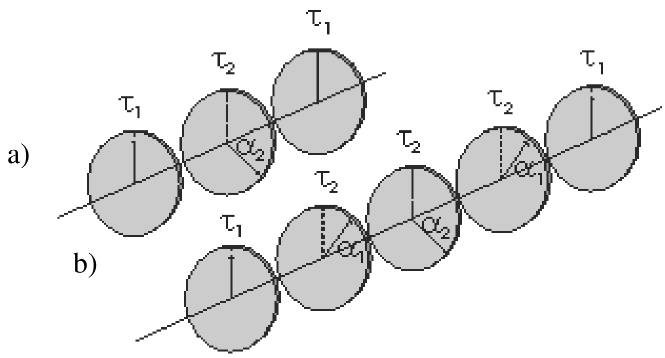
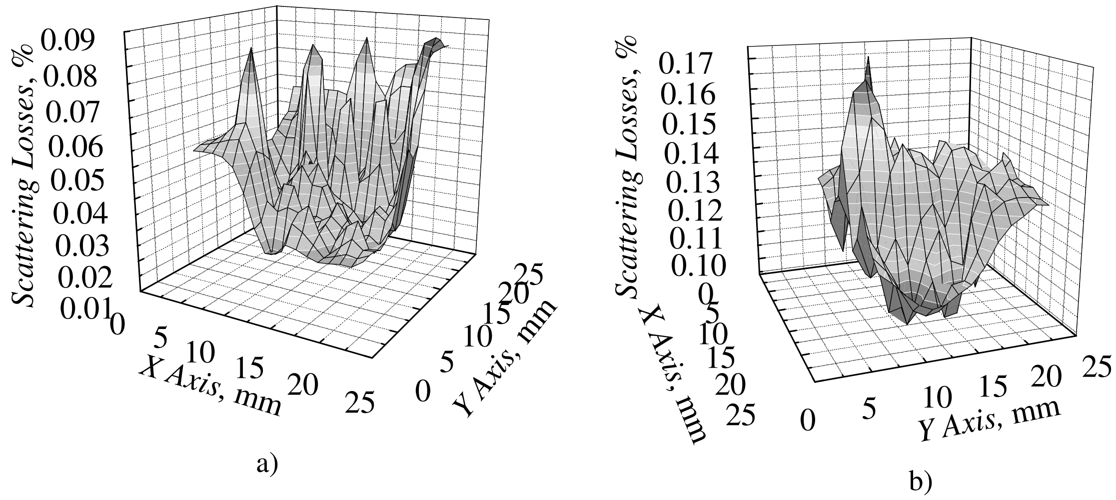


Fig. 11. Design of achromatic (a) and super-achromatic (b) waveplates

For the total light scattering mesurements in PMMA, a fundamental wave from diode pumped Nd:LSB solid-state micro laser (STA-01, Standa; Lithuania (Fig. 1)) 1062 nm radiation was used as a light source in this system. Laser beam was focused to a 1 mm spot and power was kept below 4 mW in purpose not to damage waveplates. The results are given in Fig. 12.

Table 2. Optical characteristics of tested samples

No. of sample	Scattering losses,%
1	0.04
2	0.13
3	0.11



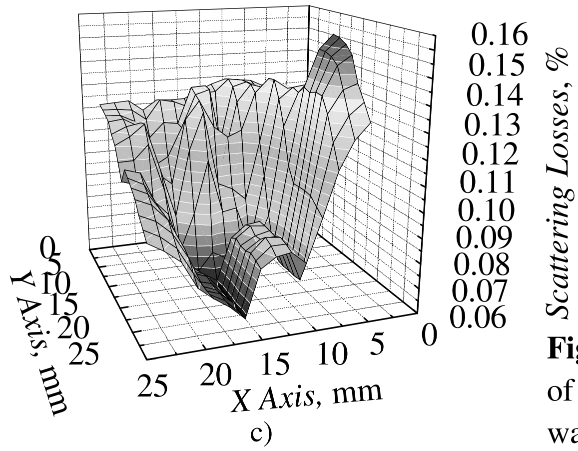
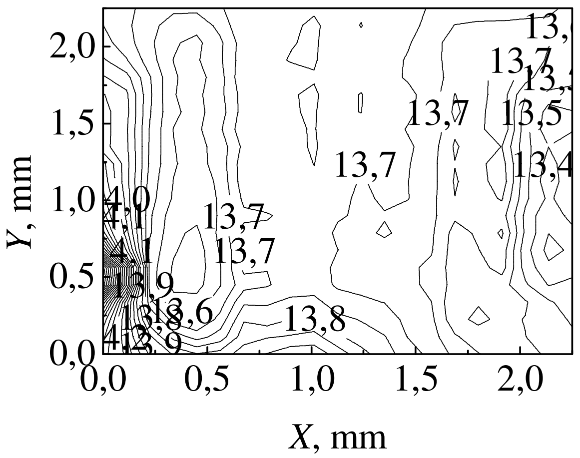


Fig. 12. Total back scattering measurements of 1st (a), 2nd (b) and 3rd (c) PMMA waveplates.

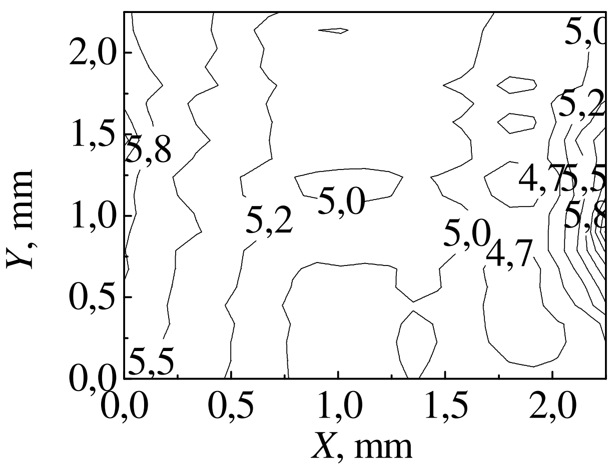
As can be seen from Table 2, all scattering losses not exceeded 0.13 % of incident laser power at 1062 nm testing wavelength. A Fig. 12 shows which places of specimens surfaces has the highest scattering. The highest scattering value (0.13 %) has waveplate without AR coating. Other samples showed a little bit lower scattering losses. These results are relative good for waveplates made of five separate components.

Chapter IV. “Tests of optical crystal quality”

In the first section of this chapter, the scattering losses measurements of the new in nonlinear optics $LiInS_2$ and $LiInSe_2$ are described. Lithium thiogallate ($LiInS_2$) and lithium selenoindate ($LiInSe_2$) were chosen for the scattering characterization. Non-linear characteristics of these crystals are close to $AgGaS_2$ and $AgGaSe_2$, but their crystal structures are different. LIS belongs to the $A^{I}B^{III}C_2^{VI}$ chalcogenide family. It is a biaxial crystal with the *mm2* symmetry, like the well-known KTP crystal. It is optically transparent from 0.4 to 12 μm. The linear and nonlinear optical properties of LIS were briefly studied by Boyd *et al.* in the 1970 [14]. Recently, renewed interest has been paid to LIS because of its attractive optical properties, such as the high optical damage threshold, large thermal conductivity, large energy band gap, and relatively high nonlinear susceptibility [15-20].



a)



b)

Fig. 13. Scattering losses (in %) of the $LiInSe_2$ (a) and $LiInS_2$ (b) crystals at various surface positions using 1.064 μm radiation.

The scattering measurements were performed using the 1064 nm laser radiation. The laser beam energy was 450 μJ per pulse at a beam diameter of 150 μm to avoid the damage of the optical components. The scattering maps of the crystals are presented in the Fig. 13. The larger scattering losses on the periphery of the samples are due the diffraction from the sample holder. Scattering losses for $LiInSe_2$ crystal was as high as 13.7 %. $LiInS_2$ crystal had 5.0 % of the total scattering losses. From the measured scattering results it can be seen that there are no intense scattering centers in these samples, ant that was confirmed by investigation of these samples by microscope. The influence of the wavelength of the radiation on the scattering correlated with the transmission spectra of these samples measured by the spectrometer.

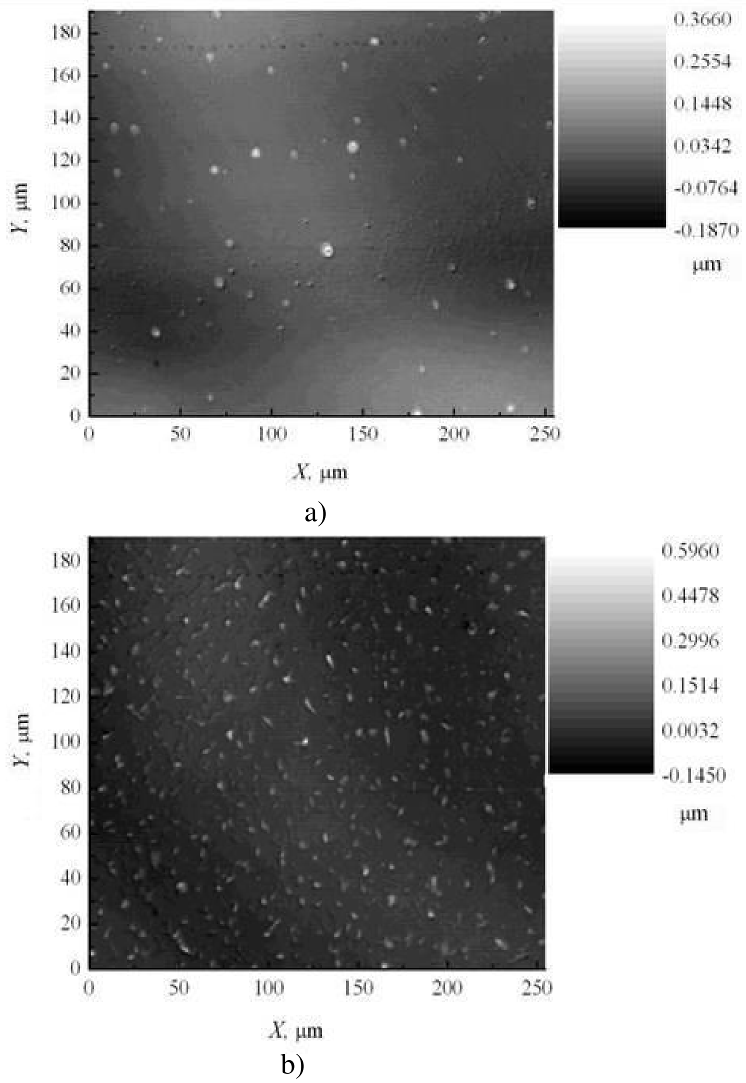


Fig. 14. Surface topography images of $LiInSe_2$ (a) and $LiInS_2$ (b) crystals.

Fig. 14 shows surface topography images of these two crystals, measured using PR2000 profilemeter. The surface roughness of $LiInSe_2$ crystal is 30 nm and 20 nm for the $LiInS_2$ crystal, therefore the surface scattering contribution is significantly smaller than the volume scattering.

In order to fully characterize the optical crystal quality, it is necessary not only to measure total scattering losses of the optical crystal, but also to investigate the defects in the crystal volume. Therefore in the second section, the researches of bulk defects in rapid-growing (r-KDP) and slow-growing (s-KDP) KDP crystals using laser scattering tomography system (Fig. 3), are described.

LST measurements were performed with a different objective magnification power, as a result we could to determine the scattering centers with dimension from 20 to 60000 μm^2. Fig. 15 shows LST images of slow-growing KDP crystal, measured using 10X microscope objective, and Fig. 16 shows images of rapid-growing KDP. In both figures picture (a) represent LST image of surface plane and (b) is image of the plane, located 3 mm inside the crystal. It can be see, that scattering centers dimensions on the crystal surface is much bigger than inside the crystal.

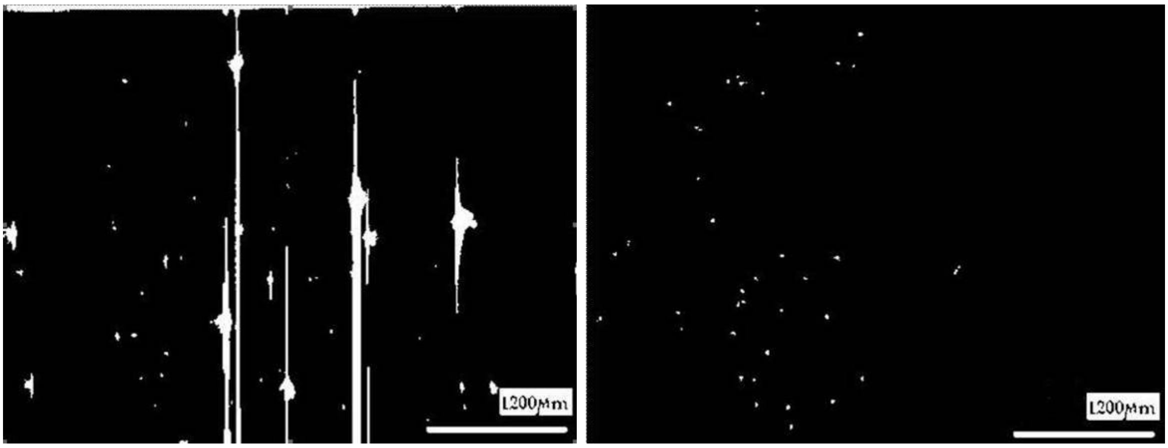


Fig. 15. LST images of slow-growing KDP crystal at surface plane (a) and in the 3 mm depth (b) using 10 X microscope objective and 532 nm illumination.

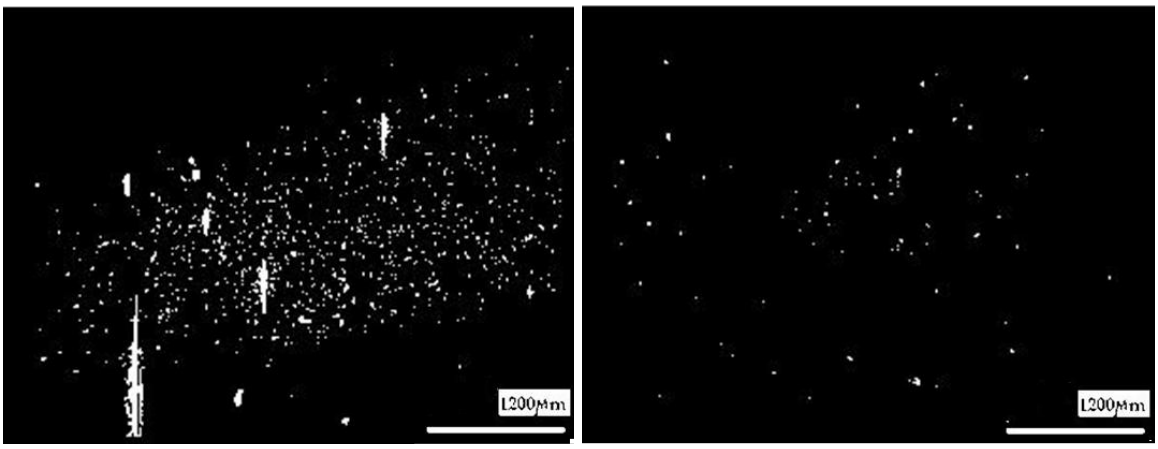


Fig. 16. LST images of rapid-growing KDP crystal at surface plane (a) and in the 3 mm depth (b) using 10 X microscope objective and 532 nm illumination.

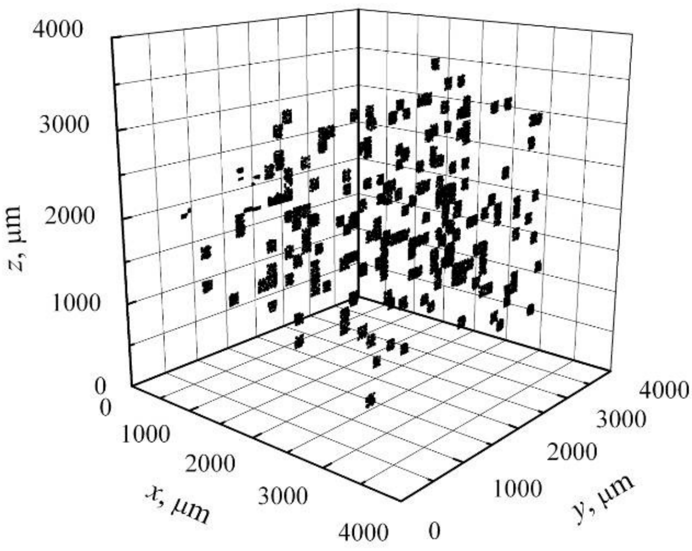


Fig. 17. 3D LST image of the r-KDP crystal scattering centres.

The average size of scattering centres (measured with 10 X microscope objective) in r-KDP surface is 6750 μm^2 and 220 μm^2 in 3 mm depth. Respectively for s-KDP they are 2606 μm^2 and 190 μm^2. Also the scattering centers density in r-KDP is 7.5 times more than in s-KDP crystal.

Layer-by-layer tomography uses images taken at discrete equidistant z-positions of the irradiation plane. Using this method it is possible 3D-image of crystal scattering centres reconstruction. Fig. 17 shows a scattering centres distribution in 4×4×4 mm^3 rapid-growing KDP crystal volume. The tomography layers of this image were taken by 1 mm equidistant distance.

Main results and conclusions

1. The measurements of light scattering losses can be performed in the spectral range from 0.4 to 4.5 µm using PPLN and BBP OPO's pumped by nanosecond pulsed lasers. The measurements sensitivity of 0.01% can be achieve without photon counting.

2. The scattering losses in optical coating, deposited by the method of thermal evaporation with ion-assistance increases by 0.01 % per film. The increase of losses is caused by the increase of surface roughness of the optical coatings. The scattering of the optical film deposited without ion assistance is higher and can be as high as 1 %.

3. The quality of the single sol-gel antireflection coating is better in comparison to the vacuum deposited films and its scattering losses are of the same magnitude as scattering losses of the ion-assisted coating.

4. The scattering losses of the antireflection sol-gel film deposited using a spinning method are in range of 0.02–0.04 %. Using this deposition method, the scattering losses are significantly higher near edges of the sample.

5. Scattering losses of the nonlinear $LiInSe_2$ and $LiInS_2$ crystals in the 1.06 µm region are much higher than that for the widely used optical mirrors and were as high as 13.7 % and 5 % in 1 cm, respectively. When radiation wavelength was approaching 2 µm, scattering losses decreased. The surface roughness for these crystals was not larger than 30 nm, therefore the surface scattering contribution was significantly smaller than the volume scattering.

6. The laser scattering tomography is the appropriate method for the observation of scattering centers in nonlinear crystals such as KDP. The method was capable to determine the scattering centers with dimension from 20 to 60000 μm^2.

7. The density of the light scattering centers in the slow-grown KDP crystal was 7.5 times less than in the rapid-grown KDP crystal.

Author's publications

The main author's results were presented at 14 International, National conferences and seminars. The results were presented also in 8 scientific publications: one publication in Lithuanian scientific journal [S8] and 7 publications in the international scientific journals [S1-S7].

Publications in the international scientific journals:

[S1]. Balachninaitė O., Eckardt R.C., Maciulevičius M., Grigonis R., Sirutkaitis V., *A coherent spectrophotometer based on a periodically poled lithium niobate optical parametric oscillator for optical characterization*. Proc. of SPIE, **5647**, p.385-393 (2005).

[S2]. Melninkaitis A., Maciulevicius M., Rakickas T., Miksys D., Grigonis R., Sirutkaitis V., Skrebutenas A., Buzelis R., Drazdys R., Abromavičius G., *Comparison of optical resistance of ion assisted deposition and standard electron beam deposition methods for high reflectance dielectric coatings*. Proc. of SPIE, **5963**, p. 429-437 (2005).

[S3]. A. Melninkaitis, K. Juškevičius, M. Maciulevičius, V. Sirutkaitis, A. Beganskiene, I. Kazadojev, A. Kareiva, and D. Perednis, *Optical characterization of antireflective sol-gel coatings fabricated using dip coating method*. Proc. of SPIE, **6403**, 64031C (2006).

[S4]. Melninkaitis A., Mikšys D., Maciulevičius M., Sirutkaitis V., Šlekys G., Samoylov A. V., *Laser-induced damage thresholds of starched PMMA waveplates*. Proc. of SPIE, **6403**, 640325 (2006).

[S5]. Balachninaite O., Petraviciute L., Maciulevicius M., Sirutkaitis V., Isaenko L., Lobanov S., Yelisseyev A., Zondy J.-J. , *Characterization of the mid-infrared nonlinear crystals $LiInSe_2$ and LiInS in the IR range*. Proc. of SPIE, **6403**, p. 64031Y (2007).

[S6]. Balachninaitė O., Petravičiūtė L., Maciulevičius M., Sirutkaitis V., Isaenko L., Lobanov S., Yelisseyev A., Zondy J-J., *Optical characterization of the $LiInS_2$ and $LiInSe_2$ crystals.* Proc. of SPIE, **6596**, 65961J-1 (2007).

[S7]. Balachninaite, O., R.C. Eckardt, M. Maciulevicius, M. Sinkevicius, V. Sirutkaitis, *Absorptance and scattering losses measurements in IR range by high average power tunable radiation of optical parametric oscillator based on a periodically poled lithium niobite.* Proc. of SPIE, **6101** p. 61011K-10, (2006).

Publications in Lithuanian scientific journal:

[S8]. Balachninaite O., Petraviciute L., Maciulevicius M., Sirutkaitis V. , Isaenko L., Lobanov S., Yelisseyev A., Zondy J.-J., *Absorptance and scattering losses measurements of the mid-infrared nonlinear crystals $LiInSe_2$ and $LiInS_2$ in the IR range*. ISSN 1392-2114 ULTRAGARSAS, **3**(60), p.7-10 (2006).

Conferences

[C1]. Maciulevičius M., Vaičaitis V., Sirutkaitis V., Measurements of scattering losses of optical elements and thin films at 1064 and 1550 nm. International workshop, Parametric processes and periodical structures. p. 101-102, 26-29 September, Vilnius, Lithuania, 2004.

[C2]. Balachninaitė O., Maciulevičius M., Melninkaitis A., Grigonis R., Sirutkaitis V., Eckardt R.C., A coherent spectrophotometer based on a periodically poled lithium niobate optical parametric oscillator for optical characterization. International workshop, Parametric processes and periodical structures. p. 71-73, 26-29 September, Vilnius, Lithuania, 2004.

[C3]. M. Maciulevicius, V. Vaicaitis, V. Sirutkaitis. Registration of scattering losses of optical elements and optical thin films at 1064 and 1550 nm XVI Lithuanian –

Belarussian seminar "Lasers and Optical Nonlinearity" Vilnius, Lithuania on 27-29 of October 2004.

[C4]. Balachninaite O., Maciulevicius M., Grigonis R., Sirutkaitis V., Coherent spectrophotometer based on a periodically poled lithium niobate optical parametric oscillator for optical characterization // In Book of Abstracts XXXVI Annual Symposium on Optical materials for High Power Lasers: Boulder Damage Symposium, NIST, document 5647- 52, p. 64-65, Boulder, Colorado, USA,2004.

[C5]. Maciulevičius M., Vaičaitis V., Sirutkaitis V., Measurements of scattering losses on the surface of optical components. Beyond Einstein Physics for the 21st Century, 13th General Conference of the European Physical Society, Conference I: Photons, Lasers and Quantum Statistics, University of Bern, Switzerland 11-15, July, 2005.

[C6]. Maciulevicius M., Balachninaite O., Sirutkaitis V. Characterization of the Optical Coatings Using Total Light Scattering Technique in Infrared Spectral Range. 7-th International Conference - School "Advanced Materials and Technologies" Palanga, Lithuania on 27th - 31st of August, 2005.

[C7]. Sirutkaitis V., Balachninaite O., Maciulevicius M., Melninkaitis A., Grigonis R., Eckardt R.C., Optical characterization in wide spectral and temporal range. Proceedings of the Symposium on Photonics Technologies for 7th Framework Program. p. 78-87, Wroclaw 12-14 October 2006.

[C8]. Balachninaite O., Eckardt R.C., Maciulevicius M., Sinkevicius M., Sirutkaitis V., Absorptance and scattering losses measurements in IR range by high average power tunable radiation of optical parametric oscillator based on a periodically poled lithium niobate, 8th International Conference on Laser Beam Control and Applications San Jose, CA, JAN 22-26, 2006.

[C9]. Balachninaite O., Petraviciute L., Maciulevicius M., Sirutkaitis V., Isaenko L., Lobanov S., Yelisseyev A., Zondy J.-J., Absorptance and scattering losses measurements of the mid-infrared nonlinear crystals $LiInSe_2$ and $LiInS_2$ in the IR range // International conference "Non-destructive Testing and Diagnostics-2006", Vilnius, 2006.

[C10]. Maciulevicius M., Sirutkaitis V., Scattering measurements of novel optical materials at visible and near infrared spectral range // XII Conference on Laser Optics 2006, III International Conference for Young Scientists on Laser Optics (LOYS 2006), St. Petersburg, 26-30 June 2006.

[C11]. Balachninaite O., Petraviciute L., Maciulevicius M., Sirutkaitis V., Isaenko L., Lobanov S., Yelisseyev A., Optical characterization of the $LiInS_2$ and $LiInSe_2$ crystals // The International Conference "Advanced Optical Materials and Devices" (AOMD-5), Vilnius 27-30 August, 2006.

[C12]. Maciulevicius M., Balachninaite O., Petraviciute L., Sirutkaitis V., Isaenko L., Lobanov S., Yelisseyev A., Zondy J.-J., Standardized Absorbance and Scattering Losses Measurements of the Nonlinear Crystals in the IR Range // XVII Belorussian-Lithuanian Workshop "Lasers and Optical Nonlinearity", Grodno 25-29 September, 2006.

[C13]. Melninkaitis A., Juskevicius K., Maciulevicius M., Sirutkaitis V., Beganskiene A., Kazadojev I., Kareiva A., Perednis D., Optical characterization of antireflective sol-gel coatings fabricated using dip coating method, Boulder Damage Symposium XXXVIII "Optical Materials for High Power Lasers" Boulder, Colorado 25–27 September 2006.

[C14]. Balachninaite O., Petraviciute L., Maciulevicius M., Sirutkaitis V., Isaenko L., Lobanov S., Yelisseyev A., Characterization of the mid-infrared nonlinear crystals $LiInSe_2$ and $LiInS_2$ in the IR range, Boulder Damage Symposium XXXVIII "Optical Materials for High Power Lasers" Boulder, Colorado 25–27 September 2006.

References

1. Standardization, I.O.f., *International Standard ISO 13696, Optics and optical instruments - Test methods for radiation scattered by optical components,* . 2002: Geneva, Switzerland.
2. Amra, C., D. Denmie, D. Torricini, P. Roche, R. Galindo, P. Dumas, F. Salvan, *Overlapping of roughness spectra measured in macroscopic (optical) and mocroscopic (AFM),* Proc. of SPIE. **2253,** p. 614-630, (1994).
3. Lutter, A., K. Ferencz, *The light scattering of dielectric films,* Thin Solid Films. **57**(1), p. 185-189, (1979).
4. Schröder, S., T. Feigl, A. Duparré, A. Tünnermann, *EUV reflectance and scattering of Mo/Si multilayers on differently polished substrates,* Opt. Express. **15**(21), p. 13997-14012, (2007).
5. Lindstrom, T., D. Ronnow, *Total integrated scattering from transparent substrates in the infrared region: validity of scalar theory,* Optical Engineering. **39**(2), p. 478-487, (2000).
6. Bennett, J.M., L. Mattson, *Introduction to Surface Roughness and Scattering*. (Optical Society of America, Washington, D.C., 1999).
7. Bennett, H.E., J. Porteous, *Relation Between Surface Roughness and Specular Reflectance at Normal Incidence,* J. Opt. Soc. Am. **51,** p. 9-15, (1961).
8. Seib, M., H. Hofler, *Optical surface and morphology inspection techniques*, in *Optical sensors*, E. Wagner, Editor. 1992. p. 552 - 587.
9. Elson, J.M., J.M. Bennett, *Vector scattering theory,* Opt. Eng. **18,** p. 116-124, (1979).
10. Hummel, R.E., K.H. Guenther, *Handbook of Optical Properties: Thin Films for Optical Coatings, Volume 1*. (CRC Press, 1995).
11. Belleville, P.F., P. Prene, F. Mennechez, C. Bouigeon, *Sol-gel antireflective spin-coating process for large-size shielding windows,* Proc. of SPIE. **4804,** p. 69-72 (2002).

12. Pancharatnam, S., *Achromatic Combinations of Birefringent Plates. Part I. An Achromatic Circular Polarizer,* Indian Acad. Sci. Proceedings **A41,** p. 130-136, (1955).
13. Samoylov, A.V., V.S. Samoylov, A.P. Vidmachenko, A.V. Perekhod, *Achromatic and super-achromatic zero-order waveplates,* Journal of Quantitative Spectroscopy and Radiative Transfer. **88**(1-3), p. 319-325.
14. Boyd, G.D., H.M. Kasper, J.H. McFee, *Linear and nonlinear optical properties of $LiInS_2$,* J. Appl. Phys. **44,** p. 2809-2812, (1973).
15. Isaenko, L., I. Vasilyeva, A. Yelisseyev, S. Lobanov, V. Malakhov, L. Dovlitova, J.J. Zondy, I. Kavun, *Growth and characterization of $LiInS_2$ single crystals,* Journal of Crystal Growth. **218**(2-4), p. 313-322, (2000).
16. Isaenko, L., A. Yelisseyev, S. Lobanov, V. Petrov, F. Rotermund, J.J. Zondy, G.H.M. Knippels, *$LiInS_2$: A new nonlinear crystal for the mid-IR,* Materials Science in Semiconductor Processing. **4**(6), p. 665-668, (2001).
17. Chen, W.D., E. Poullet, J. Burie, D. Boucher, M.W. Sigrist, J.J. Zondy, L. Isaenko, A. Yelisseyev, S. Lobanov, *Widely tunable continuous-wave mid-infrared radiation (5.5-11 μm) by difference-frequency generation in LilnS(2) crystal,* Applied Optics. **44**(19), p. 4123-4129, (2005).
18. Zondy, J.J., V. Vedenyapin, A. Yelisseyev, S. Lobanov, L. Isaenko, V. Petrov, *LiInSe2 nanosecond optical parametric oscillator,* Optics Letters. **30**(18), p. 2460-2462, (2005).
19. Fossier, S., S. Salaün, J. Mangin, O. Bidault, I. Thénot, J.-J. Zondy, W. Chen, F. Rotermund, V. Petrov, P. Petrov, J. Henningsen, A. Yelisseyev, L. Isaenko, S. Lobanov, O. Balachninaite, G. Slekys, V. Sirutkaitis, *Optical, vibrational, thermal, electrical, damage, and phase-matching properties of lithium thioindate,* J. Opt. Soc. Am. B. **21**(11), p. 1981-2007, (2004).
20. Isaenko, L., A. Yelisseyev, S. Lobanov, V. Petrov, F. Rotermund, G. Slekys, J.J. Zondy, *$LiInSe_2$: A biaxial ternary chalcogenide crystal for nonlinear optical applications in the midinfrared,* Journal of Applied Physics. **91**(12), p. 9475-9480, (2002).

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Reziumė

Šiame darbe pateikti lazerinių komponentų sklaidos tyrimai regimojoje ir artimojoje infraraudonojoje spektro srityse. Eksperimentiniai pilnutinės sklaidos matavimai buvo atliekami remiantis tarptautiniu ISO 13696 standartu, naudojant nanosekundinės trukmės impulsų lazerių bei jais kaupinamų parametrinių šviesos generatorių spinduliuotę. Sukurta sistema buvo įmanoma keisti spinduliuotės bangos ilgį 0,4 – 4,5 µm srityje, dėl ko tapo įmanomas įvairiomis optinėmis dangomis padengtų bandinių tyrimas.

Buvo ištirti didelio atspindžio SiO_2/TiO_2 dangų (1064 nm) užgarintų naudojant joninį asistavimą, taip pat dangų be joninio asistavimo ant ZnSe padėklo (1500 nm srytyje) sklaidos nuostoliai. Įvertinti sklaidos nuostoliai dangų ant CaF_2, o taip pat ant PMMA polimerų.

Nustatytas sklaidos nuostolių dydis zolių-gelių dangose, pagamintose įmerkimo ir išsukimo būdu. Sklaida šiose dangose yra mažesnė negu dangose užgarintose nenaudojant joninio asistavimo ir palyginama su joniniu asistavimu pagamintose dangose.

Buvo parodytas, kad optinių kristalų sklaidoje vyrauja tūrinės sklaidos įnašas. Buvo ištirti $LiInSe_2$ bei $LiInS_2$ sklaidos nuostoliai, taip pat, naudojant lazerinės sklaidos tomografijos metodą, buvo ištirtas tūrinių defektų pasiskirstymas greito bei lėto auginimo KDP kristaluose.

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