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# Optical properties of MgO thin films grown by laser ablation technique

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**Abstract** We have investigated the structural, linear and nonlinear optical properties of high-quality MgO thin films deposited by laser ablation technique on quartz substrates. The deposition process was carried out at various temperatures of the substrates from room temperature to 600 °C in order to investigate these properties of the films and their mutual influence. The structural and morphological properties of the films were investigated by X-ray diffraction. The quality of the films was improved with an increase of the substrate temperature. The linear optical properties of the films were studied by classic and time-resolved photoluminescence spectra in the broad range of the temperature form 13 to 300 K. An innovative time-resolved photoluminescence technique let us precisely measure the decay time in the real time. Results of these measurements reveal a simple exponential decay behavior typical for well oriented crystalline thin films. Presented spectra confirm high structural and linear optical quality of investigated films. The nonlinear optical properties of the films were investigated by third harmonic generation technique. Our results indicated that the substrate temperature slightly affected nonlinear optical properties and the values of third order nonlinear susceptibilities at the 600 °C were found to be smaller relative to RT. All these unusual properties of MgO thin films deposited by laser ablation technique made this material very interesting for the subsequent oriented growth of the other oxide a but also for potential other optical applications.

**Keywords** Magnesium oxide · Laser ablation · Time-of-flight mass spectrometry · Photoluminescence · Third harmonic generation

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## 1 Introduction

In the last two decades, materials including thin films with metal and non-metals oxides have been actively studied (Li et al. 2014; Ohnishi et al. 2004; Zawadzka et al. 2014a; Nomura et al. 2001). One of the main goals of these studies was finding the films with specific properties which allow using them as buffer layers. Magnesium oxide (MgO) seems to be one of the most interesting materials for such applications. MgO thin films attract significant attention due to their many interesting properties such as high electrical resistivity, high optical transparency, good chemical resistance, excellent thermal and thermodynamic stabilities, high secondary electron emission stability, low dielectric constant and low refractive index. These unusual properties caused that MgO films have been widely used as structural templates for the subsequent oriented growth of the other oxide films such as ferroelectrics and high temperature superconductors e.g. perovskite oxide. In many cases, the use of buffer layers allowed to decrease interdiffusion, lattice mismatch and other reactions. Moreover, MgO films have been also adopted as a protective layer in AC-plasma display panels to improve discharge characteristics and panel lifetime (Rho et al. 1999; Park et al. 1998; Sung and Kim 2000; Cho et al. 2015). Due to such applications, studies of the linear and nonlinear optical properties of MgO thin films have become very important.

The MgO thin films have been fabricated by various synthetic methods including e-beam evaporation (Kurt et al. 2010), atomic layer deposition (Burton et al. 2009), chemical vapor deposition (Wang et al. 2013), radio frequency magnetron sputtering (Yongle et al. 2014), spray pyrolysis (Bian et al. 2004) and sol gel process (Choi and Hwang 2000). However, all these methods have their limitations such as low deposition rate, low film density and poor homogeneity. Unlike these processes, laser ablation has been also successfully applied due to its flexible characteristics. For example, Tiwari et al. (Fork et al. 1991) reported a growth (1 1 1)-Textured MgO on Si (1 0 0) substrates from a magnesium oxide target. Also, epitaxial growth of (1 0 0)-oriented MgO on Si (1 0 0) using Mg metal target was realized by Fork et al. (Tiwari et al. 1991). From an application standpoint, conditions during the deposition process are extremely important because they can affect the structure of the forming film. It is well known that the structural properties of the buffer layer affect the orientation of the next deposited film. For example, MgO (1 0 0) plane has an advantage in preparing (0 0 1)-oriented tetragonal ferroelectric thin films, while the MgO (1 1 1) plane is beneficial for growing (1 1 1)-oriented rhombohedral ferroelectric thin films or (0 0 0 1)-oriented hexagonal thin films. Moreover, the laser ablation is a simple and environment-friendly method which guarantees high-quality and reproducible thin films.

In this paper, we present the growing procedure of MgO films using a simple ablation of MgO powder in vacuum environment. These structures were grown on quartz substrates by conventional method except energy of the pulse, which under experimental conditions didn't exceed 100 mJ. We have investigated influence of the different experimental conditions on quality of fabricated films. We have also performed measurements to study linear and nonlinear optical properties of these structures.

## 2 Experimental

Time distribution of ablated species inside the plasma plume is very important for understanding the PLD processes and the thin films deposition. The experimental apparatus for laser ablation process is shown in Fig. 1. A pulsed XeCl excimer laser beam

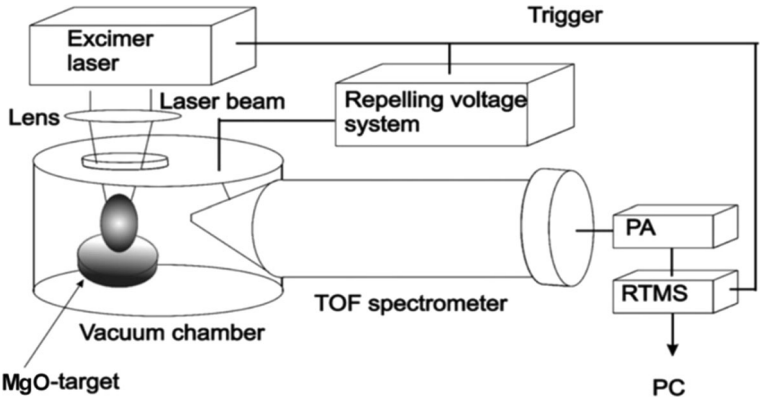


**Fig. 1** Laser ablation experimental set-up: *TOF* time of flight spectrometer, *PA* preamplifier, *RTMS* real time multichannel scaler (Rumianowski and Płóciennik 2006)

($\lambda = 308$ nm, pulse duration = 10 ns, repetition rate = 10 Hz) was incident on the rotating target at an angle of 45° with a spot diameter around 0.1 mm. A step motor was used for rotation of the target and the frequency was equal to 10 turns per minute. The distance between the target and substrate was around 4 cm. Quartz substrates were thoroughly cleaned to remove the surface native pollutions before loading into the chamber. The MgO ion–atomic plume was produced by irradiation of the MgO target by the excimer laser beam (Zawadzka et al. 2015a; Rumianowski and Płóciennik 2006; Zieliński et al. 2000). A constant field for time-of-flight (TOF) mass spectrometer was used to investigate the expansion dynamics of the ionic species ejected during ablation process (Zawadzka et al. 2015a; Rumianowski and Płóciennik 2006). Pressure within the vacuum chamber was kept at $5 \times 10^{-6}$ mbar and ablation process lasted 30 min. In order to study the structural as well as linear and nonlinear properties, the films were deposited in the substrate temperature range of room temperature (RT)—600 °C.

Ions, electrons and uncharged molecules were produced in the laser radiation interaction region at the MgO target inside the vacuum chamber. The COMSTOCK, model TOF 101 (Dygdała et al. 2000) mass spectrometer allowed us to distinguish the positive magnesium oxide ions. The positive ions were extracted into the spectrometer by an accelerating voltage between the target and the conical aperture of the spectrometer (Dygdała et al. 2009). The magnesium oxide ions were detected shot-to-shot by the tandem multichannel plate detector. The signal composed of a train of pulses was amplified and supplied to the input of the real time multichannel scaler (RTMS) (Zieliński et al. 2000; Zielinski et al. 1998). The RTMS was triggered by a fast photodiode which detects a reflection from the laser. The computer was used additionally for registering the laser power from the pulse energy meter for each laser shot.

## 3 Results and discussion

In present laser ablation process absorbed (from the laser beam) energy was sufficient to excite and ionize magnesium oxide molecules but not sufficient to break chemical bonds of the molecules. As a result, a supersonic jet of particles was ejected normal to the target

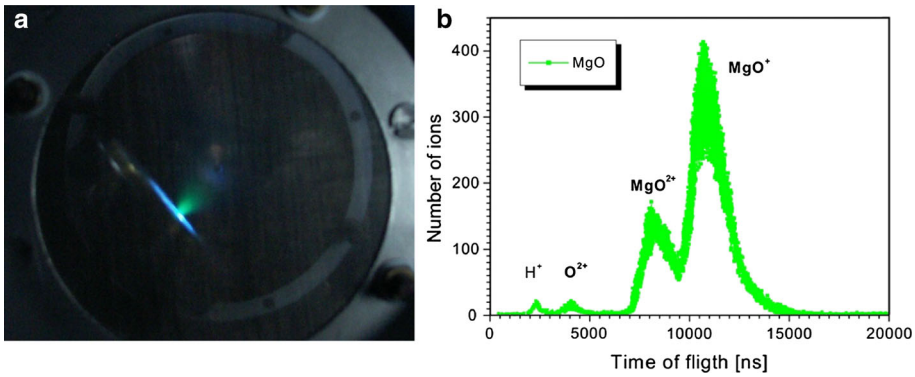


**Fig. 2** Plasma plume: typical shape after laser pulse at the energy 100 mJ (**a**), TOF spectrum for $MgO^{+}$ and $MgO^{2+}$ions (**b**)

surface giving rise a plasma plume. The plume was spreading towards the substrate with a strong forward-directed velocity distribution of the particles. Typical shape of the plasma plume during PLD process is shown in Fig. 2a. Figure 2b shows the TOF spectrum for MgO as a target material. The strong peaks assigned to $MgO^{+}$ and $MgO^{2+}$ have been observed.

### 3.1 Structural study of MgO thin films

Figure 3 shows the XRD measurement results of ablated MgO thin films as a function of the substrate temperature during the deposition process. All measurements were carried out by diffractometer using $CuK_{\alpha}$ ($\lambda = 0.1542$ nm) radiation. Two peaks attributed to diffraction from surfaces (111) and (200) were observed in most cases of XRD spectra for sample deposited at the room temperature (without heating of the substrate). Intensity of the peak (111) is about four times higher than peak (200). This result indicates that the growth of the films showed preferential orientation of crystallization. Intensity of this peak attributed to diffraction from surfaces (111) increased with increasing of the substrate temperature during ablation process, what leads to conclusion, that higher substrate's temperature causes the higher level of the internal structure orientation. This phenomenon can be explained by various molecules' mobility as a function of the substrate's temperature. The MgO molecules have higher surface mobility at higher temperature and they can move across the substrate surface and form. Hence, orientation of the internal structure obtained at higher temperature is more arranged. For lower temperatures this orientation is more accidental and weaker particles' mobility counteracts crystallization of the film.

Our results are in good agreement with that reported by Zhu et al. (2006), who found that MgO film grown at the temperature of the substrate equal to 400 °C under low oxygen atmosphere had a preferred orientation attributed to diffraction from surfaces (100) and (111). Magnesium oxide has a NaCl type crystal structure with face-centered-cubic Mg and O sublattices. The (100) planes are the cleavage planes of MgO, they are charge neutral and naturally occurring orientation, and have the lowest surface energy. Orientation attributed to surfaces (111) can be the fastest growing direction for a material with the NaCl structure (Mahieu et al. 2005). At low pressures or vacuum, the growth seems to be kinetically limited and (111) oriented grains have the largest perpendicular growth rate

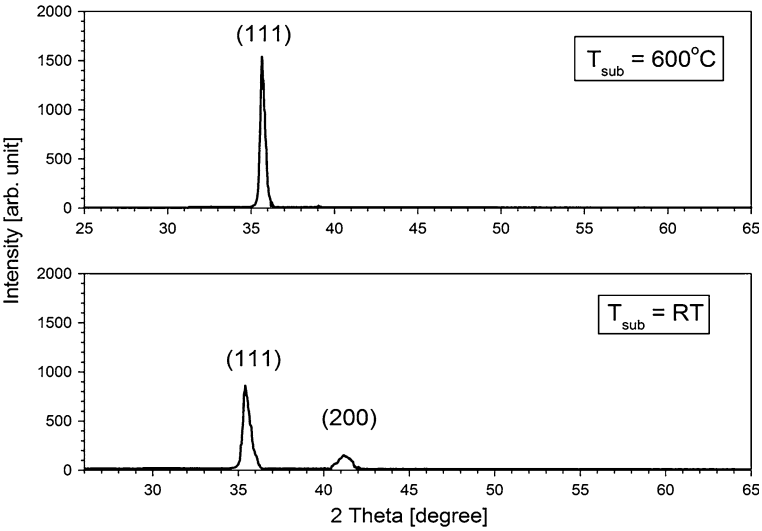


**Fig. 3** XRD measurements of MgO thin films as a function of the substrate temperature during the deposition process

over (100) oriented grains. Therefore, (111) out-of-plane orientation can develop in an evolutionary way. It is suggested that the kinetic energy of the ablated species in the plume can be changed by controlling the pressure during the deposition process, which allows the energy of the incident particles to be adjusted to obtain the expected orientations.

A half-width (FWHM) of these peaks decreased slightly with increasing temperature of the substrate. This finding indicated that the samples had a certain crystalline quality. The measured FWHMs of the sample deposited at room temperature were 0.43 and 0.78 for (111), and (200) respectively. The FWHMs of the film deposited at 600 °C were 0.29 for (111). Thus, increasing of the substrate temperature causes that the grain size becomes larger and the film exhibits a crystal structure due to the enhancement of the atomic migration ability. Therefore, the optical quality of the MgO thin films is improved by heating of the substrate during the deposition process. The temperature equal to 600 °C appears the best substrate temperature for the MgO thin films' deposition on the quartz plate by using laser ablation process. At higher temperatures, the quartz substrate is getting more plastic and the re-evaporation process occurs, which leads to re-reduce the size of grains.

### 3.2 Linear optical properties of the amorphous MgO thin films

The transmittance T(λ) of the films was measured at normal incidence in the spectral range 200–2000 nm using a double-beam spectrophotometer (Perkin Elmer Lambda 950 spectrophotometer). The spectral distribution of the transmittance for the as-deposited at different substrate's temperatures and next annealed (for 24 h at the temperature equal to 600 °C) samples is shown in Fig. 4a, b.

For most optical applications high transmittance in the UV–Vis–NIR range is very important. A relatively high transmittance in this spectral range and clear absorption edges of the films were observed for all films irrespective of the annealing process. All

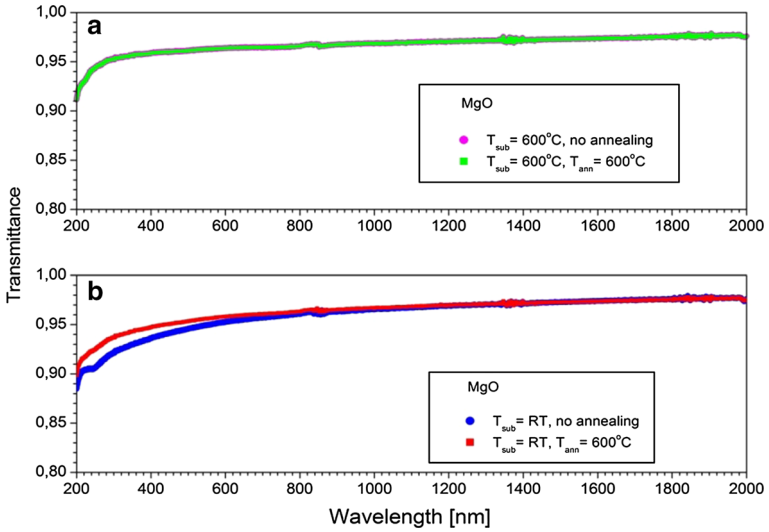


**Fig. 4** Transmittance spectra as a function of the substrate temperature and annealing temperature: **a** $T_{sub}$ = 600 °C and **b** $T_{sub}$ = RT (room temperature)

investigated magnesium oxide thin films have average transmittances >87 % in the investigated range. As the temperature of the annealing process has been increased the value of the transmittance slightly increased and reached a maximum (>91 %) after the process. The slightly weaker transmittance of the samples deposited at room temperature of the substrate results from the lower kinetic energy of the ablated species inside the plasma plume. The high transmittances of the films are attributed to larger-sized crystallites what eliminates light scattering. Investigated wavelength range did not allow determining the absorption edge of MgO films which is of about 160 nm.

Photoluminescence (PL) experiments were carried out under pulse excitation. Nitrogen laser ($\lambda$ = 337.1 nm, FWHM = 5 ns, power in pulse 20 kW and 10 Hz repetition rate) was used as excitation source. Photoluminescence signal was registered by using photomultiplier (HAMAMATSU R928) and the boxcar averager (162/164 PAR) (Zawadzka et al. 2014b). Measurements of the PL process were performed within temperature range from 13 to 300 K. Samples were placed in vacuum chamber and cooled to temperature equal to 13 K. Experimental spectra were registered after 30 min from reaching desirable temperature. Photoluminescence spectra of the MgO thin film on quartz plates recorded at different substrate's temperature equal to RT and 600 °C are shown in Fig. 5a, b.

Intensities of the photoluminescence process for all investigated samples were very low regardless of the applied substrate temperature. Photoluminescence spectra at different measuring temperature contained various emission features, which changed their intensity and positions with the temperature. The optical properties of thin films are depending on both intrinsic and extrinsic effects. Photoluminescence measurements give the possibility to determine the optical quality and the presence of impurities in the material. Precious data of the energy transport and other processes occurring within the film structure can be deducted from temperature dependent photoluminescence spectra. In the case of this study, measurements of the PL were carried out within temperature range from 13 to 300 K.

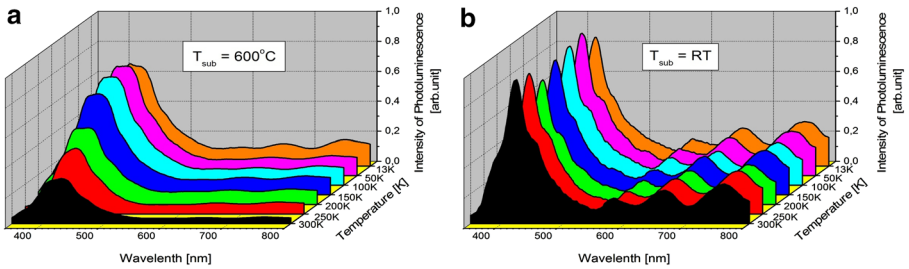


**Fig. 5** Photoluminescence spectra of MgO thin films on quartz at different measuring temperatures as a function of the substrate temperature: **a** $T_{sub}$ = 600 °C and **b** $T_{sub}$ = RT

To reconstruct the spectrum and to determine the individual peak position and its intensity within the spectrum standard multiple Gaussian fitting procedure was used. One main and five smaller bands were identified. The peak positions of the six Gaussian bands are located at about 395, 423, 464, 580, 660 and 760 nm. Intensities of the first three peaks were much greater than the other three and these three bands overlap each other. Blue and blue-green emission may originate from the defects in MgO such as oxygen and magnesium vacancies (Kar and Chaudhuri 2006) and interstitials positions of the atoms, presumably being generated during the high-temperature ablation process. The similar blue (Deepak et al. 2006) and blue-green emission (Zhang and Zhang 2002) have been observed for various MgO nanostructures. The red bands with the peaks located at 600–700 nm were reported by Zhang and Zhang (2002) and Chao (1971) and were assigned as vibrational sidebands lines in luminescence spectrum of bulk MgO with interstitials defects and/or or surface states. The band located at 760 nm may also be associated with the relaxation luminescence of the defect centers excited by mechanical stress. The relaxation luminescence of defect centers may exist because crack causes bond-breaking and nuclear motion, and promotes release of atoms and ions from the lattice sites, resulting in the creation of defects.

For MgO thin film deposited at room temperature of the substrate all six photoluminescence peaks regardless of the measurement's temperature had similar intensities. However, for the sample deposited at the substrate's temperature equal to 600 °C peaks intensities initially did not change their value but above 150 K started to fall rapidly. Additionally, the first three peaks considerably more overlap and formed one band. There were also much lower intensities of peaks associated with mechanical stresses and defects

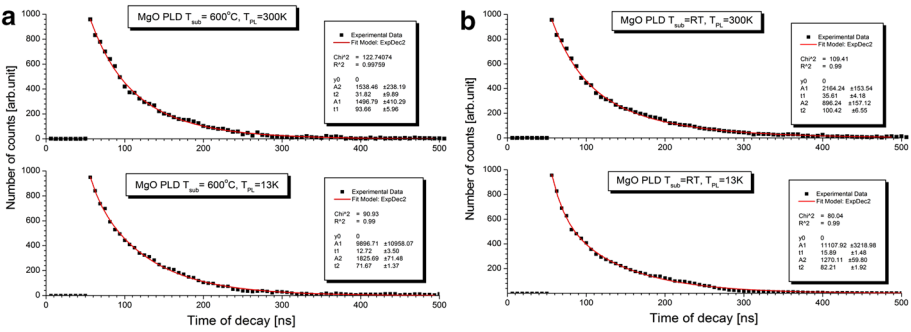


**Fig. 6** Time-resolved photoluminescence (TRPL) spectra of MgO at PL measurement's temperature 13 and 300 K: **a** $T_{sub}$ = 600 °C and **b** $T_{sub}$ = RT

of the crystal structure. This leads to the conclusion that the number of defects inside the films formed at the higher temperature of the substrate is significantly lower.

Dynamic of photoluminescence signal from MgO thin film prepared at the temperature of substrate equal to RT and 600 °C, recorded at different temperature is shown in Fig. 6a, b. These spectra were measured at the excitation energy's density equal to 460 mJ/cm$^2$. The time-resolved photoluminescence (TRPL) is a powerful, nondestructive technique commonly used for the optical characterization of the semiconductors (Zieliński et al. 2000; Zielinski et al. 1998; Zawadzka et al. 2013a). This technique allows measuring the all emission bands' lifetime. It is an important parameter related to material quality. The emission band's lifetime will vary with material phase and its crystallinity. The efficiency of the radiative recombination is strongly related to decay time of the particular transition. For all investigated samples, the decaying part of the TRPL data was well described by second order exponential decay function defined as: $A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2)$ (Płóciennik et al. 2014, 2013). The faster $\tau_1$ and slower $\tau_2$ decay constants were smaller for the sample deposited at the temperature of the substrate equal to 600 °C than that one deposited at room temperature regardless of the decay time of the PL measurements. Both of these decaying component were attributed to the blue and green–blue luminescence correspond to the vacancies, anti-site defects or deep band emission characteristic for phase containing internal strains and altered structural symmetry.

### 3.3 Nonlinear optical properties of the MgO thin films

The third order nonlinear optical susceptibilities ($\chi^{(3)}$) of investigated MgO thin films were examined by THG method. All measurements were carried out using the rotational Maker fringe technique (Maker et al. 1962) in the transmission scheme (Zawadzka et al. 2013b, 2015b; Płóciennik et al. 2015). A silica glass plate has been used as reference materials for the third harmonic generation measurements. A Q-switched mode-locked Nd: YAG laser operating at 1064 nm with 16 ps pulse duration, 1.6 mJ power per pulse and repetition frequency of 10 Hz was used as a fundamental beam. The laser beam has been found to exhibit Gaussian spatial and temporal profile. The beam's diameter was 0.4 mm at the film and the applied power density was about 5 GW/cm$^2$. The fundamental beam was focused on the sample using a lens with 250 mm focal length. A rotation stage with a resolution of 0.5° (model Standa 8MR180) with the mounted sample of magnesium oxide thin film allowed the variation of the incidence angle. After passing the sample, the transmitting filter was used to cut the pump laser beam before the photomultiplier. Detector saturation was prevented using linear neutral density filters, whose transmittance value was taken into account during data fitting. The third harmonic signals were detected by the photomultiplier tube model: HAMAMATSU R1828-01, integrated by a box-car average system and processed by a computer. A portion of the input beam was reflected and measured by a fast photodiode Ph2 to monitor the input energy. For each sample, the measurements of the third harmonic signal intensity as a function of the angle of incidence was performed on a total of 40 laser impulses for each angular position. Finally, the so-called Maker (Maker et al. 1962; Zawadzka et al. 2013b, 2015b) fringes were generated by rotating the sample through the range of ±60° to the normal and recorded.

In the THG technique, an incident laser beam of high intensity at the frequency ω interacts with a nonlinear medium and generates an additional beam at a frequency 3ω. The third harmonic beam corresponds to a pure coherent electronic nonlinearity. This technique is one of the most informative methods for evaluating the electronic contribution of the real part of the third order nonlinear optical susceptibilities $\chi^{(3)}_{\text{elec}}$. The THG technique is

sensitive to ultrafast electronic mechanisms of nonlinear response within femtosecond relaxation times and it is almost insensitive to slower effects such as thermal relaxation. Moreover, THG is a much more accurate technique because it allows to directly measure the nonlinear susceptibility and the result is undisturbed by the wavelength of fundamental laser beam. In the case of the DFWM technique, nonlinear signal from the sample is observed as an intensity decrease of the fundamental beams (all waves are focused on the sample and have the same wavelength).

A few theoretical models, using various approximations, have been described in order to determine the value of $\chi^{3}_{elec}$ from the shape of the experimental curves of Maker fringes (Maker et al. 1962; Zawadzka et al. 2013b, 2015b) obtained by the THG technique. We used comparative model for explanation of our experimental results. This model compares (Lee et al. 2001) directly the maximum of light intensities amplitudes for the third harmonic of nonlinear medium with those of the reference material used for its calibration of the experimental setup. The value of the third order nonlinear susceptibility $\chi^{3}_{elec}$ is derived by comparing third harmonic peak intensities of the sample and reference material (fused silica glass).

The comparative model gives the order of magnitude of the third order nonlinear susceptibility $\chi^{3}_{elec}$. The refractive indices and third order susceptibility are considered real, so the weak absorption of the typical nonlinear sample is ignored. The $\chi^{3}_{elec}$ value of the investigated material is calculated using the following Eq. (1):

$$\chi^{\langle 3\rangle}_{elec} = \chi^{\langle 3\rangle}_{RM} \frac{2L_{CRM}}{\pi d} \sqrt{\frac{I_{3\omega}}{I_{3\omega RM}}}, \tag{1}$$

for the thin film whose thickness $d$ is much smaller than the coherence length $L_{CRM}$ of reference material (fused silica plate). The index "*RM*" corresponds to the reference material, $\chi^{3}_{RM}$ is the third order nonlinear optical susceptibility and $I_{3\omega RM}$ is the THG intensity of the reference measured under identical conditions to the sample. $I_{3\omega}$ is the absorption-free THG signal from the fringes of the sample. The value of $\chi^{3}_{RM}$ for fused silica glass equals $2.0 \times 10^{-22}$ $m^2/V^2$ and $3.11 \times 10^{-14}$ esu (at $\lambda_{\omega} = 1064$ nm) and is reported in the literature (Chao 1971; Lee et al. 2001; Kulyk et al. 2010).

Figure 7a, b show experimental results of the third harmonic intensity of two samples deposited on 1 mm thick quartz substrate at different substrate temperature: RT and

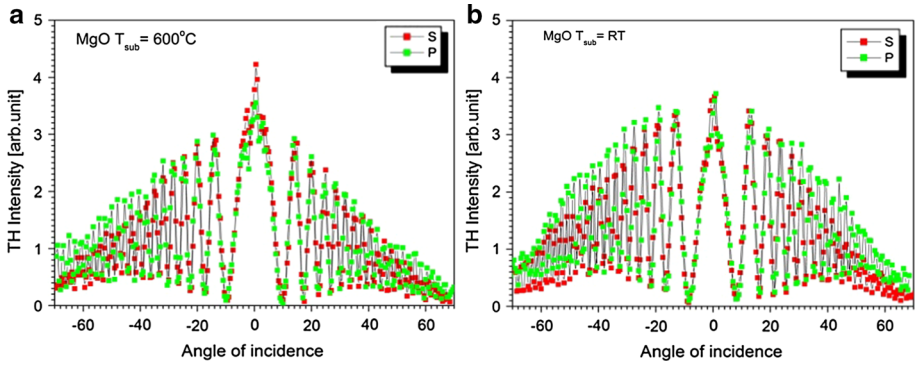


**Fig. 7** THG signal dependence as a function of the incident angle of the fundamental beam and temperature applying during the ablation process: **a** $T_{sub} = 600$ °C and **b** $T_{sub} = RT$ (S, vertical; P, horizontal polarizations of the fundamental laser beam)

**Table 1** Fundamental beam polarization, laser ablation process substrate temperature and results of nonlinear optical susceptibility $\chi^{\langle 3\rangle}_{elec}$ obtained for MgO thin films from calculation based on Comparative model

| Sample | Polarzation | Substrate temperature (°C) | $\chi^{\langle 3\rangle}_{elec}$ | | Model |
|---|---|---|---|---|---|
| | | | ($10^{-22}$ $m^2/V^2$) | ($10^{-14}$ esu) | |
| MgO | P | RT | 1.52 ± 0.22 | 2.36 ± 0.34 | Comparative |
| | S | RT | 1.59 ± 0.21 | 2.47 ± 0.33 | |
| | P | 600 | 1.44 ± 0.21 | 2.24 ± 0.33 | |
| | S | 600 | 1.41 ± 0.21 | 2.19 ± 0.33 | |

600 °C. THG signal were measured for two linear (S, vertical and P, horizontal) polarizations of the fundamental laser beam. To determine the third order optical susceptibility of the sample, the analysis of the experimental results was carried out by using Maker fringes technique. Detailed analyses of the spectra show many fringes, which become tighter with the increase of incidence angle $\theta$, because the length of interaction $L$ in sample increases nonlinearly with the angle. When the thickness of the material $d$ is higher than the coherence length $L_{CRM}$, the wave's constraint and free interfere to each other, and the intensity of third harmonic signal can pass through a series of maxima and minima.

Experimental curves display an oscillatory signal and intensities depend on the incidence angle, regardless of the substrate temperature. Good symmetry of TH signal for both deposited at RT and 600 °C samples were found and proved the smooth surface and good quality of the films. It was found that the dependence of the TH intensity on the polarization of the fundamental laser beam does not show a simply relation. Also intensity of the TH signals just slightly depends on the substrate temperature during laser ablation process. Moreover, TH intensity of the MgO thin films deposited at 600 °C was almost independent on the polarization of the laser beam for all THG measurements. In the case of RT deposited films TH intensity showed a slight dependence for all THG measurements. These measurements and theoretical calculation based on described above Comparative models allow us estimating the value of the real part of the third order nonlinear optical susceptibility.

Presented experimental results and theoretical calculation based on Comparative model allow us estimate values of the third order nonlinear optical susceptibility $\chi^{\langle 3\rangle}_{elec}$ for both temperatures of the substrate during the deposition process for Magnesium oxide thin films (Table 1). It was found, that the value of the electronic part of the third order nonlinear optical susceptibility decreased with the substrate temperature increasing for all investigated samples. Values of $\chi^{\langle 3\rangle}_{elec}$ for the sample deposited at 600 °C e are slightly smaller than for the samples fabricated at room temperature. Our calculated results are comparable to those presented in the literature (Adachi 2004; Kulyk et al. 2009).

## 4 Conclusion

This paper was focused on the growth and optical properties of magnesium oxide (MgO) thin films fabricated by laser ablation process. The improvement of the structure and the emission characteristics for MgO thin films by application proper substrate temperature during ablation process has been established. XRD spectra showed that the substrate temperature influences the FWHM of the diffraction peaks therefore also grain growth. The Photoluminescence spectra in wide range of temperatures: 13–300 K also showed

improvement in optical properties of the magnesium oxide films deposited at higher temperature of the substrate. Experimental results of TRPL shed new light on the controversial issue of the interpretation of the luminescence spectra of MgO thin films. Various physical mechanisms and their impact on observed spectra were depicted. THG experiments were performed and the values of the third order nonlinear susceptibility (electronic part) were found to be similar for well oriented crystal samples than those with the polycrystalline structure. This difference in THG intensities for the MgO thin films prompts that the well oriented crystalline thin films play a very important role as an intermediate films for various multilayer structures. Obtained results confirmed good structural and optical films' quality and the substrate temperature was crucial to controlling structure and preparing high-quality MgO thin films with a smooth surface.



## References

Adachi, S.: Handbook of Physical Properties of Semiconductors. Kluwer Academic Publishers, Boston (2004)

Bian, J.M., Li, X.M., Chen, T.L., Gao, X.D., Yu, W.D.: Preparation of high quality MgO thin films by ultrasonic spray pyrolysis. Appl. Surf. Sci. **228**, 297–301 (2004)

Burton, B.B., Goldstein, D.N., George, S.M.: Atomic layer deposition of MgO using Bis(ethylcyclopentadienyl)magnesium and $H_2O$. J. Phys. Chem. C **113**, 1939–1946 (2009)

Chao, C.C.: Charge-transfer luminescence of $Cr^{3+}$ in magnesium oxide. J. Phys. Chem. Solids **32**, 2517–2528 (1971)

Cho, S.H., Lee, S.M., Kim, W.H., Choi, K.C.: Effect of gold nanorods in the MgO protective layer of AC plasma display panels. ACS Appl. Mater. Interfaces **7**(14), 7559–7565 (2015)

Choi, H., Hwang, S.: Sol–gel-derived magnesium oxide precursor for thin-film fabrication. J. Mater. Res. **15**, 842–845 (2000)

Deepak, F.L., Saldanha, P., Vivekchand, S.R.C., Govindaraj, A.: A study of the dispersions of metal oxide nanowires in polar solvents. Chem. Phys. Lett. **417**, 535–539 (2006)

Dygdała, R.S., Krzysztof, K., Stefański, K., Zawadzka, A., Rumianowski, R., Zieliński, M.: The plasma phenomena in three-photon ionization of Ca. J. Phys. D Appl. Phys. **33**, 41–53 (2000)

Dygdała, R.S., Zieliński, M., Płóciennik, P., Zawadzka, A., Rumianowski, R.: Time-of-flight repeller circuit application in laser ablation experiment. Surf. Coat. Technol. **203**, 2328–2332 (2009)

Fork, D.K., Ponce, F.A., Tramontana, J.C., Geballe, T.H.: Epitaxial MgO on Si (001) for Y–Ba–Cu–O thin-film growth by pulsed laser deposition. Appl. Phys. Lett. **58**, 2294–2296 (1991)

Kar, S., Chaudhuri, S.: Synthesis and characterization of one-dimensional MgO nanostructures. J. Nanosci. Nanotechnol. **6**, 1447–1452 (2006)

Kulyk, B., Sahraoui, B., Figà, V., Turko, B., Rudyk, V., Kapustianyk, V.: Influence of Ag, Cu dopants on the second and third harmonic response of ZnO films. J. Alloys Compd. **481**(1–2), 819–825 (2009)

Kulyk, B., Kapustianyk, V., Tsybulskyy, V., Krupka, O., Sahraoui, B.: Optical properties of ZnO/PMMA nanocomposite films. J. Alloys Compd. **502**(1), 24–27 (2010)

Kurt, H., Oguz, K., Niizeki, T., Coey, J.M.D.: Giant tunneling magnetoresistance with electron beam evaporated MgO barrier and CoFeB electrodes. J. Appl. Phys. **107**, 083920 (2010)

Lee, G.J., Cha, S.W., Leon, S.J., Jin, J.I., Yoon, J.S.: Second order nonlinear properties of unpoled bent molecules in powder and in vacuum deposited film. J. Korean Phys. Soc. **39**, 912–915 (2001)

Li, Y., Qi, Z., Liu, M., Wang, Y., Cheng, X., Zhangand, G., Sheng, L.: Photoluminescence of monolayer $MoS_2$ on $LaAlO_3$ and $SrTiO_3$ substrates. Nanoscale **6**, 15248–15254 (2014)

Mahieu, S., Ghekiere, P., De Winter, G., Heirwegh, S., Depla, D., De Gryse, R., Lebedev, O.I., Van Tendeloo, G.: Mechanism of preferential orientation in sputter deposited titanium nitride and yttria-stabilized zirconia layers. J. Cryst. Growth **279**, 100–109 (2005)

Maker, P.D., Terhune, R.W., Nisenhoff, M., Savage, C.M.: Effects of dispersion and focusing on the production of optical harmonics. Phys. Rev. Lett. **8**, 21–22 (1962)

Nomura, K., Hoshi, S., Nakamura, Y., Izumi, T., Shiohara, Y.: Preferential growth mechanism of $REBa_2Cu_3O_y$ (RE = Y, Nd) crystal on MgO substrate by liquid phase epitaxy. J. Mater. Res. **16**(10), 2947–2958 (2001)

Ohnishi, T., Shibuya, K., Lippmaa, M., Kobayashi, D., Kumigashira, H., Oshima, M., Koinuma, H.: Preparation of thermally stable $TiO_2$—terminated $SrTiO_3$(100) substrate surfaces. Appl. Phys. Lett. **85**, 272–274 (2004)

Park, C.H., Lee, W.G., et al.: Surface discharge characteristics of MgO thin films prepared by RF reactive magnetron sputtering. Surf. Coat. Technol. **110**, 128–135 (1998)

Płóciennik, P., Zawadzka, A., Łukasiak, Z., Brodzińska, K., Korcala, A., Bartkiewicz, K.: Pulsed laser deposition of hafnium oxide on quartz substrate. In: Transparent Optical Networks (ICTON), 2013 15th International Conference, IEEE ICTON 2013, pp. 1–3. ISSN: 2161-2056 (2013). doi:10.1109/ICTON.2013.6602905

Płóciennik, P., Zawadzka, A., Strzelecki, J., Łukasiak, Z., Korcala, A.: Pulsed laser deposition (PLD) of hafnium oxide thin films. In: Transparent Optical Networks (ICTON), 2014 16th International Conference, IEEE ICTON 2014, vol. 1, pp. 1–3 (2014). doi:10.1109/ICTON.2014.6876620

Płóciennik, P., Zawadzka, A., Korcala, A.: Study of ZnO thin film deposited by PVD. In: Transparent Optical Networks (ICTON), 2015 17th International Conference IEEE ICTON 2015, vol. 1, pp. 1–3 (2015). doi:10.1109/ICTON.2015.7193661

Rho, S.J., Baik, H.K., et al.: The structural, optical and secondary electron emission properties of MgO and Mg–O–Cs thin films prepared by ion beam assisted deposition. Thin Solid Films **355–356**, 55–59 (1999)

Rumianowski, R., Płóciennik, P.: C20 clusters formation by laser ablation of graphite at 308 nm. Surf. Eng. **22**, 409–410 (2006)

Sung, M.M., Kim, C., Kim, C.G., Kim, Y.: Epitaxial growth of MgO films on Si(111) by metal organic chemical vapor deposition. J. Cryst. Growth **210**, 651–654 (2000)

Tiwari, P., Sharan, S., Narayan, J.: Growth of ceramic thin films on Si(100) using an in situ laser deposition technique. Appl. Phys. **69**, 8358–8361 (1991)

Wang, W.B., Yang, Y., Yanguas-Gil, A., Chang, N.N., Girolami, G.S., Abelson, J.R.: Highly conformal magnesium oxide thin films by low-temperature chemical vapor deposition from Mg $(H_3BNMe_2BH_3)_2$ and water. Appl. Phys. Lett. **102**, 101605 (2013)

Yongle, L., Yuming, Z., Daqing, X., Hui, G., Yimen, Z., Yuchen, L.: Influence of oxygen content on the crystallinity of MgO layers in magnetic tunnel junctions. J. Semicond. **35**(8), 083005 (2014)

Zawadzka, A., Płóciennik, P., Strzelecki, J., Łukasiak, Z., Sahraoui, B.: Photophysical properties of Alq 3 thin films. Opt. Mater. **36**, 91–97 (2013a)

Zawadzka, A., Płóciennik, P., Strzelecki, J., Pranaitis, M., Dabos-Seignon, S., Sahraoui, B.: Structural and nonlinear optical properties of as-grown and annealed metallophthalocyanine thin films. Thin Solid Films **545**, 429–437 (2013b)

Zawadzka, A., Płóciennik, P., Strzelecki, J.: Temperature-dependent luminescence dynamics for ZnO thin films. Opt. Quantum Electron. **46**, 87–101 (2014a)

Zawadzka, A., Płóciennik, P., Strzelecki, J., Sahraoui, B.: Transparent amorphous zinc oxide thin films for NLO applications. Opt. Mater. **37**, 327–337 (2014b)

Zawadzka, A., Płóciennik, P., El Kouari, Y., Bougharraf, H., Sahraoui, B.: Linear and nonlinear optical properties of ZnO thin films deposited by pulsed laser deposition. J. Lumin. (2015a). doi:10.1016/j.jlumin.2015.04.020

Zawadzka, A., Karakas, A., Płóciennik, P., Szatkowski, J., Łukasiak, Z., Kapceoglu, A., Ceylan, Y., Sahraou, B.: Optical and structural characterization of thin films containing metallophthalocyanine chlorides. Dyes Pigments **112**, 116–126 (2015b)

Zhang, J., Zhang, L.: Intensive green light emission from MgO nanobelts. Chem. Phys. Lett. **363**, 293–297 (2002)

Zhu, T.J., Lu, L., Zhao, X.B.: Orientation of MgO thin films grown by pulsed laser deposition. Mater. Sci. Eng. B **129**, 96–99 (2006)

Zielinski, M., Dygdala, R.S., Płóciennik, P.: Counting errors in a real-time multichannel scaler. Rev. Sci. Instrum. **69**(1), 299–305 (1998)

Zieliński, M., Dygdała, R.S., Karasek, K., Zawadzka, A.: Real-time multichannel scaler measurement of oscillator instabilities. Rev. Sci. Instrum. **71**, 2577–2581 (2000)