Ultrafast electron dynamics and energy deposition during IR-visible femtosecond laser ablation of fluorite

P A Danilov¹*, M L Galkin¹², M S Kovalev², A E Rupasov¹, M P Smayev¹³, A S Zolot’ko¹, A A Ionin¹, S I Kudryashov¹ and R A Zakoldaev⁴

¹Lebedev Physical Institute, Russian Academy of Sciences, Moscow, 119991 Russia
²Bauman Moscow State Technical University, Moscow, 105005 Russia
³Mendeleev University of Chemical Technology of Russia, Moscow, 125047 Russia
⁴ITMO University, 197101 Saint-Petersburg, Russia

* danilovpa@lebedev.ru

Abstract. The action of ultrashort laser pulses on dielectrics leads to the processes of redistribution of the laser pulse energy in the interaction region. The mechanisms of ultrafast energy deposition and the electron dynamics under the action of ultrashort laser pulses determine the type of final structures or elements in the transparent dielectrics. One type of these elements is a birefringent nanogratings formed with an orientation perpendicular to the laser radiation. In this work, laser-induced birefringent structures in calcium fluoride (CaF₂) bulk were obtained under the action of tightly-focused laser pulses with durations 0.3–3.8 ps and wavelengths of 515 and 1030 nm. Birefringence in modified region was analyzed using polarization contrast optical microscope with Abrio imaging system.

1. Introduction

Femtosecond (fs) direct laser writing in bulk dielectrics is broadly using in different applications: waveguide writing [1], fabrication of birefringent structures [2], holographic recording [3] and optical memory storage [4]. The underlying processes under tightly focusing ultrashort laser pulses are highly non-linear both in temporal and spatial domains [5].

Contrasting nanogrids are one type of structures formed by ultrashort laser pulses in transparent materials. These elements are formed with dash orientation perpendicular to the polarization of laser radiation [6]. Due to periodic modulation of the refractive index, the speed of light is different in directions perpendicular and parallel to the lines of the nanogratings, which leads to optical anisotropy similar to birefringent crystals. The “slow” axis of the nanogrids changes with the rotation of the polarization, leading to a periodic change in the radiation intensity in a polarizing microscope, which allows the initial identification of nanogrids. The mechanisms of the formation of nanogrids are currently associated, at the stage of electronic dynamics, with the interference of incident radiation and a plasma wave [6] followed by a multi-momentum transformation of quasiperiodic spherical nanoplasms into nanoplanes [7], the formation (for example in fused silica) of excitons and their self-localization with the formation of point defects — oxygen-deficient ODC centers [8]. The parameters of the gratings are flexibly controlled by the parameters of laser radiation — wavelength, pulse duration
and energy, their number and repetition rate, and scanning direction [9]. Notable, various glasses [1-4] and fused silica [7, 8] are most often used as samples in such studies, and less attention is paid to other crystals.

In this work, we studied the birefringence of laser-induced structures in CaF$_2$ depending on the laser wavelength, the pulse energy and the pulse duration.

2. Experimental details

In these experiments we used fundamental (FH, 1030 nm) and second-harmonic (SH, 515nm) pulses of the Yb-fiber laser Satsuma (Amplitude Systemes) with a repetition rate of 0–2 MHz and maximum pulse energy $E$ in the TEM$_{00}$ mode $10$ µJ and $4$ µJ respectively. FWHM width $\tau_{\text{las}}$ of the fundamental harmonic was varied by means of an output grating compressor over the range $0.3 – 3.8$ ps. The CaF$_2$ bulk were mounted on the PC-driven three dimensional motorized translation stage and moved with the scanning speed of $25$ µm/s. The laser pulses with variable energies ($0.06 – 0.8$ µJ) and repetition rate $100$ kHz were focused $50$ µm below the sample surface by a micro-objective with a numerical aperture $\text{NA} = 0.65$ (figure 1).

Birefringence in the modified region was analyzed by means of Olympus BX-61 optical microscope equipped with an Abrio IM 2.2 (Cambridge Research and Instrumentation, Inc.) imaging system for microanalysis of birefringence. The principle of operation of the Abrio system was described in [10]. The retardance (Ret) is characterized by the shift between two orthogonally polarized waves that propagated through an anisotropic structure expressed in units of wavelength and in given by

$$\text{Ret} = \Delta n \times L,$$

where $\Delta n = |n_e - n_o|$, $n_e$ is the refractive index of the extraordinary wave, $n_o$ is the refractive index of the ordinary wave, and $L$ is the size of the birefringent object along the direction of observation (the optical axis of the microscope). In addition, the Abrio system displays orientation of the slow axis of the birefringent object and shows the direction of the axis characterized by the highest value of the refractive index.

![Experimental setup for laser recording in CaF$_2$ volume with the computer (PC) control. EM – energy meter, BS – 50% beam splitter.](image)

3. Results

The birefringent structures in CaF$_2$ are 225 µm lines (Fig. 2 a,b) deposited at a depth of $50$ µm from the surface. We used laser pulses with durations of $0.3$, $0.9$, $1.8$, and $3.8$ ps, a repetition rate of $100$ kHz, and pulse energies of $0.064–0.4$ µJ for $515$ nm and $0.2–0.8$ µJ for $1030$ nm. The scanning speed was $25$
µm/s for all series of lines. This corresponds to an exposure of N ~ 8×10^3 pulses per spot. At least three lines were recorded for each value of energy and pulse duration.

Figure 2. Optical images of birefringent structures in CaF_2 for wavelengths 515 nm (a) and 1030 nm (b). The black arrow indicates the scanning direction from the starting point. The scale bar is 100 µm.

Then, the analysis of birefringence with the construction of a pseudo-color map of the modified area was carried out using the Abrio system (optical microscope Olympus BX-61; objective Olympus UPlanFL N 40x with NA = 0.75). The colors of the structures (red of turquoise) indicates the direction of the extraordinary wave.

Figure 3. The averaged (over five points) retardance depending on the laser pulse energies.

Figure: 3 shows the averaged (over five points) retardance depending on the laser pulse energy for different pulse durations and wavelengths. The maximum values of retardance are ~90 nm for 1030 nm, 300 fs laser pulses and ~65 nm for 515 nm and 920 fs laser pulses. The structures thickness is ~25 µm and measured by 3D-scanning confocal Raman microscope (Confotec MR350). It corresponds to the maximum change in refractive index 4×10^{-3} for 1030 nm and 3×10^{-3} for 515 nm, respectively.

4. Conclusions

In this work, the birefringence of microstructures in CaF_2 was studied depending on laser wavelengths, pulse energies and pulse durations. The maximum change in refractive index 4×10^{-3} for 1030 nm (300 fs) and 3×10^{-3} for 515 nm (920 fs). The differences in experimental values seem to be related to intensity of generated plasma wave under the tightly focused laser pulses that affects the size of the modified region with birefringent nanogratings.
Acknowledgments.
This work was funded by the grant of Russian Science Foundation (project № 20-71-10103).

References
[1] Chen F and de Aldana J V 2014 Laser & Photon. Rev. 8 (2) p. 251
[2] Shimotsuma Y, Sakakura M, Kazansky P G, Beresna M, Qiu J, Miura K, and Hirao K 2010 Adv. Mater. 22 (36), p. 4039
[3] Beresna M, Gecevičius M, Kazansky P G, and Gert t S 2011 Appl. Phys. Lett. 98 (20), p. 201101
[4] Hong M H, Luk’Yanchuk B, Huang S M, Ong T S, Van L H, and Chong T C 2004 Appl. Phys. A 79 (4-6), p. 791
[5] Couairon A, Sudrie L, Franco M, Prade B, and Mysyrowicz A 2005 Phys. Rev. B 71, p. 125435
[6] Shimotsuma Y, Kazansky P G, Qiu J, and K auoki H 2003 Phys Rev Lett; 91 p. 247405
[7] Rajeev P P, Gertsvolf M, Hnatovsky C, Simova E, Taylor R S, Corkum P B, Rayner D M and Bhardwaj V R 2007 J Phys B Atom Mol Opt Phys 40 p. 273
[8] Richter S, Heinrich M, Döring S, Tümmermann A, and Nolte S 2012 J Laser Appl. 24, p. 042008
[9] Zhang B, Liu X, Qiu J 2019 J. Materiomics, 5(1), pp. 1-14.
[10] Mehta S. B., Shribak M., and Oldenbourg, R. 2013. J. Opt., 15(9), p. 094007.