Antisymmetric Distribution of Permanent Refractive Index Change in $\beta$-Ba$_2$B$_4$O$_7$ Crystal Under Exposure of Femtosecond Pulses.

A.G. Okhrimchuk$^1$, S.G. Grechin$^2$, A.E. Kokh$^3$, V. Mezentsev$^4$

$^1$Fiber Optics Research Center of RAS, 38 Vavilova Street, Moscow 119333, Russia
$^2$Bauman Moscow State Technical University, 5 2-nd Bauman Street, Moscow, 107005, Russia.
$^3$Sobolev Institute of Geology and Mineralogy of SB RAS, 3 Acad. Koptyugina Ave., Novosibirsk 630090, Russia.
$^4$Aston University, Aston Triangle, Birmingham, B4 7ET, UK

E-mail: a.okhrimchuk@aston.ac.uk

Abstract: We have observed unusual asymmetrical refractive index change as a result of femtosecond laser inscription in a crystal without center of inversion. Profile of the refractive index change exhibits sign turn within the domain of femtosecond pulse exposure.

Femtosecond writing is a promising and flexible technology for fabrication of waveguide in transparent dielectrics. Low loss waveguides were demonstrated recently in both the rare-earth doped and non-linear (\( \chi(2) \)) crystals. Efficient lasing, frequency doubling and electro-optical modulation were achieved. The waveguide patterning is due to a phenomenon of permanent refractive index change under exposure of tightly focused beam of a femtosecond laser. The waveguides are typically formed by one or more several elementary tracks. Refractive index change inside a track can be either negative or positive depending upon material. In non-linear LiNbO$_3$ crystal sign of the index change can be different for ordinary and extraordinary waves, but up to now waveguide profile was reported to be symmetrical relative to central axis of the femtosecond writing beam at least for cases when the writing beam had a perfect symmetrical shape [1,2]. In this paper we report an antisymmetric refractive index change that is associated with intrinsic crystal symmetry and not down to a beam profile. As far as we know such phenomenon was not observed before.

We investigated the femtosecond writing in non-linear $\chi(2)$ BBO crystal in the regime of a single pulse exposure at the wavelength of 800 nm. Pulse duration was of 110 fs. We investigated X-cut plate of the crystal, thus femtosecond beam with Gaussian radial intensity distribution (\( M^2 = 1.05 \)) propagated perpendicular to the symmetry plane, which is singular in 3m space group of crystal symmetry. A lens with NA=0.55 was used to focus a beam at the depth of 130 $\mu$m under the crystal surface. Polarization of the femtosecond beam was parallel to either Z (extraordinary wave) or Y optical axes. Significantly different inscription thresholds were found in these cases at 200 nJ and 40 nJ correspondingly.

Geometry and profiles of refractive index change were investigated by measuring the phase delay under microscopic observation along [100] crystallographic axis (perpendicular to the plate). We have found a complicated geometry of a modified region, which does not follow an axial symmetry of intensity distribution in writing beam, but its structure correlated with directions of crystallographic axes. Modified regions were sufficiently differed for extraordinary and ordinary types of writing beam (Fig.1). Geometry in a form of elongated ellipsoid was found in the case of the writing beam of extraordinary type (E/Z). In this case an unusual antisymmetric profile of refractive index change with was found along the Y optical axis, that is, the refractive index change alters sign. Conversely, the profile of refractive index change was completely symmetrical along the Z axis. The refractive index change is birefringent, and the asymmetry was most pronounced for the extraordinary writing wave. Moreover a record value for refractive index changes in crystals as much as 0.07 was found at these conditions.

For writing beam of ordinary type (E/Y) a modified region has completely different shapes in dependency of polarization of viewing (Fig.1).

We associate the observed antisymmetric profile of refractive index change being originated by the charge separation due to lack of the central symmetry in the crystal investigated. The electro-optical and elasto-optical effects are eventually responsible for the observed phenomenon. Dependence of femtosecond modification upon direction of femtosecond writing was observed in LiNbO$_3$ crystal in [1] too. Although conditions of this experiment sufficiently differ from our one, nature of the phenomena could be the same, as LiNbO$_3$ crystal belongs the same space group as BBO crystal, and an anomaly was observed along the same optical axis (Y).
Fig. 1. Bright field microscopic images of modifications in a single pulse regime for extraordinary writing beam ($\mathbf{E}||\mathbf{Z}$) (a and b), and ordinary writing beam ($\mathbf{E}||\mathbf{Y}$) (c and d). a) and c) is viewed by extraordinary wave, b) and d) is viewed by ordinary wave.

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