MATEC Web of Conferences **8**, 02006 (2013)
DOI: 10.1051/matecconf/20130802006

Direct laser writing of efficient effective second order nonlinear optical properties in a tailored silver-doped phosphate glass

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This last decade, femtosecond (fs) direct laser writing (DLW) has proposed an exponentially growing number of achievements thanks to the simultaneous improvements in both fs laser technologies and material sciences, leading to innovative light/matter interactions [1]. DLW has led to micro and nano-photonics demonstrations of mechanical, chemical and optical contrast in the bulk or at the surface, as differential etching solubility, new linear absorption and fluorescence or plasmonic properties, as well as dielectric phase transition [2]. Nonlinear optical contrast can also appear from third-order nonlinear response enhancement due to density contrast, as proposed for perennial high density data storage [3, 4]. DLW also showed second-order nonlinear optical properties, even in a glassy environment, either due to the localized crystallization of non-centrosymmetric nano-crystalittes [5] or to the production of a buried static space charge separation at the root of the electric field induced second harmonic generation (EFISHG) [6].

Laser-induced optical poling, leading to the EFISHG effect, is a very promising approach. Indeed, it generates very limited index changes, no phase transition and no interface, which prevents any potential high refractive index contrast and the associated drawback of light diffusion as generally the case for nano-crystallisation in bulk glassy matrices [5]. Laser-induced optical poling is thus a powerful approach for three-dimension high density data encoding, with multiple layers in the depth of the photosensitive material, which cannot be performed under thermally-assisted electric poling where the very strong permanent static electric field, leading to a few pm/V effective EFISHG amplitude, is limited to surface structuring with depths typically limited to 10 μm [7].

We present here recent EFISHG results obtained thanks to the fs infrared DLW of a tailored silver co-doped phosphate glass. We report of significantly new EFISHG optical response with respect to previous works [6], associated to an original electric field spatial distribution with near-to-diffraction limit. Power and polarization scans confirm the EFISHG nature of the localized SHG signal. Coherent nonlinear microscopy imaging was also performed along the light propagation axis and modelled, so as to characterize the strong EFISHG signal and its spatial topology and orientation. Moreover, we report on the remarkably clear stability of the DLW-induced EFISHG signal with respect to thermal constraint below the glass transition temperature. These observations make the EFISHG behaviour a very attractive process for further perennial DLW for photonics applications.

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