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Direct Laser-Writing in silver-zinc doped phosphate glasses: correlated linear and nonlinear optical properties

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For the last decade, direct laser writing (DLW) has kept on providing impressive achievements in material processing, with a very broad range of both fundamental phenomena and subsequent applications. Among the numerous advantages of femtoseconde DLW, one can list its large versatility with respect to the irradiated material, its ability for both bulk and surface structuring, and direct experimental access to three-dimension laser-induced micro-scale modifications.

For the last years, we have been improving photosensitive glasses as tailored silver containing phosphate glasses [1], and their interaction with femtosecond infrared lasers [2, 3]. We designed laser/glass interactions to engineer laser-induced modifications of both linear and nonlinear optical responses. These modifications include absorption and fluorescence excitation and emission properties [4], but also localized nonlinear chi^3 enhancement leading to efficient third harmonic generation [5].

Recently, we reported DLW of localized second-order nonlinear response, leading to second harmonic generation in our initially centro-symmetric phosphate glass [6]. Such observation resulted from laser-induced space charge separation, at the root of a strong remnant buried static electric field whose amplitude up to $\sim 10^9$ V/m is similar to what reported elsewhere from thermal poling of glasses [7].

In this invited talk, we will provide an overview about results obtained with DLW in our tailored silver-containing phosphate glasses. We will specifically show recent progress about electric field induced second harmonic generation (EFISHG), by describing both original spatial distributions as well as detailed polarization analysis to prove the EFISHG behavior [8]. Moreover, we will present correlative microscope measurements, including both the nonlinear EFISHG signal and the linear fluorescence response, each property originating from space charge separation and from silver cluster production, respectively. Such correlative study provides new interpretation of the fluorescent silver cluster formation and stabilization since we observed that such fluorescent species appear to be a subsequent consequence of the laser-induced static electric field [8]. Silver clusters are shown to appear where DLW leads such species to be stable in adequate reduction/oxidation conditions, as seen from the correlated spatial distributions of EFISHG and fluorescence.

We will thus conclude with our new scheme of interpretation, for the understanding of the formation, stabilization and localization of the fluorescent silver clusters. Finally, DLW with our tailored glasses makes these materials promising for many applications as perennial high density 3D data storage or as integrated elementary bricks for future nonlinear photonics devices.

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