Multi-scale architected materials: from vortex-induced to dual-color direct laser writing in silver-containing phosphate glasses

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Abstract:

Phosphate glasses provide a very good solubility for silver ions, leading to an enhanced photosensitivity that allows highly interesting behaviors under femtosecond direct laser writing (DLW). In particular, laser structuring such glasses leads to strong and 3D localized optical contrasts of both linear and nonlinear optical properties, such as the fluorescence emission from silver clusters or the enhanced third-harmonic generation [1]. More recently, we have also demonstrated a strong and perennial second-harmonic generation in such glasses, originating from a laser-induced intense static electric field (up to $10^8$-$9$ V/m) buried inside the glass bulk. We reported such an observation as fs single-beam direct laser poling, whose laser-induced electro-optic coupling process leads to a significant effective second-order nonlinearity (up to 0.6 pm/V), opening the way to nonlinear photonic devices [2].

In order to better understand the mechanisms at play during DLW, but also to enlarge our abilities of material structuring, we have applied additional external constraints either simultaneously or successively to the laser writing process. Namely, we have performed dual-color DLW, which has led to the promising feedback control of the laser writing efficiency for STED-like approaches for sub-diffraction 3D laser patterning. Indeed, we have obtained up to 100% inhibition of laser writing of fluorescent silver clusters [3]. Additionally, thermal influence on laser structuring is also presented [4], to discuss the tailoring of the mechanisms at play during DLW in our silver-containing phosphate glass. Finally, we have also reported linear and nonlinear optical patterning in these photosensitive glasses with femtosecond structured light, corresponding to vortex-induced DLW. These findings provide original approaches to control the laser/matter interaction [5] in bulk and
fiber materials, for future laser-manufactured photonic applications such as composite metal-dielectric materials with plasmonic properties or artificial materials.

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**Keywords:** three-dimensional laser writing, fluorescent silver cluster, metal-dielectric composites, space-selective nanoparticles growth, super resolution microscopy, optical vortex, structured light.