FEMTOSECOND LASER STRUCTURING OF GLASSES

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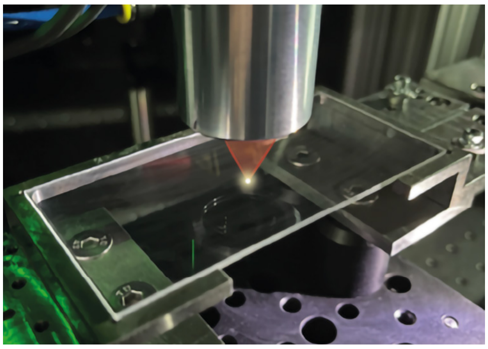

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Femtosecond lasers enable precise 3D nano- and micro-structuring in glasses, transforming photonic devices fabrication. The process relies on highly nonlinear interactions between femtosecond laser pulses and transparent materials, resulting in localized modifications of optical properties. In this article, we explore the fundamental mechanisms of laser-glass interactions, highlight advanced architectures for photonic integrated circuits, and discuss ongoing research aimed at expanding the range of achievable optical functionalities through innovative laser processing techniques and novel glass compositions.

Laser material processing has revolutionized modern manufacturing and materials science, offering precision and versatility across a vast array of applications. From cutting and polishing to surface texturing and bulk structuring, lasers, spanning continuous-wave and pulsed regimes, and wavelengths from ultraviolet to mid-infrared, enable the tailored modification of diverse materials, including metals, crystals, polymers, and glasses. Among these, femtosecond lasers have emerged as a tool of choice, particularly for the three-dimensional (3D) nano- and micro-structuring of glasses. This capability is crucial for photonics, where femtosecond laser writing facilitates the fabrication of advanced photonic integrated circuits (PICs) for quantum optics, ultracompact optical devices, archival data storage solutions with unprecedented longevity and density, and even fiber Bragg gratings (FBGs) when optical fibers are considered.

This article explores the fundamental physicochemical mechanisms underpinning femtosecond laser-induced changes in glasses, with a focus on the 3D-localized controlled modifications of optical properties. We then examine how these fundamental building blocks enable the realization of advanced photonic components and systems, bridging the gap between material science and functional devices. Finally, we highlight ongoing research efforts that aim at developing novel optical functionalities through innovative processing approaches and novel glass compositions.

FROM LASER-GLASS INTERACTION TO OPTICAL PROPERTIES

The interaction of femtosecond laser pulses with transparent materials, such as glasses, is governed by highly nonlinear processes occurring at a (sub)micrometer scale at the vicinity of the focal volume [1], as illustrated in Figure 1. In the single-pulse regime, nonlinear absorption, through quasi-instantaneous (~ fs) multiphoton absorption and tunnel ionization followed by cascade effects like avalanche ionization (~100 fs), generates a dense free electron plasma within the focal volume. Achieving the critical plasma density (~10^{21} cm^{-3}) necessary for optical breakdown typically requires intensities on the order of 10 TW·cm^{-3}, corresponding to a 100 nJ, 100 fs near infrared pulse tightly focused to a micrometer-scale volume. The ultrafast timescale of the laser pulse decouples the initial energy deposition from subsequent lattice heating, as electron-phonon coupling transfers energy to the lattice at the picosecond timescale. As a result, the energy is rapidly confined to a highly localized region, leading to extreme temperature increase (up to several 1000 °C). The relaxation of thermal stress within 0.1–1 ns generates shock waves with pressures ranging from 0.1 to 10 GPa, while thermal diffusion and re-solidification of the molten glass occur on microsecond timescales [2].

In the multiple-pulse regime, the repetition rate plays a critical role in determining the material response. At low repetition rates (typically <100 kHz), where the pulse separation exceeds the thermal diffusion time, each pulse interacts with a cooled modified material, potentially leading to incubation effects that might lower the modification threshold. Conversely, at high repetition rates (> 100 kHz), cumulative heating induces out-of-focus thermal modifications, triggering complex and rich thermodynamical and viscoelastic processes. These include melting, temperature gradient-driven material redistribution, and rapid quenching, which can result in densification, mechanical stresses, the formation of defects and colored centers, elemental migration and local phase transition.

The resulting bulk modifications in glasses span a wide spectrum of 3D-localized microstructural changes leading to a broad variety of optical contrasts, depending on the irradiation conditions and glass compositions. These range from low and smooth refractive index modifications to anisotropic birefringent nanograting structures, and highly scattering micro-voids.

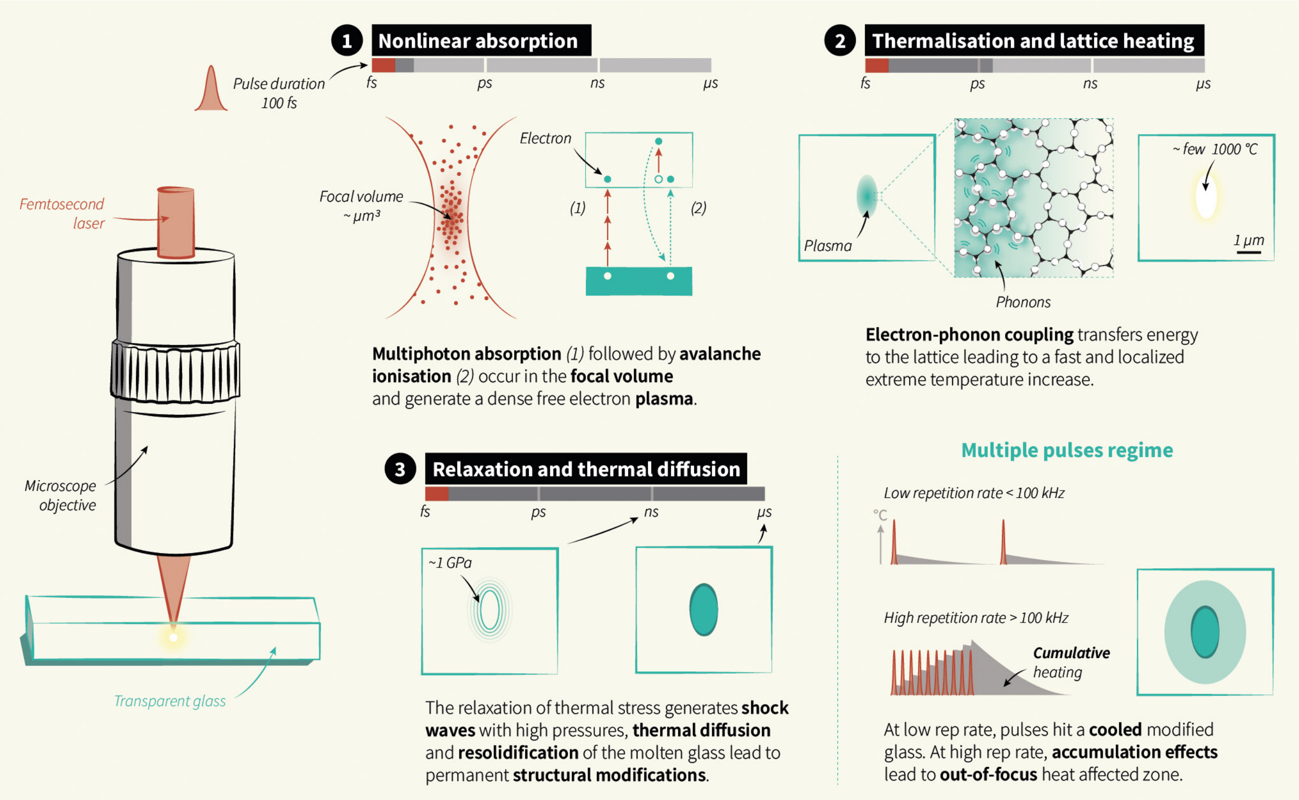
Laser-induced formation of new chemical species, space-selective crystallization, nanopores or dielectric/metallic nanoparticles growth further diversify the achievable optical properties that extend beyond linear and passive responses, including, for example, enhanced luminescence, second harmonic generation, magneto-optical responses or giant optical rotation, to cite a few.

ADVANCED PHOTONIC ARCHITECTURES

Glass nano-micro-modifications can be distributed in 3D either by moving the sample or by scanning the beam focus position allowing for the versatile prototyping of embedded multi-scale and multi-properties photonic architectures for innovative bulk optics elements, on-chip photonic integrated circuits (PICs) or fibered components (see Fig. 2).

For example, laser structuring of homogeneous smooth refractive index changes has been widely ● ● ●

Figure 1. Illustration of the laser-glass interaction mechanisms with typical timescales.



Multiphoton absorption *(1)* followed by **avalanche ionisation** *(2)* occur in the **focal volume** and generate a dense free electron **plasma**.

Electron-phonon coupling transfers energy to the lattice leading to a fast and localized extreme temperature increase.

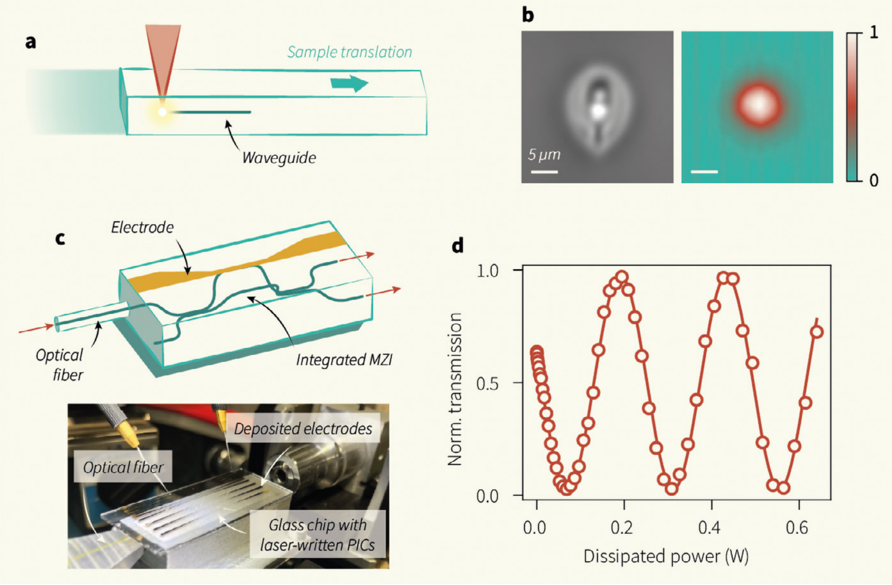
The relaxation of thermal stress generates **shock waves** with high pressures, **thermal diffusion** and **resolidification** of the molten glass lead to permanent **structural modifications**.

At low rep rate, pulses hit a **cooled** modified glass. At high rep rate, **accumulation effects** lead to **out-of-focus** heat affected zone.

used for the fabrication of low-loss (down to 0.03 dB/cm in commercial borosilicate glass and optical fibers [3]) waveguide-based PICs mainly operating at telecom wavelength with various geometries such as splitters, Y-junctions, cascaded directional couplers for (de-) multiplexing, interferometers and complex arrangement of coupled waveguides. Near-surface waveguides can also be inscribed for evanescent sensing of the environment through absorption, dephasing and/or changes in modal coupling. As illustrated in Fig. 2(c-d), realization of dynamically controllable integrated interferometers embedding active thermal phase shifting now gives access to advanced reprogrammable light management both in the classical and quantum light regime. In the latter context, the great versatility of the direct-laser-written PICs allows for the realization of hybrid multi-material quantum photonic platforms where, for example, the nonlinear generation of entangled photon pairs occurs in a periodically-poled lithium niobate crystal while the advanced complex photonic circuitry takes place in laser-inscribed glass. Complex arrangement of waveguides with tailored couplings also provides a powerful photonic platform to mimic condensed matter configurations and has been pivotal in the topological photonics community. Moreover, advanced waveguide-based architectures are also expected to play a crucial role in astrophotonics, providing ultra-stable and miniaturized devices to be implemented in telescope flux-recombining interferometric systems (see *e.g.* [4] for a review on PICs applications).

Beyond optical waveguides fabrication, femtosecond laser inscription also allows for the realization of sub-wavelength periodic refractive index modulation for the inscription of Bragg gratings in waveguides (WBGs) [5], see Fig. 3a, or in the core of optical fibers (FBGs). The resulting high spectrally-selective reflectivity can be finely tuned by controlling the grating properties (apodization, linear period chirps or even adapted phase steps). WBGs and FBGs can then be used for filtering and high-precision remote sensing applications as well as for manufacturing optical resonators in passive and active material for bulk and mostly fiber lasers devices. Laser-written periodic media also give access to the management of group velocity and slow light topics with light velocities down to a few tens of km/s.

Figure 2. Waveguide-based photonic integrated circuits (PICs). a) Sketch of the writing setup for PICs fabrication. b) White light microscope image of a single waveguide (left) and the corresponding mode at 1.55 µm. c) Sketch and actual picture of an integrated Mach-Zehnder interferometer (MZI). The dephasing is dynamically controlled with an electrode via thermo-optical effects. The normalized transmission of an MZI is plotted in d) as a function of the power dissipated by the electrode. The markers and solid line correspond to the data and fit respectively (Credits: A. Haykal, L. Bellando, I. Mouely Mouloungui, L. Raju Kalathil, L. Labonté, M. Bellec).



EXTENDING THE ENGINEER'S TOOLBOX FOR 3D OPTICAL FUNCTIONS IN GLASS

Direct-laser-writing in bulk glass is increasingly used to engineer 3D photonic microstructures that exhibit optical responses extending far beyond simple scalar refractive index modulation and conventional telecom-band applications.

Among the above-mentioned various optical contrasts offered by femtosecond laser structuring, tensorial optical responses (birefringence, fast-axis orientation, polarization-dependent scattering) are gaining strong interest. Such responses typically appear in silica when laser irradiation produces self-organized sub-wavelength structures (so called nanogratings) providing an effective uniaxial medium whose optical axis is linked to the writing laser polarization. In-volume retarders, polarization converters, and multiplexed high-dimensional optical data storage (3D positions, retardance and axis orientation) have been realized, the latter now being close to industrialization [6]. More recently, ultralow-loss birefringent regime has been identified in silica, where birefringence arises from randomly distributed elongated nanopores rather than regular nanogratings [7]. This novel microstructure type is attractive for transmissive devices requiring strong polarization control with reduced scattering, including bulk geometric-phase elements, based on spatially patterned optical-axis orientation, such as lenses, gratings, beam shapers, or multi-plane light converters.

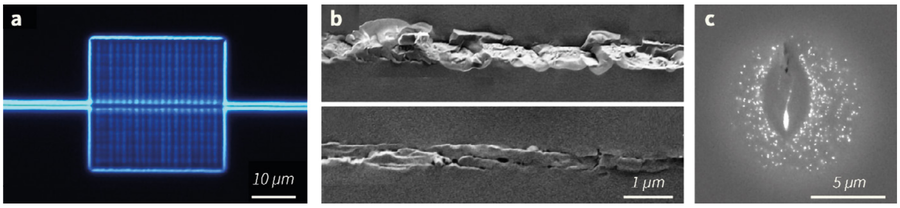


Figure 3. Examples of advanced laser-written nano-microstuctures. a) Waveguide Bragg grating written in a silver doped phosphate glass with 500 nm period [adapted from L. Loi *et al.* Opt. Mat. Exp. 14, 1837 (2024)]. b) Microstructures with chiral optical properties inscribed in silica glass with a polarized Bessel laser beam [adapted from J. Lu *et al.* APL Photonics 8, 060801 (2023)]. c) Laser-induced dissolution of dielectric nanoparticles embedded in the core of a scattering silica optical fiber (Credits: F. Orange, F. Pellerin, W. Blanc, M. Bellec).

A key perspective is to broaden the range of written properties beyond linear birefringence which includes linear and circular dichroism and other circular effects (optical activity, circular birefringence). Advanced anisotropy does not necessarily require explicit laser nano-structuring only and a promising route is to exploit the residual stress field as a design parameter: controlling the spatial distribution of stress can generate and orientate birefringence and more complex circular properties indirectly. Progress along these directions relies on spatio-temporal engineering of the writing laser field. Transversal and longitudinal spatial shaping (elongated foci, Bessel beams, vortex beams [8]) offers additional degrees of freedom by tailoring energy deposition along propagation (Fig. 3b shows an example of such laser-induced microstructures with chiral optical properties). Temporal structuring (laser pulse, pulse trains, and newly coming GHz burst regimes) provides further control over absorption dynamics, heat accumulation, and stress build-up, which collectively determines anisotropy, losses, and repeatability. Looking further ahead, emerging research highlights the potential for achieving sub-wavelength modification limits (few nm to few tens of nm). At this scale, an open question is whether laser-driven processes can be engineered to impose controlled optical anisotropy or chirality through structural rearrangements approaching interatomic-bond length scales, shifting the paradigm from writing shapes to writing material properties at the most fundamental level.

Besides laser writing processes, material science and glass composition engineering are of equal importance. While silica and borosilicate glasses remain the materials of choice for most visible and near-infrared applications, their performance is inherently limited by intrinsic absorption bands, particularly in extended spectral ranges. As the demand for advanced photonic functionalities grows, there is increasing interest in exploring alternative glass compositions, including soft-glass systems such as fluorides, heavy-metal oxides, and chalcogenides. However, their distinct thermomechanical behaviors and photoinduced responses present significant challenges for femtosecond laser structuring. Developing novel processing strategies is essential to achieve high reproducibility, low optical loss, and long-term stability of engineered optical properties.

CONCLUSION

Femtosecond laser processing has revolutionized the fabrication of 3D photonic nano-microstructures in glasses, enabling precise control over various linear, nonlinear, passive or active optical properties for advanced applications in photonics. Laser inscription is evolving to integrate subtractive and additive techniques. By inducing defects and stress, it enables laser-assisted etching for precise 3D microstructures, such as microfluidic channels and optical cavities, which can be infiltrated with active materials to incorporate functionalities in the bulk of robust glass host matrices. Additionally, laser-driven polymerization allows the fabrication of complex 3D architectures, including metamaterials and interconnects, enhancing photonic integration. Within this flourishing context, next generation of photonic platforms embedding sources, light transport, sensing and signal processing will certainly incorporate multiple functionalities obtained by combining various manufacturing abilities. ●

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