

Femtosecond Laser-Induced Bulk Modification of Transparent Materials

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ABSTRACT: Transparent materials, such as fused silica, sapphire, and lithium niobate, are widely used as windows and substrates in advanced optical and laser systems because of their high transparency, low optical loss, and excellent thermal and chemical stability. Consequently, precise bulk modification of these materials is essential for fabricating advanced internal photonic devices. However, conventional processing methods are often limited to surface processing, involve complex procedures, and lack the capability for precise three-dimensional (3D) internal structuring of the material. In contrast, femtosecond (fs) laser-induced bulk modification enables highly localized energy deposition inside transparent materials through nonlinear absorption, thereby allowing refractive-index modification and true 3D structuring with minimal impact on the outer surface. Consequently, researchers can fabricate various functional internal structures, including buried waveguides, nanogratings (NGs), and nanovoids, within transparent materials. This paper summarizes the recent progress in fs laser-induced bulk modification of transparent materials. It focuses on the underlying bulk-modification mechanisms and formation of internal structures. Finally, this paper discusses representative applications of these structures in integrated photonics, high-density optical data storage, and sensing in harsh environments.

1. INTRODUCTION

Transparent materials, such as fused silica, sapphire, and lithium niobate, are indispensable in advanced optical systems because of their high transparency and excellent mechanical, chemical, and thermal stability. With the rapid development of advanced photonics and optical physics, attention has increasingly focused on directly fabricating functional structures within these materials [1, 2]. In particular, the ability to construct 3D structures within the bulk is becoming increasingly important for next-generation photonic devices [3, 4]. However, conventional techniques, including lithography [5–7], ion implantation [8], and mechanical machining [9], are largely limited to surface or near-surface processing and are therefore not well suited for true internal structuring.

Laser processing offers non-contact, high-precision, and flexible fabrication with wider material applicability [10, 11]. Among these techniques, fs laser processing is particularly effective for localized internal modification and 3D structuring in transparent materials because of its ultrashort pulse duration and high-peak intensity [12, 13]. It can use ultrafast nonlinear absorption processes, such as multiphoton absorption and tunnel ionization, to generate initial free carriers, followed by avalanche ionization that further increases carrier density and confines the energy within a highly focused local volume [14]. Therefore, fs laser irradiation can induce localized structural and optical changes in transparent materials [15–17]. Conse-

quently, buried waveguides, NGs, and nanovoids have been fabricated in a broad range of transparent materials [18, 19].

As shown in Figure 1, this review summarizes recent progress in underlying modification mechanisms and representative internal structures enabled by fs laser-induced bulk modification. Particular attention focuses on buried waveguides and NGs, and their applications in integrated photonics, high-density optical data storage, and sensing in harsh environments.

2. BASIC PRINCIPLES

2.1. Nonlinear Absorption Mechanism

Using a high numerical aperture objective lens, an fs laser can form a highly localized, focused field within transparent materials. The instantaneous intensity in the focal volume can reach 10^{13} – 10^{15} W/cm² [20]. Because the energy of an individual photon is typically less than the bandgap of these materials, single-photon absorption is negligible. Nevertheless, the high peak intensity provided by the fs pulse is still capable of energy deposition into transparent materials through nonlinear absorption process. At the initial stage of ionization, multiphoton absorption and tunneling ionization are dominant processes. In the former process, valence band electrons simultaneously absorb multiple photons and are promoted to the conduction band through virtual intermediate states. In the latter, which becomes significant at higher electric field strengths, the intense laser field strongly distorts the atomic potential barrier, allowing the bound electrons to tunnel directly into the conduction

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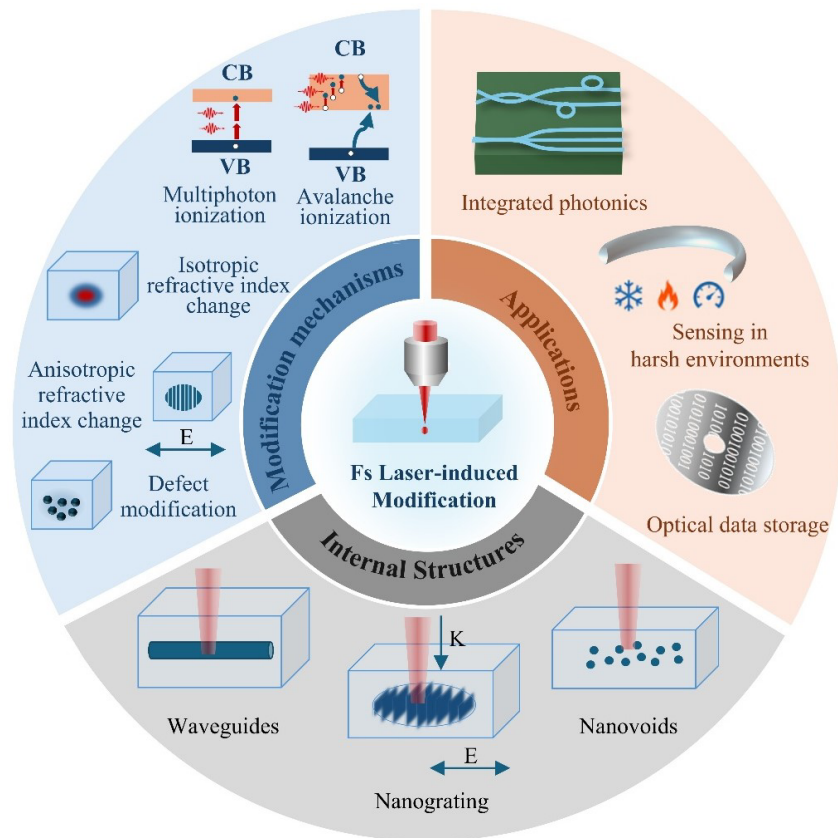


FIGURE 1. Overview of fs laser-induced bulk modification of transparent materials.

band. Both mechanisms generate free electrons in the focal region. The initially generated electrons gain energy mainly through inverse bremsstrahlung. Collisions then transfer part of this energy to the lattice and bound electrons. If they acquire sufficient kinetic energy, they can collide and ionize additional valence-band electrons into the conduction band, triggering a cascade increase in electron density. This phenomenon is called avalanche ionization. As this process continues, the free electron density within the focus area increases rapidly, eventually leading to forming a localized electron-ion plasma. After the fs laser pulse ends, the energy of the excited electron system is transferred to the lattice via electron-phonon coupling on timescales ranging from tens of femtoseconds to picoseconds, thereby inducing structural changes. Because the energy deposition of fs lasers is extremely rapid, electron excitation and thermal diffusion are effectively separated in time [21]. Consequently, the heat-affected zone is typically much smaller than that produced by picosecond or nanosecond lasers, enabling highly localized modifications while minimizing damage to the surrounding material and surface.

2.2. Local Energy Deposition and Modification Regimes

The processes discussed above determine how laser energy is coupled into transparent materials, and the resulting material response and internal modification states are governed by the level of local energy deposition. Once the fs laser energy slightly exceeded the modification threshold, a local-

ized refractive-index variation occurred, with an index change. Such refractive-index modification represents a relatively mild modification regime and provides a physical basis for buried waveguide formation [22]. When the laser energy exceeds the modification threshold but does not reach the breakdown threshold, periodic NGs with birefringence may form under suitable conditions [23]. The formation of laser-induced NGs is generally considered through several stages, as shown in Figure 2. In the initial stage, numerous nanoscale voids were generated discretely. At this stage, the voids are irregular, and the modified region does not show clear periodicity. In the second stage, these nanovoids undergo self-organized growth and interconnect with an increasing number of incident pulses. Consequently, the nanograting structure gradually emerges, and periodic ordering begins to appear. In the final stage, when the number of irradiation pulses reached several hundred, well-defined periodic NGs became clearly visible and extended throughout the irradiated region [24]. At higher energy depositions, fs laser irradiation can enter a damage-related modification regime. When the peak intensity exceeds 10^{14} W/cm², fs laser pulses can generate a highly excited electron-ion plasma inside the material through nonlinear absorption. The irradiated center then experiences extreme temperature and pressure, which induce a localized micro-explosion and an associated shock wave. Consequently, the focal region expands outward, leaving a nanovoid or low-density region at the center surrounded by a densified shell. Previous research has demonstrated that high-energy single laser pulses can be used to fabri-

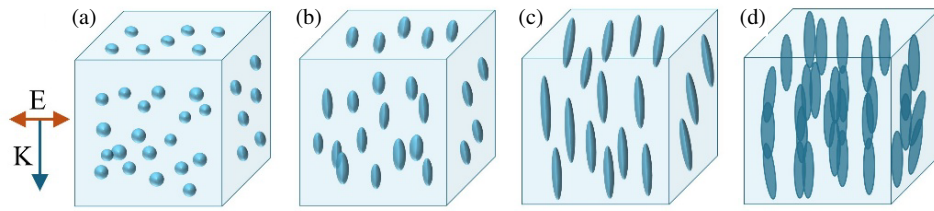


FIGURE 2. Schematic illustration of the stages of nanograting formation: (a) initial generation of dispersed nanovoids, (b) anisotropic growth, (c) interconnection of adjacent nanovoids, and (d) formation of well-defined periodic nanolayers.

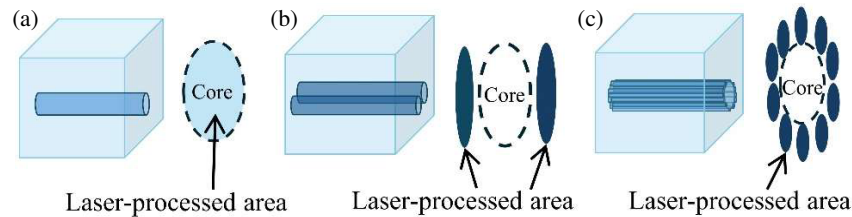


FIGURE 3. Schematic illustration of different waveguide types. (a) Single-line waveguides and their cross-section diagram. (b) Double-line waveguides and their cross-section diagram. (c) Depressed-cladding waveguides and their cross-section diagram.

cate nanovoids in the irradiated regions of transparent materials such as sapphire [25], glass [26], and siliconcarbide [27].

3. INTERNAL STRUCTURES

After discussing the interaction mechanisms between fs lasers and transparent materials and the role of local energy deposition in determining the internal modification states, this section focuses on the resulting internal structures. Refractive-index modification enables buried waveguide formation; anisotropic self-organized modification gives rise to NGs; and highly confined energy deposition can produce nanovoids through micro-explosion. The following sections discuss these structures according to their formation characteristics and their functional relevance

3.1. Waveguide

A waveguide is one of the representative structures produced by fs laser-induced bulk modifications. Davis et al. first demonstrated Fs-laser-written waveguides [28]. In this approach, controlled energy deposition changes the refractive index locally inside the material. The modified region can then guide light while the external surface remains largely unaffected. Depending on the material response and irradiation conditions, the induced refractive-index change can be positive or negative, resulting in different waveguide structures. This method has been widely used to fabricate optical waveguides in various transparent materials [29, 30].

To quantitatively describe the guiding capability of fs laser-written waveguides, the refractive-index change is often estimated under the step-index approximation as follows:

$$\Delta n = \frac{NA_{wg}^2}{2n}, \quad (1)$$

$$NA_{wg} = N_{air} \sin \theta \quad (2)$$

Here, n denotes the material's refractive index; NA_{wg} is the numerical aperture of the optical waveguide; N_{air} is the refractive index of air; and θ is the waveguide's far-field divergence angle [31].

The waveguide formation methods differ according to different types of refractive-index changes. As shown in Figure 3, there are three main types of waveguides fabricated by laser-induced modification: single-line, double-line, and depressed-cladding.

Single-line waveguides are based on Type I refractive index modification. Under focused fs laser irradiation, the refractive index increases at the focused spot ($\Delta n > 0$), while that of the surrounding unirradiated region remains unchanged. Therefore, the resulting laser-written track acts as an optical waveguide [32]. Because the waveguide is formed directly along the laser scanning path, single-line waveguides are particularly well suited for the direct writing of complex 3D waveguides. Type I modification is generally induced at relatively low pulse energies and produces a smooth, localized positive refractive-index change, making it particularly suitable for direct single-line writing. This type of waveguide is commonly fabricated using conventional glasses [33], fused silica [34], and nonlinear crystals [35]. When a Type II refractive index modification occurs inside the material, the focused fs laser induces a local decrease in the refractive index ($\Delta n < 0$). The resulting stress field subsequently leads to increasing the refractive index in the unexposed regions. Compared with Type I modification, Type II modification is typically generated at higher pulse energies and is commonly employed in crystalline materials in which a stable positive refractive-index change is difficult to obtain directly. Based on this principle, both double-line and depressed-cladding waveguides can be fabricated [36]. Double-line waveguides are formed by creating two lines of reduced refractive index within the material using an fs laser, with the light guiding region located between these two lines. In this

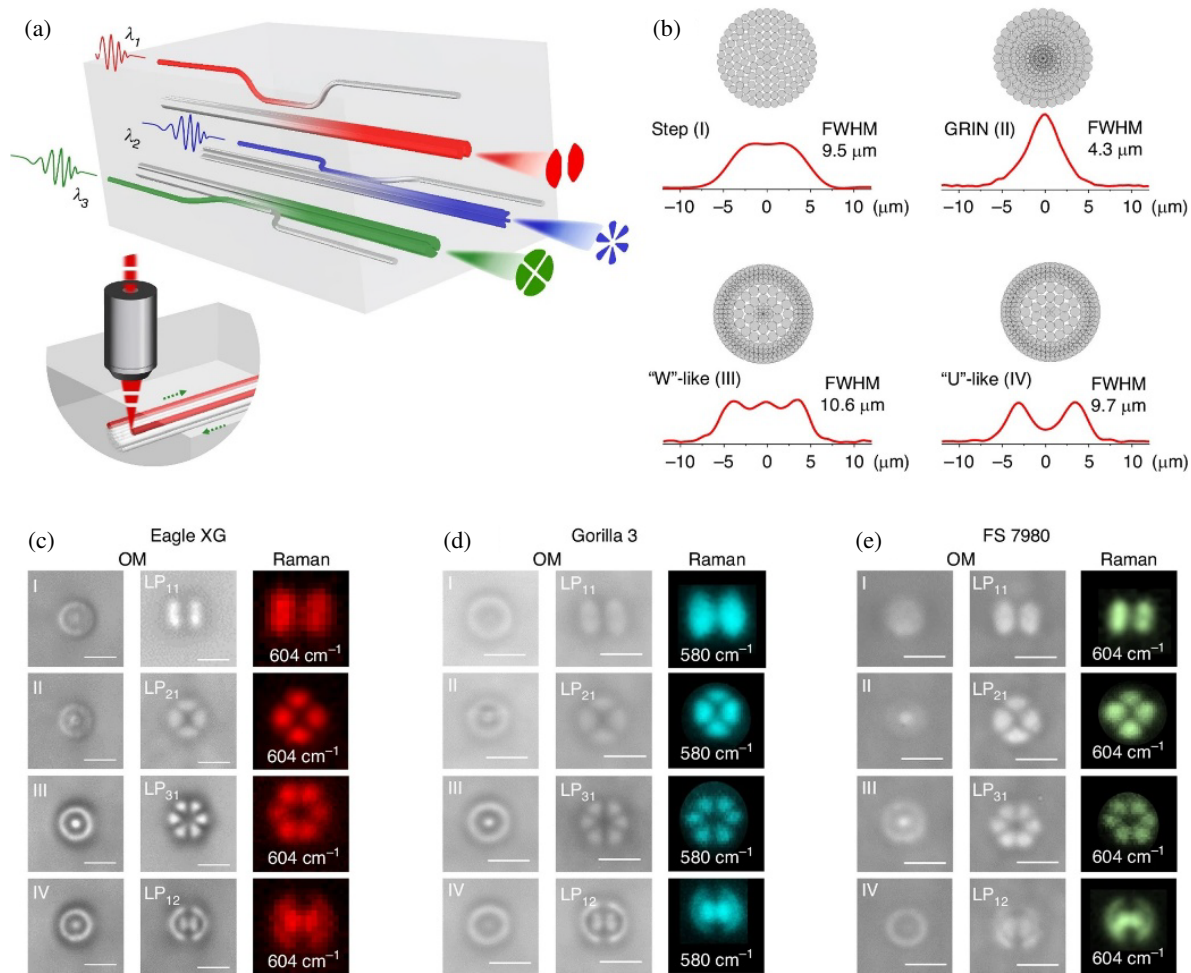


FIGURE 4. (a) Schematic showing ultrashort pulses with different central wavelengths (λ_1 , λ_2 , λ_3) being converted into higher-order spatial modes. The lower inset illustrates the fabrication concept of the OCMS method. (b) Scanline-overlap designs (top) and the corresponding measured refractive-index profiles (bottom) for four single-mode waveguide types. (c)–(e) Optical microscopy images of the four fundamental-mode waveguides (left), and the corresponding higher-order-mode waveguides supporting LP₁₁, LP₂₁, LP₃₁, and LP₁₂ modes (middle), together with Raman maps of the higher-order mode structures (right) in three representative glasses. Adapted with permission from [49] Copyright 2024, Springer Nature.

configuration, the guiding region exhibits significant birefringence owing to the compressive stress [37].

Depressed-cladding waveguides consist of low-index damage tracks surrounding an unmodified core, with flexible track arrangements that enable control over waveguide size and cross-sectional geometry. Because the guiding core is free of laser-induced refractive-index nonuniformity and defects, propagation loss is typically lower than that of single-line waveguides [38]. Moreover, they support both polarizations and can achieve polarization-independent single-mode guidance under optimized conditions, outperforming the double-line waveguides [39, 40]. In recent years, fs laser-fabricated waveguides have evolved from simple linear structures to complex 3D architectures and integrated photonic devices. Representative examples include curved, helical, and arbitrary 3D waveguides, Y-branch splitters, directional couplers, and ring resonators [41–47].

When evaluating the loss of complexly shaped waveguides, in addition to transmission loss, insertion loss and bending loss

must also be considered. With continuous optimization of laser direct-writing processing methods, bending loss can now be effectively reduced to a very low level. For example, Ross-Adams et al. [48] proposed an fs laser writing method based on a composite morphology. By optimizing the laser parameters, refractive index contrast was increased to 1.12×10^{-2} . Simultaneously, the cutoff bend radius for a bend loss of 1.0 dB cm^{-1} was reduced from 10 to approximately 4 mm. Furthermore, in 2024, Wang et al. [49] proposed the overlapping control multi-scan (OCMS) fs laser writing method, and the setup is shown in Figure 4(a). As shown in Figure 4(b), by precisely adjusting the spacing between adjacent scans to 50 nm, they achieved fine control over the refractive index and cross-section of the waveguide. Moreover, this approach can be extended to a range of glasses, as shown in Figures 4(c), (d), and (e). Based on this method, the authors fabricated low-loss waveguides with a maximum circular symmetry of approximately 99.5% and further realized a broadband, low-dispersion 3D mode-selective coupling structure.

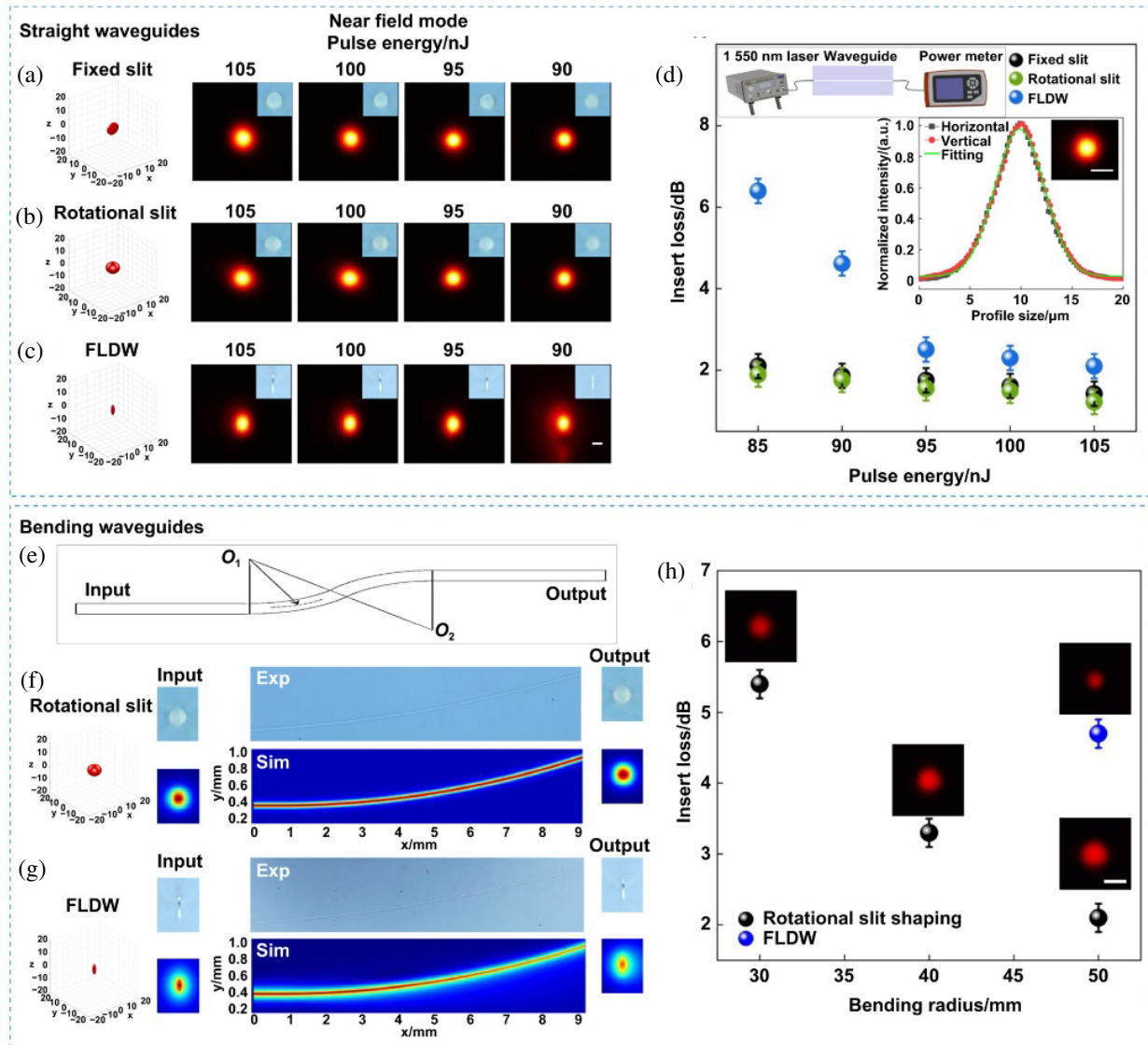


FIGURE 5. Fabrication and characterization of optical waveguides in Eagle glass. Straight waveguides fabricated by (a) fixed-slit beam shaping method, (b) rotating-slit beam shaping method, and (c) FLDW method. (d) Pulse energy-dependent waveguide insertion loss. The upper inset shows the setup, and the middle inset shows the measured mode-field distribution of the waveguide fabricated at 90 nJ. (e) Schematic layout of the curved waveguide design. Curved waveguides fabricated by (f) rotating-slit beam shaping method and (g) FLDW method. (h) Insertion loss of the curved waveguides as a function of bending radius, with the insets showing the corresponding output mode profiles. Scale bar: 10 μm . Adapted with permission from [54] Copyright 2026, IOP Publishing.

In addition to improving processing methods, the quality and efficiency of waveguide fabrication can be enhanced using fs laser beam-shaping technology [50]. During fs laser direct writing, refractive-index mismatch and nonlinear effects, such as self-focusing, often elongate the modified region longitudinally. Although transverse writing improves processing flexibility, this elongation is unfavorable for fabricating waveguides with circular cross-sections. Slit-based beam shaping effectively addresses this issue by tailoring the light field distribution [51]. Ams et al. [52] applied this technology to fs laser waveguide writing. By placing a 500 μm slit in front of the objective, they fabricated a single-line waveguide with a circular cross-section. It supported single-mode guidance at 635 nm and

showed a propagation loss as low as 0.39 dB cm^{-1} at 1550 nm. This technique enables the fabrication of various waveguides, including directional couplers for three-photon quantum interference on a waveguide platform [53].

Recently, Tan et al. [54] proposed a high-speed beam-shaping method based on a rotating slit. As shown in Figures 5(a), (b), (c), and (d), compared with the fixed-slit and conventional femtosecond laser direct writing (FLDW) approaches, this method produced more symmetric modified regions and near-field mode profiles, with nearly overlapping horizontal and vertical intensity distributions. When writing speed was set to 50 $\mu\text{m/s}$, the modified regions had circularities above 97% in two orthogonal cross-sections, and the

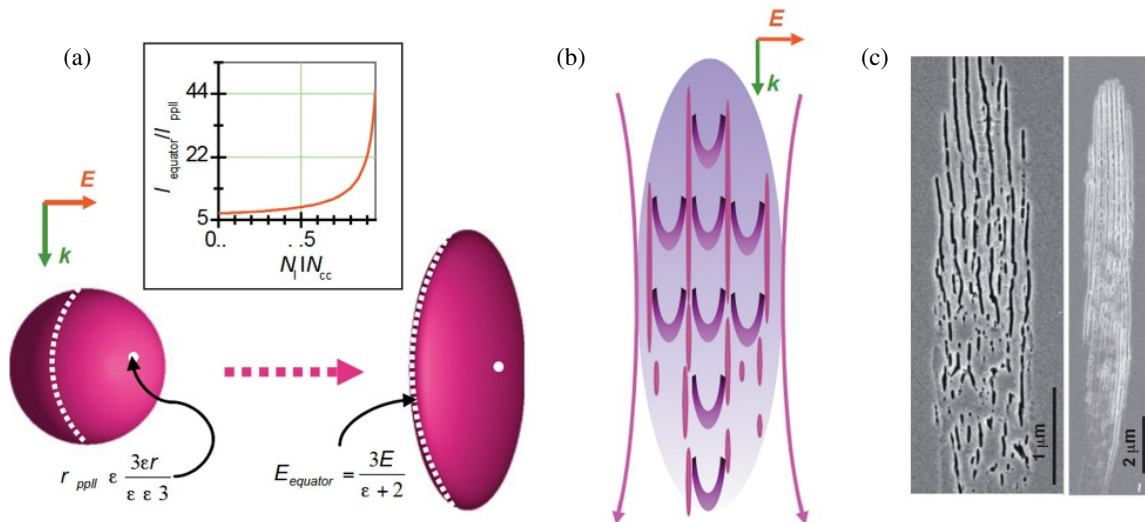


FIGURE 6. Schematic illustration of NG formation. (a) Diagram of asymmetric local field enhancement at two positions around a nanoplasma under the laser electric field, E . The relative permittivity ε is given by the ratio of the real part of the nanoplasma permittivity to the dielectric function of the surrounding medium. (b), (c) Schematic showing the evolution of periodic planar structures separated by $\lambda/2n$. Cross-sectional SEM images in the y - z plane. Adapted with permission from [58] Copyright 2008, John Wiley & Sons.

resulting straight waveguides showed insertion losses of only 1.3–2.0 dB. For the bent waveguides, Figures 5(e), (f), and (g) show the device geometry and corresponding experimental and simulated light propagation. As shown in Figure 5(h), at 100 $\mu\text{m/s}$, the rotational-slit waveguides exhibited insertion losses of 2.1, 3.3, and 5.4 dB at bending radii of 50, 40, and 30 mm, respectively, whereas a conventional FLDW waveguide already exhibited an insertion loss of 4.7 dB at 50 mm. These results indicate that rotating-slit beam shaping improves mode-field symmetry while reducing propagation loss, providing a more effective route to fabricating low-loss waveguides with improved mode symmetry.

3.2. NGs

When the deposited energy is above the modification threshold but below the breakdown threshold, repeated fs laser irradiation may induce birefringent NGs under suitable parameters. They are typically formed as laser-induced subwavelength periodic structures inside materials and exhibit significant anisotropies [55, 56]. Shimotsuma et al. first discovered NGs [57] in fused quartz in 2003 and have since become a research hotspot.

To explain how NGs form, a variety of mechanisms have been proposed, including plasmon-photon interference, asymmetric growth of nanoplasma, and exciton- or defect-assisted formation. Although plasmon-photon interference accounts for periodic energy deposition, the subsequent structural evolution is better described by the asymmetric nanoplasma growth model. Under ultrafast laser irradiation, intrinsic defects or color centers in transparent materials can act as hotspots for multiphoton ionization. Under repeated irradiation, these hotspots gradually develop into nearly spherical nanoplasmas via a feedback mechanism [58]. Local-field enhancement then becomes significant near the plasma boundary. So the

nano-plasmas grew asymmetrically and preferentially expanded perpendicularly to the laser polarization direction. Therefore, their morphology gradually changed from approximately spherical to ellipsoidal and eventually evolved into disc-shaped plasma structures, as shown in Figure 6(a). Driven by edge field enhancement, these disc-shaped structures can further merge to form nanoplanes. With continuous energy deposition, the electron density within the plasma region continues to increase and approaches the critical value, exhibiting instantaneous metallic properties. Subsequently, these nanoscale planes further interact with the incident light and eventually form periodic arrangements within the focal region, thereby evolving into NGs, as shown in Figures 6(b) and (c).

Apart from the formation mechanisms of NGs, early studies also focused on their morphological characterization, including several unique features, such as high selectivity etching rate [59], stress shearings [60], and nanoporous layers [61]. With the development of advanced characterization techniques, scientists have conducted increasingly in-depth research on these phenomena [62–66]. Shchedrina et al. [67] used scattering-type scanning near-field optical microscopy (s-SNOM) combined with synchrotron radiation-based nano-Fourier transform infrared spectroscopy (nano-FTIR) to directly confirm the presence of significant SiO_2 densification within the NGs at the nanoscale for the first time. Figure 7 presents the experimental procedure and observational data. Specifically, the authors performed s-SNOM imaging near 1130 cm^{-1} and used nano-FTIR to monitor the Si-O-Si band mode. The characteristic IR peaks in the irradiated regions shifted markedly to lower wavenumbers. It appeared at $\sim 1103\text{ cm}^{-1}$ in low-pulse-density modified areas and varied periodically across fully developed NGs, from $\sim 1095\text{ cm}^{-1}$ near the nanopore layers to $\sim 1112\text{ cm}^{-1}$ in the interlayer regions, with a period of 310 nm. Using the peak position density correlation, they concluded that NGs are not only

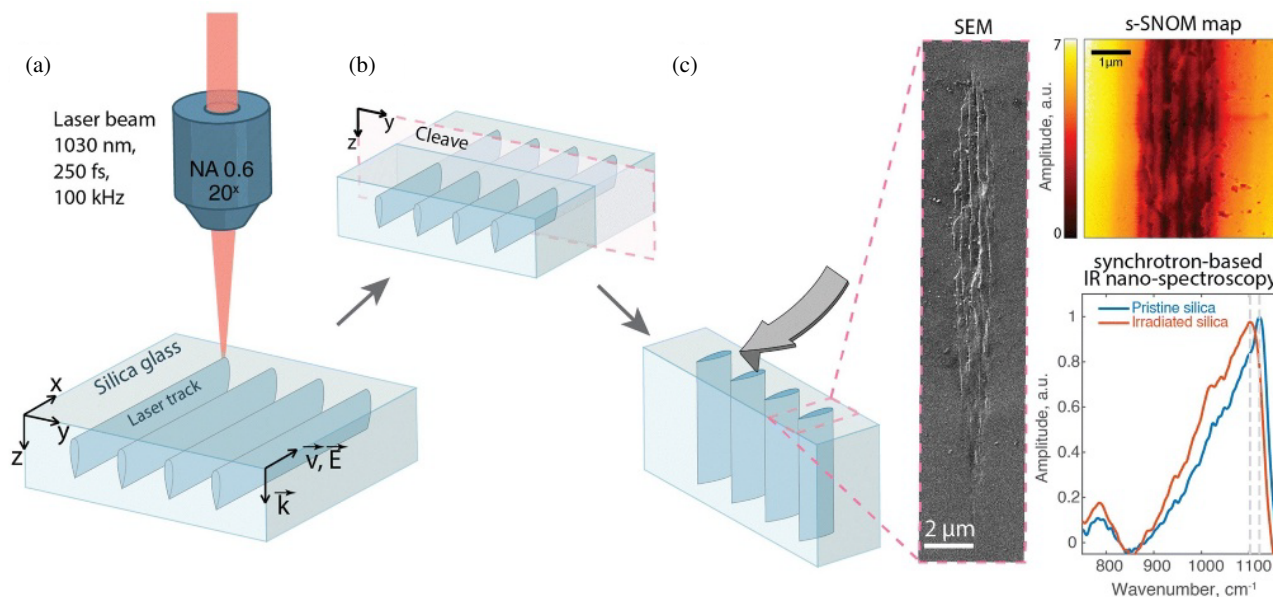


FIGURE 7. Fabrication and characterization of NGs in silica glass. (a) Schematic of the laser writing procedure, (b) sample cleaving for cross-sectional examination, and (c) Sample analysis by SEM, s-SNOM imaging, and nano-FTIR spectra. Adapted with permission from [67]. Copyright 2024, Royal Society of Chemistry (RSC).

periodic nanopore arrays but also composite structures formed by local densification.

Moreover, Xie et al. [68] further conducted direct NG characterization on a morphological scale. Using focused ion beam (FIB) sample preparation, along with scanning electron microscopy (SEM), scanning transmission electron microscopy (STEM), and high-resolution transmission electron microscopy (HR-TEM), they characterized the NGs from both cross-sectional and longitudinal perspectives. The results revealed that the NGs were not continuous planar fringes, but discontinuous nanolayers consisting of oblate ellipsoidal nanopores. These nanolayers have an average period of nearly 297 nm and a thickness of approximately 18 nm. Supported by the above observations, the authors proposed that under multi-pulse irradiation, the initially dispersed nanopores gradually grow, coalesce, and evolve into a more distinct nanolayer structure. This provides more intuitive structural evidence for NG formation and evolution.

With the deepening of related studies, NGs have been recognized as highly tunable nanostructures whose periodicity [69, 70], orientation [71], and spatial distribution [72] can be strongly modulated by laser parameters and writing strategies. Accordingly, research on NGs has gradually shifted from the initial observation of their structural features to more systematic exploration of their formation mechanisms and structural control. The polarization of the femtosecond laser determines NG arrangement, while the laser wavelength mainly determines the NG period [73]. As predicted by Bhardwaj et al. [74], the period is approximately $\lambda/2n - \lambda/n$. Here, n represents the refractive index of the glass, and λ is the laser wavelength. The writing direction is also an important influencing factor. Stankevič et al. [75] observed a larger period when scanning perpendicular to the electric field and a smaller period for parallel scanning. Increasing the pulse

energy also reduced the period. In addition to the periodicity of fully developed NGs, different anisotropic modification regimes can also emerge depending on the degree of structural organization. Type X modification is typically associated with randomly distributed, oriented oblate nanopores and exhibits relatively low optical loss, whereas Type II modification is characterized by self-organized periodic NGs with stronger form birefringence and optical scattering. Sakakura et al. [76] demonstrated that under low pulse numbers, fused silica can form a Type X modification. With further pulse accumulation, a rapid structural transition occurs from the low-loss Type X regime to the Type II NGs regime. Importantly, by precisely controlling the pulse number, low-loss birefringence modulation can be achieved before the full development of NGs. In this regime, Type X structures retain considerable retardance while delivering $\sim 99\%$ transmittance in the visible and $> 90\%$ transmittance in the ultraviolet region.

In addition to laser parameters and writing methods, the NG formation is closely related to structural characteristics of the transparent materials. Previous studies have shown that these fs laser-induced NGs can be fabricated in various glasses and crystals, such as doped silica glass [77, 78], sapphire [79], multicomponent aluminoborosilicate [80], and BaO-GeO₂-Ga₂O₃ (BGG) glasses [81].

3.3. Nanovoids

Nanovoids are typically formed under intense localized energy deposition, where strong nonlinear absorption and rapid energy release lead to irreversible structural disruptions within the material. In addition to nanovoids, this modification may also involve cavity-like regions, crack-associated zones, and porous structures.

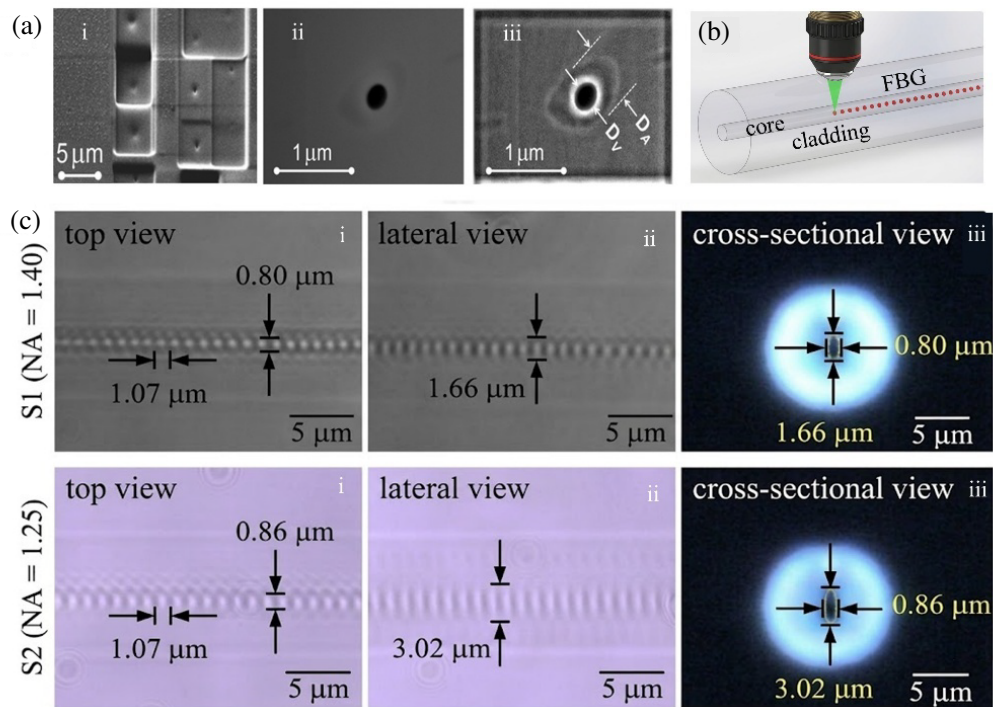


FIGURE 8. (a) SEM images of microexplosion-induced nanovoids inside sapphire revealed by FIB milling. i. SEM image of multiple voids generated by an SSTF beam. The incident pulse energy is 1.0 μJ. ii. Enlarged view of a representative void from panel (i), exposed at the optimal FIB depth. iii. Digitally enhanced image of panel (ii), showing the void (DV $\approx$ 0.3 μm) and the surrounding affected region (DA $\approx$ 0.7 μm). Adapted with permission from [85] copyright 2023, Optica Publishing Group. (b) Point-to-point fabrication of FBG gratings using an fs laser. (c) Grating morphologies fabricated using different objective lenses. i. Top-view, ii. side-view, and iii. cross-sectional microscope images of the FBGs written with different objective NAs. Adapted with permission from [93] Copyright 2022, MDPI.

Early studies of fs laser-induced nanovoids established that these features were not simply random damage but distinct internal structures that could be directly generated and manipulated inside transparent materials. In silica glass, laser-induced voids were shown to translate and merge under subsequent irradiation, and SEM observations confirmed that the optically observed dark spots were true cavities surrounded by a modified region [82]. This phenomenon has been later observed in other transparent materials [83, 84]. As shown in Figure 8(a), the formation and morphology of the nanovoids depended strongly on laser energy and focusing parameters. Nanovoids are formed only when the laser energy exceeds a threshold. Their morphology is determined mainly by the focal volume near the threshold but becomes increasingly conical at higher energies [85].

Moreover, beam shaping can tune nanovoid morphology [86]. Rapp et al. [87] used a single fs pulse Bessel beam to create nanovoids in sapphire with an aspect ratio as high as 100 : 1, which was two orders of magnitude larger than the nanovoids prepared using Gaussian beams. The laser energy used during processing was only in the range of nanojoules. At the same time, they also discovered that different laser parameters had completely different effects on the formation of nanovoids. In particular, the pulse energy and beam profile strongly influence nanovoid formation by determining the spatial distribution and density of deposited energy. Increasing the pulse energy will enlarge the void. The beam profile will

change the void geometry. For example, Gaussian beams and Bessel beams will produce significantly different void sizes and aspect ratios.

Periodic nanovoid arrays can also be produced in conventional borosilicate glass with a single fs laser beam. The voids self-organize along the propagation direction. Their period, number, and total array length can be adjusted via the laser power, pulse number, and focal position [88]. Such arrays can be used to generate periodic refractive-index modulation. This has also promoted the development of point-by-point writing strategies for structures such as fiber Bragg gratings (FBGs) [89–91]. Figure 8(b) illustrates the processing method. Sosa et al. [92] showed that the periodic nanovoid arrays are not simply damage, but functional structures that produce Bragg modulation. Chen et al. [93] further showed that the geometry of these nanovoids also affects FBG performance. As shown in Figure 8(c), the grating period under different numerical apertures remained approximately 1.07 μm, but the nanovoid shapes were different. When using a lens with a numerical aperture of 1.25, the nanovoid had a more obvious longitudinal extension. The larger longitudinal extent improved mode overlap and reduced insertion loss.

In addition, high-temperature stability also determines the working performance of FBGs. Sosa et al. [94] found that when the annealing temperature exceeded 1125°C, the nanovoids gradually deformed and lost their original periodicity. There-

fore, in the future, not only the morphology and periodicity of the nanovoids need to be optimized, but also their stability should be enhanced, so as to achieve better device performance.

4. APPLICATIONS

4.1. Optical Data Storage

Due to the rapid increase in data volume, conventional storage methods are facing increasing limitations in capacity, lifetime, and readout efficiency. In response to this demand, Glezer et al. first proposed the optical data storage relying on fs laser-induced localized modification [95] and later developed it into 3D optical storage across a range of transparent materials. However, this type of 3D storage is mainly based on nanovoids and relies largely on spatial position for encoding. Consequently, the number of available storage dimensions remains limited, and further improvement in data capacity is difficult [96].

To overcome this limitation, researchers have developed five-dimensional (5D) optical data storage based on fs laser-induced NGs, which provide additional encoding dimensions through their orientation and retardance, thereby increasing the storage density. Due to the increase in storage dimensions, it offers much higher storage capacity and greater flexibility than 3D storage. Shimotsuma et al. [97] first introduced 5D optical storage by tuning the polarization direction and phase distribution of the writing beam, achieving a storage density of 300 Gbit/cm³ in silica glass. Beresna et al. [98] further discussed the self-organization mechanism of laser-induced anisotropic nanostructures in glass, providing a physical basis for understanding the formation of birefringent structures used in multidimensional optical storage. In 2014, Zhang et al. [99] brought this strategy closer to practical application by demonstrating multilayer 5D storage in fused silica, with a projected lifetime exceeding 100 million years, a recording speed of up to 12 Mbit/s, and a capacity as high as 360 TB/disc.

Recent studies have focused more on writing speed, storage capacity, and practical usability. Ye et al. [100] achieved 5D storage in glass through a parallel writing method based on shaped voxels. The estimated data storage capacity was 1.47 TB, and the writing speed was increased to 7.5 MB/s. Moreover, the Project Silica team achieved a storage capacity of 4.84 TB in fused quartz by utilizing birefringence and phase voxels [101]. The writing speed can reach up to 25.6 Mbit/s, and the expected data lifespan exceeds 10,000 years. These results indicate that optical data storage is advancing rapidly and has the potential for practical applications.

4.2. Integrated and Quantum Photonics

Integrated photonics focuses on on-chip light transmission and integration. Quantum photonics focuses on quantum communication, sensing, and precision measurement. Although their applications differ, both require stable, low-loss, and flexibly designed optical structures.

Waveguides are an important structure in integrated photonics. It can be used for optical beam splitters [102], directional

couplers (DCs) [103], optical switches [104], and mode modulators. Beyond these simple in-chip applications, they can also serve as interfaces between chips and free space. Lapointe et al. [105] extended fs laser-written waveguides to nonreciprocal integrated photonic devices by inscribing low-loss waveguides in latched bismuth-doped iron garnet without degrading its magneto-optical response. The resulting Faraday rotator waveguide exhibited an insertion loss below 0.15 dB, and its integration with thin-film polarizers produced a compact optical isolator with an isolation ratio above 25 dB and an insertion loss below 1.5 dB across the telecom C-band. The device operates without external magnets or coupling lenses, facilitating direct hybrid integration with photonic circuits.

Furthermore, owing to their advantages such as 3D integration, high-fidelity transmission, and polarization controllability, optical waveguides have been widely employed in quantum photonic circuits and large-scale quantum networking [106]. Recently, Zhou et al. [107] fabricated a 3D waveguide lattice in glass and experimentally demonstrated three-photon quantum walks on a two-dimensional triangular lattice. The photonic chip mapped the evolution of three indistinguishable photons onto a $19 \times 19 \times 19$ Hilbert space containing 6859 nodes, demonstrating the capability of laser-written 3D circuits for large-scale multiparticle quantum simulations. In addition to waveguides, laser-induced NGs can also serve as polarization-sensitive or birefringent elements, providing complementary functionalities for polarization control and light manipulation in integrated photonic systems. Xu et al. [108] experimentally fabricated geometric-phase elements, including a polarization diffraction focusing lens and a Q-plate, based on laser-inscribed NGs in sapphire, demonstrating their capability for polarization and wavefront manipulation. These studies demonstrate that fs laser-written waveguides and nanogratings can provide complementary functionalities for integrated and quantum photonics, enabling both three-dimensional light routing and polarization-dependent optical manipulation.

4.3. Sensing in Harsh Environments

Internal structures fabricated in transparent materials are becoming increasingly important for sensing in harsh environments, as these materials can withstand high temperatures and intense radiation [109, 110]. Among these structures, NGs are particularly promising for sensors because of their unique anisotropic optical properties and excellent thermal stability. Wei et al. [111] demonstrated that the thermal stability of NGs not only is relevant to their microstructural characteristics, but also closely depends on the glass composition. Higher content of Cl⁻/OH⁻ doping significantly weakens the ability to maintain stability at high temperatures, while an appropriate amount of Ge doping has a relatively limited impact on thermal stability but does change its optical response. Beyond NGs, buried waveguide-based structures can also be employed for sensing in harsh environments because their internal guiding geometry provides optical confinement, together with protection from external damage. Wang et al. [112] constructed directional nanostructures in sapphire-derived all-glass cores using the FLDW method. They systematically studied the writ-

ing kinetics and thermal stability within the temperature range of 20–1200°C. The results demonstrate the significance of the material composition in terms of thermal stability. In addition to optimizing the material composition, the processing method can be optimized. Wang et al. [113] achieved this by optimizing the writing process, maintaining high-temperature and radiation resistance stability while minimizing losses. Eventually, they achieved an insertion loss near 1 dB m⁻¹, a single-point device loss of approximately 0.001 dB, with a temperature fluctuation of only $\pm 5.5^\circ\text{C}$ over 5 days at 1000°C, and prepared a 1.4 m long radiation-resistant distributed sensor that completed stable temperature measurements in the reactor environment. In conclusion, to enhance the environmental tolerance of the internal structure, it is necessary to simultaneously optimize the material composition and processing conditions.

5. CONCLUSIONS AND OUTLOOK

Laser-induced modification provides a direct way for fabricating internal structures in transparent materials. The thermal and chemical stability of transparent materials makes them suitable for optical and photonic applications. Conventional methods are less effective for fabricating structures inside the material. Fs laser-induced modification offers a unique route for 3D direct writing through nonlinear absorption and ultrafast energy deposition. This review summarizes underlying mechanisms, structures, and applications of fs laser-induced modification. It then reviews waveguides, NGs, and nanovoids and their applications in optical storage, integrated and quantum photonics, and harsh-environment sensing.

Several challenges still need to be addressed in this field. The physics of fs laser interaction with transparent materials is not yet fully understood. The modification process involves nonlinear excitation, free-electron generation, ultrafast energy relaxation, and lattice response, but links between these stages remain complex. This issue is particularly evident in the formation of NGs, nanovoids, and related internal structures.

Another challenge is the control and reproducibility of structure formation. In waveguide fabrication, refractive-index change, cross-sectional shape, mode matching, and propagation loss are closely coupled. This makes it difficult to obtain low loss and high symmetry while maintaining compatibility with complex 3D interconnections. For NGs, the period, orientation, and uniformity are highly sensitive to laser parameters, polarization state, and material properties, which limit their consistency. For nanovoids, issues such as size dispersion, stress concentration, and long-term reliability have not yet been fully addressed. Transparent materials differ markedly in their band structure, defect-state distribution, thermal diffusion behavior, and crystallographic anisotropy. Consequently, many existing conclusions remain material-specific, and broadly applicable principles for structural design and process control are still insufficient.

Furthermore, current research remains focused mainly on individual structures and single-function demonstrations. In comparison, the integrated fabrication of complex 3D functional systems remains relatively underdeveloped. The coordinated design and coupling of different modified units, such as

waveguides, NGs, and nanovoids, have not yet been explored to the same extent as the formation of isolated structures. In addition, the point-by-point nature of the FLDW method imposes constraints on the throughput, large-area fabrication, and batch-level consistency. These factors have become increasingly important as the field advances from the initial concept demonstration to practical device implementation.

Overall, fs laser-induced modification based on localized modification has established an important foundation for the fabrication of high-performance photonic structures in transparent materials. Future work should further clarify laser-material interaction mechanisms through a combination of time-resolved characterizations, in situ observations, and multiscale simulations. More precise processing strategies are also needed. Beam shaping, polarization control, adaptive focusing, and hybrid-assisted fabrication can improve structural controllability, uniformity, and processing efficiency. Future research should extend beyond individual structures toward complex 3D integrated systems. Such integration will also support the practical and large-scale implementation of these structures.

ACKNOWLEDGEMENT

This work was supported by the Opening Funding of the National Key Laboratory of Electromagnetic Space Security.

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