

Femtosecond laser-inscribed optical waveguides in dielectric crystals: a concise review and recent advances

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Abstract. Femtosecond laser inscription or writing has been recognized as a powerful technique to engineer various materials toward a number of applications. By efficient modification of refractive indices of dielectric crystals, optical waveguides with diverse configurations have been produced by femtosecond laser writing. The waveguiding properties depend not only on the parameters of the laser writing but also on the nature of the crystals. The mode profile tailoring and polarization engineering are realizable by selecting appropriate fabrication conditions. In addition, regardless of the complexity of crystal refractive index changes induced by ultrafast pulses, several three-dimensional geometries have been designed and implemented that are useful for the fabrication of laser-written photonic chips. Some intriguing devices, e.g., waveguide lasers, wavelength converters, and quantum memories, have been made, exhibiting potential for applications in various areas. Our work gives a concise review of the femtosecond laser-inscribed waveguides in dielectric crystals and focuses on the recent advances of this research area, including the fundamentals, fabrication, and selected photonic applications.

Keywords: femtosecond laser writing; femtosecond laser inscription; optical waveguides; dielectric crystals; laser crystals; nonlinear optical crystals; waveguide lasers; frequency/wavelength conversion; quantum photonic chip; quantum memories.

Received Oct. 10, 2021; revised manuscript received Jan. 27, 2022; accepted for publication Feb. 23, 2022; published online Mar. 29, 2022.

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[DOI: [10.1117/1.AP.4.2.024002](https://doi.org/10.1117/1.AP.4.2.024002)]

1 Introduction

Optical dielectric crystals play crucial roles in a large number of optical and photonic applications.^{1–3} For example, crystalline quartz is an ideal material for waveplates to modify the polarization state of light. Nonlinear optical crystals, such as KTiOPO_4 ,⁴ $\beta\text{-BBO}$,⁵ or LiNbO_3 ,⁶ are used to realize selected wavelength conversion in broadband covering visible to the mid-infrared (MIR). Laser crystals serve as gain media that are capable of emitting coherent laser radiation in various solid-state systems.⁷ As an emerging direction of intriguing applications of dielectric crystals, quantum photonic devices are also achievable based on specific crystal materials.^{8–15} Regardless of the attractive features of crystals, there is a strong desire to construct on-chip or three-dimensional (3D) devices to implement

multifunctions in compact systems.^{16–19} In photonics, such devices are based on structures of optical waveguides. The light propagation can be confined in waveguides in a diffraction-free way due to the total internal reflection, in which the optical intensity is able to reach a high level.^{20,21} With these advantages, the waveguide-based devices therefore have a compact size, robust functionality, high efficiency, and high-cost effectiveness.²² Optical waveguides based on crystals are attractive for a variety of applications, owing to the combination of advantages of both waveguiding structures and crystal bulk features.^{23,24} Toward this purpose, several techniques have been developed to fabricate waveguides in crystal bulks, including metal–ion indiffusion,²⁵ ion exchange,²⁶ optical induction,²⁷ ion implantation,²⁸ and femtosecond laser inscription/writing.^{29–31} However, due to the diversity of the crystal structures and physical, chemical, or optical properties, some techniques are only applicable to specific materials. Among these solutions, femtosecond laser

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writing has been recognized as an efficient tool to fabricate waveguides in versatile crystals owing to the powerful 3D engineering capability and microscale localized refractive index modification.^{23,32} The first scenario of dielectric waveguides was realized in glasses through femtosecond laser writing by Davis et al. in 1996.³³ Since then, quite extensive studies have been performed on this topic, and the materials used have also been extended to single crystals, ceramics, and organic materials.^{34–38} Various fabrication techniques of laser-written waveguides and related photonic applications have taken a considerable leap forward in the past two decades. For example, using structured light writing, the production of complex waveguides can be implemented in single-way scanning.^{39,40} The beam shaping of femtosecond laser pulses can form tracks along selected orientations inside the dielectrics.^{41,42} In addition to well-developed waveguide lasers⁷ and wavelength converters,⁴³ emerging applications in quantum photonics have been realized in laser-written waveguides, such as quantum chips^{44–49} and memories.^{50–53} There are several reviews demonstrating the research progress of the topic from different perspectives;^{9,23,24,31,34,35,37,54} however, since this is a hot topic with rapid development of technical solutions and applications, a timely concise review of the new results obtained in recent years on selected branches is desired, particularly on femtosecond laser-written crystal waveguides. In this work, we concentrate on the crystalline waveguides on the newly developed femtosecond laser inscription techniques, the diverse waveguide geometries, the engineering of waveguide modes, and selected applications by presenting an overview of the recent advances in this topic.

2 Modification Induced by Femtosecond Laser Processing of Dielectric Crystals

Focused femtosecond laser beams attain very high optical intensities. As such intense ultrafast beams irradiate the dielectrics at the focal plane inside the materials, strong-field ionization (multiphoton or tunnel ionization) occurs,^{55,56} which makes a large number of electrons break away from their original atoms. These free electrons continue interacting with the laser pulse, and more electrons are released by collisions, leading to avalanche ionization.⁵⁷ Consequently, a hot and dense electronic plasma forms.⁵⁸ If the free electrons reach a value close to the plasma density for the laser wavelength, breakdown occurs, resulting in ablation at the surface and fissures in the bulk,⁵⁹ modifying the irradiated lattices significantly. Since the interaction process happens on an ultrashort timescale, the thermal implication to the surrounding region of the focus can be very limited, particularly in regimes of low-repetition-rate pulses,⁶⁰ depending on the hot electrons coupling to the lattice and the heat relaxation, but can also modify the bulk properties.^{61,62} As a result, the femtosecond laser modification of the dielectrics is very localized, which enables “cold” micromachining of materials. The femtosecond laser modification may bring out some effects in dielectrics, such as refractive index changes,³³ stress,⁶³ birefringence,⁶⁴ nanogratings,^{65,66} voids,^{67,68} and thermal melting.⁶⁹ To promote permanent refractive index changes in the focal volumes, which are the base for waveguides, the pulse intensity has to be carefully controlled to avoid breakdown.^{70,71} Nevertheless, the processes of femtosecond laser modification are generally very complex and strongly rely on the nature of materials and parameters of laser pulses. The major parameters of laser pulses include wavelength,⁷² energy,⁷³ repetition rate,⁷⁴

focusing condition, scanning speed, and/or polarization.⁷⁵ Generally, for a specific material, it is required to be transparent at the selected laser wavelength, i.e., the linear optical absorption of the material at the used laser wavelength is minimal. Because the femtosecond laser relies on a nonlinear optical process, the strong linear absorption may bring out incubation effects and significant thermal effects (e.g., ablation at large-scale regions), which lead to modifications starting at the surface, preventing bulk machining.⁷⁶ For example, near-infrared lasers at 800 nm or 1 μm are used to process most insulating dielectrics, while for silicon the 1.5- μm laser works well for waveguide inscription.⁷⁷ In addition, waveguides have been successfully produced by green-laser writing in dielectrics with negligible absorption at the visible light band.^{78–80} In addition, low-repetition-rate ($\sim\text{kHz}$) lasers deposit less energy onto the lattice per unit length of the samples, which requires the relatively high energy of single pulses.^{74,81} High-repetition-rate (MHz) pulses induce thermal effects, and are often used for glass waveguide fabrication.⁸²

2.1 Type I and Type II Modifications

For dielectric crystals, waveguide formation depends on the efficient modification of the refractive index of the bulk. There are two major effects of refractive index changes, i.e., types I and II.⁶³ Type I refers to the laser-induced tracks with positive index changes ($\Delta n > 0$) that may be due to the slight structural change of lattices. Such variation may be correlated to the breaking of the symmetry of the irradiated lattices.^{83–85} To form type I modification, low-energy pulses are required. Recent investigation reveals that the type I modification may also be due to the phase transition of the laser-irradiated regions.^{86,87} Type II refers to laser created filaments in the material, which has negative index changes ($\Delta n < 0$) caused by localized crystalline lattice expansion in the focal volume.⁶³ The type II modification is typically correlated to highly altered lattices in which partial or complete amorphization occurs. For type I modifications, the track region is able to guide light because it has a higher refractive index than the surroundings, whereas for type II, the track has a lower index and cannot be used as a waveguide; however, the expansion of volume in the focal plane gives rise to compression of vicinity, leading to the refractive index increments in the surrounding of the track.⁶⁰ With these typical features, several configurations of waveguides have been developed based on types I and II modifications. Figure 1 shows the typical geometries of four basic laser-written waveguides: single-line, double-line, depressed-cladding, and optical-lattice-like structures. For $\Delta n > 0$ in the track, the single-line can be used to guide light since it naturally constructs a waveguide channel [Fig. 1(a)], and such structures are easy for 3D waveguide fabrication.^{88,89} However, in crystals, the single-line structure is usually polarization sensitive^{84,85} and does not possess good thermal stability.³² Another disadvantage is that the waveguide core lattice has been distorted by the laser irradiation and, for crystals, the bulk features are destroyed in the waveguide.⁹⁰ Based on type I modification, a multiscan technique can be employed for the fabrication of symmetrical waveguides in which several single-line tracks are overlapped transversal to the writing direction.⁹¹ Such a design is preferred to support waveguiding accessible to long wavelengths with low-loss propagation owing to a significant increase of the index contrast.⁸⁹ For $\Delta n < 0$, the track region has a low index in comparison with

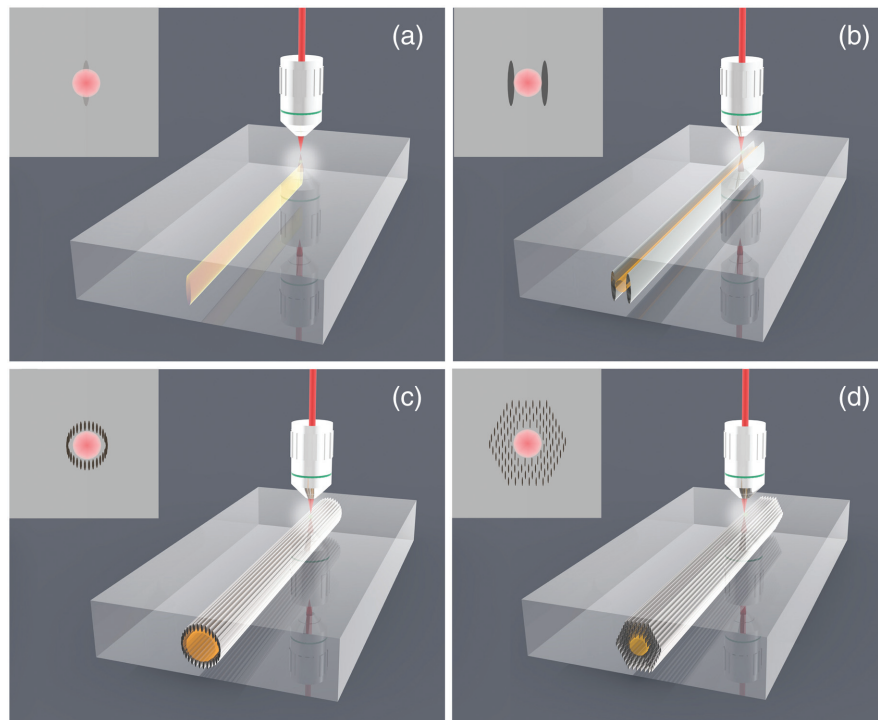


Fig. 1 Basic geometries of laser writing of waveguides: (a) single-line, (b) double-line, (c) depressed-cladding, and (d) optical-lattice-like configurations. The dark (gray) regions represent the laser-induced tracks.

the surrounds, and the vicinal zones have a relatively high index that may serve as guiding regions.⁹² In practice, to obtain a nearly symmetric waveguide core, the double-line structure is used: the guiding core is located at the region between two parallel tracks [Fig. 1(b)]. The waveguide core is not located at the tracks that have highly altered lattices. This means that the properties of the bulk crystals are well-preserved in the waveguides^{93,94} in which the laser-induced modifications of the original lattices are negligible.⁹⁵ In addition, the formed track has excellent thermal stability, enabling applications under high-power conditions.⁹⁶ There are shortcomings offered by the double-line waveguides, which cannot guide light at longer wavelengths since the dimension of the waveguide core is limited to $<20 \mu\text{m}$, and for 3D fabrication it requires a complicated design.⁹⁷ The depressed-cladding structure is a more flexible solution [Fig. 1(c)]. It is usually in the geometry of circles or rectangles,⁹⁸ which consists of a number of type II tracks. Since the diameters of cladding waveguides can be artificially determined, they are capable of guiding long-wavelength light, e.g., MIR regime.^{78,80,99} Nevertheless, for 3D waveguides, it also requires a special design. A special configuration is an “optical-lattice-like” structure,¹⁰⁰ which contains a periodically arranged array of tracks [Fig. 1(d)]. Some positions are free of any tracks, e.g., in the center of the structure, which can serve as beam propagation channels. By modulating the geometries of tracks, it is able to achieve beam transformation and shaping.^{101,102} Generally, the maximum Δn of type I modification is much lower than that of the type II modification induced by femtosecond laser irradiation.³² For instance, the typical index variation of type I waveguides in LiNbO_3 crystal does not go beyond 1×10^{-3} , whereas this value will increase to (2 to 5) $\times 10^{-3}$ at directly irradiated tracks for type II-based structures.^{23,63} For

$\text{Nd}:\text{YAG}$ double-line waveguides, Δn as high as 9×10^{-2} at type II filaments has been determined according to μ -Raman and μ -photoluminescence microscopic analysis.⁹² Most recently, quantitative phase microscopy became a popular method to recover the index contrast of the waveguides from the phase image. In particular, the index change of 2.5×10^{-3} has been maintained after heating up to 1400°C for a depressed cladding waveguide inscribed in a sapphire crystal that exhibited 4.75×10^{-3} prior to the annealing process.⁸⁰ Table 1 summarizes the advantages and disadvantages of different configurations in transparent solid materials (crystal, glass, polymer, ceramic, etc).

One could realize the evolution of laser-written tracks from type I to type II via manipulating the laser inscription parameters. For instance, in LiTaO_3 crystal, with the low-repetition rate of femtosecond laser pulses (25 kHz), the tracks can be changed from type I to type II. Figure 2(a) shows the influence of incident pulse energy on the geometry of tracks at a fixed laser scanning speed. With the increase of pulse energy, the type I region gradually moves down along the direction that the laser propagates and an elongated filamentation with type II modification appears. This can be explained by the competition of self-focusing and plasma defocusing when the peak power is higher than the critical power P_{cr} :³²

$$P_{\text{cr}} = \frac{3.77\lambda^2}{8\pi n_0 n_2}, \quad (1)$$

where λ is the wavelength, n_0 is the linear refractive index, and n_2 is the nonlinear refractive index. In addition, the laser scanning velocity can be changed to obtain different types of tracks. Figure 2(b) represents the microscope images of laser-written

Table 1 Advantages and disadvantages of different configurations in transparent material.

Waveguide configuration	Advantages	Disadvantages
Type I	<ol style="list-style-type: none"> 1. Direct writing for 3D micromachining 2. Single-mode guiding structures 3. Longer wavelength guidance using multiscan technique 	<ol style="list-style-type: none"> 1. Distorted lattices with degraded bulk features 2. Bad thermal stabilities 3. Guidance only along one polarization 4. Realizable in limited crystals
Double line	<ol style="list-style-type: none"> 1. Well-preserved bulk features 2. Single- or low-order mode structures 3. Being easily achieved in crystals 4. Excellent thermal stabilities 5. Wide applicability in crystals 	<ol style="list-style-type: none"> 1. No guidance at long wavelength (e.g., mid-IR) 2. Guidance only along one polarization in some crystals (e.g., cubic YAG) 3. Being difficult for 3D waveguides
Depressed cladding	<ol style="list-style-type: none"> 1. Well-preserved bulk features 2. Guidance till long wavelength 3. Designed geometry and adjustable diameters 4. Very good thermal stabilities 5. High coupling efficiency with fibers 6. Potential guidance along any transverse direction 7. Wide applicability in crystals 	<ol style="list-style-type: none"> 1. Relatively longer production time 2. Being difficult for 3D waveguides
Optical-lattice-like cladding	<ol style="list-style-type: none"> 1. Being similar to double line and depressed cladding 2. 3D device by special designs 	<ol style="list-style-type: none"> 1. Special design for different functions and materials

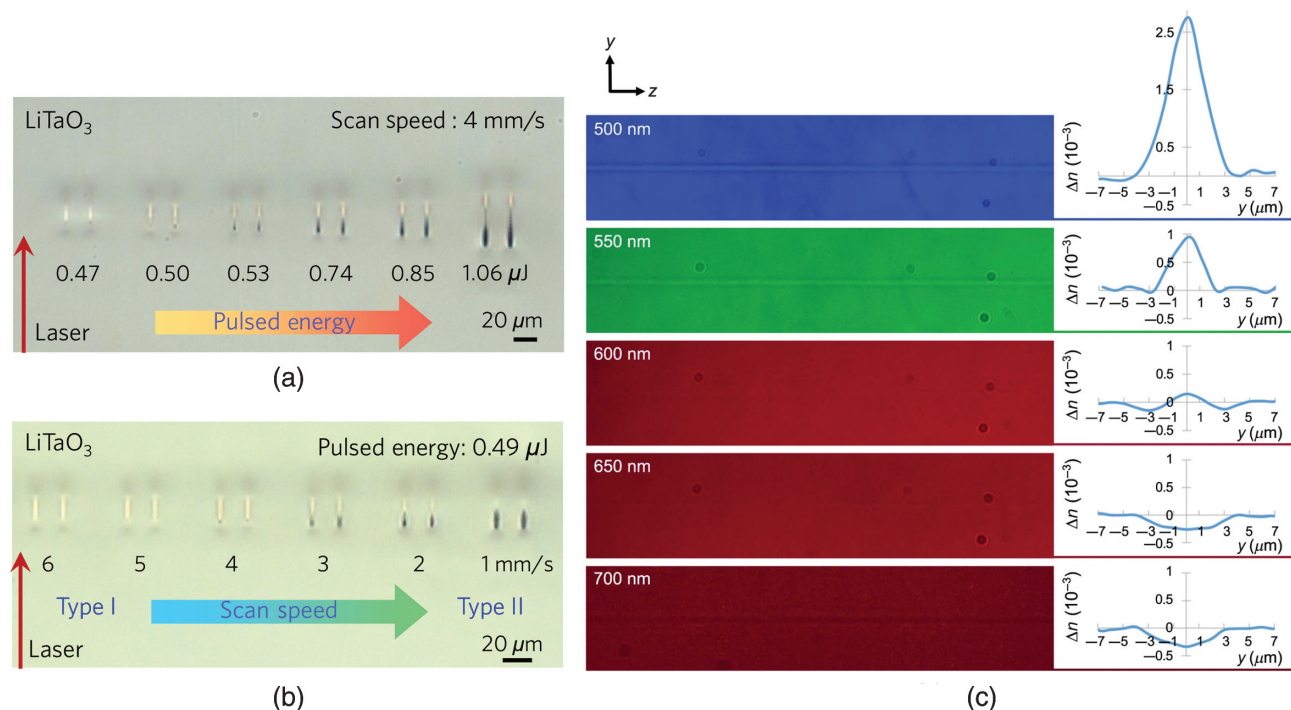


Fig. 2 Optical microscope images of laser-induced tracks with (a) different pulse energies and (b) scanning velocity in LiTaO₃ crystal. (c) Different refractive index change profiles from positive to negative, depending on the propagation wavelength in ZnSe crystal. Image (c) is reproduced with permission from Ref. 103, Creative Commons Attribution License (CC-BY).

tracks modified from type I to type II, as the scanning speed changes from 6 to 1 mm/s, whereas the pulse energy keeps a fixed value of 0.49 μJ . As the scanning speed decreases, more energy is deposited in the crystal unit volume, leading to more severe lattice distortion or damage, therefore, the slight type I modification will evolve into type II modification. More than that a new phenomenon allowing a significant increase of the refractive index contrast has been recently reported, which is called the effect of femtosecond-laser-induced electronic bandgap shift (FLIBGS).¹⁰³ When FLIBGS occurs, the absorption edge of the transmission spectrum shifts. At a propagation wavelength near the transparent material electronic resonance, the FLIBGS greatly affects the absorption and thus the refractive index, as predicted by the Kramers–Kronig relations.¹⁰⁴ By employing the effect of the FLIBGS, the tracks created by the femtosecond laser exhibit a dispersion of the refractive index, which in ZnSe crystals is the signal of the variation from positive to negative, as shown in Fig. 2(c). However, the FLIBGS effect generally happens in a narrow wavelength window. According to Lapointe et al.,¹⁰³ at a specific propagating wavelength, the refractive index modification becomes zero ($\Delta n = 0$), which means the laser inscription should be invisible, paving the way for new invisibility applications.

2.2 Control of Laser-Induced Tracks

The track morphology is crucial for laser inscribed waveguides because the track geometry plays an important role in the refractive index changes and stress fields. In addition, track engineering is also essential for nonlinear domain modification and well-defined microstructures.

2.2.1 Effect of writing parameters

For a given laser writing system, the main parameter that determines the localized modification in focal volume is the pulse intensity (or fluence).³² At low pulse intensity but beyond the optical modification threshold, an isotropic refractive index change is achieved,¹⁰⁵ whereas a birefringent refractive index change has been obtained at intermediate intensity.¹⁰⁶ Higher pulse intensity will lead to empty voids caused by microexplosions,¹⁰⁷ depending on the properties of the material. The transverse intensity distribution of the laser beam could be written as¹⁰⁸

$$I(r, z) = \left(\frac{w_0}{w(z)} \right)^2 \exp\left(-\frac{2r^2}{w^2(z)} \right), \quad (2)$$

where r is the radial distance and z is the axial distance from the beam waist. $w(z)$ is the variation of the laser beam spot size (the distance from the beam axis where the intensity drops to $1/e^2$ of the maximum value) and is given by

$$w(z) = w_0 \sqrt{1 + \left(\frac{z}{z_0} \right)^2}, \quad (3)$$

and w_0 is the diffraction-limited minimum beam waist radius:

$$w_0 = \frac{M^2 \lambda}{\pi \text{NA}}, \quad (4)$$

where M^2 is the Gaussian beam quality factor and NA is the numerical aperture of the focusing objective. The Rayleigh range

of the focus within a transparent material having a refractive index n is given by

$$z_0 = \frac{M^2 n \lambda}{\pi \text{NA}^2}. \quad (5)$$

The laser beam focus size at the focal plane is simply given by the w_0 and z_0 in the transverse and axial directions of the incident beam when neglecting optical aberrations and nonlinear effects. The peak intensities reached at the focal volume of the transparent material are:

$$I_p = \frac{E_p}{\pi w_0^2 \tau_p}, \quad (6)$$

where E_p is the pulse energy and τ_p is the pulse duration. As we can see, the beam waist determines the transversal size of the modifications induced in the dielectrics; together with the scanning velocity, they define the pulses superposition and the total amount of the energy delivered at a focal spot. It also defines the pulses that have enough intensity to promote changes in the material.

Generally, the higher the energy of laser pulses is, the more modified the tracks are.⁹⁰ In addition, higher-speed scanning creates weaker tracks.²³ This is easily understandable because the track geometry is determined by the average energy deposition of the unit volume in the samples. Figure 3(a) shows the laser-induced tracks in a Nd:YVO₄ crystal at different energies and scanning rates (the repetition rate of the laser is set at 1 kHz). Similar effects are observed for other crystals. Therefore, the choice of energy and scanning rate for a successful waveguide fabrication is to balance both effects on dielectric materials. The energy of pulses is typical of micro-Joule (μJ) or sub- μJ , and the used scanning rate ranges from tens of $\mu\text{m/s}$ to tens of mm/s. The polarization of femtosecond laser pulses may also affect the track writing, although in most cases, the effect is negligible. For example, in an optical-lattice-like structure of Nd:YAP crystal,⁷⁵ as femtosecond laser polarization is perpendicular to the scan direction, the waveguide cores are located in the regions between adjacent tracks, while in the center of the structure in the case of polarization parallel to the scan orientation. Figure 3(b) shows the optical microscope image of the lattice-like structure in a Nd:YAP crystal and the guided modes obtained under different laser polarizations.

2.2.2 Properties of crystal materials

The unique characteristics of crystal materials, such as birefringence and optical nonlinearities will affect the track morphology. For example, in Nd:GdVO₄, along the crystalline orientation of the c axis, the incident laser is split into two beams inside the crystal due to the high birefringence; as a result, two tracks are created at one shot of a single-laser pulse [Fig. 3(c)]. Nevertheless, such behaviors strongly depend on the used laser parameters and the bulk features. In many cases, nonlinear optical effects highly influence the propagation of femtosecond laser pulses in crystals, resulting in the morphology of tracks that are more likely to be an elongated filamentation or structure with multifoci not only a single focus.^{110,111} It is a very complex dynamic process. As the light propagates through a dielectric, the optical response is described as a macroscopic polarization induced by displacement of bound charges. For the simplified case of a centrosymmetric material that nullified the second order

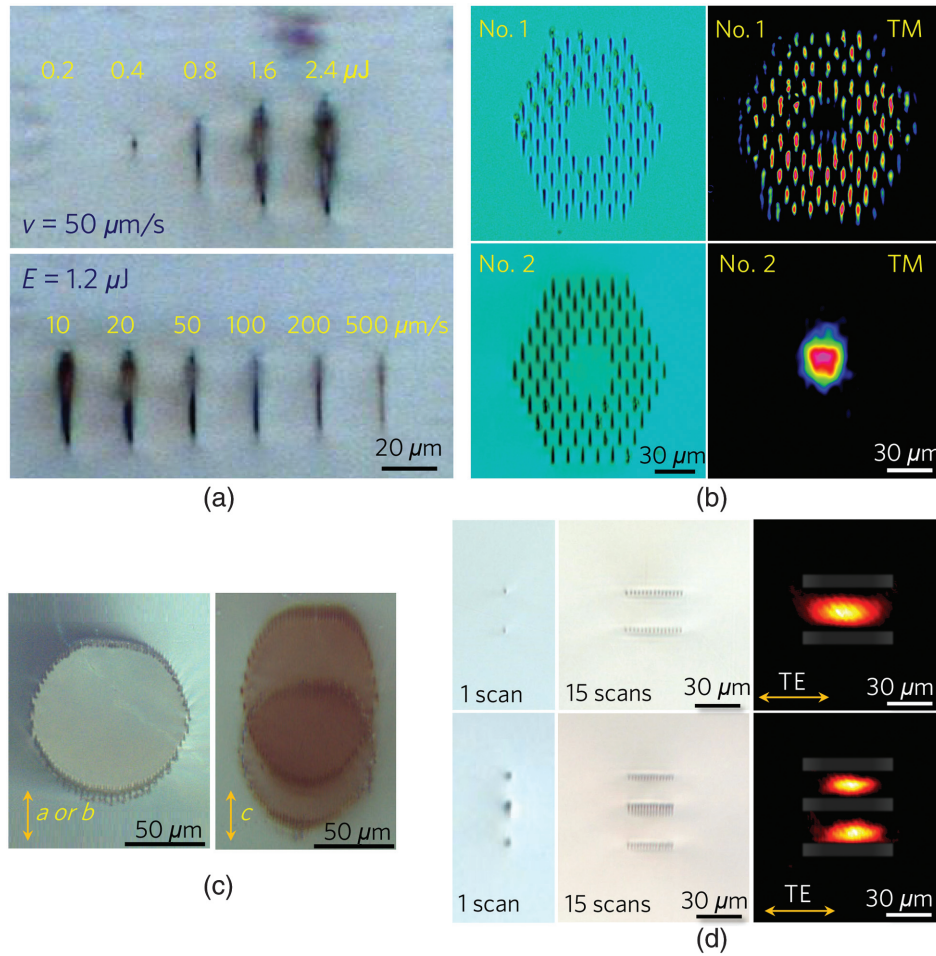


Fig. 3 (a) Microscope images of tracks with different pulse energies and scanning rates in Nd:YVO₄. (b) Influence of different laser polarizations on the tracks and modal profiles in Nd:YAP, perpendicular to the scans (No. 1) and parallel to the scans (No. 2), respectively. Images reprinted from Ref. 75, © 2016 The Optical Society (OSA). (c) Optical images of tracks as the laser writing along different crystalline orientations in Nd:GdVO₄. (d) Laser-induced multifoci in LiTaO₃, corresponding horizontal waveguides, and modal profiles. Reprinted with permission from Ref. 109, © 2019 IEEE.

(an all even order), if fourth and higher order nonlinear effects are neglected, the polarization of a centrosymmetric medium can be written as¹¹²

$$\mathbf{P}(t) = \epsilon_0 \left(\chi^{(1)} + \frac{3}{4} \chi^{(3)} |\mathbf{E}|^2 \right) \mathbf{E}(t), \quad (7)$$

where $\chi^{(i)}$ is the i th order nonlinear susceptibility and $\mathbf{E}(t)$ is the incident electric field. The refractive index of the material is defined by n_0 and n_2 (optical Kerr effect) when the linear part dominates over the nonlinear part:³²

$$n = \sqrt{1 + \chi^{(1)} + \frac{3}{4} \chi^{(3)} |\mathbf{E}|^2} = n_0 + n_2 I, \quad (8)$$

where $n_0 = \sqrt{1 + \chi^{(1)}}$, $n_2 = \frac{3}{4} \frac{\chi^{(3)}}{\epsilon_0 c n_0^2}$, and $I = \frac{1}{2} \epsilon_0 n_0 c |\mathbf{E}|^2$ is the laser intensity. Thereby, a spatially dependent intensity of the Gaussian laser beam modifies the effective refractive index of

dielectrics. In most media, the n_2 is positive and the refractive index at the center of the beam is higher than in the wings, acting as a positive lens to self-focus the light, as if the femtosecond laser pulse exceeds the P_{cr} , a collapse occurs at a focal point. Additionally, the convex effect induces sufficient intensity to create ionization and other nonlinear effects, forming an electron plasma that acts as a concave lens to defocus the laser beam. The self-focusing comes again provided the intensity is higher than the P_{cr} . Such a spontaneous multiple focusing can be realized in both type I and type II regimes. For example, in LiNbO₃ crystals, under the high-repetition-rate of femtosecond laser (700 kHz) for type II modification, the altered track consists of three point-like structures at a single shot, which is identified with a periodic distribution of lattice defects.¹¹³ Furthermore, the laser-induced grating structure of a period $\sim 1.2 \mu\text{m}$ is also implemented. In addition, in the regime of low repetition rate (8 kHz), the multifoci tracks based on type I modification can be demonstrated as the energy increases and the track morphology is determined by the energy of laser pulses.⁹¹ Another interesting sample is LiTaO₃ crystal, where the multiple-focusing

spots with different numbers are also realized, depending on the laser-irradiated parameters.¹⁰⁹ Using the multiscan technique, researchers have fabricated desired structures to obtain horizontal tracks for waveguide-guiding along with TE polarization, whereas the normal double-line waveguide only supports TM polarization [Fig. 3(d)]. In this design, the layer of the waveguide is composed of a number of modified points, forming a dotted line, and the total length can be controlled by the laser scans. Theoretically, more layers of waveguides can be possibly obtained using this approach.

2.2.3 Beam shaping techniques

During the process of femtosecond laser irradiation, the laser beam is generally fixed and focused at a certain depth beneath the sample surface. The sample is usually mounted on a computer-controlled XYZ linear stage to realize an arbitrary shape, and a CCD system is used to monitor the waveguide writing process. The sample can be translated along the beam propagation direction, i.e., longitudinal writing, or orthogonally to, i.e.,

transverse writing.⁶⁰ The waveguide formed by longitudinal writing has intrinsic circular symmetry, while it is restricted by the working distance of the objective lens and it has limitations in 3D layouts.¹¹⁴ In contrast, the design flexibility of transverse writing is enormous and allows for the fabrication of 3D structures,^{23,60} in spite of their asymmetric cross sections.

Over the past decades, beam-engineering techniques have been developed to improve the shape of the laser-written waveguides, such as the double-beam technique,¹¹⁵ adding cylindrical lenses,¹¹⁶ the slit technique,⁴¹ using a nondiffractive Bessel beam,¹¹⁷ and using a spatial light modulator (SLM).³⁹ Figure 4(a) illustrates the writing process of depressed cladding waveguides utilizing an ellipsoidal focal spot, where a number of tracks are inscribed one by one, thus, resulting in a time-consuming process. Nowadays, SLMs have been used widely in femtosecond laser processing. From the work of Qi et al., the fabrication times have been considerably reduced,⁴¹ as shown in Fig. 4(b). The phase mask for writing horizontal lines is presented in Fig. 4(c). Thereby, horizontal tracks can be employed to construct the

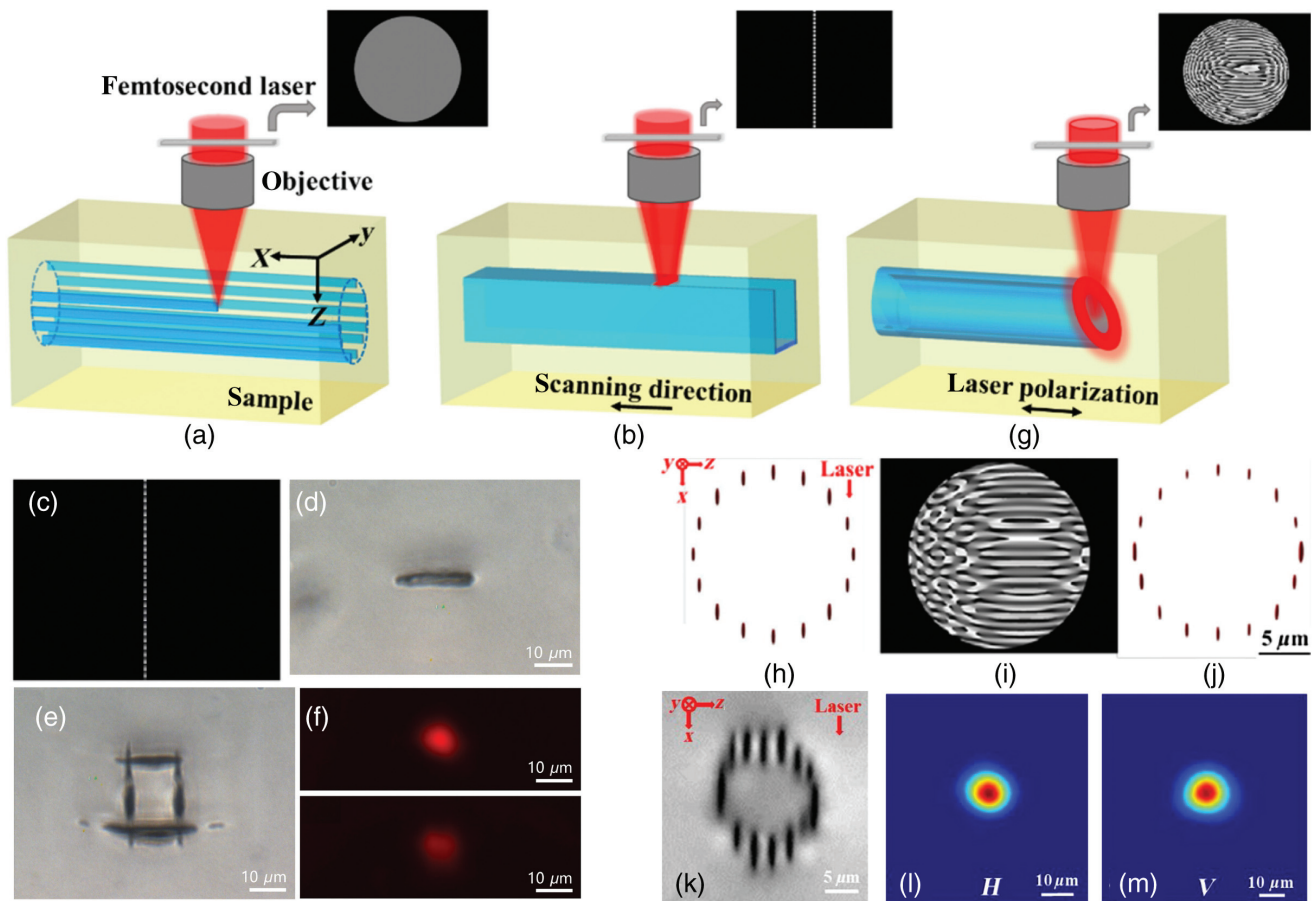


Fig. 4 Schematic diagram of direct laser-written cladding waveguides (a) with an ellipsoidal focal spot and (b) with a slit-shaped beam focus. Images (a), (b), and (g) are reprinted with permission from Ref. 40, © 2017 OSA. (c) The phase mask for writing horizontal lines. Microscope image of (d) horizontal tracks, (e) waveguide, and (f) near-field profiles. Images (c)–(f) are reproduced with permission from Ref. 42, CC-BY. (g) Schematic plot of single-scan cladding waveguides utilizing a longitudinal ring-shaped focal field. (h) Calculated 3D isosurface, (i) phase mask, and (j) simulated focal intensity profile. (k) Microscope image and (l), (m) corresponding modal profiles. Images (h)–(m) are reprinted with permission from Ref. 118, © 2019 Chinese Laser Press (CLP).

cladding of waveguides in a LiNbO₃ crystal that exhibit the polarization-independent guidance⁴² [Figs. 4(d)–4(f)]. Later in 2019, Zhang et al.¹¹⁸ proposed a single-scan method to fabricate cladding waveguides using 3D engineered focal field [Figs. 4(g)–4(j)]. The longitudinally oriented ring-shaped focal field is composed of 16 discrete spots, allowing for the generation of annular cladding waveguides at a single shot of laser writing, demonstrated in Figs. 4(k)–4(m). Most recently, Sun et al.³⁹ developed a new and efficient multifoci-shaped femtosecond pulsed method to write circular symmetric waveguides with an SLM.

2.2.4 Nonlinear domain engineering

The morphology of tracks is crucial for not only waveguide configuration but also for engineering the crystal properties to achieve modulation of nonlinearity polarization. Especially for type II modification, focused laser pulses will cause highly damaged lattices, where an amorphous volume forms that reduces the nonlinear coefficient. Such a modification can be periodically introduced to realize spatial modulation of the second-order nonlinear coefficient $\chi^{(2)}$, enabling quasi-phase-matching (QPM) structures,¹¹⁹ also known as nonlinear photonic crystals (NPC).¹²⁰ Thereby, the researchers have fabricated 3D NPCs by selectively erasing the nonlinear coefficients of a LiNbO₃ crystal.¹²¹ The recently developed 3D NPCs present efficient generations of second-harmonic vortex and Hermite–Gaussian beams, showing the potential for nonlinear beam-shaping devices⁶ and nonlinear volume holography^{122,123} in comparison to the two-dimensional (2D) case. Additionally, the huge flexibility of direct laser writing enables complex 3D domain structures,^{43,120} allowing their monolithic integration with other phase modulators and switches, and improving the performances and functions of on-chip devices. Using the same strategy of domain erasing in a quartz crystal, researchers from Shandong University have realized tunable second harmonic

generation (SHG) down to the ultraviolet wavelength.¹²⁴ In addition, there will be an obvious thermal effect at the high-repetition rate of femtosecond laser pulses, which can be utilized to realize reversed polarization. The group from Australia reported on the similar 3D domain structures in a BCT crystal by femtosecond laser direct writing through the thermoelectric-field poling,¹²⁵ allowing for precise domain inversion with high resolution. This laser engineering strategy can be extended to other systems of crystals.

Moreover, laser-induced quasi-phase-matching (LiQPM) grating structures inside the waveguide's core allow efficient frequency conversion, which opens new avenues for advanced all-integrated nonlinear devices. For instance, using femtosecond laser-induced domain inversion in LiNbO₃ waveguides, a conversion efficiency of 17.45% for frequency doubling of 815 nm has been demonstrated.¹²⁶ In addition, a depressed cladding waveguide-integrated LiQPM grating scheme has been proposed by Denz et al.¹²⁷ With the precise and flexible 3D domain engineering, as shown in Figs. 5(a) and 5(b), they have realized broadband and multiwavelength frequency conversions.⁴³ Particularly, the second harmonic vortex can be demonstrated by a helical twisted domain grating into a waveguide, as depicted in Figs. 5(c)–5(e).

2.2.5 Well-defined micro- and nanostructures

The laser-written track is also promising for creating spatially well-defined microstructures in combination with chemical wet etching. In the laser-modified volumes, the femtosecond pulses have driven the crystal network to a predamaged state that shows a larger selective etching than the unperturbed bulk lattices.¹²⁸ A good sample is the YAG crystal, as shown in Fig. 6(a), where the optical-lattice-like waveguide is fabricated by direct laser writing, and then the sample is immersed into the selected acid (e.g., phosphoric acid).¹²⁹ As one can see, after some time,

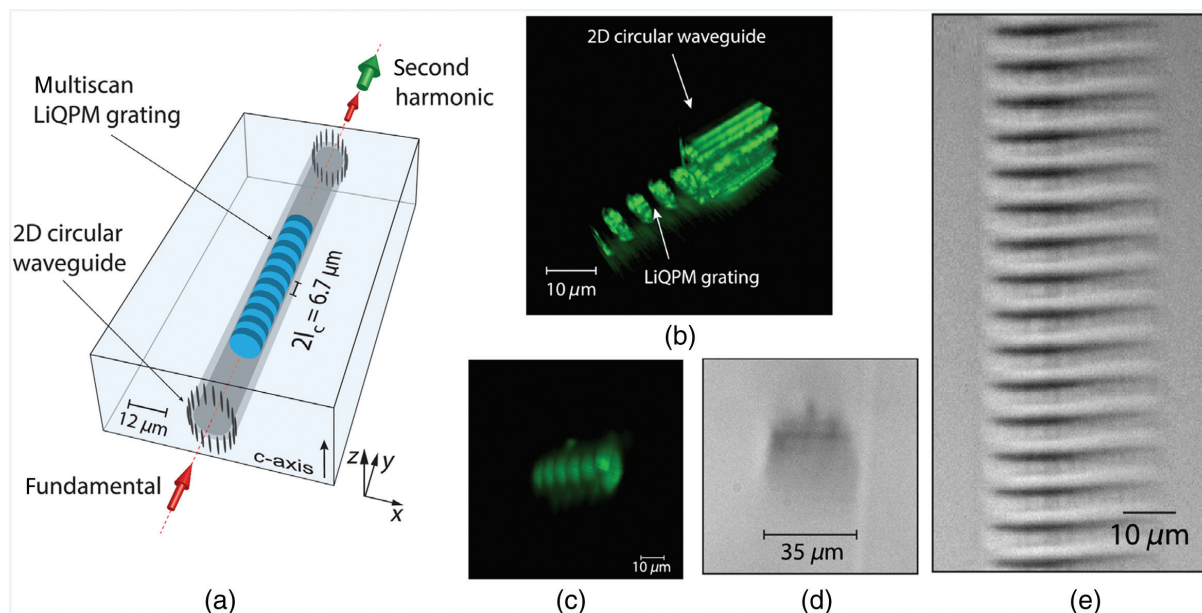


Fig. 5 (a) Schematic design of a waveguide-integrated LiQPM grating; (b) SH microscope image of LiQPM grating and waveguide; and (c) helical grating structure. Microscope image of the helical structure: (d) the front face and (e) top view. Images reprinted with permission from Ref. 43, © 2020 OSA.

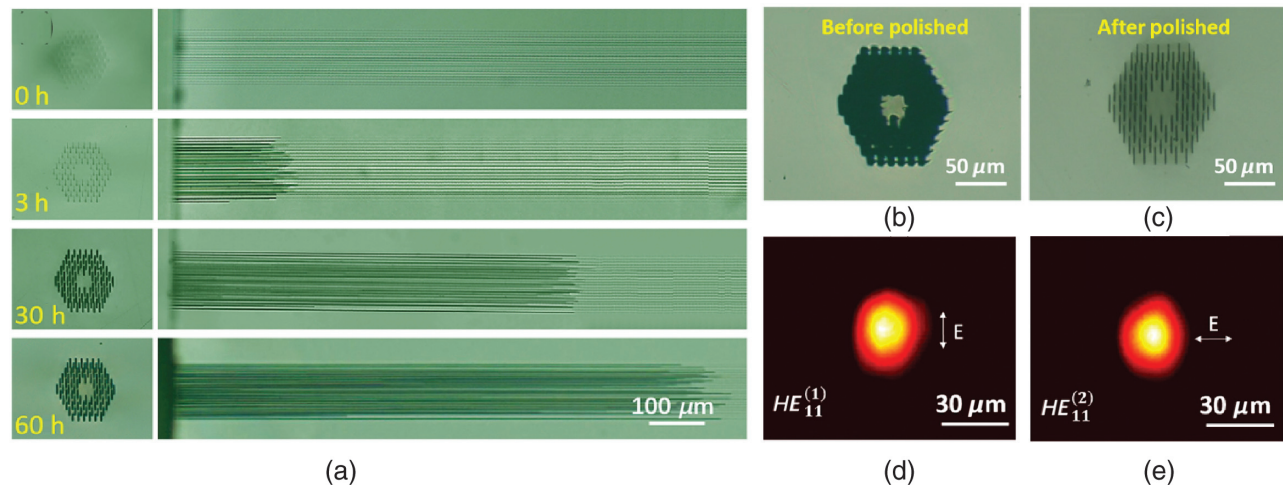


Fig. 6 (a) Microscope images of hollow optical-lattice-like structures at different etching times in YAG crystal. (b) Before polished and (c) after polished. Near-field modal profiles at $4 \mu\text{m}$ along (d) TM and (e) TE polarization, respectively. Images reproduced with permission from Ref. 129, © 2020 CLP.

the etched portion of the damaged track is filled with air, thus resulting in a larger refractive index change than the unetched region [Figs. 6(b) and 6(c)]. Therefore, the confinement of the light field will be enhanced. In this work, the etched microstructured optical waveguide supports good guidance at the MIR wavelength of $4 \mu\text{m}$ [Figs. 6(d) and 6(e)], which is impossible for structure before etching. Actually, early in 2013, Kar et al. produced large microfluidic channels within YAG by selectively wet etching laser-written type I tracks.¹³⁰ The technique is very interesting and offers an efficient solution to 3D nanolithography of crystals. In 2019, Osellame et al.¹³¹ reported on the nanopores with mm-scale length. By implementing auxiliary vertical etching pores to build a 3D-connecting etching architecture, they have achieved 3D nanolithography with a long length. Using this strategy, they have produced subwavelength diffraction gratings and microstructured waveguides in YAG crystals.

2.3 Mode Guidance Correlated to Crystal Systems

Dielectric crystals are important media for optical and photonic applications, which can be roughly cast into a few groups, such as laser crystals, nonlinear optical crystals, and electrooptic crystals.²³ Unlike the amorphous substrates, crystalline material entails structures of long-range orders of lattices, offering unique physical properties that are not exhibited in glass, such as even order optical nonlinearities and birefringence.¹ There are seven crystal systems to characterize the crystal lattices or structures of different geometries, i.e., cubic, tetragonal, hexagonal, trigonal, orthorhombic, monoclinic, and triclinic. Up to the current date, a wide range of crystals has been used for the fabrication of direct laser-written waveguides; for example, YAG crystal belongs to the cubic, LiNbO_3 , β -BBO, and sapphire crystals belong to the trigonal, and 6H-SiC crystal belongs to the hexagonal crystal family. To some extent, the symmetry of each lattice determines the waveguide properties, such as polarization dependence and propagation loss. Table 2 summarizes the waveguide configurations and guiding properties of typical crystals in different crystal systems. For each lattice family, a brief overview will be given below.

For cubic systems, YAG is one of the most well-studied crystals in the direct laser writing of waveguides. For the depressed-cladding and optical-lattice-like waveguides, symmetric guidance for any polarization, i.e., both transverse magnetic (TM) and electric (TE) are supported,^{100,132} whereas the dual-line geometry only allows TM-polarized guidance,⁷⁹ which is mainly related to the high symmetry of the cubic system that possesses three fourfold axes of rotation. The cubic-lattice crystals exposed to laser irradiation undergo a severe modification that results in a positive index change in a specific polarization. A similar polarization-dependent guiding behavior is also found in Nd:GGG crystal,¹³⁴ another typical cubic crystal. Although for the tetragonal system, such as Nd:YVO₄ and Nd:GdVO₄, only one fourfold axis of rotation is contained, leading to a lower symmetry than that of the cubic system. As a result, the dual-line structure supports guidance along both TE and TM polarizations^{138,141} but is still of slightly anisotropic guidance. The depressed-cladding waveguides in the tetragonal is guided in an anisotropic way as well, which is different from the cubic YAG crystal. As for the hexagonal system, up to now, only 6H-SiC crystal is employed for laser writing of waveguides.¹⁴³ Like the cubic system, both TM and TE guidances are determined in rectangular cladding waveguides, whereas only TM guiding mode is supported in dual-line structure. In hexagonal system, there is one six-fold axis of rotation, thus the symmetry is between the cubic and tetragonal systems.

LiNbO_3 , LiTaO_3 , β -BBO, and sapphire, as the typical crystals belonging to the trigonal system, exhibit unique nonlinearities and birefringence. Therefore, as these crystals are processed under laser writing, there will be some different structural modifications due to the high-order threefold axis of rotation. Various configurations have been inscribed in LiNbO_3 crystal by femtosecond lasers, such as type I modified single-line and multiline structures, type II modified dual-line waveguides, depressed-cladding, optical-lattice-like, and ridge waveguides. Generally, in LiNbO_3 and LiTaO_3 crystals, type I and type II waveguides would suffer from serious polarization-dependent guiding, which only permits polarization along the orientation parallel to the laser-induced tracks,^{91,147} whereas

Table 2 Summary of latest published works about waveguide configuration and properties of typical crystals in different crystal systems.

Crystal system	Material	Waveguide configuration	Guiding properties		Ref.	
			Polarization dependence	Minimum propagation loss (dB/cm)		
Cubic crystals	Nd:YAG	Type I (single line)	TE and TM	5@632.8 nm	87	
		Dual line	TM	0.21@632.8 nm	79	
		Double cladding	TE and TM	1.3@632.8 nm	132	
		Optical-lattice like	TE and TM	0.7@1064 nm	100	
		Cladding + dual line	TE and TM	—	133	
	Nd:GGG	Dual line	TM	2.0@632.8 nm	134	
		Depressed cladding	TE and TM	1.7@632.8 nm	135	
	BGO	Type I (multiscan)	TE and TM	3.22@4 μ m	89	
		Type I (ring shaped)	TE and TM	1.56@1550 nm	136	
		Dual line	TE and TM	0.5@632.8 nm	137	
	Tetragonal crystals	Nd:YVO ₄	Depressed cladding	TE and TM	2.1@632.8 nm	137
			Dual line	TE and TM	0.8@632.8 nm	138
			Depressed cladding	TE and TM	1.1@632.9 nm	139
Nd:GdVO ₄		Optical-lattice like	—	—	140	
		Dual line	TM	0.5@1064 nm	141	
		Depressed cladding	TE and TM	0.7@632.8 nm	142	
Hexagonal crystals		6H-SiC	Dual line	TM	0.78@1064 nm	143
			Rectangular cladding	TE and TM	1.62@1064 nm	143
Trigonal crystals		LiNbO ₃	Type I (single line)	TM	2.22@1064 nm	144
			Type I (multiline)	TM	1.98@632.8 nm	91
			Dual line (vertical)	TM	0.6@1064 nm	63
			Dual line (horizontal)	TE	3.25@1550 nm	41
			Depressed cladding	TE and TM	1.25@1550 nm	118
	Optical-lattice like		TE	1.27@1550 nm	145	
	Ridge configuration		TM	3.28@4 μ m	146	
	LiTaO ₃	Type I (single line)	TM	2.67@632.8 nm	147	
		Dual line (horizontal)	TE	1.7@632.8 nm	109	
		Depressed cladding	TE and TM	1.56@1550 nm	148	
		Rectangular cladding	TE and TM	0.12@1550 nm	149	
		Depressed cladding	TM	0.19@800 nm	150	
		Depressed cladding	TE and TM	2.3@633 nm	151	
Orthorhombic crystals	KTP	Dual line	TM	0.65@798.5 nm	152	
		Depressed cladding	TE and TM	0.37@2850 nm	80	
		Optical-lattice like	TE and TM	2.9@1064 nm	153	
		Type I (multiline)	TM	1.0@980 nm	154	
		Dual line	TE and TM	0.8@633 nm	155	
		Depressed cladding	TE and TM	1.7@632.8 nm	156	
		Optical-lattice like	TE and TM	1.2@632.8 nm	157	
	Nd:YAP	Depressed cladding	TE and TM	0.15@1064 nm	158	
		Optical-lattice like	TE and TM	1.11@1064 nm	159	
	Monoclinic crystals	BiB ₃ O ₆	Depressed cladding	TE and TM	0.6@1064 nm	160
			Type I	TM	1,1@1550 nm	85
		Nd:YCOB	Depressed cladding	TM and TE	—	161
			Double cladding	TM and TE	0.65@633 nm	162
Nd:KGW		Dual line	TM and TE	2.0@632.8 nm	163	
		Depressed cladding	TM and TE	1.8@1064 nm	164	

the depressed-cladding waveguide enables isotropic guiding as well as in Ti-sapphire crystal.¹⁶⁵ However, for β -BBO crystal, only TM-polarized guiding is supported in depressed-cladding waveguides.¹⁵⁰ In addition, polarization-dependent guiding also happens in both optical-lattice-like¹⁴⁵ and ridge configurations¹⁴⁶ in a LiNbO₃ crystal, which only allows n_0 -polarized guidance.

In addition to the crystal system mentioned above there are lower-symmetry crystal families that do not contain a higher-fold axis of rotation, such as orthorhombic system and monoclinic system. In such crystal systems, laser-written waveguides present isotropic guiding behavior. For instance, KTiOPO₄ (KTP) is a typical crystal belonging to the orthorhombic system; the guidance for dual-line,¹⁵⁵ depressed-cladding,¹⁵⁶ and optical-lattice-like waveguides¹⁵⁷ is almost polarization independent. In addition, BiB₃O₆,¹⁶⁰ Nd:KGW,¹⁶⁴ and KLu(WO₄)¹⁶⁶ crystals belong to the monoclinic system, another member of the lower-symmetry family. In these crystals, femtosecond laser-written waveguides mainly focus on the depressed-cladding configuration, which allows symmetric guidance for both TM and TE polarizations due to the poor lattice symmetry. These features imply the unique modification of laser pulses on the crystals with different symmetries.

2.4 Mode Modulation

In recent years, numerous newly developed waveguide geometries that allow mode modulation and guiding polarization engineering have been demonstrated, benefiting from the rapid and direct prototyping of femtosecond laser writing.

For type I modification, the multiscan technique offers a flexible manner to tailor the mode profiles by controlling the number and separation of laser-induced tracks⁹¹ [Fig. 7(a)]. In addition, by arranging the laser-modified tracks into an annular ring, the ring-shaped mode profiles in BGO crystal have also been obtained,¹³⁶ as shown in Fig. 7(b). In particular, such a design allows for polarization-independent guiding, which is different from single- and multiline type I waveguides that only support one special polarized guiding. In addition, the rings can be inscribed in the desired diameter to meet the practical applications. Theoretically, one could realize mode modulation with arbitrary geometries by proper design and arrangement of the laser-induced tracks.

As for type II modification, one can fabricate a series of dotted lines along the horizontal direction via multiscan, modulating the modal profiles along the TE polarization owing to the changed stress field distribution,¹⁶⁷ as shown in Fig. 7(c). At recent reports in SiC crystal, the mode modulation of waveguides has also been achieved by controlling the morphology of laser-induced tracks.¹⁴³ In addition, femtosecond laser-written waveguide arrays based on type II modification can implement mode modulation via the effect of evanescent coupling. For example, the one-dimensional planar waveguide laser arrays in Nd:YAG crystals support flexible discrete diffraction by adjusting the coupling condition.¹⁷⁰ Zhang et al.³⁷ reported the 2D waveguide arrays in LiNbO₃ crystal, where the output mode profiles can be modulated as the pump beam goes into the array at different positions.

Beyond the above reports, some designs based on the depressed-cladding structures also enable mode modulation. Sun et al. proposed an “ear-like” waveguide structure with extra tracks at the sides of cladding, which is an evolved scheme of double-line and depressed-cladding waveguides that was first

reported by Okhrimchuk et al.¹⁷¹ Such a design is intended to further enhance the optical confinement of the light field. Therefore, more symmetric mode profiles and lower propagation losses are preferred compared with the normal cladding waveguides,¹⁶⁸ as shown in Fig. 7(d). Another interesting sample is the double-cladding waveguide [Fig. 7(e)], which is composed of two concentric tubular shapes with different diameters.¹⁷² One of the advantages is that the outer cladding could enhance the coupling efficiency of the pump beam and inner cladding permits an output beam with low-order modes, i.e., generating an integrated single-mode laser system.¹⁶⁹ Moreover, using a third-cladding structure, Wu et al.¹³² demonstrated annular ring waveguide lasers, promising for complex integrated pump sources to alleviate thermal lensing.

3 Designs for Laser-Written 3D Crystalline Photonic Devices

New emerging photonic devices with 3D configurations have attracted considerable attention. Femtosecond laser writing, as the most efficient technique for true 3D microfabrication, allows an arbitrary geometry in almost any transparent substrate. Although various laser-written 3D structures have been successfully fabricated in glass,^{34,173–177} the complexity of lattice and anisotropies make it much difficult to manipulate in crystals. Special designs are, therefore, required by varying the writing parameters and track geometries. In this section, we will overview the recent reports on waveguide-based 3D photonic structures, such as Y-branches, beam steering elements, ring-shaped transformers, polarizers, tapered structures, and directional couplers.

3.1 Y-Branched

As the basic 3D components, a Y branch-based splitter allows for dividing the single-input beam into several output beams in different dimensions. For example, the 3D waveguide beam splitters (1 × 4) based on type-I modification have been realized in LiNbO₃⁸⁸ and BGO crystals,⁸⁹ whereas the polarization-dependent properties are very different for the two crystalline waveguides. The LiNbO₃ only supports light propagation along particular orientations, whereas wave guidance in BGO is isotropic for any transverse polarization. As for type-II modification, Y-branches work well in a relatively simple way for 2D structures, as reported in diamond¹⁷⁸ and YAG crystals.⁹⁷ However, in the 3D case, disadvantages are emerging due to the unbalanced guiding properties and additional architecture required to connect separate channels. Moreover, the geometry of depressed cladding supports wave-guiding along any polarization, and for 3D fabrication it also requires a complicated design. The buried Y-junctions based on circular and rectangular depressed-cladding geometries have been proposed in YAG^{179,180} and Ti:sapphire crystals,¹⁸¹ as shown in Figs. 8(a)–8(e), which produce smooth transitions and preserve modal profiles. Furthermore, Ajates et al. reported the novel 3D beam splitting based on depressed-cladding architecture in LiNbO₃ crystals [Fig. 8(f)], realizing the desired 3D guiding path.¹⁸² Although the TM-polarized modes have higher losses than that of TE, the 3D structures work well in the near-infrared band. It is possible to implement any arbitrary 3D photonic circuit in crystalline materials based on circular cladding waveguides.

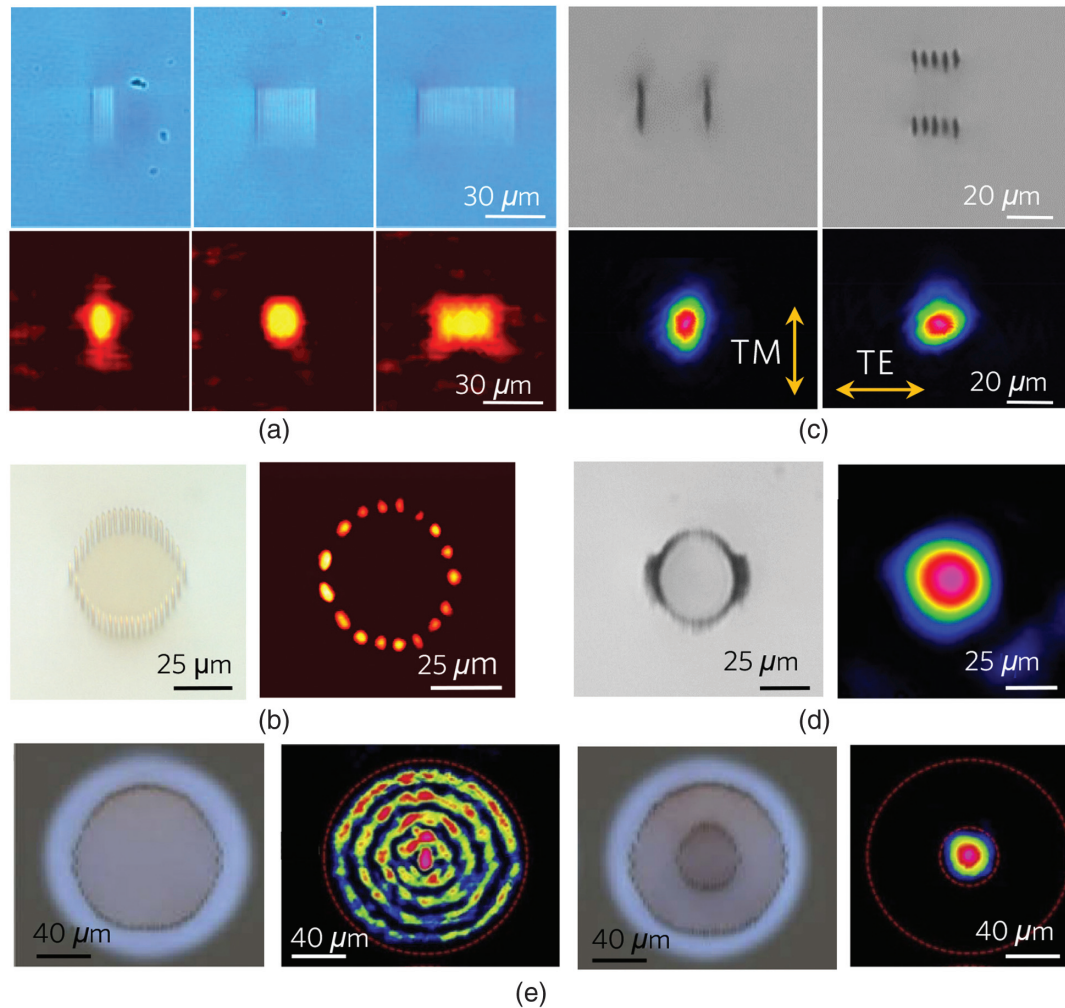


Fig. 7 (a) Microscope images and modal profiles of tailored multiline waveguides in a LiNbO_3 crystal, reproduced with permission from Ref. 91, © 2018 Elsevier. (b) Ring-shaped waveguide based on type-I modification in a BGO crystal, reprinted with permission from Ref. 136, © 2017 OSA. (c) Polarization engineering for dual-line waveguides in a LiNbO_3 crystal, reproduced with permission from Ref. 167, © 2020 Elsevier. (d) The “ear-like” waveguide in Nd:YAG crystal, reprinted with permission from Ref. 168, © 2021 OSA. (e) Double-cladding waveguide in Nd:YVO₄ crystal, reprinted with permission from Ref. 169, © 2019 OSA.

3.2 Photonic-Lattice-Like Structures for 3D Microfabrication

Additional branched boundaries are essential for beam splitters in Sec. 3.1; as a result, the fabrication processing is usually time-consuming. To this end, the family for 3D microfabrication is designed, known as an “optical-lattice-like” photonic structure, which contains a number of periodic tracks with geometry defects. A typical structure has a track layer of hexagonal layout, where the light field is confined at enclosed track-free regions. Particularly, better guiding confinement is followed at multilayers rather than single layer. Tailored beam steering in the 3D manner, such as beam splitting or ring-shaped transformation,¹⁰⁰ can be realized by directly introducing additional defect tracks at certain positions. These defect lines served as beam blocks or confiners that smoothly reshape the guiding profiles during the propagation; therefore, no additional losses are introduced.¹⁰² Based on this strategy, 3D beam steering of

ring-shaped transformation has been successfully inscribed in YAG crystals,¹⁰⁰ as shown in Figs. 9(a) and 9(b). In addition, highly compact 3D beam splitters ($1 \times N$) have been implemented in many platforms, such as LiNbO_3 ,¹⁴⁵ KTiOPO_4 ,¹⁵⁷ Nd:YAG, and Ti-sapphire crystals¹⁵³ [Figs. 9(c)–9(e)]. By carefully considering the crystalline properties, more advanced applications, such as nonlinear frequency conversion¹⁸³ and miniature laser sources have been demonstrated.¹⁰¹ Such 3D designs could efficiently improve the integration of complex photonic circuits. Considering the huge differences in the characteristics of crystals, however, each type requires a different 3D inscription and feasible optimization scheme.

3.3 3D Polarizers

In photonic integrated circuits (PICs), the undesirable effect of polarization dependence limits the practical applications of polarization-division multiplexed systems. Hence, polarization

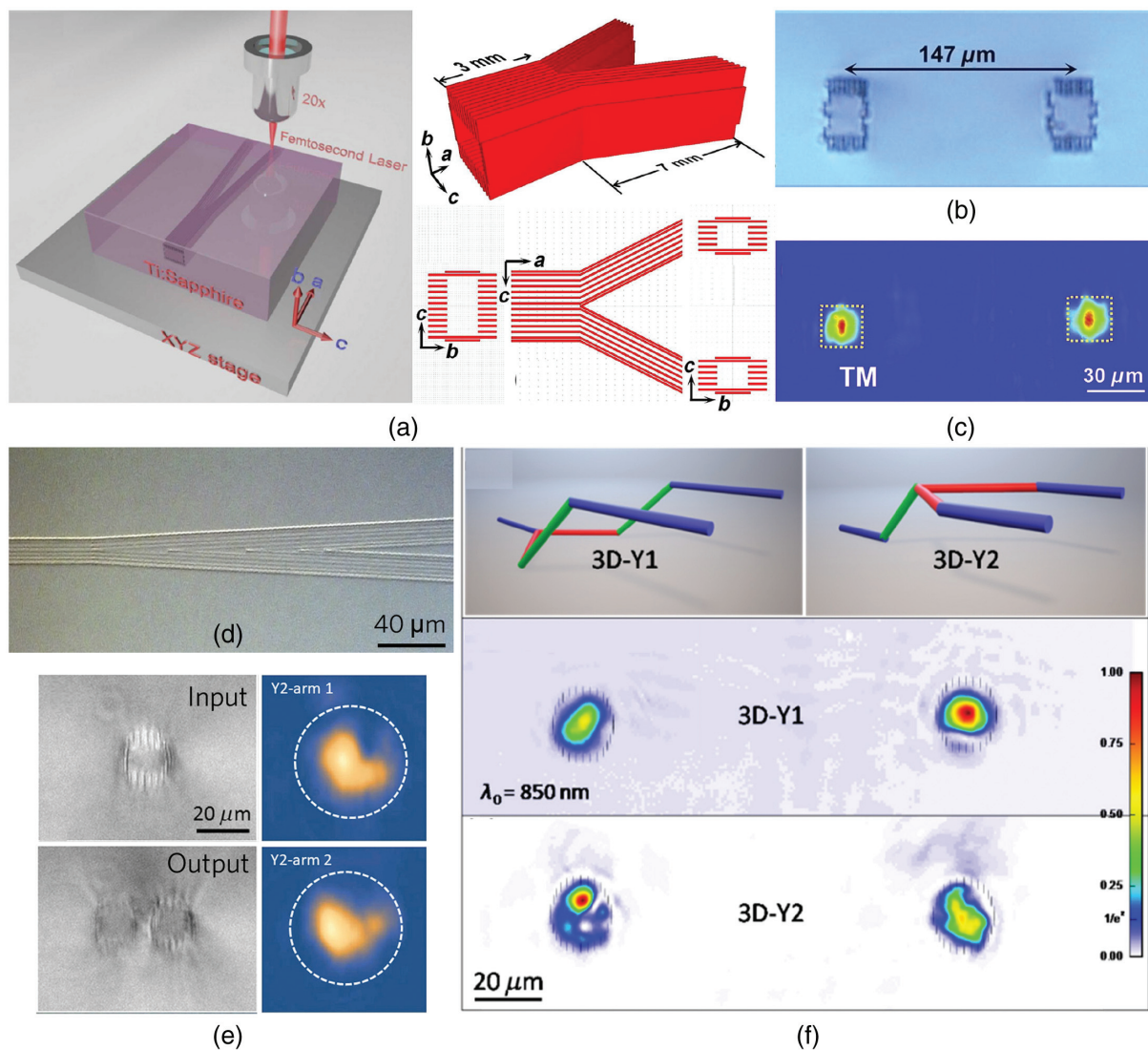


Fig. 8 (a) Fabrication and 3D schematic diagram of Y-splitters based on rectangular cladding geometry in Ti:sapphire crystal, (b) microscope image of 1-deg branching angle, and (c) intensity distributions at 1064 nm. Images (a)–(c) are reproduced with permission from Ref. 181, © 2018 Elsevier. Microscope images of Y-branch with circular cladding structure (d) in top view and (e) in cross section, as well as modal profiles of two arms. Images (d) and (e) are reproduced with permission from Ref. 179, © 2017 Elsevier. (f) 3D beam-splitting structures in a LiNbO_3 crystal, reprinted with permission from Ref. 182, © 2018 Optica.

diversity devices including polarizers, polarization beam splitters (PBSs), and polarization rotators attract researchers' interest. Femtosecond laser writing provides an on-demand fabrication method for polarization photonic devices. Recently, a new structure of polarizer has been employed to engineer the guiding polarization in LiNbO_3 crystals,¹⁸⁴ as depicted in Figs. 10(a) and 10(b), which is composed of three parts: a rectangular-shaped channel, a 1×2 beam splitter acting as the connection element, and two output channels. In such a structure, the output will be well-separated into TE and TM polarized light as the polarized light or hybrid light of TE plus TM modes enters the waveguide [Fig. 10(c)]. The polarization extinction ratio and insertion loss can be comparable with that of fused-silica-based PBS. In addition, a 3D polarizer in a cross-shaped geometry has been produced in LiNbO_3 crystal,¹⁶⁷

consisting of a square cladding waveguide and four vertically oriented double lines [Figs. 10(d) and 10(e)]. This 3D configuration enables specific polarized-light output at different waveguide arrays, i.e., the TM or TE-polarized light can be controllably coupled into vertical or horizontal waveguide arrays, respectively.

3.4 Other Schemes

Tapered waveguide structures, like the tapered optical fibers, have different circular radii of input and output arms. In 2020, Romero et al.¹⁸⁵ proposed a 3D tapered waveguide by smoothly decreasing the separation between the tracks that consisted of the depressed-cladding waveguide, as shown in Fig. 11(a). This approach enables the desired reduction of the input/output

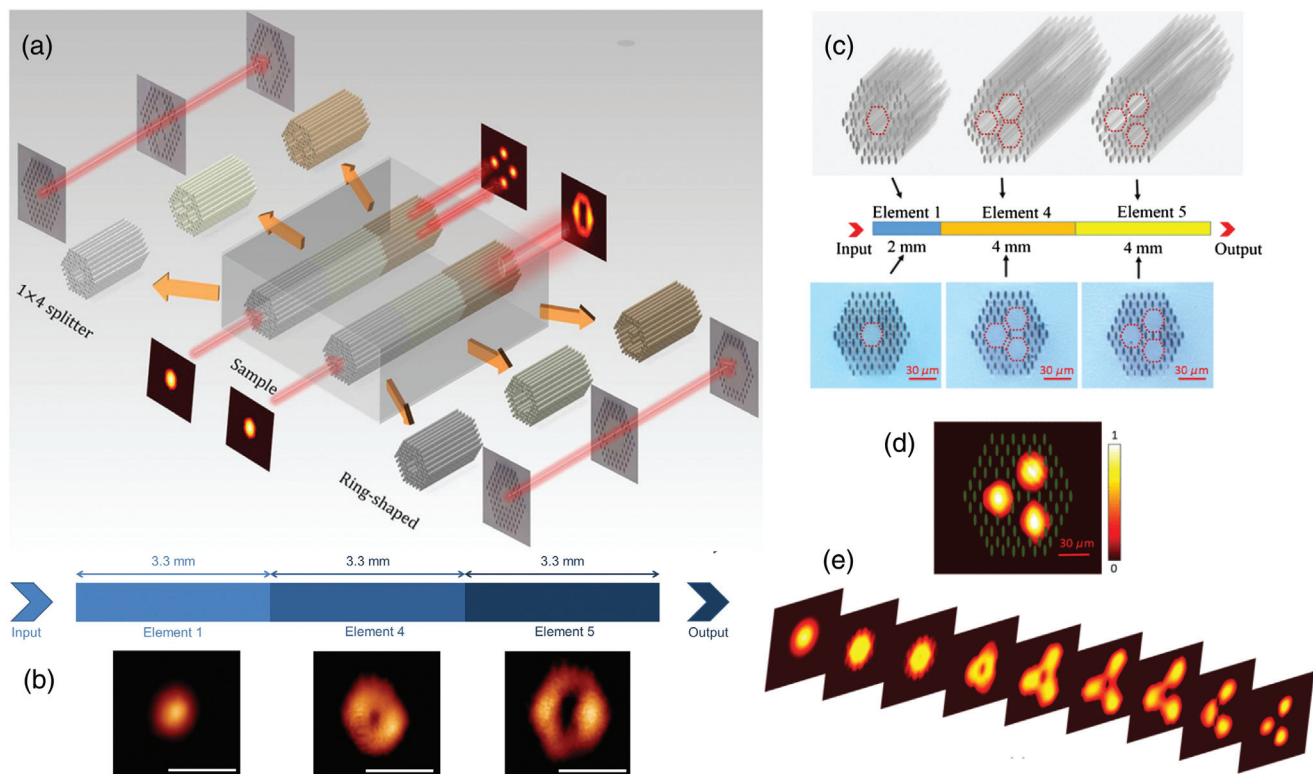


Fig. 9 (a) Schematic illustration of 1×4 beam splitting and ring-shaped transformation based on photonic-lattice-like structures. Image (a) is reproduced from Ref. 102. (b) Measured evolution of ring-shaped transformation in a Nd:YAG crystal. The scale bar is $50 \mu\text{m}$. (c) Prototype design and microscope images of 1×3 beam splitters in a LiNbO₃ crystal, (d) measured and (e) simulated modal profile. Images (c)–(e) are reprinted with permission from Ref. 145, © 2016 IEEE.

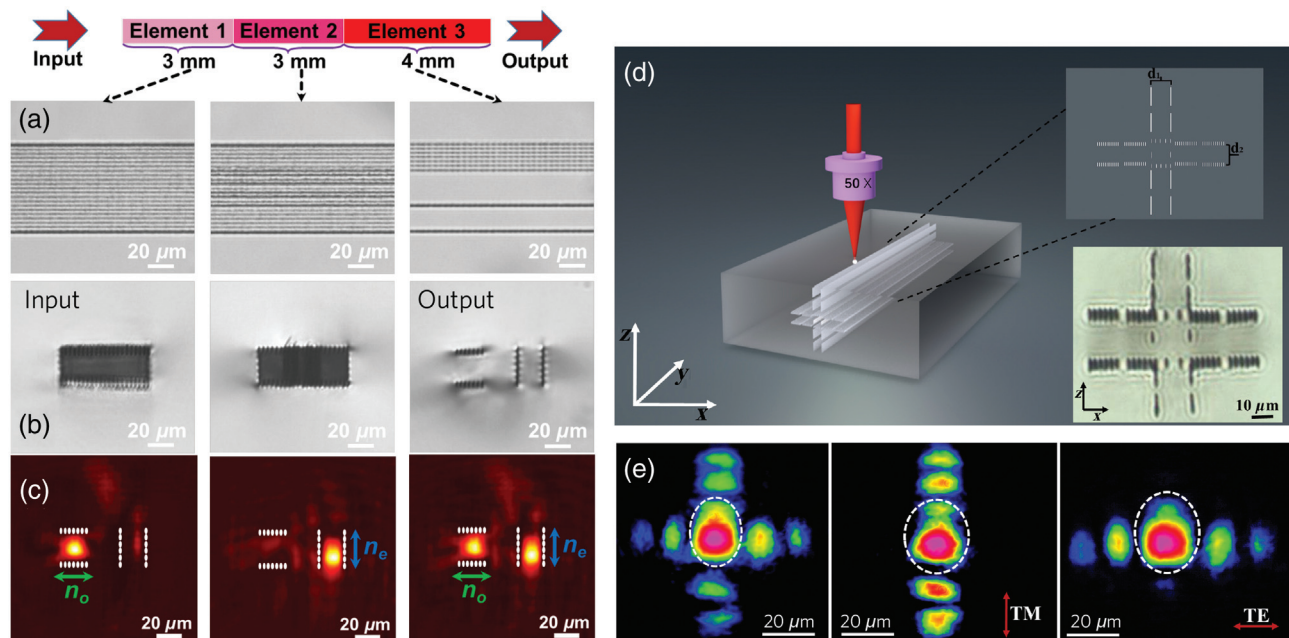


Fig. 10 (a) Microscope images in top view, (b) end-face of polarization beam splitters, and (c) modal profiles along n_o , n_e , and circular polarizations, respectively. Images (a)–(c) are reprinted with permission from Ref. 184, © 2020 IEEE. (d) Schematic plot and microscope images of 3D polarizer. (e) Modal profiles at different polarizations. Images (d) and (e) are reprinted with permission from Ref. 167, © 2020 IEEE.

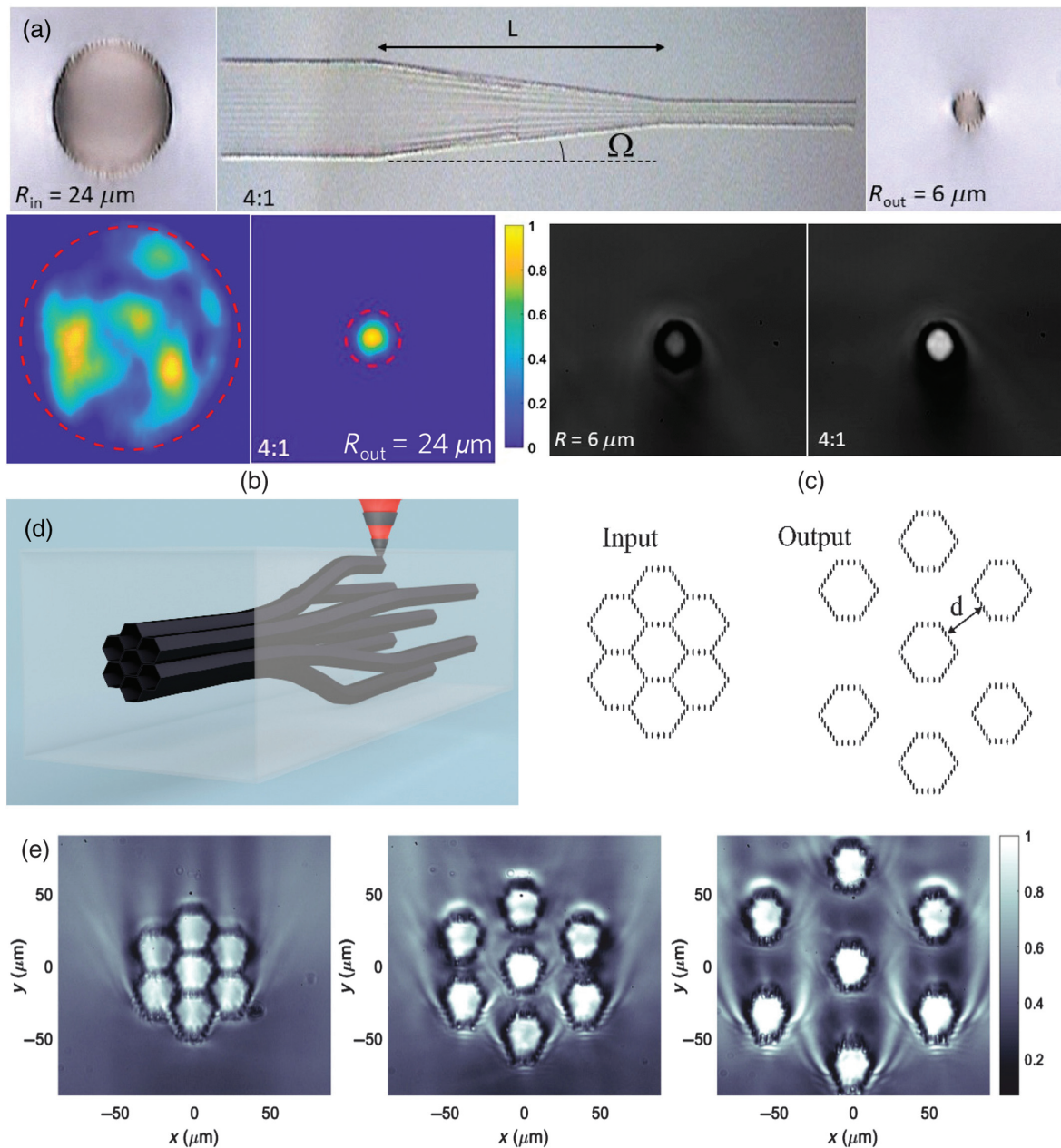


Fig. 11 (a) Microscopic pictures of a tapered cladding waveguide in a Nd:YAG crystal and (b) modal profiles at the input radii of $24\ \mu\text{m}$ and output of $6\ \mu\text{m}$, respectively. (c) Modal profiles of a straight and tapered waveguide at the same output radii from an incident LED light, reproduced with permission from Ref. 185, CC-BY. (d) Prototype of depressed-cladding 3D waveguide arrays. (e) Optical micrographs at the output face for different separations between the central and adjacent waveguides. Images (d) and (e) are reprinted with permission from Ref. 186, © 2017 IEEE.

radius, allowing highly multimodal coupling into single-mode waveguides, as depicted in Fig. 11(b). It is particularly interesting for waveguide laser systems, which reaches a higher coupling efficiency of the pump beam and enhanced intensity of the output power compared to a straight waveguide with the same radius. In addition, such a 3D design offers an efficient manner for modal formatting or light concentration [Fig. 11(c)]. Another promising scheme is the 3D waveguide array,³⁷ which has a potential application in 3D photonic devices, such as

photonic lanterns. Figure 11(d) illustrates the prototype of 3D waveguide arrays in Nd:YAG crystals,¹⁸⁶ which are composed of seven adjacent hexagonal cladding structures at the input face and have each waveguide separated radially from the center at a certain distance at the output face, as shown in Fig. 11(e). This design shows a good wave guiding for both the visible and near-infrared bands. The directional coupler is the basic building block for integrated classical and quantum photonic circuits.⁴⁵ Figure 12(a) presents the 3D design of a 3×3

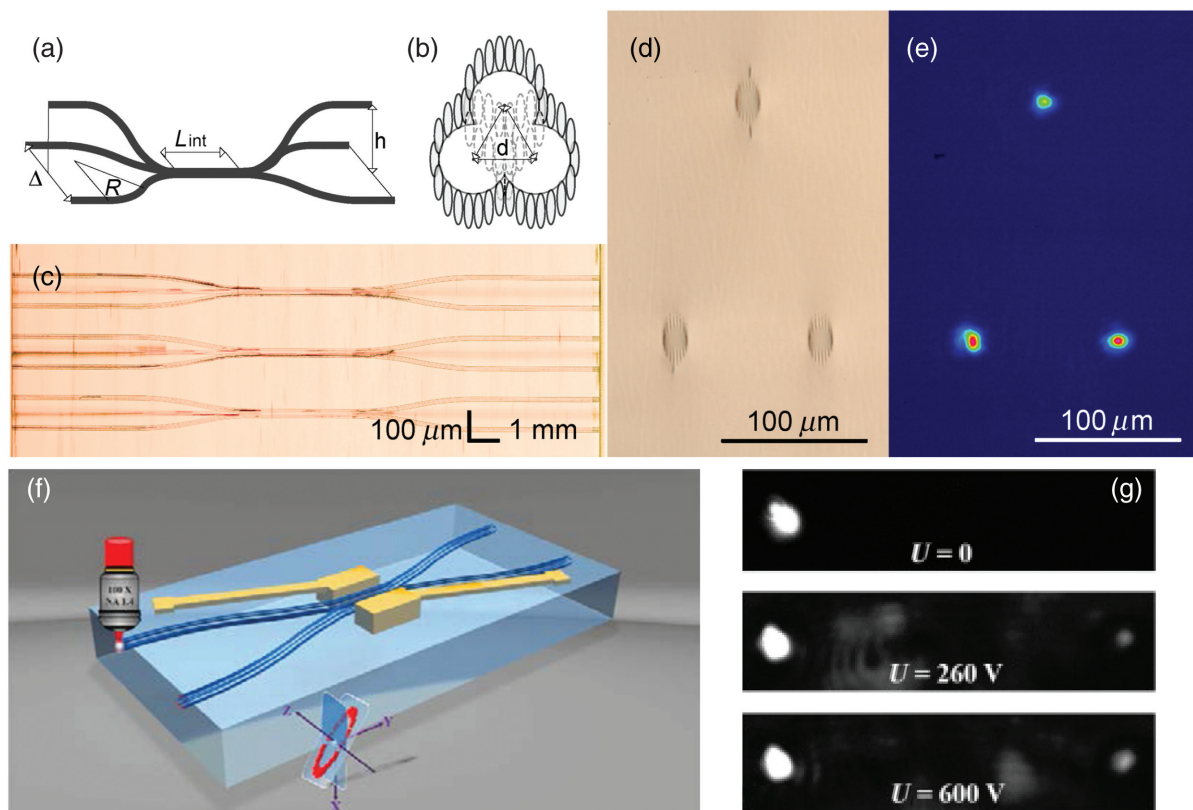


Fig. 12 (a), (b) Geometry and cross-section design in the interaction region of the 3×3 directional coupler in a $\text{Tm}^{3+}:\text{YAG}$ crystal. (c), (d) Top and output view microscope images and (e) output intensity distribution. Images (a)–(e) are reproduced with permission from Ref. 187, CC-BY. (f) Schematic plot of 2×2 directional coupler integrated with 3D microelectrodes in a LiNbO_3 crystal. (g) Output intensity profiles with different voltages. Images (f) and (g) are reprinted with permission from Ref. 118, © 2019 CLP.

directional coupler based on a depressed-cladding waveguide in a $\text{Tm}^{3+}:\text{YAG}$ crystal¹⁸⁷ in which several cladding tracks are removed to form the coupling region [Fig. 12(b)]. The top and cross-section view microscope images are shown in Figs. 12(c) and 12(d). Single-mode guidance with an output splitting ratio of 19:40:41 at 810 nm was demonstrated in this work [Fig. 12(e)], indicating the potential applications in quantum storage devices. By integrating embedded 3D electrodes on both sides of the coupling region of the buried directional coupler in a LiNbO_3 crystal,¹¹⁸ as shown in Fig. 12(f), the efficiency of electro-optic modulation was finely improved [Fig. 12(g)].

4 Selected Applications

In recent years, direct femtosecond laser writing of dielectric crystals enables various waveguiding structures, offering an ideal platform for a broad range of photonic applications by considering the bulk properties of the materials used. In this section, selected advances in recent applications of laser-written crystal-line waveguides are overviewed.

4.1 Waveguide Lasers

Benefitting from the high optical confinement, femtosecond laser-written waveguides provide efficient miniaturized laser sources that cover the spectral range from visible to MIR (530 nm to 4 μm) with many advantages, such as reduced lasing

thresholds, enhanced slope efficiencies, compact size, nondiffraction beams, and multifunctional integration.⁷ Up to now, some notable improvements have been achieved, depending on the active ions and the mechanisms for lasing. Table 3 summarizes the reported results for waveguide lasers emitting at different wavelengths based on various cavity designs fabricated in femtosecond laser writing.

Laser emission at visible wavelengths mainly relies on Pr^{3+} -doped gain media, such as $\text{Pr}:\text{SrAl}_{12}\text{O}_{19}$, $\text{Pr},\text{Mg}:\text{SrAl}_{12}\text{O}_{19}$, and $\text{Pr}:\text{LiYF}_4$ crystals. For instance, using well-established dual-line structure, a green waveguide laser operating at 525.3 nm was first demonstrated in $\text{Pr},\text{Mg}:\text{SrAl}_{12}\text{O}_{19}$ crystals.¹⁹⁰ Since then, based on the rhombic cladding geometry, orange and deep red laser oscillations at 604 and 720 nm were produced in $\text{Pr}:\text{LiYF}_4$ crystal.¹⁹¹ Moreover, laser-written active dual-line waveguides in $\text{Ti}:\text{sapphire}$ have offered tunable laser operating from 700 to 870 nm.¹⁹² In addition, with the same dual-line configuration in $\text{Ti}:\text{sapphire}$, Grivas et al.¹⁵² reported a femtosecond continuous-wave mode-locked (CWML) laser emitting at 790 nm. The generated waveguide laser was operated at pulses of 41.4 fs with a fundamental repetition rate up to 21.25 GHz.

Near-infrared spectral range is the most common laser emission band, which is gained by writing an active waveguide into well-known Nd^{3+} - and Yb^{3+} -doped crystals. Beyond a normal straight laser cavity, it is also possible to achieve efficient optical

Table 3 Summary of reported results for waveguide lasers emitting at different wavelengths based on various laser-cavity designs.

Wavelength band	Gain media	Working wavelength (nm)	Cavity configuration	Operation regime	Laser performance			Ref.	
					Lasing threshold (mW)	Max. output power (mW)	Slope efficiency		
Visible	Nd:YCOB	531	Cladding	CW	5	0.1	—	161	
		532	Dual line	CW	—	0.032	—	188	
	Pr: SrAl ₁₂ O ₁₉	634.5	Dual line	CW	190	28.1	8%	189	
		Pr, Mg: SrAl ₁₂ O ₁₉	525.3	Dual line	CW	1088	36		190
	644				516	1065	37%		
	724.9				885	504	25%		
	Pr: LiYF ₄	604	Rhombic cladding	Rhombic cladding	CW	360	25	5.6%	191
		720				243	12	2.0%	
	Ti: sapphire	700 to 870	Dual line	CW	84	143	23.5%	192	
			Dual line	CWML (21.25 GHz)	1160.1	87.48	—	152	
Near-infrared	Yb, Na: CaF ₂	1013.9 and 1027.9	Cladding	CW and Q-switched	152.2	26.6	10%	193	
		Yb: YAG	1030	S-curved dual line	CW	141	1 W	79%	194
			Y-branch dual line	CW	271	2.29 W	52%	97	
			Dual line	Q-switched	102	5.6 W	74%	195	
			Dual line	QML (2 GHz)	1800	322	11.3%	196	
			Double cladding	CW	401.7	45.8	38%	172	
	Yb: KLuW	1040	Surface cladding	Q-switched	491	680	61%	197	
			Nd: YAG	1064	Annular ring shaped	CW	191	84	20%
		Ear-like cladding		CW and Q-switched	10	327	34.4%	168	
		Cladding		Q-switched	287	102.3	11.9%	198	
				QML (8.8 GHz)	74	127	26%	199	
				Cladding	CWML (9.8 GHz)	—	530	—	200
	Nd: YVO ₄	1061.58 and 1064.18	1064	Y-branch cladding	CW	231	172	22.4%	180
				1 × 2 splitters	CW	90	333	34%	101
				1 × 4 splitters		90	217	22%	
				Ring shaped	CW and Q-switched	148	224	22%	100
				Cladding	CW	10.3 W	3.4 W	36%	139
		1064			Q-switched	57.4	275	37%	201
					QML (6.5 GHz)	65	424	56%	202
					CWML (6.5 GHz)	19.3	259	30.6%	203
			Double cladding	Q-switched	59	397	46%	169	
			Optical-lattice like	Q-switched	—	85	20%	140	
Nd: YAP	1064 and 1079	Cladding	CW	243	199.8	33.4%	158		
		S-curved cladding	QML (7.9 GHz)	196	77	14.1	159		
		S-curved optical-lattice like		228	57	10.69			
Nd: GdVO ₄	1072 and 1079	Optical-lattice like	CW	384.5	101.3	30.9	75		
		1063.6	Dual line	CW	52	256	70%	141	
		1064.5	Cladding	CW and Q-switched	178	570	68%	142	
Nd: GGG	1061	Dual line	CW	29	11	25%	134		
		1063	Cladding	CW	270	209	44.4%	135	
Nd: LuVO ₄	1066.4	Dual line	CW	98	30	14%	204		
Nd: KGW	1065	Dual line	CW	141	33	52.3%	163		
	1067	Cladding	CW	120	198.5	39.4%	164		

Wavelength band	Gain media	Working wavelength (nm)	Cavity configuration	Operation regime	Laser performance			Ref.	
					Lasing threshold (mW)	Max. output power (mW)	Slope efficiency		
MIR	Tm:KLu (WO ₄) ₂	1847.4	Surface cladding	CW	52	171.1	37.8%	205	
		1846.8		Q-switched	500	150	34.6%		
		1849.6	Cladding	CW	45	247	48.7%	206	
		1844.8		Q-switched	—	24.9	9.3%		
		1847	Optical-lattice like	CW	21	46	9.9%	207	
			1841 to 1848	Y-branch cladding	CW	280	460	40.6%	208
		Tm : YAG	1943.5	Cladding	QML (7.8 GHz)	665	6.5	2%	209
		Ho:KGd (WO ₄) ₂	2055	Cladding	CW	180	212	67.3%	210
		Tm:MgWO ₄	2080	Surface cladding	CW	120	132	38.9%	211
		Ho : YAG	2091	Cladding	QML (5.9 GHz)	—	170	6.8%	212
			2096		CW	100	1775	16%	213
		Cr : ZnS	2333	Cladding	CW	450	101	20%	214
		Cr : ZnSe	2522	Cladding	CW	—	5200	41%	215
	Fe : ZnSe	4070	Cladding	CW	—	995	58%		

feedback for lasing oscillation in curved and branched cavity geometry owing to an enhanced optical gain of a waveguide laser. For example, in femtosecond laser-written 3D structures, compact waveguide lasers at 1 μm have been proposed in different functionalities, such as S-curved,^{159,194} Y-branches,^{97,180,208,216} 1 \times 2 and 1 \times 4 beam splitting,¹⁰¹ ring-shaped beam transformation,^{100,132} and optical-lattice-like lasing⁷⁵ [Figs. 13(a)–13(e)]. Laser performance in these reports is comparable to that of straight waveguides, suggesting a superior flexibility for prototyping laser sources using femtosecond laser writing. By further integrating a suitable absorber, passively Q-switched lasing has been realized. For example, using a high gain Yb:YAG channel waveguide, an efficient Q-switched laser has shown a slope efficiency of 74% with an average power up to 5.6 W in a single-pass optical pumping without any reflective mirrors.¹⁹⁵ Most recently, mode-locked waveguide lasers operating at multi-gigahertz (GHz) regimes have attracted increasing interest. For instance, 6.5 GHz Q-switched mode-locked (QML) lasers have been demonstrated in a femtosecond laser-written Nd:YVO₄ cladding waveguide using 2D materials as the saturable absorber.²⁰² Another recent sample is Nd:YAP crystal : 31.6-GHz QML lasers have been demonstrated in an S-curved waveguide,¹⁵⁹ as shown in Fig. 13(f). In particular, both single- and dual-wavelength (1064 and 1079 nm) lasing can be achieved by tuning the pump polarization [Figs. 13(g) and 13(h)].

Recently, MIR ($\sim 2 \mu\text{m}$) waveguide lasers, typically obtained from Tm³⁺- and Ho³⁺-doped materials, have attracted much attention from researchers. For a gain medium of Tm:KLu(WO₄)₂, efficient waveguide lasers at $\sim 1.8 \mu\text{m}$ operating both in the CW and Q-switched regimes have been reported by Kifle et al. in various laser-written waveguide structures, such as Y-branch splitters,²⁰⁸ surface cladding,²⁰⁵ buried circular cladding,^{166,206} and photonic-lattice-like cladding prototypes.²⁰⁷ In addition, their group has demonstrated a low-loss ($<0.1 \text{ dB/cm}$)

waveguide laser in a Tm³⁺:MgWO₄ crystal at $\sim 2.02 \mu\text{m}$ with a slope efficiency of 64.4%.²¹¹ Also, in the multi-GHz operation regime, 5.9- and 7.8-GHz QML waveguide lasers at $\sim 2 \mu\text{m}$ have been successfully achieved in Ho³⁺:YAG²¹² and Tm : YAG,²⁰⁹ respectively. Cr:ZnS is another host medium for MIR laser generation. Based on the cladding channel waveguide, an efficient 2333-nm waveguide laser with an output power of 101 mW has been reported by Macdonald et al.²¹⁴ Moreover, waveguides written in Fe:ZnSe crystal have shown a slope efficiency of 58%, offering a working wavelength up to 4070 nm.²¹⁵

4.2 Frequency Converters

Frequency conversion displayed in femtosecond laser-written waveguides coming from second and higher-order nonlinearities offers an efficient manner to access special wavelengths. Similar to waveguide lasers, frequency converters in waveguide structures provide enhanced conversion efficiencies compared to bulk material, as a result of strong optical confinement and good overlap between the fundamental and nonlinear polarization waves over the whole interaction length. Femtosecond laser-written waveguides allow for nonlinear optical conversion, mainly SHG, at a wavelength range between ~ 400 and 790 nm.

One of the most well-studied nonlinear crystals is LiNbO₃, since the work of Burghoff et al.²¹⁷ in 2007, which enabled efficient frequency doubling of 1064 nm with a conversion efficiency of 49% in laser-written dual-line waveguides. In 2021, using the regime of birefringent phase matching (BPM), waveguide geometries of multiline, vertical dual line, and depressed cladding have also been reported in LiNbO₃ crystals for SHG of 1064 nm,²¹⁸ whereas BPM is difficult to realize in some crystals with weak birefringence. In recent years, LiQPM grating structures have attracted a particular interest in frequency doubling, beginning from the first proof in 2013 reported by Thomas

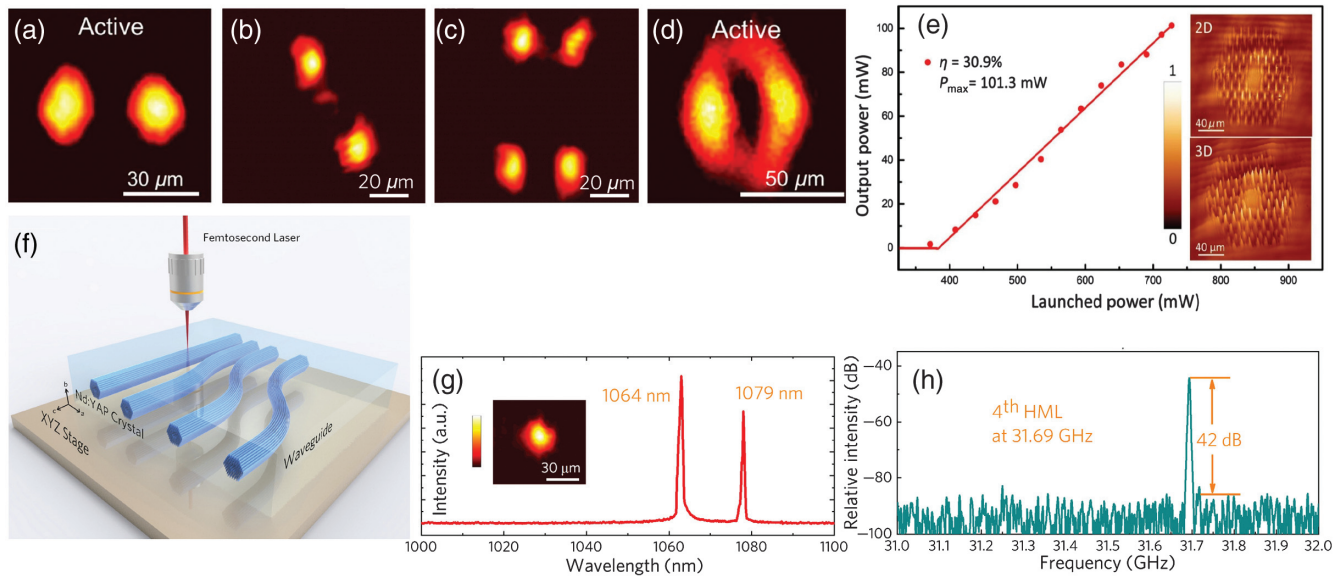


Fig. 13 Complex waveguide laser modal profiles at $1\ \mu\text{m}$: (a), (b) Y-branches, (c) 1×4 -branch, (d) ring-shaped transformation, and (e) optical-lattice-like. Images (a) and (d) are reproduced with permission from Ref. 100. Images (b) and (c) are reprinted with permission from Ref. 101, © 2016 IEEE. Image (e) is reprinted with permission from Ref. 75, © 2016 OSA. (f) Schematic illustration of the fabrication process of an S-curved waveguide, (g) laser spectra of dual-wavelengths at 1064 and 1079 nm, and (h) RF spectrum of modelocking at 31.69 GHz. Images (f)–(h) are reprinted with permission from Ref. 159, © 2020 IEEE.

et al.¹¹⁹ though with a poor conversion efficiency. Later in 2015, an increased conversion efficiency of 5.72% was reported by Kroesen et al., which introduced QPM grating structure inside a depressed-cladding waveguide.¹²⁷ In 2020, a waveguide-integrated 3D QPM grating scheme was proposed, as shown in Fig. 14(a), thus multiwavelength SHGs of 1065.3, 1064, 1061.6, and 1060.5 nm radiation have been demonstrated⁴³ [Fig. 14(b)]. Using this technique, Wei et al.¹²¹ produced 3D NPC in LiNbO₃ crystals, as well as realizing an SHG of 829 nm with an enhanced second-harmonic power compared with the on-engineered area. Figure 14(c) illustrates the setup of the SHG process using LiQPM structures in quartz crystal, and a deep-ultraviolet generation of 177.3 nm has been achieved with a high efficiency of 1.07%¹²⁴ [Fig. 14(d)]. In addition, based on a type I single-line waveguide, the green-up conversion at 550 and 528 nm and NIR emission at 1550 nm have been reported in Er:MgO:LiNbO₃ crystals.¹⁴⁴ Moreover, spontaneous parametric down-conversion from 780 to 1560 nm has also been demonstrated in laser-written multiline waveguides of PPLN crystal, giving access to photon entanglement in quantum technologies.⁴⁹

Frequency doubling down to ultraviolet bands has also attracted much attention. BiB₃O₆ cladding waveguide is a good sample, and a violet SHG at 400 nm has been realized.²¹⁹ LiTaO₃ crystal is another promising material owing to its shorter cut-off wavelength (0.28 μm) and high optical damage threshold. In 2019, a second harmonic power up to 8.5 W was realized in laser-written circular-cladding waveguides under a CW pump at 1050 nm in a periodically poled MgO:LiTaO₃ (PPMgSLT) crystal.²²⁰ Figure 14(e) depicts the schematic plot of femtosecond laser direct writing cladding waveguides in fan-out PPMgSLT crystals, corresponding to different poling periods. Temperature-tuned SHG for different QPM periods has been

demonstrated in Fig. 14(f) with a comparable normalized conversion efficiency of 3.55% $\text{W}^{-1}\text{cm}^{-2}$.¹⁴⁸ Later in 2020, a tunable violet SHG over the range of 396 to 401 nm was demonstrated by varying the QPM condition in fan-out PPMgSLT crystals,²²¹ evincing efficient frequency converters based on femtosecond laser-written waveguides.

KTiOPO₄ (KTP) is another excellent nonlinear platform for frequency conversion. In 2007, the laser-written multiline waveguide based on type I modification permitted SHG down to 400 nm in periodically poled:KTiO_pO₄ (PPKTP) crystals.¹⁵⁴ Lately, a further improvement of conversion efficiency (4.6% $\text{W}^{-1}\text{cm}^{-2}$) has been proposed in type II dual-line waveguides in Rb-doped PPKTP crystals.²²² In particular, due to the true 3D micromachining of femtosecond laser writing, optical lattice-like 1×4 beam splitters in KTP crystals have enabled SHG of 1064 nm by BPM.¹⁵⁷ In addition, the optical-lattice-like microstructures offer an efficient manner for beam mode controlling, providing a simple way to realize single-mode frequency doubling at 532 nm with a comparable conversion efficiency.¹⁸³ Table 4 summarizes the latest results for frequency converters based on femtosecond laser-written waveguides.

4.3 Quantum Memories

To date, femtosecond laser direct writing has played an important role in integrated quantum information processing,^{225,226} such as integrated sources of entangled photons,⁴⁹ high-dimensional transmission of quantum twisted light states,⁴⁷ 2D quantum walk,⁴⁶ and the rotated polarization directional coupler.⁴⁵ Quantum memories (QMs) are essential components in large-scale quantum networks, especially in quantum repeaters. Rare-earth ion-doped crystals are promising candidates of integrated

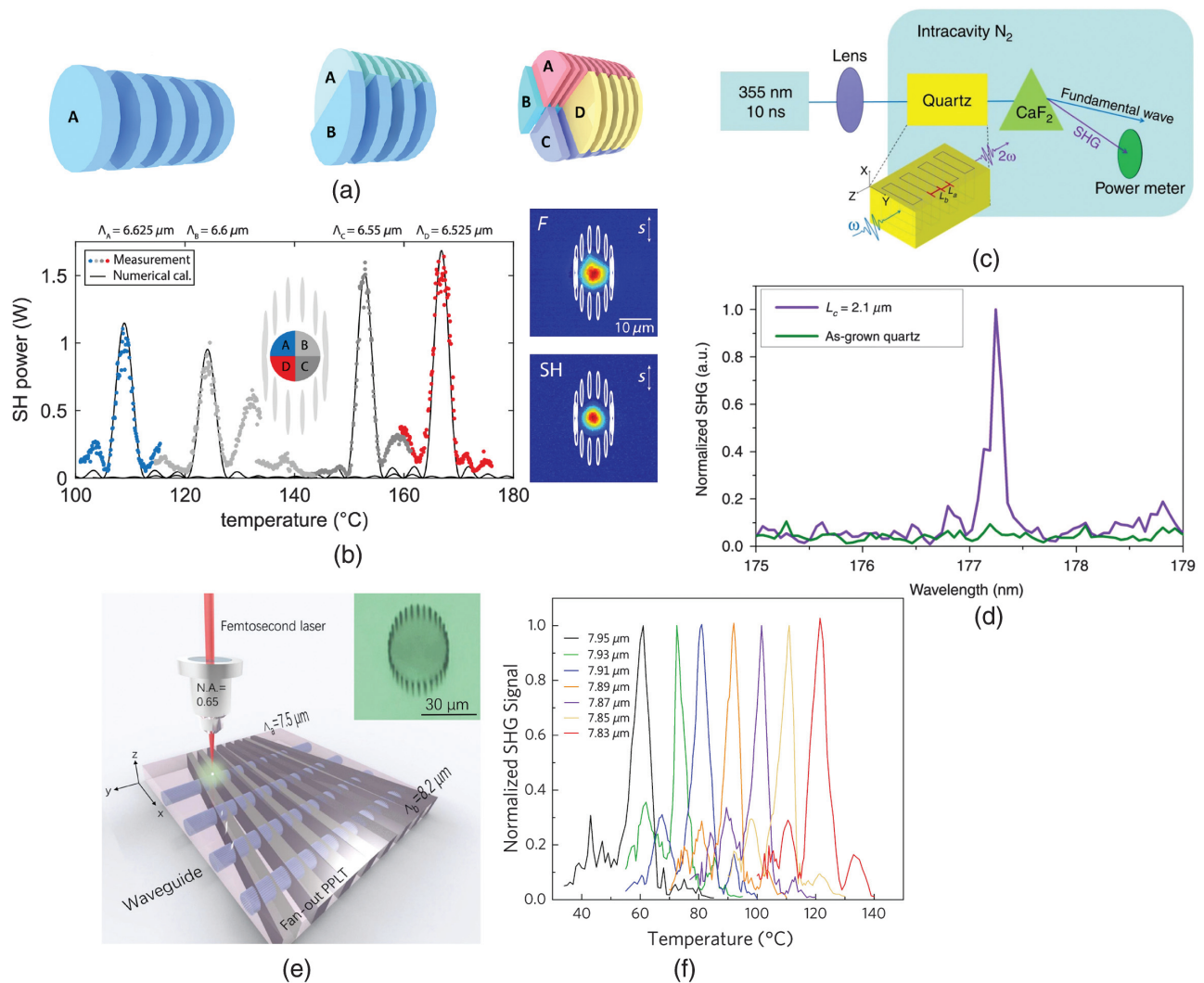


Fig. 14 (a) Waveguide-integrated 3D LIQPM scheme, one period, two periods, and four periods in a LiNbO₃ crystal. (b) Simultaneous SHG of four wavelengths and the fundamental and second harmonic modal profiles of a single period. Images (a) and (b) are reprinted with permission from Ref. 43, © 2020 OSA. (c) Experimental setup of the ultraviolet SHG process using LIQPM structure in a quartz crystal and (d) the SHG response signal of 177.3 nm. Images (c) and (d) are reproduced with permission from Ref. 124, CC-BY. (e) Schematic diagram of femtosecond laser-written cladding waveguide in a fan-out PPLT crystal and (f) temperature tuning curves of seven waveguides with different poling periods. Images (e) and (f) are reprinted with permission from Ref. 148, © 2019 OSA.

QMs due to their large bandwidth, high multimode capacity, and high storage fidelity.²²⁷ Two approaches have been employed to manufacture integrated QMs in rare-earth ion-doped crystals, such as Ti-indiffusion in LiNbO₃¹⁵ and focused-ion-beam milling.²²⁸ Nevertheless, the storage time and efficiencies in these two systems are significantly reduced. In recent years, femtosecond laser direct writing has allowed channel waveguides with improved capabilities, where the active ions are directly coupled to the light, rather than via evanescent coupling.^{52,225,229} As a result, spin-wave storage with an extended lifetime, which enables long-term storage and on-demand read-out, has been achieved in waveguide-integrated QMs.^{50,51,53,230} A great amount of effort has been devoted to this topic, mainly focused on the geometry of type I, type II, and ridge waveguides.

The first proof of QMs based on laser-written dual-line waveguides was reported in Pr³⁺:Y₂SiO₅ crystals, where the interaction with the active ions is increased by a factor of 6 compared to bulk, and demonstrated a spin-wave optical memory,²²⁹ as shown in Figs. 15(a)–15(d). Later in 2020, using the type II waveguides in Eu³⁺:Y₂SiO₅ crystals, on-demand light storage was demonstrated via the spin-wave atomic frequency comb (AFC) and the storage fidelity was quantitatively characterized for the first time.⁵⁰ Compared to type II waveguides, the fabrication of type I waveguides in crystalline substrates is still a very challenging task, as it requires a very narrow writing window. In 2018, Seri et al.⁵² produced high-quality type I waveguides in Pr³⁺:Y₂SiO₅ crystals, demonstrating efficient storage times 100 times longer than previous demonstrations [Figs. 15(e)–15(g)]. In the next year, their group reported more than 130

Table 4 Summary of latest results for frequency converters in femtosecond laser-written waveguides.

Crystal	Waveguide configuration	Laser regime	λ_{ω} (nm)	$\lambda_{2\omega}$ (nm)	SHG configuration	P_{out} ($W^{-1} cm^{-2}$)	Norm. efficiency ($W^{-1} cm^{-2}$)	Ref.
BBO	Cladding	CW	800	400	BPM	1.05 mW	0.98%	219
PPKTP	Type I (multiscan)	CW	800	400	QPM	51 μ W	0.02%	154
PPSLT	Cladding	CW	800	396 to 401 (tunable)	QPM	0.37 mW	0.39%	221
PPKTP	Dual line	CW	943.18	471.59	QPM	76 mW	4.6%	222
LiNbO ₃	Cladding	Pulsed	1030	515	BPM	—	—	223
	Type I (multiline)	Pulsed	1064	532	BPM	12.45 W (peak)	0.27%	218
	Dual line	Pulsed	1064	532	BPM	4.95 W (peak)	0.14%	
	Cladding	Pulsed	1064	532	BPM	40.40 mW (peak)	0.87%	
	Cladding	Pulsed	1064	532	LiQPM	25.1 W (peak)	0.0637%	127
	Cladding	Pulsed	1065.3, 1064, 1061.6, and 1060.5	532.65, 532, 530.8, and 530.25	LiQPM	1.33 W (peak)	0.64% ($P_{2\omega}/P_{\omega}$)	43
PPMgSLT	Cladding	CW	1064	532	QPM	17.3 μ W	0.74%	224
	Cladding	CW	1050	525	QPM	8.5 W	0.16%	220
	Cladding	CW	1064	532	QPM	14.87 mW	3.55%	148
	Cladding	Pulsed	1064	532	QPM	153 W (peak)	54.3% ($P_{2\omega}/P_{\omega}$)	
KTiOPO ₄	Optical-lattice like (1 \times 4 splitters)	CW	1064	532	BPM	0.65 mW	1.5%	157
	Optical-lattice like (straight)	CW	1064	532	BPM	0.67 mW	0.87%	
	Hybrid optical-lattice	CW	1064	532	BPM	0.8 mW	1.1%	121

individual storage modes in laser-written type I waveguides in $Pr^{3+}:Y_2SiO_5$ crystals, among the temporal, spectral, and spatial domains, demonstrating the capability of both time and frequency multiplexing single photon.⁵³ Such high multimodality will be necessary to realize an efficient quantum repeater scheme.

Considering that the channel waveguides (type I and type II) are fabricated at a depth beneath the crystal surface, ridge waveguide-based QMs are easily integrated with other on-chip devices, allowing for constructing large-scale quantum networks. In 2020, a laser-written ridge waveguide was successfully fabricated in an $Eu^{3+}:Y_2SiO_5$ crystal, in which the properties of the Eu^{3+} ions were well-preserved.²³⁰ The spin-wave AFC storage was implemented, confirming high-interference visibility [Figs. 16(a)–16(d)]. In 2021, their group achieved a better fabrication parameter, realizing 40% end-to-end device efficiency, while the typical coupling efficiency is 10% in LiNbO₃ waveguide memory. Combined with on-chip electrodes, a high storage fidelity of $99.3\% \pm 0.2\%$ and on-demand storage of time-bin qubits were demonstrated,⁵¹ far beyond that value of the classical measure-and-prepare strategy.

4.4 On-Chip Devices

Thin-film lithium-niobate-on-insulator (LNOI) has attracted significant interest in the field of PIC for its favorable low-loss limit

and electro-optic coefficients.^{17,18,231} With the recent revolution in fabrication techniques of on-chip photonics, great progress has been sparked in producing ultrahigh performance building blocks, such as waveguides^{16,232} and microcavities,²³³ for scalable and high-density LNOI PICs. In 2015, Lin et al.²³⁴ first reported on the fabrication of high- Q LiNbO₃ microresonators by femtosecond laser direct writing, followed by focused ion beam milling. In 2018, their group proposed an efficient LNOI fabrication technique termed photolithography-assisted chemomechanical etching (PLACE).²³⁵ Figures 17(a)–17(e) schematically illustrate the process of PLACE, including mainly five steps. Notably, femtosecond laser micromachining is used to pattern the Cr thin film into a waveguide- or circular disk-shaped mask with the advantages of high scanning speed and high quality. The key in this step is to carefully choose a peak intensity of the femtosecond laser, in which the Cr film can be completely removed without damaging the underneath LNOI, since the ablation threshold of LNOI is significantly higher than that of the metal Cr.²³³ In addition, chemomechanical polishing (CMP) is employed to selectively etch smooth sidewalls capable of low scattering loss. Using this technique, a 10-cm-long LNOI waveguide with an ultralow loss of 0.027 dB/cm²³² [Figs. 17(f)–17(h)] and an LNOI microresonator with an ultrahigh Q factor of 4.7×10^7 by improving the surface roughness of ~ 0.115 nm²³³ [Fig. 17(i)] have been demonstrated. In addition, by adding a layer of Ta₂O₅ film on the waveguides,

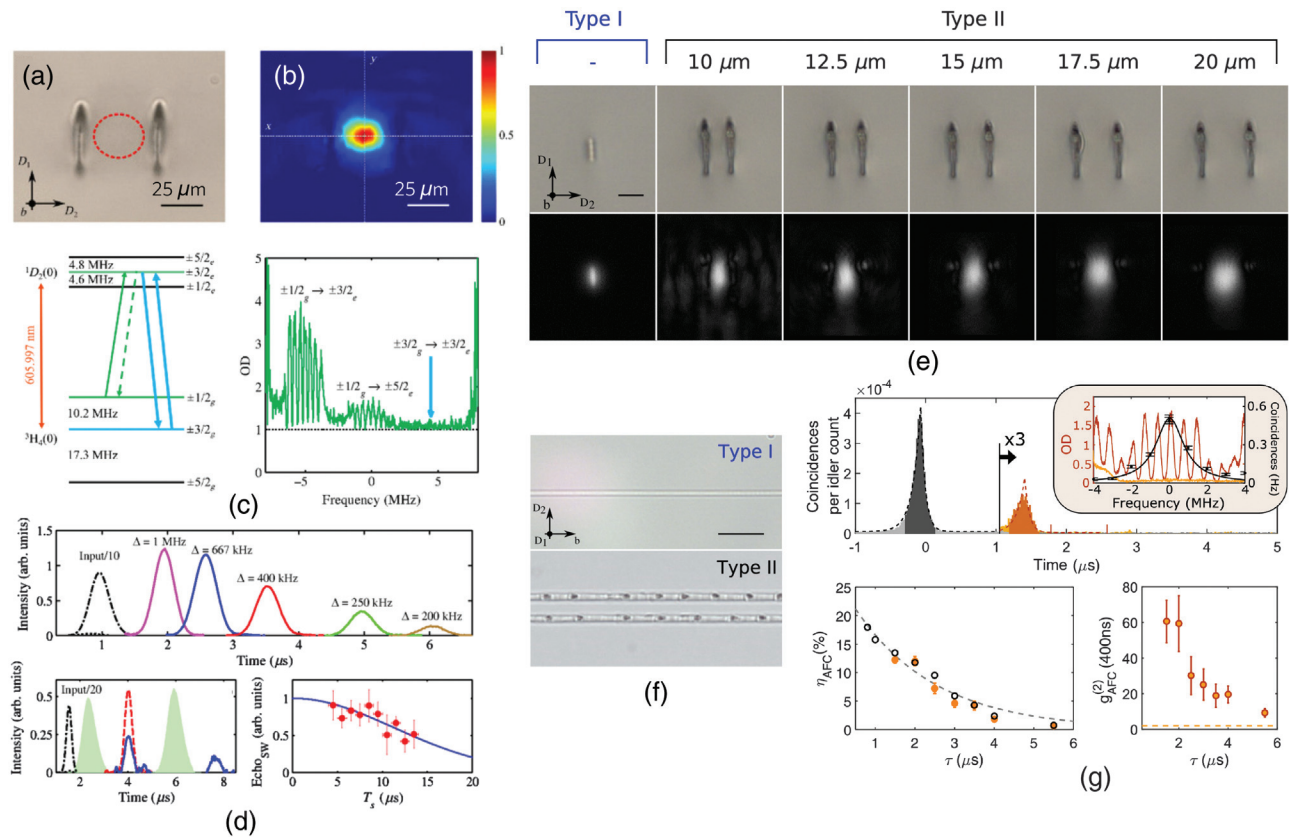


Fig. 15 (a), (b) Microscope image and near-field intensity profile of a type-II waveguide in a $\text{Pr}^{3+}:\text{Y}_2\text{SiO}_5$ crystal. (c) Energy-level structure of 3H_4 ground and 1D_2 excited manifolds. (d) Light-storage experiments using the AFC protocol. Images (a)–(d) are reproduced with permission from Ref. 229, © 2016 American Physical Society (APS). (e), (f) End-face and top-view microscope images of type I and type II waveguides and modal profiles in $\text{Pr}^{3+}:\text{Y}_2\text{SiO}_5$ crystal, respectively. (g) Time-resolved histogram for signal photons, internal storage efficiency η_{AFC} at different storage times, and cross-correlation values between idler photons and stored signal photons. Images (e)–(g) are reprinted with permission from Ref. 52, © 2018 Optica.

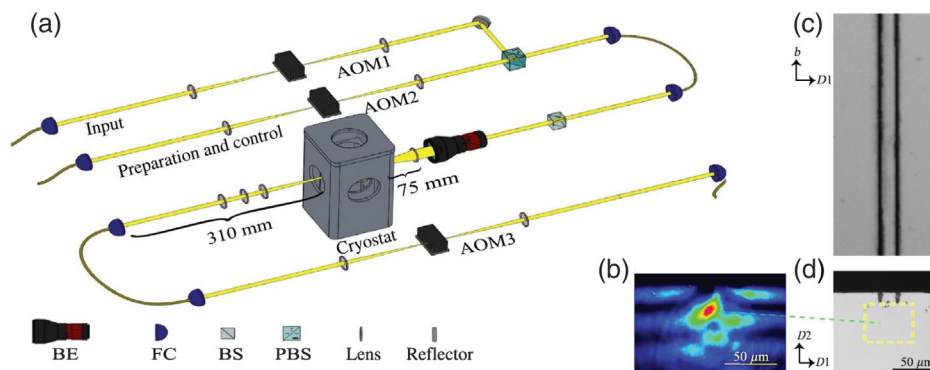


Fig. 16 (a) Experimental setup of coherent optical memory based on an on-chip waveguide. (b) Guided mode intensity distribution of laser-written ridge waveguides in an $\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$ crystal. (c), (d) Top and front view microscope images. Images are reproduced with permission from Ref. 230, © 2020 APS.

a single-mode LNOI waveguide with a propagation loss of 0.029 dB/cm at 1550 nm has also been discussed.²³⁶ Based on these ultralow-loss waveguides, Wang et al.²³⁷ demonstrated the fabrication of LNOI beam splitters and Mach–Zehnder

interferometers. Later in 2020, Zhou et al.²³⁶ successfully designed and fabricated optical true delay lines on the LNOI chip, realizing a relative time delay of 2.285 ns. Various waveguides with lengths from ~ 10 to ~ 100 cm have been fabricated with

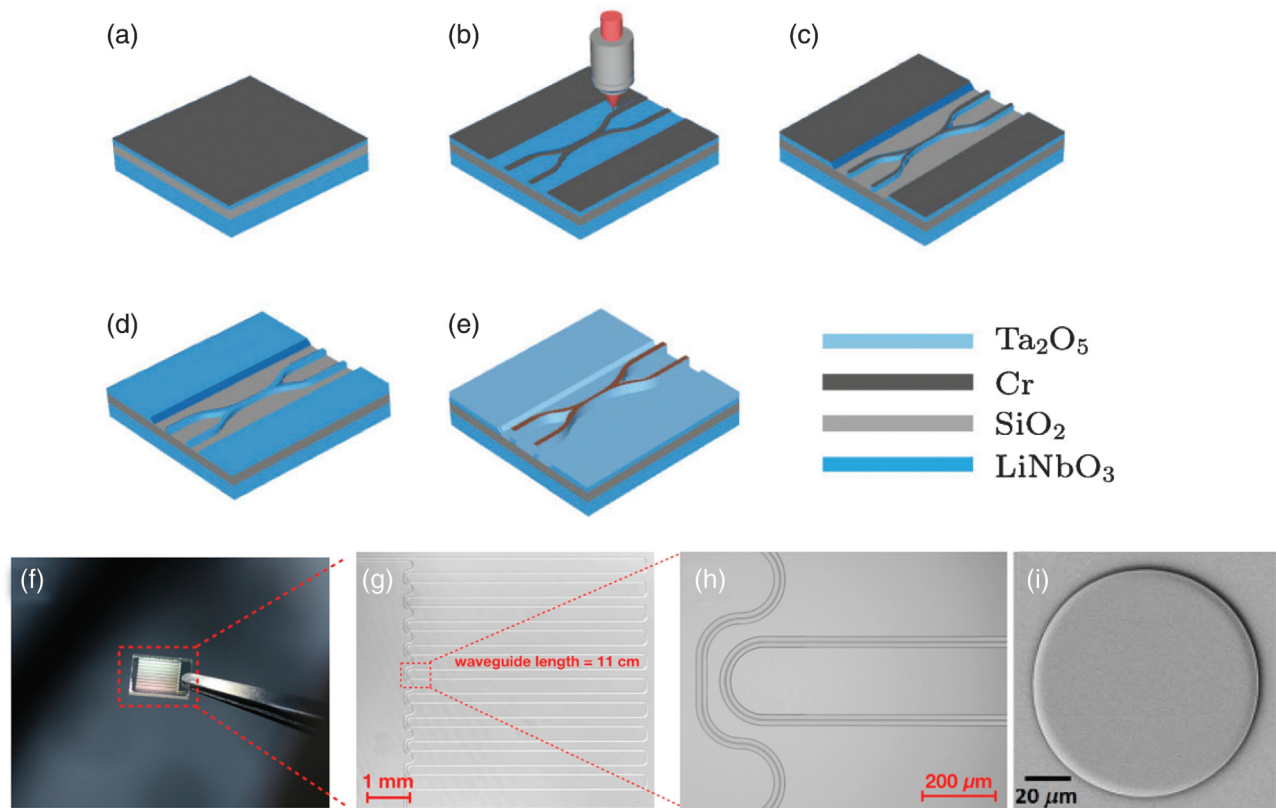


Fig. 17 Schematic illustration of the PLACE fabrication process. (a) Cr thin-film deposition, (b) Cr patterning, (c) CMP, (d) chemical wet etching, and (e) coating Ta_2O_5 film. Images (a)–(e) are reproduced with permission from Ref. 236, © 2020 Chinese Physical Society (CPL). (f) Camera photo of the 11-cm-long LNOI waveguide, (g) microscope image, and (h) enlarged image. Images (f)–(h) are reproduced with permission from Ref. 232, CC-BY. (i) SEM image of LNOI microdisk. Image (i) is reproduced with permission from Ref. 233, CC-BY.

losses below 0.03 dB/cm, providing the large-scale on-chip integration of numerous unit microdevices.

To date, some active components, such as on-chip microlasers and amplifiers, have also played an important role in the monolithic integration of PICs. In 2021, Wang et al.²³⁸ demonstrated a tunable microring laser on Er^{3+} -doped LNOI, enabling a low threshold of 400 μW . In the same year, by integrating the microelectrodes, an electro-optically tunable microring laser has been achieved on an LNOI platform on which the laser wavelength can be tuned with an electro-optic coefficient of 0.33 pm/V.²³⁹ Specifically, their group provided an alternative pumping manner, using an undoped LNOI waveguide directly bonded above the Er^{3+} -doped microring, rather than the tapered fibers. In addition, Zhou et al.²⁴⁰ proposed an on-chip waveguide amplifier on Er^{3+} -doped LNOI, which allows a net gain of 18 dB for a length of 3.6 cm emitting of 1530 nm, indicating an efficient broadband amplification.

5 Summary and Perspectives

Femtosecond laser inscribed crystalline waveguides play increasingly significant roles in modern photonic circuits, offering an attractive platform for multifunctional integrated optical applications. The unique 3D microfabrication of femtosecond lasers is currently driving the success of new devices and

functionalities, such as quantum and on-chip photonics. Recent progress and achievements in this research field are briefly overviewed, whereas a few challenges and potential topics remain to be further investigated.

First, the underlying mechanism of different refractive index modifications induced by femtosecond lasers is not entirely understood, which strongly depends on the parameters of the laser processing system and the physical properties of crystals. For example, an interesting phase transformation from YAG to YAP crystal is observed by Okhrimchuk et al.⁸⁶ under direct laser writing, relating to rapid quenching after overheated crystal melting. Future investigation towards the mechanisms of refractive index modification and the phase-change transitions is planned. Second, a severe limitation of femtosecond laser inscription is related to the relatively lower refractive index contrast than that of bulk, usually on the magnitude of 10^{-3} in crystals. Thus the miniaturization of femtosecond laser-processed photonic devices and many applications are limited. Further work will focus on the formation of waveguides with an enhanced photoinduced refractive index contrast while preserving the morphology and uniformity of laser-induced modifications. Third, waveguide-integrated photonic devices have a significant interest in large-scale on-chip circuits, thus the waveguide performances, including propagation loss, bending radii, and coupling efficiencies with other optical components, need to be

further improved via optimizing design and fabrication. Benefitting from the 3D capability of femtosecond laser writing, more complex structures are expected to improve the integration density for multifunctional devices in crystals, such as lantern waveguides,¹⁷⁴ high-channel interposers,¹⁷⁶ and helical waveguide arrays¹⁷⁵ that have been achieved in glass. Fourth, some femtosecond laser micromachined on-chip LNOI devices as mentioned in Sec. 4.4 are opening an avenue for scalable integrated applications. Notably, the outstanding nonlinear properties of LNOI compared with silicon on insulator will be very attractive for future PICs. Therefore, the performance of LNOI requires further investigation by carefully designed domain structures for enhancing nonlinear conversion efficiency, whereas femtosecond laser-engineered domain modification is an efficient manner. Considering a relatively large nonlinear coefficient ($d_{33} \approx 26$ pm/V), LT-on-insulator (LTOI) is another fascinating alternative for on-chip photonics who possesses a higher optical damage threshold (240 MW/cm²) and a greater UV transparency (0.28 to 5.5 μ m), whereas there is little research in LTOI devices and their nonlinear characterization.²⁴¹ Fifth, laser-written waveguides may receive wider applications in the field of quantum photonics. Most recently, using a single-mode waveguide and a doughnut-shaped waveguide fabricated by femtosecond laser direct writing, Chen et al.⁴⁸ demonstrated the vector vortex beam emitter carrying spin and orbital angular momentum mode inside a photonic chip, allowing for high-dimensional quantum information processing. Further optimizations concern the research of indistinguishable vortex photonic states and on-chip quantum interference. In addition, laser-written waveguides play an important role in single-photon sources, and QMs based on rare-earth doped waveguides, promising potential applications at the visible wavelength band. Large memory arrays and matrices in one crystal can be expected for massively multiplexed QMs in the near future.

In summary, recent advances of femtosecond laser-written waveguides in crystalline materials are overviewed. First, this review presents an introduction to the modification induced by femtosecond laser processing of dielectric crystals, including the fundamentals, control of laser-induced tracks, beam shaping techniques, waveguide geometries, and mode modulation. Track morphology engineering is crucial for not only tailoring the properties of the waveguide but also for the fabrication of novel photonic structures, such as 3D NPCs, 3D domain grating structures, nanopores, and microstructured optical waveguides. Then progress in 3D laser-written photonic devices is discussed, including 3D beam-splitters, tapered waveguides, 3D waveguide arrays, and 3D directional couplers. Last but not least, the selected applications, such as waveguide lasers, frequency converters, QMs, and on-chip devices, have been reviewed in detail. Future challenges and potential topics that could be realized in laser-written waveguides have also been proposed. Overall, it can be concluded that femtosecond laser micromachined waveguides in crystals are still rapidly developing and expected to present the possibility for truly 3D-integrated on-chip photonics in the near future.

Acknowledgments

This work was financially supported by the National Natural Science Foundation of China (Nos. 12174222 and 61775120), the Natural Science Foundation of Shandong Province (ZR2021ZD02), and Taishan Scholars Program of Shandong

Province. The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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